Department of the Environment, Transport and the Regions Commissioned research for Radioactive Substances Division

Report Title

Natural Radioactivity in Private Water Supplies in Devon

DETR Report No: DETR/RAS/00.010

Contract Title: Assessment of hazard from radon from private water supplies

DETR Reference: RW5/2/299 (EPG1/4/51)

Sector: Band C

Contractor's Reference: E60V

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Date approved by DETR: Nov 2000

Period covered by report:

Abstract (100-200 words as desired):

This report details a study of the occurrence of natural radioactivity in private water supplies in West Devon. Supplies sourced from wells, springs boreholes and a small number surface supplies were sampled. The findings of a laboratory simulation of the radon content in drinks such as tea, coffee and squash are also presented.

Of supplies sampled in phase one of the work approximately 8% of tap water and 9% of samples directly from the supply contained radon at concentrations exceeding the draft European Union Commission Recommendation action level of IOOOBq/1 for individual and public water supplies. In a small number of supplies ^{238}U is present at levels exceeding $2\mu g/l$, the World Health Organisation (WHO) provisional guideline value for uranium in drinking water.

The final aspect of the study looked at seasonal variation in the radon content of selected supplies. This showed considerable variability in radon concentration over the course of a week and between studies carried out several months apart.

Keywords (5 Maximum): Radon, drinking water, ground water, uranium, Devon

The results of this work will be used for the formulation of Government policy, but views expressed in this report do not necessarily represent Government policy.

BRITISH GEOLOGICAL SURVEY

Natural radioactivity in Private Water Supplies in Devon.

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Bibliographic reference

D K Talbot, J R Davis and M P Rainey, 2000. Natural radioactivity in Private Water Supplies in Devon. Department of the Environment, Transport and the Regions Report Number DETRJRAS/OO/OIO

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1) INTRODUCTION

Between 1986 and 1996 the Department of Environment (DOE) commissioned annual radiological assessments on various private water supplies to households in England and Wales. Each study examined samples from approximately 100 different households in a selected area for a range of natural and artificial radionuclides and, out of the samples studied, about 20% were also analysed for ²²²Rn. In each study, committed effective dose equivalents (CEDEs) were calculated for the observed radionuclides for hypothetical critical groups of householders based on the consumption of a year's supply of drinking water. Three groups of consumers were studied - infants $($ \leq 1 year old), children (1-10 year old) and adults. The most recent dose conversion factors (ie. Dose per unit uptake factors) were used. Some of the results obtained for areas studied in 1990 (e.g. Kerrier and Restormel districts in Cornwall), exceed the World Health Organisation's (WHO's) guideline activity concentration value for radioactivity in drinking water. The WHO recommend a reference level of committed effective dose of 0.1mSv (for adults) for one year's consumption of drinking water (based on the consumption of two litres of drinking water per day). Although the WHO guideline does not differentiate between natural and man-made radionuclides, the CEDEs calculated for the areas in Cornwall were predominantly due to the naturally occurring radionuclide ²²²Rn and, to a lesser extent, ²¹⁰Po. The WHO recognise the difficulty in applying its guideline to ²²²Rn

An area of West Devon around Tavistock was selected by the Department of the Environment, Transport and the Regions (DETR) for further, more focused, research. The aim of the research, as identified in the project specification, is to define:

- the concentration of 222 Rn in 100 private water supplies from West Devon;
- temporal and spatial variations (within the household supply and distribution system) of 222 Rn and 210 Po content in the three water supplies selected from the detailed study;
- the dose received by members of the household taking into account their actual drinking water consumption patterns rather than by assuming the 2 litres per day estimate given by WHO.

The DETR awarded the British Geological Survey (BGS) a contract, reference number RW 5/2/299, EPG 1/4/51, to carry out this work. As the first phase of the project the BGS arranged for 128 private supplies in West Devon to be sampled and analysed for 222Rn , 226Ra , 238U and 232Th . Three supplies with very high radon concentrations have also been sampled for 210 Po. Although the original work programme envisaged sampling only springs, wells and boreholes, discussions with West Devon Borough Council (WDBC) revealed a number of other private water supply types in the district. With the agreement of the DETR, a small number of these supplies, including river/stream extractions, mine drainage from adits and open, artificial waterways (known in the area as leats) were included in the sampling programme. Under subcontract to BGS WDBC staff liaised with local householders to obtain the samples.

2) SAMPLING PROCEDURES

2.1 Sampling for 222 Rn and 226 Ra.

Although radon analysis by liquid scintillation counting (LSC) only requires 10ml of sample experience has shown that such small samples can be difficult to take in the field. Better results are obtained by taking 200 ml samples into glass bottles in the field and then sub-sampling 10ml aliquots in the laboratory. The 200ml sample size allows for duplicates and aliquots to be taken for the subsequent analysis of 226 Ra. Each 200 ml sample bottle was carefully pre-rinsed with the sample, filled and capped. Excessive disturbance and agitation of the sample was avoided during sampling to prevent the loss of 222 Rn.

After analysis of ²²²Rn the vials containing the mixed water samples and LSC cocktails are stored for a period of time sufficiently long to allow the 222 Rn unsupported by an equivalent activity of its parent isotope, 226 Ra, to decay to levels below the detection limit. 226 Ra in the vials can then be determined by analysing the residual concentration of 222 Rn, since the activity of the two radionuclides will now be in equilibrium.

2.2) Sampling for 2^{10} Po.

Previous work has shown glass bottles to be unsuitable for the storage of samples awaiting analysis for 210 Po (Flynn, 1968). An approximate 2 litre sample was, therefore, collected from each specified location into fresh 2 litre polyethylene terphalate (PET) sample bottles. Each sample was acidified with 5mls of concentrated Analar grade hydrochloric acid to prevent sorption of the $^{210}P_o$ onto the sample bottle. On receipt in the radiochemical laboratories each sample was split and spiked with ²⁰⁹Po to monitor recovery of 210 Po during the subsequent pre-concentration and plating process..

2.3) Sampling Uranium and Thorium.

One 30ml sample of water was collected at each sampling location in a fresh, pre-rinsed, 30ml Nalgene(TM) bottle. Experience has demonstrated that the typical blank obtained from such sample vessels is less than 0.05 µg/l U and Th. On return to BGS, and prior to analysis, each sample was acidified by the addition of 3 ml of concentrated Aristar nitric acid, recapped, agitated and left to stand for one week prior to analysis to allow for the re-solution of any sorbed Uranium or Thorium.

3) ANALYTICAL PROCEDURES

3.1) 222 Rn and 226 Ra.

The concentration of radon in water was determined using alpha particle liquid scintillation counting (LSC) (Talbot, 1994). A 10ml aliquot of water is placed in a low background glass LSC vial which already contains 10ml of a toluene-based LSC cocktail. Immediately prior to analysis the vial is shaken to extract the radon into the organic layer. The sample is counted on an LBK Wallac Rackbeta liquid scintillation counter using pulse shape analysis to differentiate alpha and beta scintillation events. Duplicates, standards, and blank samples are included in each batch. At an activity of 1 Bq/l analytical error (2σ) is typically of the order of $5 - 10$ %, for a 1 hour count time, though this will vary slightly depending on physical characteristics of the sample such as salinity and discolouration.

A ²²⁶Ra standard is used since its long half life (1602 years) means it provides a stable source of ²²²Rn within the time frame of this project. This solution has been compared to a certified standard solution at Central Mining Institute, Poland and to a certified standard solution in the Water Authority of Jordan (Certificate No. 425-56-3, Isotope Products Laboratory, Burbank, California, USA). Once the samples from the initial screening survey had been analysed a standard 226 Ra solution of an activity reflecting that found in the screening programme was ordered from AEA Technology QSA. Work has also been undertaken outside this project to verify the uncertified standard and provide more direct traceability to an internationally recognised standard.

3.2) 210 Po

The likelihood of any interfering ions in a potable water supply are low, as compared to a biological or mineralogical sample, therefore a simple pre-concentration by evaporation was used. This evaporation step both cuts down on solvent use and generation of solvent vapour waste streams as used in traditional methods (Shannon and Orren, 1970), and on expense in use of state of the art resins (Vadja et al., 1995), for a small loss in recovery.

After acidifying the approximately 2 litres of collected sample, the sample was split. A spike of 0.16Bq ²⁰⁹Po was added to each 800ml water split. Each sample was slowly evaporated down to about 30ml. In sequence, 2ml of Analar grade hydrochloric acid, 5ml of 2M hydroxylamine hydrochloride (w/v) and 2ml of 25% sodium citrate, were added. The pH was adjusted to 2 with ammonia and the whole diluted to 50ml. ²¹⁰Po and the ²⁰⁹Po spike were then chemically plated out onto a silver disc, as described by Flyn (1968). Samples were counted by alpha spectroscopy which allowed the quantitative determination of both the 210 Po and 209 Po (spike). Within each batch of ten samples, two were determined in duplicate and two blanks were measured.

 Typical background measurements made using the BGS alpha spectroscopy system yielded count rates of less than 1 count per day. This gave a theoretical detection limit of <0.0001 Bq/l (2σ). Extraction and plating efficiencies obtained during ²¹⁰Po analysis by this method were typically $> 85\%$, about 7% less than the other methods outlined above. Counting efficiencies when monitored against a range of standard alpha emitters (Amersham, NBS/NAMAS certified mixed alpha source, 0020RN) were 20-22%. With such a low background, precision was directly dependent upon counting time, for an activity of 0.01 Bq/l a precision of 6% (2σ) was achieved by counting for 4 days.

3.3) Uranium and Thorium.

Uranium and Thorium can be quantitatively determined at concentrations between 1000 and 0.01 µg/l by ICP-MS using a VG Plasma quad PQ2 plus. In terms of radioactivity $1\mu\text{g}/\text{l}$ corresponds to approximately 0.01Bq/l. This technique does not provide isotopic information on the Uranium and Thorium analysed. The accuracy of Uranium and Thorium determinations was validated through the analysis of an international standard basalt (BCR1) as U and Th do not constitute part of the Aquacheck scheme to which BGS subscribes. Standardisation of the ICP-MS was achieved through certified commercially available multielement solutions (Spex Industries). During analysis, each batch of 50 samples contained 5 blanks, 5 duplicates and 5 internal QC solutions.

4) FIELD STUDY

4.1 Radon and Radium Screening Study

Arrangements were made for staff of the Environmental Health Department of WDBC to sample private water supplies at 128 properties in the Borough. It proved impossible to sample a number of sealed sources directly and a few samples were delayed or damaged in transit. This resulted in a final total of 105 samples from the sources and 116 tap water samples being analysed for radon. In addition the 114 samples of tap water were analysed for uranium and thorium as two samples were damaged in transit.

Details of the number and age of occupants of the house and of the supply system were also recorded. Although the normal situation in this area is for a private supply to provide water for single properties, four of the sources sampled supply water to several homes. Full details are provided in Appendix 1.

Care was taken to avoid sampling waters that were obviously highly turbid as these would not normally be consumed by the householder. Where there were obvious large particulates (such as orange iron precipitates) the sample was carefully decanted into the sampling vessel to avoid the transference of such material, which again would not normally be consumed by the householder.

The distribution of sample sites and their relationship to geology is illustrated in Figure 1. For clarity only those geological formations which contain samples are included in the key. A variety of types of groundwater source are in use in the study area. In addition to the more common source types of wells springs and boreholes a number of leats, river extractions and mine adits were sampled.

The full analysis results are given in Appendix 2. Statistical summaries, analysed by source type and underlying geology, are given in Tables 1 to 4. Geology based on the BGS 1:250 000 scale mapping has been assigned to sample points using a GIS. Although only 105 properties have both source and tap analysis, all available results are included in the tables, giving 105 analyses from the source and 116 analyses of tap water. Where duplicate analyses are available for a sample the mean of the two determinations has been used in the production of the statistical summaries.

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Figure 1. Simplified geological map showing sample locations. Based on BGS 1:250000 digital cartography. Black diamonds represent sampling sites.

Simplified 1:250 000 Scale Geology of West Devon

- CRACKINGTON FORMATION
- LOWER CARBONIFEROUS ROCKS [argillaceous rocks and chert]
- UPPER DEVONIAN AND LOWER CARBONIFEROUS ROCKS [UNDIFFERENTIATED]
- SILURIAN IGNEOUS ROCKS
- BASIC INTRUSIVE IGNEOUS ROCKS
- DARTMOOR GRANITE

Table 1. Radon in tap water (Bq/l) by source type.

Table 2. Radon in tap water (Bq/l) by geology

Table 3. Radon at source (Bq/l) by source type

Table 4. Radon at source (Bq/l) by geology

 222 Rn was found to be above the draft European Union Commission Recommendation action level of 1000Bq/l for individual and public water supplies in 8% of the tap water samples and 9% of the source water samples analysed. In terms of the geology at the source only the Upper Devonian etc and Granite rocks yielded tap waters over 1000 Bq/l. Dartmoor Granite produced the highest average radon levels of over 700 Bq/l with the single highest value in the whole screening study resulting from a borehole source in this formation and the other highest values found in springs and wells also coming from these rocks. The few examples of surface water supplies sampled during the study, such as those drawn from streams and leats, all show low radon concentrations (<35 Bq/l).

For water from groundwater sources, mean values (by source type) at the tap are consistently lower than those at the source. This is consistent with loss of radon due to degassing as a result of water turbulence within the supply system and natural radioactive decay while the water is resident in the household supply system. Appendix 2 shows that in the majority of individual cases 222 Rn concentrations in tap water for a particular supply are lower than, or similar to that at the source. In a number of individual instances water from the tap contains higher levels of 222 Rn than the source. This may indicate that there is considerable short term variability in 222 Rn concentrations in the supply or may relate to degassing, and so loss of radon, occurring while sampling the source. There are also reported instances of radium building up in pipe scale, which could then release radon into the water flowing through them (Field et al, 1995). Further work would be necessary to determine if this is a factor in the case of these supplies.

The samples collected during the screening study were also analysed for 226 Ra. In all cases no 226 Ra was detected above the detection limit of approximately 0.1 Bq/l

4.2 Seasonal Variations in Radon

Three sites were selected for a more detailed study, designed to allow the short term and seasonal variation in radon at the source to be assessed. One of these sites had a borehole supply, one a well and one a spring. The borehole site gave the highest radon concentration found in the screening study. The spring site had a radon concentration close to the mean value for all sites studied. The occupants of well site included an infant less than 1 year old, it was also the only property with an infant occupant where radon in tap water exceeded 200 Bq/l. Figures 2 - 5 show the variability in radon at the sites of the detailed study.

During the summer study samples were taken morning and afternoon at the source, tap and in the case of the spring supply from the storage tank on the system. For the winter study a single sample was collected from the source in the morning and evening.

Figure 2, Well summer study

Figure 3, Spring summer study.

Figure 4, Borehole summer study

Figure 5, Winter Study (source sampling)

Figures $2 - 5$ show there to be no constant change in radon between morning and evening or evening to following morning, error bars indicate the mean sampling errors determined in the screening study. The borehole site consistently has higher radon at the tap than at the source. This observation, coupled with the short distribution system at this property indicates that radon is lost due to degassing at the borehole sampling point, or gained from radium bearing pipe scale, rather than indicating a large, fast variation in radon in the water An increase in radon is apparent at all three sites in the summer study (Figs2-4) around the 27th of August, this coincides with a change in weather from showers, prevalent during the first part of the sampling period, to dry weather for the second part of the week.

It appears that the tap and source samples from spring site on the afternoon of $27th$ August have been mislabelled by the sampling subcontractor, all checks on the samples and sample cards show the situation to be as shown in Figure 3, however the results are consistent with a switch between the samples.

	Summer		Winter			
Source type	Samples	Mean	RSD(%)	Samples	Mean	RSD (%)
Borehole	14	5520	35.8	14	3080	13.2
Spring	14	661	34.9	14	509	10.3
Well	14	644	52.4	14	1070	15.2

Table 5. Summary of summer and winter radon measurements at source (Bq/l)

All the sources sampled showed large variability in radon concentration over the summer sampling period. Less pronounced variability was observed during the winter sampling. Maximum values were observed during the summer sampling and winter values were generally within the range of those found during the summer. Although no clear trends are visible in this data a more intensive and longer term sampling programme would be needed to reveal any systematic variability.

4.3 Uranium and Thorium

The full analysis results for uranium and thorium analyses are given in Appendix 2. Statistical summaries, analysed by source type and underlying geology, are given in Tables 6 to 9. Where duplicate analyses are available for a sample the mean of the two determinations has been used in the production of the statistical summaries.

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Table 6. Uranium in tap water (μg/l) by source type.

Table 7. Uranium in tap water (μg /l) by geology.

Table 8. Thorium in tap water $(\mu g/l)$ by source type.

Table 9. Thorium in tap water (μg /l) by geology.

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4.4 Polonium Analysis

Single samples for polonium analysis were collected during the summer detailed study from the well and spring sites found to have the highest radon concentrations in the screening study. A series of samples were taken from the borehole site, at the tap and the source during the summer and at the source during the winter. Analysis results are shown in Figure 6. The analysis method has a lower limit of detection of 0.01 Bq/l.

Table 10. 210 Po. (Bq/l)

Figure 6. Summer polonium analysis, Borehole site

Polonium concentrations were found to be markedly lower during the winter study than the summer study for reasons which are not apparent in this study.

5) CALIBRATION AND ERROR ANALYSES.

5.1) Activity calibration

Throughout the 9 radon analysis runs undertaken for this stage of the project, standard radium solutions with a nominal activity of 200 Bq/l were counted. The mean decay corrected count rate is shown in Figure 8. The overall mean standard count rate was 269.7cps corresponding to an activity calibration of 1.35cps per Bq/l. Long term monitoring of the standard has yielded an overall mean count rate of 270.1 +/- 6.3 counts per second.

Analysis of the BGS standard against a traceable international standard has shown its value to be correct to within analytical errors.

5.2) Duplicate samples

Throughout the screening programme duplicate samples were collected at approximately 15% of sites. Duplicate analyses for ²²²Rn show a sampling variability between samples ranging from 0.5 to 11%, with all but four of the pairs displaying a difference of less than 4.4% (Table 11). Analytical errors for the individual samples range from 1.1% to 5.6%. Analysis for ²³⁸U of nine sets of duplicate samples shows a sampling error between 2.4% and 11% (Table12). For 232 Th, all of the duplicate samples have activities lower than the detection level.

Figure 9 shows a comparison between duplicate analyses results and the theoretical 1:1 match assuming no sampling or analytical errors for radon. Error bars on the individual data points represent the 3σ analytical error. Figure 10 shows the comparison between duplicates for uranium.

Figure 9. Duplicate analysis $(^{222}Rn, Bq/l)$.

	% Error		
Duplicate	Between A and B	Analytical A	Analytical B
1	10.7	4.0	5.0
\overline{c}	8.0	5.5	5.4
3	2.9	5.4	5.6
4	0.5	3.1	3.4
5	0.8	4.3	4.8
6	3.7	2.2	2.3
7	6.1	2.3	2.1
8	7.1	3.2	$\overline{3.5}$
9	4.3	1.4	1.5
10	3.3	1.7	1.9
11	2.1	1.6	1.8
12	3.4	1.2	1.3
13	2.1	1.1	1.1
14	2.2	1.2	1.2
15	3.4	1.2	1.1
16	1.3	1.1	1.1

Table 11. Percentage errors in duplicate analysis of radon samples

	% ERROR BETWEEN DUPLICATES		
Duplicate	Uranium	Thorium	
	\ast	\ast	
$\overline{2}$	5.7	\ast	
3	3.8	\ast	
$\overline{4}$	9.5	\ast	
5	4.6	\ast	
6	6.8	\ast	
7	4.1	\ast	
8	2.4	\ast	
$\mathbf Q$	10.7	\ast	

Table 12. Percentage errors in duplicate analysis of uranium and thorium sample.

* Measurements of one or both duplicate samples are below detection levels.

Figure 10. Duplicate analysis $(^{238}U, \mu g/l)$

Uranium

6) Reduction due to drink preparation study.

6.1 Rationale to study

Studies on the inhalation of radon are well documented given that inhalation accounts for 89% of 'risk' arising from radon while ingestion accounts for only 11% (USEPA, 1999). The aim of this part of the project was to further refine the understanding of radionuclide uptake with a brief study to determine the quantity of 222 Rn lost during the preparation of various beverages and storage of water within household containers.

The proportion of the original concentration of radon released from water is dependent on (Becker and Lachajcyk, 1984):

- Surface Area
- Duration of air : water contact
- **Agitation**
- Temperature

A radium standard was purchased to use in creating a synthetic radon containing water. The solution was diluted, maintaining acidity, to a level were the radon generated from the radium would be at a concentration similar to the levels found in the preliminary sampling exercise. A bulk sample of radonbearing groundwater unsupported by radium was also used. This was needed since a radium-supported standard cannot be used for any experiments which involved storage of the 'drink' as any radon lost due to process will rapidly be replaced from radium. The radon-bearing groundwater came from a source, of negligible radium content, sampled for previous work at the BGS, and a repeat analysis of the sample collected for this work confirmed radium concentration to be below the lower limit of detection.

The study consists of sub-sampling of water, spiked with 222 Rn, and calculating simple loss as a result of combinations of filling a kettle, boiling a kettle, pouring, stirring and storage associated with the making of both hot and cold drinks. Whilst most previous research on the risks of radon have centred mostly on inhalation problems and degassing during showering, studies relevant to the current one have concluded;

- Filling a kettle from a tapped source results in 0-50% loss, with USEPA 1991 stating 20% as a good average;
- Normal exchange to air very low $(-0.15\%$ per minute from a still sample), this rate increased by up to 20 times that if the sample was agitated, (Gesell and Pritchard 1980);
- Ratio of air to water exchange has been found to double from 10° C to 30° C, and double again from 30°C to 40°C over long term (Hess, 1987). This study was performed using a water bath and sampling under a layer of mineral oil to minimise Radon loss during sampling;

• In well experiments Tedesco et al (1996) found an order of magnitude difference in radon levels, using etched track detectors, in well bores at a temperature of 44-60°C c.f. well bores with a temperature range of 20-27°C.

The experimental outline is described in Appendix 3.

6.2 Results

The data from the experiments was used to calculate the 'removal factors', ρ , used in dose calculations, as described in section 7 of this report. The experimental results can be summarised as follows:

- Over the series of experiments loss on pouring ranged from 4% to 25%. The mean was 10.6% and the standard deviation 7.6%. These numbers are similar to those reported in previous studies (USEPA 1991)
- Over the series of experiments loss on reaching boiling for a few moments (as occurs in modern automatic electric kettles) from the ²²²Rn left after pouring ranged from 63-83%. The mean was 73.4% with a standard deviation of 9.7%. Most literature values predict total, or near total, loss on boiling.
- 8 hour storage of an unsealed water sample resulted in a 60% loss of radon whether refrigerated or not.
- 24-hour storage of an unsealed sample resulted in total loss from the non-refrigerated samples and 95% loss in the refrigerated samples. The rate of loss is similar to that reported by Gessel and Pritchard (1980).
- Samples which were boiled, and then sealed and allowed to cool before storing, were used to simulate the content of bottles used for bottle fed infants. After boiling only 25% of the remaining 222 Rn is lost during this cooled and sealed storage.
- After boiling no significant losses occurred on pouring and stirring.

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7) Estimation of adjusted Committed Effective Dose Equivalents

The IAEA Rasanet web-site (Radiation and Waste Safety Division, 1998) outlines the rationale for the use of CEDE. A measure of the total risk of specified somatic and hereditary effects to an average individual and progeny from an intake of a radioactive material, including the risk from irradiation in the subsequent 50 years resulting from the intake is given by the committed effective dose equivalent (CEDE), H_{50} . Values are calculated taking into account a number of factors such as gut and blood transfer parameters and chemical form of the ingested radionuclide.

7.1 Calculation and Rationale of dose parameters

The values for CEDE in this study were obtained by multiplying the dose equivalents resulting from ingestion only, by appropriate weighting factors and summing. When calculated for a unit intake of radioactivity i.e. 1 Bq, the result is a dose conversion factor (DCF) f in Sv/Bq.

 The values used for (DCF)f [in this study are taken from the IAEA Rasanet web-site (Radiation and Waste Safety Division, 1998) http://www.iaea.org/ns/rasanet/information/doselim.htm. The parameters for Committed Effective Dose per Unit Intake via Ingestion in (Sv/Bq) for members of the public were used as recommended by the NRPB 1999. Many literature values exist for Radon (DCF)f by ingestion and some of these are shown in Table 14, with the value used in the study of $1x 10^{-8}$ being a good approximation.

Table 13. Dose Conversion Factors used in the current study.

A transfer parameter, termed P(i)09, relating the dose to humans from intake of drinking water is calculated as:

$$
P(i)_{0} = \rho k''_{W} I_{W} (DCF)_{f}
$$

where (DCF) $f =$ dose conversion factor for intake by ingestion (Sv/Bq),

 $I_{\rm w}$ = intake of drinking water (l/a),

 k''_w = fraction of the intake of drinking water arising from the contaminated source,

 ρ = removal factor to account for a process, such as sedimentation and removal of radio-nuclides by water treatment processes.

The default value for k''_W is 1.

The value of ρ is site specific and in this case will be assumed to be unity.

The value of I_w intake of water is taken from the survey.

A number of assumptions are made in calculating CEDEs:

-The same dose conversion factor for infant and child groups was used

-In the absence of a definitive literature value, the same dose conversion factor is used for all age groups with regard to 222 Rn, though the effective dose is expected to be higher per unit intake for infants and young children (Crowford-Brown, 1987). Recent literature values for dose from ingested 222 Rn include:

The newly released NRPB (2000) booklet on Health Risks on Radon also contains a brief appendices on non-lung cancers, gives some dose equivalent details, and stresses the substantial uncertainties in all the estimations of dose.

 The drinks survey undertaken by BGS included collection of data on consumption rates. Most literature CEDE values work on an assumption that an adult drinks 0.5l/day, with some values as high as 1l. The survey shows this value can be higher and varies by season. All members of the households involved in the detailed study were asked to complete a questionnaire detailing their water consumption during the study periods. It is worth noting that the water consumption reported for Infant 1 is exceptionally high. This data can then be used to calculate the CEDE arising due to water consumption Table 15 summarises these surveys. Although one aspect of the lab. experiments simulated the storage of water before making drinks all those completing the questionnaire reported that water was always drawn fresh from the tap prior to preparing a drink.

Table 15. Observed water consumption patterns (l/day), based on householders completion of a questionnaire

	Summer			Winter		
	Boiled	Tap	Total	Boiled	Tap	Total
Child 1	0.5	0.25	0.75	0.25		0.25
Child 2	0	0.3	0.30	$\boldsymbol{0}$	0.3	0.3
Infant 1	0.564	0.564	1.13	0.564	0.564	1.128
Infant 2	0.5		0.50	0.4		0.4
Adult 1	0.75		1.75	0.75	0.75	1.50
Adult 2	0.75	1.41	2.16	0.75	0.94	1.70
Adult3	1.25	0.25	1.50	1.50	0.00	1.50
Adult4	1.43	0.25	1.68	1.43	0.00	1.43
Adult5	2.33	0.53	2.86	2.13	0.28	2.41
Adult6	0.25	1.25	1.50	0.25	1.11	1.36

7.2 Doses derived from field study data.

The calculation of potential internal ionising radiation doses received has been calculated for each sample, and these are shown in Appendix 4. The average value for the whole study area was 1.10 mSv. The average contribution of each study radionuclide is shown in Table 16 as well as its maximum and minimum contribution. Radionuclide contributions in this table are based on analytical data from the screening samples

Table 16. Average contribution to annual dose by individual radionuclide.

The vast majority of dose from consumption of water comes from 222 Rn, with significant contribution from 210 Po. The higher the overall potential CEDE, the greater the contribution of total dose is from radon.

7.3 Incorporation of reductive factors derived from drink making study

Table 17 gives the average water consumption patterns of those surveyed in the detailed study. The adults involved in this study consumed noticeably more tap water than the 1.138 l/day suggested to be the national average by the Environment Agency (1998),

Table 17. Average water consumption from the survey.

No person detailed in the consumption survey drank water that had been stored in cold storage without preboiling and only the bottle fed infant consumed water other than freshly drawn from the tap.

The loss due to process and consumption data was then combined with analytical results to attain an estimate of actual dose which would be received based on average water consumption at each of the properties in the detailed study and this is given in Table 19. A worst case taking into account the losses predicted from the drink making study, using the highest values of water consumption (summer consumption by adult 5), and the highest analytical results would give a CEDE potential of 22.5 mSv for an adult. In comparison 2.6mSv is the average dose received from background sources in the UK. The significance of the high dosage from 222 Rn observed in this study is evidenced by the fact that ingested radon normally only contributes about 10% of all risk from indoor radon, and radon itself only contributes 50% of background dose. On 'average' a householder would therefore get a CEDE from ingestion of water of only about 0.1mSv, the WHO recommended guideline from ingestion of radionuclides in groundwater. The average potential CEDE from householders in this study is 1.1 mSv (calculated before any losses due to process).

	Borehole site	Spring site	Well site
Adult	13.56	1.80	2.49
Child	3.93	0.51	0.67
Bottle-fed infant	9.43	1.28	1.87
Breast-fed infant	1.60	0.21	0.30

Table 18. Dose based on observed average consumption, mSv per annum.

Table 19. Dose based on individual consumption patterns, mSv per annum.

	Borehole site	Spring site	Well site
Adult1	16.73	2.27	3.32
Adult2	22.53	3.06	4.47
Adult3	7.83	1.06	1.56
Adult4	8.45	1.15	1.68
Adult5	15.51	2.11	3.08
Adult6	18.53	2.52	3.68
Child1	5.25	0.71	1.04
Child ₂	3.53	0.48	0.70
Bottle-fed infant	9.43	1.28	1.87
Breast-fed infant	7.07	0.96	1.40

Tables 18 and 19 assume, based on the findings of the drink making experiments.

- 10% of radon is lost on pouring
- No significant further loss occurs prior to cold drink consumption (90% of original radon content is consumed).
- 73% of what remains is lost on boiling (total loss of 76% from original)
- 10% is lost on pouring (total loss of 78% from original)
- No significant further loss occurs prior to hot drink consumption $(22% of original radon content is$ consumed).
- After sealing and cooled storage 25% is lost prior to consumption by a bottle fed infant (16% of original radon content is consumed).

The dose values calculated for the study only account for radionuclides physically ingested. The values may be raised significantly by inhalation of radon degassing from water supplies by processes in the household such as during drinks making, filling of baths, water storage tanks and showering. Radon has a greater dose conversion factor for inhalation than ingestion (Cross et al. 1985). There have been a number of studies on degassing effects of radon in the household (Hess et al., 1987, Fitzgerald et al. 1997). These studies derive transfer parameters from water-air during water processing. Using the data from the current project it may be possible to scale up from smaller scale water processes to larger scale (e.g. from kettle to bath). The overall 'dose burden' for a dwelling can than be calculated by including gas levels from ground intrusion.

8) CONCLUSIONS

²²²Rn was found to be above the draft Commission Recommendation action level of 1000 Bq/l in 8% of the tap water samples analysed.

 In terms of the geology at the source Dartmoor Granite and Undifferentiated Upper Devonian & Lower Carboniferous rocks yielded tap water with radon concentrations over 1000 Bq/l, while some water with a source on the Lower Carboniferous argillaceous rocks and chert had a radon concentration of over 1000 Bq/l at the source. Dartmoor Granite produced the highest average radon levels with the single highest value resulting from a borehole source in this formation and the highest values found in springs and wells coming from these rocks. The high degree of variability observed during the detailed study make it impossible to estimate the proportion of supplies which exceed the advisory level for what proportion of the time. The few examples of surface water supplies such as those drawn from streams and leats all show low radon concentrations (<35 Bq/l).

As a baseline all private water supplies would need to be sampled. More detailed sampling over a longer time period may clarify the relationship between 222 Rn and weather conditions indicated by the summer sampling so enabling a detailed picture of radon levels throughout the year to be built up.

For water from groundwater sources, mean values (by source type) at the tap are consistently lower than those values at the source. This is consistent with loss of radon due to degassing, as a result of water turbulence within the supply system and natural radioactive decay while the water is resident in the supply system.

Appendix 2 shows that in the majority of cases 222 Rn concentrations in tap water for a particular supply are lower than, or similar to, that at the source. However in a number of cases 222 Rn is significantly higher in the tap water sample than in the source sample. This may indicate very large short-term variability of 222 Rn levels in the supply. However at the particular borehole site, radon was consistently higher at the tap than the source during both of the detailed sampling exercises indicating the discrepancy is due to the loss of radon from the water while sampling the source, or an increase in radon while the water is in the supply pipe work, possibly due to the presence of radium bearing pipe scale.

The potential internal ionising radiation doses received has been calculated for each site involved in the screening study based on water consumption patterns found in the detailed study. In these cases the majority of dose from consumption of water arises from 222 Rn, with a small contribution from 210 Po, and insignificant dose from other uranium series elements. In all cases where there is a potential CEDE of above 1mSv at least 95% of the dosage will be from 222 Rn.

 In the locations with the highest radon values experiments have shown the dosage was well in excess of the WHO recommendation of 0.1mSv even after reduction due to the drink making process are taken into account. In the most extreme case it is over 13mSv per annum. Were a person to receive a dose at a place of employment(eg. in a hotel) of greater than 6mSv they would need to be a classified radiation worker (IRR 1999), which would then ensure work was conducted under a regime of monitoring, medical checks, personal protective equipment.

 To fully understand the exposure received by individuals it would also be necessary to incorporate dose due to inhalation of ²²²Rn. Workers in areas having greater than 400Bq/m3 are also covered by the procedures of IRR 1999.

Although ²²²Rn is the principal isotope contributing to radiological dose it is worth noting that ²³⁸U is present in a number of supplies at levels exceeding 2μg/l, the World Health Organisation (WHO) provisional guideline value for uranium in drinking water (WHO, 1998).

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Appendix 1. Sample site details.

Appendix 2. Screening study analysis results.

Appendix 3.

Methods used for study of loss due to drink making process.

Purchase a standard radium supported, radon-bearing solution and have a radon-bearing groundwater unsupported by radium. The two differing solutions are needed, as a radium-supported standard cannot be used for any long-term experiments as any radon lost due to process will rapidly be replaced from radium. The radon-bearing groundwater came from a source of negligible radium content The protocol below was used. Sub-sampling involved using a pipette to gently remove 10ml of sample and, without degassing effects, transfer the sample under the solvent scintillation fluid, see 3.1

Appendix 4 Complete list of adjusted CEDE's

The dose rates presented in this table are based on the average adult water consumption

pattern observed in the study

