

Requirements for the radiological monitoring of drinking water: Guidance provided to the Drinking Water Inspectorate

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ABSTRACT

This report begins by summarising the approach to the monitoring of drinking water given by the European Commission (EC) in its Drinking Water Directive. Guidance issued subsequently by the Drinking Water Inspectorate (DWI) is also brought together. A process is then developed that guides the water companies through the various stages of investigation following an exceedance of a criterion on gross activity. This process conforms with the guidance issued by the EC and draws on that provided by DWI. Advice is provided on sampling philosophy and the development of a suitable analytical strategy.

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EXECUTIVE SUMMARY

In 1998, the European Commission issued its Drinking Water Directive. This covers a wide range of potential pollutants. For radionuclides, the Directive gives values for two radiological indicator parameters. These are a concentration of tritium of 100 Bq l^{-1} and a total indicative dose (TID) of 0.1 mSv y^{-1} . The Directive does not specify the frequency of monitoring that is required, nor the scope of monitoring needed for the estimation of TID. The Directive does specify however that tritium, ^{40}K , radon and radon decay products should not be included in the estimation of TID.

In England, the Drinking Water Directive (DWD) has been implemented by means of the Water Supply (Water Quality) Regulations 2000, published by the Drinking Water Inspectorate (DWI). For radioactivity, these require monitoring for tritium together with measurements of gross alpha and gross beta activity. The Regulations also include a provision that allows the Secretary of State to permit water companies not to monitor concentrations of tritium or to estimate TID if there is satisfactory evidence to show that the actual values are well below the specified levels.

In 2005, DWI placed guidance on the Regulations on its website (www.dwi.gov.uk). This related to both England and Wales. The guidance largely echoed the draft material on monitoring issued by the EC. However, it covered all potential pollutants in drinking water, and information on radioactivity was widely distributed between various parts of the guidance. Shortly after its guidance had been issued, DWI recognised that further advice on radioactivity was needed, since for example no guidance was provided on which radionuclides should be considered in the event that a screening level was exceeded. This was a particularly important factor because many water companies were gathering significant amounts of monitoring data, and there had been a few occasions on which the screening value for gross alpha activity has been exceeded. The types of further investigation that had been carried out in these cases had varied between different companies. DWI therefore commissioned the Radiation Protection Division of the Health Protection Agency (HPA-RPD) to provide specialist advice. The output from this work is described in this report.

The report begins by summarising the guidance contained within the EC Drinking Water Directive and then draws together the material on sampling, analysis and assessment issued by DWI. For many potential pollutants, DWI already requires sampling to be undertaken at a point of supply ie after any water treatment has been carried out. The same approach needs to be adopted for radioactivity. This is because the criterion on TID relates to the consumer, and so the sample needs to come from water that will actually be drunk. Some other monitoring programmes are based on sampling from sources of raw water. The resultant analytical data should not be used to estimate TID because various water treatments can be effective in removing radionuclides.

The guidance issued by the EC and DWI has been used to develop a process for the analysis of drinking water and the subsequent assessment. The process is based on the measurement of gross alpha and beta activities, a procedure that has been in widespread use in the water industry for many years. The criteria that would trigger further investigation are 0.1 Bq l^{-1} for gross alpha and 1 Bq l^{-1} for gross beta, which are

the values specified by the EC. Practical guidance is provided on the likely causes of the criteria on gross activity being exceeded, and on the implications for further investigation and the estimation of TID. In the United Kingdom, the main contributors to gross alpha activity are likely to be naturally - occurring radionuclides such as radium-226 (^{226}Ra) and isotopes of uranium. For gross beta, potassium-40 (^{40}K) could be important. This radionuclide is of particular interest because it is among those that the Drinking Water Directive specifies should not be included in the estimation of TID.

Reference concentrations for selected radionuclides have been issued by the EC, and these provide an intermediate step between measurements of gross activity and the estimation of TID. This type of approach is very helpful to non-specialists in radiological protection because reference concentrations can be compared directly against measured values. For this reason, this report contains reference concentrations for an expanded range of radionuclides, derived using the parameter values specified by the EC.

Exceedances in the criteria on gross activity would require individual radionuclides to be determined specifically. This report gives advice on the selection of radionuclides for specific analysis. It also contains guidance on the development of an analytical strategy and the resources required.

The EC Directive emphasises that the TID refers to a dose received over a full year. Consequently, a single measurement need not necessarily be a cause for concern. It should however be a trigger for further sampling and analysis. Guidance is provided on how such work could be structured.

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1 INTRODUCTION

The Drinking Water Directive issued by the European Commission (EC) gives values for two radiological indicator parameters (European Council, 1998). These are a concentration of tritium of 100 Bq l^{-1} and a total indicative dose (TID) of 0.1 mSv y^{-1} . The Directive does not specify the frequency of monitoring that is required, nor the scope of monitoring needed for the estimation of TID. The Directive does specify however that tritium, ^{40}K , radon and radon decay products should not be included in the estimation of TID.

In England, the Drinking Water Directive has been implemented by means of the Water Supply (Water Quality) Regulations 2000 (DWI, 2000). These require monitoring for tritium together with measurements of gross alpha and gross beta activity. An audit frequency is specified and monitoring must be carried out either at supply points (usually a water treatment works) or in water supply zones (which would normally mean at consumers' taps). The Regulations also include a provision that allows the Secretary of State to permit water companies not to monitor concentrations of tritium or to estimate TID if there is satisfactory evidence to show that the actual values are well below the specified levels.

In 2005, the Drinking Water Inspectorate (DWI) placed guidance on the Regulations on its website (www.dwi.gov.uk) (DWI, 2005). This related to both England and Wales. The guidance largely echoed the draft monitoring directive issued by the EC. However, it covered all potential pollutants in drinking water, and material on radioactivity is spread throughout the document. In addition, DWI recognised that further advice on radioactivity was needed, since for example no guidance was provided on which radionuclides should be considered in the event that a screening level was exceeded. The guidance did however state that further investigation need not necessarily involve a comprehensive analysis of all radionuclides.

Many water companies have now gathered significant amounts of monitoring data, and there have been a few occasions on which the screening value for gross alpha activity has been exceeded. The types of further investigation that have been carried out in these cases have varied between different companies. DWI considered that it needed specialist advice to enable it to decide whether such further investigations were adequate in defining whether the specified requirements on TID had been met and whether there were any risks to public health from the radionuclides detected. The Radiation Protection Division of the Health Protection Agency (HPA-RPD) was asked to provide such advice. The work is set out in the remainder of this report.

2 THE APPROACH PUT FORWARD BY THE EC

2.1 Screening

The system put forward by the EC enables Member States to use simple screening methods involving the determination of gross alpha or beta activity as a first step towards conformance with the criterion on TID. If the gross alpha and gross beta activities are less than 0.1 Bq l^{-1} and less than 1 Bq l^{-1} respectively, then the Member State may assume that the parametric indicator value for TID of 0.1 mSv y^{-1} has not been exceeded and no further radiological investigation is needed. There are however two important caveats. These are as follows.

- (i) Any contributions from tritium, ^{40}K , radon and radon decay products are excluded from the estimations of gross alpha and beta activity.
- (ii) Even if the criteria on gross activity were met, further investigation would be needed if other sources of information indicated that specific radionuclides were present in the water supply and that activity concentrations were such that a TID value of 0.1 mSv y^{-1} might be exceeded.

Tritium is an isotope of hydrogen and emits a beta particle that is low in energy. In practice, the methods for the determination of gross beta activity that are in common use would not take account of tritium because they involve the removal of radionuclides from water. In addition, the counting procedures used are generally only sensitive to more energetic beta particles, and so tritium along with other low-energy beta emitting radionuclides such as ^{14}C and ^{35}S would not be included in the measured value. Consequently, if there are reasonable grounds to expect that radionuclides are present in the drinking water and that these would not be detected using the established analytical methods for gross activity, then a more specific analytical technique would be required.

The other radionuclides that are excluded in the estimation of TID also deserve comment. Potassium-40 (^{40}K) emits beta particles as well as gamma rays and occurs naturally. Potassium is under homeostatic control in the human body and so the dose from ^{40}K is not dependent on the amount of activity that is ingested. Radon is a noble gas that is readily expelled from water on aeration. It also occurs naturally, being a decay product of ^{238}U (uranium-238), and is relatively short-lived, having a radioactive half-life ($T_{1/2}$) of about 3.8 days. It decays via a series of short-lived progeny to ^{210}Pb (lead-210) ($T_{1/2}$ about 22 y) and thence via ^{210}Bi (bismuth-210) and ^{210}Po (polonium-210) ($T_{1/2}$ about 138 d) to the stable isotope ^{206}Pb (lead-210). All of these radionuclides are classed as radon decay products.

2.2 Radionuclide-specific analyses

If either the gross alpha activity exceeds 0.1 Bq l^{-1} or the gross beta activity exceeds 1 Bq l^{-1} , then the EC requires the Member State to carry out radionuclide specific analyses. The choice of radionuclide to be determined is left to the Member State, and

selection should take account of all relevant information about the likely source of the radioactivity. The EC notes that since elevated levels of tritium may indicate the presence of artificial radionuclides, then concentrations of tritium and gross alpha and beta activity should be made on the same sample.

The EC also gives Member States the discretion to adopt a different approach to initial screening. There may be situations where the presence of particular radionuclides indicates reliably that radioactivity is present in the drinking water. In such cases, the Member State may choose to determine these radionuclides specifically, rather than carrying out measurements of gross activity.

For selected radionuclides, the EC has derived so-called reference concentrations. These are activity concentrations of a particular radionuclide that, if maintained in drinking water over a full year, would give a dose of 0.1 mSv^{*}. These reference concentrations are based on an adult consuming 2 l of water per day over a year, and make use of the relevant coefficients published in a Euratom Directive that relate intakes of activity with dose (European Council, 1996).

If the concentration of one of the specific radionuclides being measured exceeds 20% of the reference concentration, or if the concentration of tritium exceeds 100 Bq l⁻¹, then the analysis of further radionuclides is required. The 20% value could be regarded as a trigger level above which further investigation is needed, and so for convenience this term has been used in the remainder of this report. The choice of which radionuclides to consider is a matter for the Member State, and the decision should take account of all relevant information about the likely source of the radioactivity. Both natural and artificial radionuclides should be taken into account, although as noted in Section 2.1, tritium, ⁴⁰K, radon and radon decay products are excluded from the estimation of TID.

The trigger level has been set well below the reference concentration because the TID refers to all of the radionuclides present in the drinking water. The EC provides guidance on the summing of the effects of several radionuclides, making use of the quotients of the measured value and the relevant reference concentration. If the sum of these quotients is less than one and all appropriate radionuclides have been considered, then Member States may assume that the TID is less than 0.1 mSv. No further investigation is then required.

Trigger levels and reference concentrations are useful because they provide quantities that can be compared directly with measured values. Provided that none of the measured values exceeds the relevant trigger level then there is no need to carry out the more complex process of evaluating the TID. This can be advantageous for those monitoring teams that have limited expertise in radiological assessments. It should be noted however that the EC provides reference concentrations (and hence trigger levels) for a relatively small number of radionuclides, albeit those that are likely to be most commonly encountered. Similar guidance has been issued by the World Health

^{*} In this report, unless otherwise stated the term "dose" refers to the effective dose. This quantity takes account of the type of radioactivity emitted by a particular radionuclide (alpha, beta, gamma), its behaviour within the human body and the sensitivity of different organs in the body to radiation. It enables the effects of different radionuclides to be brought on to a common basis.

Organisation (WHO) (WHO, 2004). However, although WHO provides reference concentrations for a wide range of radionuclides the numerical values are presented simply in terms of orders of magnitude, ie, 1, 10, 100 Bq l⁻¹ etc. Consequently, they cannot be used within the system set out by the EC.

2.3 Estimation of TID

The TID is estimated using the measured activity concentrations of all of the appropriate radionuclides together with the relevant dose coefficients for adults and a water consumption rate of 730 l y⁻¹ (2 l per day). This approach has been widely used in the development of international recommendations on drinking water quality for many years (WHO, 2004). The assumed consumption rate is considerably greater than the value considered appropriate for typical adults in the UK (NRPB, 1994). This has implications for situations where the criterion on TID is exceeded, and these are discussed later in this report (Section 4.6)

The EC has used the dose coefficients laid down in the 1996 Euratom Directive (European Council, 1996) in the calculation of reference concentrations but states that more recent data can be used if these are recognised by competent authorities in the Member State. The system proposed by the EC does not specifically state the need to estimate the TID. However, this would need to be done if there were radionuclides present for which no reference concentrations were available.

The EC does note that, in cases where a set of measurements indicates that the TID criterion will be exceeded, an annual dose of greater than 0.1mSv criterion will only occur if the activity concentrations are maintained over a full year. It is for the Member States to decide on the frequency of sampling needed to provide a reasonable estimate of the actual annual dose.

3 GUIDANCE PROVIDED BY DWI

The DWI provides guidance to the water industry via its website (www.dwi.gov.uk). This covers a wide range of potential pollutants, and so the guidance on sampling, analysis and assessment of radionuclides is not all in the same section.

For many potential pollutants, DWI requires the water companies to collect samples from supply points, ie after any water treatment has been carried out. The same approach needs to be adopted for radioactivity. This is because the criterion on TID relates to the consumer, and so the sample needs to come from water that will actually be drunk. The concentrations of many of the radionuclides that might be encountered in raw water are likely to be reduced as a result of the types of treatment commonly employed in the UK (Brown et al, 2008). The water companies may well carry out sampling and analysis within and upstream of the treatment plant for other purposes, but the data should not be used for comparison with criteria on reference concentrations and TID.

DWI also specifies a frequency of sampling, based on the amount of throughput. It would be reasonable to adopt this frequency for the monitoring of radionuclides, at least until a reasonable amount of monitoring data had been accumulated over a period of at least one year. A review of these data might then indicate that a change in sampling frequency was warranted.

The guidance on the subsequent analysis for radioactivity begins with a requirement to assess gross concentrations of alpha and beta activity against the criteria specified by the EC. This is a sound approach because the majority of water companies in the UK have established an extensive capacity to carry out such monitoring, whereas the capacity for radionuclide-specific analyses is too limited to accommodate all of the monitoring needed. In addition, the monitoring data accumulated so far both by the water industry and other organisations such as the Environment Agency (EA) indicate that in most cases the criteria on gross activity are met.

In the event of a criterion on gross activity being exceeded, the guidance specifies the need to establish which radionuclides are present, taking account of any relevant information on potential sources. It points out that these further analyses should not necessarily involve a comprehensive evaluation of all radionuclides. For most of the sources of drinking water in the UK, most if not all of the radioactivity is expected to be of natural origin. Compilations of monitoring data support this view (Environment Agency et al 2007). Consequently, in the event of an exceedance of the gross alpha activity criterion, DWI recommends that in the first instance the water companies should consider the possible presence of naturally-occurring isotopes of uranium as well as radium-226 (^{226}Ra). The guidance states that if most of the gross activity can be accounted for by such naturally-occurring radionuclides, then the calculation of TID should be based on these radionuclides alone. It should be emphasised at this point that even if all alpha-emitting radionuclides are determined specifically, the sum of their activities is very unlikely to agree with the result from a gross alpha measurement because of differences in the counting techniques employed, together with the associated uncertainties in the measurements. A similar situation applies to the analysis of beta emitting radionuclides.

The guidance moves directly from radionuclide-specific analyses to an estimation of TID, with the exclusions specified by the EC, although the DWD reference concentrations are given in an appendix. Some specific guidance is given concerning the analysis of tritium. In common with the EC, DWI emphasises that the TID is specified in terms of the dose over a year. The Inspectorate also notes that there may be variability in the observed activity concentrations over that period, due for example to natural processes. An increase in river flows may result in a dilution of any discharges of artificial radionuclides, while flood conditions may increase the amount of run-off from soil with a consequent increase in concentrations of some naturally-occurring radionuclides. The implication therefore is that if a single sample gives data that would result in the criterion on TID being exceeded, then further sampling and analysis would be needed for a period of up to one year before a definite view on exceedance can be obtained.

The guidance also distinguishes between on-going situations and short term increases due to radiological incidents. In the event of an incident, activity concentrations in

drinking water should be assessed against the intervention levels issued by the former National Radiological Protection Board (NRPB) (now the Radiation Protection Division of HPA) (NRPB, 1994), which have been placed in guidance issued by DWI (1995). The present report focuses however on the routine situation.

4 RECOMMENDED PROCESS FOR THE ROUTINE MONITORING OF DRINKING WATER

This section sets out a proposed analytical and assessment process. It has been assumed that sampling has been carried out at the point of supply, as already specified by DWI. This differs from the approach taken by Government Departments and Agencies, where the raw water is sampled prior to treatment (Environment Agency et al, 2007). This difference needs to be taken into account if measurement data between these two programmes are compared. Differences are likely for the majority of radionuclides if chemical processes have been used in the treatment of the drinking water. A compilation of information has been published on the likely levels of removal that can be expected (Brown et al, 2008).

It has also been assumed that any treatment required to maintain sample integrity has been carried out. Thus, for example, unless elements such as iodine are to be determined the sample might be acidified or amended with a carrier solution in order to prevent adsorption of radionuclides on to the surface of the container. No further guidance is given in this document on sample stabilisation and preparation. The process is summarised in the flowchart given in Figure 1.

The main aspects are discussed further in the remainder of this section.

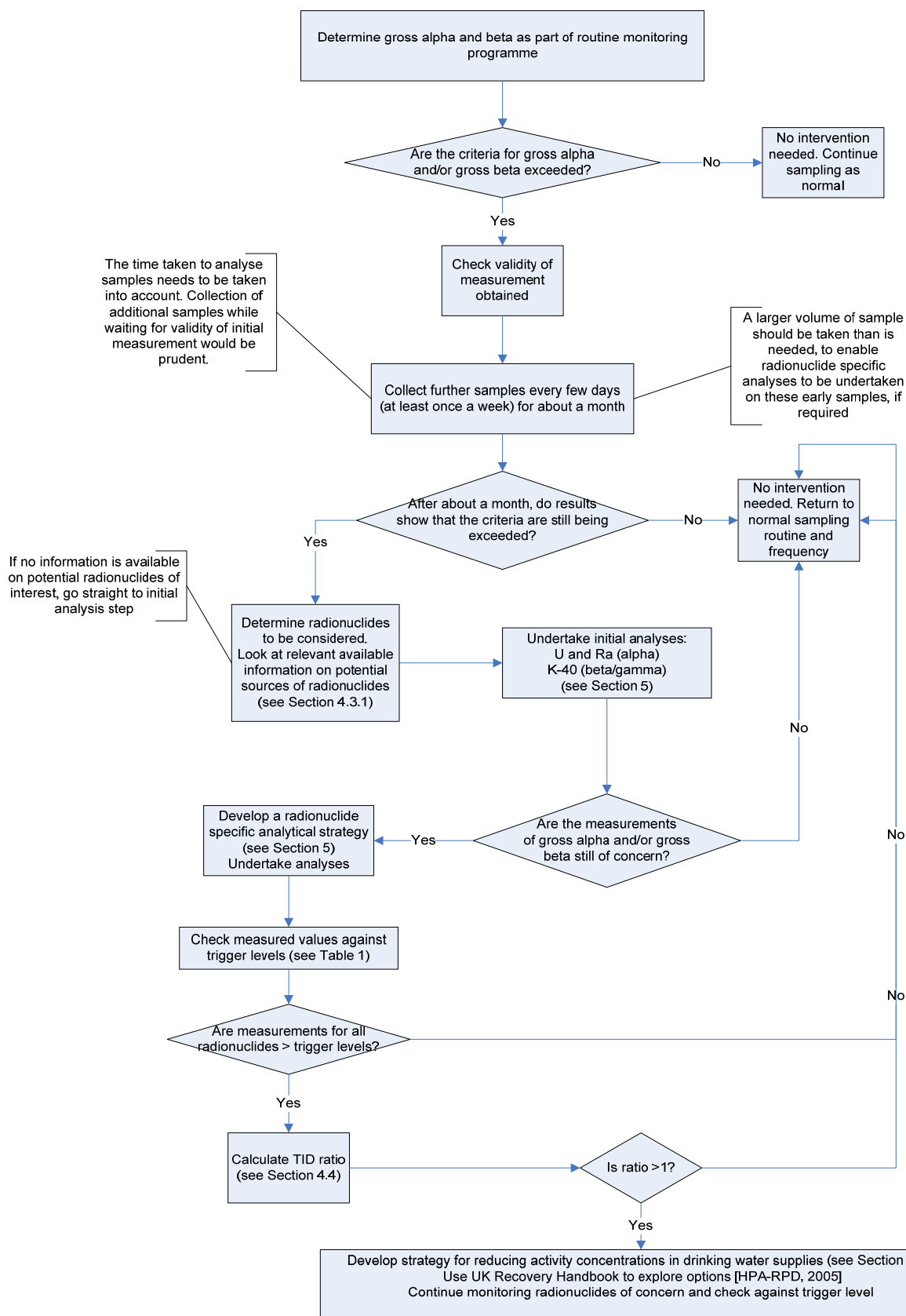


Figure 1: A process for the routine monitoring of drinking water

4.1 Determination of gross concentrations of alpha and beta activity

As noted in Section 3, the water industry in the UK is already well-equipped to carry out its own measurements of gross activity, whereas the capability for radionuclide-specific analyses is much more limited. The equipment needed for the determination of specific radionuclides is expensive and requires dedicated experienced staff. The industry is required to carry out extensive monitoring on a range of potential contaminants and given that the majority of samples conform to the criteria on gross activity there seems no good reason to move to a system based on radionuclide-specific analyses. The existing guidance from DWI already takes this view.

The existing statutory monitoring focuses on the measurement of gross alpha activity. This is understandable given that most supplies of raw water are more likely to be affected by naturally-occurring alpha emitting radionuclides such as ^{234}U , ^{238}U or ^{226}Ra . However, many radionuclides that might be found in water supplies emit beta particles. Examples would be the naturally occurring radionuclides ^{40}K , ^{210}Pb and ^{210}Bi , all of which are excluded from the estimation of TID, and artificial radionuclides such as ^{60}Co (cobalt-60), ^{90}Sr (strontium-90) and ^{137}Cs (caesium-137). Strontium-90 and, to a much lesser extent, ^{137}Cs are detectable in sources of raw drinking water in England and Wales (Environment Agency et al, 2007). However, the observed concentrations are very small, well below the criterion on gross beta activity of 1 Bq l^{-1} . The gross beta activity tends to be dominated by ^{40}K . Nevertheless, a requirement for the water industry to determine gross beta activity should be retained in order to demonstrate the overall potability of the water supply. In addition, many of the radionuclides that might be released in nuclear reactor accidents or from incidents involving industrial sources emit beta particles. Measurements of gross beta activity can be useful in such circumstances, since they may remove the need for more radionuclide-specific analyses (Environment Agency, 2002). It is important therefore that the capability to measure gross beta activity is maintained.

4.2 The action required if a criterion on gross activity is exceeded

If the criterion on either gross alpha or gross beta activity is exceeded, then the first task would be to check the validity of the result. It may be possible to use the data from other samples analysed in the same batch to demonstrate that the procedure and the measurement equipment itself are working properly. Checks on instrument calibration and background would also be needed. If a sufficient amount of the original sample is available, then a repeat analysis should be carried out. It is recognised however that water companies already collect large numbers of samples for a wide range of analyses. Consequently, the archiving of sufficient volumes of all samples, even for a relatively short time, may not be practicable.

As noted earlier, the criterion on TID relates to an annual dose, and so if the criteria on gross activity are only exceeded for a short time then this does not necessarily imply that the TID will be exceeded. Once the validity of the initial measurement data have been established, the next task is to collect further samples from the relevant supply and

carry out further measurements of gross activity. A reasonable amount of data is needed quickly in order to assess the situation. As the time between collecting the original sample and having the analytical data could be up to a few days (Environment Agency, 2002), it would be prudent to collect a further sample before any checks on the original sample were completed. Samples should therefore be taken every few days and not less than once per week. However, if the gross activity concentrations fell below the criteria then it might not be necessary to carry out analyses on all of the samples that had been collected. It does follow therefore that the water companies do need the capability to increase sample throughput, albeit for a relatively short period.

The amount of sample collected during this phase of the investigation also deserves comment. If the criteria on gross activity continue to be exceeded, then radionuclide-specific analyses will be needed. It is worth making sure that a sufficient volume of sample is collected each time so that if necessary these types of analysis can be carried out later.

As was emphasised in Section 2.3, the TID relates to an annual dose and concentrations of radionuclides will need to remain elevated over that sort of period if the criterion is to be exceeded. If measurements taken over a period of about 4 weeks fell below the criteria for gross alpha and / or beta, then no intervention would be needed to reduce doses to members of the public. Some occasional sampling might still be needed to provide reassurance that the low concentrations were being maintained. If the measured gross concentrations remained in excess of the criteria over a period of around 4 weeks, then sampling should continue on at least a weekly basis, depending on the concentrations being found. At this stage consideration would also need to be given to more specific analyses, as described below in Section 4.3.1.

4.3 The development of a strategy for radionuclide-specific analyses

A comprehensive determination of all radionuclides is neither practicable nor desirable. This section outlines how the radionuclides of interest can be selected and how to develop an analytical strategy.

4.3.1 The use of relevant information to determine likely radionuclides causing concern

The EC comments that all relevant information should be taken into account when deciding on the radionuclides to be studied. This should take account of other monitoring data within the area of interest. For drinking water, this would include the catchment from which the raw water was drawn. There are various sites across the UK that are licensed to discharge small quantities of radioactivity into the environment. These include the nuclear fuel reprocessing plant at Sellafield, nuclear power stations and radiopharmaceutical facilities such as that at Cardiff. The site operators are required to undertake environmental monitoring, and independent programmes are also carried out by government agencies. Monitoring also takes place around some landfill sites. More general nationwide programmes dealing with diet, milk and water are also in

operation. The most comprehensive source of monitoring data is that issued annually by a consortium consisting of the Environment Agency, the Food Standards Agency, the Scottish Environment Protection Agency and the Northern Ireland Environment and Heritage Service. The report is given the acronym RIFE (Radioactivity in Food and the Environment) and the most recent edition deals with 2006 (Environment Agency et al, 2007). This gives data for national surveys of milk, drinking water and some foodstuffs, as well as information generated by these agencies from around specific sites. Some data on discharges are also included. Further environmental monitoring data may be available from site operators or from local authorities.

These data can be used to decide whether the analysis of a particular radionuclide is warranted. As an example, monitoring data for the radiopharmaceutical facility at Cardiff include the concentrations of several radionuclides in run-off water that then enters the River Taff. However, concentrations were extremely small and the very large dilution in the river itself would mean that the corresponding values in any water abstracted further downstream would be even lower. On this basis, the expenditure of a great deal of effort on the analysis of these radionuclides in drinking water abstracted downstream would not be warranted.

4.3.2 First stage of radionuclide analysis

DWI has already advised that, in terms of gross alpha activity, isotopes of uranium and radium are most likely to account for exceedances. Unless there is specific information to the contrary these radionuclides should be determined first together with ^{210}Po . Polonium-210 is a radon decay product and so strictly should not be included in the estimation of TID. However, it is also sometimes found in water supplies (Environment Agency et al, 2007) and would contribute to the gross alpha activity. Artificial alpha emitting radionuclides such as plutonium-239 and -240 ($^{239,240}\text{Pu}$) and americium-241 (^{241}Am) generally become strongly sorbed on to soil or sediment and so would not then be expected to be found in drinking water. In the case of an incident involving the accidental or deliberate discharge of such radionuclides directly into the drinking water supply, the water companies might reasonably expect information on the radionuclides involved to come from other organisations. Incidents such as these are not dealt with specifically in this report.

Many of the radionuclides that emit beta particles also emit gamma photons. The energy of these photons characterises the radionuclide. Consequently, when the criterion on gross beta activity is exceeded, high-resolution gamma-ray spectrometry provides a powerful way of determining the presence or absence of a wide range of both natural and artificial radionuclides. Potassium-40 emits a characteristic gamma photon, and so the radionuclide most likely to account for exceedance of the criterion on gross beta can be determined very conveniently. There are other radionuclides of potential importance, notably ^{90}Sr , that do not emit gamma photons, for which radiochemical isolation would be needed.

The role of reference concentrations needs to be explored before further guidance on analytical methods can be given.

4.4 The use of reference concentrations in determining whether there is a problem

Reference concentrations can provide a very useful input to radiological assessments because they are related to a primary criterion based on dose, in this case the TID of 0.1 mSv y^{-1} . They are expressed in terms of activity concentrations in drinking water and so can be compared directly with measured values. They represent the activity concentration of a specific radionuclide that, when taken with a consumption rate of 2 l per day, would give a dose of 0.1 mSv. Actual doses from the consumption of drinking water containing these concentrations are likely to be less than the 0.1 mSv criterion (Section 2.2).

The DWI guidance reproduces the reference concentrations for a small range of radionuclides. Table 1 gives reference concentrations for an expanded range, based on those radionuclides that might be encountered in the environment in the UK. The additional radionuclides were selected on the basis of data published in RIFE for 2006 (Environment Agency et al, 2007). The additional reference concentrations have been derived in an identical manner to that used by the EC, ie they refer to an adult consuming 730 l of drinking water each year. In addition, they are based on the current dose coefficients published by ICRP (ICRP, 1996), which are identical to those in the EC Basic Safety Standards.

It must be emphasised that the presence of a radionuclide in Table 1 does **not** imply that all of the water companies must carry out an analysis for that specific radionuclide. That choice needs to be made on the basis of the available information for the catchment in question, as set out in Section 4.3.

Reference concentrations have also been derived for ^{210}Pb and ^{210}Po . These radionuclides are part of the chain of radon decay products and so strictly would be excluded from any estimation of TID. However, they are determined routinely in various monitoring programmes and may make contributions to the overall gross activities in a sample of drinking water. It was therefore considered that reference concentrations for these radionuclides would help to put the measured values in context.

The EC guidance states that if the measured concentration of any *one* radionuclide exceeds a trigger level of 20% of the reference concentration, or if the reference value for tritium of 100 Bq l^{-1} is exceeded, then the concentrations of all radionuclides should be determined. Within the system proposed here, if the criteria based on gross activity are exceeded then the next step is to identify the specific radionuclides of interest and to put the necessary analyses in hand.

Table 1 Radionuclides and Reference Concentrations for drinking water

| Radionuclide | | | Reference concentration ^{a,b} , Bq l ⁻¹ | |
|---|----------------------------|---------------------------------------|---|-------------|
| Half-life | | | 20% of reference conc | |
| ¹⁴ C | Carbon-14 | 5730 y | 240 | 48 |
| ³² P | Phosphorus-32 | 14.29 d | 57 | 11 |
| ³³ P | Phosphorus-33 | 25.4 d | 571 | 114 |
| ³⁵ S | Sulphur-35 | 87.44 d | 1054 | 211 |
| ⁶⁰ Co | Cobalt-60 | 5.27 y | 40 | 8 |
| ⁹⁰ Sr | Strontium-90 | 29.12 y | 4.9 | 1 |
| ⁹⁵ Zr | Zirconium-95 | 63.98 d | 144 | 29 |
| ⁹⁵ Nb | Niobium-95 | 35.15 d | 236 | 47 |
| ⁹⁹ Tc | Technetium-99 | 213000 y | 214 | 43 |
| ^{99m} Tc | Technetium-99m | 6.02 h | 6227 | 1245 |
| ¹⁰⁶ Ru | Ruthenium-106 | 368.2 d | 20 | 3.91 |
| ¹²⁵ Sb | Antimony-125 | 2.77 y | 125 | 24.91 |
| ¹²⁵ I | Iodine-125 | 60.14 d | 9 | 1.83 |
| ¹²⁹ I | Iodine-129 | 1.57 10 ⁷ y | 1 | 0.25 |
| ¹³¹ I | Iodine-131 | 8.04 d | 6.2 | 1.2 |
| ¹³⁴ Cs | Caesium-134 | 2.062 y | 7.2 | 1.4 |
| ¹³⁷ Cs | Caesium-137 | 30 y | 11 | 2.1 |
| ¹⁴⁴ Ce | Cerium-144 | 284.3 d | 26.34 | 5.27 |
| ²¹⁰ Pb | Lead-210 | 22.3 y | 0.20 | 0.04 |
| ²¹⁰ Bi | Bismuth-210 | 5.012 d | 105.37 | 21.07 |
| ²¹⁰ Po | Polonium-210 | 138.38 d | 0.11 | 0.02 |
| ²²⁶ Ra | Radium-226 | 1600 y | 0.5 | 0.10 |
| ²²⁸ Ra | Radium-228 | 5.75 y | 0.2 | 0.04 |
| ²³⁴ U | Uranium-234 | 244500 y | 2.8 | 0.6 |
| ²³⁸ U | Uranium-238 | 4.468 10⁹ y | 3.0 | 0.6 |
| ²²⁸ Th | Thorium-228 | 1.913 y | 0.60 | 0.12 |
| ²³⁰ Th | Thorium-230 | 7.7 10 ⁴ y | 2.80 | 0.56 |
| ²³² Th | Thorium-232 | 1.405 10 ¹⁰ y | 3.04 | 0.61 |
| ²³⁹ Pu/ ²⁴⁰ Pu | Plutonium-239 / 240 | 2.41 10⁴ y / 6537 y | 0.6 | 0.1 |
| ²⁴¹ Am | Americium-241 | 432.2 y | 0.7 | 0.1 |
| a) Reference concentration corresponds to a dose of 0.1 mSv to an adult (based on an ingestion rate of 730 l y ⁻¹). | | | | |
| b) Values in bold are for radionuclides included in the EC drinking water directive draft; data for reference concentrations are taken from that document. | | | | |

Once the activity concentrations of all of the radionuclides of interest have been determined, the overall radiological impact can be evaluated in the manner set out by the EC (see Section 2.2). If the sum of the quotients is less than 1, then there would be no need for further action. It is conceivable that, by this stage, radionuclide specific data could be available for samples collected over a period of a few weeks. The EC emphasises the importance of activity concentrations remaining elevated over considerable periods of time (Section 2.3), and so for this part of the process it would be

more appropriate to consider the mean values over the sampling period rather than the maxima observed. This could be an ongoing process as more data are accumulated.

4.4.1 The use of reference concentrations in developing an analytical strategy

One primary requirement in the development of an analytical strategy is the sensitivity of the analytical method, ie the limits of detection that are needed. The reference concentrations are related directly to the TID. Consequently, to be of use in the radiological assessment process proposed here, any detection limits would need to be much less than the corresponding reference concentration. As an illustration, if the detection limit for a given radionuclide was equal to the trigger level of 20% of the reference concentration, and the value for the detection limit was included in the summation process (Section 4.4), then this might be the largest contributor. This would then distort the true radiological situation significantly. Therefore for any radioanalytical method to be of use to the water company, the detection limit needs to be below 20% of the relevant reference concentration. This idea forms the basis for the advice given here on analytical strategy. It is difficult to be prescriptive, but a detection limit of between 5 and 10% of the reference concentration is likely to be the minimum target. If a water company is considering a contractual arrangement with an analytical laboratory, then the detection limits required must be discussed and agreed in advance.

Guidance on the selection of the most appropriate analytical techniques for the radionuclides listed in Table 1 is given in Section 5.

4.5 Action needed if the reference concentrations are exceeded

Some of the detection limits needed for the monitoring system proposed here are low in environmental terms and require considerable analytical expertise. If the measurements made on a particular sample indicate that the criterion on TID of 0.1 mSv per year might be exceeded, then the actions that should be undertaken are similar to those given earlier for exceedances in the criteria for gross activity. These actions are set out below.

- (i) Check that the sample has been taken from the supply point. If it has not, then collect a further sample from the correct location and repeat the analysis. An evaluation of the TID must relate to water from a supply point.
- (ii) Carry out checks on instrument calibration and background if these have not been done recently; examine data for samples that were analysed at the same time but which have come from other locations; examine data on any reagent blanks that have been analysed at the same time. Together, these data will provide information on whether there are any problems with the overall analytical procedure.
- (iii) If there is sufficient sample left, carry out another analysis for those radionuclides that contribute most to the exceedance of the TID. This will determine whether or not there was anything unusual about the first analysis. It

would be helpful to analyse a reagent blank alongside this repeat sample to evaluate whether any cross-contamination had taken place.

- (iv) Collect a further sample from the location of interest, ensuring that sufficient volume is collected so that repeat analyses can be carried out if necessary. For many radionuclides, it is likely that this second sample can be collected a few days after the first. However, there are some radionuclides for which the analyses can take several days or weeks. In these cases, it would be worthwhile taking several samples at intervals of a few days, so that data on temporal changes can be monitored. Depending on the results, it may not be necessary to analyse all of these samples.

This part of the process should establish whether there is an ongoing problem. If activity concentrations remain elevated, then sampling needs to continue on a regular basis with a minimum frequency of one or two samples per month. A higher frequency might be needed, depending on the activity concentrations and the resultant estimated doses. The measurement data should be kept under continual review to inform any decisions to reduce the sampling frequency. This increased monitoring effort should enable the water company to decide whether the annual dose criterion of 0.1 mSv is being exceeded.

Reasons for periodic increases in activity concentrations of some radionuclides are given in the DWI Guidance Document. Sustained increases, particularly of artificial radionuclides, should prompt a further investigation. This could initially involve a check on monitoring data from other organisations such as the Environment Agency.

4.6 Action in the event that the TID is exceeded

The EC guidance requires action to be taken if the evidence clearly indicates that an annual dose of 0.1 mSv is being exceeded. Typical consumption rates of tap water in the UK are considerably lower than the value assumed in the DWD, the annual values being 391 l, 197 l and 172 l for adults, children and infants, respectively (NRPB, 1994). For adults, actual doses from the consumption of tap water should therefore be less than those estimated using the approach adopted by the EC. For children and infants, the actual doses for some radionuclides could be higher than the corresponding values for adults as a consequence of the higher dose coefficients for these younger age groups. The differences could be up to a factor of around 4, and are dependent on the radionuclide involved (HPA-RPD, 2005). Once it becomes clear that the criterion on TID has been exceeded, the first step should therefore be to estimate the actual doses to the three different age groups, based on the annual consumption rates given here and the dose coefficients specified by the EC. This provides a more realistic view of the radiological situation, and the water companies may wish to seek specific radiological protection advice at this point.

The DWI guidance states if the criterion on TID is exceeded then medical advice should be sought. However, this level of dose is many orders of magnitude below that which would be needed to cause short term observable health effects. For comparison, the annual dose that an average person in the UK receives from all sources of natural

radiation is about 2.2 mSv (Watson et al, 2005). The principal annual dose limit for members of the public recommended by ICRP, which relates to doses arising only from controlled releases into the environment, is 1 mSv (ICRP, 1991; ICRP, 2007). The value of 1 mSv y^{-1} is set well below any level at which there is a risk to public health.

Overall therefore although action to reduce activity concentrations in drinking water should be considered it need not be implemented urgently if the criterion on TID has only been marginally exceeded. The types of action to be considered would be a matter for DWI and the water company, and would depend on the specific circumstances. Some information on the practicalities of various courses of action has been published in the context of recovery after accidents and incidents (HPA-RPD, 2005). The installation of completely new and large-scale treatment steps is expensive and it may be many years before these can be brought on-stream. This option is unlikely to be viable when dealing with a short-term breach in the TID criterion.

5 SELECTION OF APPROPRIATE ANALYTICAL TECHNIQUES FOR RADIONUCLIDE SPECIFIC ANALYSIS

This section gives some broad guidance on the selection of the most appropriate analytical techniques that would achieve detection limits that were well below the relevant reference concentrations derived by HPA-RPD (Table 1). The following objectives need to be considered when deciding on the suitability of an analytical procedure for monitoring purposes.

- Able to provide radionuclide-specific data, either via spectrometry and / or via radiochemical isolation.
- Able to achieve the required limits of detection.
- Able to achieve a high throughput of samples and provide reliable data on a timely basis.

It is not possible to provide a detailed review of analytical methods for all of the radionuclides considered in this report. However, the following sections contain some examples that illustrate how the objectives above can be evaluated.

5.1 High resolution gamma-ray spectrometry

This technique is particularly attractive if it can be applied directly to the sample of drinking water, ie without any processing of the sample at all apart from dispensing an aliquot into a suitable container. For the purposes of this evaluation, detection limits have been derived based on a one-litre sample counted for a period of 80,000 seconds, or about 22 hours. This time period means that samples would be changed on a daily basis. A counting period longer than this would mean that the throughput of samples would be much reduced, which is an important consideration when dealing with a large monitoring programme.

Typical detection limits that would be obtained at the HPA-RPD Chilton laboratory are given in Table 2. For many of the radionuclides of interest, the detection limits are a small fraction of the reference concentration, which reinforces the potential usefulness of this technique. In such cases, a suitable detection limit might be achievable with a shorter counting time, which would in turn mean that throughput of samples could be increased. It should be emphasised however that the cost of setting up a gamma-ray spectrometry facility large enough to cope with a large monitoring programme is considerable. Such a facility also needs to be maintained, and this requires professional staff. On the basis of current monitoring data for the UK, radionuclide-specific analyses are only likely to be needed infrequently, and so the purchase of this type of equipment is unlikely to be warranted for each individual water company. Possible options might be that several companies make use of a single facility, or that a specialist laboratory carries out measurements on a contract basis.

Table 2 Minimum Detectable Activities for counting samples for 80,000 seconds

| Nuclide | Minimum detectable activity ¹ , Bq |
|-------------------|---|
| ¹⁴ C | Not possible by gamma spec |
| ³² P | Not possible by gamma spec |
| ³³ P | Not possible by gamma spec |
| ³⁵ S | Not possible by gamma spec |
| ⁶⁰ Co | 0.1 |
| ⁹⁰ Sr | Not possible by gamma spec |
| ⁹⁵ Zr | 0.2 |
| ⁹⁵ Nb | 0.2 |
| ⁹⁹ Tc | Not sensible by gamma spec, very low gamma emission probability |
| ^{99m} Tc | 0.2 ² |
| ¹⁰⁶ Ru | 0.8 |
| ¹²⁵ I | 0.1 ³ |
| ¹²⁹ I | 0.2 ⁴ |
| ¹³¹ I | 0.1 |
| ¹³⁴ Cs | 0.1 |
| ¹³⁷ Cs | 0.1 |
| ¹⁴⁴ Ce | 0.5 |
| ²¹⁰ Pb | 2 ⁵ |
| ²¹⁰ Bi | Not possible by gamma spec |
| ²¹⁰ Po | Not sensible by gamma spec, very low gamma emission probability |
| ²²⁶ Ra | 0.2 ⁶ |
| ²²⁸ Ra | 0.3 ⁷ |
| ²²⁸ Th | 0.2 ⁸ |
| ²³⁰ Th | Cannot be measured directly, daughter is Ra-226 |
| ²³² Th | Cannot be measured directly, daughter is Ra-228 |
| ²³⁴ U | Not sensible by gamma spec, very low gamma emission probability ⁹ |
| ²³⁸ U | Not sensible by gamma spec, very low gamma emission probability ¹⁰ |
| ²³⁹ Pu | Not sensible by gamma spec, very low gamma emission probability |
| ²⁴⁰ Pu | Not sensible by gamma spec, very low gamma emission probability |
| ²⁴¹ Am | 0.4 |

¹ MDA is the amount we can be 95% confident of detection; except where stated the detection limits are based on counting a one-litre sample in a re-entrant container configuration. i

² MDA assumes counting made very soon after collection as half-life is only 6 hours

³ Approximate MDA assumes count in a Pot configuration (sample volume 100 mls) in a well shielded area with a low energy detector

⁴ Approximate MDA assumes count in a Pot configuration (sample volume 100 mls) in a well shielded area with a low energy detector

⁵ Approximate MDA assumes count in a Pot configuration (sample volume 100 mls) in a well shielded area with a low energy detector

⁶ By counting Pb-214 or Bi-214, assumes equilibrium through Rn-222

⁷ By counting Ac-228, assumes equilibrium

⁸ By counting Pb-212/Bi-212

⁹ If uranium is a natural source U-234 can be determined by counting U-235 (MDA 0.1 Bq) and assuming natural isotopic ratios

¹⁰ If uranium is a natural source U-238 can be determined by counting U-235 (MDA 0.1 Bq) and assuming natural isotopic ratios

Some of the radionuclides of interest emit characteristic photons that are of low energy. These are noted in Table 2. In such cases, the more commonly-used gamma-ray

spectrometry equipment would not produce valid data. Instead, measurements would need to be made in a well-shielded, low-background area using a low-energy detector. Detection limits have been derived for a sample volume of 100 cm³. These have not been derived from actual measurements at HPA-RPD and the associated uncertainties are considerable. However, they are considered adequate for the purposes of this evaluation. In these cases, the detection limits are not sufficiently low to be of use in assessing the radiological situation, and consideration needs to be given to ways in which these radionuclides can be concentrated or isolated for a more sensitive measurement.

The original volume of sample needed to produce useful measurements can be estimated roughly using the data in Tables 1 and 2. As an example, for ¹²⁵I, the reference concentration is 9 Bq l⁻¹ (Table 1), and so to produce meaningful data a detection limit of around 0.5 Bq l⁻¹ would be needed (Section 4.4.1). From Table 2, the instrument detection limit is about 0.1 Bq in a volume of 100 cm³, and so the volume of original sample needed would be small, less than 0.5 l. Applying the same approach to ²¹⁰Pb, around 200 l would be needed, a consequence of the very low reference concentration (Table 1). Such a volume would be difficult to handle, especially if a large number of samples required analysis, and a completely different analytical strategy would be preferable (Section 5.2).

Radium-226 deserves special mention, because in many cases in the UK it is expected to be an important contributor to the gross alpha activity. With some concentration of the sample, a workable detection limit can be achieved using gamma-ray spectrometry to determine the activity of the decay products ²¹⁴Pb or ²¹⁴Bi. This approach is based on the assumption that ²²⁶Ra is in equilibrium with its decay products. One of these is radon gas, ²²²Rn. Consequently, after any concentration step each sample has to be stored in a suitably sealed container so that no radon is lost and equilibrium can be established. This means that no monitoring data would be available until several weeks after the samples have been sealed. An alternative approach would be to expel the radon gas, trap it and count the alpha particle emissions – the so-called radon emanation procedure. However, this is a complicated procedure involving low-temperature vacuum distillation, and there is still a delay between sampling and the production of data. It is unlikely to be of general use to the water industry.

For involatile radionuclides, evaporation of the sample is a simple procedure. Basic equipment can be set up that will reduce around 10 litres of water to a volume suitable for gamma-ray spectrometry in a few hours. Evaporation might therefore provide a practical option for monitoring purposes provided that enough samples can be processed simultaneously. This would not however be suitable for isotopes of iodine. In these cases, evaporation would need to follow the method recommended for the determination of gross beta activity by the Standing Committee of Analysts (SCA) (SCA, 1985 – 1986). Alternatively, a straightforward concentration procedure involving precipitation or ion exchange might be more appropriate (Wilkins et al, 1982).

Potassium-40 has not been included in Tables 1 or 2 because it is excluded from the estimation of TID. However, it is likely that this radionuclide will be an important contributor to the gross beta activity (Section 4.1). For a direct count of 80,000 seconds and a 1 l sample, the detection limit estimated by HPA-RPD would be about 1.5 Bq l⁻¹.

This is a relatively high detection limit compared with some of the other radionuclides of interest. This is because the gamma-ray emission for this radionuclide is close to a peak in the background. The implications are therefore that some concentration of the sample may be needed if the criterion on gross beta activity is exceeded.

5.2 Beta counting

The emission of beta particles does not occur at a discrete energy. Instead, there is a characteristic maximum energy but emissions occur over a continuum of energies below that maximum value. In general, this means that radionuclides need to be isolated using chemical separation procedures before the activity can be measured.

Tritium (^3H) has a very low maximum energy and is normally measured using liquid scintillation counting. A method for the determination of tritium in water has been issued by SCA (SCA 2005). With this technique, the amount of sample that can be used is relatively small, not more than 10 cm^3 , but with the sensitive equipment currently available detection limits of around 10 Bq l^{-1} can be achieved with counting times of a few hours. Liquid scintillation counters are able to distinguish between tritium with its low energy beta emission and radionuclides such as ^{14}C for which the energy of the beta particles emitted is much higher. These two radionuclides could therefore be determined in the same sample. The detection limit for ^{14}C is comparable with that for tritium, and so is well below the 20% trigger level given in Table 1. Liquid scintillation counting could therefore give useful information with the minimum of sample preparation. There would however need to be evidence to demonstrate that other radionuclides with beta emissions similar in energy to ^{14}C were absent.

Two isotopes of phosphorus, ^{32}P and ^{33}P , are included in Table 1. This is because they are used in the production of radiopharmaceuticals. Both have short radioactive half-lives, and can be measured using liquid scintillation counting for ^{33}P or, for ^{32}P , a similar technique known as Cerenkov counting that makes use of the same counting equipment. Both analyses require some complex chemical isolation procedures. Unless a very clear need for such analyses has been demonstrated, these procedures are unlikely to be amenable for use in a water company laboratory.

Gas-flow proportional counters provide an alternative means of measuring beta activity, and these can achieve much lower limits of detection than liquid scintillation counters. They are generally the preferred option when determining a radionuclide such as ^{90}Sr . In this particular case, the 20% trigger value is relatively small (Table 1). To obtain the necessary limits of detection the analysis would require the chemical isolation of ^{90}Sr , followed by a period of storage to allow the ingrowth of the decay product ^{90}Y . Consequently, monitoring data would not be available until some weeks after the analysis began.

As noted in Section 4.3.2.1, the determination of ^{210}Pb requires radiochemical separation. Methods for the analysis of this radionuclide have been reviewed (Clayton and Bradley, 1994). A method based on the ingrowth of the decay product ^{210}Bi followed by beta counting might be a possibility, but the chemical isolation procedure is complex and data would not be available for about 4 weeks. A more straightforward

method involves the ingrowth of ^{210}Po followed by alpha spectrometry (Section 5.3), but again data would not be available for about 4 weeks. This approach is much more sensitive than beta counting, and would be the preferred option.

Both liquid scintillation counters and gas flow proportional counters are expensive to buy and require specialist staff to maintain and use them. In addition, the chemical procedures needed to isolate a particular radionuclide are often complex, again requiring specialist staff and laboratory space and equipment. It may not always be viable for a water company to undertake such work in house. Before embarking on radionuclide-specific analyses, the need for such work should be firmly established and specialist advice sought on the most effective way of generating monitoring data. The potential for using commercial contract analytical facilities should be explored.

5.3 Alpha counting

Alpha particles are emitted with discrete energies, and these can be used to identify a particular radionuclide. However, in contrast to gamma-ray emitting radionuclides the range of energies is small, and in many cases the elements of interest need to be isolated chemically before a source is prepared for counting. In addition, alpha particles are readily attenuated. It is essential therefore that a very thin source is prepared, otherwise the peak in the alpha spectrum will be broad, which in turn may make the interpretation of the data difficult. The general approach is to isolate each element using techniques such as ion exchange or solvent extraction, and then to prepare a source by electrodeposition or spontaneous deposition on to a suitable metal disc. A suitable tracer is used so that the chemical recovery can be estimated.

An alpha spectrometry system is expensive to purchase and requires specialist staff to operate it. The chemical isolation procedures also require specialist staff and laboratory facilities even with drinking water, which is a relatively simple material to handle analytically. As in previous cases, unless there was a clear need for the ongoing analysis of a large number of samples, it is unlikely that an individual water company would undertake the analysis of alpha-emitting radionuclides in house.

5.4 Non-radiometric methods

Inductively-coupled plasma-mass spectrometry (ICP-MS) is an attractive option for the monitoring of drinking water because both stable elements and some radionuclides can be determined simultaneously. This technique depends on the mass of the element of interest. Consequently, in terms of the limits of detection that can be achieved ICP-MS tends to be an effective option for those radionuclides with long half-lives, ie those that produce a small number of radioactive decays per unit mass of the isotope. For the monitoring of drinking water, uranium would be an element of particular interest because of its likely contribution to the gross alpha activity.

The capital cost of ICP-MS equipment has decreased in recent years. The viability of a water company having an ICP-MS facility is enhanced because of the potential to carry out a wide range of analyses, not just radionuclides.

6 FINAL REMARKS

A process has been developed that guides the water companies through the various stages of investigation following an exceedance of a criterion on gross activity. The process conforms with the guidance issued by the EC in its Drinking Water Directive and draws on guidance issued subsequently by DWI. The DWI guidance necessarily covers a wide range of potential pollutants, and as a consequence the material on any given topic can become fragmented. DWI might wish to consider using this report as a basis for an annex dealing specifically with radionuclides in drinking water.

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