BEHAVIOR OF TRITIUM IN ENVIRONMENTAL SAMPLES AROUND NUCLEAR FACILITIES AT TOKAI-MURA

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1.INTRODUCTION

Many kinds of nuclear facilities such as Power Reactors, Research Reactors, Reprocessing Plant etc. are operated in Tokai-mura, located 140 km north of Tokyo. From a point of view of tritium discharge to the environment, Heavy Water Reactors and a Reprocessing Plant are noteworthy.

Tritium concentrations in drinking water, river water, rain water and sea water have been measured in a series of environmental monitoring around the fuel reprocessing plant at Tokai-Works, Power Reactor and Nuclear Fuel Development Corporation (PNC). Besides this monitoring, a measurement method of tritium concentration in the air has been investigated. This report summarizes the results of tritium concentration in the air at Tokai-mura.

2.METHOD

Atmospheric tritium occurs mainly in two chemical forms which are tritiated water vapor (HTO) and tritium gas (HT). We developed an atmospheric tritium sampler which can differenciate between HTO and HT (Fig.1). These samplers have been set at three monitoring points around Tokai-mura and the concentrations of HTO and HT in the air have been measured separately since 1983; while only HTO had been measured by using the other samplers since 1976.

The procedure of differenciating between HTO and HT is as follows:

Water vapor is adsorbed on the first molecular sieve column. Tritium gas passed through this column is oxidized by the palladium (Pd) catalyzer and adsorbed on the last molecular sieve column (HT column in Fig.1). HTO free water is decomposed into hydrogen and oxygen electrolytically in an Electrolysis cell to give a hydrogen carrier for HT. Then, the decomposed hydrogen with the sampled HT in the air is oxidized by the Pd catalyzer. The produced H_2O and HTO are adsorbed on the HT column and the Pd catalyzer column. By knowing the ratio of an amount of water adsorbed by both columns to the electrolyzed water, the efficiency of Pd catalyzer is obtained as follows;

$$\text{(Efficiency of oxidation)} = \frac{W_{Pd} + W_{HT}}{W_E - W_D} \times 100 \quad (\%)$$

W_{Pd} : Gain in weight at Pd Column (gram)
W_{HT} : Gain in weight at HT Column (gram)
W_E : Loss in weight at Electrolysis Cell (gram)
W_D : Gain in weight at Drying Column (gram)

Efficiency of oxidation is, on average, almost 100%

After the sampling, each column is heated about 450°C in a furnace. Water trapped by each column is desorbed and recovered by a cold trap. Forty milliliters of sampled water is mixed with 60 ml of scintillator (AQUASOL-2). After that, the mixture is kept in a cooler (about 10 °C) for about one day. Then, tritium concentration in the recovered water is measured by a low background liquid scintillation counter. That measurement is repeated ten times, counting unit being 50 minutes. The detection limit is about 20-30 pCi/ℓ.

3.RESULTS and DISCUSSION

As an example of seasonal variation of HTO concentration in the air, the data measured at one monitoring station are shown in Fig.2. There are two kind of units to show tritium concentration in the air by pCi/m^3, and in the water vapor by pCi/ℓ. Although tritium concentrations in water vapor are stable, the tritium concentrations in the air show high in the summer and low in the winter because of humidity. The chronological change of annual tritium concentration in water vapor is shown in Fig.3 and that in the air is shown in Fig.4. Each geometrical mean (m_g) plotted on Figs.3 and 4 is obtained based on about fifty individual values measured at each place during a year, respectively. Figures 3 and 4 also show the values of '$m_g \times \sigma^3$' where σ is a geometric standard deviation. The measured data at monitoring station-1 (ST-1) have practically shown the same values around 140 pCi/ℓ in water vapor and 1 pCi/m^3 in the air, respectively . On the other hand, the measured data at monitoring station-2 and 3 (ST-2,3) have shown a decreasing tendency. The decreasing rates calculated by the least square method, based on the measured data at ST-2 and 3 from 1980 to 1986, are about 5 pCi/ (ℓ·year) and about 0.05 pCi/(m^3·year), respectively. These values correspond to the half-life of 10-11 years which are almost the same as the decay half-life of tritium.

The chronological concentration of HT in the air is plotted on Fig.5. The geometric mean based on the measured data at ST-1, 2, and 3 show the same values and tendency. There is no difference among these data. The individual data throughout the year also present no seasonal variation as in the case of HTO.

The measured data at ST-3 in 1984 are plotted on a log-normal distribution paper as shown in Fig.6. This indicates that the data of HTO and HT in the air fit the log-normal distribution. Figure 6 shows that the geometric mean of HTO is 0.45 times lower than that of HT, while the standard deviation of HTO is 1.5 times higher than that of HT.

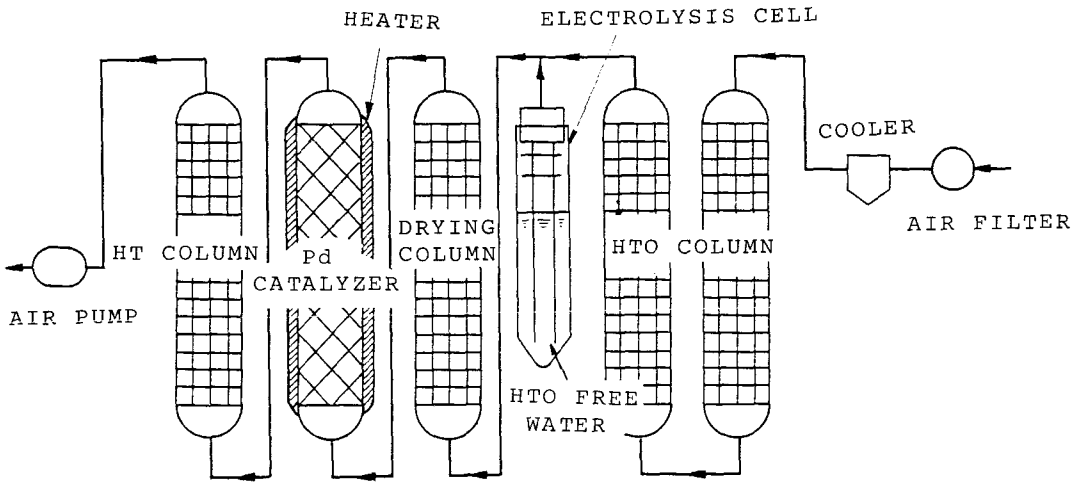

Fig.1 Atmospheric HTO and HT Sampler

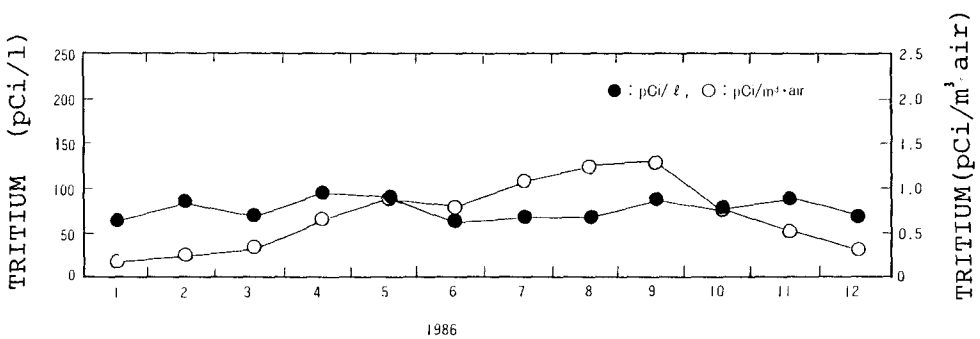

Fig.2 Monthly Tritium Concentration in Air around Tokai-mura

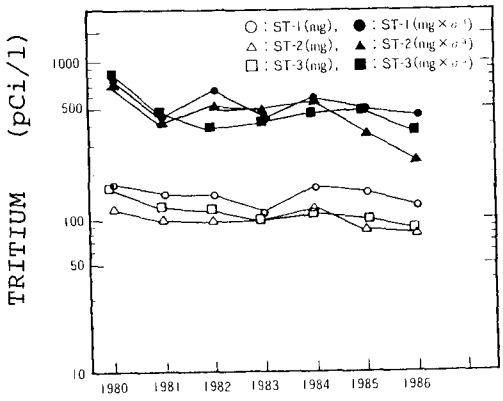

Fig.3 Annual Tritium Concentration in Water Vapor (pCi/l)

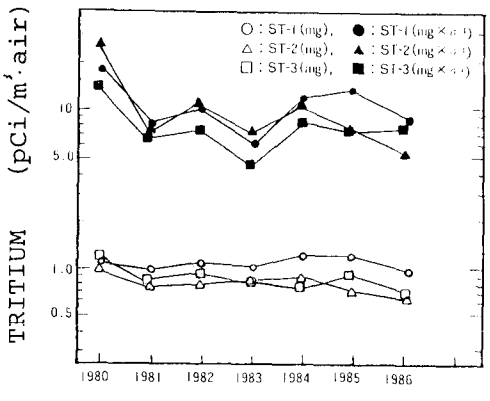

Fig.4 Annual Tritium(HTO) Concentration in Air (pCi/m³·air)

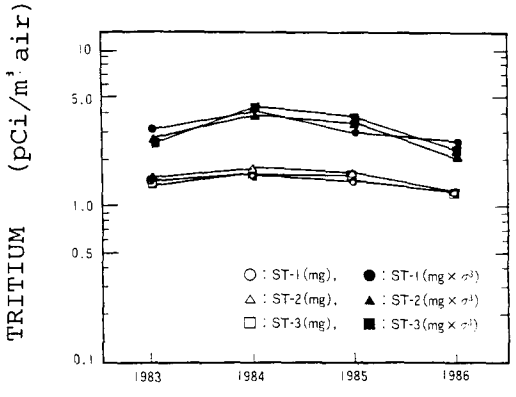

Fig.5 Annual Tritium(HT) oncentration in Air (pCi/m³ air)

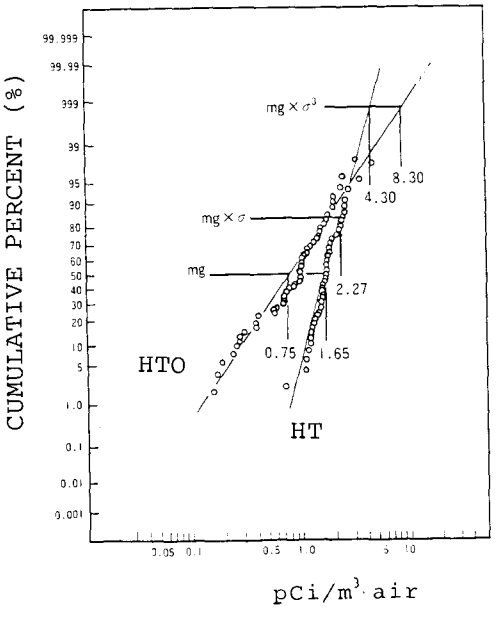

Fig.6 Measured HTO and HT Concentration plotted according to an assumed log-normal distribution