LONG-TERM OBSERVATION OF TRITIUM IN PINE NEEDLES NEAR NUCLEAR FACILITIES

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INTRODUCTION

An interest has been focussed to tissue bound tritium (TBT) in foods, because it may be directly assimilated in the bound compartment of tissues, and increase cumulative total body dose[1]. Transfer from environmental tritiated water to the tissue bound fraction in plants had been investigated through many laboratories and relatively lower percentage fixation was suggested[2]. On the other hand, analysis of various kind of environmental samples which were considered to be affected by fallout tritium only showed high specific activity ratio of TBT to tissue water tritium (TWT)[3,4]. This phenomenon had been attributed to either following cause:a) long retention of tritium previously incorporated in the soil during the period of maximum fallout[3], or b) an incorporation of high specific activity elemental tritium in the atmosphere into the soil microbes and plants [5]. More studies have to be performed in the environment as well as in the laboratories on the behaviour of TBT in different types ecosystem in order to solve this problem, because few literatures have been available on the TBT behaviour in the chronically contaminated environment [6,7]. Therefore, a long-term study was conducted on the behaviour of TBT and TWT in plants in a location which had been affected mainly by tritiated vapour discharged into the atmosphere.

METHODS

Pine trees were selected for the investigation, because of their availability regardless places and seasons in Japan and a capability of simultaneous collection of present and old pine needles back to a few years ago from a branch. Samples had been collected in their growing seasons on the monthly basis from 1981 to 1986 except 1984 at several points in a down-wind area within 2km from two heavy-water moderated research reactors JRR-2 and JRR-3 of JAERI in Tokaimura, Ibaraki Prefecture. Monthly accumulated precipitation, groundwater and surface soil to 1m depth were also collected. Detail discription of sampling points, location of nuclear facilities, meteorological conditions, tritium deposition-source distance relationship in this area and the time-variation in tritium concentration of water form in various samples during 1981 and 1983 were presented previously[8,9].

Freeze-dried pine needles were plasma-ashed[10] and tritium concentrations of both freeze-dried water and oxidation water

were determined by means of liquid scintillation counters (LSCs). The tritium concentration of both waters are expressed in pCi/l The detection limit for 500 minutes of counting lies at approx. 20 pCi/l for 40ml freeze-dried water by a LSC ALOKA-LB1, and 50 pCi/l for 10ml oxidation water by a PACKARD TRICARB 2000CA/LL.

RESULTS AND DISCUSSIONS

Time-variations of tritium concentrations in the tissue water (TWT) and the tissue bound form (TBT) of the pine needles and in monthly accumulated precipitation collected at about 700m south-west from JRR-2 and JRR-3 were shown in Fig.1 with that of sum values of monthly discharge rates of tritiated vapour into the atmosphere from the two reactors. It is shown that several curies of tritiated vapour were discharged every month under their normal operation except June 1982 when abnormal discharge rate 44Ci was recorded due to a small leakage of heavy water from JRR-3. The tritium concentration of monthly precipitation varied from about 50 pCi/l to about 600 pCi/l except an abnormal high value of 2570 pCi/l in June 1982. The tritium concentration of tissue water varied from about 100 pCi/l to about 1000 pCi/l throughout 1981-1985 except a value of 2500 pCi/l in June 1982. On the contrary, the concentration of tissue bound tritium varied from about 250 pCi/l to about 1400 pCi/l without exception. The TBT data in 1984 were estimated by oxidizing pine needles having grown in 1984 which were separated from branches collected in 1985 and were plotted inside a rectangle in Fig.1 in order of their collection date. This memory effect of TBT in the pine needles has been almost verified with agreement in TBT concentrations between some pine needles of a same growth year but different collection years.

The concentrations of TWT and TBT in the pine needles at the point mentioned above proved to be 10 to 20 times higher than natural environment[11]. The tritium discharge rates, the tritium concentrations in both the monthly precipitation and tissue water of the pine needles fluctuate larger than the TBT concentrations, however annual average of these four items seemed to correlate each other. Therefore correlation was examined between annual mean TWT or TBT concentration and each of following three factors:a) annual mean tritium concentration in monthly precipitation, b) annual tritium deposition and c) annual discharge rate. The results were shown in Fig.2 for TWT and in Fig.3 for TBT . From Fig.2, the TWT concentration proved to correlate well with the three factors a, b and c with correlation coefficients from 0.95 to 0.99. From Fig.3, the TBT concentration proved to correlate fairly well with the three factors with correlation coefficients from 0.75 to 0.92. These results suggest that tritium in the pine needles at the point originated from the atmospheric discharges mainly from JRR-2 and JRR-3 and was incorporated mainly through the root after washout deposition as well as through the air. It is worth noting that the ratio of annual mean specific activity of TBT to TWT was about 1.7 from 1981 to 1983 and 1.1 in 1985, and the ratio of

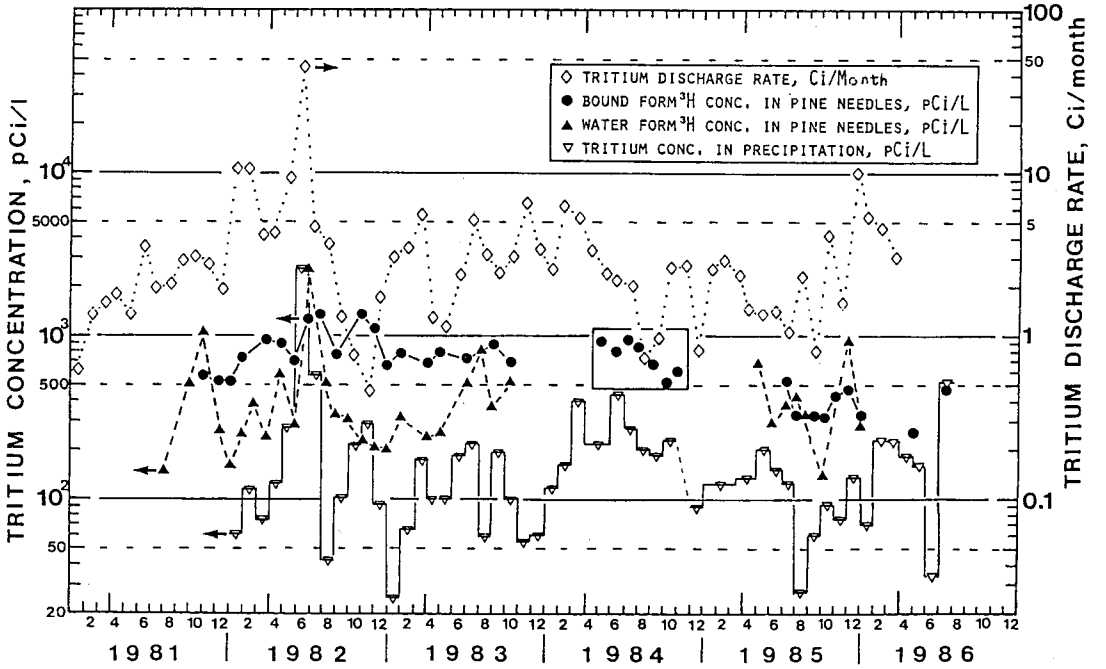
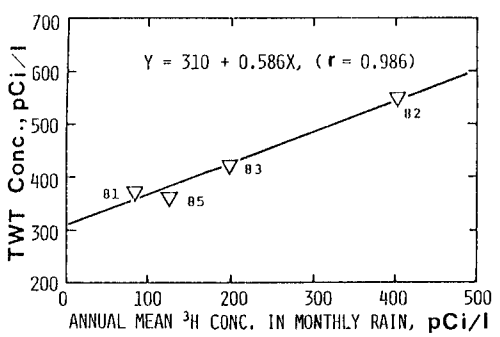


Fig.1 Time variations of TWT and TBT concentrations in the pine needles, ^{3}H concentration in rain and ^{3}H discharge rate.



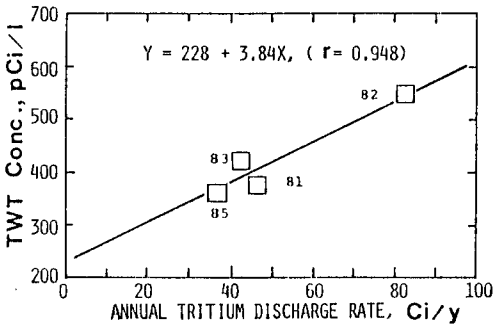
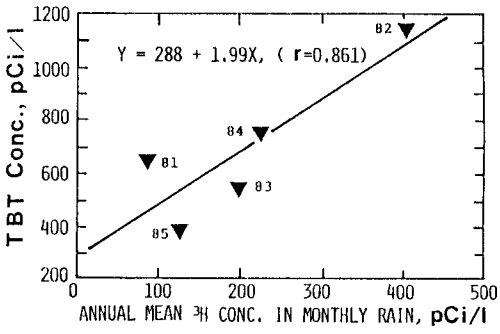


Fig2. Correlation of TWT conc. with ^{3}H conc. in rain (↑) and with annual ^{3}H discharge rate (↓).



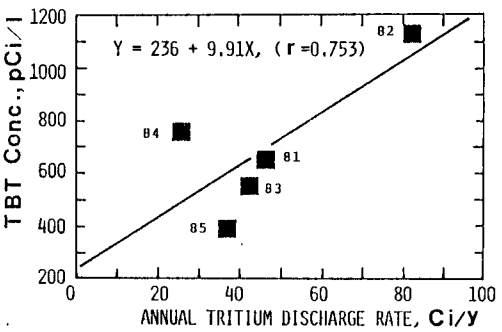


Fig.3 Correlation of TBT conc. with ^{3}H conc. in rain (↑) and with annual ^{3}H discharge rate (↓).

the annual mean specific activity of TWT to the precipitation was ranged from 1.4 to 4.5 with an average 2.7 throughout 1981-1985 except 1984.

Weekly monitoring data of HT in air having been obtained by PNC and JAERI in this area showed its level of 1-2pCi/m^3 through several recent years which was the same level with that in natural environment. The mean specific activity of TBT in pine needles in whole Japan was reported 66 $\pm$18 pCi/l whereas that of TWT was 46 $\pm$15 pCi/l[11]. Therefore, the elevated specific activity of TBT observed at the point can not be attributed to HT in the air. The mean specific activity of tritium in the soil moisture in the area appeared to be medium between those of the precipitation and the tissue water in the pine needles. The monthly mean specific activity of tritium in the air vapour was reported to be about twofold higher than that of the monthly precipitation at the monitoring station No.7 of JAERI which was located close to the point of the study. The elevated specific activity of TBT can not be explained by the tritium level of monthly mean or chronically contamination both in the soil moisture and the air vapour.

It has been demonstrated under non-equiribrium conditions at Karsruhe that after leaves were exposed temporarily high specific activity tritiated vapour, the specific activity of TBT in leaves reached approx. one tenth of vapour and it was retained with a considerably high ratio of TBT to TWT until the end of the vegetation period due to a retention effect of TBT[6]. An exposure to abnormally high level of tritiated vapour had supposedly happened also to the area in Tokaimura in the middle of June 1982 accompaning with the high level of tritium washout deposition. In addition to this event, temporal elevation of tritium level in the air vapour might had happened repeatedly at the area along the main downwind direction from JRR-2 and JRR-3 and resulted in the long-term retention of higher level of TBT in the pine needles.

REFERENCES

1) Etnier, E.L.,et al.: Radia. Res. 100, 487-502, 1984.
2) Tritium in some typical ecosystems, Technical Reports Series No. 207, IAEA VIENNA, 1981.
3) Bogen, D.C., et al.: Health Phys., 30, 203-208, 1976.
4) Belot,Y.,et al.: Radia. Protec. Dosi.,16, 111-113,1986.
5) Sweet, C.W., et al.: Environ. Sci. Technol. 18, 358-361, 1984.
6) Strack, S: Proceedings of IRPA 6, Vol.I, 135-138, 1984.
7) Murphy, C.E, Jr.: Radia. Protec. Dosi. 16, 51-58, 1986.
8) Inoue,Y.,et al.: Proceedings of IRPA 6, Vol.I, 180-183, 1984.
9) Inoue, Y., et al.: NIRS-M-52, 296-315, 1985.
10)Strack,S., et al.: KfK-report 3249, 1981.
11)Takashima, Y. et al.: Appl. Radiat. Isot., 38, 255-261, 1987.