

§3. Measurement of Atmospheric Tritium in the Chemical Form and Analysis of Factors Controlling the Variation

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National Institute of Fusion Science (NIFS) is now constructing a new fusion device and related facilities at Toki city in Gifu prefecture, Japan. Although no tritium burning experiments will be carried out in the Toki site, it should be appropriate and useful to establish the general database on behavior of atmospheric tritium in and around an actual place of the experiments. Atmospheric tritium consists of three different chemical forms, tritiated water vapor (HTO), tritiated hydrogen (HT) and tritiated hydrocarbons (primarily tritiated methane, CH₃T). Thus, tritium concentrations of atmospheric HTO, HT and CH₃T have been measured in the Toki site a few times a year from 1991 to the present.

The present HTO concentrations vary within a range of 1.1 to 2.8 and are close to the tritium concentration in Kobe in 1953. Since the Japanese islands are surrounded by oceans with low tritium levels, atmospheric HTO would be easily diluted by water vapor from sea water and its concentration would become low quickly. Thus we conclude that the present HTO concentrations have already decreased to the levels before nuclear tests.

HT concentrations gradually decreased from 1992 to 1998 and residence time for atmospheric HT are calculated to be about 9 years for 1992-98. Mason and Östlund reported an average residence time for atmospheric HT of 4.8 years. This value would result from the physical decay of tritium and atmospheric HT removal processes. The dominant phenomenon among these removal processes is the conversion of HT to HTO by oxidation in surface soils. HT oxidation in soil is usually parameterized by a deposition velocity and the deposition velocity is mainly dependent on the water content of the soil, while temperature is only of secondary importance. Assuming that the average water content of soil was almost constant for 1963-1998 on the whole

earth, removal of atmospheric HT by the soil must have remained constant. Therefore, the residence time of 9 years longer than the 4.8 years suggest that atmospheric HT has been supplied from various sources.

HT concentrations have decreased by a factor of about seven since the high tritium level in 1963. If, during the period 1963-1998, HT concentrations had decreased at a rate corresponding to Mason and Östlund's residence time estimation of 4.8 years, then currently we would expect a level of about 3 mBq/m³-air. However, the present HT levels of about 24 mBq/m³-air are distinctly higher than those expected. Since atmospheric HT has the highest specific activity of the three tritium species, transformation of HT from the other tritium species can not possibly explain the present HT concentrations in the atmosphere. These facts would indicate the supply of HT from tritium sources to the atmosphere. The present HT concentration is still higher by a factor of about 120 than that before the nuclear tests.

On the other hand, the present CH₃T concentrations are higher by a factor of about 30 compared to values observed in the southern hemisphere in 1953. Methane gas is chemically degraded in the atmosphere by the reaction with OH radicals and it has a residence time of 12 years. Therefore, the residence half life of atmospheric CH₃T is estimated to be about 6 years in addition to the physical decay of tritium. Decrease in atmospheric CH₃T concentrations was not apparent from our observations. The fact that, unlike HT concentrations, CH₃T concentrations have not been decreasing is one of the most interesting problems in our study. The causes could be the transformation of HT into CH₃T with less specific activity or a continuous release of CH₃T from tritium sources. The oxidation of atmospheric HT by microorganisms in soil has continuously added HTO to the soil surface and the HTO has high specific activity of tritium. When the HTO was transformed into CH₃T by methane-producing bacteria in the soil, the resultant CH₃T should have high specific activity and could be a source of atmospheric CH₃T. This is reasonable, because the specific activity of HT is one order of magnitude higher than that of CH₃T.