

## S26. Study on Environmental Behavior of Tritium

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### i) Variation of atmospheric tritium concentrations

Major chemical species of atmospheric tritium have been measured at Kumamoto. The concentrations of water vapor (HTO) changed in accordance with that of water content in the atmosphere (Fig. 1), showing high concentrations in summer and low in winter. The tritium concentration of water vapor revealed no distinct seasonal variation, rather showing a large scattering in concentrations. The tritium concentrations in rain collected at Kumamoto was significantly lower than that of water vapor, about half in concentration. The origin of tritium in the recent rain would be natural production, thus higher tritium concentration in water vapor at earth surface suggesting an existence of additional sources of tritium for water vapor in the atmosphere. The tritium concentrations of atmospheric hydrogen (HT) and hydrocarbon ( $\text{CH}_3\text{T}$ ) are shown in Fig. 2 and 3. The concentrations of both species have decreased and fairly constant concentrations in recent years, however the decrease of  $\text{CH}_3\text{T}$  is much larger than HT. The atmospheric concentrations of  $\text{H}_2$  and  $\text{CH}_4$  are 0.5 ppm and 1.7 ppm respectively. The specific activities (T/H) calculated are significantly different among the chemical species as shown in Table 1. Since the sources of environmental tritium are natural production, nuclear detonation and nuclear facilities. The residence times of atmospheric HT and  $\text{CH}_3\text{T}$  would be less than several years because the physical half-life of tritium is 12.34 year. Nuclear detonation is not likely the source of HT and  $\text{CH}_3\text{T}$  because atmospheric nuclear detonations were carried out at dense schedule in early 1960s, more than 40 years ago. Recent source of chemical forms of HT and  $\text{CH}_3\text{T}$  must be nuclear facilities for peaceful and military uses.

Table 1 Atmospheric concentrations and specific activity

	concentration (mBq/m <sup>3</sup> )	Specific activity (TU)
HT	$12.5 \pm 6.9$	$2.6 \times 10^5$
$\text{CH}_3\text{T}$	$9.0 \pm 8.2$	$3.4 \times 10^3$
HTO	$6.7 \pm 5.4$	
HTO	$0.69 \pm 0.35$ Bq/L	5.8

$$\text{TU} = \text{T}/\text{H} = 1 \times 10^{-18}$$

### ii) Model calculation

Tritium transfer model was constructed, which was developed to explain the change of natural tritium concentrations at Toki area. The model consists in underground reservoirs and rivers which defined as water catchments for rain. The input of tritium to Toki area was considered only from rain and the amount of tritium input

gave yearly or a few month averages. The observed tritium concentrations in the river network at Toki area and in the soil water of the deep soil cores were compared with the predicted values. The time course of tritium concentration in the rivers as well as the soil core waters shows in good agreement with the predicted values as a hole, however, large out flows from underground reservoir to river occurs at high precipitation events. An improvement of model adoptable to the situations of strong rain is necessary.

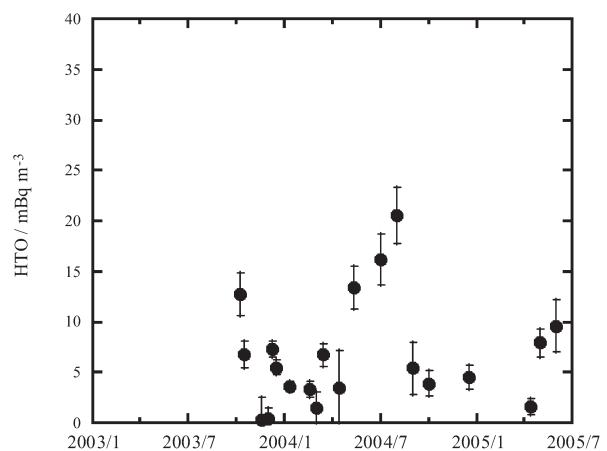


Fig. 1 Change of HTO in the atmosphere

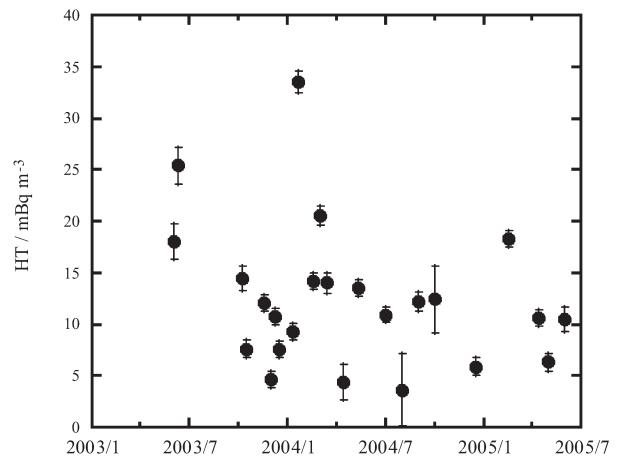


Fig. 2 Change of HT in the atmosphere

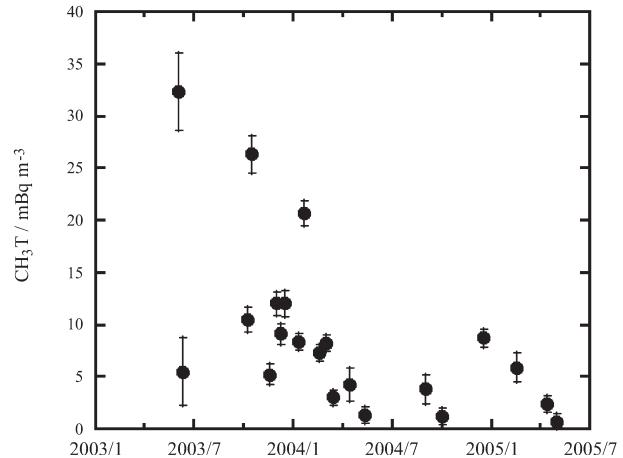


Fig. 3 Change of  $\text{CH}_3\text{T}$  in the atmosphere