

Measurement of Carbon-14 in Air Released from a Research Reactor

IHSANULLAH

Health Physics Division,

Pakistan Institute of Nuclear Science and Technology

(PINSTECH) P.O. Nilore, Islamabad, Pakistan

(Received 3rd February, 1994.)

Summary: Measurement of ^{14}C in air, generated from a low power (300 kW) training reactor, has been carried out. A simple method has been applied using barium hydroxide for the absorption of CO_2 , followed by conversion of barium carbonate to benzene for analysis using liquid scintillation counting. Air from the reactor stack and from various sites in the immediate vicinity has been analysed for the determination of ^{14}C . In order to assess the environmental hazards to the population, the collected data has been used to calculate radiation dose due to ^{14}C .

Introduction

The long-lived radioactive isotope of carbon, ^{14}C , is a soft beta emitter with a half-life of 5730 years [1]. ^{14}C is formed naturally in the upper atmosphere by the interaction of neutrons of cosmic ray with nitrogen and, to a lesser extent, with oxygen and carbon. Artificially, large amounts of ^{14}C have also been released to the atmosphere as a result of nuclear weapons testing, nuclear reactors and preparation of labelled materials [2-4]. ^{14}C is produced in nuclear power reactors from absorption of neutrons by carbons, nitrogen, or oxygen. Carbon and oxygen can occur in the coolant (H_2O and CO_2), moderator (graphite and H_2O) or in the fuel (UO_2). ^{14}N is the major contributor to ^{14}C production because of its high neutron capture cross-section and high isotope abundance in natural nitrogen. Nitrogen can present as an impurity in the fuel or in the structural material. Different possible neutron induced reactions for production of ^{14}C alongwith their cross-sections are reported earlier [5].

The significance of the contribution of ^{14}C towards doses is due to its (a) long half-life, (b) easy access via the food chain to all key molecules of body tissues, and (c) relatively long residence times in both stratosphere and troposphere of four and eight years respectively [6]. However, because this isotope is produced in quite large quantities in reactors, it is desirable to determine the contribution of this type of contamination in the local environment [7]. The emission rate of ^{14}C from different types of nuclear reactors are given for comparison [8]. It is interesting to note that the total installed power of research in the world is

equal to one large nuclear power station [9]. However, the increased awareness about the harmful effects of radiation demands that every efforts should be made to reduce undue exposure of radiation and even the low level activities should not be neglected.

Different techniques i.e. sample combustion, solid source counting, internal gas counting, direct mass spectrometry, laser absorption spectroscopy, isotopic enrichment and liquid scintillation counting (LSC) are used for the determination of ^{14}C [3]. Amongst these, LSC is the simple, highly sensitive and the most popular technique for the measurement of ^{14}C (as CO_2) produced by various reactors [8,10-12].

In the present study, determination of ^{14}C in air from a low power training reactor, the Scottish Universities Research and Reactor Centre (SURRC), East Kilbride, Glasgow, UK, has been carried out.

Result and Discussion

SURRC houses a research reactor of the Argonaut type, the UTR (Universities Teaching Reactor) 300, which can operate at a maximum power of 300 kW (thermal). The enriched uranium fuel (~ 95% ^{235}U) is contained within two tanks in a graphite reflector, which is surrounded by a 2 m thick concrete as a biological shielding. The light water acts as the primary coolant as well as the moderator. The UTR 300 is situated far closer to large population areas i.e. East Kilbride with

population of 65,000 and Glasgow with population of 800,000 persons [5]. Therefore, monitoring of ^{14}C is important in relation to the dosimetry of the local population.

The results obtained from the present research work are given in Table 1, 2 and 3. The following observations are obtained from Table 1:

Table-1: ^{14}C concentrations in the samples

Sample No.	Date of sampling	Reactor operat.	Total air passed (l)	^{14}C concentration $\text{mBq l}^{-1} \pm 1\sigma$
Site I				
1.	27/3-20/4/90	ON	5645	0.084 ± 0.002
2.	20/4-23/4/90	OFF	5879	0.053 ± 0.001
3.	23/4-25/4/90	OFF	3113	0.061 ± 0.003
Site II				
4.	30/4-15/5/90	ON	3642	0.074 ± 0.003
5.	27/4-30/4/90	OFF	5798	0.069 ± 0.004
6.	04/5-08/5/90	OFF	5888	0.069 ± 0.004
Reactor hall				
7.	21/5-13/6/90	ON	4185	0.175 ± 0.005
8.	18/5-21/5/90	OFF	6251	0.082 ± 0.001
9.	25/5-28/5/90	OFF	6276	0.094 ± 0.001
Reactor				
10.	22/5-24/5/90	ON	1073	173.7 ± 0.9
11.	28/5-13/6/90	ON	2726	216.2 ± 1.0
12.	18/5-21/5/90	OFF	6263	2.4 ± 0.01
13.	25/5-28/5/90	OFF	6276	2.9 ± 0.001
14.	19/2-07/3/88	ON/OFF	36883	41 ± 6
15.	07/02/1990	ON/OFF	458	46 ± 6
16.	09/02/1990	ON	533	139 ± 12
Block C				
17.	03/2-08/3/90		70956	0.03 ± 0.18

Table -2: ^{14}C activity released per year of reactor stack.

Sample No.	kwhrs during sampling	mBq (total air of reactor stack, litres)	mbq per year (released)
10.	3575	0.18 (1012261)	10.6
11.	9085	0.56 (2572418)	13.3
14.	15125	1.42 (34622166)	20.3*
15.	975	0.02 (431804)	4.4*
16.	1775	0.07 (502591)	8.5

* = when reactor was ON/OFF during sampling.

Table-3: The production rate (mBq kWhr^{-1}) at different locations.

Site	Total activity (mBq)	Total power (kWhr)	mBq kWhr^{-1}	Ratio of mBq kWhr^{-1} to R. Stack
Site I	468	17700	0.026	4.7×10^{-7}
Site II	276	12015	0.023	4.1×10^{-7}
R. hall	734	13950	0.053	9.6×10^{-7}
R. stack	5.6×10^8	9085	6.1×10^4	1
	1.8×10^2	3575	4.9×10^4	1
Average, R. stack			5.5×10^4	

Sample No. 17 was run in a laboratory (Block C) to check the chemical yield. The percent chemical yield was found to be 85 ± 15 . The sample was also analysed for ^{14}C . Samples from Sites I and II showed enhancement when the reactor was in operation. Site I showed greater

activity of ^{14}C than that at Site II when the reactor was in operation and vice versa when the reactor was off. This indicates that ^{14}C might be dispersed to some distance from the release point probably due to speed and direction of wind. The stack and, to a lesser extent, the reactor hall showed small activity of ^{14}C even when the reactor was off. The wide variations in the results of samples 14 and 15 as shown in Table 2 may be explained as below:

i) large differences in the sampling time i.e. 24455 min and 305 min for samples 14 and 15 respectively, affecting the chemical yield due to losses of BaCO_3 in various steps, ii) very large differences in the time when reactor was on, iii) ^{14}C was present even after the reactor was shutdown, and (iv) if the power of the reactor is not considered in calculations, the differences are small as ^{14}C concentrations are 41 and 46 mBq l^{-1} for samples 14 and 15 respectively (see Table 1).

The total annual output of ^{14}C was calculated by taking the integrated operating power of 216000 kWhrs y^{-1} (based on four years reactor operation record i.e. from 1985 to 1988). The average activity released per year from the reactor was found to be 12 MBq for 216000 kWhrs .

It was assumed that all ^{14}C was released as CO_2 . On the basis of an average reactor power, 12 MBq y^{-1} corresponds to $0.04 \text{ TBq GW}(\text{th})^{-1} \cdot \text{y}^{-1}$, which is equal to $0.12 \text{ TBq GW}(\text{e})^{-1} \cdot \text{y}^{-1}$. Comparing this values with ^{14}C emission rate of other reactors [5,13], the production rate of the radionuclide at the UTR reactor is less than all of them except the FBR* and the HTGR** types. The levels of ^{14}C in terms of mBq kWhr^{-1} at different locations are given in Table 3 for comparison.

Radiation doses due to ^{14}C

The overall radiological impact from ^{14}C releases is assessed by means of collective dose commitment [14] using the following equation:

$$D_c = D_B \cdot Q/B$$

where D_c = the dose commitment due to release Q,

D_B = the dose rate due to natural ^{14}C ,

B = the rate of production of natural ^{14}C , and

*FBR = Reactor Fast Breeder.

**HTGR = High Temperature Gas Cooled Reactor.

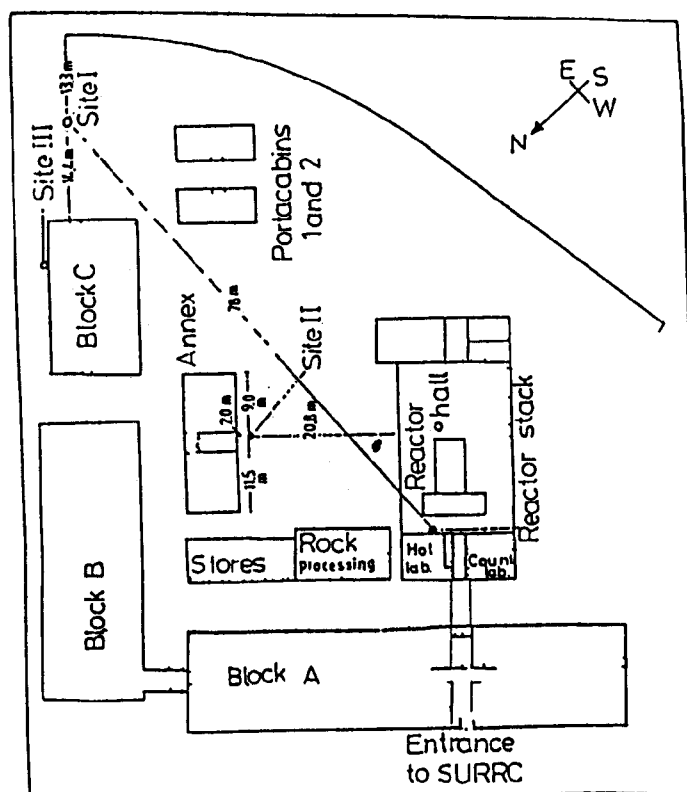


Fig. 1: Sampling sites.

Q = the activity of ^{14}C released into the environment.

A value of $1.4 \times 10^{-18} \text{ Sv. y}^{-1}$ is obtained for 12 MBq. y^{-1} .

Experimental

Sampling sites

The sampling sites i.e. the reactor stack, reactor hall, site I and site II are shown in Figure 1.

Procedure

A Dymax model 2A pump was used to draw air at a flow rate of 1.5 l/min through 2.5 litres of 0.1 M $\text{Ba}(\text{OH})_2$ solution contained in a closed container. The inlet point (through filter paper) was positioned 50 cm above the ground surface. The air through the reactor stack was again discharged in the stack after passing through $\text{Ba}(\text{OH})_2$ solution. Sampling was carried out at various locations when the reactor was in operation as well as when it was shutdown. The samples were

collected in duplicate. The BaCO_3 precipitates were filtered and dried. The BaCO_3 was converted to a suitable form for ^{14}C analysis by LSC, the following steps of conversion were carried out.

i) Preparation of carbon dioxide

The prepared BaCO_3 sample was placed in a flask and wetted with demineralised H_2O . From a specially designed dropping funnel, freshly prepared dilute HCl was added dropwise until all the carbonate had been hydrolysed. The excess CO_2 evolved was collected in liquid nitrogen.

ii) Production of acetylene

The C_2H_2 synthesis involved two steps in which CO_2 was reacted with molten Li metal to form lithium carbide (Li_2C_2), and the Li_2C_2 was hydrolysed with H_2O to yield C_2H_2 . The generated C_2H_2 , and the H_2 produced from hydrolysis of excess Li, were passed through a recirculating condenser (to retain LiOH), a P_2O_5 trap (to retain water vapour) and then into two traps cooled by liquid nitrogen for C_2H_2 collection.

iii) Conversion to benzene

Acetylene was converted to benzene by letting the frozen C_2H_2 slowly sublime on to an activated vanadium catalyst. The column was sealed and allowed to stand overnight at room temperature. Then the column was heated to $110^\circ C$ and the benzene collected under vacuum in a liquid nitrogen trap. The frozen benzene was melted at room temperature and transferred to a storage vial and refrigerated ($-15^\circ C$) until required for counting.

Instrumentation

^{14}C was measured by LSC using a Tricarb 2260XL, Packard Instrument Company.

Conclusion

^{14}C is measured for the first time in the air, contaminated from the UTR 300. Although a few sites are considered, they give useful information about dispersion of the radionuclide. The plume line passes above site II and comes down at site I. Calculations of collective dose show that the exposure of the global population by the ^{14}C from this research reactor is almost negligible.

Acknowledgement

I am grateful to Prof M. S. Baxter and Dr. B. W. East at SURRC, under whom supervision the research work has been carried out. I am also grateful to Dr. N.M. Butt, Dr. M.A. Atta and Mrs. J. Akhtar for their guidance and cooperation.

References

1. C.M. Lederer, J.M. Hollander, I. Perlman, Table of isotopes, 7th ed. John Wiley & Sons, New York, (1978).
2. C. Keller, Radiochemistry, Ellis Horwood Ltd, England, p. 196 (1988).
3. NCRP Report No. 18, Carbon-14 in the environment, Recommendations of the National Council on Radiation Protection and Measurements, Bethesda, Maryland (1985).
4. M. McCartney, M.S. Baxter, E.M. Scott, *J. Environ. Radioact.* **8**, 157 (1988).
5. M. McCartney, Ph.D. Thesis, Glasgow University, UK, (1987).
6. M.J. Stenhouse, M. S. Baxter, *Nature* **267**, 828 (1982).
7. M. Chudy and P. Povinec, Radiocarbon production in a CO_2 coolant of nuclear reactor. Acta Facultatis Retun Naturalim Universitatis Comenianae, *Physica XXII*, 127 (1982).
8. T.M. Krishnamoorthy, V.N. Sastry, T.P. Sarma, *Int. J. Pur. Appl. Phys.*, **20**, 119 (1982).
9. M. Tschurlovits, K. Pfeiffer, D. Rank, *Health Phys.* **82**, 398 (1982).
10. M.L. Joshi, B. Ramamirtham and S.D. Soman, *Health Phys.* **52**, 787 (1987).
11. R.L. Otlet, A.J. Walker, H. Longley, *Radiocarbon* **25**, 593 (1983).
12. T. Kato, *Int. J. Appl. Radiat. Isot.* **31**, 349 (1979).
13. C. Kunz, *Health Phys.* **49**, 25 (1985).
14. D.J. Beninson, A.J. Gonzalez, Application of the dose limitation system to the control of carbon-14 releases from the heavy-water moderated reactors, Proc. of IAEA, Symp. IAEA/SM/258, p. 1-19 (1981).