§30. Study on Environmental Behavior of Tritium

Momoshima, N. (Fac. Sci. Kumamoto Univ.)
Okai, T. (Fac. Eng. Kyushu Univ.)
Amano, H., Atarashi, M. (JAERI)
Miyamoto, K. (NIRS)
Takahashi, T. (Kyoto Univ.)
Uda, T., Sakuma, Y., Yamanishi, H., Sugiyama, T.

i) Tritium measurement

Atmospheric concentrations of three different chemical forms of tritium were measured at Kumamoto (Kumamoto Univ.) and Toki (NIFS) to elucidate a dependence of concentration upon sampling locations and a seasonal variation. In the atmosphere, water vapor (HTO), hydrogen gas (HT) and hydrocarbon, mostly methane (CH_3T) is the major species related to tritium. Sampling apparatus developed for successive collection of three species was applied in which at first HTO was collected by adsorption on molecular sieve (MS), followed by HT by oxidation to water and adsorption on MS, finally CH_3T by oxidation to water and adsorption on MS. During oxidation of HT and CH_3T tritium free hydrogen evolved by electric decomposition of tritium free water and bomb CH_4 were added to increase water to be adsorbed on MS. Waters adsorbed on MS were recovered by heating the MS and flowing N_2 gas and tritium activity in the recovered water was measured by liquid scintillation counting.

The concentrations of HTO, HT and CH_3T between Kumamoto and Toki were not so different, indicating no difference with sampling location. Considering the geographical situation of Kumamoto in south of Japan and Toki in central Japan, sources of three chemical forms were not a local origin. The concentrations (Bq/m^3) was in the order HT> CH_3T >HTO, indicating a large difference of specific activity, tritium atom to hydrogen atom, due to large difference of water vapor, hydrogen and CH_4 concentrations in the atmosphere. Significantly high specific activity in hydrogen and methane speculate their origin from nuclear industry.

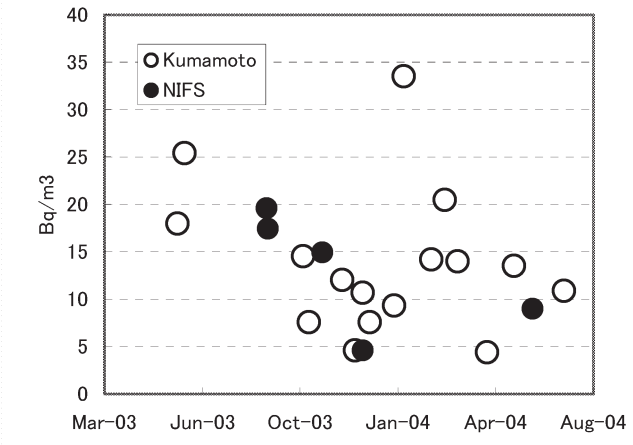

Fig. 1. Atmospheric HT concentrations collected at Kumamoto and NIFS.

Measurement of atmospheric tritium species at Fukuoka, Japan from 1984 suggests that overall continuous decrease in their concentrations. The HT reveled temporal increases in 1985 and 1988; however from 1989 a concentration seems to has decreased. The apparent half time of decline was 11.8 year in the period from 1984 to 2002. While CH_3T showed a half time of 26.6 year in the period from 1984 to 2002, and highest concentration was recorded in 1988 as HT. A latitude dependency was reported on HTO concentrations in rain and atmospheric vapor and increased HTO concentration was observed at north area of Japan. It would be necessary to carry out measurement of atmospheric tritium species at northern area of Japan.

ii) Model calculation

In evaluation of radiation dose of tritium that is released from nuclear installations to the environment, model calculation would be an only possible powerful tool. At NIFS site 1g of tritium release was assessed using a guideline for commercial nuclear power plant, in which reemission form surface ground was not considered, though reemission is a characteristic phenomenon in tritium dynamics in the environment. The reported maximum atmospheric concentration was 20 Bq/m^3 for 1 g annual release of tritium. We evaluated an effect of reemitted tritium on atmospheric concentration; reemission should elevate tritium concentration by release from the ground surface. The evaluation code of ETDOSE, which was developed for modeling of tritium behavior in the environment, was used and atmospheric concentrations were calculated using the meteorological date taken at Toki. The ground was divided into equal size meshes and influence of tritium reemitted from each mesh on interested point was accumulated with assuming that all of the tritium was reemitted when the interested point was located downward of wind for some period. A height of a stack was 40m and 50% of the released tritium was HTO were used as described in the guideline. Deposition velocities of HTO and HT were assumed to be $3x10^{-3}$m/s and $3x10^{-4}$m/s, respectively.

The evaluation was carried out for northwest wind direction, which gave the maximum atmospheric tritium concentration of 20 Bq/m^3 for the calculation based on the guideline. For the case of HTO release tritium concentration in the atmosphere was not increased clearly within 1km from the release point of NIFS compared to the calculation based on the guideline and maximum concentration of about 20 Bq/m^3 was evaluated, suggesting validity of evaluation based on the guideline. However, a clear increase in atmospheric tritium concentration was observed beyond 1 km due to reemission of tritium. For the case of HT release, elevation of atmospheric tritium concentration with HTO released from ground surface was insignificant. The HTO was derived by oxidation of HT and deposition to ground and reemission to atmosphere. The evaluation based on the guideline was confirmed to be valid for HTO and HT release because the maximum tritium concentration were appeared at the same distance from the NIFS and the same level for both evaluations.