

Low-level measurement of organically bound tritium in environmental samples with noble gas mass spectrometer

H. Kakiuchi^{a,*}, H. Hasegawa^a, N. Akata^a, S. Hisamatsu^a, T. Tamari^b, H. Kawamura^b,
N. Baglan^c, N. Momoshima^d and T. Uda^e

^a*Institute for Environmental Sciences, Rokkasho, Aomori 039-3212 Japan*

^b*Kyushu Environmental Evaluation Association, Matsukadai 1-10-1, Higashi-ku, Fukuoka 813-0004 Japan*

^c*CEA/DIF/DASE/SRCE, 91297 Arpajon Cédex France*

^d*Radioisotope Center, Kyushu University, Hakozaki 6-10-1, Higashi-ku, Fukuoka 812-8581 Japan*

^e*National Institute for Fusion Science, Oroshi-cho 322-6, Toki, Gifu 509-5292 Japan*

Accompanied with release of ^3H from nuclear power plants and spent nuclear fuel reprocessing plants to the environment, increased ^3H concentration is observed in various environmental materials around them. Tritium in environmental samples consists of free water ^3H (FWT) and organically bound ^3H (OBT). OBT has a longer biological half time and higher assimilation to organic materials than FWT. Therefore, the concentration of OBT should be separately measured from that of FWT for radiation dose estimation from ^3H in environmental samples. OBT has been generally measured by a liquid scintillation counting of combustion water from freeze-dried sample. The alternative method is to measure the decay product of ^3H , ^3He , with a noble gas mass spectrometry (NG-MS). Although the method is readily adaptable to a wide variety of sample matrices, there is no report to measure the low-level OBT concentration less than 1 Bq L^{-1} combustion water. We developed the measurement method adoptable to the sample with the low-level OBT concentration less than 0.1 Bq L^{-1} combustion water.

Fresh pine needles and humus samples which have OBT concentration of around $0.5\text{-}0.9 \text{ Bq L}^{-1}$ combustion water were collected from a forest floor in Rokkasho, Japan ($40^{\circ}58'\text{N}$, $141^{\circ}22'\text{E}$) in July 2002. The samples were cryogenically processed to separate free water and dried residue. A portion of the residue sample was sealed in an evacuated Al-Si glass ampoule (8252, SCHOTT). To remove atmospheric He as completely as possible to lower background of ^3He from ^3H , a small amount of acetone was added to the sample, and the ampoule was evacuated to expel adsorbed gases in the sample before sealed. The ^3He was measured with a NG-MS (VG-5400, Micromass) after the sealed ampoule was stored at -30°C over 90 d. The other portion of the residue sample was combusted, and the ^3H concentration in combustion water was measured by a liquid scintillation counter after electrolytic enrichment. Both OBT concentrations agreed well with each other, which showed that this method was practical and effective.

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