RELATIONSHIPS EXISTING BETWEEN TRITIUM RELEASES FROM DIFFERENT SOURCES AND THE CONTAMINATION OF AIR, WATER AND PLANTS

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1. INTRODUCTION

Several thousands curies of tritium are released every year from the Karlsruhe Nuclear Research Center via the effluent air and water (1, 2). Therefore, it is obvious to investigate thoroughly the consequences of these releases. The liquid scintillation technique was chosen as the method of measurement. It offers the advantage that a great number of samples can be measured at relatively low expenditure. However, within the range of background concentrations, the accuracy of measurements is not very high, which is a drawback of this method. Nevertheless, it is adequate in this range to fulfill the main objective of these measurements, namely monitoring of the environmental radioactivity with a view to radiation protection.

2. TRITIUM EMISSIONS FROM THE KARLSRUHE NUCLEAR RESEARCH CENTER

Tritium is released to the atmosphere mainly via several exhaust stacks and it is almost exclusively in the form of tritiated water vapor. Different forms of tritium released have not yet been detected and are estimated to be insignificant. In the effluent water tritium from all sources in the Nuclear Research Center is discharged via the sewage treatment plant (3). Passing a 2.9 km long sewer, the liquid effluents reach the 'Altrhein', a former branch of the river Rhine and then they flow, mixed with surface water, over a distance of 23.6 km to be discharged into the river. Table 1 shows the tritium releases for the years 1975 and 1976 and the different emitters of the Nuclear Research Center as well as the heights of sources above the ground. In case that direct measurement is impossible, tritiated water vapor released to the atmosphere is measured by means of the liquid scintillation measurement technique on continuously condensed humidity samples of exhaust air (4).

3. MEASUREMENT PROGRAMS AND MEASUREMENT RESULTS

To be able to record contaminations of the environment caused by discharges, it was necessary to determine the present background due to fallout. A comprehensive measurement program serves this purpose, which covers essentially the region of the Upper Rhine Valley between Kehl and Mannheim. Details of this long-term program were reported recently in two publications (2, 5).

Diffusion of the tritiated water vapor released into the atmosphere can be calculated with a satisfactory accuracy. To determine the site specific parameters to be employed in the diffusion calculation (6, 7), diffusion experiments were carried out at the Karlsruhe Nuclear Research Center in which HTO and, subsequently, inactive chemical compounds had been used as the tracers (8-11). The most significant results of previous diffusion experiments has been that at a 100 m emission height the concentration maxima get considerably shifted toward the source as a result of surface roughness and that they are higher in amount than calculated with the help of the dispersion parameters used so far (6, 7).

To the extent of manpower availability, special measurement programs were performed the main results of which will be reported here. One example will be treated in more detail. The positions of the main tritium emitters and of sampling locations for the two programs described here are indicated in Fig. 1.

Emitter	Height of Emission in m	Tritium Release in Ci 1975	1976
FR 2 (44 MW_{th}, D_2O-moderated)	99	285	170
MZFR (200 MW_{th}, D_2O-moderated)	99.5	765	703
WAK (Reprocessing Plant, 40 t/a throughput, $3x10^4$ MWd/t)	60	1000*	1000*
FERAB (incineration facility for solid radioactive wastes)	70	467	213
Decontamination plant for liquid wastes	19	53	73
Sewage treatment plant			
a) evaporations	0	∿2**	∿3**
b) liquid effluents	-	2821	4024
Several minor emitters	∿10	5	20

Table 1 Tritium releases by the Karlsruhe Nuclear Research Center in 1975 and 1976.

*authorized value

**estimate based on average evaporation rate of 1 mm/d for an 800 m^2 uncovered surface

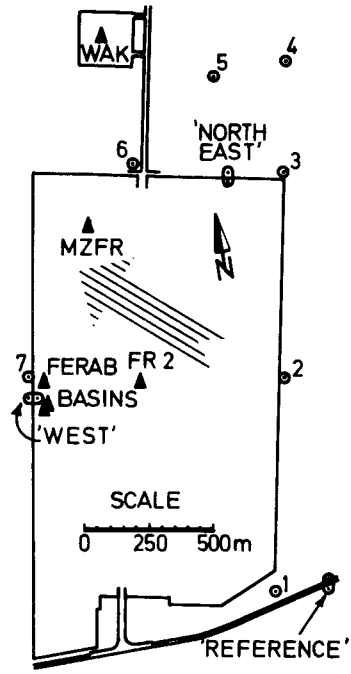

Fig. 1 Site plan indicating the main tritium emitters of the Karlsruhe Nuclear Research Center ▲ and the sampling locations covered by the initial program ⊙ and the follow-on program ⊂⊃, respectively. ≡≡ direction of ground water flow

3.1 Initial Program Covering Seven Sampling Locations at the Periphery of the Karlsruhe Nuclear Research Center

Within a measurement program limited in time the tritium concentration in the tissue water of plant samples was determined at seven sampling locations and at two of them also in the humidity of the air. The results have been summarized in Table 2. For the sake of clarity, only maximum and minimum values are indicated. Some of the individual values agree very well for samples of different types of plant taken at one sampling location, but partly they also show deviations by 30 - 50 % from each other. In very rare cases the ratios of concentration values are greater than the factor 2. According to expectations, the sampling location 1 yielded the lowest results because relative to the main emitters it lies outside the main wind directions. By contrast, clearly higher values were found at the sampling locations 3 to 6 whose positions relative to the emitters are in one of the two main wind directions. Last but not least, the highest values were measured at the sampling location 7 in the vicinity of the final basins of the sewage treatment plant.

3.2 Follow-on Program Covering Three Selected Sampling Locations

In another program limited in time the tritium concentrations were measured simultaneously at three sampling locations of pine (pinus sylvestris) and spruce (picea abies) needles, hornbeam (carpinus betulus) leaves, the humidity of the air, precipitations and of the ground water, if applicable. The samples were taken on working days only.

One sampling location lies in a region in which the ground water is contaminated (12). It is termed here sampling location 'West'. In the same region, also the humidity of the air is contaminated by the evaporation of chemical effluent water. The second location, termed 'North-east,' lies in the north-eastern part of the Karlsruhe Nuclear Research Center in the main wind direction relative to the tritium releasing FR 2 and MZFR reactors. At this point a back-

Sampling Location	Sampled Material	Tritium Concentration pCi/ml maximum	minimum
1	grass	2.9	0.3
	foliage	2.3	0.6
	air humidity	1.6	0.1
2	foliage	3.5	0.7
3	foliage	17	0.8
4	foliage	10	0.8
5	grass	5.3	0.8
	foliage	7.8	0.7
	air humidity	7.9	0.6
6	foliage	3.2	1.0
7	grass	21	1.9
	foliage	29	1.5

Table 2 Results of simultaneous measurements of tritium concentration at seven selected sampling locations (18 samples each). The values were measured in the period from August 1 to September 23, 1975.

Sampling Location		Air Humidity pCi/ml	Precipitations pCi/ml	Precipitations nCi/m²/d	Ground Water pCi/ml	Needles pCi/ml pinus sylvest.	Needles pCi/ml picea abies	Foliage pCi/ml carpinus
Reference Location	Maximum	6.2±0.4 (2.8±0.3)*	3.5±0.3	25±2	0.59±0.17	3.0±0.3	2.8±0.3	1.6±0.2*
	Minimum	0.41±0.21 (0.2±0.2)*	0.2±0.2	0.07±0.02	< 0.15	1.0±0.2	0.84±0.20	0.88±0.17*
Sampling Location 'West'	Maximum	455±13 (47.4±1.5)*	48.8±0.5	124±4	576±16	61.5±1.9	45.2±1.4	70+2*
	Minimum	2.7±0.2 (4.1±0.3)*	1.5±0.2	0.56±0.10	37.6±2.2	13.1±0.6	2.5±0.3	15.2±0.6*
Sampling Location 'North-east'	Maximum	37.5±1.3 (5.4±0.5)*	4.5±0.3	27±2	not measured,	9.7±0.5	12.6±0.6	4.9±0.3*
	Minimum	0.5±0.2 (0.6±0.2)*	0.40±0.17	0.12±0.05	background expected	2.0±0.2	2.4±0.2	2.3±0.2*

Table 3 Results of simultaneous measurements of tritium concentration in air humidity, precipitations, ground water, foliage as well as in needles at selected sampling locations. The values were measured in the periods from March 3 to April 8, 1976 and from May 3 to May 26, 1976. The air humidity samples the measured values of which are given in parentheses, are taken at 15 m height above ground. The values marked by an asterisk refer only to May 1976.

ground level of tritium concentration has to be expected in the ground water. The third location, termed 'Reference', was so selected that it is exposed but rarely to the wind coming from the tritium emitters. Besides, it is opposed to the direction of ground water flow relative to the Nuclear Research Center.

The results of measurements have been compiled in Table 3. Again only the maximum and minimum values of tritium concentrations are indicated. In qualitative terms, the results correspond to that in Table 2.

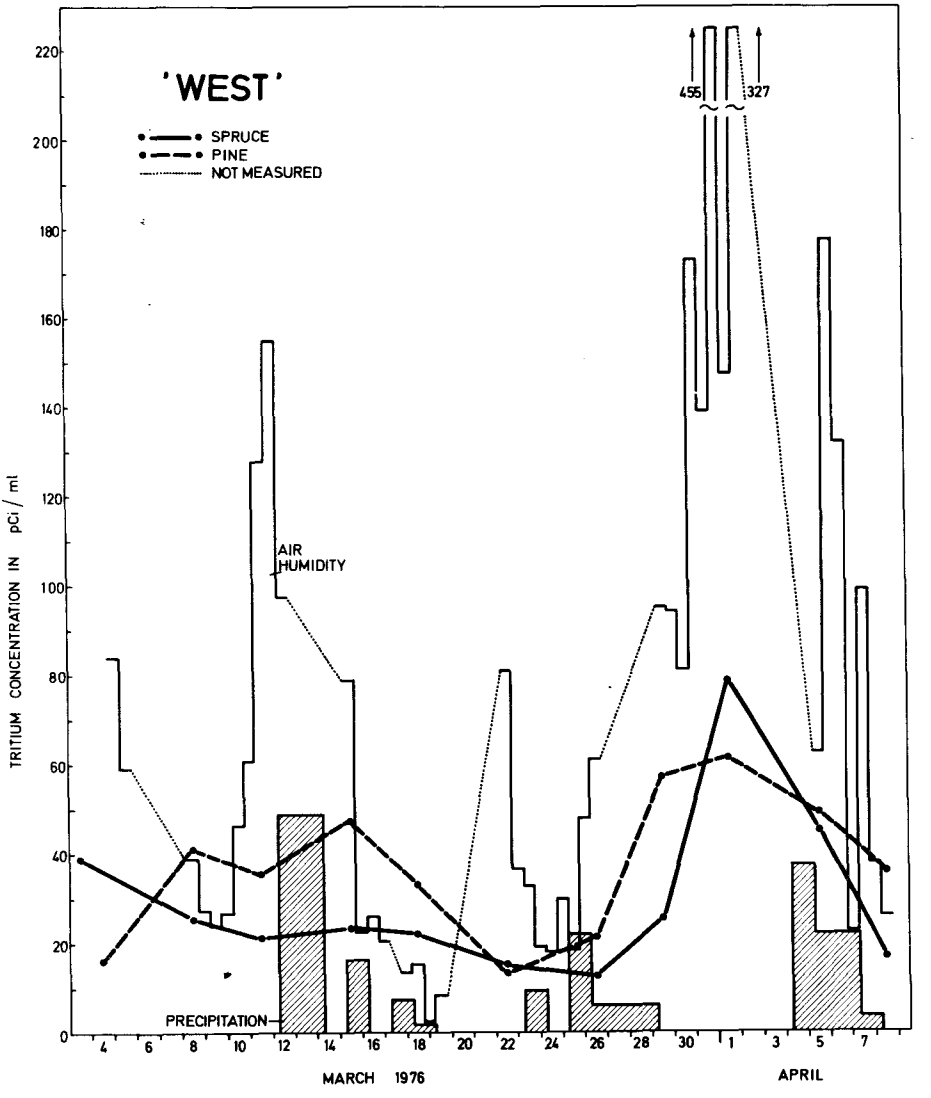

Fig. 2 Representation of the variation with time of the tritium concentration in spruces, pines, air humidity and precipitations, measured at the sampling location 'West' (see Fig. 1).

Fig. 2 shows the results obtained at the sampling location 'West' for the period from March 1 to April 7, 1976. The highest tritium concentrations were found at this sampling location. The results unambiguously show that the precipitation is not the main cause of tritium contamination of the needles. Since around the sampling location 'West' the ground water had been contaminated by about 400 pCi/ml of tritium within the period of reporting, both this contamination and that of the humidity of the air must be considered as causes of pine and spruce needle contamination. However, the rapid variations with time observed for the tritium contamination in tissue water, which correspond to that of the humidity of the air, indicate that the humidity of the air is one of the reasons of contamination.

Since the roots of pines extend to much lower depths than that of spruces, the contamination of the ground water should become apparent in differing tritium contaminations of the two types of needles. As a matter of fact, the expected differences can be observed. Due to the close vicinity of the final basins, where the tritium bearing effluent waters evaporate at ground level, the branches of spruces growing near the ground level (sampling height about 2 m) are exposed to higher tritium concentrations of the humidity of the air than the branches of pines beginning to grow at an about 12 m higher level. The fact that the higher tritium concentrations were generally found in pine needles also makes visible the influence of ground water contamination. However, the results measured in early April 1976 (see Fig. 2) likewise show that the contamination of spruces can be higher than that of pines in case of very high air humidity concentration near the ground level.

Fig. 3 shows the measured values for the sampling location 'North-east' applicable to the same time interval. As a result of tritium emissions from the exhaust stacks, the tritium concentrations of the humidity of the air are sometimes considerably enhanced, to which both types of needles are equally exposed. The ground water not contaminated at this place seems to dilute in this example the tritium concentration in pine needles.

At the so-called 'Reference' location neither the ground water is contaminated nor does a comparably high contamination of the air humidity prevail. For this reason, differing contaminations of pine and spruce needles have not to be expected here which is confirmed by Fig. 4. The observations made at the 'Reference' location do not allow a conclusion to be drawn relative to the path via which the needles measured had been contaminated.

The time plot of variations of the tritium concentration in tissue water allows to make conclusions with respect to the time constant and the half-life, respectively, which determines this plot. Both for pine needles and for spruce needles time constants and half-lives, respectively, of 4 ± 2 d are found for the increase and reduction of the tritium concentration. The time sequence of measurement points and the experimental conditions were not sufficient to make a more accurate statement. In case of soil contamination by precipitation a slower reduction with time can be expected whereas rapid changes can be associated with the humidity of the air.

Besides the relationship existing between the tritium contaminations of the humidity of the air and of plants the relationship between the tritium emissions and the tritium contamination of the humidity of the air is also of some interest. In Fig. 5 the product of tritium emission rate and of the frequency of exposure to tritium from the individual emitters has been represented for the three sampling locations. The frequency is that at which the wind reaches the sampling location when it blows from an emitter. Since one day has been chosen as the unit of time, no high expectations should be made with respect to such a representation. Moreover, the diffusion category has not been

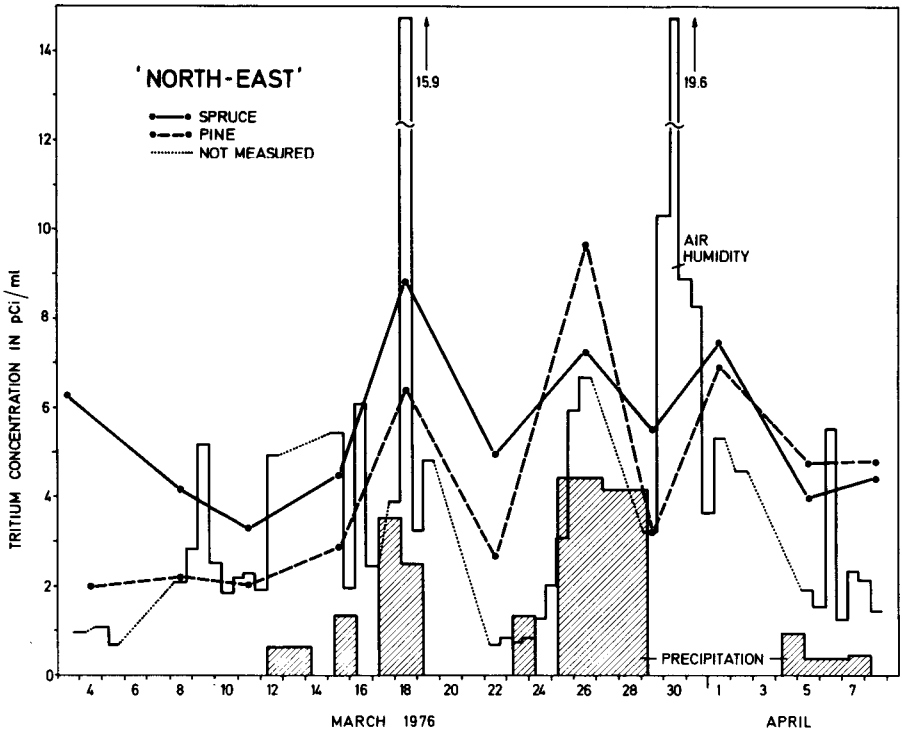

Fig. 3 Representation of the variation with time of the tritium concentration in spruces, pines, air humidity and precipitations, measured at the sampling location 'North-east' (see Fig. 1).

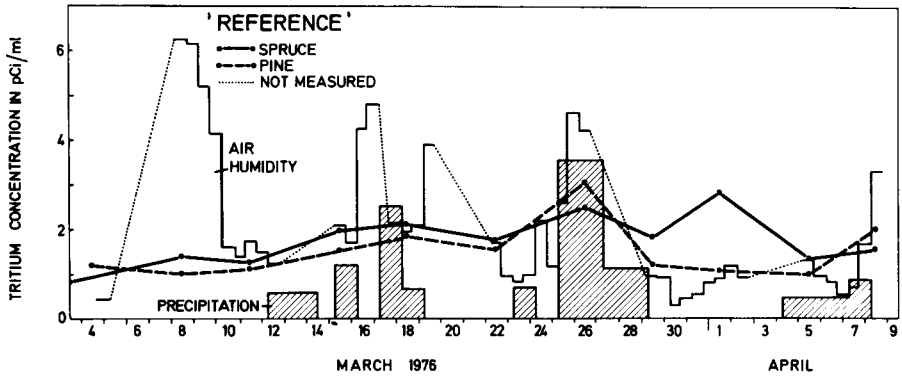

Fig. 4 Representation of the variation with time of the tritium concentration in spruces, pines, air humidity and precipitations, measured at the 'Reference' sampling location (see Fig. 1).

taken into account in this representation, which has a considerable influence on the concentration prevailing on the soil. Comparison of Fig. 5 with Figs.2 to 4 shows that some peak values visible in Fig. 5 are matched by peak values found in the representations of the humidity of the air.

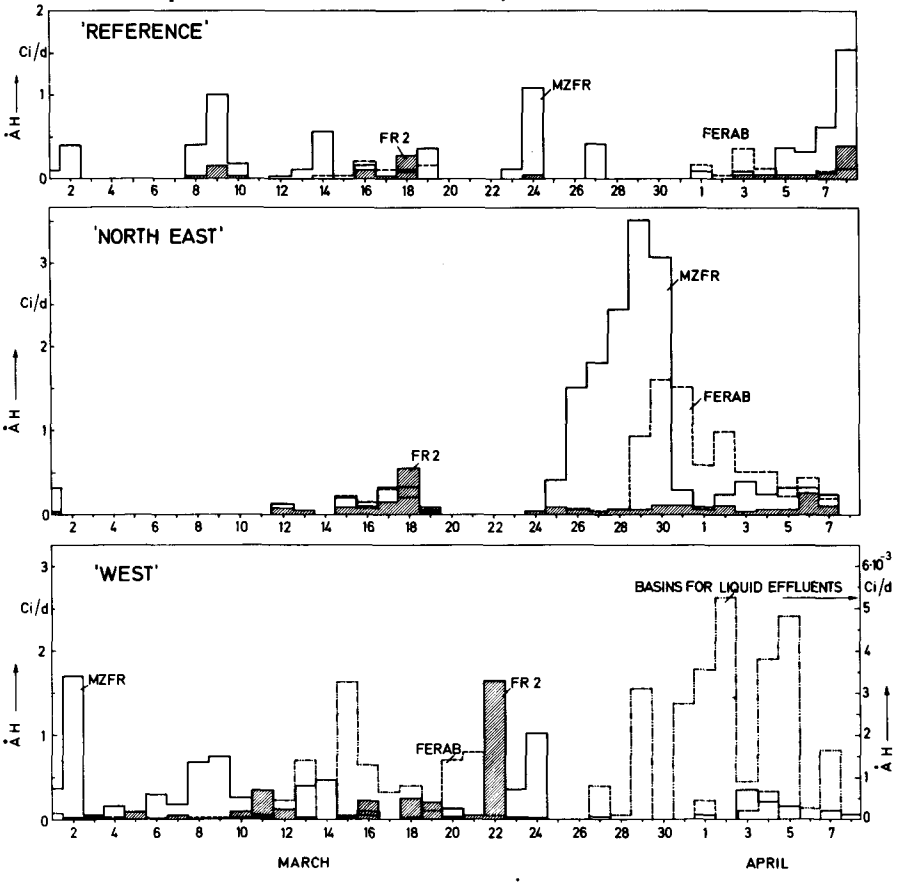

Fig. 5 Products formed of the tritium emission rate (A) of the main emitters of the Karlsruhe Nuclear Research Center and of the frequency (H) of those 10 minutes intervals on a day in which exposure of the sampling location under consideration was possible on account of the wind direction.

Also the days are of interest on which the sampling locations are not exposed at all to tritium from the emitters and yet tritium has been found in the humidity of the air. This has to be explained partly by contamination of the vegetation and the soil both of which release tritium by transpiration also on such days. The fact that a peak value of tritium concentration was observed in needles at the sampling location 'North-east' on March 26, 1976, without a corresponding peak value detected in the humidity of the air, is probably due to the fact that a peak value of tritium concentration in the air humidity was missed.

The difficulties in the evaluation of measured values are still increased by the dependency on height of the wind direction. Experience gathered at the site has shown that the wind direction can change substantially with the height and that eddies caused by buildings and vegetation essentially influence the diffusion of pollutants in the atmospheric layer near the

ground. So, the tritium concentration in the humidity of the air can be dependent on the height.

It can be concluded from results available so far that the tritium concentration of plants is largely determined by the tritium content of the humidity of the air. It has to be clarified by which path the tritium is taken up by the plants.

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