Optimization for tritium radioactivity analysis in rain water by liquid scintillation counter

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1. Introduction

About 600 kg Tritium (^{3}H also written as T) has been generated in the atmosphere from 1952 to 1962 due the nuclear tests performed during the same period[1]. In 1963, ^{3}H content in the rainwater reached 10,000TU (Tritium Unit). However the Nuclear Test Ban Treaty reduced the ^{3}H content in the atmosphere greatly, to the environmental level currently. ^{3}H generated artificially (e.g. ^{3}H from nuclear facilities) will be finally distributed on a global scale[2]. However it will be first distributed in the working environment and local environment, thus it is essential to consider the radiation exposure for the foregoing reason. The dependency on nuclear power is increasing worldwide and the amount of ^{3}H to handle is increasing due to the operation of heavy water reactor and nuclear reprocessing facilities. Moreover the commercialization of fusion reactor will cause the amount of ^{3}H discharged into the atmosphere to increase.

At present, tritium in the environment is equal to the previous background concentrations. Taking into account the tritium low level in the environment, even if the method for ^{3}H analysis is now routine, special conditions have to be fulfilled in order to obtain accurate and reliable tritium measurements. The activity concentration of natural water samples is so low, that for obtaining accurate measurements, any counting system must have a high and stable over all efficiency for the detection of low energy beta particles together with a low and stable background [3].

The aim of study was to investigate the effect of vial type, combination of scintillant/vial for measured sample, comparison of few different scintillation cocktails in our routine measurements according to its efficiency, figure of merit (FOM) value, background and minimal detectable activity (MDA).

2. Methods and Results

2.1 Procedure

The rain water was distilled to remove impurities before the dilution procedure of sample and scintillation cocktail. Distillation was performed by adding 0.5 g of $KMnO_4$ and 0.5 g of Na_2O_2 into 500 ml of the rain water and heating the mixed rain water. The first 5 ml of distillate was discarded and 350 ml of distilled rain water was collected. 8 ml of rain sample was mixed with each 12 ml scintillation cocktail of Ultima Gold LLT, OptiPhase HiSafe 2, or OptiPhase HiSafe 3. All samples were stored for 24 hours in a dark cool place before measurement.

2.2 Cocktails

Ultima Gold LLT, determine low levels of 3H in a wide range of water samples without requiring distillation. It accepts up to 54% tap water, river water, rain water, and even sea water, with ^{3}H counting efficiencies of approximately 30% and with very low background levels. *OptiPhase HiSafe 2* is a general-purpose liquid scintillation cocktail. It combines very high counting efficiency with moderate to high sample holding capacity for a wide range of aqueous and non-aqueous solutes. *OptiPhase HiSafe 3* is a liquid scintillator that handles a broad range of solutes. Used for a variety of scintillation applications, it combines good counting efficiency with a very high level of sample acceptance, particularly for high ionic strength solutes.

2.3 Vials

In this study we used three vial types. 20 ml *teflon vials*, 20 ml *glass vials* (low potassium borosilicate glass), which provide advantage of transparency for cocktail, and 20 ml *plastic vials* (low diffusion polyethylene antistatic vials) whi are permeable to benzene, toluene, and xylene based cocktails, resistant to diffusion with environmentally friendly cocktails (Ultima Gold LLT, Opti-Fluor etc.)

2.4 Standard quenching parameter

A quenching calibration curve method was applied to determine the detection efficiency. Eight standard tritium samples with different quenching levels were prepared. The same tritium concentration was used in all the samples. (figure 1)

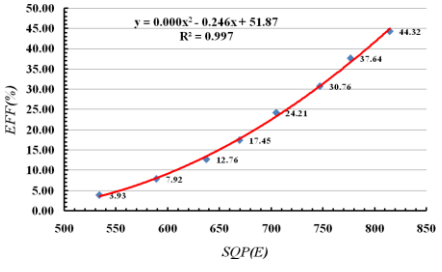


Figure 1. Efficiency dependence of SQP(E) parameter.

2.5 Expression

The sample activity was obtained by the following expression:

$$A(Bq/ml) = \frac{R_s - R_b}{60 \cdot \varepsilon \cdot V}$$

Where R_b is is the net count rate, in pulses per minute, of the tritium- free water, V is the sample volume.

The inaccuracy due to statistical nature of radioactive decay and background radiation is evaluated using a confidence level of 95% ($k=2$) using the following expression:

$$\sigma(Bq/ml) = \frac{2}{60 \cdot \varepsilon \cdot V}\sqrt{\frac{R_s + R_b}{t}}$$

Where t is the summed counting time, in minutes, of the tritium-free water, equal to the summed counting time of the sample counting vials.

L_D is expressed as counts per minute; in order to transform this value in minimum detectable activity, which can be directly compared with the measured activity of the sample, the following equation can be applied:

$$MDA = \frac{2.71 + 4.64 \cdot \sqrt{R_b \cdot t}}{60 \cdot \varepsilon \cdot V \cdot t}$$

The critical level defined by Currie (1968) is the one used to establish if measured sample is distinct from the background. The formula for the critical level is

$$L_c = 1.64 \cdot \sqrt{\frac{2R_b}{t}}$$

If $R_s - R_b > L_C$ with a probability error of 5%, the measured rate include a sample contribution. In that case the tritium activity of the sample can be calculated.

2.6 Results

- Background values

Background values of the glass vials were almost 7 time higher than those of plastic vials. It is clear that a glass vial surely contains some radioactivity. It is likely a result of the presence of natural ^{40}K which emits X-rays from ^{40}Ar, a ^{40}K decay product.

Table 1. Background values in tritium for three kinds of vials.

Cocktail	Teflon vials			Glass vials			Plastic vials		
	cpm	*SQP(E)*	*efficiency*	*cpm*	*SQP(E)*	*efficiency*	*cpm*	*SQP(E)*	*efficiency*
U-LLT	1.24	768.84	35.74	7.84	765.71	35.17	1.09	763.16	34.68
O-HS2	0.71	763.77	34.79	6.85	764.97	35.02	0.58	762.48	34.55
O-HS3	1.14	746.91	31.59	7.50	751.09	32.38	1.04	746.73	31.56

**U-LLT: Ultima Gold LLT, O-HS2: OptiPhase HiSafe 2, O-HS3: OptiPhase HiSafe 3*

- Effect of cocktail quantity and vial types

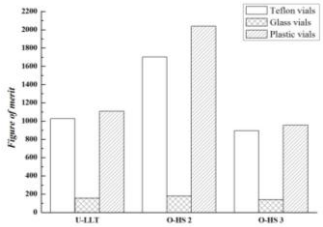
Table 2. Comparison of count rates(cpm) for three different scintillation cocktails and three types of 20 ml vials with rain water samples.

Samples	Cocktail								
	Ultima Gold LLT			OptiPhase HiSafe 2			OptiPhase HiSafe 3		
	Teflon vials	*Glass vials*	*Plastic vials*	*Teflon vials*	*Glass vials*	*Plastic vials*	*Teflon vials*	*Glass vials*	*Plastic vials*
#1	1.403	8.255	1.061	1.114	5.838	0.663	1.269	7.339	1.089
#2	1.341	8.209	1.125	0.911	6.418	0.746	1.045	7.384	1.039

- Figure of merit

An important parameter which describes a measurement quality of liquid scintillation method is a figure of merit. It combines the information about the efficiency and background and is described by equation below:

$$FOM = \frac{(Efficiency[\%])^2}{Background[cpm]}$$



- Tritium activity concentration

Table 3. Tritium activity concentration in sample(three different scintillation cocktails and three types of 20 ml vials with rain water samples.).

Cocktail	Vial types													
	Teflon vials					Glass vials					Plastic vials			
	A	*±σ*	*MDA*	*Rs-Rb*	*Lc*	*A*	*±σ*	*MDA*	*Rs-Rb*	*Lc*	*A*	*±σ*	*MDA*	*Rs-Rb*	*Lc*
U-LLT	0.93	0.55	0.91	0.16	0.15	2.44	1.38	2.26	0.41	0.37	-0.15	0.51	0.87	-0.03	0.14
O-HS2	2.65	0.48	0.73	0.43	0.11	-6.72	1.36	2.35	-0.10	0.35	0.50	0.41	0.69	0.08	0.10
O-HS3	0.99	0.57	0.94	0.16	0.14	-1.07	1.43	2.39	-0.17	0.37	0.31	0.57	0.95	0.05	0.14

**U-LLT: Ultima Gold LLT, O-HS2: OptiPhase HiSafe 2, O-HS3: OptiPhase HiSafe 3*

3. Conclusions

To evaluate ^3H concentration in the atmosphere more accurately compared to the conventional methods, the author of this paper intended to suggest more improved analytical methods and derived the elements which might occur during analysis or required improvements.

The method suggested in this study is able to reduce the uncertainty and errors which may be existent in evaluating the ^3H concentration of environmental sample s and thus will serve as the best solution in the technical and economic point of view.

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