

# Electrolytic enrichment method for tritium determination in the Arctic Ocean using liquid scintillation counter

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## Abstract

A method of measuring the tritium in seawater based on electrolytic enrichment and ultra-low background liquid scintillation counting techniques was established. The different factors influencing the detection limit were studied, including the counting time, the electrolytic volume of the seawater samples, the selection of background water, scintillation solution and their ratio. After optimizing the parameters and electrolyzing 350 mL volume of samples, the detection limit of the method was as low as 0.10 Bq/L. In order to test the optimization of system for this method, of the 84 seawater samples collected from the Arctic Ocean we measured, 92% were above the detection limit (the activity of this samples ranged from 0.10 Bq/L to 1.44 Bq/L with an average of  $(0.30 \pm 0.24)$  Bq/L). In future research, if we need to accurately measure the tritium activity in samples, the volume of the electrolytic samples will be increased to further reduce the minimum detectable activity.

**Key words:** tritium, liquid scintillation, cocktails, electrolytic enrichment

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## 1 Introduction

Tritium ( $^3\text{H}$ ) is a radioactive isotope of hydrogen with a half-life of 12.3 years (Gröning and Rozanski, 2003), which emits low-energy beta particles with a maximum energy of 18.6 keV (Carsten, 1979). It is a naturally occurring radionuclide through the interaction of high-energy cosmic rays with oxygen and nitrogen atoms in the upper atmosphere (Popoaca et al., 2014). The environmental levels of tritium increased after nuclear weapon tests between 1945 and 1963 (Jacobs, 1968; Hua and Wen, 2008), and after that it is mainly released from nuclear facilities, especially the heavy water reactor (HWR) (Jacobs, 1968; Carsten, 1979).

Dramatic changes have been observed in the hydrological cycle in the Arctic over the last century, such as changes in the magnitude and timing of ice melting, precipitation and surrounding river discharge, and sea level changes in the Arctic Ocean (Peterson et al., 2002; Aagaard and Carmack, 1989; Carmack, 2000; Serreze et al., 2006). Multiple tracers (e.g.,  $\delta^{18}\text{O}$ ,  $\delta^2\text{H}$  and  $^3\text{H}$ ) have been widely used to identify and quantify the freshwater contribution in studies of water mass compositions and movement in the Arctic Ocean (Yi et al., 2012; Cooper et al., 2008), which provides insight into the origin of the isotopic signals and the link between geomorphic features and hydrologic variations.

Liquid scintillation counting (LSC) is the most common technique for measuring low-energy beta emitters, especially for triti-

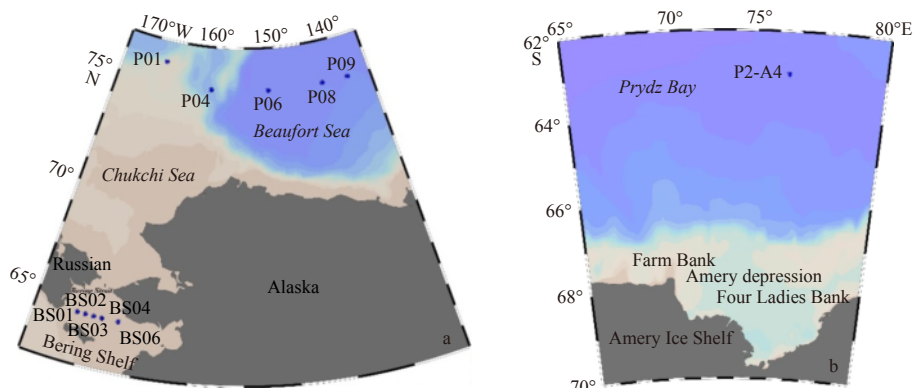
um. Examples include measuring anthropogenic inputs in natural environments, such as global fallout from nuclear weapons test (Kern et al., 2009), routine monitoring of nuclear power reactors (Janovics et al., 2014; Nikolov et al., 2013), and assessing the release from nuclear fuel reprocessing facilities to surrounding environments (Koarashi et al., 2008). The radioactive sample is combined with a liquid scintillation cocktail, and the radionuclide decay produces an ionizing particle. At present, due to the natural decay of tritium and the prohibition of nuclear activities, the activity of tritium in water decreases year by year, and the concentration of atmospheric tritium has gradually returned to the background level since the 1990s (Carsten, 1979). The average tritium activity in marine and groundwater is 0.11 Bq/L and 0.15 Bq/L, respectively, which makes it difficult to directly measure the concentration of tritium in water using the LSC method. While measuring the tritium in the sample, if the radioactive activity of the sample is less than 10 Bq/L, the net count rate of the sample is the same order of magnitude as the background count rate, or even lower (Wang et al., 2011), so the accuracy of the measured data cannot be guaranteed. The International Atomic Energy Agency recommends the addition of an electrolytic concentration pretreatment, which can significantly reduce the detection limit of the liquid scintillation to accommodate measurement of tritium in the environment.

The principle of electrolytic concentration of tritium is to use

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the isotope fractionation effect of hydrogen isotopes in the gas and liquid phases. HTO is more difficult to electrolyze than  $H_2O$ . The solid polymer electrolyte concentration method (SPE) mainly uses a special kind of electrolyte (Nafion membrane), directly catalytic electrolysis produce  $H_2$  and  $O_2$ , to achieve the goal concentration. It is characterized by the absence of any solute residue after electrolysis and an unlimited volume of concentrated water. The purpose of this study was to establish a method for the measurement of low-activity tritium in seawater, which can not only accurately determine the tritium activity in the Arctic Ocean, but can also provide accurate signals for multi-tracer studies used to identify and quantify water mass compositions.



**Fig. 1.** Sampling station. a. Arctic and b. Antarctic.

## 2.2 Experimental procedure

### 2.2.1 Seawater distillation

Generally, sample treatment consists of distillation to desalt and remove any impurities and interfering radionuclides to reduce quenching. The seawater is filtered through a 0.45- $\mu$ m membrane (Waterman, 47 mm) to remove suspended particles, followed by storage in brown glass bottles. One liter of filtered seawater was added to a distillation bottle along with 4 g of anhydrous sodium carbonate, 2 g of potassium permanganate, and 2 g of copper powder. The ground glass plug was covered and the serpentine condensing tube was installed, followed by the heating distillation. The first 10 mL of the distillate was discarded (used to clean the distillation lines), and the distillate was collected and sealed in a frosted glass bottle.

### 2.2.2 Background water and cocktail

A large number of commercial scintillation cocktails are available for tritium counting; however, only a few hold a large amount of water and an even smaller number have good specifications for level counting (Varlam et al., 2009; Jakonić et al., 2014). The cocktails available in our laboratory are Ultima Gold LLT and OptiPhase HiSafe 3. A comparison of their efficiencies was conducted in this study.

Furthermore, a blank was also analyzed to determine the background of the detector. We selected pure laboratory water (A), several brands of commonly available pure water (B, C, D, E), and distillates (F) extracted from the deep seawater (3 200 m) in the outer waters of Prydz Bay, Antarctica (Section 2.1) as the blanks. Twenty-four polyethylene scintillation bottles were used and divided into six groups with four parallel samples in each group. Eight milliliters of each of these six background water samples were mixed with 12 mL of scintillation solution in sealed

## 2 Materials and methods

### 2.1 Sampling

The samples were collected onboard the icebreaker *Xuelong* from July to October 2017 during the 8th Chinese National Arctic Research Expedition (CHINARE) (Fig. 1a). Deep seawater was collected as the background during the 30th CHINARE in the Prydz Bay (Fig. 1b).

Seawater samples from the surface to the ocean bottom were collected using a CTD/Rosette system. The samples were placed in clean, well-sealed, 1 L, polyethylene bottles and were stored in a refrigerator before further processing in the laboratory.

bottles.

### 2.2.3 Counting time

Four parallel tritium-free water samples were selected and counted for 50 min, 100 min, 200 min, 300 min, 500 min, 800 min and 1 000 min after being stored for 24 h in darkness.

### 2.2.4 Ratio of sample to scintillant

To optimize the volume ratio between the sample and the scintillation liquid solution, the tritium standard solution ((33.5 $\pm$ 0.7) MBq/g, (2.013 1 $\pm$ 0.001 7) g, Physikalisch-Technische Bundesanstalt, Germany) was diluted to 92.1 Bq/L to make the standard solution for measurement. We added 0 mL, 2 mL, 4 mL, 6 mL, 7 mL, 8 mL, 9 mL, 11 mL, 13 mL, 15 mL and 17 mL of tritium standard into individual scintillation bottles, followed by the addition of scintillation solution to reach a total volume of 20 mL.

## 2.3 Electrolytic enrichment

In order to concentrate the tritium level to make measurement easier, electrolytic enrichment was conducted (Nikolov et al., 2013). In this study, an electrolytic instrument (ECTW-1, Hapstar, China) based on SPE technology was used, which only required the addition of the samples directly into the electrolytic instrument. The cooling temperature of the instrument can reach to 5–10°C. And the standard deviations (SD) for electrolysis time and tritium recovery were less than 1.5% and 10%, respectively. Samples of 100 mL, 200 mL, 350 mL, 500 mL and 700 mL of tritium standard solution (4.36 Bq/L) was added to the electrolytic instrument and its enrichment coefficient was measured.

The specific steps used are as follows.

(1) After the conductivity of the distilled and purified sample was less than 5  $\mu$ S/cm, the sample was placed in a washed glass bottle.

(2) The deionized water previously added to the electrolyzer was removed.

(3) The pipeline of the instrument containing the sample was cleaned by 50 mL of purified sample water.

(4) About 50 mL of purified sample water was poured into the instrument again, after electrolyzing for about 10 min. The sample water was emptied.

(5) A certain amount of purified water sample was poured into the instrument, and the electrolysis was conducted.

(6) After the electrolysis, the concentrated liquor was collected in the collection bottle for storage.

(7) After the experiment, the instrument pipeline was cleaned with deionized water.

#### 2.4 Measurement

All of the prepared samples were measured using an ultra-low-level LSC Quantulus 1220 spectrometer (Perkin Elmer, Waltham, MA, USA). The instrument includes its own background reduction system around the vial chamber, which consists of both an active and a passive shield (Nikolov et al., 2013). The detector has a multi-channel analyzer that separates the tritium signals in the sample from those produced by the chemiluminescence into two different spectra. The window for tritium was set between Channels 30 and 250.

After shaking and mixing the measured samples, they were placed in the chamber of the ultra-low background liquid scintillation spectrometer for dark adaption for 24 h before counting, which allowed for the full decay of the chemiluminescence and the photo-stimulated luminescence. Then, the samples were measured.

#### 2.5 Data analysis

The minimum detectable activity (MDA, Bq/L) is usually used to discriminate the measurement from the background. The MDA was evaluated using the following equation:

$$\text{MDA} = \frac{3.29 \sqrt{cpm_{bk}/t_{bk} + cpm_{bk}/t_s}}{60\varepsilon \cdot V \cdot EF} \quad (1)$$

The sample activity ( $A$ , Bq/L) was obtained from the following equation:

$$A = \frac{cpm_s - cpm_{bk}}{60\varepsilon \cdot V \cdot EF} \quad (2)$$

The uncertainty was evaluated using a confidence level of 95% ( $K=2$ ) and the following equation:

$$\sigma = \sqrt{\frac{1}{cpm_s^2} \left( \frac{cpm_s + cpm_{bk}}{t_s} + \frac{cpm_{bk}}{t_{bk}} + \sigma_{EF}^2 + \sigma_\varepsilon^2 \right)} \quad (3)$$

In the above equations,  $cpm_{bk}$  is the net count rate of the background water;  $cpm_s$  is the net count rate of the sample;  $t_{bk}$  and  $t_s$  are the background and sample counting times, respectively;  $V$  is the sample volume in liters;  $\varepsilon$  is the counting efficiency;  $\sigma_\varepsilon$  is the standard deviation of  $\varepsilon$ ;  $EF$  is the enrichment factor; and  $\sigma_{EF}$  is the standard deviation of  $EF$ .

### 3 Results and discussion

The determination of the tritium activity in seawater samples can be conducted using different parameters. We optimized the parameters that directly affect the radioactive counting.

#### 3.1 Selection of the background water

Water that has received no tritium for a period of time equivalent to more than five times the half-life of tritium (i.e., approx. 60 years) can be used as suitable background water, such as water from the deep ocean or from petroliferous wells. Additionally, due to the difficulty of obtaining this type of water, a variety of commercial mineral bottled waters were also considered, of which those containing the lowest tritium abundance served as the blank. The activities of the background samples are showed in Fig. 2. They vary from 1.349 counts per minute (CPM) to 1.563 CPM, with a range of 19% change. Pure water Sample B had the lowest activity, while pure water Sample E had the lowest value.

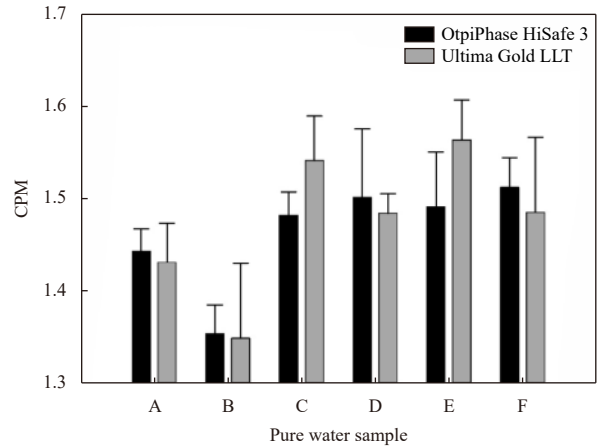


Fig. 2. Effect of the different background water and scintillation cocktails on the counting rate.

According to Eq. (3), when the measuring time of the sample was consistent with that of the background sample, the MDA was directly proportional to  $\sqrt{cpm_{bk}}$ . In order to reduce the MDA, the CPM of the selected background water should be as low as possible. Therefore, during the measurement of seawater samples, pure water Sample B was selected as the background water.

#### 3.2 Selection of the scintillation solution

At present, there are many commercial scintillation solutions available for tritium measurement. Nevertheless, only a few scintillation solutions can be mixed with a large amount of water, and even fewer have low tritium levels. In this study, the background value and efficiency of the tritium measurement of OtpiPhase HiSafe 3 and Ultima Gold LLT were compared. As is shown in Fig. 2, there was no significant difference between the two solutions (SPSS,  $I>0.05$ ). However, the Ultima Gold LLT scintillation solution can be used with a wider range of salinity, pH, and total dissolved solids than OtpiPhase HiSafe 3 based on their product specifications. Since our experiment mainly involved seawater samples, Ultima Gold LLT was selected as the scintillation solution for the follow-up experiment.

#### 3.3 Optimization of the sample to the scintillant ratio

Water is a strong quenching agent, and the quenching will be stronger if the water-scintillant mixed solution has a high water abundance (Thomson, 2002). In contrast, a low water/scintillant ratio can allow low radioactivity to be detected. Thus, the ratio of the water sample to the scintillation solution is critical for the measurement of tritium (Zhang et al., 2010).

Each standard sample was counted for three cycles and 5 min for each cycle. Our results show that the count rate increased gradually with the volume of tritium solution, reaching the maximum value at 9 mL followed by a decreasing trend with increas-

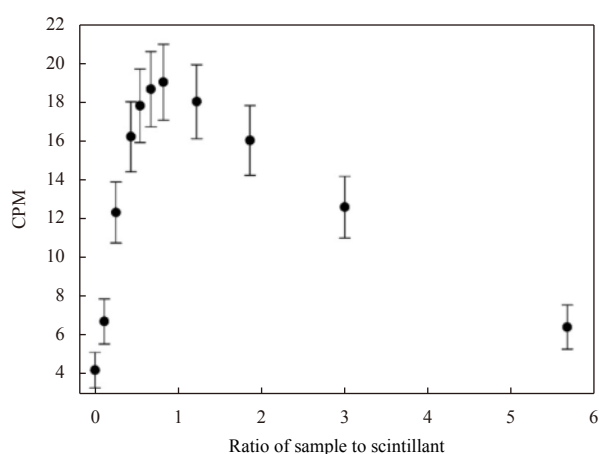
ing volume of tritium solution. The sample had the highest count rate (18.156 CPM) at a ratio of 9:11 (water/scintillant), indicating that the scintillation solution reached the highest rate in the dissolved water. However, through calculation, it was found that the measurement efficiency was the highest when the water/scintillant ratio was 8:12, with a counting rate of 17.739 CPM. Considering the inverse relationship between the measurement effect and the detection limit (Pujol and Sanchez-Cabeza, 1999; Varlam et al., 2009), we determined that the optimal water/scintillant ratio was 8:12 (Fig. 3).

### 3.4 Counting time

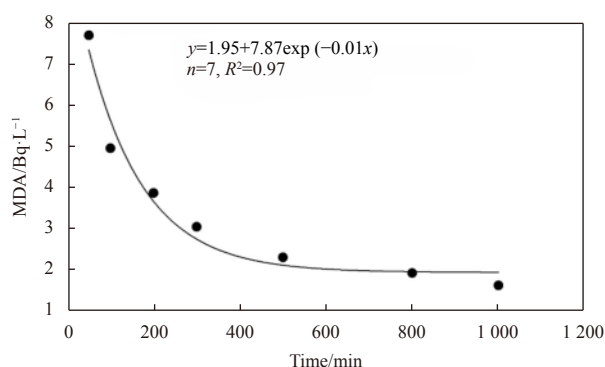
The detection limit was evaluated at different counting times using Eq. (1). As can be seen from Fig. 4, the detection limit decreased exponentially with increasing counting time, decreasing from 7.68 Bq/L at 10 min to 1.63 Bq/L at 1 000 min. Based on the variation trend of the curve, the change in the detection limit was minor when the counting time exceeded 1 000 min, similar to the observations of Pujol and Sanchez-Cabeza (1999) and Jakonić et al. (2014). Therefore, the counting time for tritium measurements should be increased to 1 000 min for low-tritium-level seawater samples.

### 3.5 Electrolytic enrichment

Based on the experimental results presented above, we optimized the tritium-free water, the scintillation solution, their volume ratio, and the counting time (1 000 min). Using these optimized conditions, the detection limit of the tritium in the water



**Fig. 3.** Variation in counting rate as a function of the ratio of sample to scintillant.



**Fig. 4.** Change in the minimal detectable activity (MDA) as a function of time.

sample can be reduced to 1.07 Bq/L. In order to test the accuracy of this method, our laboratory participated in the International Atomic Energy Agency's (IAEA) radionuclide measurement and inter-comparison in the Asia-Pacific oceanic region in November 2017. Using our established method, we obtained  $(3.09 \pm 0.52)$  Bq/L of tritium, and the relative deviation from the IAEA's given value  $((3.12 \pm 0.06)$  Bq/L) was only 0.9%, which proves the accuracy of this method in the determination of tritium in natural seawater.

In addition, 10 seawater samples from the Arctic Ocean were also tested using our established direct methods (Table 1). However, they were all found to be undetectable (i.e., below the MDA), which means that electrolytic enrichment of the samples is required for the accurate measurements of tritium activity in these Arctic seawaters. Electrolytic enrichment would lower the MDA by an order of magnitude. Therefore, the stability of the electrolytic enrichment method as well as the electrolytic volume and enrichment coefficient were tested in this study.

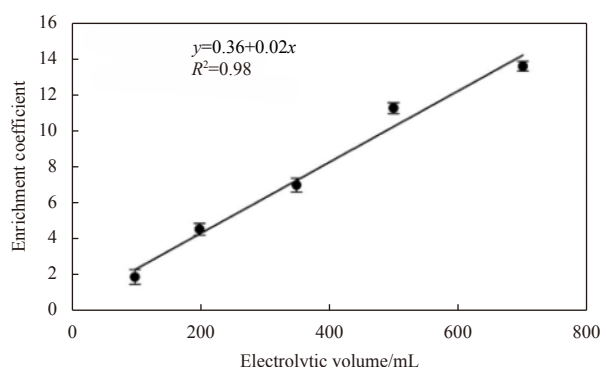
As can be seen from Fig. 5, the enrichment coefficient increased with increasing electrolytic volume, exhibiting a significant linear relationship ( $p < 0.002$ ). In addition, as the electrolysis volume increased from 100 mL to 700 mL, the electrolysis time also increased from about 7 h to 50 h. Thus, the appropriate electrolytic volume can be selected depending on the tritium activity range of the actual samples.

For the tritium measurements of the Arctic seawater, the volume of the electrolytic sample was first selected to be 350 mL, for which the MDA decreased to 0.10 Bq/L.

Therefore, we re-tested 10 samples of Arctic sea water using the electrolysis enrichment method (electrolysis volume of

**Table 1.** The results of the direct and electrolytic enrichment methods used to measure the Arctic seawater

Sample	Specific activity	
	Direct measurement/Bq·L <sup>-1</sup>	Electrolytic enrichment/Bq·L <sup>-1</sup>
P01 (0 m)	<1.07	0.19±0.09
P04 (0 m)	<1.07	0.26±0.07
P06 (0 m)	<1.07	0.21±0.08
P08 (1 000 m)	<1.07	0.22±0.10
P09 (0 m)	<1.07	0.36±0.08
BS01 (0 m)	<1.07	<0.10
BS02 (0 m)	<1.07	<0.10
BS03 (0 m)	<1.07	0.23±0.10
BS04 (0 m)	<1.07	0.15±0.08
BS06 (0 m)	<1.07	0.21±0.09



**Fig. 5.** Variation in the enrichment coefficient with electrolytic volume.

350 mL). The test results reveal that except for Samples BS01 (0 m) and BS02 (0 m), which were below the detection limit, the activity of the remaining eight samples ranged from 0.15 Bq/L to 0.36 Bq/L with an average of  $(0.23 \pm 0.06)$  Bq/L. This result is far lower than the measured values of Dorsey and Peterson (1976) for samples from the Arctic Ocean (up to 5.9 Bq/L) and from the East Greenland Current (up to 8.3 Bq/L).

In addition, Fourré et al. (2006) extrapolated a decreasing trend in the samples from the Summit during 1984–1992. They concluded that the present day levels should be close to the natural background of 1.2 Bq/L to 2.4 Bq/L. Considering that our samples were collected in 2017, no new atmospheric nuclear explosion has occurred, and half-life of tritium is 12.3 years, if the average value of the observation of Fourré et al. (2006) was 1.8 Bq/L, the tritium activity should be 0.42 Bq/L after about two half-lives, which is on the same level as our current measured value. It has also been shown that the tritium activity in the Arctic region is less than 1.0 Bq/L.

Therefore, we conducted electrolytic enrichment of several subsequent samples of Arctic seawater (with an electrolytic volume of 350 mL). Of the 84 samples we measured, 92% were above the detection limit (the activity of this samples ranged from 0.10 Bq/L to 1.44 Bq/L with an average of  $(0.30 \pm 0.24)$  Bq/L), while for the other 8% of the samples, 700 mL will be used for the electrolysis in subsequent research, which will help us further reduce the MDA to 0.05 Bq/L.

#### 4 Conclusions

A method of measuring the tritium in seawater was established using electrolytic enrichment and ultra-low background liquid scintillation in this study. The various factors affecting the detection limit were evaluated, including the counting time, the electrolytic volume of the water sample, and the selection of the background water and scintillation solution. The results show that the lowest detection limit is reached when the samples are counted for 1 000 min. The background water is a key factor influencing the detection limit. After optimizing the parameters, the detection limit of the method was as low as 1.07 Bq/L. The feasibility of this method was demonstrated by comparing our measurements with IAEA measurements of seawater samples, showing only 0.9% of the relative deviation from the average. An electrolytic enrichment step was needed to further reduce the detection limit of the tritium measurements of the seawater from Arctic regions. For example, 350 mL of Arctic seawater samples were electrolyzed for the tritium measurements, which allow 0.10 Bq/L of tritium to be detected. Of the 84 Arctic seawater samples measured, 92% were above this limit. In future research, the volume of the electrolytic samples will be increased to further reduce the MDA.

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