High sensitivity measurement of ^{224}Ra and ^{226}Ra in water with an improved hydrous titanium oxide technique at the Sudbury Neutrino Observatory

B. Aharmim [c], B.T. Cleveland [d,e,f], X. Dai [b,d,e,1], G. Doucas [d], J. Farine [c], H. Fergani [d], R. Ford [f], R.L. Hahn [a], E.D. Hallman [c], N.A. Jelley [d], R. Lange [a], S. Majerus [d], C. Mifflin [b], A.J. Noble [e], H.M. O'Keeffe [d], R. Rodriguez-Jimenez [c], D. Sinclair [b], and M. Yeh [a]

[a] *Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973-5000, USA*
[b] *Ottawa-Carleton Institute for Physics, Department of Physics, Carleton University, Ottawa, Ontario K1S 5B6, Canada*
[c] *Department of Physics, Laurentian University, Sudbury, Ontario P3E 2C6, Canada*
[d] *Department of Physics, University of Oxford, Denys Wilkinson Building, Keble Road, Oxford, OX1 3RH, UK*
[e] *Department of Physics, Queen's University, Kingston, Ontario K7L 3N6, Canada*
[f] *SNOLAB, Sudbury, Ontario P3Y 1M3, Canada*

Abstract

The existing hydrous titanium oxide (HTiO) technique for the measurement of ^{224}Ra and ^{226}Ra in the water at the Sudbury Neutrino Observatory (SNO) has been changed to make it faster and less sensitive to trace impurities in the HTiO eluate. Using HTiO-loaded filters followed by cation exchange adsorption and HTiO co-precipitation, Ra isotopes from 200-450 tonnes of heavy water can be extracted and concentrated into a single sample of a few millilitres with a total chemical efficiency of 50%. Combined with beta-alpha coincidence counting, this method is capable of measuring 1.5×10^{-3} μBq/kg of ^{224}Ra and 3.3×10^{-3} μBq/kg of ^{226}Ra from the ^{232}Th and ^{238}U decay chains, respectively, for a 275 tonne D$_2$O assay, which are equivalent to 4×10^{-16} g Th/g and 3×10^{-16} g U/g in heavy water.

Key words: Radium, Thorium, Sudbury Neutrino Observatory, Hydrous titanium oxide, Ion exchange, Liquid scintillation

1. Introduction

SNO is a heavy water Cherenkov detector situated at a depth of about 2100 m in Vale Inco's Creighton mine in Sudbury, Canada [1]. It was built to detect neutrinos from the Sun and to understand the origin of the "Solar Neutrino Problem" - the longstanding disagreement between the measured neutrino flux from several previous solar neutrino experiments [2,3,4,5,6,7] and theoretical predictions from the Standard Solar Models (SSM) [8]. The SNO detector consists of an inner neutrino target of 1000 tonnes of ultrapure D$_2$O held in a 12 m diameter acrylic sphere which is surrounded by a shield of 7000 tonnes of H$_2$O

[1] Corresponding author. Tel: 1-613-533-2694; Fax: 1-613-533-6813; E-mail: xdai@owl.phy.queensu.ca.

contained in a 34 m high barrel-shaped cavity of maximum diameter 22 m. An array of approximately 9500 inward-looking photomultiplier tubes, mounted on a geodesic sphere of diameter 17.8 m, detects the Cherenkov light produced as a result of neutrino interactions occurring in the D_2O.

Solar neutrinos are detected in SNO through three distinct reactions: the charged current (CC) reaction, $\nu_e + d \rightarrow p + p + e^-$; the neutral current (NC) reaction, $\nu_x + d \rightarrow p + n + \nu_x$, ($x =$ e, μ, τ); and the elastic-scattering (ES) reaction, $\nu_x + e^- \rightarrow \nu_x + e^-$. The CC reaction is only sensitive to electron neutrinos — the neutrino type produced by fusion reactions in the Sun, whereas the NC reaction is equally sensitive to all active neutrino flavours.

There were three phases in the SNO experiment: pure D_2O (November 1999 to April 2001), salt (May 2001 to September 2003) when 2 tonnes of NaCl were added to the heavy water to improve the detection of neutrons from neutrino NC interactions, and the Neutral Current Detector (NCD) phase (November 2004 to November 2006). The total flux of all active neutrinos measured during the first two phases of the experiment agreed very well with the predictions made by the SSM, while only one third of the expected solar neutrinos were detected by the CC reaction [9,10,11,12]. This provided direct evidence that neutrino flavour change is the cause of the Solar Neutrino Problem.

The water used in the SNO detector had to be extremely clean to minimize background signals from naturally occurring radioactivity. The ^{232}Th and ^{238}U decay chains both contain gamma rays with energies greater than 2.22 MeV. These gammas produced by ^{208}Tl from the ^{232}Th chain and ^{214}Bi from the ^{238}U chain could photodisintegrate a deuterium nucleus, producing a neutron that was indistinguishable from a NC event. Radiopurity limits in the water were set by constraining the backgrounds from each decay chain to be less than one photodisintegration neutron per day in the SNO detector, compared with ~12.5 neutrons per day from solar neutrino NC interactions.

To measure the amounts of ^{208}Tl and ^{214}Bi, SNO developed three separate techniques to assay and purify ^{224}Ra (the ^{208}Tl precursor), and ^{226}Ra and ^{222}Rn (the ^{214}Bi precursors). These were MnOx

[13] and HTiO [14] for ^{224}Ra and ^{226}Ra, and degassing for ^{222}Rn [15]. The ^{232}Th and ^{238}U chains were not in secular equilibrium in SNO water due to leaching of radium isotopes from trace contamination of materials in contact with the water and from ingress of ^{222}Rn (half-life of 3.8 d). However, to facilitate comparisons between different assay methods, concentrations are quoted in terms of the equivalent amounts of ^{232}Th and ^{238}U in a chain in equilibrium. For a limit of one photodisintegration neutron per day the equivalent limits on ^{232}Th and ^{238}U were 3.8×10^{-15} g Th/g and 3.0×10^{-14} g U/g in the heavy water. The requirement in the light water surrounding the heavy water was less stringent, with upper limits of 3.7×10^{-14} g Th/g water and of 4.5×10^{-13} g U/g water.

In the original HTiO D_2O assay procedure [14], HTiO adsorbent was deposited onto a pair of 1 m long filters. The loaded filters were connected to the detector water systems underground and Ra from a few hundred tonnes of D_2O was extracted. Once the assay was complete, the Ra was removed by eluting the filters with 15 L of 0.03 mol/L of HNO_3. In the subsequent secondary concentration step, the HNO_3 eluate was further concentrated to a small sample volume for counting by co-precipitating radium with HTiO and passing it through a series of three MediaKap-10 filters; each filter was then eluted three times to ensure all of the activity was removed. The three eluates from each MediaKap-10 filter were combined, thus producing three samples per assay. This method was time consuming and required many samples to be measured in the beta-alpha counting system to determine the amount of radium extracted.

During the salt phase of the SNO experiment, a significant decrease in the secondary concentration efficiency was observed. This was attributed to increased levels of Mn in the D_2O from leaching of the beads used in the MnOx technique, which was extracted by the HTiO columns, forming hydrous manganese oxide during the co-precipitation step in the secondary concentration stage. The hydrous manganese oxide absorbed some of the Ra present in the sample, which could not be removed during the subsequent HCl elution of the MediaKap filters, resulting in a decreased secondary concentration efficiency.

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In addition, Ni was expected to leach slightly from the NCD detectors that were deployed in the D_2O region. Levels of less than about 1 ppb Ni were expected (and found) in the D_2O during the NCD phase. While this concentration would not affect the optical properties of the SNO detector it could, similar to manganese, decrease the secondary concentration efficiency of the HTiO method. Therefore, the effect of Ni on the new technique was studied.

To overcome the problem of interference by trace elements in the HTiO eluate, new elution and secondary concentration procedures were developed. This method is faster than the original method and it also reduces the number of samples produced and counted for each assay. In this paper, the details of the new method, which was used in the NCD phase of SNO, and evaluation of its efficiency are presented. All modifications are equally applicable to both light and heavy water assays.

2. The new HTiO procedure

An overview of the HTiO assay procedure is shown in Fig. 1. It consists of five steps: HTiO deposition, Ra extraction, elution, secondary concentration and counting. The HTiO adsorbent, formed as a white colloidal suspension by hydrolysing $Ti(SO_4)_2$, is deposited onto a Memtrex pleated filter (Osmonics, Inc., USA) made from polyethersulfone or polypropylene (PP) with a Ti coverage of 2.5 g/m^2. To achieve this, 15 L of dilute HTiO solution is recirculated through the filter at approximately 80 L/min for 10 min. A typical D_2O assay used a pair of 1 m long filters each with a surface area of 2.08 m^2 and a pore size of 0.1 μm. For H_2O assays, 0.25 m long filters were used with a surface area of 0.52 m^2 and a pore size of 0.1 μm. The filters used in D_2O assays must be deuterated before use in an assay to maintain the isotopic purity of the SNO heavy water. The HTiO production, deposition and deuteration procedures are described in detail in [14].

The filters are housed in PP columns and are loaded with HTiO before being transported underground. For an assay, the filters are attached to the SNO water circulation system where between 200 and 450 tonnes of D_2O (20-40 tonnes of H_2O) are passed through them at a typical rate of 22 $L/(min \cdot m^2)$. After extraction, Ra is eluted from the HTiO-loaded filters by circulating 15 L of 0.1 mol/L HCl through the columns at a flow rate of approximately 80 L/min for 20 min. The use of 0.1 mol/L HCl, rather than 0.03 mol/L HNO_3 as in the original procedure, is because the cation exchange resin used in the subsequent step extracts Ra in HCl with a higher efficiency; it also removes Th as well as Ra from the HTiO-loaded filters. In the secondary concentration step, Ra (and any Th if present) is extracted onto a cation exchange resin by flowing the 15 L HTiO eluate at a rate of 250 mL/min through a polymethylpentene (PMP) column (1.0 cm diameter, 18.0 cm length) containing 12.0 g of Dowex 50WX8 resin (H^+-form, 100 mesh). The resin is mixed with 50 mL of 0.1 mol/L HCl and added to the column, followed by rinsing with 50 mL of 0.1 mol/L HCl before the eluate is passed through it. To minimise the procedural background, the resin is only used once for each assay or background experiment.

The resin column is rinsed with ultrapure water (UPW) to neutral pH and then turned in reverse flow direction for Ra elution. Approximately 100 mL of 0.25 mol/L of disodium ethylenediaminetetraacetate (EDTA) is passed through the column at pH 10 and a flow rate of 5 mL/min to remove Ra. About 15% of any Th extracted by the resin is also removed. The EDTA eluate is collected in a PTFE beaker. Upon addition of 8 mL of concentrated HNO_3 to the eluate, the EDTA precipitates. This EDTA is decomposed by boiling the solution for 15 minutes after which time a second 8 mL of concentrated HNO_3 is added, and boiling is continued for a further 15 minutes. The next step involves coprecipitation with HTiO. Approximately 250 mL of UPW is added to the residual solution along with 2 mL of 15% $Ti(SO_4)_2$ (60 mg of Ti). This solution is titrated to pH 9 with NaOH which causes coprecipitation of the HTiO and Ra. In the final step, the solution is centrifuged at 3400 rpm for 3 min. The supernatant is decanted and the HTiO precipitate is dissolved in 1.5 mL of concentrated HCl. This final sample is diluted with UPW to 8 mL and mixed with 42 g of Optiphase HiSafe 3 liquid

scintillator cocktail (purchased from PerkinElmer, Inc.) in a 60 mL PMP jar. The amount of [224]Ra and [226]Ra is inferred by counting the samples using a beta-alpha coincidence counter for 10-14 days.

The beta-alpha coincidence counting system is described in detail in [14]. The background of this counting system was measured using a blank sample (a mixture of 10 mL of 0.5 mol/L HCl and 42 g of liquid scintillator cocktail) and the count rate was found to be very low, 0.03 h^{-1} for [224]Ra and 0.3 h^{-1} for [226]Ra. The [226]Ra background is mainly contributed by the Optiphase HiSafe 3 liquid scintillator cocktail. The counting efficiencies were also measured to be 45±5% for [224]Ra and 60±10% for [226]Ra. These efficiencies were regularly checked using [226]Ra and [228]Th calibration sources. Throughout the paper ± values are standard combined uncertainties (k=1).
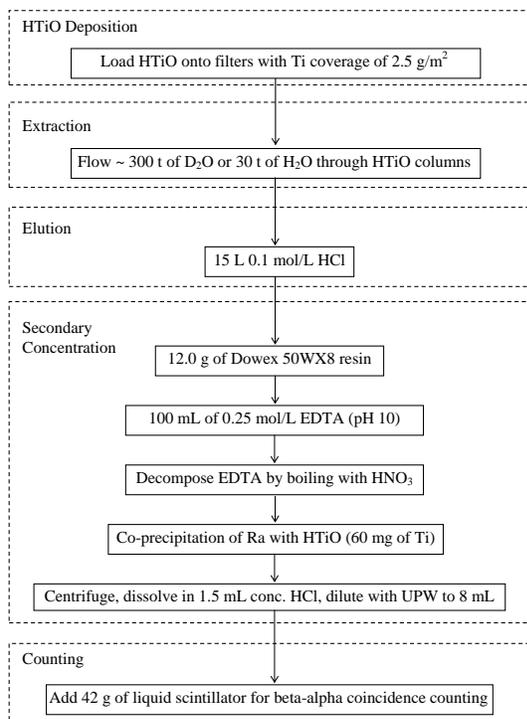

Fig. 1. The new Ra procedure for an HTiO water assay.

Table 1
Summary of efficiencies of Ra for the new procedure.

Efficiency	[226]Ra	[224]Ra
Extraction ε_{ext}	95±5%	
Elution ε_{elu}	90±10%	
Secondary concentration ε_{conc}	58±6%	
Total chemical ($\varepsilon_{ext}\cdot\varepsilon_{elu}\cdot\varepsilon_{conc}$)	50±8%	
Counting ε_{count}	60±10%	45±5%
Total ($\varepsilon_{ext}\cdot\varepsilon_{elu}\cdot\varepsilon_{conc}\cdot\varepsilon_{count}$)	30±7%	22±4%

3. Results and discussion

3.1. *Efficiencies for the new procedure*

The Ra efficiencies for the new procedure were determined by spike experiments using 0.01-0.5 Bq of [226]Ra tracer and are given, along with the uncertainties, in Table 1. The total chemical recovery for Ra was determined to be 50±8%. Combined with the counting efficiencies, the total procedural efficiencies are 30±7% for [226]Ra and 22±4% for [224]Ra. In the following sections, the methods used to obtain the efficiency for each step are discussed in detail.

3.1.1. *Extraction*

In previous studies [14,16], the extraction efficiency for Ra was carefully examined, and for a typical HTiO water assay in the pure D_2O phase ($\sim$200 tonnes for D_2O), the extraction efficiency was determined to be 95±5% at a flow rate of 19 L/(min·m^2). This was measured by flowing D_2O through two sets of HTiO-loaded filters (each set consisting of two parallel filters), connected in series. The activities extracted by the two sets were compared. The formula used to calculate the extraction efficiency, ε_{ext}, is

$\varepsilon_{ext} = 1 - \frac{A_d}{A_u}$,

where A_d and A_u are the activities on the downstream and upstream columns, respectively, and where the first two columns are the upstream columns.

During the NCD phase the heavy water was passed through a reverse osmosis unit from time to time to maintain a low level of metallic ions as in the pure D_2O phase, so an extraction of 95±5% was assumed. Checks of the extraction efficiency

made during the NCD phase were consistent with this value.

3.1.2. Elution

To investigate the effect of HCl concentration on the Ra and Th elution efficiencies, several small scale spike experiments were carried out using HTiO-loaded (2.5 g Ti/m^2) MediaKap filters. These have identical pore size, but smaller surface area (0.015 m^2) than the Memtrex filters used for assays. The elution efficiency of Ra was found to be 90±10% for HCl concentrations ranging from 0.1-0.5 mol/L, while the elution efficiency of Th was decreased from 95±5% to 65±5% with HCl concentrations decreasing from 0.5 to 0.1 mol/L. Higher concentration of HCl would also dissolve more Ti from the loaded filters which could reduce the Ra extraction efficiency by the cation exchange resin in the secondary concentration step. Therefore, 0.1 mol/L of HCl was chosen to elute Ra and Th from the HTiO filters. The Ra elution efficiency for this acidity was determined to be 90±10% which is a slight improvement on the use of HNO$_3$ in the previous method.

3.1.3. Secondary concentration

One major concern for the secondary concentration efficiency of water assays was the presence of impurities such as Mn and Ni in the water, which could be observed in the HTiO eluate along with Ti stripped from the HTiO-loaded filters during elution. By adding a known amount of ^{226}Ra and ^{228}Th radiotracers, spike experiments were conducted to determine the efficiency for each step of the new secondary concentration procedure in the presence of Mn, Ni and Ti. In each spike experiment, 130 mg of Mn, 140 mg of Ni and 1500 mg of Ti were added to 15 L of 0.1 mol/L HCl to mimic a typical assay eluate. The amounts of Mn and Ni were based on the estimated maximum adsorption capacities for these elements by a pair of 1 m long HTiO loaded filters measured in an assay during the transition from the salt to the NCD phase of SNO. The amount of Ti was based upon test elutions of HTiO-loaded filters. In these spike experiments, ^{226}Ra and ^{228}Th were counted using the beta-alpha coincidence counters, while all of the

stable tracers, including Ba, Mn, Ni and Ti, were measured using inductively coupled plasma mass spectrometry (ICP-MS) at the Geoscience Laboratories, Sudbury, Ontario.

The secondary concentration procedure consists of two steps. In the first step, Ra is adsorbed onto an ion exchange (IX) resin and eluted into a relatively small volume. The second step further concentrates Ra, resulting in an 8 mL sample. The Dowex 50WX8 cation exchange resin is widely used for the pre-concentration of Ra in environmental samples [17,18,19] due to its strong retention ability and fast adsorption equilibrium. The adsorption and elution behaviour of Ra from this resin were carefully examined for extraction of Ra from 15 L of the HCl eluate. The extraction efficiency for Ra by the IX resin was measured to be greater than 90% in the presence of Mn, Ni and Ti from the spike experiments (see Table 2) (the Ra is preferentially adsorbed by both the HTiO-loaded filter during extraction and by the IX resin over Mn, Ni or Ti). The elution of Ra from the IX resin at various EDTA concentrations and pHs was also studied. It was found that 90% of the extracted Ra was eluted into 100 mL of 0.25 mol/L EDTA at pH 10. Most of the Ti remained on the resin (see Table 2). An HTiO co-precipitation method is used to further concentrate the Ra from 100 mL of EDTA eluate for beta-alpha coincidence counting. Since co-precipitation would not occur in the presence of a high concentration of EDTA, the EDTA must be decomposed by boiling with concentrated HNO$_3$ prior to this step. The spike experiments indicated that an average of 69% of the eluted Ra from the resin could be concentrated into the final counting sample (see Table 2). Tests were also performed in the absence of Ni and Mn and no observable difference in the secondary concentration efficiency was found.

3.1.4. Counting

In principle high amounts of Mn or Ni could reduce the counting efficiency as both have absorption bands between 350 and 500 nm, the region of wavelengths to which the photomultiplier tubes used in the beta-alpha coincidence counters are most sensitive. However, as only very small quanti-

Table 2

Efficiencies of Ra, Th, Ni, Mn and Ti in the secondary concentration step from the spike experiments.

| | Efficiencies in secondary concentration step | | | Impurity contents (mg) | |
	IX extraction	IX elution	HTiO co-precipitation	HCl eluate	Final sample
^{226}Ra	93±4%	90±6%	69±5%		
^{228}Th	98±2%	15±2%	48±10%		
Ba	94±4%	95±5%	15±1%		
Ni	33±3%	67±7%	<0.2%	140	<0.06
Mn	33±3%	75±9%	<7%	130	<3
Ti	51±9%	3.0±0.4%	∼75%	1500	∼62

ties of Mn and Ni were present in the final sample, the counting efficiency was unaffected. A typical assay sample looks milky white after mixing with liquid scintillator cocktail due to up to ∼62 mg Ti in the final sample. This does not affect the counting efficiency significantly as the scintillation light is only scattered but not absorbed; as a result the position of the alpha peak is not altered appreciably. Tests were performed to verify this and no obvious decrease in counting efficiency was observed (see Fig. 2). The counting efficiencies were found to be $45 \pm 5\%$ for ^{224}Ra and $60 \pm 10\%$ for ^{226}Ra. Combining all of the individual efficiencies, the total secondary concentration efficiency is found to be 58±6% for Ra (see Table 1).

3.1.5. ^{228}Th measurement

As the HCl elution of the HTiO-loaded filters removes Th with an efficiency of 65±5%, Th will be extracted by the IX resin, along with the Ra. Approximately 15% of the Th is removed from the IX resin when the Ra is eluted with EDTA (see the IX elution efficiency for ^{228}Th in Table 2). Following the EDTA elution the column is sealed and left moist and Ra builds up from the Th that remains on the resin. Approximately 9-10 days after the initial EDTA elution, the equilibrium between ^{228}Th and ^{224}Ra is well established and the resin may be re-eluted with EDTA. The secondary concentration proceeds as before, and the efficiencies of all subsequent stages are identical to those given in Tables 1 and 2. As the elution efficiency is lower for Th compared to Ra and as approximately 15% of the Th is removed with the initial IX elution, the overall chemical efficiency is approximately 27% for a second IX elution.
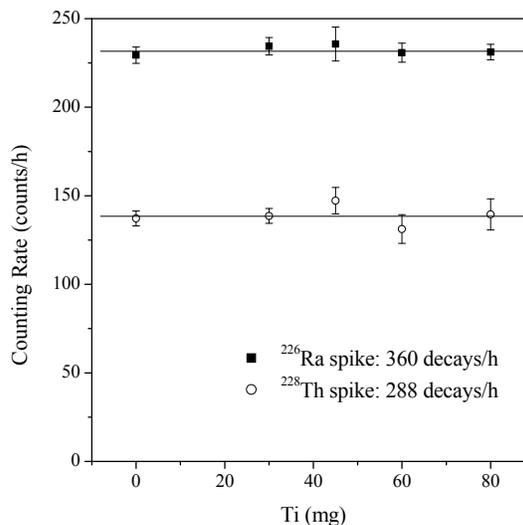

Fig. 2. Effect of various Ti concentrations on the counting efficiencies.(Sample matrix: titanium, in the form of HTiO was dissolved in 2.0 mL of concentrated HCl + 6.0 mL of UPW + 42 g of Optiphase HiSafe 3 liquid scintillation cocktail.)

3.2. Background and detection limit

To determine the contribution of the equipment and reagents to the radioactive signal observed for an assay, a background measurement must always be carried out. This takes place typically one week before the assay. For a background measurement, the filters are loaded with HTiO and processed in an identical manner to an assay. Underground extraction does not occur during the background measurement. Following this measurement the filters are cleaned using 0.5 mol/L HCl and re-loaded with HTiO for the assay. The count rate for a typical background signal was

found to be 0.25 ± 0.1 h^{-1} for ^{224}Ra (^{232}Th chain) and 0.85 ± 0.3 h^{-1} for ^{226}Ra (^{238}U chain). This background arises from Ra leaching from the apparatus and from the reagents used in the elution and secondary concentration processes. Taking the detection limit as 3.29 times the standard deviation of the background rate [20], the minimum detectable concentrations of the new procedure for a 275 tonne assay are 1.5×10^{-3} μBq/kg of ^{224}Ra and 3.3×10^{-3} μBq/kg of ^{226}Ra, which are equivalent to $\sim4\times10^{-16}$ g Th/g and $\sim3\times10^{-16}$ g U/g in heavy water.

3.3. Assay results

A 275 tonne assay of the heavy water in the NCD phase of SNO gave a (signal minus background) count rate of 0.5 ± 0.2 h^{-1} for ^{224}Ra (^{232}Th chain) and 0.0 ± 0.4 h^{-1} for ^{226}Ra (^{238}U chain). These were the rates at the start of counting. The Ra activity in the water was deduced after allowing for the time of extraction ($\sim$3 d) and the delay between the end of extraction and counting ($\sim$5-6 h). Assuming radioactive equilibrium in the ^{232}Th and ^{238}U chains, these rates correspond to a value of $8\pm4\times10^{-16}$ g Th/g water and less than 3×10^{-16} g U/g water. The systematic error arises mainly through the uncertainties of $\sim$20% for ^{224}Ra and $\sim$24% for ^{226}Ra in the efficiencies of the chemical procedures and counting (see Table 1), and to a lesser extent from the uncertainties in the background subtraction and in the volume of water sampled. The systematic uncertainty contributions (k=1) are estimated to be $\pm$24% for the ^{232}Th chain and $\pm$29% for the ^{238}U chain. This procedure was used for light water as well as heavy water HTiO assays in the NCD phase of the SNO experiment.

The new procedure was also used to assay trace amounts of ^{224}Ra and ^{226}Ra on the surface of sections of one of the NCDs. Each section was placed in a column, connected to the elution rig and the outer surface rinsed with 0.1 mol/L HCl at 80 L/min for 20 minutes. Following the elution, the concentration of Ni in the 15 L eluate was very high (approximately 80 mg of Ni was present). The new secondary concentration procedure was followed to extract Ra. To ensure that the secondary concen-

tration efficiency was unchanged, the procedure was tested using a number of test sections which had been plated with ^{228}Th. The secondary concentration efficiency derived from these tests was in agreement with that given in Table 1. The final results were 0.91 ± 0.21 μg of ^{232}Th and 0.11 ± 0.03 μg of ^{238}U, in good agreement with measurements obtained by two other independent techniques [21].

4. Conclusions

We have developed and implemented a new secondary concentration stage for the HTiO-based assay procedure used to determine the Ra concentration in both the light and the heavy water of the SNO detector. Compared to the previous HTiO assay method [14], the new procedure offers the following advantages:

(1) It is less sensitive to impurities in the HTiO eluate and was unaffected by concentrations of 9.3×10^{-6} g Ni/mL, 8.7×10^{-6} g Mn/mL and 1.0×10^{-4} g Ti/mL in the 15 L HTiO eluate.

(2) It requires fewer beta-alpha coincidence counters; only two samples (one for assay plus one for background) are produced for one water assay using the new procedure while six samples (three assay, three background) were produced using the previous method.

(3) The time required to process the filters once the assay has been completed is reduced from 12 hours to around 5-6 hours.

The new HTiO procedure has been used to assay the light and heavy water of the SNO detector, with detection limits of 1.5×10^{-3} μBq/kg for ^{224}Ra and 3.3×10^{-3} μBq/kg for ^{226}Ra, which are equivalent to 4×10^{-16} g Th/g and 3×10^{-16} g U/g for a 275 tonne heavy water assay. It has also been used to assay for trace amounts of ^{224}Ra and ^{226}Ra on sections of the NCDs. With appropriate validations, the technique could have a variety of applications in the measurement of low-level natural radioactivity in different environmental and biological samples.

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