Rapid radionuclide specific screening procedures in drinking water: alternative options to replace inaccurate gross activity measurements

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Abstract

It was concluded from two European wide proficiency tests that the gross alpha/beta methods used for drinking water analysis have fundamental pitfalls regardless of the specific gross-counting methods. The majority of gross-counting methods suffer from serious trueness and repeatability issues. To replace inaccurate gross activity measurements an alternative rapid radionuclide specific screening procedure for water analysis is proposed. This procedure considers liquid scintillation counting, alpha-particle- and gamma-ray spectrometry. The proposed procedure is more robust and can achieve lower uncertainties than gross-counting methods. Furthermore, qualitative and quantitative analytical data can be obtained with turnaround times comparable to the gross-counting methods.

Keywords Gross alpha/beta activity · Drinking water · Radionuclide specific analysis · Alpha spectrometry · Liquid scintillation counting · Environmental radioactivity

Introduction

In Europe, it is required from a health protection point of view, that water intended for drinking purposes is checked for radioactivity content before supplying it to the population [1]. Among the specific radioactivity parameters, gross alpha/beta activity concentration is also included in the current Euratom-Drinking Water Directive [1] as a screening parameter to monitor. The most common methods for determination of gross alpha/beta activity concentration in waters are using (1) thick source method [2, 3], (2) thin source method [4] and (3) liquid scintillation counting (LSC) [5]. The difference between thick and thin source method is the deposited residue (surface density) on a counting planchette. Further differences of thick and thin source methods are related to sample preparation. Thick source approach includes four stages: concentration, ignition, sulfation and counting source preparation. Thin source approach follows either evaporation or co-precipitation. Both method can use the same instrumentation for counting (mainly gas-flow proportional counter or solid state scintillation counters).

Regarding liquid scintillation counting, sample preparation is simpler than that of the thin- and thick source method, based either on direct counting or thermal preconcentration. The possible advantages of LSC method is that water sample is simply directly evaporated into a small volume and no conversion of liquid sample into other phase (solid) is required. During preparation of calibration sources, the standardised radioactive solution is introduced in the same phase (liquid) as the water sample, no loss of radioactivity can be expected in the procedure and dissolved materials, radionuclides distributed homogeneously as well as in the sample and in the calibration source. This is not the case when thin- or thick source methods are used because the radionuclides may behave differently in sulfation (distribution on the formed sulphate residue crystals) or co-precipitation process (depends on the valence state).

The claimed arguments for gross activity measurement methods in drinking water analysis are that they are fast, relatively cheap and simple [6]. For these reasons, gross methods are often considered and used as the first or only analytical technique to check radioactivity in drinking waters. However, after the evaluation of two European wide proficiency tests on gross alpha/beta activity concentration measurements in water, it was found that these methods are not always fit for purpose [7–9]. Gross activity measurements are probably the most criticised radioanalytical methods due
to their fundamental pitfalls that lead to serious trueness, repeatability, reproducibility issues and in addition can corrupt the comparability of the measurement results [10, 11]. These pitfalls and sources of interferences were discussed in numerous studies in the past decades [7, 12–15] and called for using radionuclide specific measurements instead [10, 11].

Conventional routine radiochemical preparation methods are considered time consuming and expensive. Due to this outdated assumption, radiochemical methods are still not preferred for drinking water analysis by many laboratories. Also by policy makers and authorities, simplified methods are often preferred where the method and interpretation of results can be quickly understood. Furthermore, management prefers cost and time efficient methods where human and financial resources can be saved. For these reasons, gross alpha/beta measurement methods have been the preferred options in drinking water analysis at many laboratories despite their general pitfalls that lead to inaccuracy under a majority of conditions [7, 11]. However, in the past three decades analytical methods based on radiometric and non-radiometric detection techniques for radionuclide specific analysis were developed enabling high sample throughput and measurement results within 2 days or even a few hours from sample reception [16, 17]. It is still valid that radiochemical methods require proficiency and thorough chemical knowledge of the analysts but some of the recently developed radiochemical methods require less reagents, analyst time and energy consumption than traditional radiochemical methods or the gross methods. In addition, they enable the laboratories to deliver more accurate and true metrological traceable results.

Apart from the technical and metrological aspects, there is another indisputable argument in favour of radionuclide specific methods. Dose coefficients do not exist for gross alpha and gross beta activities, thus the calculation of indicative dose (ID) is possible only from the individual radionuclide activity concentrations. Otherwise, the dose exposure remains just an assumption. Furthermore, the Euratom-Drinking Water Directive [1] clearly mentions, “The ID shall be calculated from the measured radionuclide concentrations…”.

The work in this paper was motivated by the JRC proficiency tests in the field and a previous study by Surbeck [18] where a detailed analytical protocol was proposed to determine individual natural radionuclides in drinking water. In this recent study, the author made an attempt to provide rapid, cost effective radionuclide specific procedures for drinking water analysis using the already existing measurement infrastructure as alternative options to replace inaccurate gross activity measurements. The relative costs and time demand of the gross measurements and radionuclide specific methods are also discussed. Based on the method tested at JRC laboratories or skimming through the available literature a simplified analytical sequence for the determination of mainly natural radionuclides in drinking water is proposed. In addition, the current state-of-the-art analytical techniques are also briefly introduced considering that these methods will soon be or are already used routinely in certain fields.

Last but not least, this paper intends to initiate discussions and motivate the scientific community to propose/use water analytical protocols that could be alternatives for gross methods.

**Experimental**

All used reagents were of analytical grade. Deionized laboratory water, ISO 3696 grade 3 [19] was used for the dilutions. For the chemisorption methods, discs covered with manganese dioxide and Diphonix resin were purchased from Triskem International. The certified radioactive standard solutions were purchased from the National Physical Laboratory (NPL, UK) or from Eckert and Ziegler Analytics (EZAG) or the Czech Metrological Institute (CMI) with traceable massic activities. All uncertainties in the paper are given as standard uncertainties (at $k = 1$) unless otherwise stated.

The $^{209}$Po tracer (Eckert and Ziegler) with original activity concentration of standard solution (37.0 ± 0.7) Bq/g. A general remark: despite the recent updates of the $^{209}$Po half-life value [20, 21], still the old erroneous half-life value can be found on certificates issued by metrology institutes or technical documents.

The original $^{232}$U (NPL) tracer solution from NPL had an activity concentration of (1.02 ± 0.01) Bq g⁻¹. The $^{90}$Sr/$^{90}$Y solution with massic activity of (121.4 ± 1.0) Bq g⁻¹, and $^{241}$Am (80.99 ± 0.40) Bq g⁻¹, radioactive solutions were standardized at the Czech Metrology Institute (CMI, Eurostandard). The certified $^{226}$Ra radioactive solution containing Ba-carrier was provided by the Czech Metrological Institute (CMI, Eurostandard) with traceable massic activity of (3.526 ± 0.018) kBq g⁻¹ ($k = 1$).

$^{40}$K was obtained in the form of KCl (Merck, analytical grade, K assay content 99.5%). The following data were used for $^{40}$K activity calculation: $^{40}$K specific activity considering the beta particle emission probability was (27.9 ± 0.7) Bq g⁻¹ of natural K [7], and the natural isotopic abundance was taken from the Commission on Isotopic Abundances and Atomic Weights website [22]. The beta particle emission probability data was taken from the Decay Data Evaluation Project [23, 24].
Chemisorption and alpha-particle counting

The chemisorption methods were based on the radionuclide specific adsorbing discs developed by Heinz Surbeck [25]. The discs were placed in a polyethylene holder and immersed in the gravimetrically weighed, approximately 100 g of water sample. Samples contained either the dispensed 226Ra or 232U standard solutions or naturally occurring radionuclides. The sample solutions were stirred with approximately 200 rpm for 6 h and 20 h for radium and uranium specific disc, respectively.

Quantification of the different alpha emitting radionuclides was carried out by using high-resolution alpha spectrometry. The alpha spectrometer was equipped with passivated ion-implanted planar silicon (PIPS) detectors with an active area of 450 mm² and 19 keV nominal resolution. The alpha sources (i.e. the discs with manganese dioxide and Diphonix resin) were placed on the stainless steel source holder centered to the detector at a distance of (5.0 ± 0.2) mm. The energy calibration of the alpha-particle detectors was performed by measuring a mixed source (148Gd, 240Pu, 241Am, 244Cm) with a known radionuclide composition that was standardized at JRC-Geel. The alpha counting efficiencies of the PIPS detectors in the chosen configuration and geometry were determined by measuring a standardized single nuclide source of 236U (A = 105.1 ± 0.3 Bq). The alpha counting efficiencies of the PIPS detectors varied between 24 and 28% with an uncertainty of 0.2%. The alpha spectra were analyzed by using commercially available software algorithm.

Gross alpha/beta activity measurements

The method for the gross alpha/beta activity concentrations measurements in water samples was based on the ISO 11704:2018 standard thermal preconcentration approach [5]. In the sample concentration step, an aliquot of 250 g of water was gravimetrically weighed into a glass beaker and acidified to approximately pH 2 using nitric acid if not acidified before. The sample was gently evaporated to approximately 20–30 mL on an electrical plate keeping the temperature at maximum of 80 °C. After the beaker was cooled down and weighed again on an analytical balance, an aliquot of 10 mL of water sub-sample was dispensed into a 20 mL low-diffusion polyethylene liquid scintillation vial already containing 10 mL of Ultima Gold AB liquid scintillation cocktail and mixed thoroughly. The LSC vial with sub-sample was placed into a cooled tray of the liquid scintillation (LS) counter for minimum 3 h to reduce photoluminescence. The samples were measured for typically 6 h and the alpha/beta spectrum was recorded in a previously defined settings using a low background Quantulus 1220 counter (Perkin Elmer).

Before measuring a batch of samples, the pulse shape analyser (PSA) value of the alpha/beta discriminator was adjusted. Alpha and beta counting efficiencies, alpha to beta and beta to alpha spillover were determined by dispensing known activities of alpha emitting (241Am) and beta emitting (90Sr/90Y) radionuclide standard solution to an acidified grade 3 laboratory water and measuring alpha and beta spectra. Blank samples were prepared the same way as the routine samples and were measured before and after measuring a batch of samples.

Radon measurements using gamma-ray spectrometry and liquid scintillation counting

Determination of 222Rn massic activity in water samples was done using a standard gamma-ray spectrometry method [26] based on high purity germanium (HPGe) detectors and liquid scintillation counting [27]. In case of the gamma-ray spectrometry, the measurements were performed on non-opened glass bottles in order to minimize radon-loss from the sample (direct measurement method). For liquid scintillation counting an aliquot of sub-sample was mixed with a non-water miscible cocktail and counted with an ultra-low background counter. The detailed measurements conditions and procedure are described in a previous study [28].

Results and discussion

The performance of rapid methods were compared to the results from standard analytical methods where either reference materials produced for proficiency tests or commercially available reference materials were used.

First, the method based on chemisorption using radium and uranium specific disks were tested for accuracy and recovery. According to literature > 90% of the radium and uranium activity present in the sample can be recovered after 6 h and 20 h of exposure, respectively [29]. To verify the U and Ra recoveries, replicates of proficiency test reference materials and/or spiked samples, spiked blank solutions, were prepared using standardised 232U and 226Ra solutions. The recovery results for uranium and radium are presented in Fig. 1 and Table 1, respectively.

It was observed that the experimental recovery values differed from the literature recovery values. Especially the radium recovery showed a large variation, in general much lower values than that from the literature. One of the reasons for lower uranium recovery from the mineral water samples could be the type and composition of water samples. If waters contain carbonates, then uranyl ions form complexes with them resulting in decreasing chemisorption. However, as part of our standard sample preparation procedure, carbonates are eliminated by acidification.
and heating the water samples. Another reason could be that nitric acid was used to adjust pH instead of the recommended formic acid. For the lower radium recovery, we could not explain the reasons since these waters were either spiked (i.e. without chemical interferences) or natural waters with low Ba concentration. In case of natural mineral water, the radium recovery value was close to the value (75%) in the study by Surbeck [18].

However, from an accuracy point of view, this recovery variation underpins the importance of using internal tracers to be able to determine correctly the recoveries and eventually the uranium/radium activity concentrations. The massic activity of \( ^{234+238}U \) including recovery correction are presented in Fig. 2.

**Table 1** Radium as \(^{226}\)Ra recoveries from different water samples

| Sample type                                           | \(^{226}\)Ra recovery (%) |
|-------------------------------------------------------|---------------------------|
| IAEA ALMERA PT 2019 reference sample 01 (spiked tap water) | 45                        |
| Spiked demineralised water                            | 10–50                     |
| Natural low salinity mineral water                    | 73                        |
| Radon in water PT reference sample (University of Cantabria 2019) | 62                        |
It can be noted that the massic activity values from the JRC reference alpha spectrometry method and the replicates using U specific disc are within the measurement uncertainties. If the total uranium activity concentration is compared, then the difference is smaller. Therefore, the method using U specific discs can be considered reliable.

As far as the spectral properties are concerned, the typical resolution of alpha sources were compared. Alpha sources made by chemisorption was worse (55–100 keV) than that made by micro-coprecipitation (40–75 keV) or electrodeposition (20–40 keV). The resolution itself just increased the uncertainty from spectral analysis due to low energy peak tailing but did not influence the accuracy of the method.

It has to be stressed that the afore discussed rapid radionuclide specific methods cannot replace the more accurate and efficient full scale radionuclide specific methods based on sample preconcentration, separation and source preparation. There are two important beta particle emitting radionuclides ($^{228}$Ra, $^{210}$Pb) that can be analysed by gamma ray spectrometry but due to the detection limit of the method, additional radiochemical analysis may be needed. In addition to the aforementioned straightforward measurement methods, there are other radiochemical methods that enable doing simultaneous multi-radionuclide analysis focusing on the naturally occurring radionuclides that are of interest from a health risk point of view ($^{226}$Ra, $^{228}$Ra, $^{210}$Po, $^{210}$Pb, and the uranium isotopes). These simultaneous methods can provide metrological traceable qualitative and quantitative measurement results with a comparable...
speed, detection limits and uncertainty to gross alpha/beta methods as described by [25, 29–33].

Cantaluppi et al. [34] compared the advantages and disadvantages of different ISO standards dealing with water radioactivity measurements focusing on total sample processing time (i.e. sum of sample preparation and counting time of a single sample). Comparison of the different features of measurement methods can give useful technical information to the analysts. Therefore, they can better plan their work and estimate sample throughput (i.e. how much samples can be processed within a measurement cycle using certain methods). The absolute analytical costs vary from one country to another, influenced also by the local economy. Therefore, it is difficult to give this estimation. However, an attempt is made to make a relative cost estimation (mainly focusing on reagents, lab ware, energy consumption, occupancy of human resources) and comparison between the analytical techniques. Furthermore,

### Table 2 Typical features of the different detector systems [6]

| Detection system                      | Gas-flow proportional counter | Scintillation counter | Low-level liquid scintillation counter | Alpha-particle spectrometry |
|---------------------------------------|------------------------------|-----------------------|----------------------------------------|-------------------------------|
| Energy discrimination                 | No                           | No                    | Yes but bad resolution (300–500 keV)   | Yes resolution: 20–60 keV (depends on source preparation) |
| Simultaneous α/β counting            | Yes                          | Yes                   | Yes                                    | No                           |
| Counting efficiency (%)              | α: 19–42; β: 24–47           | α: 1–30; β: 10–35     | α: > 99; β: > 90                       | α: 20–35                     |
| α/β spillover (%)                    | 0–3                          | 1–5                   | < 1                                    | Not applicable               |
| Self-absorption                      | Yes                          | No                    | Yes                                    | Yes⁹                         |
| Detection of ²²²Rn and ³H             | No                           | No                    | Yes                                    | No                           |
| Automatic sample changer             | Possible                     | No                    | Possible                               | No                           |

*Less or no self-absorption can be obtained, depending on the source preparation approach

### Table 3 Overview of the typical characteristics of the different measurement methods, the gamma-ray spectrometry data partly from [35]

| Methods                                      | Time demand | Manipulations, extra resource needs* | Relative cost per sample# | Typical rel. uncertainty (%) | Detection limit |
|----------------------------------------------|-------------|--------------------------------------|---------------------------|------------------------------|-----------------|
| ²²²Rn measurement by gamma-ray spectrometry | SP: 0–10 min MT: 20–60 h C, QC: 10 min | Direct measurement, sample container | Low                       | 3                            | 2 Bq L⁻¹         |
| ²²²Rn measurement by LSC                     | SP: 5 min, + 3 h waiting time first sample (photoluminescence) MT: 2–6 h C, QC: 10 min | LSC cocktail and vial, syringe | Low                       | 3                            | 4 Bq L⁻¹         |
| Gross activity measurements by LSC           | SP: 4–8 h (evaporation from 250 mL to 40–50 mL) MT: 4–10 h C, QC: 15 min | Evaporation, LSC cocktail, hot plate | Moderate                   | 3–30                        | α: 20–33 mBq L⁻¹; β: 50–125 mBq L⁻¹ |
| ²²⁸Ra and ²¹⁰Po by gamma-ray spectrometry     | SP: 0–8 h MT: 30–150 h; up to weeks C, QC: 30 min | Direct measurement/sample transfer/evaporation, hot plate | Low/moderate               | 5–20                        | 1–60 mBq L⁻¹     |
| ²²⁶Ra + ²¹⁰Po chemisorption and alpha spectrometry | SP: 6.5 h MT: 12–24 h C, QC: 15 min | ¹³⁷Ba and ²⁰⁹Po tracers, magnetic stirrer | Low                       | 4–15                        | < 10 mBq L⁻¹     |
| U chemisorption and alpha spectrometry       | SP: 20.5 h MT: 12–24 h C, QC: 20 min | ²³²U tracer, magnetic stirrer | Low                       | 4–15                        | < 10 mBq L⁻¹     |

*Apart from the normal operation of the already existing measurement system
#Analyst labour cost excluded

SP sample preparation time, MT measurement time, C, QC time for calculation, quality control

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the typical time demand is also studied. The typical features of the different detection systems and the analytical methods including resource requirements are presented in Tables 2 and 3, respectively.

Based on the literature comparison of the analytical methods and our experimental data, the following analytical sequence for drinking water is proposed as a rapid quantitative screening approach.

**Step 1.** Gamma-ray spectrometry for $^{40}$K and $^{222}$Rn, simultaneous sub-sampling for LSC $^{222}$Rn measurement. Direct gamma-ray spectrometry measurement can be sufficient for $^{222}$Rn analysis but needs longer measurement time than LSC to achieve the desired detection limits. Standards to follow: ISO 10703:2021 [36] and ISO 13164-2:2013 [26] for gamma-ray spectrometry and ISO 13164-4:2015 [27] for liquid scintillation counting.

**Step 2.** Evaporation of an aliquot of water sample for more accurate and detailed low-background gamma-ray spectrometry analysis (e.g. $^{40}$K, $^{228}$Ra and $^{210}$Pb) and for gross activity measurements by LSC. Standards to follow: ISO 10703:2021 [33] for gamma-ray spectrometry and ISO 11704:2018 [5] for liquid scintillation counting.

The evaporation speed depends on the power of the hot plate, draft under the fume hood (cooling effect) and the relative humidity in the laboratory air. If more samples are processed together, extra attention has to be paid to avoid cross contamination from splashing samples due to overheating (temperature control, covering samples).

**Step 3.** Radionuclide specific measurements by alpha-particle spectrometry based on chemisorption (nuclide specific discs). Radium and uranium specific disks are applied for the determination of $^{226}$Ra+$^{210}$Po and U isotopes respectively [18, 25].

The schematic overview of the afore-described analytical procedure is presented in Fig. 3.

Of course, these three steps can be simultaneously performed to reduce laboratory analytical time if there is sufficient volume of water sample (500 mL) available that is usually the case.

### State-of-the-art rapid analytical methods

The conventional manual sample preparation and separation steps are usually tedious and time-consuming, sometimes they require up to few days of intense work to complete. To overcome these constraints, the application of automated sample preparation and separation of radionuclides using “on-line” methods (flow injection, sequential injection) have been developed in conventional analytical chemistry. These “on-line” methods were also introduced to radioanalytical procedures but with limitations [16]. Some of the main advantages of such systems are identified:

1. rapid separation,
2. possible to connect directly to the measurement instrument (e.g. ICP-MS),
3. can be fully automated (less human resources and costs needed),
4. saving chemicals,
5. reducing waste production and risk of cross contamination,
6. lower risk of chemical exposure to the laboratory staff,

(+ 1) they could provide very rapid (within few hours) quantitative analytical results in case of nuclear emergency.

As far as the instrumentations are concerned, the techniques of choice for future ultra-low-level analyses of long-lived radionuclides are accelerator mass spectrometry (AMS) and inductively coupled plasma mass spectrometry (ICP-MS). These techniques can achieve detection limits for $^{232}$Th and $^{238}$U at the order of magnitude of nBq g$^{-1}$ [37] while alpha-particle spectrometry can achieve a typical detection limit of 1–10 mBq kg$^{-1}$ for water samples.

The mass spectrometry techniques are considered more sensitive than radiometric measurement techniques when the half-life of a radionuclide is longer than 10 years [38]. For the radionuclides with half-life shorter than 10 years, radiometric methods are more advantageous.

As a conclusion, on the one hand, automated and mass spectrometry based techniques are becoming more and more popular and routinely used in certain fields at many laboratories. On the other hand, these commercially offered options are still too expensive and not available for many laboratories.
Conclusions

The main aim of this paper was to provide an alternative radionuclide specific procedure to replace popular but inaccurate gross activity measurement methods of drinking water. An attempt was made to propose rapid, cost effective radionuclide specific procedures in drinking water analysis using the already existing measurement infrastructure. A simplified protocol for the determination of mainly natural radionuclides in drinking water has been introduced. Based on the relative costs, additionally required resources and time demand of the different methods, it was found that certain radionuclide specific methods (e.g. nuclide specific disks combined with alpha-particle spectrometry) does require much less time, human resources and energy consumption (electrical or gas) than gross measurement methods based on evaporation. The experience from the tests described in this paper is that replacing gross methods with radionuclide-specific methods for drinking water analysis does not require extra resources or investments. It is rather the contrary; they would save analyst time and laboratory resources. These radionuclide specific measurements could be performed either by the same instrumentation (gas-flow proportional counter, liquid- and solid scintillation counter) that are used for gross alpha/beta analysis or instruments that are essential parts of a routine radiometric laboratory nowadays high-resolution gamma-ray spectrometer using high-purity germanium detectors (HPGe) and alpha-particle spectrometer. Still, the role of gross methods could be considered as complimentary analytical methods to the radionuclide specific methods in the future.

Furthermore, there are three main undisputable advantages of the proposed methodology (applicable to radionuclide specific methods in general) that gross-counting methods cannot meet:

1. establishing metrological traceability,
2. clear identification of analytes/measurands,
3. calculation of indicative dose from the individually determined radionuclides.

One of the few investments laboratories would have to make is to develop expertise/proficiency of the laboratory staff. Another important requirement towards quality assured work is that methods shall be validated but usually in radiochemistry laboratories working according to ISO 17025 standard [39], method validation already form an essential part of the quality system. Routine laboratory measurement protocols must be up to date to comply with the recent food safety-, health- and environment-related challenges where rapid, specific and accurate measurement methods are required, that can provide true measurement results and not just numbers or semi-quantitative values.

In the near future, more extended involvement of mass spectrometry methods in environmental sample analysis, also in water quality analysis, is expected. Although they are still expensive instruments, the scientific community has to be prepared and open to make fundamental changes in the analytical protocols and application of routine methods.

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