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Tritium concentration analysis in environmental water samples of centro nuclear ARAMAR (CTMSP—Brazil)

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Abstract

The experimental results obtained with liquid scintillation technique for the measurement of tritium (^{3}H) radiation background in surface water samples collected in Centro Experimental ARAMAR and surroundings, from 1990 to 1999 are described. This centre is subordinated to Centro Tecnologico da Marinha (CTMSP—Sao 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 Environmental Protection Agency—USA, and indicate a low natural background tritium radioactivity in Centro Experimental ARAMAR and surrounding region is indicated.

Keywords: Tritium; Liquid scintillation; Water samples

1. Introduction

Tritium, the heaviest isotope of hydrogen, is naturally formed due to cosmic ray interactions in nuclear reactors by ternary fission and by the activation of light elements such as boron and lithium. The ^{3}H 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 (IAEA, 1975). Therefore, Nuclear facilities release small amounts of tritium into the surrounding environment in the vapour and liquid form, contributing to the human annual average radiation dose (IAEA, 1975). The tritium concentration limit in drinking water established by (Environmental Protection Agency—USA (EPA) is 740 Bq/l (ASTM, 1995).

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 propulsion programmes (Hiromoto et al., 1988; LARE, 1999).

The investigation of background tritium concentrations in samples from the Centro Experimental ARAMAR and surrounding region is of great significance because an industrial nuclear research programme is being developed in this centre (LARE, 1999; CTMSP, 1997). The tritium measurements were performed in addition to the Environmental Monitoring Programme carried out by the Radioecological

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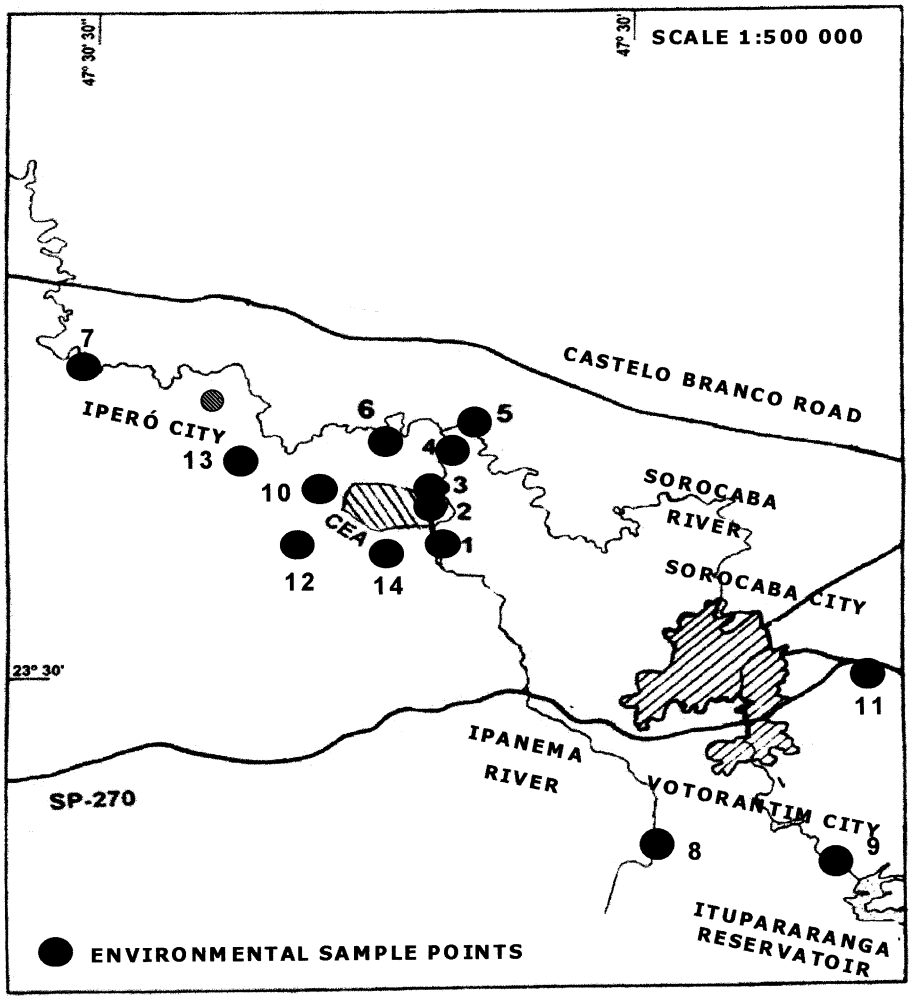

Fig. 1. Environmental sample points.

laboratory in this region (CTMSP, 1997; Sartoratto, 1995; Moraes and Daltro, 2000). This Environmental Monitoring Programme has been conducted by the Nuclear Safety Department (CTMSP) and is being systematically carried out in this centre and surrounding region by collecting and analysing soils, fish, grass, water, milk, harvest and air samples in 124 environmental stations, since 1988 (Hiromoto, 1988; Moraes and Daltro, 2000). This study provides a reference level for the purposes of comparative monitoring and, specifically, knowledge of radioactive concentrations as a basic task in determining the background levels, transfer and dosimetry implications.

2. Materials and methods

Surface water samples from environmental stations have been collected and analysed systematically, since 1990 (second semester), by using a LS-5801 Beckmann liquid scintillation detector system (CTMSP, 1997).

The liquid scintillation measurement is a well-known technique employed for several kinds of analyses (Pane, 1978; EPA, 1975). The abbreviated procedure adopted for ^{3}H 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 centre (12 sample points).
- Reagents: all the reagents used were of analytical grade: sodium hydroxide (NaOH), potassium permanganate ($KMnO_4$) and the liquid scintillation solution was Ultima Gold XR from Packard Co. USA.
- Equipment: Beta counting was performed with a Beckmann (model LS-5801) liquid scintillation spectrometer using polyethylene vials.

- Preparation of samples: from the surface water samples collected with volumes usually higher than 1 l, separated 20 ml was 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 was carried out till the temperature range of 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 sample were added. Then the samples were counted for 100 min. Establishment of counting regions was made by taking into account the quench level of the samples. Previously, the tritium spectrum was calibrated using BECKMANN/SPECTRUM ANALYSIS software calibration, by measuring a set of ^{3}H standards [(761 ± 5%) dps/activity on 01/02/1991] with different quenching levels (called #H number).

The frequency of sampling and analysis has not been the same along the years, owing to technical conditions. In the last 3 years only one ^{3}H determination per year has been done. Localization of the environmental stations for tritium sampling are given in Fig. 1. Those stations are located mainly near Sorocaba and Ipanema rivers.

The scintillation system calibration has been periodically checked by participating in a National Intercomparison Programme (PNI) for tritium samples, conducted by Secondary Standard Dosimetry Laboratory (IRD/CNEN/BRAZIL) (IRD, 1999).

3. Results and discussion

Measurement of the ^{3}H standards allowed us to obtain the efficiency curves shown in Figs. 2 and 3.

The reproducibility of counting efficiency has been obtained earlier and its value is 1%, and the low level

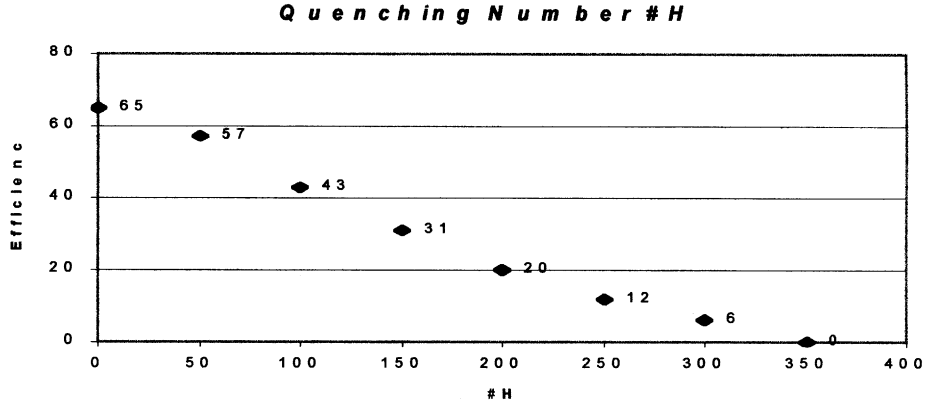

Fig. 2. Tritium spectrum obtained with spectrum analysis software in Beckman LS-5801 liquid scintillation equipment.

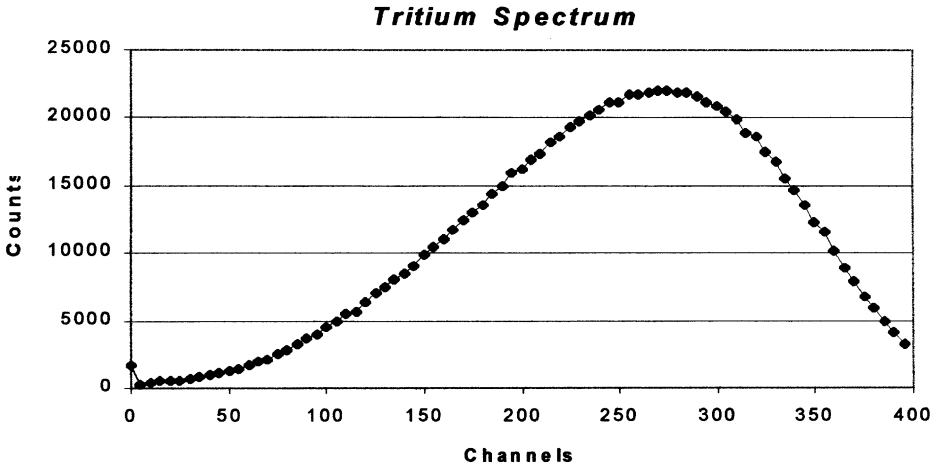

Fig. 3. Tritium detection efficiency as a function of quenching number #H.

Table 1
Results of tritium concentrations (Bq/1) obtained by Laboratorio Radioecologico (CTMSP) in samples prepared by the Secondary Standard Dosimetry Laboratory (IRD-Brazil)

Year	LARE (Bq/l)	IRD (Bq/l)
April 97	501 ± 12	423 ± 85
August 97	278 ± 12	228 ± 46
December 98	516 ± 12	493 ± 99
April 98	300 ± 12	300 ± 60
December 98	385 ± 16	399 ± 80
April 99	265 ± 3	234 ± 47
August 99	134 ± 8	152 ± 30
December 99	518 ± 2	416 ± 83
April 2000	483 ± 18	408 ± 82

Table 2
Tritium concentrations in the 14 environmental stations as a function of the collected period

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	26.7	LLD	20.1	28.5	25.8	—
1992	25.0	29.2	27.0	33.0	38.1	39.2	83.7	32.9	48.6	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														

LLD indicates values lower than 14.8 Bq/l. Values with dash 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.

detection (LLD) limit of this technique was calculated to be 14.8 Bq/l, by analysing background samples (Sartoratto, 1995).

The comparative results obtained in the National Intercomparison Programme for tritium sample 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 (IRD, 1999).

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 which are practically the same for all environmental station points. Another noteworthy finding is that the average values obtained in the two points located inside the centre (2 and 3) and in the 12 points located outside the centre, are practically the same, respectively 27.9 ± 4.4 and 25.6 ± 1.6 Bq/l.

4. Conclusions

Tritium activities (Bq/l) in surface water samples were measured in the period from 1990 to 1999 in Centro 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. This average value obtained is compared with the tritium limit concentration of 740 Bq/l established in drinking water by EPA (USA) (ASTM, 1995), indicating a low natural tri-

tium concentration in the waters of Centro Experimental ARAMAR and surrounding region.

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