TRITIUM CONCENTRATION ANALYSIS IN ENVIRONMENTAL WATER SAMPLES OF CENTRO NUCLEAR ARAMAR

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ABSTRACT

The experimental results are obtained with liquid scintillation technique for the measurement of tritium (^3H) radiation background in surface water samples collected in Centro Experimental Aramar and surroundings , from 1990 to 1999. This centre is subordinated to Centro Tecnologico da Marinha (CTMSP – São Paulo – Brazil) located in S. Paulo city ,a military research organisation whose objectives are to develop nuclear and energy systems for Brazilian naval ship propulsion. The estimated background average tritium concentration in this region is (25.9 ± 2.1) Bq / L . This value is compared with tritium concentration limit in drinking water established by E.P.A (Environmental Protection Agency - U.S.A), and indicate a low natural background tritium radioactivity in Centro Experimental Aramar and region.

Keywords : tritium, liquid scintillation, water samples

I. INTRODUCTION

Tritium, the heaviest isotope of hydrogen, is being naturally formed due to cosmic ray interactions ,in nuclear reactors by ternary fission and by activation of light elements such as boron and lithium. The ^3H physical half-life is 12.3 years and its activity is calculated from the beta decay with a maximum energy of 18 keV of this nuclide [1] Therefore , nuclear facilities release small amounts of tritium into the surrounding environment in the vapor and liquid form, contributing to the human annual average radiation dose [1] .The tritium concentration limit in drinking water established by E.P.A (Environmental Protection Agency - U.S.A) is 740 Bq / L [2] .

The Centro Tecnológico da Marinha (CTMSP) is a military technological research and development centre, located in São Paulo and Iperó (Centro Experimental ARAMAR) cities, whose objectives are to develop nuclear and energy systems for Brazilian naval ship propulsions programmes [3, 4] .

The investigation of background tritium concentrations in samples of the Centro Experimental ARAMAR and region is of great significance due to an industrial nuclear research programme is being developed in this centre [3, 4] . The tritium measurements were performed in addition to the Environmental Monitoring Programme carried out by the Radioecological laboratory in this region [5,6,7]. This Environmental Monitoring Programme has been conducted by the Nuclear Safety Department (CTMSP) and is being systematically carried out in this centre and region by collecting and analysing soils, fish, grass, water, milk, harvest and air samples in 124 environmental stations, since 1988 [4, 6] . This study provides a reference level for the purposes of comparative monitoring and , specifically , knowledge of radioactive concentrations is a basic task in determining the background levels, transfer and dosimetry implications.

II. MATERIALS AND METHODS

Surface water samples from environmental stations have been collected and analysed sistematically, since 1990 (2^{nd} semester), by using a LS-5801 Beckman liquid scintillation detector system [5] .

The liquid scintillation measurement is a well known technique employed for several kinds of analysis [8 ,9]. The abbreviated procedure adopted for ^3H activity measurement by using this technique is described below :

- Surface water samples are collected in 14 environmental stations , distributed inside (two sample points) and in the vicinity of this nuclear research center (twelve sample points) ;
- Reagents : all the reagents used were of analytical grade: sodium hydroxide (NaOH) , potassium permanganate ($KMnO_4$) and the liquid scintillation cocktail was Ultima Gold XR from Packard Co.. U.S.A ;
- Equipment: Beta counting was performed with a Beckmann (model LS-5801) liquid scintillation spectrometer, using polyethylene vials ;.
- Sample Preparation: From the surface water collected samples with volumes usually higher than one liter, were separated 20 mL and placed in a distillation glass of 50 mL .Following ,0.1 g of sodium hydroxide and 0.1 g of potassium permanganate were added and the distillation process ocurred until the temperature (100 – 105) °C . Then, 10 mL of the solution is separated for counting analysis.The same process is repeated for background estimations by using de-ionized water samples ;

- Sample Counting: In a polyethylene scintillation vial , 10 mL of Ultima Gold XR and 1 mL of the distillated sampled were added. Then , the samples were counted during 100 minutes. Establishment of counting regions was made by taking into account the quench level of the samples. Previously, the tritium spectrum was calibrated using BECKMAN / SPECTRUM ANALYSIS software calibration , by measuring a set of ^3H standards [(761 ± 5%) d ps / activity in 01 / 02 /1991] with different quenching levels (called #H number).

The frequency of sampling and analysis has been not the same along the years, owing by technical conditions. In the last three years only one ^3H determination per year has been done. The environmental stations localization for tritium sampling are given in references [3,4,5,6] . Those stations are located mainly nearby Sorocaba and Ipanema rivers.

The scintillation system calibration has been periodically checked by participating in a National Intercomparison Programme (P.N.I.) for tritium samples, conducted by Secondary Standard Dosimetry Laboratory (IRD / CNEN / BRAZIL)[10].

III. RESULTS AND DISCUSSION

Measurement of the ^3H standards allowed us to obtain the efficiency curve shown in figure 1.

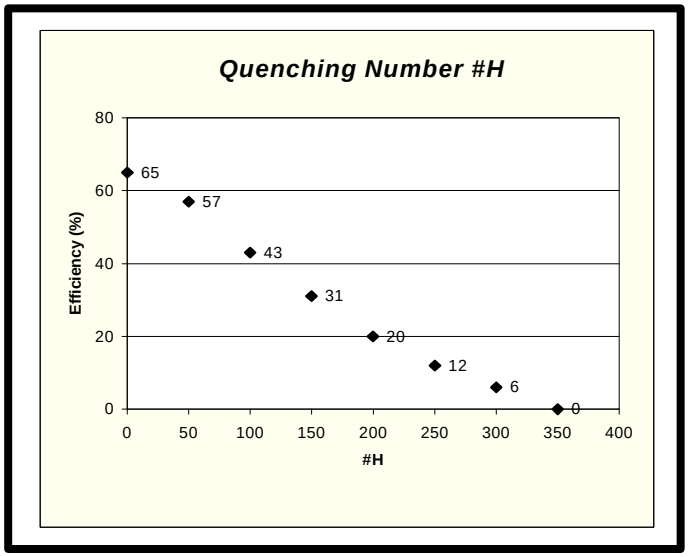

Figure 1: tritium detection efficiency as a function of Quenching number #H

The reproducibility of counting efficiency has been obtained earlier and its value is 1 % , and the low level detection limit (L.L.D.) of this technique was calculated as being 14.8 Bq / L, by analysing periodically background samples [7].

The comparative results obtained in the National Intercomparison Programme for tritium samples analysis are shown in Table 1 as a function of the period of analysis. All the results obtained are in good agreement with the average and standard deviation values obtained by other laboratories with the same samples [10] .

The calculated values of tritium concentrations (Bq / L) obtained for each one of the 14 environmental stations in Centro Experimental Aramar are given in Table 2 as a function of the sampling period and analysis. The average values and respective standard deviations for the period from 1990 to 1999 , for each environmental point , are also given. Standards deviations ,calculated for each sample point were about 20 -30 % indicating reasonable homogeneous data along the years, as can be also observed in the average measured values of tritium concentrations Bq / L per year , obtained in each environmental station ,. The range of these average values is (22.9 – 31.0) Bq / L, and the total average and standard deviation is (25.9 ± 2.1) Bq / L , as shown in the last row of Table 2. The low standard deviation value indicates homogeneous data , or tritium concentration values practically the same for all environmental station points. Another noteworthy finding is that the average value obtained in the two points located inside the center (2 and 3) and in the 12 points located outside the center, are practically the same , respectively (27.9 ± 4.4) Bq / L and (25.6 ± 1.6) Bq / L.

TABLE 2 – Tritium concentrations in the 14 environmental stations as a function of the collected period,L,Ld, indicates values lower than 14,8 Bq/l, Values in black mean lost data, Average value is obtained for each environmental point, Total average value is obtained by taking into account the 14 environmental point average data

Year	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11	#12	#13	#14
1990	16,4	LLD	32,3	15,8	LLD	28,1	15,9	LLD	40,5	LLD	40,1	20,7	LLD	
1991	37,7	35,9	21,8	LLD	26,1	24,3	19,5	24,7	25,7	LLD	20,1	28,5	25,8	
1992	25,0	29,2	27,0	33,0	38,1	39,2	33,7	32,9	48,5	33,7	26,3	46,2	39,6	44,4
1993	30,3	30,0	22,5	27,2	27,2	22,0	23,2	22,5	27,3	19,8	26,3	23,5	23,5	21,7
1994	30,8	31,8	25,3	26,7	32,7	26,8	22,3	25,7	25,8	33,6	26,8	30,8	31,1	20,2
1996	24,5	39,2	33,4	26,5	15,5	36,5	30,8	28,6	27,9	31,7	21,3	27,2	32,2	LLD
1997	26,2	28,5	23,4	23,5	25,6	21,7	24,7	23,5	21,4	22,0	21,6	25,2	24,5	20,8
1998	23,2	29,4	20,8	25,4	24,5	16,8	23,4	22,5	15,0	21,4	16,3	24,5	25,2	20,1
1999	25,7	26,4	20,9	27,2	22,0	19,4	25,9	25,8	16,3	20,2	18,4	21,0	27,3	22,1
Average	28,0	31,0	24,8	26,8	27,2	25,3	23,5	24,6	27,2	23,9	24,7	26,3	26,4	22,9
Standard	7,8	9,1	6,9	6,2	6,3	7,4	6,6	8,0	8,9	7,0	8,3	7,4	6,1	6,7
Total Average							(	25,9	+/-	2,1	)	Bq/L		

TABLE 1 – Results of tritium concentrations (Bq / L) obtained by Laboratorio Radioecologico (CTMSP) in samples prepared by the Secondary Standard Dosimetry Laboratory (IRD- BRASIL) .

YEAR	LARE (Bq/L)	IRD (Bq/L)
APRIL97	501 ± 12	423 ± 85
AUGUST97	278 ± 12	228 ± 46
DECEMBER97	516 ± 12	493 ± 99
APRIL98	300 ± 12	300 ± 60
DECEMBER98	385 ± 16	399 ± 80
APRIL99	265 ± 3	234 ± 47
AUGUST99	134 ± 8	152 ± 30
DECEMBER99	518 ± 2	416 ± 83
APRIL2000	483 ± 18	408 ± 82

IV. CONCLUSIONS

Tritium activities (Bq / L) in surface water samples were measured in the period from 1990 to 1999 entro Experimental Aramar by using the liquid scintillation technique..Fourteen environmental stations were studied and the results obtained provide an average of (25.9 ± 2.1) Bq / L [11]. This average value obtained is compared with the tritium limit concentration in drinking water of 740 Bq / L. established by E.P.A (U.S.A) [2] , indicating a low natural tritium concentration in waters of Centro Experimental Aramar and region.

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