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Comparison of radium-228 determination in water among Australian laboratories

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ABSTRACT

The National Health and Medical Research Council and Natural Resource Management Ministerial Council of Australia developed the current Australian Drinking Water Guidelines which recommend an annual radiation dose value of 1 mSv year^{-1}. One of the potential major contributors to the radiation dose from drinking water is radium-228, a naturally occurring radionuclide arising from the thorium decay series. Various methods of analysing for radium-228 in water have been established and adapted by analytical radiochemistry laboratories. Seven laboratories in Australia participated in analysing radium-228 spiked water samples with activity concentrations ranging from 6 mBq L^{-1} to 20 Bq L^{-1}. The aim of the exercise was to compare and evaluate radium-228 results reported by the participating laboratories, the methods used and the detection limits. This paper presents the outcome of the exercise.

1. Introduction

Radium isotopes are routinely analysed in drinking water as they can contribute to the radiation dose to humans from drinking water sourced from groundwater. The Australian Drinking Water Guideline (ADWG) recommends the determination of radium-226 (^{226}Ra), if gross alpha activity (minus radon-222 contribution) in drinking water exceeds 0.5 Bq L^{-1}, and radium-228 (^{228}Ra) if the gross beta (minus potassium-40 contribution) exceeds 0.5 Bq L^{-1} (NHMRC, NRMMC, 2011).

The determination of ^{226}Ra activity in water samples using co-precipitation with barium sulphate followed by alpha-particle spectrometry developed by Sill (1987) is widely used by analytical radiochemistry laboratories for its reliability and relatively low detection limits (down to 1 mBq L^{-1}).

The determination of low activity concentrations of ^{228}Ra is more challenging. ^{228}Ra can be analysed by gamma spectrometry, using High Purity Germanium (HPGe) detectors, from the activity of its progeny actinium-228 (^{228}Ac) in Marinelli beaker geometries, with sample volumes ranging from 0.5 L to 4 L (IAEA, 2010). Gamma-ray spectrometry is a non-destructive technique which does not require complex and time consuming radiochemical separation procedures. However, typical detection limits are relatively high due to the low detector efficiency and high background.

More sensitive methods of analysing ^{228}Ra have been developed and adapted by radiochemistry laboratories. These methods involve radiochemical processes to isolate ^{228}Ra or its progenies ^{228}Ac or thorium-228 (^{228}Th). The isolated radionuclides of interest are analysed using one of the radiation detection techniques: alpha, beta or gamma spectrometry. The ADWG (USEPA, 1980) does not specify the detection limit required for the determination of ^{228}Ra. However, it states for ^{228}Ra "The estimated limit of determination is 20 mBq/L". Thus, one of the objectives of this exercise is to determine which techniques are suitable for achieving analytical detection limits close to 20 mBq L^{-1}.

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2. Materials and methods

2.1. Sample preparation

The water samples used for the determination of ^{228}Ra activity exercise were prepared at the Australian Nuclear Science and Technology Organisation (ANSTO) Environmental Radioactivity Measurement Centre (ERMC) laboratory. An uncertified thorium-232 (^{232}Th) stock solution with an activity of 1.8 ± 0.05 Bq g^{-1}, reference date 3 September 1992, was diluted to prepare six water samples with ^{232}Th activity ranging from 6 mBq L^{-1} to 20 Bq L^{-1} in 1 L volumes (Table 1). It was assumed the activity of ^{232}Th in the stock solution was in secular equilibrium with the activity of its progeny ^{228}Ra.

The ^{228}Ra activity concentration in the ^{232}Th stock solution was independently verified by analysing 6 diluted aliquots of the solution via ^{228}Th analysis by alpha-particle spectrometry (Martin and Hancock, 2004). A diluted certified thorium-229 solution (Eckert and Ziegler Isotopes Products Ref. no.: 1725-57) was used as the tracer to determine the ^{228}Th recoveries. The measured ^{228}Th activity concentrations in the diluted stock solutions were plotted against the expected ^{228}Ra activity concentrations (Fig. 1) showing a good correlation (R^2 = 0.9978). The expected ^{228}Ra activity concentrations were calculated based on the uncertified activity value of the ^{232}Th stock solution. This verification corroborates the accuracy of the ^{232}Th stock solution reference activity. Six spiked water samples and a blank sample, preserved in 1% concentrated nitric acid, contained in 1 L acid washed bottles, were packed and sent to each of the participating laboratory in March 2016.

2.2. Sample analysis instruction

Participants were requested to use their established analytical methods to determine the activity concentrations of ^{228}Ra in each of the water samples. Laboratories equipped with a gamma-ray spectrometry facility were requested to perform the analysis by the non-destructive method first using gamma-ray spectrometry for 80,000 s and 200,000 s live times, followed by the destructive method by radiochemical processing. Detailed sample analysis instructions are shown in Appendix A.

2.3. Evaluation of reported results

Reported analytical results received from the participating laboratories were evaluated following the International Atomic Energy Agency (IAEA) Terrestrial Environment Laboratory proficiency test scoring method (IAEA, 2007; Shakashiro et al., 2006), which takes into consideration the Trueness and the Precision of the expected and the reported data, including their uncertainties. The first step in the evaluation of each reported result was to determine the relative bias between the expected value and the value reported by the analyst, expressed as a percentage:

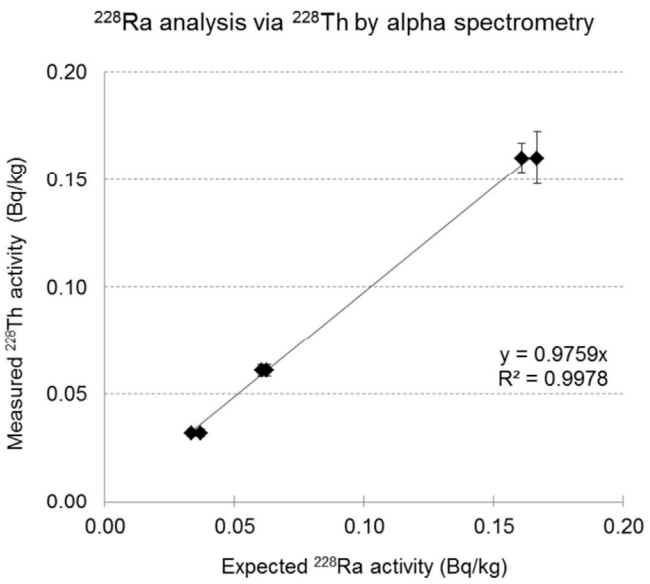

Fig. 1. Measured ^{228}Ra activity concentrations in six diluted ^{232}Th stock solution analysed via ^{228}Th by alpha spectrometry plotted against the expected ^{228}Ra activities.

$$Relative\ \ bias = \frac{Value_{analyst} - Value_{expected}}{Value_{expected}} \times 100\% \quad (1)$$

To evaluate the Trueness of the reported value, the following equations were calculated:

$$A1 = \left| Value_{expected} - Value_{analyst} \right| \quad (2)$$

$$A2 = 2.58 \times \sqrt{Unc._{expected}^{2} + Unc._{analyst}^{2}} \quad (3)$$

The participant result was assigned “Acceptable” for the Trueness test if:

$$A1 \leq A2 \quad (4)$$

To evaluate the Precision of the reported value, the following equation was calculated:

$$P = \sqrt{\left(\frac{Unc._{expected}}{Value_{expected}}\right)^{2} + \left(\frac{Unc._{analyst}}{Value_{analyst}}\right)^{2}} \times 100\% \quad (5)$$

The Limit of Acceptable Precision (LAP) for the ^{228}Ra comparison exercise was set at 20%, a conservative value used by the IAEA in their proficiency tests, which reflects the complexity or difficulty in the measurement of the radionuclide of interest. The participant result was assigned “Acceptable” for the precision test if:

Table 1
Samples prepared for the ^{228}Ra determination in water exercise (^{232}Th stock solution activity: 1.8 ± 0.05 Bq/g).

Sample ID	1st dilution		2nd dilution		Final activity
	^{232}Th stock taken (g)	Diluted to (mL)	Dilution from (mL)	Diluted to (mL)	^{232}Th and ^{228}Ra (Bq/L)
S111	0.453	250	25	1000	0.082 ± 0.002
S112	0.035	250	25	1000	0.0063 ± 0.0002
S113	1.09	250	25	1000	0.20 ± 0.01
S114	11.2	250	25	1000	2.0 ± 0.1
S115	56.1	250	25	1000	10.1 ± 0.3
S116	113.2	250	25	1000	20.4 ± 0.6

$$P \leq 20\% \quad (6)$$

In the final evaluation, both scores for Trueness and Precision were combined. A result must obtain "Acceptable" scores in both criteria in order to obtain a final score of "Acceptable" (A). If a score of "Not acceptable" was obtained for both criteria, the final score will also be "Not acceptable" (N). In cases where either Precision or Trueness score was "Not acceptable", a further check was applied. The relative bias (Equation (1)) of the reported result was compared with the Maximum Acceptable Bias (MAB), also set at 20% for this exercise. The participant result was assigned "Warning" (W) if the relative bias was less than 20%, otherwise the final score was assigned "Not acceptable" (N).

3. Results and discussion

Twenty sets of analytical results were received from seven laboratories, employing a range of analytical techniques to measure ^{228}Ra in the water samples (see Table 2). Five out of seven laboratories reported more than one set of results analysed by several techniques.

Four laboratories performed ^{228}Ra analysis using the Marinelli beaker geometry analysed by gamma spectrometry. The Marinelli beaker volume ranged from 0.5 L to 2 L capacity. One laboratory evaporated the sample from 1 L to 35 mL volume, set the evaporated liquid sample in agar and analysed by gamma spectrometry. The 911 keV line for ^{228}Ac was used by most laboratories as a proxy for ^{228}Ra activity. The less abundant lines 965, 969 and 338 keV were also used by some laboratories.

Five laboratories reported ^{228}Ra results processed by radiochemical separation techniques, followed by analysis by gamma-ray spectrometry (Medley et al., 2015), liquid scintillation counting (LSC) spectrometry (Cook and Kleinschmidt, 2011; IAEA, 2014), gas flow proportional counting (Eichrom, 2014) and alpha-particle spectrometry (Martin and Hancock, 2004). The barium sulphate ($BaSO_4$) co-precipitation method (Sill, 1987) was used by four laboratories to isolate radium from the sample matrix. One laboratory used Eichrom Technologies, Inc. MnO_2 and DGA resins to isolate ^{228}Ac which was then measured using a Gas Flow Proportional Counter (GFPC) (Eichrom, 2014).

All laboratories reported satisfactory results for the 10 and 20 Bq L^{-1} ^{228}Ra samples. The activity concentrations in these samples were relatively high compared to drinking water samples. Activity concentrations of 10 and 20 Bq L^{-1} of ^{228}Ra in drinking water would yield an annual radiation dose well above the levels recommended in the ADWG by about 5 and 10 times, respectively, not accounting for contributions from other radionuclides. It was expected at these activity concentrations that all laboratories should be able to report results in agreement with the expected activity concentrations. The reported results from each laboratory, using varying techniques, for the analysis of the 20 and 10 Bq L^{-1} ^{228}Ra water samples are shown in Figs. 2 and 3. All reported results scored "Acceptable" for the 20 Bq L^{-1} samples. Two results scored "Warning" for the 10 Bq/L-1 samples (results 18 and 19), others scored "Acceptable". Results 9, 10, 11 and 16 reported higher uncertainty values compared to other results, between 8 and 12%; others reported between 1 and 6%. Results 2 and 17 had the lowest uncertainties, less than 1% for the 20 Bq L^{-1} ^{228}Ra samples.

Table 2
Analytical techniques used by the participating laboratories for the determination of ^{228}Ra activity in water samples.

Laboratory	Result no.	Sample preparation	Detection technique	Count time (seconds)
A	1	Isolate radium by preparing $BaSO_4$ micro-precipitates collected on 0.1 μm 25 mm membrane filter papers	Gamma spectrometry ^{228}Ac at 911, 965, 969 keV	80,000
	2	Isolate radium by preparing $BaSO_4$ micro-precipitates collected on 0.1 μm 25 mm membrane filter papers	Gamma spectrometry ^{228}Ac at 911, 965, 969 keV	200,000
	3	Isolate radium by preparing $BaSO_4$ micro-precipitates collected on 0.1 μm 25 mm membrane filter papers	Gamma spectrometry ^{228}Ac at 911 keV	80,000
	4	Isolate radium by preparing $BaSO_4$ micro-precipitate collected on 0.1 μm 25 mm membrane filter papers	Gamma spectrometry ^{228}Ac at 911 keV	200,000
	5 & 6	Isolate radium by preparing $BaSO_4$ micro-precipitate analyse for ^{228}Th ingrowth after 6 months	Alpha spectrometry via ^{228}Th analysis	400,000 to 1,000,000
B	7	Contain sample in a 500 mL Marinelli beaker geometry	Gamma spectrometry ^{228}Ac at 911 keV	200,000
C	8	Isolate Ac using MnO_2 and DGA resins	Gas flow proportional counter	600 (5 cycles)
D	9	Contain sample in a 500 mL Marinelli beaker geometry	Gamma spectrometry ^{228}Ac at 911 keV	200,000
	10	Evaporate sample to less than 35 mL and set in agar in 65 mm × 10 mm Petri dish	Gamma spectrometry ^{228}Ac at 911 keV	200,000
E	11	Isolate radium by preparing $BaSO_4$ micro-precipitates collected as a thin layer at the bottom of a 20 mL vial	Gamma spectrometry ^{228}Ac at 911 & 338 & 969 keV	200,000
	12	Contain sample in a 1 L Marinelli beaker geometry	Gamma spectrometry ^{228}Ac at 911 keV plus others	80,000
	13	Contain sample in a 1 L Marinelli beaker geometry	Gamma spectrometry ^{228}Ac at 911 keV plus others	200,000
F	14	Contain sample in a 2 L Marinelli beaker geometry	Gamma spectrometry ^{228}Ac at 911 keV	80,000
	15	Contain sample in a 2 L Marinelli beaker geometry	Gamma spectrometry ^{228}Ac at 911 keV	200,000
	16	Isolate radium by preparing $BaSO_4$ micro-precipitates dissolved in EDTA and addition of a scintillation cocktail	Liquid Scintillation Counting	3600 (1 cycle)
G	17	Isolate radium by preparing $BaSO_4$ micro-precipitates collected on 0.1 μm 25 mm membrane filter papers	Gamma spectrometry ^{228}Ac at 911 & 965 keV	300,000
	18	Isolate radium by preparing $BaSO_4$ micro-precipitates collected on 0.1 μm 25 mm membrane filter papers	Gamma spectrometry ^{228}Ac at 911 & 965 keV	80,000
	19	Isolate radium by preparing $BaSO_4$ micro-precipitates collected on 0.1 μm 25 mm membrane filter papers	Gamma spectrometry ^{228}Ac at 911 & 965 keV	200,000
	20	Isolate radium by preparing $BaSO_4$ micro-precipitate analyse for ^{228}Th ingrowth after 6 months	Alpha spectrometry via ^{228}Th analysis	300,000 to 500,000

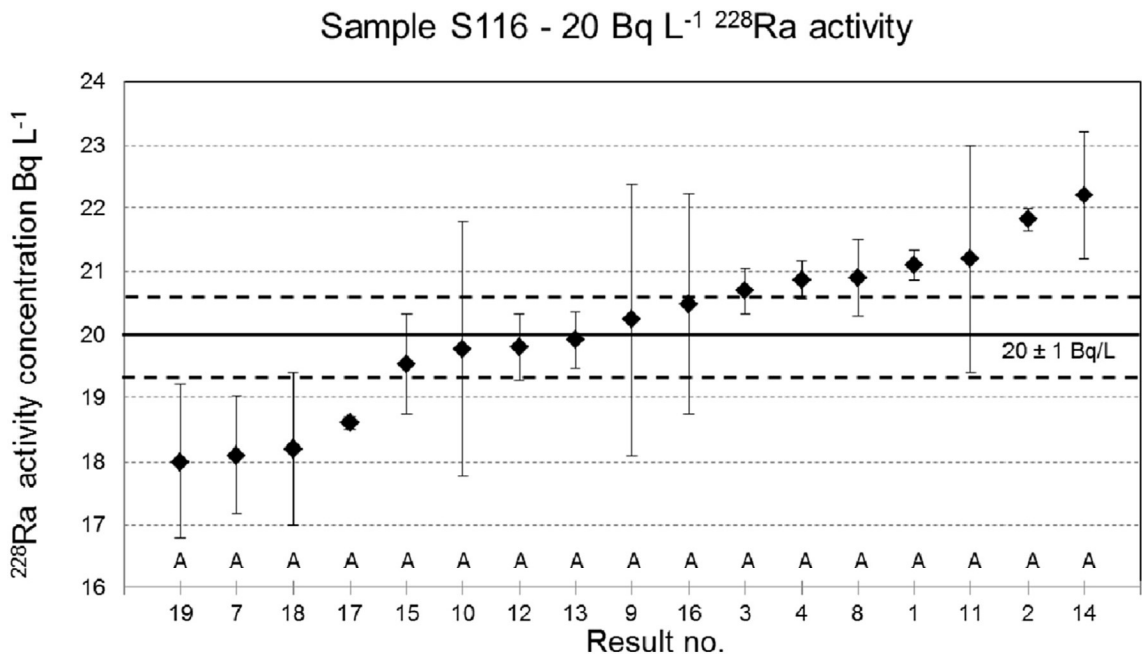

Fig. 2. Reported ^{228}Ra results for the 20 Bq L^{-1} activity samples, compared to the expected activity and the evaluated result scores (A-Acceptable, N-Not acceptable, W-Warning).

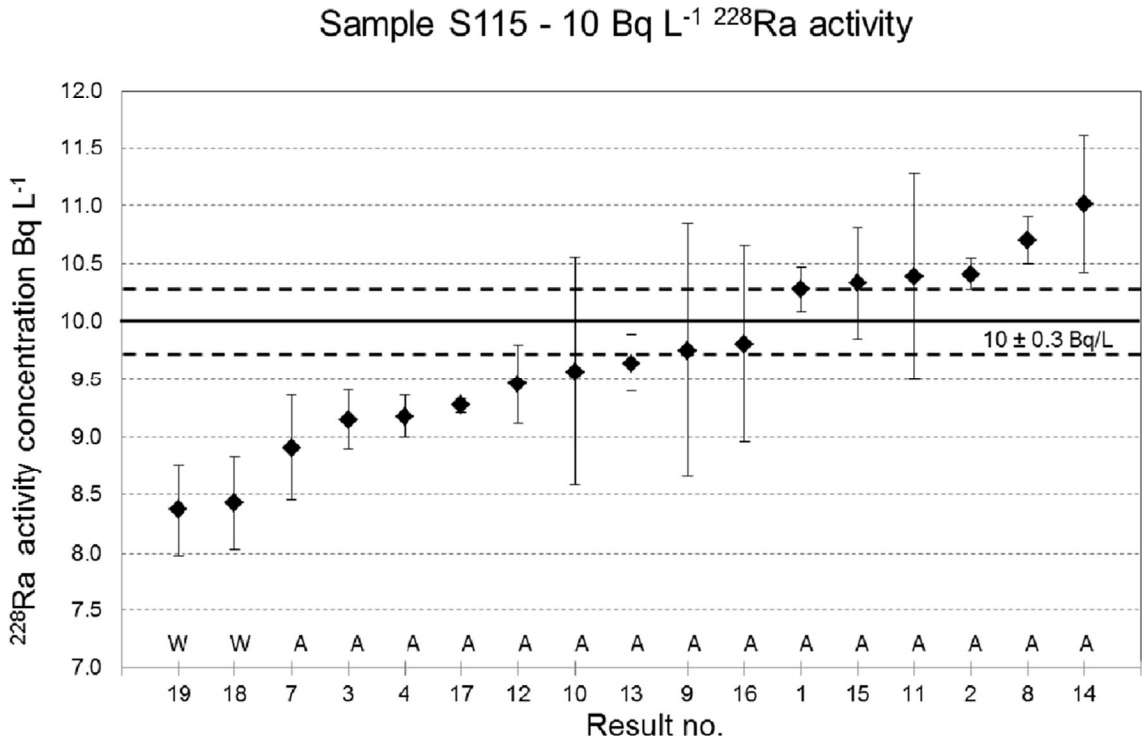

Fig. 3. Reported ^{228}Ra results for the 10 Bq L^{-1} activity samples, compared to the expected activity and the evaluated result scores (A-Acceptable, N-Not acceptable, W-Warning).

One result was given a "Not acceptable" score for the 2 Bq L^{-1} ^{228}Ra sample analysis, whilst all other results scored "Acceptable" (Fig. 4). ^{228}Ra activity at this level in drinking water would yield an annual dose of 1 mSv yr^{-1}. The reported uncertainty values ranged from 2 to 20%. The uncertainty values reported from the Marinelli beaker geometry analysed by gamma-ray spectrometry method (Results, 7, 9, 12, 13, 14 and 15) ranged from 4 to 20%.

Four out of twenty reported results for the 0.2 Bq L^{-1} ^{228}Ra samples were below the method detection limits (Fig. 5). These results were from the Marinelli beaker geometries analysed by gamma-ray spectrometry (Results 7, 9, 14 and 15). Only one result scored "Acceptable" from this technique, with a longer counting time of 200,000 s (Result 13). The result from a shorter counting time (Result 12), performed by the same laboratory for 80,000 s, scored "Not acceptable". The lowest detection limit reported from the Marinelli geometry analysed by gamma-ray spectrometry

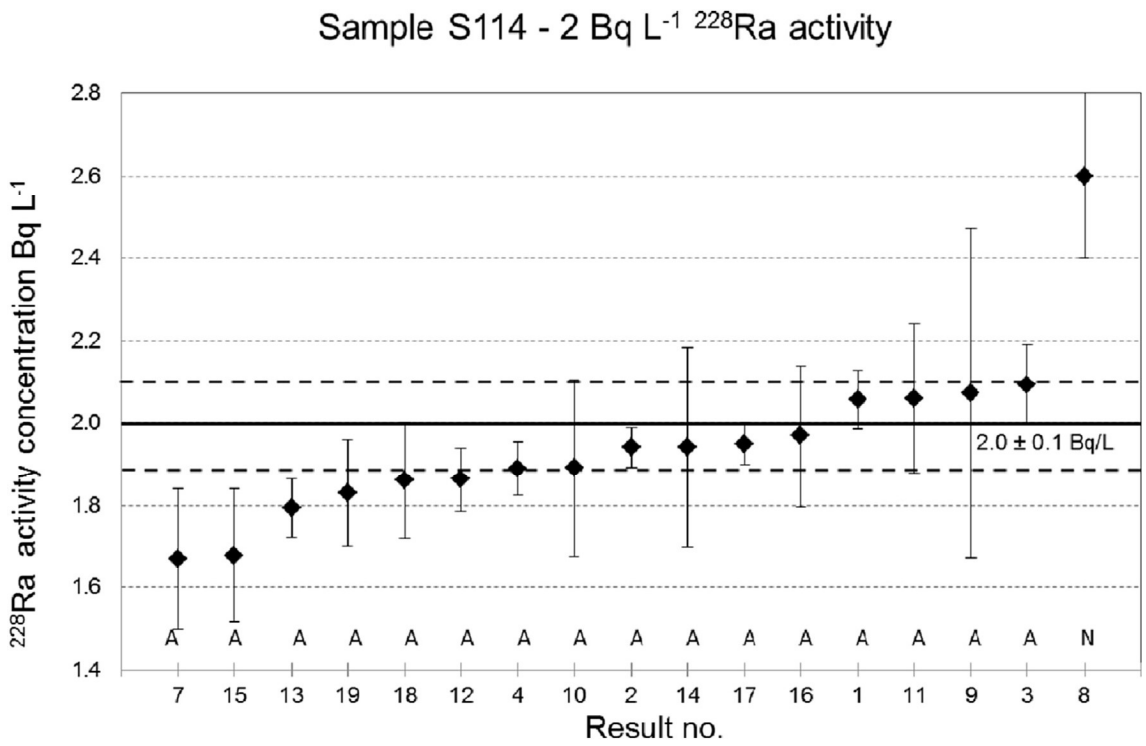

Fig. 4. Reported ^{228}Ra results for the 2 Bq L^{-1} activity samples, compared to the expected activity and the evaluated result scores (A-Acceptable, N-Not acceptable, W-Warning).

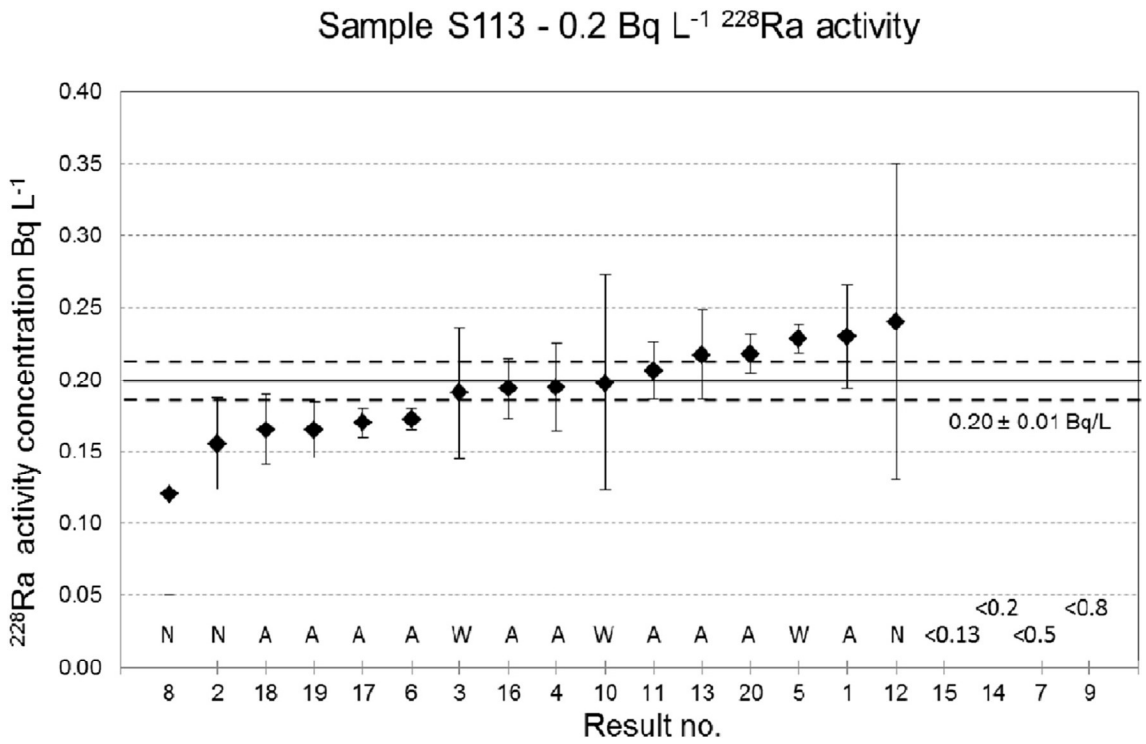

Fig. 5. Reported ^{228}Ra results for the 0.2 Bq L^{-1} activity samples, compared to the expected activity, the evaluated result scores (A-Acceptable, N-Not acceptable, W-Warning), and reported detection limits.

method was 0.13 Bq L^{-1} for 1 L volume and 200,000 s counting time. Other reported results were from techniques which required radiochemical processing. Ten out of sixteen reported results above the detection limits scored “Acceptable”, three scored “Warning” and three scored “Not acceptable”.

Only eleven out of twenty reported results for the 0.08 Bq L^{-1} ^{228}Ra samples were above the method detection limits (Fig. 6). All these results were from techniques which required radiochemical

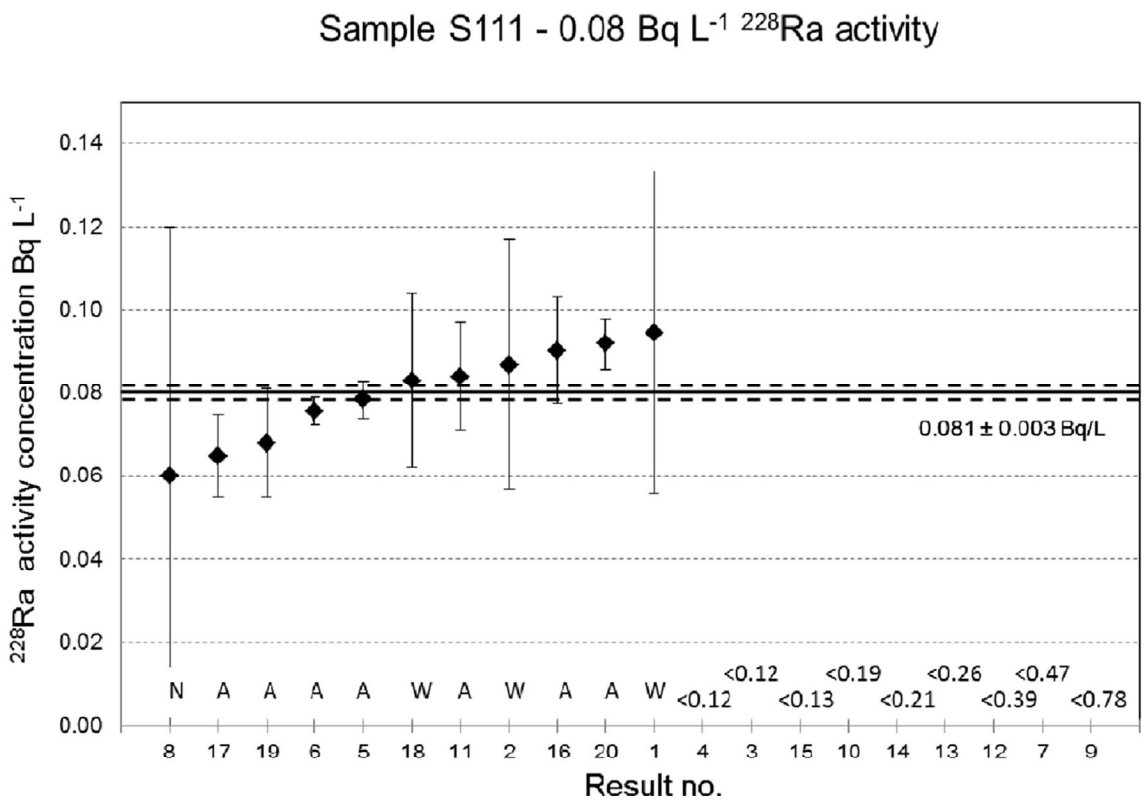

Fig. 6. Reported ^{228}Ra results for the 0.08 Bq L^{-1} activity samples, compared to the expected activity, the evaluated result scores (A-Acceptable, N-Not acceptable, W-Warning), and reported detection limits.

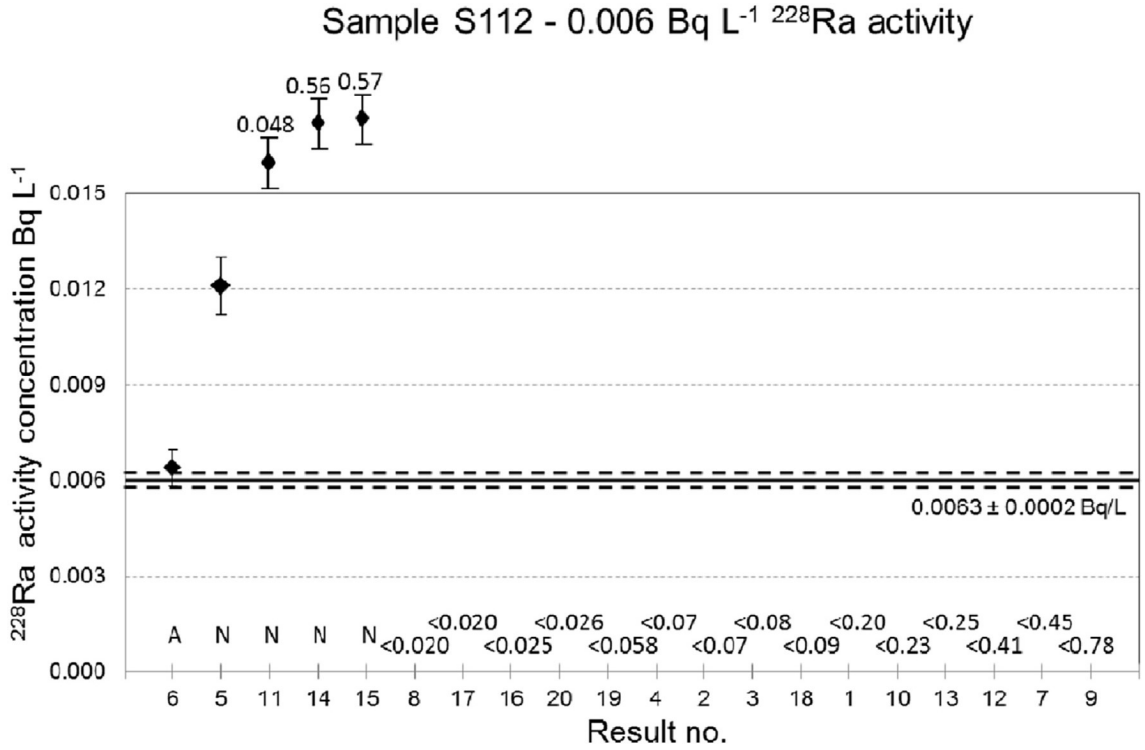

Fig. 7. Reported ^{228}Ra results for the 0.006 Bq L^{-1} activity samples, compared to the expected activity, the evaluated result scores (A-Acceptable, N-Not acceptable, W-Warning), and reported detection limits.

processing. Nine results were processed via the $BaSO_4$ co-precipitation method (Sill, 1987), analysed by gamma-ray spectrometry (Results 1, 2, 9 and 15), 3 results were determined via ^{228}Th analysis by alpha-particle spectrometry (Results 5, 6, 20). The GFPC method (Eichrom, 2014) scored "Not Acceptable" (Result 8). Three results scored "Warning" due to the high uncertainty values (Results 1, 2 and 18) and seven results scored "Acceptable". Results 3 and 4 were below the method detection limits of 0.12 Bq L^{-1}. These results were performed by the same laboratory which reported Results 1 and 2. Results 3 and 4 used a single ^{228}Ac energy at 911 keV, whereas Results 1 and 2 used the combined ^{228}Ac energies at 911, 965 and 969 keV (Medley et al.), which yielded lower detection limits.

Five out of twenty reported results were above the method detection limits for the 0.006 Bq L^{-1} ^{228}Ra sample, however, only one result scored "Acceptable", the rest scored "Not acceptable" due to the overestimation of the activities. The reported detection limits ranged between 0.02 and 0.78 Bq L^{-1} (Fig. 7).

All participants reported below the method detection limits for the blank sample (Table 3). The lowest detection limits, between 0.0006 and 0.016 Bq L^{-1} (Result 5, 6 and 20) were reported from the $BaSO_4$ co-precipitation technique, followed by ^{228}Th analysis, after an ingrowth period of six months, by alpha-particle spectrometry (Martin and Hancock, 2004; Medley et al., 2015). The $BaSO_4$ co-precipitation method analysed by LSC (Cook and Kleinschmidt, 2011) yielded a detection limit of 0.025 Bq L^{-1} (Result 16). The $BaSO_4$ co-precipitation technique analysed by gamma-ray spectrometry (Medley et al., 2015), counted for 200,000 s, yielded detection limits of between 0.047 and 0.09 Bq L^{-1} (Results 2, 4, 11 and 19); counted for 300,000 s the detection limit was 0.026 Bq L-1 (Result 17). The GFPC method (Eichrom, 2014) also yielded a relatively low detection limit of 0.030 Bq L^{-1}(Result 8).

Table 3
Reported ^{228}Ra activity concentrations for the blank sample.

Result no.	^{228}Ra activity Bq L^{-1}
5	<0.0006
6	<0.0008
20	<0.016
16	<0.025
17	<0.026
8	<0.030
19	<0.047
11	<0.07
2	<0.07
4	<0.09
18	<0.10
1	<0.11
3	<0.15
15	<0.18
10	<0.23
13	<0.25
14	<0.29
12	<0.37
7	<0.46
9	<0.78

4. Conclusions

The evaluation of the reported results from the inter-laboratory comparison revealed the techniques used by the participating laboratories in the comparison exercise were satisfactory for the higher activity samples above 2 Bq L^{-1}, with the exception of one technique.

The comparison exercise results showed that radiochemical processing techniques were required to determine ^{228}Ra activities below 0.2 Bq L^{-1}. The $BaSO_4$ co-precipitation method (Sill, 1987) was used by a number of laboratories as the method of choice for the isolation of radium, followed by gamma-ray spectrometry analysis, liquid scintillation counting or alpha spectrometry. The reported results from these techniques were satisfactory down to 0.08 Bq L^{-1} activities. A few results were given "Warning" scores due to the relatively high uncertainty values.

A summary of the detection limits reported for the blank samples, from various techniques used in the comparison of ^{228}Ra determination in water exercise are shown on Table 4. The lowest detection limits of 0.6–16 mBq L^{-1} were reported from the $BaSO_4$ co-precipitation technique, followed by ^{228}Th analysis, after an ingrowth period of six months, by alpha-particle spectrometry (Martin and Hancock, 2004). The main disadvantage with this technique is the long waiting time for the ingrowth of ^{228}Th from ^{228}Ra, which takes at least 6 months.

The $BaSO_4$ co-precipitation method, followed by LSC analysis (Cook and Kleinschmidt, 2011), yielded relatively low ^{228}Ra detection limits of 25 mBq L^{-1}. One advantage of this technique is that it can be set up to analyse ^{228}Ra as well as ^{226}Ra simultaneously.

The $BaSO_4$ co-precipitation method, followed by gamma-ray spectrometry analysis (Medley et al., 2015) yielded relatively low detection limits down to 70 mBq L^{-1}. In order to achieve lower detection limits down to 20 mBq L^{-1}, the sample size and counting time would need to be increased.

This comparison exercise showed the Marinelli beaker geometry analysed by gamma-ray spectrometry technique, without any sample processing, would not be suitable to achieve low detection limits down to 20 mBq L^{-1}. The lowest reported detection limit for the blank sample using this technique was 180 mBq L^{-1} in 1 L marinelli beaker geometry, counted for a longer time of 200,000 s.

The ^{228}Ra results from the GFPC method (Eichrom, 2014), in this comparison exercise, were not satisfactory for samples below 10 Bq L^{-1} activities. However, the detection limit reported from this technique was relatively low, down to 30 mBq L^{-1}.

This comparison exercise also revealed there was a wide range

Table 4
A summary of ^{228}Ra analysis detection limits by various techniques, based on the detection limits reported in the comparison exercise for the blank water samples.

Method	Sample preparation	Detection limit (Bq/L)	
		80,000 s	200,000 s
Gamma spectrometry	0.5 L in marinelli beaker	0.8	0.5
Gamma spectrometry	1 L in marinelli beaker	0.3	0.18
Gamma spectrometry	Evaporation from 1 L to a smaller geometry (35 mL set in agar)	0.3	0.2
Gamma spectrometry	Ra isolation via $BaSO_4$ co-precipitation	0.12	0.07
Gas Proportional Counting	Ra isolation followed by Ac isolation using ion chromatography	0.03	
Liquid Scintillation Counting	Ra isolation via $BaSO_4$ co-precipitation	0.025	
Alpha spectrometry	Ra isolation via $BaSO_4$ co-precipitation Analyse for ^{228}Th ingrowth after 6 months Isolate Th and prepare alpha sources	0.0006–0.016	

of uncertainty values reported by the participating laboratories, between 4 and 20% for the determination of the 2 Bq L^{-1} ^{228}Ra samples analysed by gamma-ray spectrometry in the Marinelli beaker geometries. The varying uncertainty values may be due to varying detector efficiencies and detector backgrounds and/or possibly varying methods of uncertainty determinations among the participating laboratories.

This comparison exercise has shown various techniques used by the participating laboratories to analyse ^{228}Ra in water samples, their reliability in determining activity concentrations from low levels to high and the detection limit for each technique. The data shows the levels of ^{228}Ra detection limits which can be achieved by varying the sample size and counting time, particularly with the alpha-particle and gamma-ray spectrometry techniques. Thus the data can be used to assist analytical radiochemistry laboratories investigating to develop methods for ^{228}Ra determination in water samples.

The current ADWG does not specify required minimum detection limits (MDL) for the determination of radionuclides, including for ^{228}Ra. The radium isotopes MDL value specified in the United State Safe Drinking Water Act is 37 mBq L^{-1} for both ^{226}Ra and ^{228}Ra (USEPA, 2002), and in the European Union (EU) council directive 2013/51/EURATOM is 20 mBq L^{-1} for ^{228}Ra (EropeanUnion, 2013). The data presented in this study showed that only those techniques employing radiochemical separation of radium would be capable of achieving these levels of detection limits (Cook and Kleinschmidt, 2011; Martin and Hancock, 2004; Medley et al., 2015). The ^{228}Ra method analysed by GFPC (Eichrom, 2014) would require further method developments to yield more accurate results.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2017.05.012.

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