RADIOACTIVITY RATIO OF HTO TO HT IN AIR OF KEK 12 GeV PS BEAM LINE

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ABSTRACT

The tritium produced by nuclear spallation reactions in the air of high energy accelerator tunnels is present in chemical forms of HTO, HT etc.. By using a simple tritium collection system, the tritium concentrations of these individual species in the air around the KEK 12 GeV PS Beam Line (EP-2 tunnel) were measured and compared with those obtained for atmospheric air. The tritium concentrations of HTO and HT in EP-2 air were about 10^4 and 10^2 times higher than those for atmospheric tritium, respectively. A big difference was observed between the HTO/HT ratio for EP-2 and atmospheric air, the HTO/HT ratio for the atmospheric air was about 1.4, while the one for EP-2 air was extremely large, being approximately 50. Most of the tritiums produced by spallation reactions in EP-2 tunnel were found to be present in the form of water vapour.

INTRODUCTION

With the enlargement of peaceful uses of atomic energy and advance of study for nuclear fusion, the environmental contamination of tritium has been coming into important problem. Recently, in order to elucidate the behavior of environmental tritiums the tritium distribution for individual chemical species were measured for various kind of environmental samples extensively[1].

From the viewpoint of radiation control, tritium is one of the most important nuclides at the 12 GeV proton synchrotron of the National Laboratory for High Energy Physics (KEK) because of its long half-time (12.34 y) and difficulty to control discharge. Operation of 12 GeV proton synchrotron at KEK results in the production of tritium in the accelerator tunnel air by nuclear spallation reactions of high energy primary and secondary particles mainly with oxygen and nitrogen in the air, and the tritium thus formed is considered to be present in the form of aerosols and/or gaseous species (HTO, HT, etc.). Recently, ICRP has introduced a new system of limits for radioactive nuclides intaken by workers, published in the ICRP Publication 30. The annual limits on intake (ALI) and derived air concentration (DAC) of radioactive nuclides for inhalation depends very much on their chemical forms. The values of ALI for tritium are 5×10^{11} pCi/m^3 for elemental tritium and 2×10^7 pCi/m^3 for tritium water vapour, respectively. Construction of a high energy and high intensity proton accelerator is planned in Japan, which is called Japanese Hadron Project, and if this machine comes into operation in future, the tritium concentration is considered to be much higher than that for KEK PS and is one of the important parameters limiting radiation works and accelerator operation. From the point of radiation control, it is therefore very important to obtain the information on the tritium concentrations and chemical forms of tritium formed in the accelerator tunnel air.

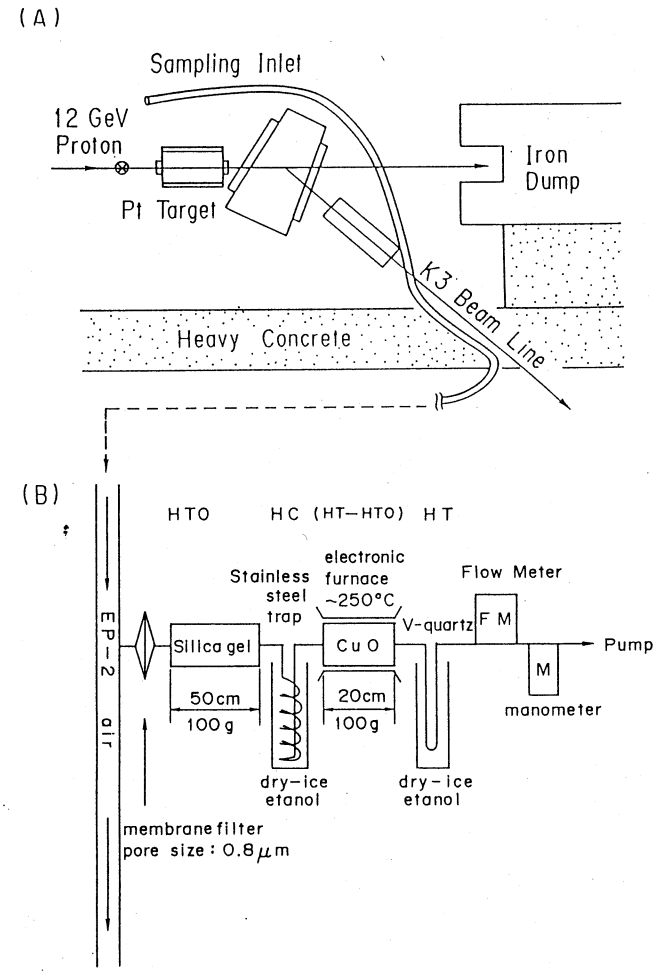

Fig. 1 (A) Plane view around the sampling inlet at the slow extracted proton beam line and (B) schematic diagram of sampling apparatus.

In this study, the chemical form of tritiated compounds and their concentrations were measured for the accelerator tunnel air and atmosphere at KEK and the results were compared with each other.

Table 1

Various sampling conditions

	Silica gel (g)	CuO (g)	CuO temperature (℃)	sampling rate (ℓ/min)	total sampling Air (ℓ)	temperature (℃)	humidity (mg/ℓ)
EP-2 air	100	100	250	~ 1.5	174 ~ 234	24.8 ~ 26.8	9.5 ~ 10.8
Atmosphere	200	100	250	~ 1.5	~ 2500	23.0 ~ 23.8	8.8 ~ 10.5

EXPERIMENTAL

Sampling of EP-2 air and tritium measurement

The sampling point at EP-2 tunnel (slow extracted proton beam line) of 12 GeV proton synchrotron is shown in Fig. 1(A). The sample air was introduced to the measurement station through a polyethylene tubing of 50 mm in diameter at a flow rate of about 1.6 m^3/min and returned to the EP-2 tunnel after measurement. The water vapour (HTO), hydrogen (HT) and compounds with melting point over -76°C (HC) were collected successively from EP-2 air at the measurement station during the accelerator operation. The beam intensity was monitored with SEC (Secondary Emission Chamber) and was approximately 1.5 x 10^{12} particles per second.

The schematic diagram of sampling apparatus is shown in Fig. 1(B). First, dust and aerosols were collected on the membrane filters (Millipore Co. Type HA) of 25 mm diameter with a pore size of 0.8 μm. The water vapor was collected by silica gel in a quartz glass tube of 16 mm inside diameter. Then, the compounds with melting point over -76°C was collected in the stainless steel trap cooled at -76°C. Finally the air was passed through a copper oxide furnace heated at 250°C to oxidize hydrogen to water, and the water vapour effluented was trapped by a quartz glass tube cooled at -76°C. The flow rate of air was about 1.5 l/min which was corrected by measuring the pressure differences with Hg manometers, and the total volume of sampled air was estimated to be about 200 l. The sampling conditions were listed in Table 1.

Each fraction thus trapped was washed out by about 2 ml of acetone after sampling, and then the acetone solution was mixed well with 16 ml of scintillation cocktail, INSTAGEL (PACKARD INSTRUMENT COMPANY, INC.) in a 20 ml low potassium glass vial. On the other hand, the silica gel adsorbed tritium water was desiccated by heating at 110°C with a flow of water-free helium gas at flow rate of 0.5 ml/min. The tritium water thus desorpted was collected in the similar manner as above. Two ml of tritium water thus collected and 16 ml of scintillation cocktail, INSTAGEL were mixed well in a counting glass vial. The tritium activity measurement was carried out by a low background liquid scintillation counter, 2000CA/LL (PACKARD INSTRUMENT COMPANY, INC.), and was cycled ten times by setting the counting time to 50 minutes. The counter efficiency and quenching correction were made by using so-called external standard method.

Sampling of atmosphere and tritium measurement

The atmosphere was directly sampled in the laboratory room at KEK. The sampling apparatus was fundamentally the same as that for EP-2 air. The flow rate was about 1.5 l/min and the total sampled volume was about 2500 l. The sampling conditions were listed in Table 1 together with those for EP-2 air.

HTO, HT and HC were separated and collected as the same manner. Each fraction were washed out by the water with low tritium concentration instead of acetone, which is mineral water sampled at Takahagi in Ibaraki prefecture. Then 60 ml of liquid scintillator, AQUASOL-2 (New England Nuclear Co. Ltd.) was added, and the total volume was adjusted to 100 ml in a teflon vial. After shaking well, it was stood in an air bath regulated at 50°C for one day in order to remove bubbles in the gelled sample. The tritium activity was measured by a low background liquid scintillation counter, LB-1 (Aloka Co. Ltd.). A set of samples was cycled five times, and in each cycle the sample was measured ten times repeatedly for 50 minutes. The counter efficiency and quenching correction was made by using external standard method[2].

Table 2

Tritium concentration in EP-2 air and atmosphere, HTO: water vapour, HT: hydrogen and HC: compounds with melting point over -76 °C

		HT pCi/m³	HTO pCi/m³	HC pCi/m³	HTO/HT
EP-2 air	1	209	1.15×10^4	BG	55.0
	2	230	1.09×10^4	BG	52.4
	3	208	9.93×10^3	BG	47.7
	4	164	7.62×10^3	BG	46.5
	5	245	1.20×10^4	BG	49.0
	Av.	211	1.04×10^4	BG	51.0
Atmosphere at KEK		0.92	1.25	BG	1.36
Atmosphere*		1.27	0.59	0.39	0.465

* Annual average tritium concentrations in the Atmosphere reported by Momoshima et al.[3].

RESULTS

The measurement results are listed in Table 2 together with the data for atmosphere obtained by Momoshima et al.[3]. The tritium concentration of HTO in atmosphere was about 2 times as large as the annual average of HTO concentration reported by Momoshima et al.[3]. They have also reported that the average tritium concentration of HTO in a summer season is 2 or 3 times higher than the annual average HTO concentration. In this experiment, the atmosphere was sampled in a period of July to September in 1987. So the concentration of HTO of 1.25 pCi/m^3 for atmosphere is nearly in accord with the values reported.

On the other hand, the tritium concentrations of HTO and HT in EP-2 air were about 10^4 and about 10^2 times higher than those for atmosphere, respectively. In atmosphere the tritium concentration of HTO and HT is nearly equal, but the radioactivity ratio of HTO/HT in EP-2 air is very large, being approximately 50. Most of tritiums produced by spallation reactions in EP-2 tunnel are present in the form of water vapour. From the viewpoint of radiation protection the present result is very useful in estimating the tritium concentrations and tritium distributions of individual species in the air of high energy and high intensity hadron accelerators such as Japanese Hadron Accelerator, which is now projected.

REFERENCES

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