

Production and release of tritium from a research reactor

Masami Fukui
Ron Grazioso

1 Introduction

The Kyoto University Research Reactor (KURR) is a light water moderated research reactor. It commenced operation in 1964 at 1 MW_{th} and the power was raised to 5 MW_{th} in 1968. The ventilation system in the containment building works at ca. 350 m³.mn⁻¹ order to cover the duration of the reactor operation cycle of 70-80 h.wk⁻¹ from Tuesday through Friday. Using a Ge(Li) detector the concentrations of radionuclides in the exhaust air during operation at 5 MW_{th} have been confirmed to be in the order of 10⁻² Bq.cm⁻³ for ⁴¹Ar, 10⁻⁶ Bq.cm⁻³ for tritium, 10⁻⁸ Bq.cm⁻³ for ¹³¹I and 10⁻⁹ Bq.cm⁻³ for ¹³¹I. As a result of the routine monitoring by a 22-L ionization chamber set for the stack exhaust, the presence of most radionuclides in the stream is usually masked by the reading of ⁴¹Ar; this is especially true for tritium vapor.

As shown in Figure 1 the reactor core is submerged in a moderating pool of 30 m³ of demineralized light water. The reactor supports two major sources of tritium: a D₂O facility (Installed in 1964: ca. 2 m³) and a Cold Neutron Source (CNS) facility (Installed in 1986: ca. 40 dm³ liquid deuterium).

This report is divided into three major sections: 1) monitoring the tritiated water (HTO) vapor in the stack exhaust in order to assess

the annual quantity discharged, 2) monitoring the HTO concentrations in the air within the containment building in order to get insight into the distribution and variations in the controlled area for radiation workers, and 3) monitoring the HTO concentrations in the exhaust from ⁴¹Ar decay tanks of both the D₂O and CNS facilities (Figure 1) set near the reactor core in order to assess the HTO production of major sources. The goal of the last section is to attempt to locate a possible tritium leak to decrease radiation exposure to both radiation workers and the public.

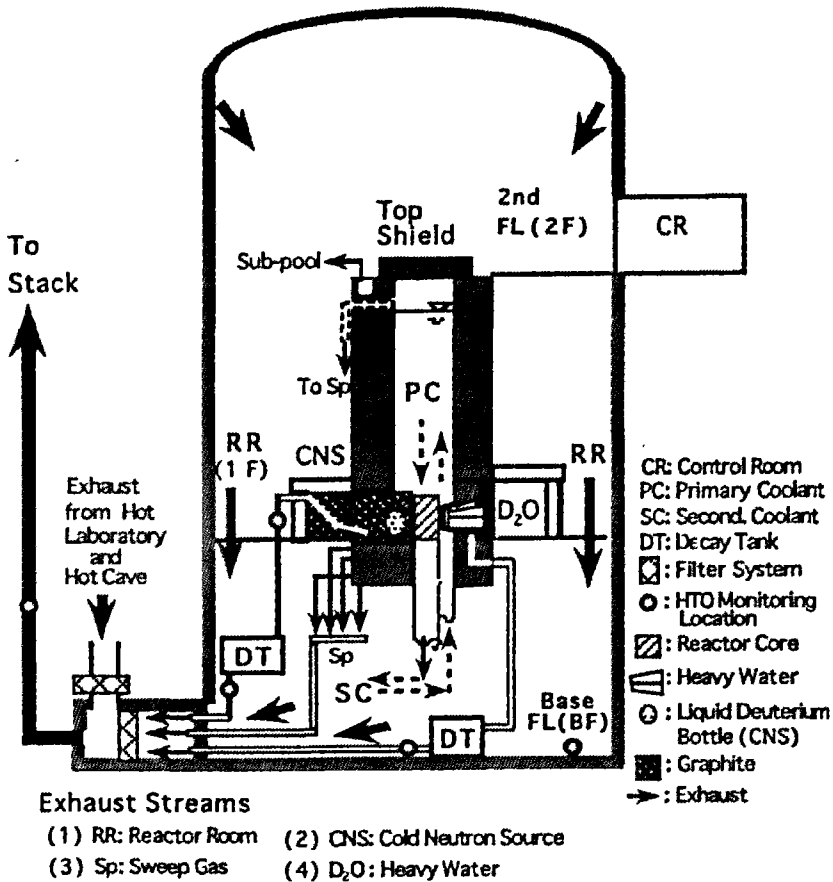


Figure 1
 Cross section of the KURR with major exhaust streams.

All concentrations reported here are measured by the liquid scintillation counting (LSC) method for a cocktail of 10 ml scintillator and 1 ml condensate, collected using condensers and/or dehumidifiers for a few hours.

Monitoring HTO concentrations in stack exhaust

Figure 2 shows the HTO concentrations in condensate of the stack exhaust monitored routinely for twenty years using LSC. This Figure indicates that the concentration has not increased so much since the installation of these HTO source facilities except for a term of unexpected release of heavy water. During a one and half

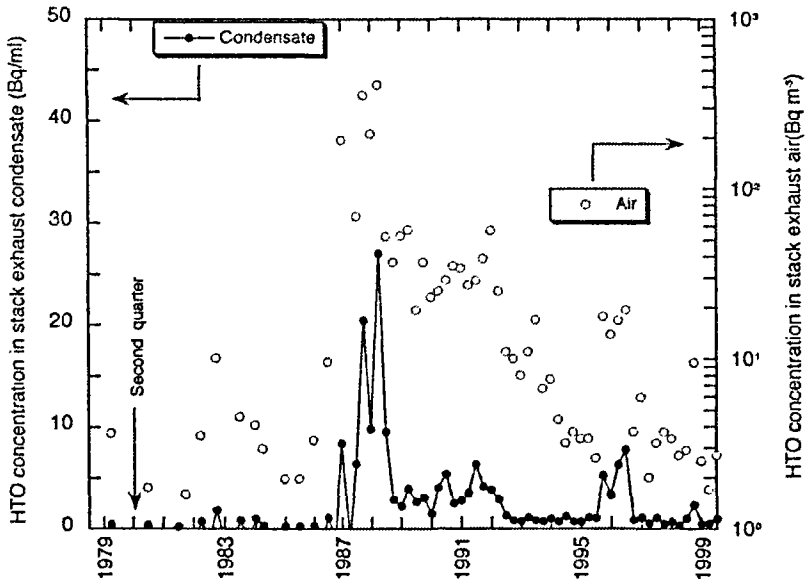


Figure 2
Evolution of HTO concentrations in the KURR stack exhaust over the last twenty years.

year period from May 1987 to November 1988, the D₂O facility leaked approximately 0.34 TBq of heavy water (ca.30 dm³) into the KURR containment building (Fukui, 1992). As shown in Figure 2, peak concentration, 27 Bq.ml⁻¹ in the stack condensate (ca.400 Bq.m⁻³ in air), was observed for the routine monitoring of the third quarter of 1988. After the leak event, monitoring of HTO vapor in the stack exhaust has been carried out on a quarterly basis during the operation of the KURR. A few times after 1990, ranges from 5 to 10 Bq.ml⁻¹ were recorded due to the small leak of the heavy water during the period of renewal work of distribution pipes of D₂O during 1990 to 1992 and the storage tank in 1996. As recognized the exhaust monitoring is on a batch basis for the water samples

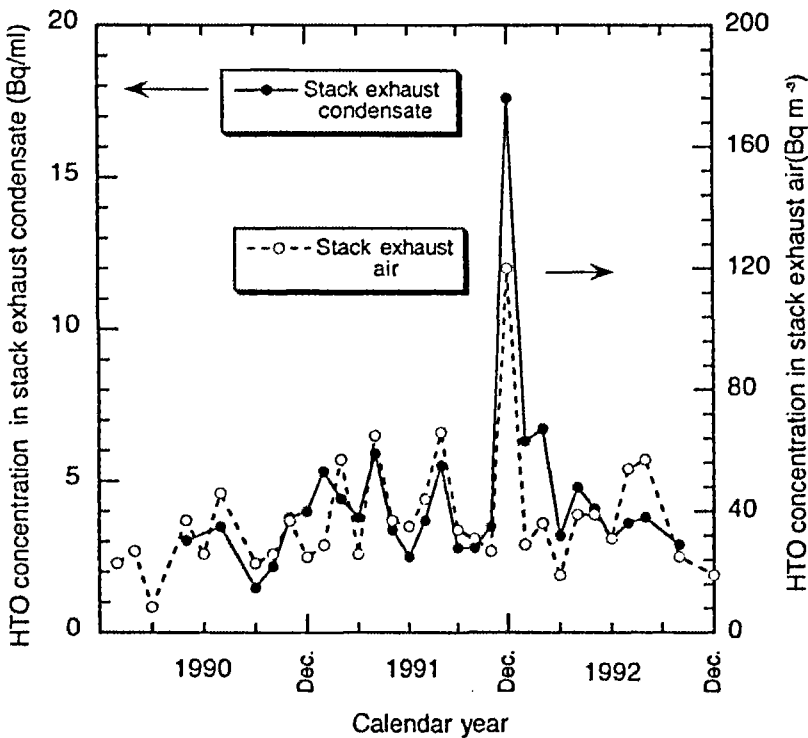


Figure 3
HTO concentrations in the stack exhaust monitored on a monthly basis during 1990-1992.

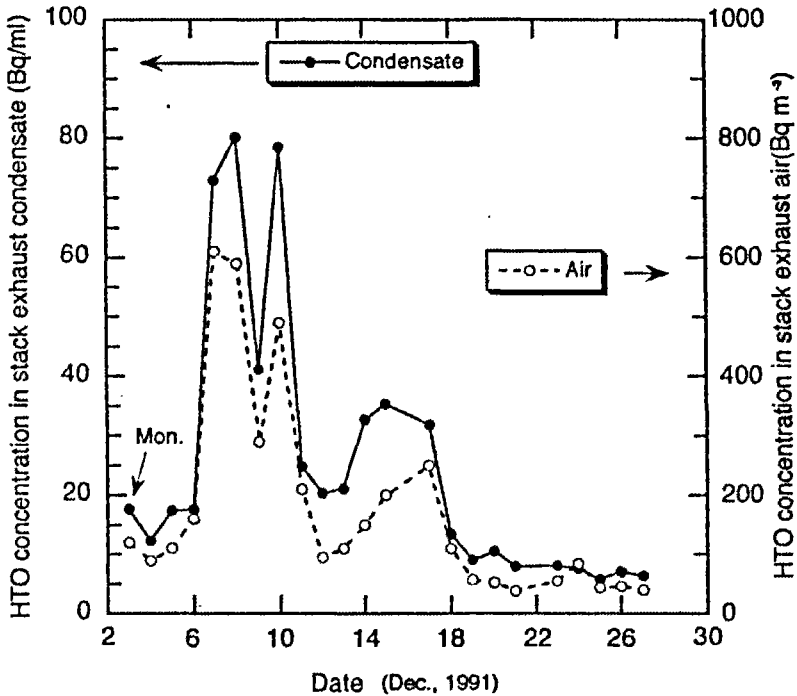


Figure 4
HTO concentrations in the stack exhaust monitored on a daily basis in December 1991.

collected as condensate and may overlook more high concentrations in the exhaust. This is confirmed by the results that showed the peak concentrations of 18 and 80 Bq.ml⁻¹, respectively, for monthly (1990-1992; Figure 3) and daily (December 1991; Figure 4) basis monitoring. This suggests that a greater frequency of sampling condensate is required (at least on a daily basis) in addition to the routine monitoring on the quarterly basis in case of a probable leakage of tritium.

Recent monitoring data show that the HTO concentrations in vapor are less than a few Bq.ml⁻¹, resulting in the annual HTO discharge from the KURR to be estimated as ca. 1 GBq.yr⁻¹.

Monitoring HTO concentrations in containment building air

Procedure for assessment of HTO concentrations in the air using laboratory dishes

The transfer of HTO vapor, C_v , from the air into dish water is primarily a function of two processes: exchange and evaporation. Exchange refers to the net transport of HTO into or out of the water dish. Exchange is dependent on the difference in specific activities between the water in the dish and the vapor in the air. Evaporation affects the amount of HTO that leaves the water and returns to the air. These phenomena were modeled mathematically (Fukui, 1992). The change in the HTO concentration, C_w , in the dish water exposed to the HTO concentration in vapor, C_v , is expressed as:

$$C_w = C_v \{ 1 - (1 - k_e t / H)^{k_c/k_e} \} / \bar{\alpha}, \quad (1)$$

where:

- C_w : the HTO concentration of the water sample (Bq.ml⁻¹);
- $\bar{\alpha}$: the ratio of isotropic concentrations (H^3)/(H^1) in the saturated vapor and liquid phases of tritiated;
- C_v : the HTO concentration in vapor (Bq.ml⁻¹);
- k_c : the exchange rate constant (cm.h⁻¹);
- k_e : the apparent evaporation rate constant (cm.h⁻¹);
- t : the elapsed time after exposure (h), and
- H : the initial depth of water in the dish (cm).

Thus from eqn. (1), the average concentration in vapor during exposure time, t , is given by:

$$C_v = \bar{\alpha} C_w / \{ 1 - (1 - k_e t / H)^{k_c/k_e} \}, \quad (2)$$

Finally, the concentration of HTO in air, C_a , is related to C_v by the following equation:

$$C_a = K C_v, \quad (3)$$

where:

$$K = 2.9 \times 10^{-4} P^{-1} (273 + T)^{-1} R / 100, \quad (4)$$

P is the saturated vapor pressure (mmHg) at temperature $T(^{\circ}\text{C})$ (Anazawa *et al.*, 1972), and R is the relative humidity (%). The isotope ratios, $\hat{\alpha}$, were found by measurement to be approximately 0.90 to 0.92 (Sepall, 1960). The ratio k_c/k_e is estimated approximately as 1.8 for the given ambient temperature and humidity in the containment building. Estimation of other parameter values including k_c and k_e is described elsewhere (Fukui, 1993). Thus, all constants can be calculated or are given in order to monitor C_a .

A liquid scintillation counter was used to determine the specific activity of the water sample, from which the tritiated water vapor concentration was calculated. The samples were counted for three 10-minute cycles for tritium (^3H) and carbon 14 (^{14}C) as a reassurance that tritium was the main radioactive nuclide in the water samples.

Distribution of HTO concentration in the containment building air during ventilation cessation using laboratory dishes

The distribution of the HTO concentration in the air of the containment building was determined from 30 laboratory dishes which were filled with 25 ml of tap water and placed on the floor and/or desks. The reactor was shutdown at 16:00 on Fri., July 18, 1997 and ventilation in the building ceased at 18:00 that day. After 40 hours the ventilation cessation, the dishes were exposed for 24 hours until July 21, then they were weighed in order to estimate the evaporation rate and water sampling for radiometry on July 21. The average concentration of HTO, C_v , estimated by the methodology described above was 54.5 Bq.ml^{-1} in the containment building and is shown for each of the locations as the difference (Bq.ml^{-1}) from the average concentration in Figure 5 together with the location numbers. The HTO concentrations in vapor in Figure 5 show a little dependence on the location of the sample. The values range from 30 (No.24) to 75 Bq.ml^{-1} (No.8) with the lowest concentrations coming from the basement (heat exchange room). The heat exchange room is isolated by a wall in the basement and the lowest downstream of the containment building, i.e. is connected to the exhaust stack, suggesting air exchange between the heat exchange room in the containment building and field during venting cessation. The highest

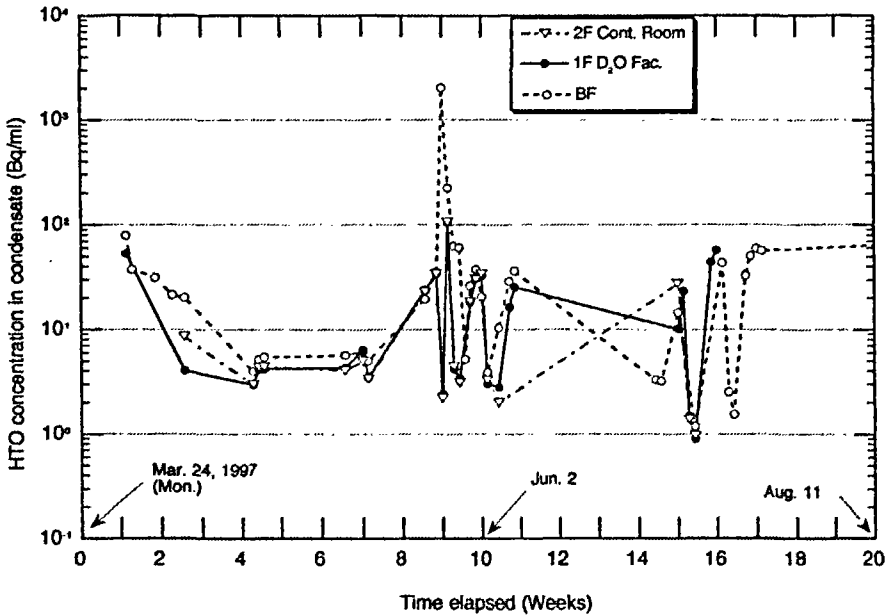


Figure 5

Distribution of HTO concentrations in the vapor, C_V , estimated using laboratory dishes filled with tap water (25 ml) set at 30 locations for 24 hrs during venting cessation in the KURR containment building. Bars indicate the difference in $\text{Bq}\cdot\text{m}^{-1}$ from the average concentration for the 30 locations ($54.5 \text{ Bq}\cdot\text{m}^{-1}$).

concentration (No.8) that appeared near the D_2O facility on the first floor may link to No.16 in the basement, the location with the second highest concentration via. An open gallery under the A stairs. Thus, the local high concentration was found to be attributable to the small leak of heavy water and/or HTO impregnated wastes related to the D_2O facility. From the above, this technique using laboratory dishes provides an adequate distribution of HTO concentrations within the three floors of the containment building. Figure 6 shows the HTO concentrations in dish water, C_w , at 30 locations, the vapor concentration estimated from eqn. (2), C_V , and the HTO concentration in air, C_a , converted from eqn. (3). This indicates that the measured concentrations in dish water exposed for 24 hours fall

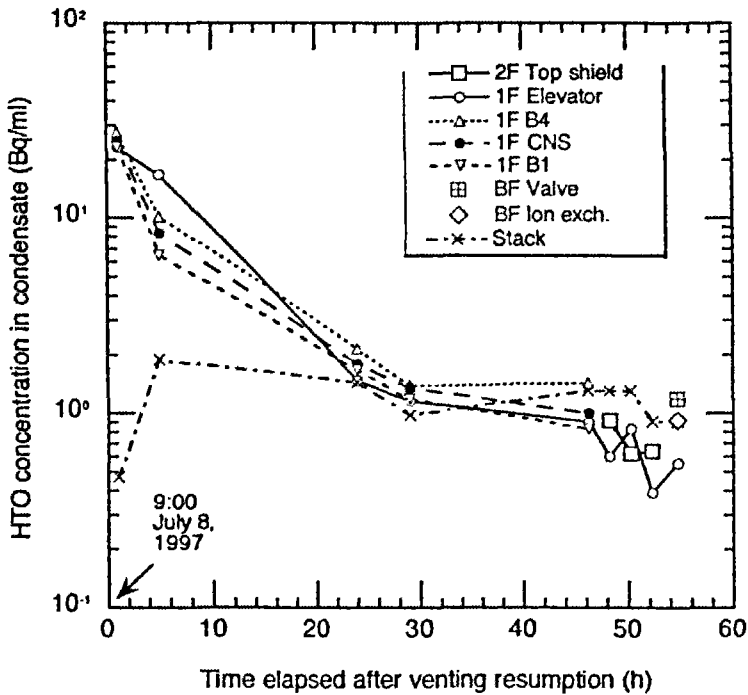


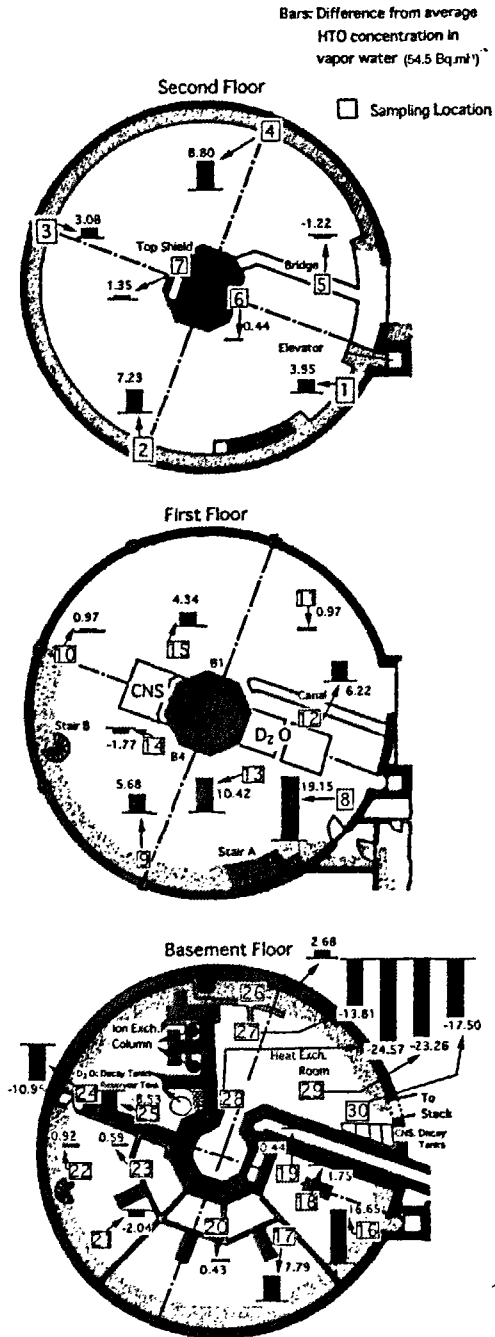
Figure 6
HTO concentrations in dish water, vapor and air
at the 30 locations.

from 40 to 80% of C_V , depending on the temperature and humidity of each location. The highest concentration in air, $1.3 \text{ kBq}\cdot\text{m}^{-3}$, was found at location No.16 in the basement. It is below the maximum concentration limit for an average over three months, restricted by the law, $700 \text{ kBq}\cdot\text{m}^{-3}$.

Changes in HTO concentration in vapor in the containment building after venting resumption

Four dehumidifiers were set in the containment building to investigate the changes in concentrations with time after venting resumption. As seen in Figure 7, the concentrations at the four locations on

Figure 7
Changes in the HTO concentration in condensate within the containment building after venting resumption.



the first floor (No. 8-11 in Figure 5) decreased from a few ten to a few $\text{Bq}\cdot\text{ml}^{-1}$, i.e., one order of magnitude less, 24 hours after venting resumption. No apparent differences can be seen in the concentrations in the air of the three floors. This indicates that if one detects a concentration exceeding $10 \text{ Bq}\cdot\text{ml}^{-1}$ in vapor 24 hours after venting resumption, it may come from an artificial cause of contamination.

Figure 8 shows the HTO concentrations monitored over period of four months from late March to late July 1997 using a dehumidifier on three floors (Base FL: No.19, 1st FL: No.12, 2nd FL: near No.1 in Figure 5). The highest concentration, more than $2000 \text{ Bq}\cdot\text{ml}^{-1}$, was detected in late May 1997, resulting from a spill of heavy water during the renewal of a distribution pipe linked to the D_2O tank. The

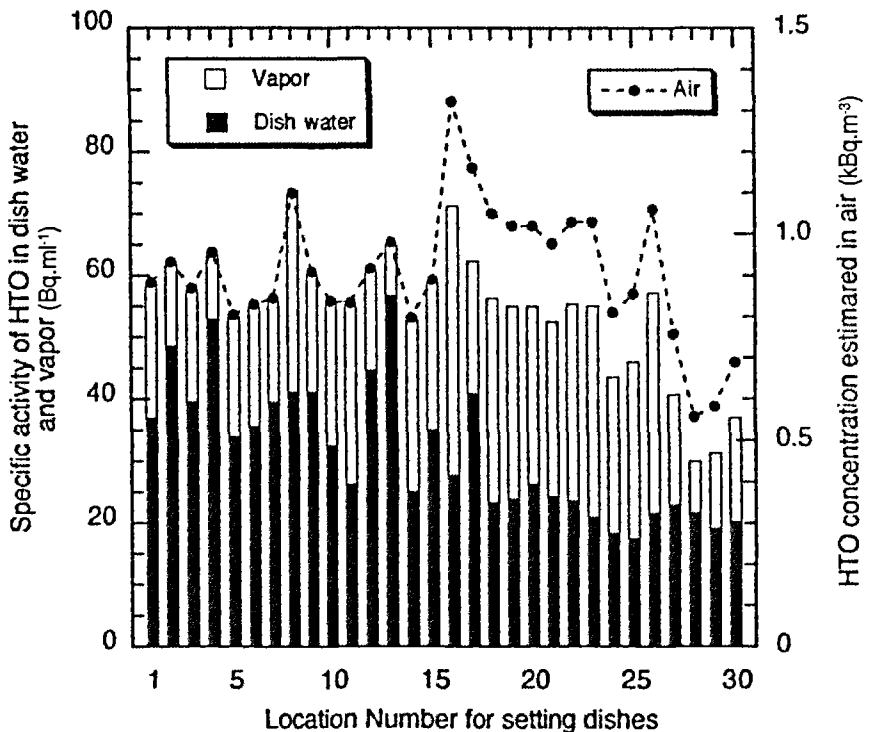


Figure 8
HTO concentrations in condensate at three floors
for four months in 1997.

work for the renewal of the distribution pipe and tank finished in late July 1997. Nevertheless, ingrowth of the HTO concentration in vapor was shown in the containment building during venting cessation after reactor shutdown. Monitoring the concentrations in vapor near the reactor core was then planned to locate a possible tritium leak other than that from the D₂O facility.

Monitoring HTO concentrations in vapor in decay tank exhaust

Many source points in KURR were sampled in order to locate a higher source of tritium activity or any other radionuclide that could be causing a higher specific activity in the air. The sources that were sampled included the: exhaust stack, reactor room exhaust, reactor room filter chamber, experimental pipe to the biological shielding, exhaust from the CNS and D₂O facilities and air samples from all three floors of KURR. Among these, only the CNS and D₂O facilities showed an abnormally high specific activity. The 4 dm³ liquid deuterium in a bottle as a neutron moderator set near the reactor core is isolated by three layered aluminum conduit (5 mm thick) including a vacuum, a He and a Ne gas layer. The CNS system surrounded by a graphite block layer is isolated by sealing from which exhaust air is introduced to decay tanks (1.3 m³) at a rate of ca. 2 dm³.mn⁻¹ in order to lower ⁴¹Ar concentration. The exhaust line pump for the CNS and D₂O (flow rate: 0.53 dm³.mn⁻¹, decay tank volume: 0.56 m³) facilities only works to cover the reactor operation. Figure 9 shows the activity of the CNS and D₂O exhaust vapor. The higher specific activities ranging from 130 to 200 kBq.ml⁻¹ were found at the inlet of the decay tank from the CNS graphite. These values do not fluctuate because of the presence of the graphite which can absorb tritium near the CNS facility and release it depending on venting resumption by the pump set in the exhaust line. That is, the graphite may normalize the tritium concentration with time. On the other hand, the concentrations at the outlet of the

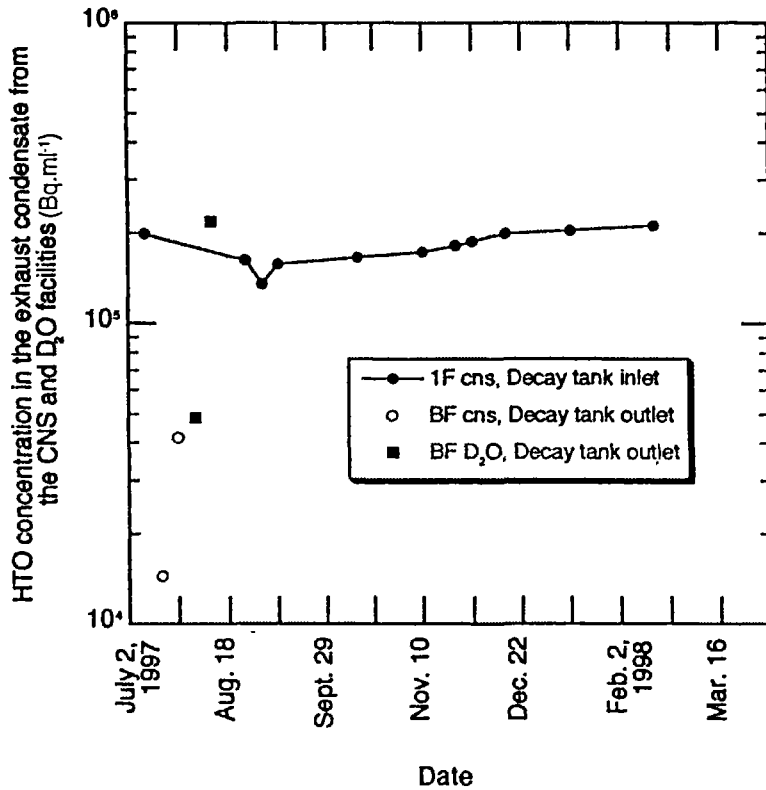


Figure 9
HTO concentrations in condensate
in the CNS and D₂O exhaust lines.

decay tanks for both facilities fluctuate so much due mainly to inhaling downstream air near the outlet of the exhaust line pipes because of pumping by the ionization chamber system from the line during exhaust line pumping cessation.

There may be two reasons why the tritium concentrations from the CNS and D₂O lines could be so high: 1) the leak of liquid deuterium and/or heavy water could exist, therefore expelling a large quantity of tritium, or 2) the leak could be negligible, but a higher concentration of tritium exists in the facilities that was produced by a nuclear reaction. That is, the ingrowth of HTO vapor during vent-

ing cessation in the containment building may be caused by the $^{14}\text{N}(n, ^3\text{H})^{12}\text{C}$ reaction of the irradiation of nitrogen in air. Assuming the constant concentration of $200 \text{ kBq}\cdot\text{ml}^{-1}$ is in a steady state in the exhaust line vapor which is trapped by the CNS condenser, the production rate of HTO from the CNS line is estimated as $0.6 \text{ GBq}\cdot\text{yr}^{-1}$. Considering the above and HTO sources of other exhaust lines from the KURR, the annual release rate based on the monitoring data from the stack exhaust, $0.6 \text{ GBq}\cdot\text{yr}^{-1}$, may increase by a factor. The production of a large content of tritium from the D_2O facility could not be discarded through the $^6\text{Li}(n, \alpha)^3\text{H}$ reaction. The probable source of tritium, Li, was removed from the experimental devices in July 1997. All of the water samples collected were also counted for ^{14}C , which was thought to have contributed to the higher than expected activities. But the constituent of ^{14}C in the activity of the samples was less than a few percent and, therefore, considered to be minimal.

Precise estimation for the amount of discharge from the KURR is under investigation using not only a LSC method but also a 1.5 l portable ionization chamber focused on near core exhaust.

Conclusions

The use of laboratory dishes in estimating the HTO concentration in air proved to be very simple and effective. The laboratory dish method did provide us with the useful information of a low HTO concentration in the containment building air. Unfortunately, since the HTO concentrations within the containment building air were not so different, a positive location of a tritium leak could not be identified. Even after the renewal of distribution pipes, valves and tank for heavy water, which were likely points of HTO leakage, higher than concentrations were found in the exhaust air from facilities adjacent to the reactor core. This was a cause of ingrowth in the HTO concentration in the containment building during venting cessation. Our next step must be to concentrate the sampling around the reactor core in order to pinpoint the precise location of the leak

and/or to estimate the production rate by the neutron reaction. The production and leak should be located and rectified in order to lower the tritium concentration throughout the containment building. Though the annual discharge of tritium from the KURR was estimated to be around 1 GBq.yr^{-1} , This must be done in accordance with ALARA. It is hoped that the leak is small, and that a large percentage of the tritium leaking is from a nitrogen reaction not an artificial one.

Acknowledgements

This research was supported, in part, by Grant-in Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture, Japan.

A portion of this work was done while R. Grazioso, University of New Mexico, USA was an exchange student at the Kyoto University. Mr. T. Yoshimoto provided technical assistance in monitoring activities.

Bibliography

ANAZAWA Y., KOKUBU M., FUJITA K., 1972 — Monitoring of Tritium in the Working Environment. *Hoken Butsuri (Japan J. Health Phys.)*, 7: 27-35.

FUKUI M., 1992 — Modeling the Behavior of Tritiated Water Vapor in a Research Reactor Containment Building. *Health Phys.*, 62: 144-154.

FUKUI, M., 1993 — Development of a Convenient Monitoring Method for Tritiated Water Vapour in Air using Small Water Dishes as Passive Samplers. *Radiat. Prot. Dosimetry*, 48: 169-178.

SEPALL O., MASON S. G., 1960 — Vapor/Liquid Partition of Tritium in Tritiated Water. *Can. J. Chem.* 38: 2024-2025.