■ 論　文 ■

Tritium Concentrations of Tritiated Water Vapor and Tritiated Hydrogen in the Atmosphere in Taejon

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대전지역 대기중 수증기상태 (HTO) 및 가스상태 (HT) 삼중수소의 농도

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Abstract – During the period of March 1995 to December 1995, tritium concentrations of tritiated water vapor (HTO) and tritiated hydrogen (HT) in the atmosphere in Taejon were measured to evaluate present background levels of tritium in the atmosphere. Air samples were collected continuously for three weeks with a sampling system for tritium in the atmosphere and were analyzed by a liquid scintillation counting system.

The range of the atmospheric HTO concentrations was 3.2 - 36 mBq m^{-3} with a mean value of 16.2 mBq m^{-3}. The atmospheric HTO concentrations were the highest in summer and the lowest in winter. This trend was similar to the variation of atmospheric absolute humidity. The specific activities of tritium in atmospheric water vapor in Taejon ranged from 0.62 Bq L^{-1} to 3.82 Bq L^{-1} with a mean value of 2.04 Bq L^{-1}. The atmospheric HT concentrations were in the range of 35.7 mBq m^{-3} to 48.9 mBq m^{-3} with a mean value of 41.1 mBq m^{-3}.

Key words : tritiated water vapor (HTO), tritiated hydrogen (HT), atmosphere, Taejon

요약 – 대기중 삼중수소에 대한 국내 기초 준위를 파악하기 위하여 1995년 3월부터 12월까지 대전지역 대기중 수증기상태의 삼중수소(HTO) 및 가스상태의 삼중수소(HT)의 농도를 측정하였다. 대기시료는 삼중수소 포집장치를 이용하여 3주간 계속적으로 채취하였으며, 대기중 삼중수소는 액체섬광계수기(Liquid scintillation counter)를 이용하여 계측하였다.

대기중 HTO의 농도는 3.2 - 36 mBq m^{-3} (평균 : 16.2 mBq m^{-3})이었으며, 여름철에 높고 겨울철에 낮은 농도를 나타내었다. 이러한 경향은 대기중 절대습도의 변화와 유사하였다. 한편, 대전지역 대기 수분중 삼중수소의 비방사능 농도는 0.62 - 3.82 Bq L^{-3} 범위의 값을 나타내었으며, 대기중 HT는 35.7 - 48.9 mBq m^{-3}(평균 : 41.1 mBq m^{-3})의 농도범위를 나타내었다.

중심어 : 수증기상태 삼중수소(HTO), 가스상태 삼중수소(HT)

INTRODUCTION

In addition to natural sources, tritium has been artificially released to the atmosphere from a variety of sources including nuclear detonations, nuclear power reactors and fuel reprocessing plants. At present, tritium discharge from nuclear facilities for peaceful use of nuclear energy has become more significant than that from nuclear weapons tests[1]. Fusion reactors, which use tritium as fuel, will become another source of atmospheric tritium in the future[2]. Therefore, it is very important to survey the background levels of atmospheric tritium before large-scale tritium handling begins.

There are three major chemical forms of tritium in the atmosphere, tritiated water vapor (HTO), tritiated hydrogen (HT) and tritiated hydrocarbons, primarily tritiated methane (CH_3T). Migration behavior of tritium is strongly affected by chemical and physical forms. HT discharged into the environment is converted into HTO mainly by activity of microorganisms in soil. Discharged HTO enters directly into water cycles in the environment. So, chemical forms of tritium at a discharge point are very important for the estimation of the tritium migration in the environment and dose to the public. From an atmospheric discharge point of view, HT and HTO from nuclear facilities are dominant forms of discharged tritium[3]. We determined HTO and HT concentrations in the atmosphere in the Taejon area as the first step to obtain background data of the atmospheric tritium concentrations in Korea, which will eventually be used for evaluation of the environmental impact from nuclear facilities.

EXPERIMENTAL

Sampling

Air samples were collected on the roof of the research building #3 of the Korea Atomic Energy Research Institute in Taejon during the sampling period of March 1995 to December 1995. The sampling was carried out continuously for three weeks with a sampling system for tritium in the atmosphere [4,5].

Reagent and apparatus

Molecular sieves (Yakuri chemical Ltd. one-sixteenth inch 4A pellet) were used as adsorbents for water vapor. Palladium catalyst (0.5% palladium-alumina pellet) was used for hydrogen oxidation. For the correction of counting efficiency of tritium, quenched standard samples were prepared from the tritium standard solution (Amersham, Ltd.).

A sampling system for tritium in the atmosphere was designed (Fig. 1) and set in a case (90 cm depth×60 cm width×40 cm depth). HTO and HT only were collected for up to three weeks with the system. The system was composed of HTO collecting system, oxygen supplying system, HT oxidation system, and oxidized HT (HTO) collecting system. The HTO collecting system was consisted of six columns filled with 500 g of molecular sieve. The water vapor in the atmosphere was adsorbed onto the molecular sieve in the columns. From the oxygen supplying system, oxygen was supplied into the HT oxidation system continuously by electrolysis of tritium-free water. For the drying system, two 500 g molecular sieves containing columns were used to remove the mist from the electrolytic cell. Two columns filled with 700 g palladium catalyst were used to oxidize HT. The oxidized HT (HTO) collecting system consisted of two 500 g molecular sieve columns. The HTO oxidized from HT was adsorbed on the molecular sieve. A dry test meter was used to calculate the integrated air volume collected during the sampling period. A small diagram-type air pump was used to introduce air into the sampler. The flow rate of the air was controlled from 0.5 to 1 L min-1 using a gas flow meter.
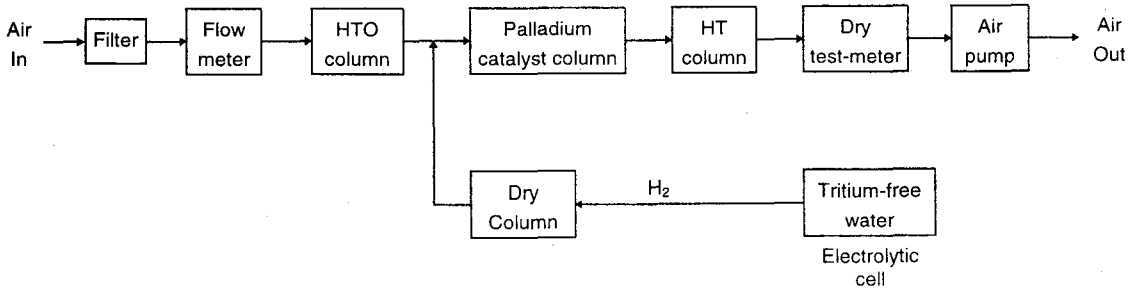
Fig. 1. Flow diagram for stepwise collections of atmospheric HTO and HT.

Method for atmospheric tritium analysis

Throughout the three week sampling period, air samples ($15 m^3$ in summer ; 24 m^3 in winter) were first passed through a filtering system at a flow rate of 0.5-1 L min^{-1}. Water vapor in the air was adsorbed on the molecular sieve in the HTO collecting system, and then dried air and the oxygen generated from the electrolysis of tritium-free water were introduced into the HT oxidation system where HT was oxidized to the water form (HTO). After catalytic oxidation to water, HTO was taken up into the oxidized HT (HTO) collecting system. The quantities of water collected in the HTO collecting, HT oxidation, and oxidized HT (HTO) collecting systems were determined by weighing each column before and after sampling. Each column in the system was heated to 450℃ using an electric heater with high purity N_2 gas (99.99%) flowing through the column at a rate of 1 L min^{-1}, and then water samples from the column were recovered into a cold trap. Eight-mililiter of recovered water from the HTO collecting, HT oxidation, and oxidized HT (HTO) collecting systems and 12 ml of scintillation cocktail (Insta-gel XF, Packard Ltd.) were mixed in 20 ml Teflon vials. After cooling in the counter at 13℃ under dark conditions for 3 days, tritium concentrations in the samples were measured by the liquid scintillation counter (LKB Wallac, Quantulus 1220) for 1000 min (50 min×20 times). The correction curve for the counting efficiency was prepared by measuring a series of quenching standard samples of the same activity at different water contents.

RESULTS AND DISCUSSION

Calculation of HTO and HT activities

HTO and HT activities in the atmosphere were calculated by the following equations :

$$C_{HTO} = (C_1 \times M)/(Q \times 1000)$$
$$C_{HT} = \{C_2 \times (P+N) \times 100\}/(Q \times E_{ox} \times 1000)$$

where, C_{HTO} is the atmospheric HTO concentration (Bq m^{-3} - air), C_1 is the tritium concentration in the water recovered from the HTO collecting system (Bq L^{-1}), M is the amount of water collected from the HTO collecting system (g), Q is the integrated air volume (m^3), C_{HT} is the atmospheric HT concentration (Bq m^{-3} - air), C_2 is the tritium concentration in the water recovered from HT oxidation and oxidized HT (HTO) collecting systems (Bq L^{-1}), P is the amount of water collected from the HT oxidation system (g), N is the amount of water collected from the oxidized HT collecting system (g), and E_{ox} is the hydrogen oxidation efficiency (%), which can be calculated from the following equation :

$$E_{ox} = |(P+N)/(C-D)| \times 100$$

where, C is the reduced weight of the electrolyte in the electrolysis system (g), and D is the amount of water collected from the HTO collecting system.

Adsorption efficiency of water vapor on molecular sieve

All of the water vapor in the air passed through the sampling system should be adsorbed by the molecular sieve in the HTO collecting system so that excess water vapor is not introduced into the HT oxidation and oxidized HT collecting systems. Figure 2 shows the adsorption efficiency of water vapor into the molecular sieve columns which were connected in series. For 15 m^3 of air, above 95% of the total water vapor in the atmosphere was collected by four molecular sieve columns during summer when the absolute humidity in the atmosphere was high, whereas above 95% of total water vapor was adsorbed on three molecular sieve columns during spring, autumn and winter, when absolute humidities were relatively lower than that during summer. Therefore, at least three molecular sieve columns were required to adsorb more than 90% of total water content in 15 m^3 of air throughout 3 week sampling period.
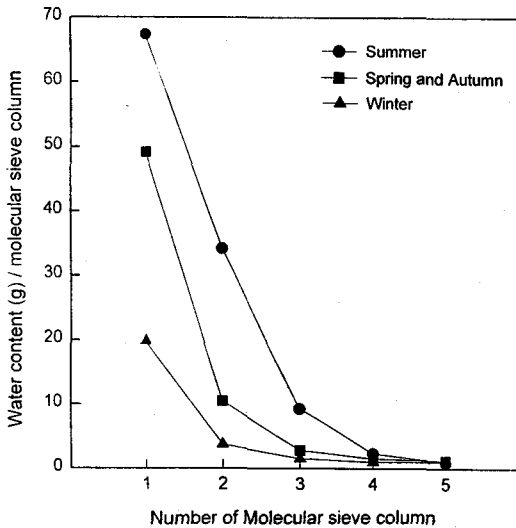

Fig. 2. Adsorption efficiency of atmospheric water vapor on molecular sieve.

Atmospheric tritium concentrations in Taejon

The range of atmospheric HTO concentrations was 3.2 - 36 mBq m^{-3} with a mean value of 16.2 mBq m^{-3} (Table1 and Fig. 3). The atmospheric HTO concentrations were the highest in summer and lowest in winter. This trend was similar to the variation of atmospheric absolute humidity, which varied from about 2 g m^{-3} in winter to about 20 g m^{-3} in summer.

Table 1. Annual average values of atmospheric tritium concentrations at Taejon in 1995

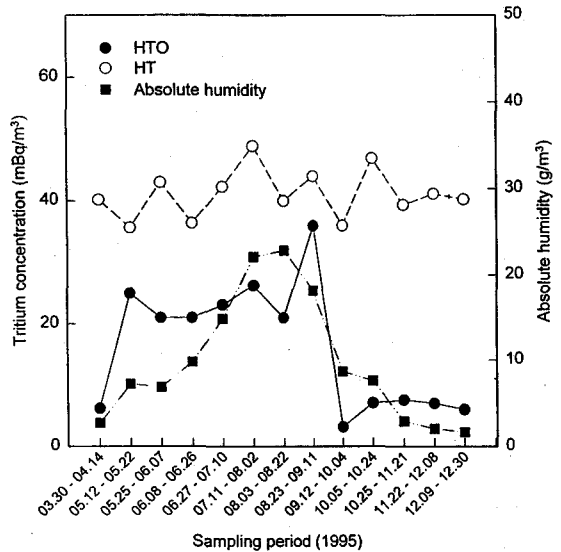| Chemical form	Tritium concentration
HTO(Bq m^{-3} - air)	16.2± 10
HTO(Bq L^{-3} - H_2O)	2.04± 1.13
HT(Bq m^{-3} - air)	41.1± 3.8

Fig. 3. Atmospheric tritium concentrations and absolute humidity in Taejon.

Consequently, the higher the absolute humidity is, the higher the atmospheric HTO concentration is. This resulted from the fact that the specific activities of tritium in the atmospheric water vapor did not show much seasonal variation as shown in Fig. 4.

The specific activities of tritium in atmospheric water vapor in Taejon ranged from 0.62 Bq L^{-1} to 3.82 Bq L^{-1} with a mean value of 2.04 Bq L^{-1}, which is close to the values of 0.8 - 3.2 Bq L^{-1} observed in Fukuoka, Japan from 1984 to 1988[6]. An assessment of the seasonal variation of tritium in the atmosphere, however, can be achieved through the long-term observation of the atmospheric HTO concentration. Therefore, many studies on atmospheric tritium should be done continuously.
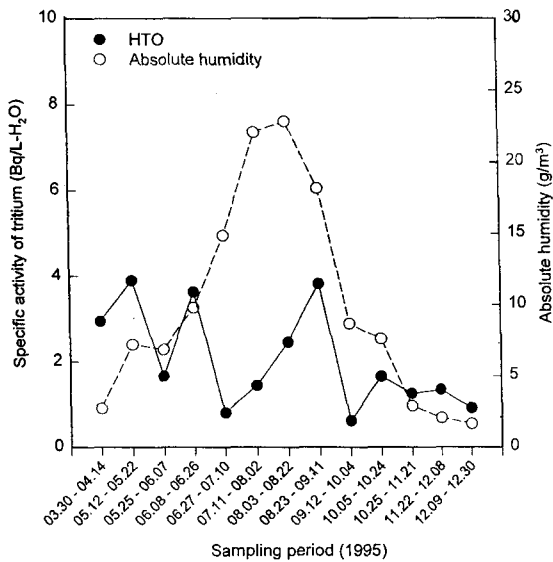

Fig. 4. Specific activities of tritium in atmospheric water vapor and absolute humidity in Taejon.

On the contrary, the atmospheric HT level did not show any remarkable seasonal variation (Fig. 3). The atmospheric HT concentrations ranged from 35.7 mBq m^{-3} to 48.9 mBq m^{-3} with a mean value of 41.1 mBq m^{-3}. These values are similar to those observed in Fukuoka, Japan from 1984 to 1988[6], whereas it is lower by more than one order of magnitude than the values of 22-110 Bq L^{-1} observed in Tokyo in 1966[7], and about 50% of those obtained by Mason

et al.[8] in 1978. This means that the atmospheric HT concentration has decreased since 1963 when the atmospheric weapons tests ban treaty became effective. However, the present HT concentration is still 200 times higher than the HT level before the nuclear era[9].

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