tritium

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CHAPTER TEN
Tritium Health Physics

RADIOBIOASSAY AT THE KARLSRUHE NUCLEAR RESEARCH CENTER DURING THE YEARS 1967 - 1970

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Radiobioassay serves the purpose of controlling the observance of radiation protection measures. As soon as the body burden surpasses specific limits, working conditions must be changed to avoid further incorporations or at least to reduce them to an acceptable level.

For tritium, in particular for tritiated water generated in heavy water moderated reactors, radiation protection measures consist of keeping the concentration of tritium in the respiratory air below the maximum permissible value of 2 pCi/ml. This is done with adequate ventilation of the work area. In the absence of this precaution, the worker must be protected from exposure by respirators or protective suits. Measurement of the concentration of tritium in respiratory air by appropriate monitors is necessary, since this concentration determines the number of hours the worker may spend in the work area. The hours should be arranged to safely avoid exposures exceeding the maximum permissible continuous body burden.

Using man as a biological indicator, i.e., adopting health physics measures after the events, is a method which should never be adopted. Hence, postexposure surveillance through urine analyses should be used only to confirm the success of the measures and for additional control.

In this report, an account will be given of the experience gathered at the Karlsruhe Nuclear Research Center with respect to Radiobioassay.

Determination of the Minimum Activity Detectable by the Analytical Method Used

The method of detection to be used in routine analyses should be as simple and timesaving as possible. Expensive preparation procedures should be avoided. According to the Recommendations of the International Commisssion on Radiological Protection (ICRP), Publication 10, at least 1/20 of the maximum permissible activity should be detectable, which is about 1.2 µCi of tritium per liter of urine for 500 µCi of tritiated water in the body.

Using a liquid scintillation spectrometer and a scintillator consisting of 4 g 2,5-diphenyloxazole (PPO), 0.05 g 2,2'-p-phenylenbis (4-methyl-5-phenyloxazol) (dimethyl POPOP), 120 g naphthalene for scintillator purposes, and one liter dioxane p.a. [1], the smallest detectable quantity can easily be reduced to 1 nCi/ml. Extended periods of measurement and larger sample volumes allow even low-level detection of tritium in urine with detection limits of 0.2 nCi/l [1]. It was questioned whether this sensitivity of measurement was reasonable in radiation protection for radiobioassay of persons working at the Nuclear Research Center. In this range of measurement, disturbing effects may

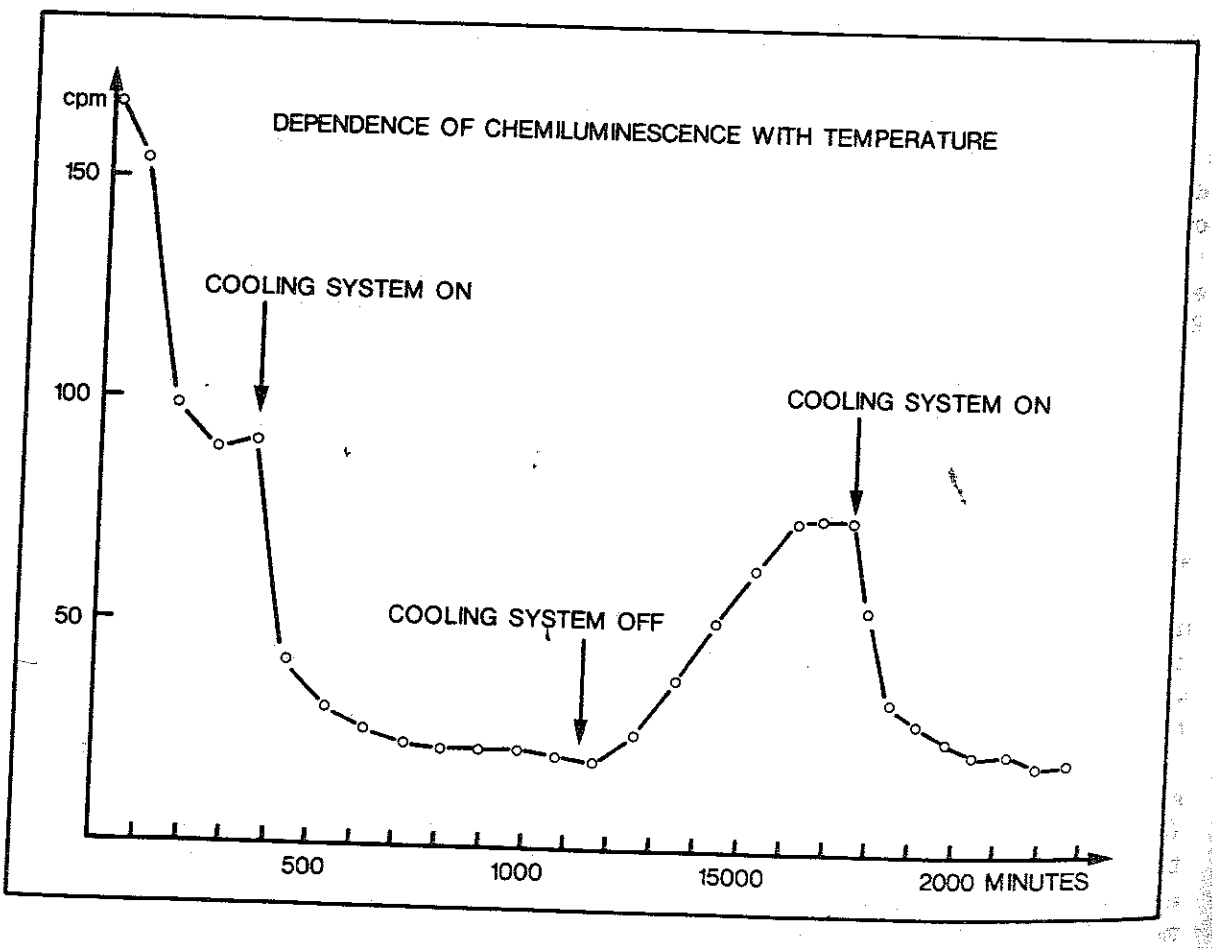
occur, as was learned from experience.

In the urine of some unexposed persons, count rates were measured which could not be attributed to tritium. In the direct measurement of urine, chemiluminescence is generated; this was considered as the major source of the erroneous counts. As a result of pathologic changes of metabolism, metabolites seem to be excreted in the urine of some persons; these react chemically either with each other or with the scintillation agents, causing chemiluminescence which is measured at room temperature.

The presence of chemiluminescence in urine samples was confirmed by experiment. The count rate of a freshly prepared urine sample, as represented in Figure 1, is characterized by a strong decrease with time. After 400 minutes, this count rate has approached a limit which is characteristic for a specific temperature. When the sample is cooled, the count rate again decreases, approaching a value which corresponds to the background count rate. Gradual reheating, even if performed after a longer period of time, again raises the count rate to another limit and subsequent cooling reduces it to background.

Figure 1

Dependence of Chemiluminescence with Temperature



It was not possible to elucidate the cause underlying this effect, since it is not observed with urine samples of all subjects. However, when measurements are carried out at room temperature, this effect falsely indicates tritium activities up to 0.1 μCi of tritium per liter of urine and, consequently, greatly interferes with the measurement. These errors could be reduced by cooling the samples during measurement. However, the appearance of chemiluminescence cannot be precluded even at lower temperatures. Consequently, the samples are measured at a temperature of about + 15°C. In addition, each lot of samples is measured at least three times for about 10 minutes. Count rates corresponding to specific activities below 20 nCi/l are considered with reservation only.

Therefore, the limit of detection was arbitrarily assumed to be 23 nCi of tritium per ml or 1/1000 of the maximum permissible continuous tritium body burden. This limit of detection in radiobioassay can be safely adopted since, according to Bond [2], the initially feared hazard to men by the organically bound form of tritium and the resulting longer action of tritium upon the human organism can be neglected. There is no need in radiobioassay to achieve excessively high sensitivities at considerable expense. In most occupational cases, the body burdens will be higher than 23 nCi/ml.

We use 1 ml of urine and add it directly to the scintillator solution described above. The effect of direct sunlight is avoided to prevent phosphorescence. The samples are measured at least three times, in a cycle, so that a greater time interval exists between these three measurements in which the disturbing effects are allowed to fade away. The duration of the measurement is 10 minutes [1].

In addition to radiobioassay of exposed persons, the surveillance of the unexposed population becomes more and more interesting. Here, urine analyses cannot be performed in the manner described, because it is too insensitive.

We employ the method of liquid scintillation measurement proposed by Lieberman and Moghissi [3] and use Instagel (Packard) as the scintillator solution to which 10 ml of a distilled urine sample can be added. However, we are faced with the difficulty that, meanwhile, there is no more water which is free from tritium. We have adopted the practice of considering pure scintillator solution as the background. The role of chemiluminescence in this low-level range of measurement is still controversial, which means that a final limit of detection cannot yet be stated. Work on this subject is still going on.

Determination of Tritium Equivalent Doses from Urine Analyses

The equivalent dose D for the whole body can be calulated from the tritium concentration A of urine according to equation (1), assuming that tritiated water is uniformly distributed in the body water [1].

$$D\ [\text{mrem/day}] = 0.51 \times A \qquad (1)$$

To obtain the factor 0.51, the value of "effective energy of tritium for total body $\Sigma EF(RBE)n = 0.010$ MeV," as stated in reference [4] (corresponding to a Q-value of 1.7), is multiplied by the converting factors

$3.7 \times 10^4 \hat{=}$ dis/sec per μCi

$1.6 \times 10^{-6} \hat{=}$ ergs/MeV

$8.64 \times 10^4 \hat{=}$ sec/day

$10^{-3} \hat{=}$ kg/g

when A is expressed in μCi/l urine or body water.

If urine samples are analyzed daily for tritium, the annual dose D_a can be calculated, using modified equation (1) for the number of days per year as follows:

$$D_a \text{ [mrem/a]} = 0.51 \times 365 \times A_m \tag{2}$$

where A_m represents the average value of all urine measurements, expressed in μCi/l urine.

The practice of daily urine measurement, as described for instance by Osborne [5], is not applied everywhere. Mostly, there is a period of several days between the individual measurements, and sometimes only random samples are taken at very long intervals. This implies that in calculating the dose, the elimination of tritiated water from the body and a possible new incorporation at a specific time must be taken into account.

For one single incorporation, the total equivalent dose is calculated from one urine analysis by integration of equation (1) over time

$$D_\infty = 0.51 \int_0^\infty A \times e^{-\lambda_b t} \, dt \tag{3}$$

This results in

$$D_\infty \text{ [mrem]} = 0.74 \times T_b \times A \tag{4}$$

where A is the initial tritium concentration in urine, expressed in μCi/l, $\lambda_b = \ln 2 / T_b$ (T_b represents the biological half-life in days) and t is the time in days.

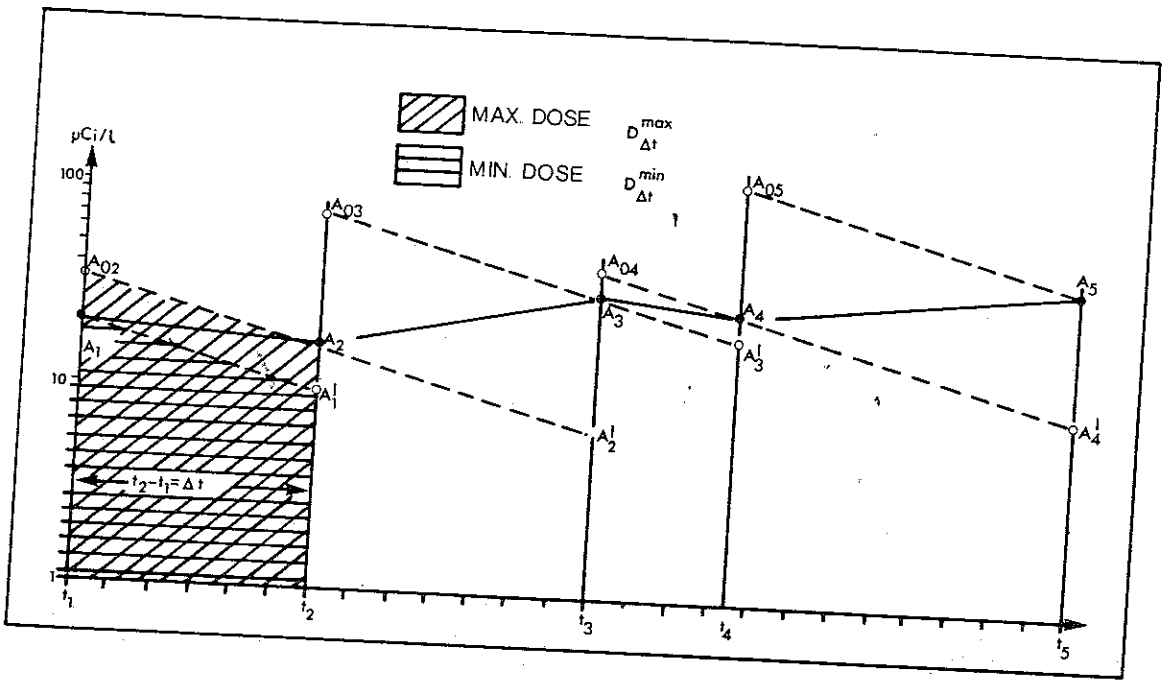
However, in most cases tritium incorporation will not be a unique event but an irregular sequence of individual incorporations. This implies that a dose calculation according to Eq. (4) cannot be performed on the basis of individual analyses and that regular urine analyses must be carried out. Considerations were concerned with the frequency required for such urine analyses to allow adequate radiobioassay and, hence, dose estimate.

If a urine analysis has revealed a certain activity of tritium, this activity will decrease with time according to the biological half-life of tritium. If, upon repetition of the analysis A_1, see Figure 2, a tritium activity is found which is higher (A_2) than the expected value A_1', a pessimistic assumption will lead to the conclusion that an incorporation took place the day after the last urine analysis. It should be large enough (A_{02}) to decrease to the value A_2 found in the second analysis in the interval $t_2 - t_1 = \Delta t$ between the two analyses.

According to the optimistic assumption, the incorporation would have taken place the day preceding the last sampling. This may,

Figure 2

Example for calculation of equivalent doses from tritium concentration in urine



therefore, result in two different dose values, when dose has to be calculated for the period between t_1 and t_2. With this optimistic assumption, there will be a minimum dose because the second control-measurement by urine analysis will not be included in the calculation. This "minimum dose," $D^{min}_{\Delta t}$, is calculated from the value A_1 by modifying equation (3) and taking the time integral between $t_1 = 0$ and $t_2 - t_1 = \Delta t$ only, as described below:

$$D^{min}_{\Delta t} = 0.51 \int_{t=o}^{t=\Delta t} A_1 \, e^{-\lambda_b t} \, dt \qquad (5)$$

$$D^{min}_{\Delta t} = \frac{0.51}{\lambda_b} A_1 \left(1 - e^{-\lambda_b \Delta t}\right) \qquad (6)$$

According to the pessimistic assumption, an incorporation would have taken place the day following the last sampling, which would have resulted in a hypothetic concentration in urine of A_{02}. Thus, from the value A_{02} a maximum dose D^{max}_{t} can be calculated for the time between t_1 and t_2, where the value A_1 can be left unconsidered. A_{02} can be calculated from A_2, using the biological half-life $T_b = \ln 2/\lambda_b$, as follows:

$$A_{02} = A_2 \times e^{\lambda_b \Delta t} \tag{7}$$

The maximum dose $D_{\Delta t}^{max}$ is calculated similarly to equation (5), viz.:

$$D_{\Delta t}^{max} = 0.51 \int_{t=o}^{t=\Delta t} A_{02} \times e^{-\lambda_b t} \, dt \tag{8}$$

This results in

$$D_{\Delta t}^{max} = \frac{0.51}{\lambda_b} A_{02} \left(1 - e^{-\lambda_b \Delta t}\right) \tag{9}$$

and finally, using equation (7), in

$$D_{\Delta t}^{max} = \frac{0.51}{\lambda_b} A_2 \left(e^{\lambda_b \Delta t} - 1\right) \tag{10}$$

The dose values from equations (6) and (10) will be expressed in mrem/Δt, if A_1 and A_2 are specified in μCi/l, and λ_b in day^{-1}.

The difference between the maximum and minimum doses are the greater, the higher, the second value, and the longer the interval Δt between the two urine analyses. A criterion must be found which fixes this interval in such a way as to permit sufficiently accurate radiobioassay and dose estimate.

The biological half-life of tiitium of about 10 days is a reasonable limit to the interval in which to carry out radiobioassay; for this reason, urine samples are examined on a weekly basis at the Karlsruhe Nuclear Research Center if an exposure must be anticipated as a result of work performed. However, routine surveillance is performed monthly.

Calculation of the body doses according to (6) and (10) requires the calculation of e-functions, which involves more expenditure. For this purpose, a programmable desk calculator is used. To make calculations easier, the terms

$$K_{min} = \frac{0.51}{\lambda_b} \left(1 - e^{-\lambda_b \Delta t}\right) \text{ and } K_{max} = \frac{0.51}{\lambda_b} \left(+\lambda_b \Delta t - 1\right)$$

were calculated for T_b = 10 days and for Δt from 1 to 60 days and are compiled in Tables 1a and 1b.

The computing program is conceived in such a way that only the tritium concentrations A_1, A_2, A_3, etc., and the constants K_{min} and K_{max} must be introduced.[1] The constants applicable to the respective interval Δt between the individual analyses can be taken from Tables 1a and 1b. The mean value between maximum and minimum dose is considered to be probably the proper dose, when more specific data on the exact time of incorporation are not known.

With the help of this computing program, the necessary dose calculations are readily performed by one person. However, this

Table 1a

Calculation of K_{min} and K_{max}

Δt (days)	$K_{min} = \frac{0.51}{\lambda_b}\left(1 - e^{-\lambda_b \Delta t}\right)$	$K_{max} = \frac{0.51}{\lambda_b}\left(e^{\lambda_b \Delta t} - 1\right)$
1	0.493	0.528
2	0.952	1.094
3	1.381	1.700
4	1.781	2.351
5	2.155	3.048
6	2.503	1.156,
7	2.828	4.595
8	3.131	5.453
9	3.415	5.066
10	3.679	7.358
11	3.925	8.414
12	4.155	9.546
13	4.370	10.759
14	4.570	12.059
15	4.756	13.453
16	4.930	14.946
17	5.093	16.548
18	5.245	18.263
19	5.387	20.102
20	5.518	22.072
21	5.641	24.185
22	5.757	26.449
23	5.863	28.875
24	5.963	31.476
25	6.057	34.263
26	6.144	37.251
27	6.225	40.452
28	6.301	43.884
29	6.372	47.561
30	6.438	51.502

Calculated on the basis of ($\lambda_b = \frac{\ln 2}{T_b}$ and T_b = 10 days).

Table 1b

Calculation of K_{min} and K_{max}

Δt (days)	$K_{min} = \frac{0.51}{\lambda_b}\left(1 - e^{-\lambda_b \Delta t}\right)$	$K_{max} = \frac{0.51}{\lambda_b}\left(e^{\lambda_b \Delta t} - 1\right)$
31	6.499	55.727
32	6.556	60.255
33	6.610	65.108
34	6.660	70.309
35	6.707	75.883
36	6.751	81.857
37	6.791	88.261
38	6.829	95.123
39	6.865	102.479
40	6.897	110.362
41	6.928	118.811
42	6.957	127.867
43	6.984	137.572
44	7.009	147.974
45	7.032	159.123
46	7.206	171.072
47	7.074	183.878
48	7.094	197.604
49	7.111	212.314
50	7.127	228.081
51	7.143	244.978
52	7.157	263.089
53	7.171	282.500
54	7.183	303.304
55	7.195	325.600
56	7.205	349.498
57	7.216	375.110
58	7.225	402.562
59	7.234	431.982
60	7.242	463.515

Calculated on the basis of ($\lambda_b = \frac{\ln 2}{T_b}$ and T_b = 10 days).

relatively expensive calculation is done only in cases where more than 10 percent of the maximum permissible continuous body burden is measured. If the tritium concentration found in the urine is always less than 10 percent of the maximum permissible body burden, the dose is estimated according to Eq. (2). Using this equation, one may safely assume that an excessive incorporation would have been detected.

Results

In the years from 1967 to 1970, three groups of workers were surveyed for tritium. They included the staff employed at the two heavy water moderated reactors FR 2 and MZFR as well as those in the Decontamination Services.

Body equivalent doses were calculated from urine analyses as described in this paper, and their frequency distribution was divided into five logarithmically graded classes. Table 2 and Figure 3 represent the results of the investigations. Percentages refer to the number of persons examined. It appears as a general finding that in no case the maximum permissible continuous burden of tritium in the body was surpassed.

Due to the smaller quantity of tritium handled in the Decontamination Services, the burden of the staff working there is smaller than with personnel in the reactor operating departments. Only a few of them had body burdens of 1 to 10 percent of the maximum permissible level. The majority were practically unexposed.

The reverse tendency is observed with staff working at the two reactors. The fraction of unexposed persons is low; most of the incorporations fall within the range of 1 to 10 percent of the maximum permissible body burden. It is a noticeable fact that the fraction contributed by the group with 10 to 100 percent maximum permissible body burden is higher in MZFR than in FR 2, although the tritium content of MZFR, as shown in Figure 4, had been lower than in FR 2 over the whole period of surveillance.

The higher exposure was caused by early repair and maintenance work. Initially, repairs had been carried out without the use of respirators. This is the reason for the comparatively high incorporation in MZFR in 1967/68. Respirators are now being used for maintenance work, decreasing the level of incorporations in MZFR since 1969. A new increase in incorporation found in 1970 under identical working conditions is primarily due to the increase in tritium content of the heavy water. This requires more rigorous radiation protection measures. In FR 2, recent major repair work on an experimental heavy water loop is the only explanation for a higher incorporation rate. Hence, a comparison with pure power reactors is not possible. In principle, radiation protection measurements were adequate for all work, which was confirmed by regular radiobioassay of the reactor staff. There was no excessive incorporation.

Table 2

Distribution of Equivalent Doses from Tritium Urine Analyses

	Equivalent Doses mrem/a	Distribution in percentage of surveyed persons			
		1967	1968	1969	1970
FR 2	<5	0.0%	15.4%	4.4%	1.1%
	5 - 50	27.5%	20.5%	19.6%	33.8%
	50 - 500	47.5%	41.0%	60.8%	53.5%
	500 - 5000	25.0%	23.1%	15.2%	11.6%
	>5000	0.0%	0.0%	0.0%	0.0%
number of persons		40	39	46	86
MZFR	<5	2.5%	0.7%	5.9%	0.0%
	5 - 50	12.5%	5.6%	24.8%	11.0%
	50 - 500	62.5%	45.8%	55.0%	70.0%
	500 - 5000	22.5%	47.8%	14.3%	19.0%
	>5000	0.0%	0.0%	0.0%	0.0%
number of persons		80	144	238	109
Decont. facility	<5	72.0%	54.3%	70.0%	82.8%
	5 - 50	21.3%	35.4%	27.1%	14.3%
	50 - 500	6.6%	10.3%	2.9%	2.9%
	500 - 5000	0.0%	0.0%	0.0%	0.0%
	>5000	0.0%	0.0%	0.0%	0.0%
number of persons		61	48	60	70

Figure 3

Distribution of equivalent doses to man
from tritium-urine-analyses during 1967-1970
(in percentage of surveyed persons)

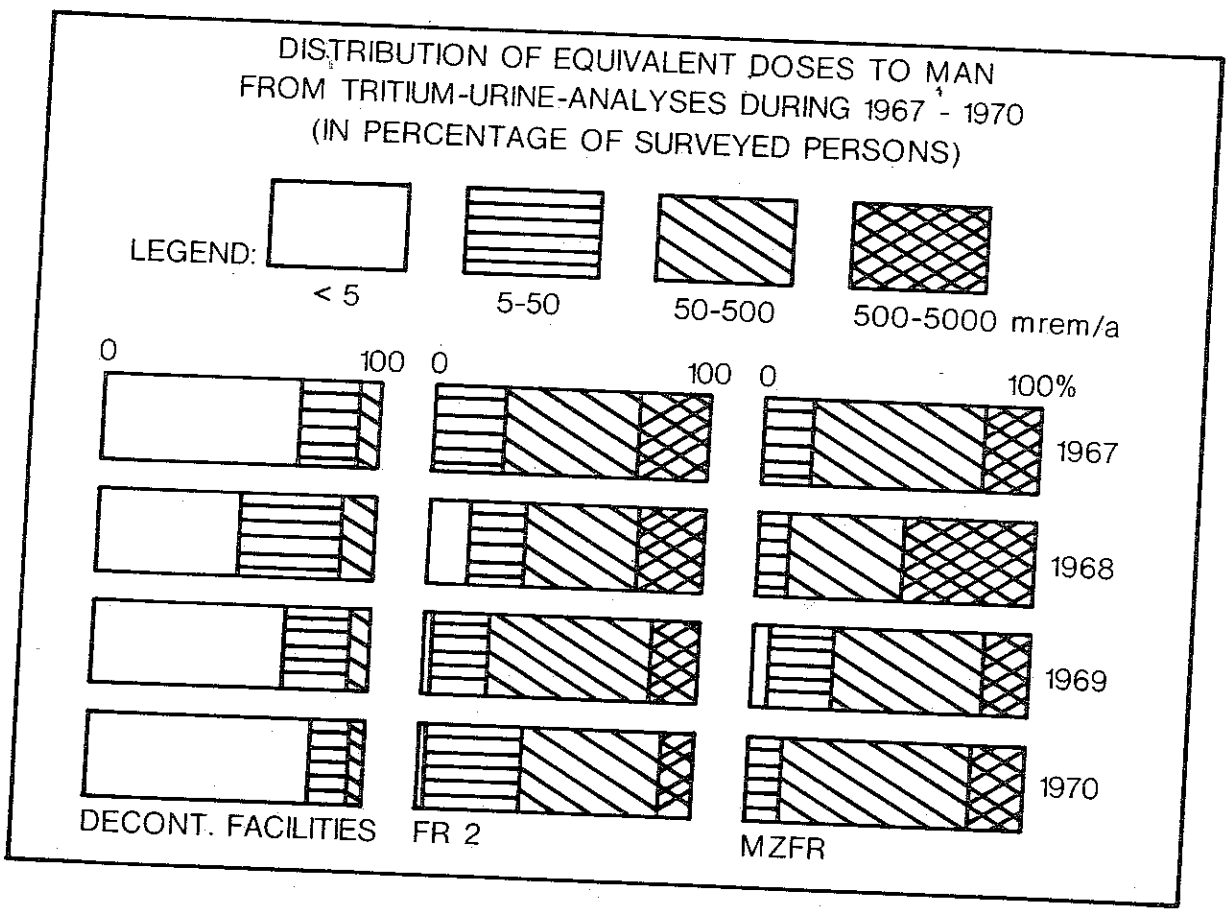
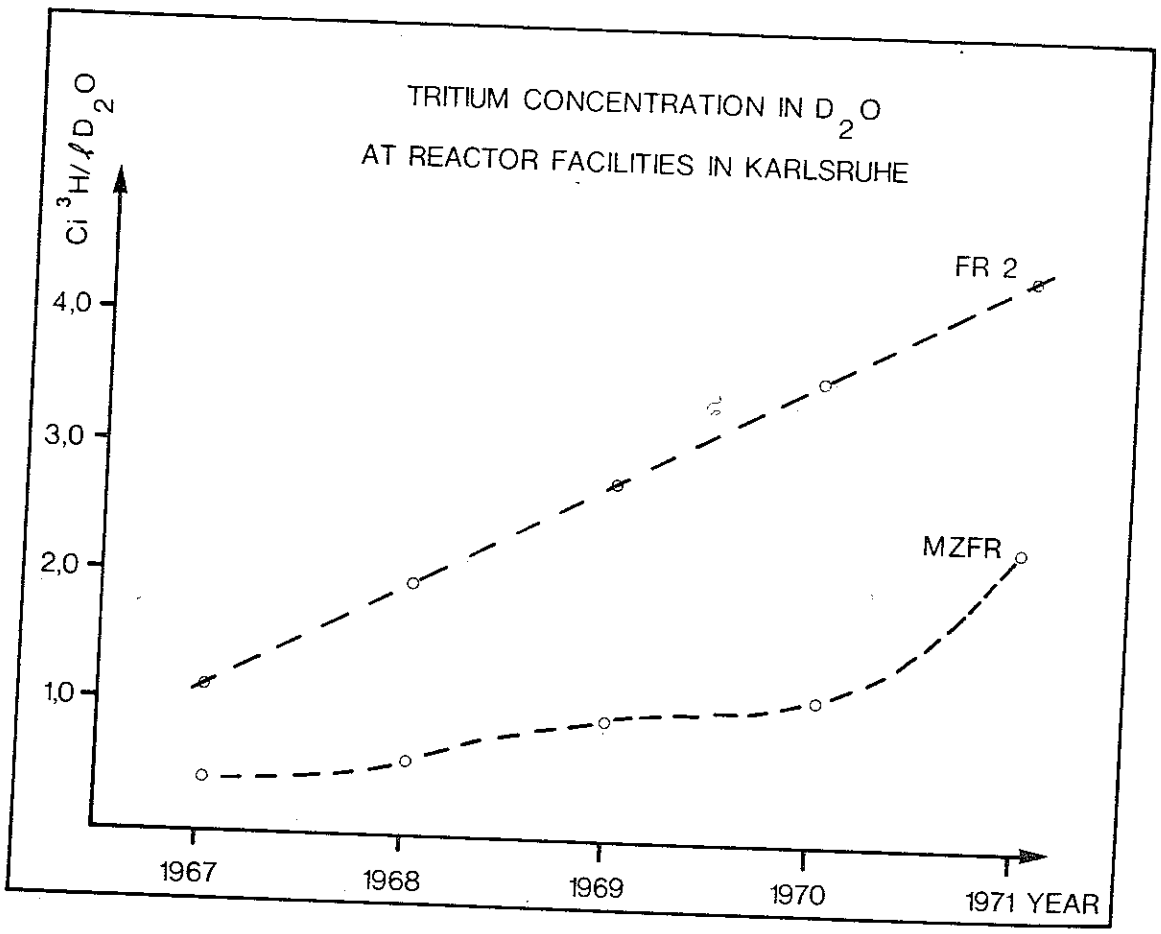


Figure 4

Tritium concentration in D_2O at reactor facilities in Karlsruhe



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