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Biological and Chemical Research, Volume 3, 191-199 | Science Signpost Publishing

Procedure for Determining of Tritium Concentration in the Hanoi Atmosphere

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Received: April 08, 2016 / Accepted: May 05, 2016 / Published: July 25, 2016

Abstract: Environmental radiation monitoring around nuclear power plants is a mandatory task, an indispensable content of any nuclear power plants yet. The radiation measurement techniques using for environmental radioactivity monitoring are continuously being improved. One of the radioisotopes that should be regularly monitored is tritium (H-3), especially in the atmosphere. Tritium presents in the atmosphere mainly in the vapor (HTO) and gaseous (HT) forms. In this study the HTO and HT in the Hanoi atmosphere are collected simultaneously by the MARC-7000 Tritium sampler. The sampler was specifically designed for capturing Tritium by using a series of four bubbling bottles, a cooling system and an oven packed with Palladium-Alumina catalyst to oxydize HT. Sampling was carried out a flow rate of 30 L/h continuously for 10 to 14 days which depends on the atmosphere humidity. The trapping yields of HTO and HT were higher than 99% and 96%, respectively. The collected atmospheric moisture samples have been enriched by electrolysis then were measured for tritium activity on a Liquid Scintillation Analyzer Tri-carb 3170 TR/SL. Counting time was 100min x 10 cycles to achieve an accounting precision of less than 10%. The detection limit of the procedure was estimated as low as 0.46 TU. Average tritium radioactivity in the Hanoi atmosphere was found to be 16.94 (mBq/m^3).

Keywords: HTO and HT in the atmosphere, MARC-7000 Tritium sampler, TRI CARB 3170 TR/SL LSA.

1. Introduction

Tritium is the radioactive isotope of hydrogen. Tritium exists in various forms in atmospheric plumes released from nuclear facilities, the most important being tritiated water (HTO) and tritiated hydrogen gas

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(HT). The relative proportion of these various forms of tritium in air around a facility depends on that released from the facility, and also on atmospheric chemical processes, the most important being the water vapor content of air (Momoshima et al., 2007) [1]. Because water vapor saturation is temperature dependent, the water vapor content and consequently the conversion of HT to HTO in the summer will be higher than in the winter.

The form of tritium is important to both environmental partitioning and radiological dose impacted to people living around a facility. Gaseous HT reacts more slowly than HTO with other environmental compartments, and impacts a very low radiological dose relative to HTO because it is only weakly absorbed by the body. The HTO behaves like water in the atmosphere, partitions easily to soil and plants, and impacts a larger radiological dose in the body. As noted by Spencer and Vereecken-Sheehan (1994) [2], HTO is usually the more abundant chemical form in plumes arising from nuclear facilities.

Vietnam is preparing to build nuclear power plants in Ninh Thuan province. Before building nuclear power plants, it's necessary to set up a data base of environmental radiation background. We need to know initial radiation background data to monitor their movements due to the operation of the plant and compliance with safety radiation standards. In fact, some radioactive isotopes such as U, Ra, Th, K-40, Be-7, Cs-134, Cs-137, Sr-90, Ra-226, Rn, ... in the other objects are currently also being analyzed. However, in Vietnam, up to now there is no study of ^{3}H in the air.

In this paper a procedure for determining tritium content in the Hanoi atmosphere is presented.

2. Instruments and Procedure

1. Tritium sampler: Tritium sampler, MARC 7000

Tritium sampler, MARC 7000, is designed and built by SDEC France following ISO 2889 (Fig 1). The tritium sampler can trap tritium in the environment (gas and vapor forms) quickly but this trapping depends on the quantities extracted during the sampling time. Thus, when working in a hot environment (ambient temperature above $+25^{0}C$), we strongly recommend to use a sampler fitted with a "cooling" system. By reducing the losses of water in the bottles (by evaporation), the trapping yield will increase and almost tritium in the sampled air will be trapped.

Figure 1. Tritium sampler, MARC 7000

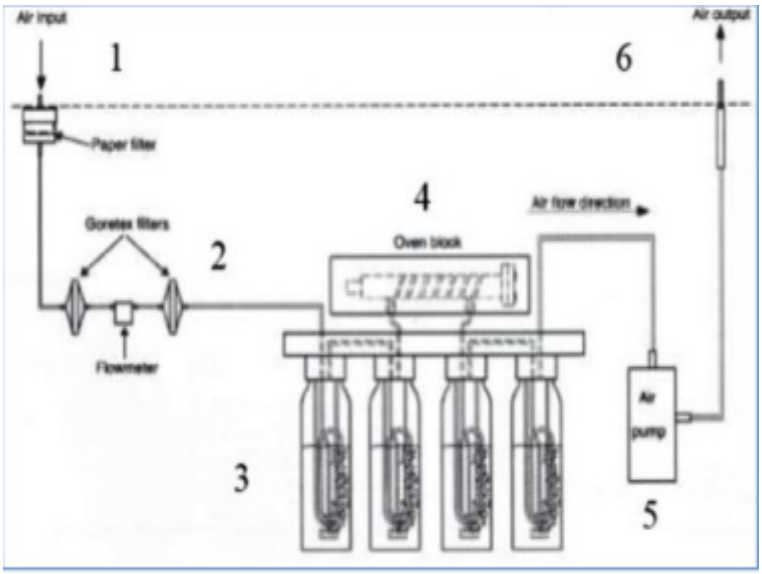

Figure 2. A block diagram of tritium sampler

where:

1: air inlet with 6.4mm diameter connected sampling tube. It can be easily take off the cylinder part and see a filter paper supported on wire grid (Ø=45mm).

2: air flow meter can be adjusted from 10 to 50(L/h),

3: four sampling bottles with 250mL capacity,

4: oven block has a tubular form in stainless steel equipped with a catalyst cartridge (palladium on alumina). The oven temperature can be adjusted from 200^0C to 500^0C. Under the oven block, a cooling system is designed to avoid the evaporation of the demineralized water contained in the bottles. The bubbling bottles temperature must be controlled and maintained on average 7^0C for an ambient temperature of 20^0C.

5: air pump go through the system,

6: air outlet.

2. Tritium electrolytic enrichment

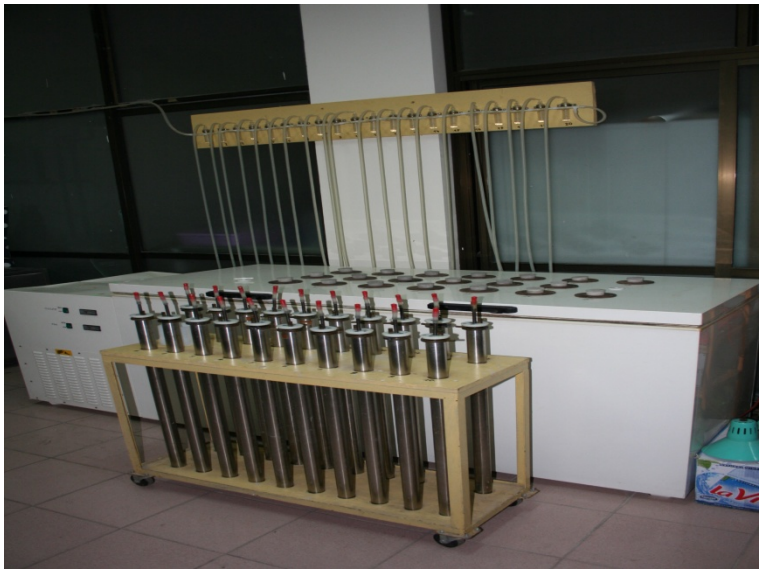

Figure 3. Electrolytic enrichment system

To increase the sensitivity of H-3 analysis, samples have been enriched by electrolysis. The isotope fractionation due to mass differences between H-1 and H-3, resulting differences in physical chemical properties and increase 3H concentration in the samples after electrolysis.

3. Liquid scintillation analyzer system

The Tri-Carb 3170TR/SL is a computer-controlled benchtop liquid scintillation analyzer, specially configured for detection of extremely low level alpha and beta radioactivity with BGO detector.

Figure 4. LSA system, Tri-Carb 3170TR/SL

4. Procedure for determining tritium in the Hanoi atmosphere

Reagents, equipment and instruments: Sodium peroxide, Na_2O_2; lead oxide, $PbCl_2$, Ultima Gold LLT; Spike solution (SPK), demineralized water (DIW), dead water (DW)…Tritium secondary standards, MARC-7000; Electrolytic enrichment system; Liquid Scintillation Analyzer System, Tri-Carb 3170TR/SL.

Sampling procedure by MARC-7000 :

- Fill up the sampling bottles with DW.
- The bubbling bottles must be filled with 150ml DW.
- Fix the four bottles in their units, switch on the Sampler.
- Set the airflow at 30L/h,
- Switch on the Cooling System,
- Switch on the Oven,
- The last switch on the air Pump.
- After sampling, the air pump was switched off
- The sampling bottles were removed.
- The collected samples mixed,
- Taken out 500 ml approximately for electrolysis enrichment.

Tritium electrolytic enrichment: System of electrolytic enrichment consists of 20 cells of 500 ml volume (anodes: stainless steel, cathodes: mild steel). The method is described in 4 steps: sample treatment, electrolytic enrichment, sample neutralization, and measurement by LSC. The sample treatment consists of water distillation to remove any impurities and interfering radionuclides to reduce quenching. The first step of sample pretreatment is primary distillation. The electrolytic enrichment is carried out by adding 1,5 g of sodium peroxide (Na_2O_2) to each 500-mL water sample aliquot. The sodium hydroxide (NaOH) formed is used as an electrolyte. For each enrichment run 8 spike and 8 dead-water (DW) samples are used for system control. Enrichment procedure lasts for 164 hours (1426 Ah). Finally, the sample is neutralized by addition 6 g of lead chloride ($PbCl_2$) and a new distillation is performed in order to separate the lead oxide

(PbO) and other impurities from the water. Final volume of water sample after electrolysis is higher 12g water, and the enrichment factor is 31.54±0.61.

Sample measurement by LSC system, Tri-carb 3170TR/SL: Mixture of 10 ml of water and 10 ml of scintillation cocktail Ultima Gold LLT in plastic vials is used for counting in LSC. The measurement run contains 12 samples, 3 standards, 3 DW samples. The limit of detection is 0.46TU. Counting is performed for 10 cycles of 100 min and the tritium activity is calculated for each sample by averaging the counting values.

The concentration activity of tritium at the counting time is calculated using the following expression:

$$A_T = \frac{N_{SA}.A_{ST}}{N_{ST}.Z_I}$$

where

A_T: concentration activity of tritium at the counting time (Bq/L)

N_{SA}: net count rate of the sample (cpm).

N_{ST}: net count rate of the standard (cpm).

A_{ST}: tritium activity of the standard at the counting time (TU).

Z_I: enrichment factor.

3. Results and Discussion

1. Sampling result

Table 1. Sampling result of tritium in the Hanoi atmosphere

Name	Sampling time	Time (h)	V (m3)	HTO (ml)	HT(ml)	HTO+HT (ml)	RH (%)
H1	Oct. 18-31, 2013	330.20	9.88	330	310	640	74
H2	Oct. 31-Nov.14, 2013	330.20	10.06	372	310	682	73
H3	Nov. 14-28, 2013	334.97	9.86	330	310	640	74
H4	Nov. 28-Dec.13, 2013	355.00	10.44	308	315	623	73
H5	Dec. 13-30, 2013	405.68	11.94	299	312	611	68
H6	Dec.30, 2013-Jan.15, 2014	380.52	11.2	302	308	610	70
H7	Jan. 15-27, 2014	286.30	8.42	291	304	595	72
H8	Jan. 27-Feb. 17, 2014	507.42	14.93	326	318	644	71

H9	Feb. 17-28, 2014	269.30	7.93	315	308	623	79
H10	Feb. 28-Mar. 13, 2014	313.93	9.24	340	334	674	78
H11	Mar. 13-28, 2014	364.11	10.71	350	310	660	87
H12	Mar. 28-Apr.08, 2014	256.95	7.56	381	332	713	88
H13	Apr. 08-26, 2014	427.18	13.57	455	336	791	89
H14	Apr. 26-May.09, 2014	316.27	9.31	385	306	691	87
H15	May. 09-30, 2014	504.13	14.84	482	310	792	77
H16	May. 30-Jun. 13, 2014	330.61	9.72	437	316	753	78
H17	Jun.13-30, 2014	408.16	12.01	456	320	776	78
H18	Jun. 30-Jul. 15, 2014	375.00	10.51	442	314	756	76
H19	Jul. 15-31, 2014	386.92	11.39	459	320	779	82
H20	Jul. 31-Aug. 14, 2014	341.73	10.05	437	313	750	81
H21	Aug. 14-31, 2014	400.88	11.79	463	317	780	83
H22	Aug.31-Sep.15, 2014	361.90	10.60	434	318	752	82
H23	Sep.15-30, 2014	361.84	10.65	307	427	734	73
H24	Sep.30-Oct.15, 2014	293.41	8.91	318	364	682	75
H25	Oct.15-31, 2014	380.22	10.91	393	316	709	78

We found that amount of HTO and HT in the samples in the Rainy season (from April to September) higher than that in the Dry season.

2. Determining minimum detectable activity (MDA) of tritium in the Hanoi atmosphere

Table 2. Analysis result of dead water samples to determining MDA

No.	Name	Count rate (cpm)	Time (minute)	V(l)	ε	Z	Ld	MDA (Bq/l)	MDA (TU)
1	DW	1.10	1000.00	0.01	0.56	26.02	0.51	0.06	0.49
2	DW	1.06	1000.00	0.01	0.56	26.02	0.51	0.06	0.49
3	DW	0.90	1000.00	0.01	0.56	26.02	0.47	0.05	0.45
4	DW	0.92	1000.00	0.01	0.56	26.02	0.47	0.05	0.45
5	DW	0.87	1000.00	0.01	0.56	26.02	0.46	0.05	0.44
6	DW	0.93	1000.00	0.01	0.56	26.02	0.48	0.05	0.46
7	DW	0.82	1000.00	0.01	0.56	26.02	0.45	0.05	0.43
8	DW	0.94	1000.00	0.01	0.56	26.02	0.48	0.05	0.46

The detection limit of the procedure (MDA) was estimated as low as 0.46 TU [7].

3. Result of Tritium concentration in the Hanoi atmosphere

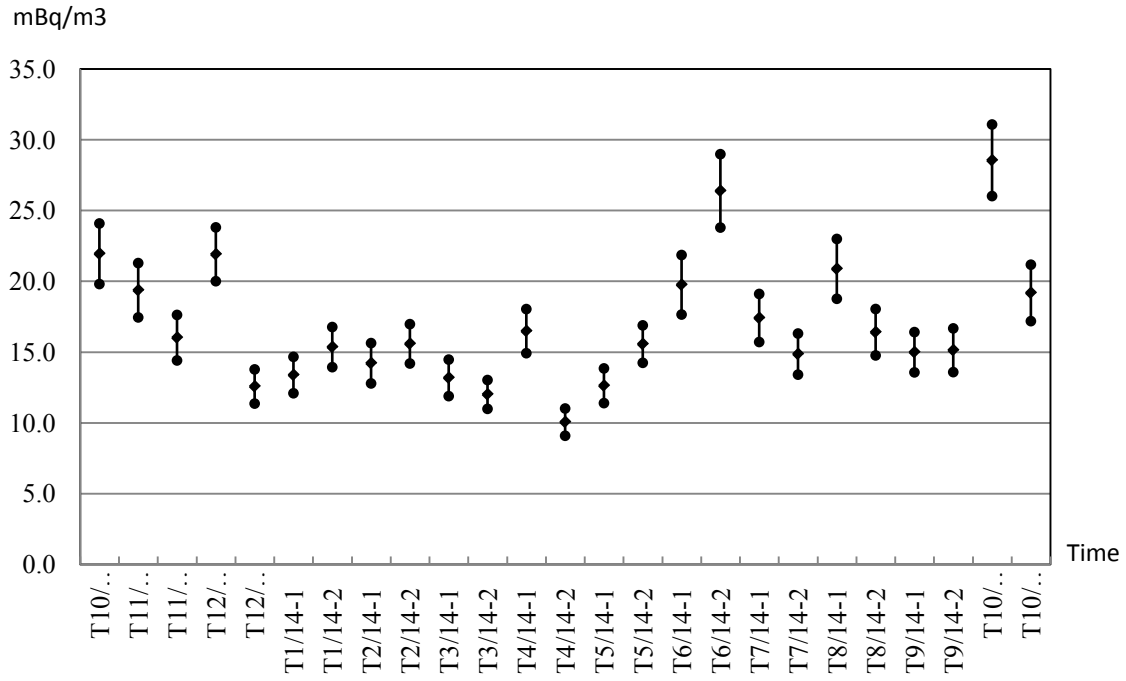

Figure 5. Relationship between Tritium activity concentration and sampling time in the Hanoi atmosphere

From figure 5 we found that the lowest and highest activity concentration of tritium was 12.61 ± 1.23 (mBq/m^3) and 28.54 ± 2.53 (mBq/m^3) in the sampling time (from April 26, 2014 to May 9, 2014) and (from September 30, 2014 to October 15, 2014).

4. Conclusion

1- The optimum tritium sampling conditions in Hanoi atmosphere for MARC-7000:

-Dead water volume in each bottle before sampling was 150ml

- Oven Temperature was 450^0C
- Cooling circuit temperature was 5^0C
- Air flow setting was 30L/h
- At least sampling time was 15 days
- At least air volume in sampling time was $9m^3$.

2- The amount of HTO and HT in the samples in the Rainy season (from April to September) higher than that in the Dry season.

- The detection limit of the procedure (MDA) was estimated as low as 0.46 TU.

- The lowest activity concentration of tritium was 12.61 ± 1.23 (mBq/m^3) in sampling time from April 26, 2014 to May 09, 2014.
- The highest activity concentration of tritium was 28.54 ± 2.53 (mBq/m^3) in sampling time from September 30, 2014 to October 15, 2014.

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