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Abteilung Strahlenschutz und Sicherheit

Tritium in Water and Plants —Summary Report of Results Obtained in a Measuring Program of Several Years Duration—

L. A. König, M. Winter

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Tritium in Water and Plants*

- Summary Report of Results Obtained in a Measuring Program of Several Years Duration -

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*Part of a Co-ordinated Program of Research under the Sponsorship of the IAEA (Research Agreement No. 1302/CF)

Summary

A summary is presented of the results obtained in programs on the measurement of the tritium concentration in the immediate and more distant environment of the Karlsruhe Nuclear Research Center. The main objectives of these programs consisted in the determination of the environmental burden caused by tritium released from the Karlsruhe Nuclear Research Center as well as in the study of the radioecological behavior of tritium.

The amount of tritium released with the waste water and the exhaust air from the Karlsruhe Nuclear Research Center in the years 1969 until 1976 is indicated. The total releases ranged from 2000 Ci/a to 5200 Ci/a in the period of reporting. The fractions contained in the exhaust air and in the waste water were subjected to considerable variations over the years.

The results of measurements for water samples are presented as annual mean values. The annual values of measured results obtained for precipitations on the site clearly exceed the values measured at sampling stations outside the Karlsruhe Nuclear Research Center. Of the surface waters monitored the rivers Rhine and Neckar showed the highest tritium values. In smaller flowing waters the tritium concentration is also influenced by the percentage of waste water from communities. Obviously the tritium content of the ground and drinking water depends on the depth of sampling. Drinking water raised from a small depth in the vicinity of the Rhine is subjected to the same variations as the water from the river Rhine.

To find out relations to the tritium offer of the relevant media close to the plants, the tritium concentrations in tissue water of plants and in air humidity, ground water and precipitations were investigated. Variations of the tritium concentration in air humidity correlate with the variations of the tritium concentration in the tissue water of plants. The tritium concentration level in the tissue water of plants is close to the tritium concentration in air humidity. The following time constants and half-lives, respectively, are found: for oak and hornbeam leaves 2±1 days, for spruce needles 3±1.5 days, for pine needles 6±3 days.

The dispersion of tritium released into the air and into the water is dealt briefly.

Tritium in Wasser und Pflanzen

- Zusammenfassender Bericht über Ergebnisse eines mehrjährigen Meßprogrammes -

Zusammenfassung

Es wird eine Zusammenfassung der Ergebnisse aus Meßprogrammen zur Bestimmung der Tritiumkontamination in der näheren und weiteren Umgebung des Kernforschungszentrums Karlsruhe gegeben. Hauptziele dieser Programme waren die Ermittlung der Umgebungsbelastung infolge der Tritiumfreisetzungen aus dem Kernforschungszentrum Karlsruhe und das Studium des radioökologischen Verhaltens von Tritium.

Die Höhe der Tritiumfreisetzungen aus dem Kernforschungszentrum Karlsruhe in Abluft und Abwasser wird mitgeteilt. Die totalen Freisetzungen lagen im Berichtszeitraum zwischen 2000 Ci/a und 5200 Ci/a. Die Anteile der Freisetzungen in Luft und Wasser schwankten dabei erheblich.

Die Ergebnisse der Messungen an Wasserproben werden als Jahresmittelwerte dargestellt. Die im Kernforschungszentrum gesammelten Niederschlagsproben zeigen gegenüber den außerhalb gesammelten Proben deutlich erhöhte Werte. Von den Oberflächengewässern weisen Rhein und Neckar die höchsten Tritiumwerte auf. In kleineren Fließgewässern wird die Tritiumkonzentration vom Abwasseranteil aus Gemeinden beeinflußt. Offensichtlich hängt der Tritiumgehalt des Grund- und Trinkwassers von der Tiefe der Probenahme ab. Trinkwasser aus geringeren Tiefen aus der Nähe des Rheins zeigt die gleichen zeitlichen Schwankungen wie das Rheinwasser.

Um Beziehungen zwischen dem Tritiumangebot aus den Medien in der unmittelbaren Umgebung der Pflanzen zu der Tritiumkonzentration in Pflanzen zu finden, wurden gleichzeitig die Tritiumkonzentrationen in Luftfeuchte, Grundwasser und Niederschlägen mituntersucht. Schwankungen der Tritiumkonzentration in der Luftfeuchte korrelieren mit jenen im Gewebewasser. Der Konzentrationspegel im Gewebewasser der Pflanzen liegt in der Nähe jener in Luftfeuchte. Folgende Zeitkonstanten bzw. Halbwertszeiten wurden gefunden: für Eichen- und Hainbuchenblätter 2±1 Tage, für Fichtennadeln 3±1,5 Tage und für Kiefernnadeln 6±3 Tage.

Die Ausbreitung von über den Luft- und Wasserpfad freigesetztem Tritium wird kurz behandelt.

Contents

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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. Therefore, it is obvious to investigate thoroughly the consequences of these releases in a comprehensive monitoring program. This program is extended beyond the immediate and more distant environment of the Karlsruhe Nuclear Research Center. The main objectives of the program consist in the determination of the environmental burden by tritium released from the Karlsruhe Nuclear Research Center as well as in the study of the radioecological behavior of tritium.

Due to the shortage of manpower it was not possible to establish a long-term program for tritium measurements in plants. However, it was possible to set up several ad hoc programs carried out by some guest scientists and students.

The liquid scintillation technique was chosen as the method of measurement for all samples. It offers the advantage that a great number of samples can be measured at relatively low expenditure.

At various previous occasions results were reported of the measurements carried out within the framework of the monitoring program [1-12]. This paper is a summary of the most important results from these reports and it will be presented at the IAEA Research Coordination Meeting in Mexico, October 3-7, 1977.

2. Tritium Emissions from the Karlsruhe Nuclear Research Center

Tritium is released to the atmosphere mainly via several exhaust stacks and is almost exclusively in the form of tritiated water vapor. Different forms of tritium released have not been detected and are estimated to be insignificant. In the effluent water tritium from all sources at the Nuclear Research Center is dis-

Pathway	Tritium release in Ci							
	1969	1970	1971	1972	1973	1974	1975	1976
exhaust air	1600	2100	1900	1200	1900	1600	1600	1200
liquid effluents	440	602	735	2226	1580	770	2821	4024

Table 1 Tritium releases from the Karlsruhe Nuclear Research Center via effluent air and water for the period from 1969 to 1976.

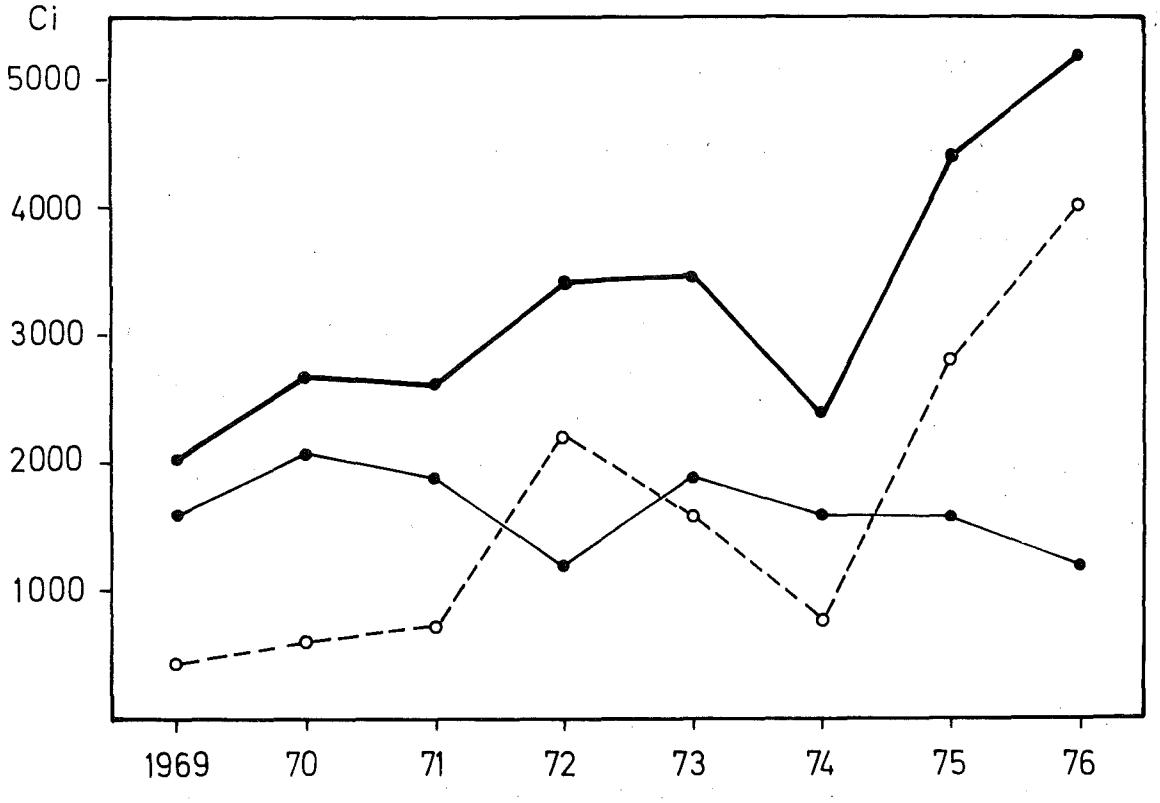

Fig. 1 Tritium releases from the Karlsruhe Nuclear Research Center for the period from 1969 to 1976.

——— total releases
——— releases in exhaust air
- - - - releases in waste water

charged via the sewage treatment plant. 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 and Fig. 1 give a survey of the tritium release via effluent air and water for the period from 1969 to 1976. Table 2 is a breakdown of releases referring to the different emitters of the Nuclear Research Center and it indicates the heights of the sources above the ground.

Comprehensive monitoring routine programs have been carried out to determine all the tritium releases from the different facilities of the Center.

In case that direct measurement [13, 14] is impossible, tritiated water vapor is measured by means of the liquid scintillation measurement technique on continuously condensed humidity samples of exhaust air [15].

3. Tritium in Water

In this chapter measuring results are reported of tritium in ground, drinking and surface waters and in precipitations. Because of the abundance of data only annual mean values are given. More details can be found in earlier publications [1, 8, 11]. Table 3 gives a breakdown by types of samples of the present state of the program. More of the sampling locations for the types of water sample mentioned above are shown in Figs. 2 to 5.

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) evaporation b) liquid effluents	 ∿2 -	 ∿2** 2821	 ∿3** 4024
Several minor emitters	∿10	5	20

Table 2 Tritium releases by the Karlsruhe Nuclear Research Center in 1975 and 1976 [9]

*authorized value

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

Type of sample	Number of sampling locations		Frequency of sampling	Samples investigated number	Samples investigated percent
ground water	28	2 21 4 1	weekly monthly quarterly semiannually	373	13.4
drinking water	23	23	twice per month	446	16.0
surface water	58	10 45 3	weekly twice per month annually	1 558	55.8
precipitations	14	3 2 6 3	each working day after precipitation three times per month twice per month monthly	414	14.8
t o t a l	123			2 791	100

Table 3 Survey of the monitoring program (status 1977)

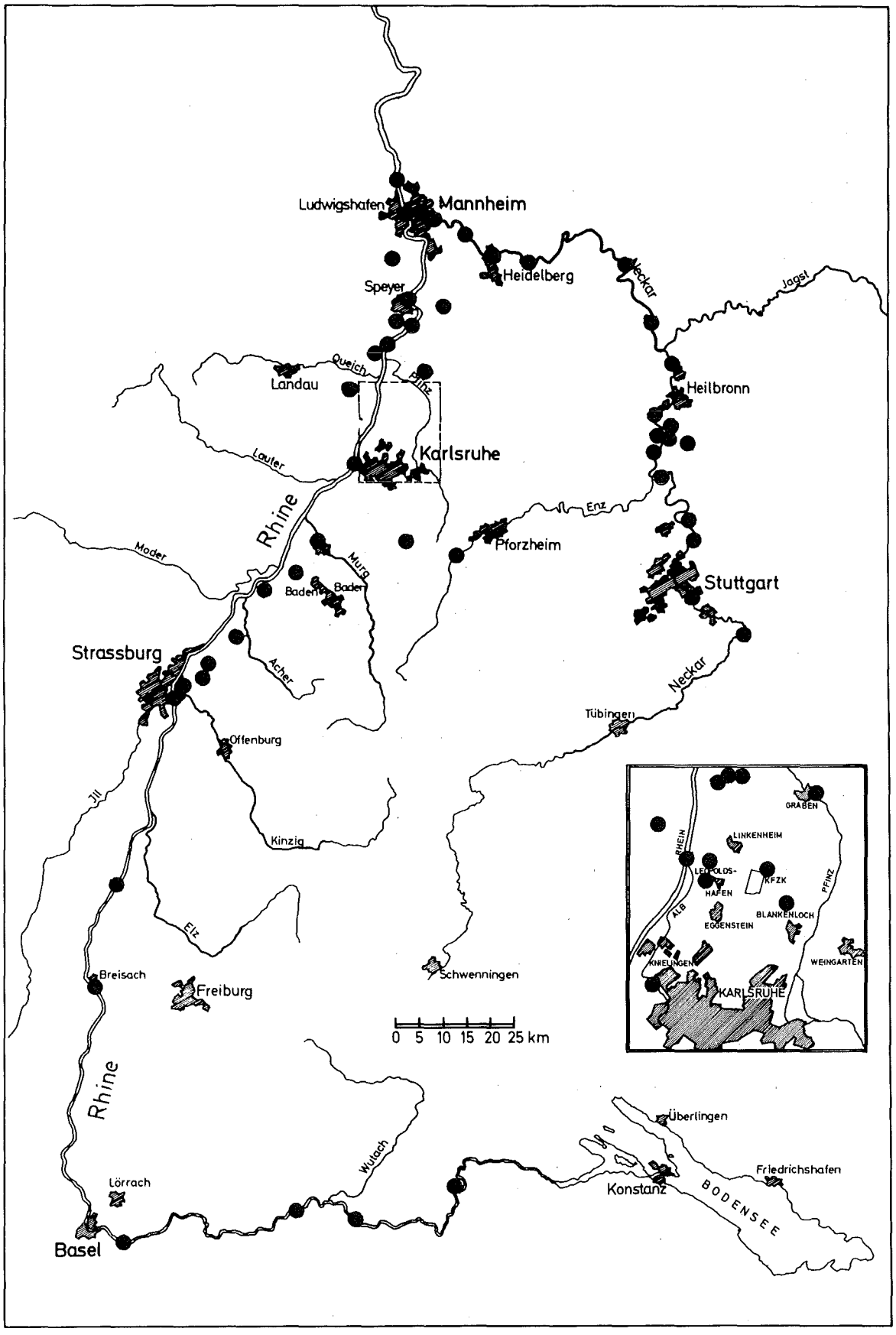

Fig. 3 Map of sampling locations for surface water
(KFZK: Karlsruhe Nuclear Research Center)

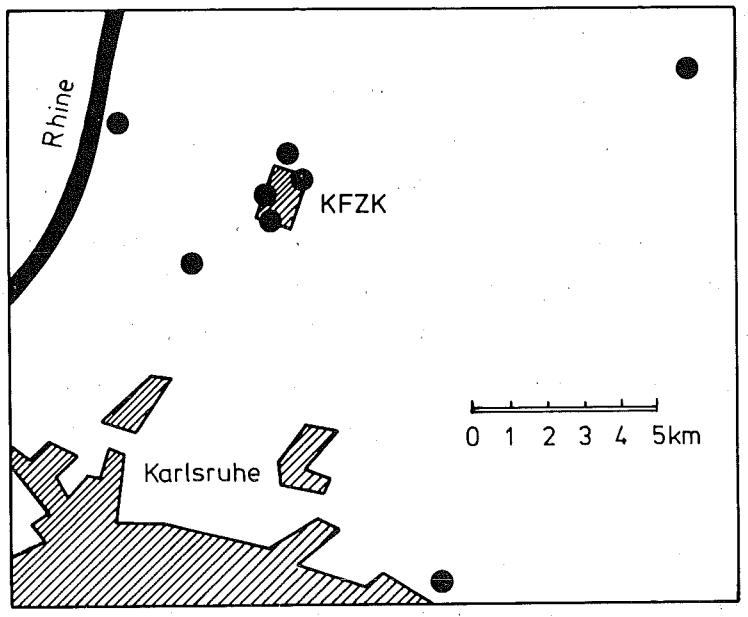

Fig. 2 Map of sampling locations of precipitations (KFZK: Karlsruhe Nuclear Research Center)

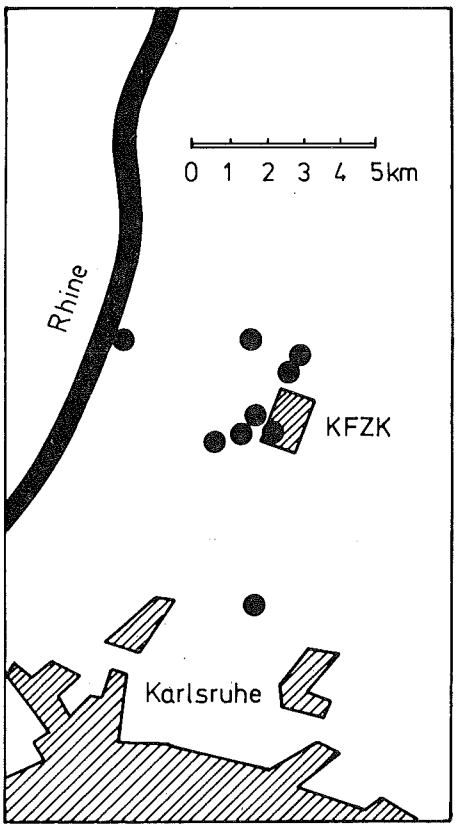

Fig. 4 Map of sampling locations for ground water

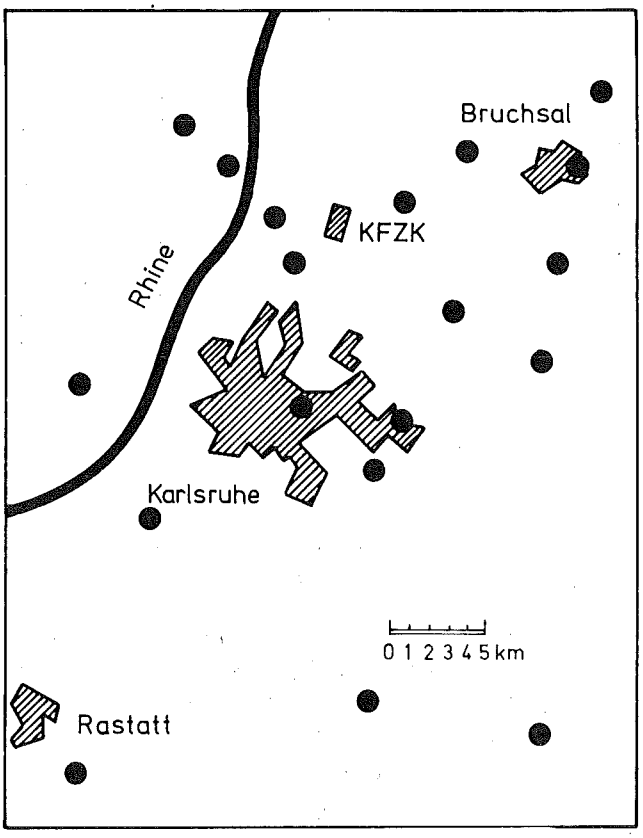

Fig. 5 Map of sampling locations for drinking water

3.1 Tritium in Precipitations

Precipitation samples (see Fig. 2) are of special interest because their tritium concentration together with the amount of precipitation govern the tritium activity fed into the soil and, consequently, have an impact on all the other media. Table 4 is a compilation of values measured in the years 1973 until 1976. The mean values of surface load at the four collecting stations located on the site of the Karlsruhe Nuclear Research Center clearly exceeded the values for the more distant environment (see Figs. 6 and 7). This is due to the tritium emissions from the Nuclear Research Center.

Establishing the difference of surface load values applicable on the site and outside the Nuclear Research Center gives a mean surface load of about 0.3 Ci/km^2 due to releases. The major part of surface load is caused by fall-out.

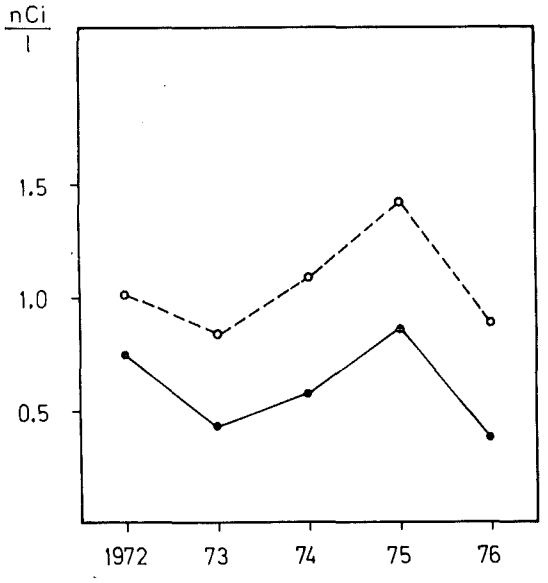

Fig. 6 Mean values of tritium concentration in precipitation for the period from 1972 to 1976

----- within KFZK
——— outside KFZK

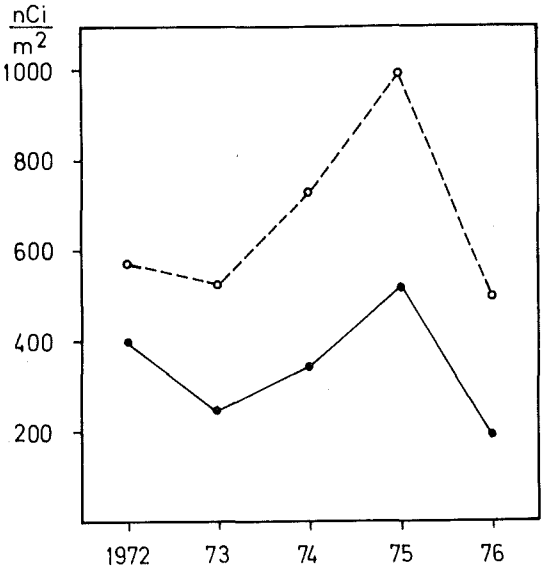

Fig. 7 Mean values of tritium surface load for the period from 1972 to 1976

----- within KFZK
——— outside KFZK

Locations of precipitation collectors	1972			1973			1974			1975			1976		
	M mm	$\bar{a}$ nCi/l	A_F nCi/m^2	M mm	$\bar{a}$ nCi/l	A_F nCi/m^2	M mm	$\bar{a}$ nCi/l	nCi/m^2	M mm	$\bar{a}$ nCi/l	A_F nCi/m^2	M mm	$\bar{a}$ nCi/l	A_F nCi/m^2
within Karlsruhe Nuclear Research Center															
roof of building 123	359	1.63	585	538	0.84	452	623	1.12	698	599	1.87	1118	469	1.02	478
measuring hut west	620	0.81	504	661	0.70	466	566	0.84	647	736	1.18	869	571	0.66	375
measuring hut north-east	612	1.07	656	632	1.20	760	718	1.19	857	733	1.49	1088	605	1.26	764
Karlsruhe Reprocessing Plant	640	0.84	537	681	0.62	425	771	0.95	728	734	1.23	898	580	0.64	372
outside Karlsruhe Nuclear Research Center															
Augustenberg (Grötzingen)	623	0.67	426	619	0.41	255	614	0.62	381	698	0.93	648	657	0.42	277
Bruchsal	429	1.02	438	687	0.35	242	530	0.52	306	675	0.86	581	597	0.37	217
Eggenstein	424	0.87	369	590	0.47	278	754	0.65	486	647	0.90	579	553	0.43	238
Tiefgestade waterworks (Leopoldshafen)	650	0.55	358	652	0.43	278	657	0.58	380	580	0.89	516	544	0.40	215
Philippsburg	-	-	-	688	0.46	314	584	0.64	374	-	-	-	-	-	-
Speyer	-	-	-	453	0.43	196	544	0.55	298	-	-	-	-	-	-
Niederstotzingen	-	-	-	456	0.45	205	550	0.55	303	467	0.85	395	364	0.24	85
Buchmühle	-	-	-	460	0.42	192	514	0.42	216	507	0.78	396	327	0.31	100

Table 4 Amount of precipitation M, average tritium concentration $\bar{a}$ and tritium surface loading A_F at sampling locations within and outside the Karlsruhe Nuclear Research Center from 1972 to 1976.

If the calculations are based on a mean surface load of 0.3 Ci/km^2 due to releases, a total load of the order of 1 Ci is obtained for an area of 1 km radius around the Nuclear Research Center. This means that the portion of 5×10^{-4} of tritium released to the atmosphere is dispersed by precipitation in this area. It is not possible to detect a surface load due to releases at a greater distance from the Nuclear Research Center because the variations as a function of time and location of surface load due to fall-out are greater than the little influence anticipated to be exerted by the Nuclear Research Center in this more distant area. The situation will be the same in case that the measuring accuracy can be substantially improved.

3.2 Tritium in Surface Waters

In Table 5 and Fig. 8, respectively, the annual mean values of the rivers Rhine and Neckar and of some affluents are compared for the years 1972 until 1976. The map of sampling locations is shown in Fig. 3.

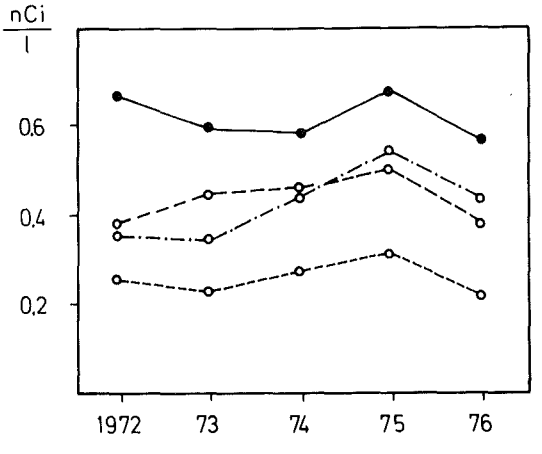

Fig. 8 Tritium concentration in surface water for the period from 1972 to 1976

—— Rhine
---- Neckar affluents to the Rhine
-·-·- between Kehl and Karlsruhe
----- between Karlsruhe and Mannheim

Waters investigated	Average tritium concentration of all sampling locations in nCi/l				
	1972	1973	1974	1975	1976
Rhine	0.66	0.59	0.58	0.68	0.56
Neckar	0.37	0.44	0.45	0.50	0.37
surface waters between Karlsruhe and Kehl (without Rhine)	0.35	0.34	0.44	0.54	0.43
surface waters between Mannheim and Karlsruhe (without Rhine and Neckar)	0.25	0.22	0.27	0.31	0.21

Table 5 Comparison of annual mean values of tritium concentration evaluated from the Rhine and Neckar rivers and above all from right affluents to the Rhine for the years 1972 to 1976.

Values for various sampling locations on the rivers Rhine and Neckar are listed in Table 6 for the years 1975 and 1976. At most of these locations samples have been taken continuously by the 'Institut für Wasser- und Abfallwirtschaft' of the 'Landesanstalt für Umweltschutz Baden-Württemberg.' Fig. 9 shows the profiles of the mean tritium concentration along the river Rhine between Öhringen and Mannheim for the years 1975 and 1976.

The differences of the annual mean values shown in Tables 5 and 6 are due to the varying mixing proportions of precipitations and ground water in flowing surface waters. Waterworks often process ground water with tritium concentrations lower than those of precipitations. The amounts of waste water so produced and discharged into the surface water are sufficiently high to influence the tritium concentration of the mixture.

RHINE			
km	Sampling locations	^{3}H concentration in nCi/l 1975	1976
29	Öhringen-Stiegen	0.63	0.48
91	Reckingen	0.68	0.50
113	Albbrück-Dogern	0.66	0.47
155	Wyhlen	0.62	0.46
224.7	Breisach	0.64	0.48
248.9	Weisweil	0.65	0.48
294	*Kehl	0.64	0.54
362.2	Maxau	0.67	0.48
372	*Leopoldshafen (ferry)	0.73	0.49
392.6	*downstream of Rheinschanz isle	0.99	1.23
400	*Speyer	0.62	0.49
426.2	Mannheim	0.71	0.64
432	*Mannheim	0.63	0.59
mean value		0.68	0.56

NECKAR			
km	Sampling locations	^{3}H concentration in nCi/l 1975	1976
0	*Mannheim	0.53	0.39
8.1	Feudenheim	0.51	0.35
18.1	Schwabenheim	0.50	0.37
21.0	*Heidelberg	0.58	0.37
31.0	Neckargemünd	0.47	0.44
72.5	Guttenbach	0.48	0.40
93.9	Gundelsheim	0.47	0.34
104.1	Kochendorf	0.49	0.35
118.0	Horkheim	0.49	0.33
125.5	Lauffen	0.47	0.35
143.1	Hessigheim	0.50	0.32
165.2	Poppenweiler	0.52	0.36
172.3	Aldingen	0.53	0.38
187.0	Stuttgart-Untertürkheim	0.49	0.40
199.7	Deizisau	0.54	0.39
mean value		0.50	0.37

*At these sampling locations two samples are taken per month. For all other sampling locations the mean values are derived from concentration values of samples collected continuously over a fortnight.

Table 6 Average tritium concentrations from the Rhine and Neckar rivers for 1975 and 1976

The tritium concentration in the surface water samples taken between Kehl and Karlsruhe is distinctly higher than that of the samples taken between Karlsruhe and Mannheim. This results from the higher contribution of precipitation water to flowing surface water and the lower density of population in the Black Forest area where the right affluents to the river Rhine south of Karlsruhe have their sources.

It should be mentioned that the tritium concentration in the Rhine river downstream of the Rheinschanz isle is remarkably higher than upstream of this isle (see Table 6). This is due to liquid tritium discharges from the Nuclear Research Center, which arrive near this sampling location (see Fig. 9). Only 8 km downstream of this location average concentrations were found.

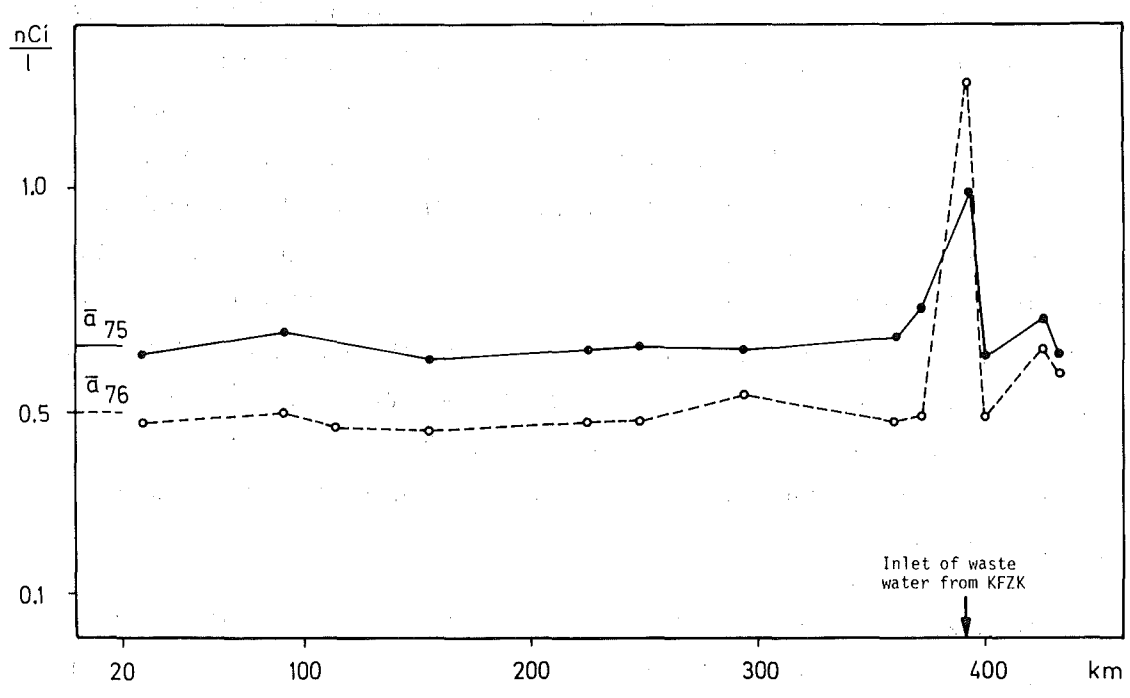

Fig. 9 Tritium concentration in the river Rhine as a function of location

—— 1975
---- 1976
ā annual mean value

3.3 Tritium in Ground and Drinking Waters

The sampling program makes a distinction between ground and drinking water samples. Pumped or scooped samples collected from wells at different depths are termed ground water samples whilst all the samples taken from the tap water system are termed drinking water samples. The tap water system is frequently supplied by two or more waterworks, even in small communities, so that the drinking water is often composed of a mixture of ground water from different depths and points of sampling and from spring waters, whose tritium concentration may differ quite markedly.

The tritium concentration of the ground water depends both on the tritium supplied by precipitation and from the tritium supplied by surface water filtered on the river banks in case that the ground water is raised by waterworks in the direct vicinity of a river. Besides, the tritium concentration of the ground water depends on the depths of sampling and on the vertical rate of migration of precipitations in the soil down to the ground water carrying stratum. The migration of the ground water is normally very slow and depends on the composition of the soil. For all these reasons and if detailed knowledge of local conditions is not available, it is generally impossible to establish quantitative relationships between the tritium concentration of ground water, precipitations and possible neighbouring surface waters. It is important above all in such investigations to observe the development with the time of tritium concentration over extended periods of time.

The ground water samples investigated under this measuring program were rarely taken from waterworks. The majority of ground water samples were taken from wells sunk in the vicinity of nuclear facilities. These observation wells were constructed in many cases for the specific purpose of background radioactivity measurements and routine investigations performed under the environmental monitoring of nuclear power stations.

In Tables 7 and 8 the annual mean values for the years 1973 until 1976 have been compiled of tritium concentration in ground and drinking water.

Sampling location	Annual mean values of the tritium concentration of ground water in nCi/l			
	1973	1974	1975	1976
Vicinity of the Karlsruhe Nuclear Research Center (KNRC) and reference locations Baden-Baden				
Southern waterworks in the KNRC	0.19	0.16	0.17	0.14
Leopoldshafen waterworks	0.44	0.32	0.51	0.38
Linkenheim waterworks	0.33	0.11	0.21	0.25
Karlsruhe-Hardtwald waterworks	0.37	0.21	0.30	0.12
Well A (Karlsruhe Reprocessing Plant)	0.32	0.32	0.45	0.37
Well B (Karlsruhe Reprocessing Plant)	0.48	0.27	0.47	0.37
Observation well Nr. 16	0.27	0.32	0.39	0.29
Observation well Nr. 20	0.26	0.24	0.36	0.30
Tiefgestade waterworks	0.50	0.49	0.57	0.44
'Fettquelle' Baden-Baden	0.12	-	-	-
'Reiherbrunnen' Baden-Baden	0.17	-	-	-
Vicinity of Nuclear Power Plant Niederaichbach*				
Dingolfingen	0.17	0.11	0.10	0.08
Aumühle	0.17	0.09	0.13	0.09
Kehl - Wörth	0.15	0.12	0.14	0.08
Wörth	0.24	0.17	0.24	0.10
Premises of power plant P I	0.28	0.27	0.26	0.14
Premises of power plant P II	0.28	0.25	0.18	0.16
Vicinity of Nuclear Power Plant Gundremmingen				
Well Guf 32 Gundelfingen	0.66	0.58	0.63	0.44
Well Lau 3 Lauingen	0.47	0.54	0.60	0.50
Well Lau 7 Lauingen	0.60	0.45	0.61	0.49
Gundremmingen waterworks	0.57	0.44	0.52	0.40
Dillingen waterworks	0.54	0.37	0.47	0.22
Conveyer Niederstotzingen 1	0.17	0.18	0.41	0.22
" " 4	0.15	0.14	0.42	0.34
" " 6	0.23	0.24	0.33	0.27
" Schalthof 2	0.14	0.14	0.17	0.08
" " 3	0.19	0.22	0.22	0.15
" " 5	0.51	0.51	0.52	0.46
" Buchmühle	0.35	0.32	0.40	0.22
Well, premises of power plant	0.53	0.54	0.52	0.35

Table 7 Annual mean values of tritium concentration in ground water from 1973 to 1976
*decommissioned

Sampling locations	Annual mean values of tritium concentration in nCi/l				
	1972	1973	1974	1975	1976
Marxzell	-	-	0.25	0.31	0.27
Neuenbürg	-	-	0.19	0.22	0.17
Heidelberg	0.27	0.30	0.14	0.23	0.17
Mannheim	0.19	0.17	0.15	0.19	0.10
Ludwigshafen	0.25	0.15	0.10	0.10	0.08
Speyer	0.20	0.14	0.12	0.16	0.11
Kehl	0.64	0.55	0.51	0.66	0.50
Baden-Baden	0.28	0.30	-	-	-
Bruchsal	0.40	0.20	0.16	0.16	0.10
Eggenstein	0.63	0.24	0.21	0.27	0.18
Förch-Niederbühl	0.46	0.48	0.35	0.42	0.33
Friedrichstal	0.37	0.37	0.31	0.35	0.24
Jöhlingen	0.41	0.13	-	0.14	0.13
Hagenbach	0.29	0.18	0.17	0.19	0.09
Karlsruhe	0.38	0.21	0.18	0.27	0.21
Durlach	0.56	0.33	0.30	0.34	0.23
Kuhardt	0.18	0.14	0.10	0.08	0.08
Leimersheim	0.31	0.14	0.09	0.08	0.08
Leopoldshafen	0.49	0.43	0.32	0.37	0.33
Mörsch	0.42	0.18	0.14	0.25	0.19
Neuthard	0.46	0.25	0.15	0.22	0.28
Obergrombach	0.38	0.32	0.28	0.37	0.29
Ubstadt	0.29	0.25	0.19	0.25	0.17
Weingarten	0.29	0.15	0.11	0.19	0.16
Wolfartsweier	0.52	0.58	0.41	0.48	-

Table 8 Annual mean values of tritium cocentration in drinking water from 1972 to 1976

Maps of the sampling locations are shown in Fig. 4 for ground water and in Fig. 5 for drinking water. Fig. 10 shows a graph with mean values of the tritium concentration of drinking water measured at 23 locations during the last five years.

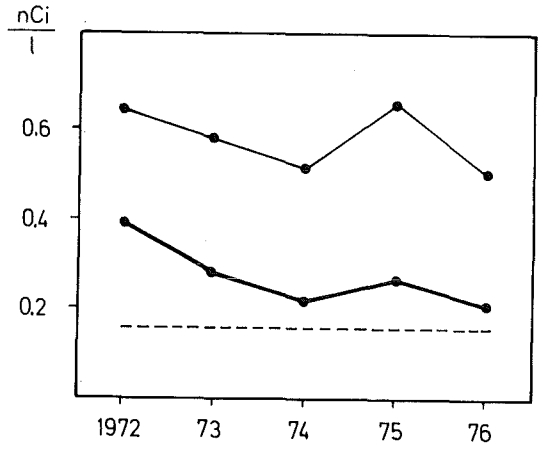

Fig. 10 Tritium concentration in drinking water, averaged over 23 sampling locations

—— maximum annual mean values
—— average annual mean values
---- detection limit

3.4 Summary of Measuring Results of Tritium in Waters

A comparison of the mean values of tritium concentrations for the different types of waters might be of interest in a summarizing review of measured results. Such a summarizing representation of measured results for precipitations, surface, ground and drinking waters is shown in Fig. 11 for the years 1973 through 1976. In this graphical plot the variations of annual mean values found for the individual sampling locations have been shown for different sampling areas. The mean values of concentrations for the years 1973 to 1976

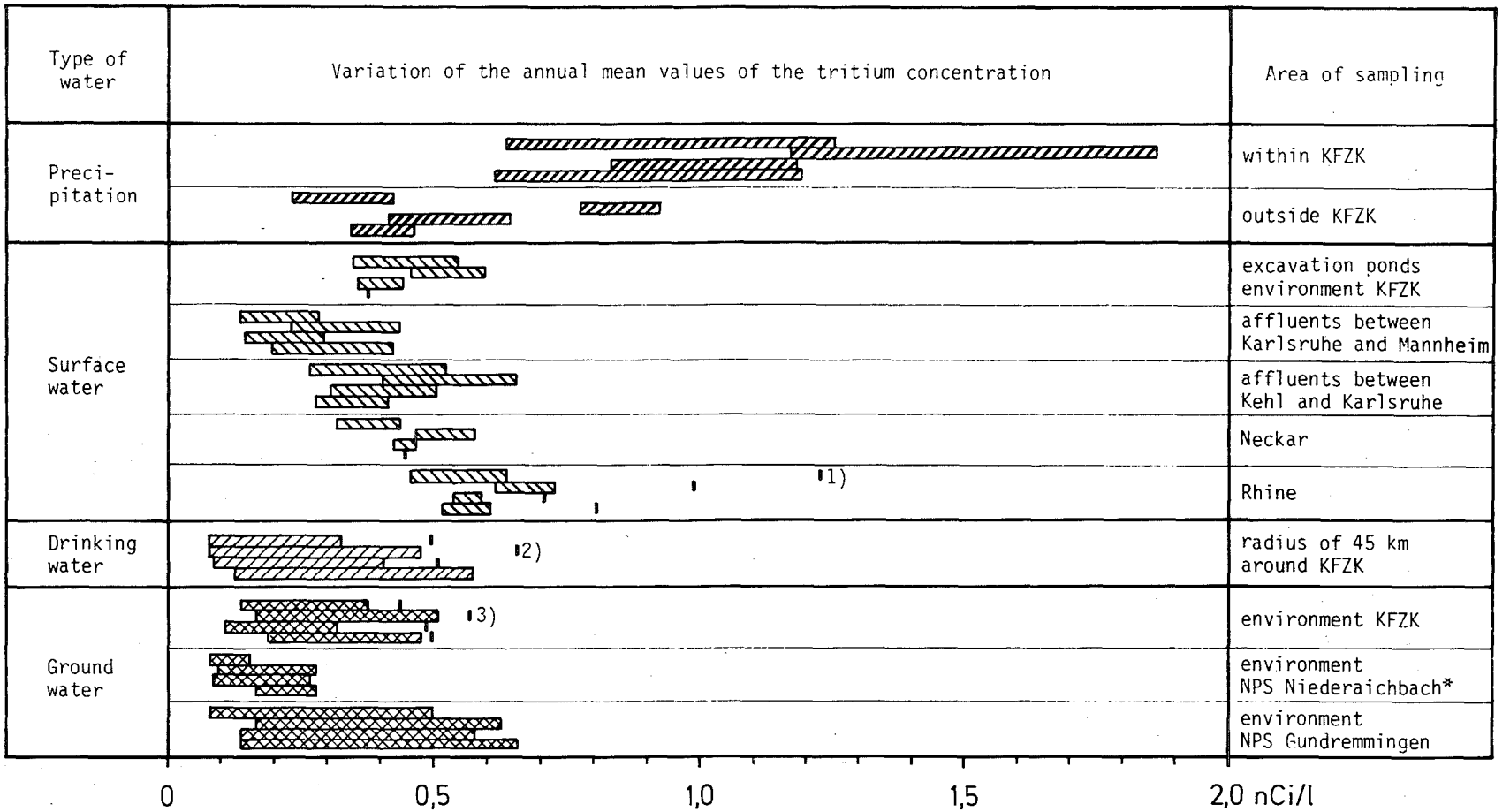

Fig. 11 Summary of measuring results of tritium in water for the years 1973 through 1976. For each year the variation of the annual mean values is given in the arrangement from top (1976) to bottom (1973). Outstanding values are represented separately.
KFZK: Karlsruhe Nuclear Research Center; NPS: Nuclear Power Station.
1) Inlet waste water from KFZK; 2) Kehl; 3) 'Tiefgestade' waterworks.
*decommissioned

with respect to the different types of water range from 0.08 nCi/l* for ground water to 1.9 nCi/l for precipitations inside the Karlsruhe Nuclear Research Center where the maximum annual mean values of tritium concentrations were found in 1975. Of the surface waters investigated the river Rhine shows by far the highest annual mean values of tritium concentration (0.46 nCi/l - 0.73 nCi/l). For the river Neckar and all other affluents to the Rhine between Kehl and Mannheim markedly lower annual mean values were obtained.

Depending on the sampling region, annual mean values between 0.08 nCi/l* and 0.66 nCi/l were measured for ground water in the period from 1973 to 1976. In the same period the annual mean values for drinking water ranged from 0.08 nCi/l* to 0.58 nCi/l, except for the city of Kehl where higher values were found, obviously due to the high portion of Rhine water.

4. Tritium in Plants

As already mentioned in the introduction only three limited programs could be carried out in 1975 and 1976 to study the tritium contamination of the free tissue water of plants. To find out relations to the tritium offer of the relevant media close to the plants, the tritium concentrations in air humidity, ground water and precipitations were investigated simultaneously.

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*When calculating the mean values half the value of the detection limit (ca. 0.15 nCi/l) was used for samples with concentrations below the detection limit.

4.1 First Period of Investigation [9]

Under an initial measurement program (from August 1 to September 23, 1975) 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 (see site plan, Fig. 12). The results have been summarized in Table 9. For the sake of clarity, only maximum and minimum values are indicated. 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.

4.2 Second Period of Investigation [9]

Under another program (March 3 to May 26, 1976) the tritium concentration of pine and spruce needles, hornbeam leaves, the humidity of the air, precipitations and ground water were measured simultaneously at three selected sampling locations (map of sampling locations, see Fig. 12).

One sampling location lies in a region in which the ground water is contaminated. It is termed here sampling location 'West.' At the end of August 1975 a leakage was found in the piping system of the sewage treatment plant about 3 m deep in the ground by which low level effluent water from laboratories, after having passed the decontamination facility, could escape into the soil. In the region of the so-called location 'West' also the humidity of the air is contaminated by the evaporation of chemical effluent water.

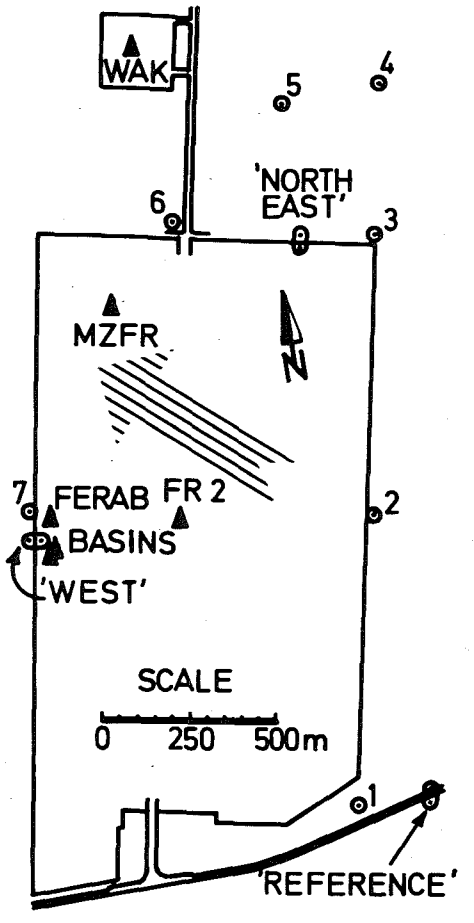

Fig. 12 Site plan indicating the main tritium emitters from the Karlsruhe Nuclear Research Center ▲ and the sampling locations covered by the first ⊙ and the second period ⊙ of investigation
Direction of ground water flow.

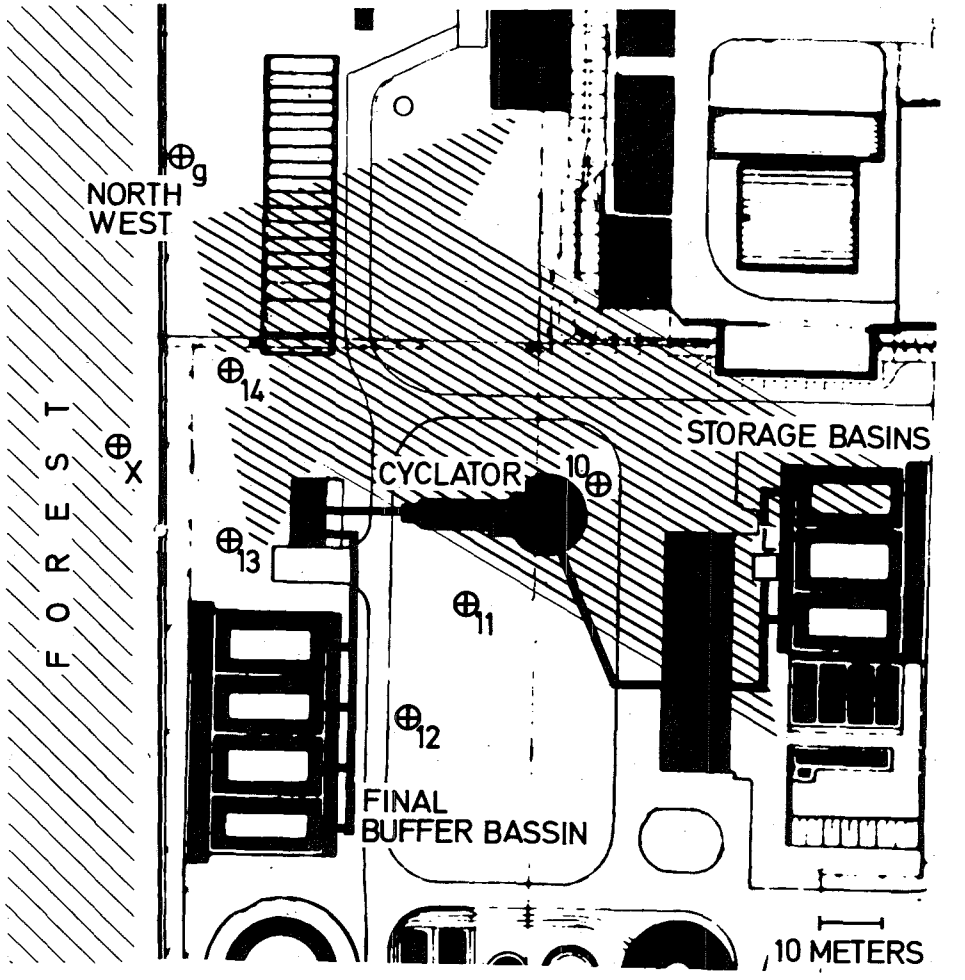

Fig. 13 Site plan of the sewage treatment plant of the Karlsruhe Nuclear Research Center

□L Point of leakage, ⊕ Locations of ground water drillings and index numbers.
Direction of ground water flow.

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 9 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.

Fig. 13 is a site map of sampling locations where samples have been taken regularly since the tritium ground water contamination was detected. Fig. 14 shows the development with time of measured results [10]. It is represented here to show above all that an existing tritium contamination of the ground water undergoes but relatively slow variations.

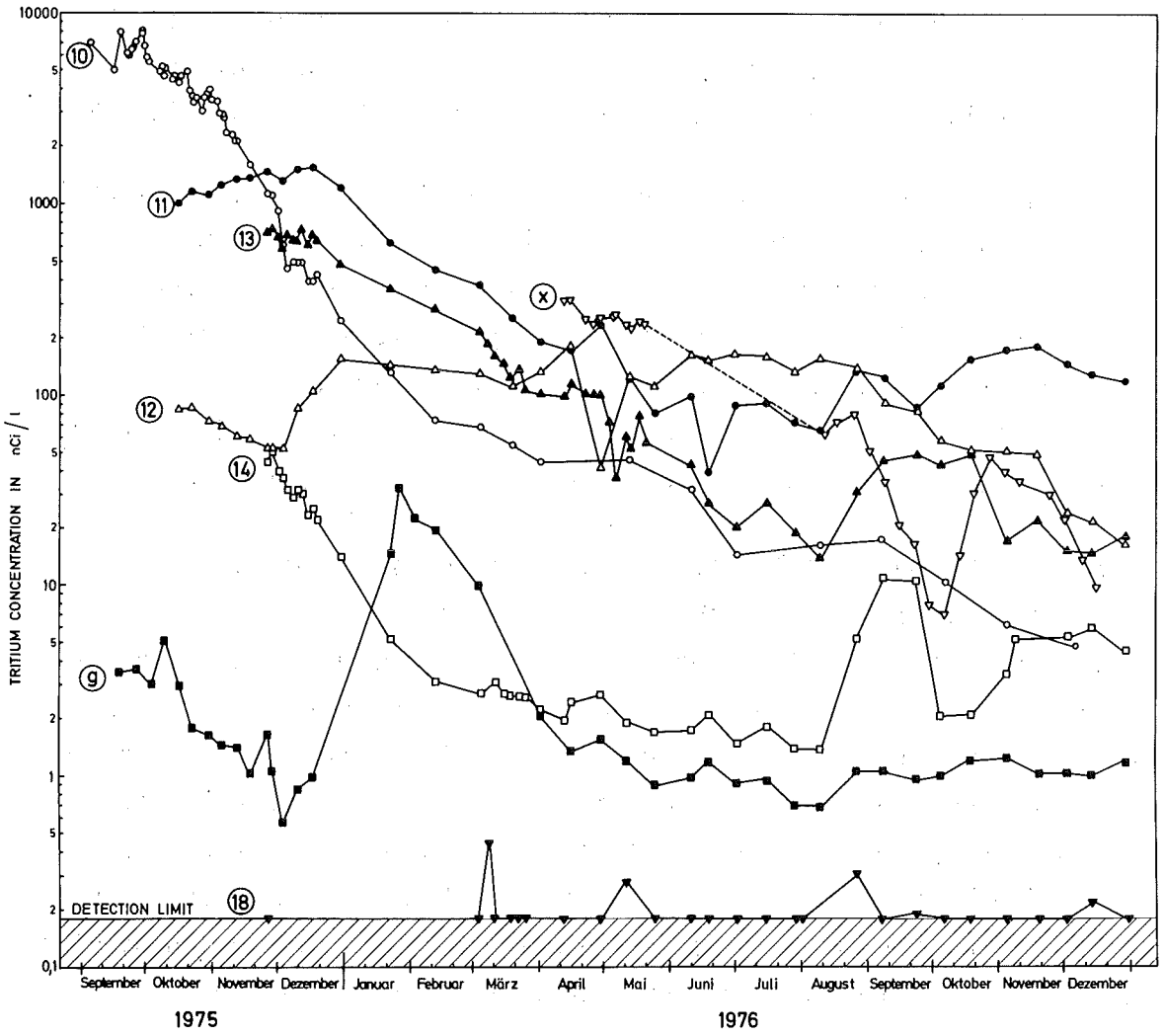

Fig. 14 Development of tritium concentration of the ground water in different observation wells in the vicinity of the sewage treatment plant

The second location, termed 'Northeast,' lies in the northeastern 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 background 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 are shown in detail in Figs. 15 to 17 and have been compiled in Table 10. Again only the maximum and minimum values of tritium concentrations are indicated in this table.

The highest tritium concentrations were found at the sampling location 'West.' The results (see Fig. 15) unambiguously show that the precipitation is not the main cause of tritium contamination of the pine and spruce needles. Since around the sampling location 'West' the ground water had been contaminated within the period of investigation, both this contamination and that of the humidity of the air must be considered as causes of pine and spruce needle contamination.

Since at the sampling location 'West' the roots of the pines extend to much lower depths than that of the 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.

Fig. 16 shows the measured values for the sampling location 'Northeast' 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.

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±0.17	background expected	2.0±0.2	2.4±0.2	2.3±0.2*

Table 10 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.

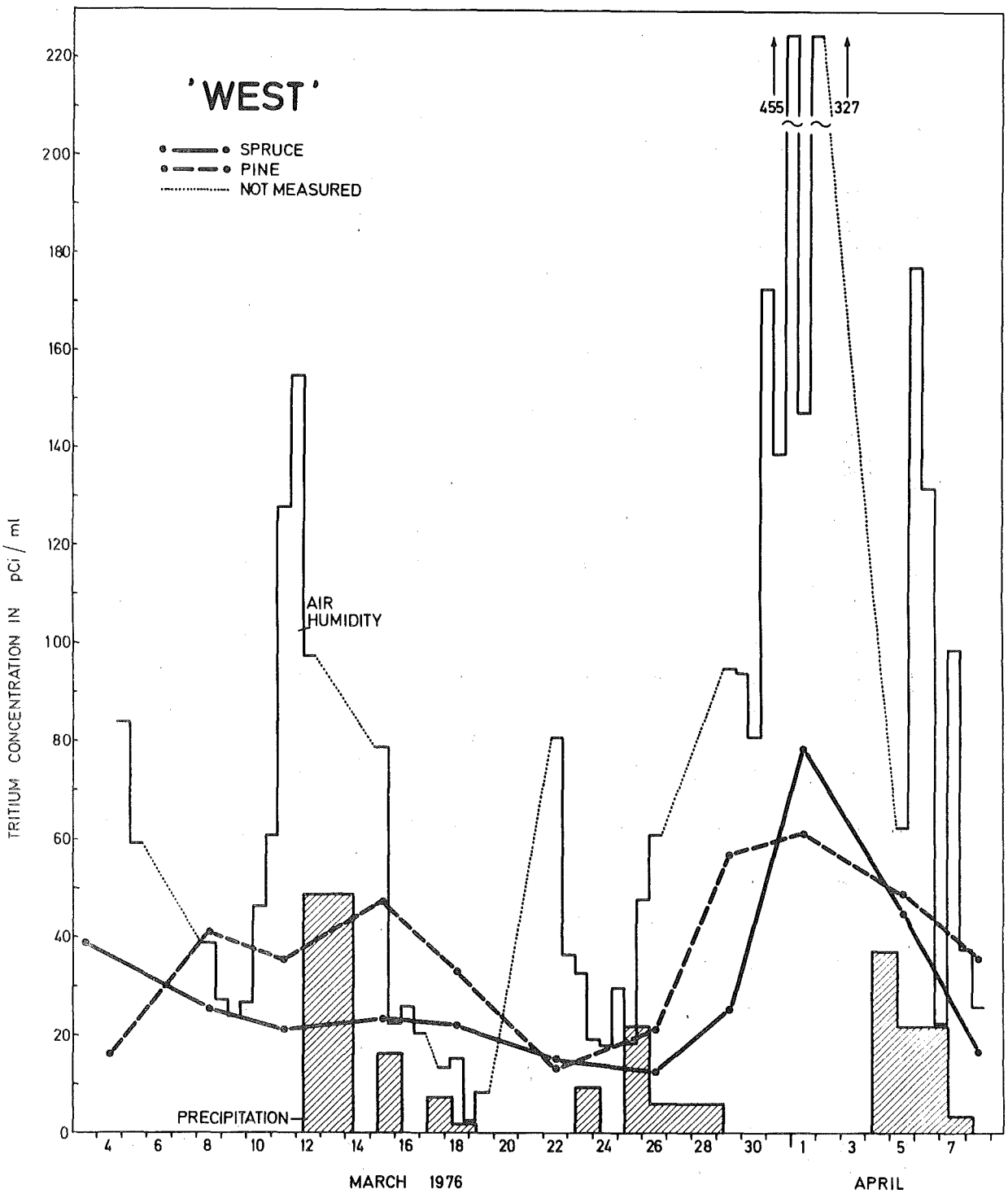

Fig. 15 Representation of the variation with time of the tritium concentration in spruces, pines, air humidity and precipitations, measured at the sampling location 'West' (see Fig. 12).

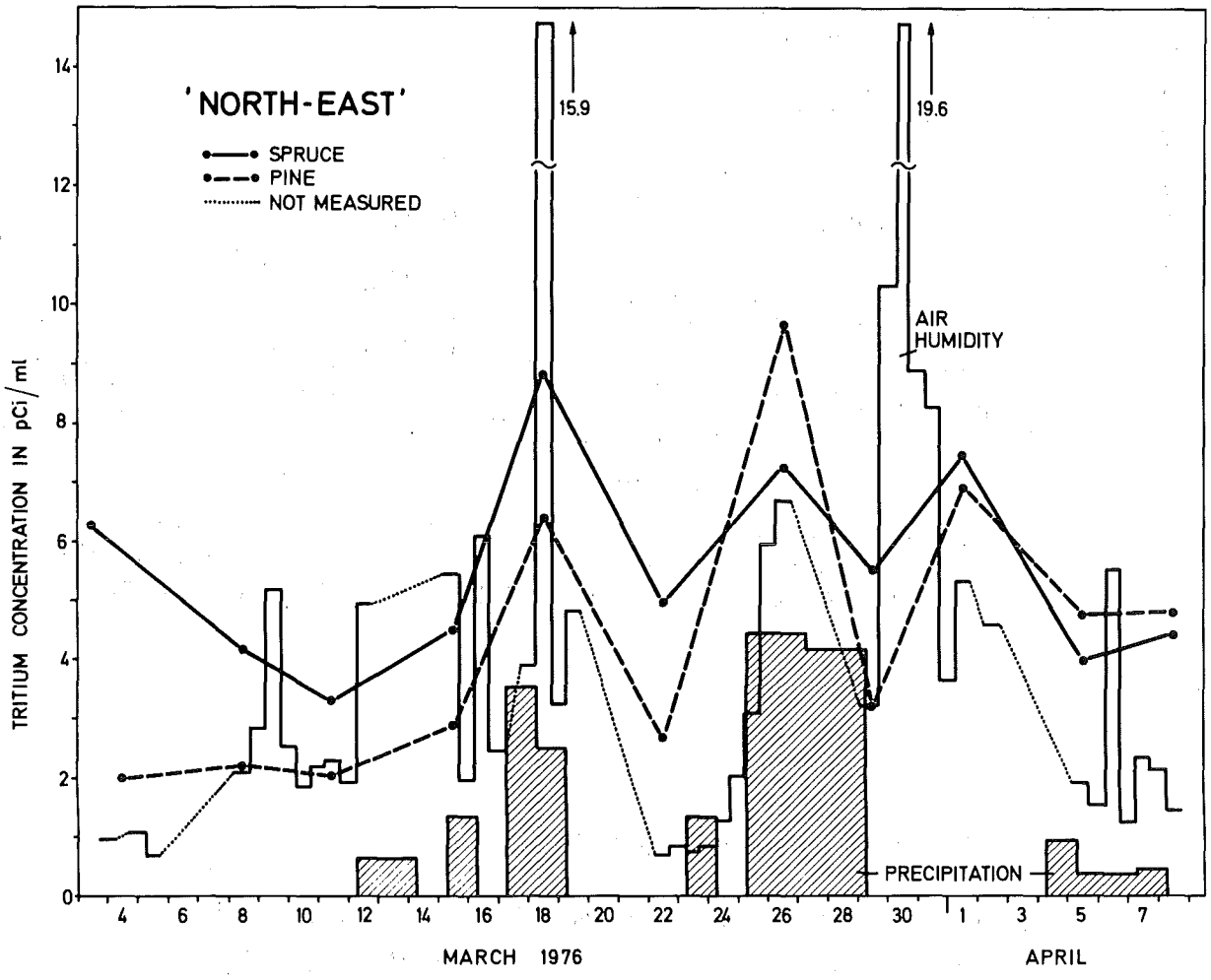

Fig. 16 Representation of the variation with time of the tritium concentration in spruces, pines, air humidity and precipitations, measured at the sampling location 'Northeast' (see Fig. 12).

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. 17.

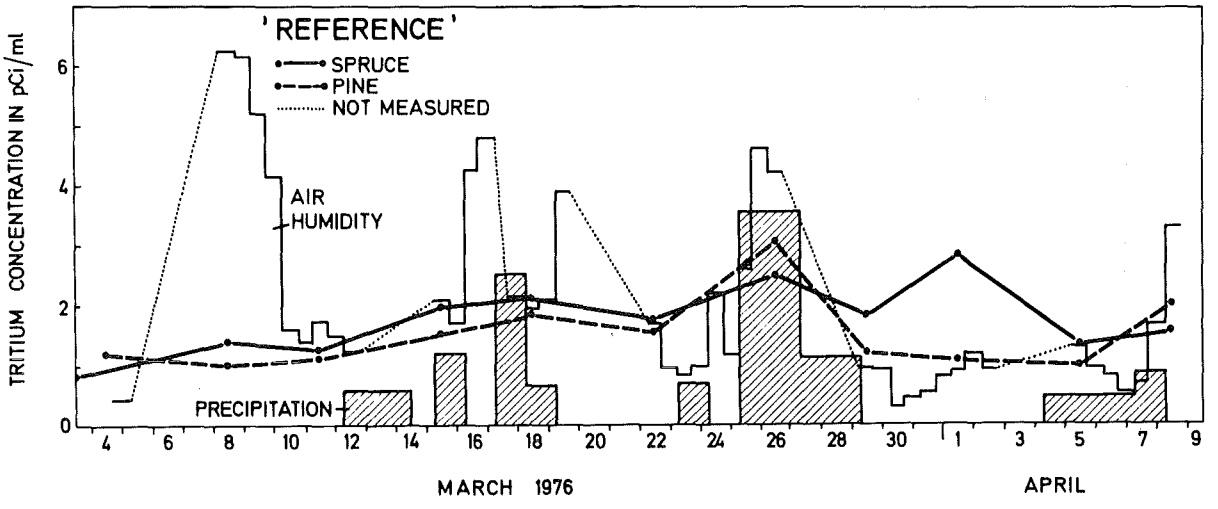

Fig. 17 Representation of the variation with time of the tritium concentration in spruces, pines, air humidity and precipitations, measured at the 'Reference' sampling location (see Fig. 12).

4.3 Third Period of Investigation [12]

To collect a more significant amount of information, it was decided to limit the investigation to location 'West' (see Fig. 12) in the third measurement program (from August 9 to November 10, 1976).

Samples were taken at this place from pine needles (once a week from a height of about 12 m), spruce needles, oak and hornbeam leaves (three times a week), air humidity (twice a day), ground water (once a week) and precipitations (each working day after rainfall). During the period in which the program was carried out samples were taken only from the trees selected. Within the period of this program about 200 plant samples were investigated in total.

The maximum and minimum values measured during the period of investigation have been compiled in Table 11. The results of this investigation are shown in Fig. 18 and discussed in detail in ref. [12]. In

Fig. 19 the variation with time of the tritium concentration in air (pCi/m^3), air humidity (pCi/ml) and ground water (pCi/ml) is represented.

Tritium concentration	Air humidity pCi/ml	Ground water pCi/ml	Precipitation pCi/ml	Precipitation $nCi/m^2/d$
Maximum	277±8	80.3±3.2	61.0±2.5	230±10
Minimum	0.29±0.18	7.02±0.35	0.16±0.16	0.18±0.15

Tritium concentration	Needles Spruce pCi/ml	Needles Pine pCi/ml	Leaves Hornbeam pCi/ml	Leaves Oak pCi/ml
Maximum	71.2±2.9	63.6±2.6	234±6	235±9
Minimum	1.44±0.21	5.42±0.34	0.95±0.19	1.11±0.21

Table 11 Results of measuring tritium concentration in air humidity, ground water, precipitation, plants at the sampling location 'West.' Table 11 consists of maximum and minimum values measured during the investigation period from August 9 to November 10, 1976.

4.4 Comparison of Vacuum Distillation and Azeotropic Distillation. Organically Bound Tritium

By comparison, 40 samples were subjected simultaneously to vacuum distillation and azeotropic distillation by xylene to extract the tissue water from needles and leaves of different trees. Azeotropic distillation allows to obtain the distilled product within a short period whereas the vacuum distillation process takes more time.

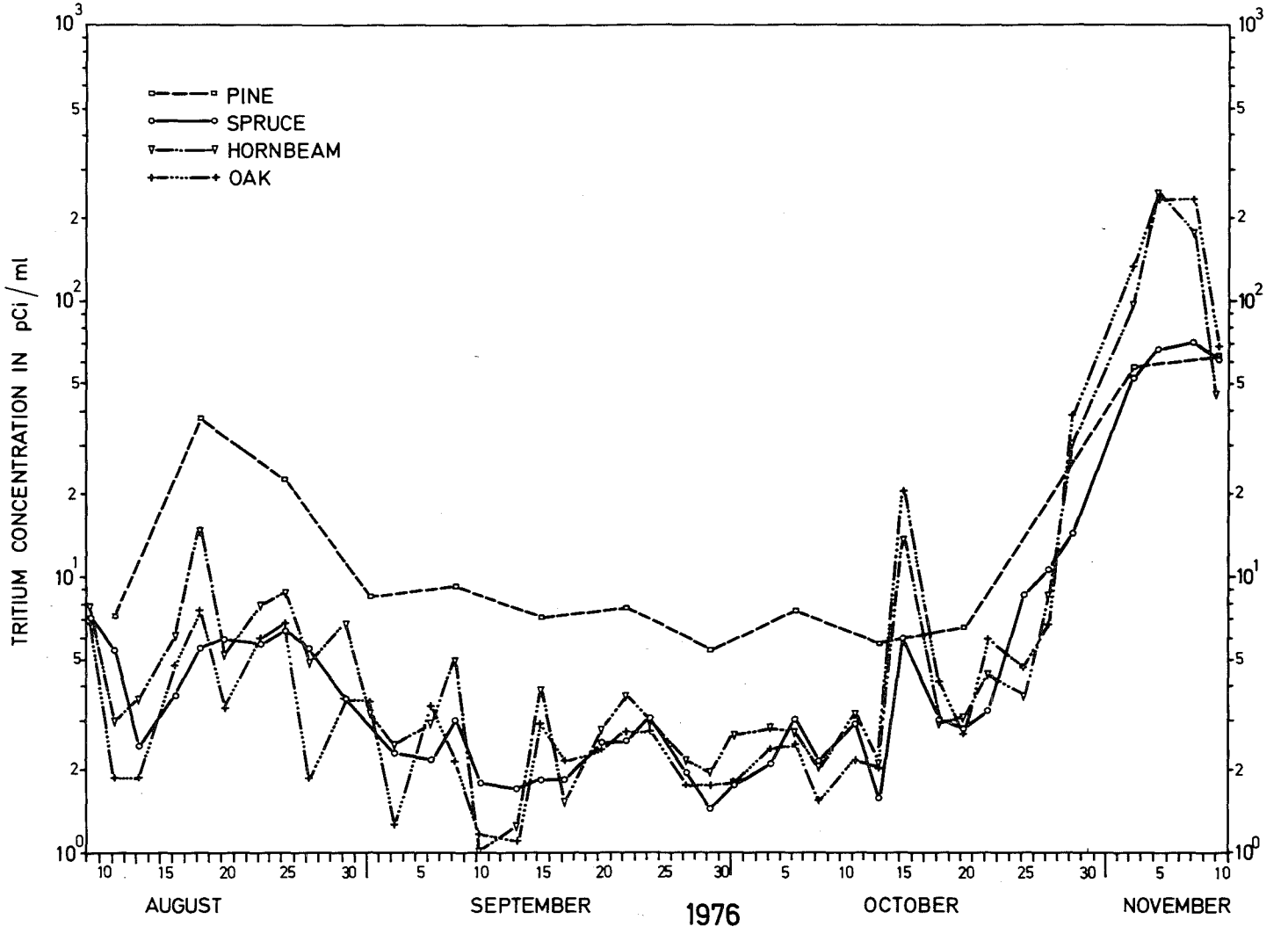

Fig. 18 Representation of the variation with time of the tritium concentration in pine and spruce needles, hornbeam and oak leaves.

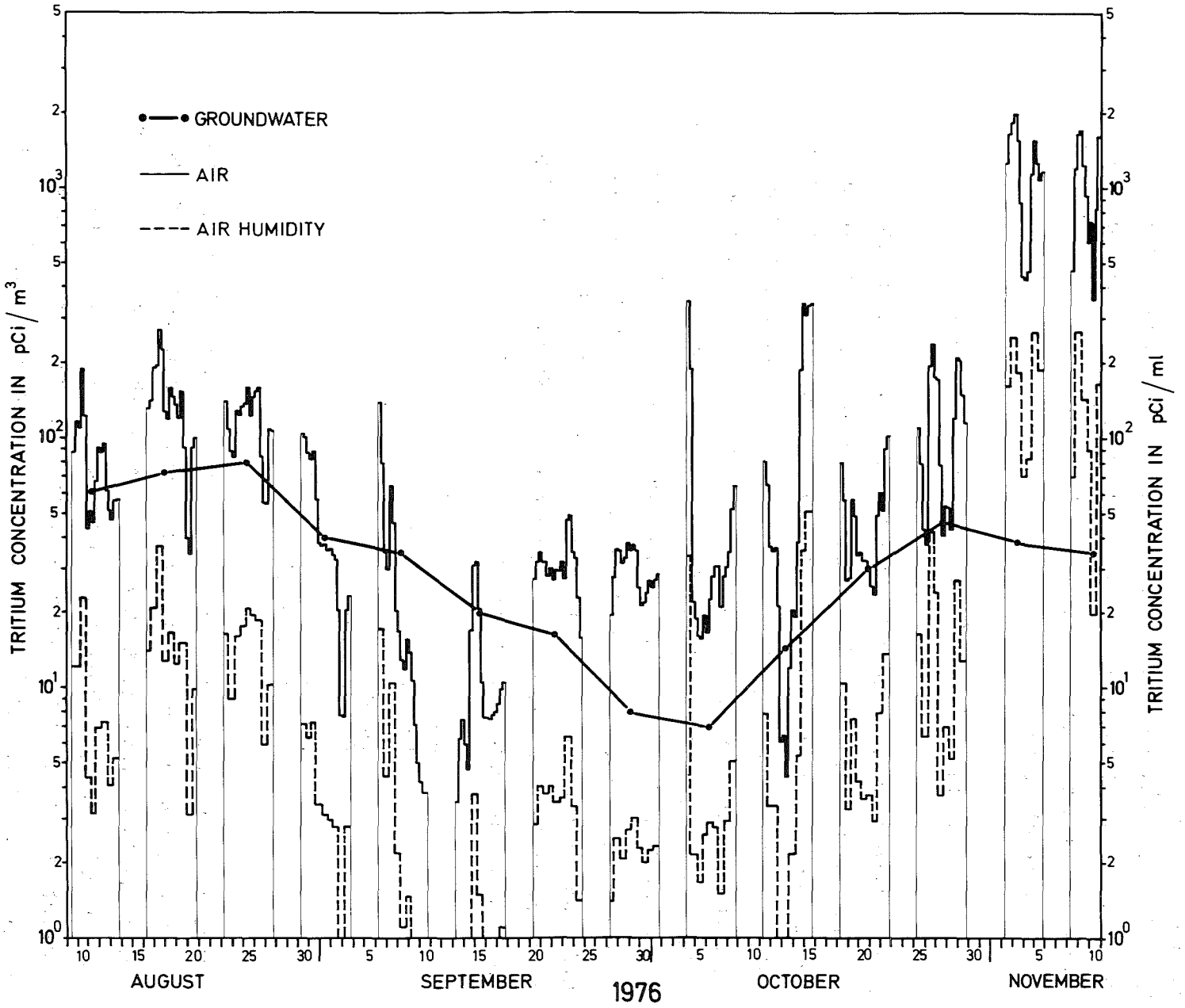

Fig. 19 Representation of the variation with time of the tritium concentration in air (pCi/m³), air humidity (pCi/ml) and ground water (pCi/ml).

Vacuum distillation gives two products, a distillate and dry biological matter enabling to determine organically bound tritium, which is not possible with the azeotropic method. With the 40 couples of samples an average value of 0.95 was found for the ratio of tritium concentrations

$$a_{\text{vacuum distillation}} / a_{\text{azeotropic distillation}} \cdot$$

It is interesting to compare tritium bound in the tissue water with tritium bound organically. Dried masses of selected plant samples were measured by R. Kirchmann, Department of Radiobiology at CEN in Mol, Belgium, after dehydration, using vacuum distillation. Table 12 is a comparison of values measured by Kirchmann with our own measured values for tissue water. It was found that the amounts of tritium in water obtained from the dried mass by incineration are clearly higher than that in tissue water. With the ratio of tritium concentration in HTO from organic material to tritium concentration of tissue water distinct differences were found between the different species. The mean values of these ratios and the errors were for

pine (pinus sylvestris)	2.04 ± 0.13
spruce (picea abies)	4.8 ± 1.1
hornbeam (carpinus betulus)	9.7 ± 2.3

Since only few values are available until now, one should not be rash in drawing conclusions. It should be considered in particular that fresh plants contain about 80 % of water and that with a two-compartment model already differences can be easily explained between the concentrations of tritium bound in water and bound organically. Considering the longer biological half-life of the organically bound tritium, the finding could be explained by a preceding tritium contamination. However, this explanation is put in doubt by the time interval and the development of measured values. In any case, further investigations are necessary to clarify the situation.

Date of sampling	Tritium activity tissue water pCi/ml	organically bound pCi/ml	organically bound pCi/g	Yield ml/g
p i n e (pinus sylvestris)				
11.8.1976	7,92 ± 0,45	20,2	12,2	0,606
18.8.1976	13,76 ± 0,60	27,6	16,9	0,612
25.8.1976	12,00 ± 0,51	22,8	13,8	0,604
1.9.1976	7,73 ± 0,40	17,6	10,7	0,605
8.9.1976	9,52 ± 0,46	14,4	8,3	0,576
15.9.1976	6,95 ± 0,31	14,0	8,5	0,606
h o r n b e a m (carpinus betulus)				
13.8.1976	2,99 ± 0,26	14,4	7,9	0,550
20.8.1976	3,90 ± 0,29	26,4	14,4	0,548
27.8.1976	5,17 ± 0,34	19,5	10,8	0,555
3.9.1976	1,39 ± 0,20	15,5	8,4	0,542
10.9.1976	0,88 ± 0,19	16,4	9,1	0,556
17.9.1976	1,52 ± 0,22	19,4	10,8	0,556
s p r u c e (picea abies)				
16.8.1976	3,74 ± 0,28	14,1	8,2	0,582
23.8.1976	5,34 ± 0,34	17,7	9,8	0,554
30.8.1976	3,89 ± 0,30	14,7	8,3	0,562
6.9.1976	1,74 ± 0,21	16,1	9,1	0,566
13.9.1976	1,55 ± 0,21	5,9	3,2	0,537

Table 12 Comparison of tritium content of leaves and needles, respectively, in tissue water and in organic compound for selected samples.

4.5 Discussion of the Measuring Results

The following general conclusions can be drawn:

The decisive element with respect to the type of tritium contamination in leaves and needles is the influence of the water vapor exchange as well as the direct influence of precipitation and dew on the surfaces of leaves and needles.

Variations of the tritium concentration in air humidity affect the variations of the tritium concentration in the tissue water of plants. These changes correlate with each other. The tritium concentration level in the tissue water of plants is close to the tritium concentration in air humidity. The variation with the time of the tritium concentration in leaves and needles depends on the water circulation between the root system and branches and on the process of water evaporation through the leaves and needles into the air.

In the period of leaf fall a change of 80 to 90 % of the color of leaf surfaces was reached on November 8, 1976. This period does not seem to be affected by the loss of exchange of water between the atmosphere and the volume of the leaves because, throughout this period up to leaf fall on November 12, 1976, a process of change in the tritium concentration in leaves took place (see Fig. 18).

Precipitation has a direct influence on the changes in tritium concentration in leaves and needles. The changes in these levels depend on both the tritium concentration in leaves and needles and in precipitations.

The measured results do not indicate any direct relationship between the tritium concentrations in ground water and in plants. The influence of tritium in the soil gets effective through the capillary water at different tritium contamination levels arising from the slow penetration of precipitations into the soil and through capillary water con-

taminated by tritium in the ground water. The water taken up through the root system at different depths with varying levels of tritium contaminations has an average tritium concentration which results from the different levels of contamination in the soil.

The minimum levels of tritium concentration in the leaves are about 10 to 30 times lower than the tritium concentration of the ground water. At the same time, the tritium level in leaves is close to the background level of tritium concentration in air humidity. On the other hand, in accordance with the contamination level of air humidity there are maximum levels of tritium concentration in the tissue water of plants much higher than the tritium level in ground water. The fact that the tritium levels in leaves are close to the tritium levels of air humidity demonstrates that water vapor is quickly exchanged between the atmosphere and the leaves.

The variation of the tritium concentration in tissue water of plants allows to determine the time constants and the half-lives of the process. The following values are found: for oak and hornbeam leaves 2±1 days, for spruce needles 3±1.5 days, for pine needles 6±3 days. In reference [9] a value of 4±2 days was reported both for pine and spruce needles. The time sequence of measurement points and the experimental conditions were not sufficient to make more accurate statements.

5. Movement of Tritium in the Environment

Tritium released into the environment gets into the water cycle and thus is dispersed worldwide. Fig. 20 presents a simplified model of the transport of tritium in the environment.

The influence on precipitations of tritiated water vapor released from the Karlsruhe Nuclear Research Center was treated already in chapter 3.1. When investigating the tritium content of surface waters (see chapter 3.2) some experience was gathered on the dispersion of

tritium in surface waters. The impact of environmental tritium on different plant species was reported in chapter 4. The results obtained provide some indications of contaminations of precipitation and plants to be expected upon the release of tritiated water vapor.

Within the framework of studies performed beside the tritium measuring programs dealt with here the dispersion was investigated of tritium released to the atmosphere as well as to the ground and surface water from the Karlsruhe Nuclear Research Center. Some of the results found in these investigations will be shortly described now.

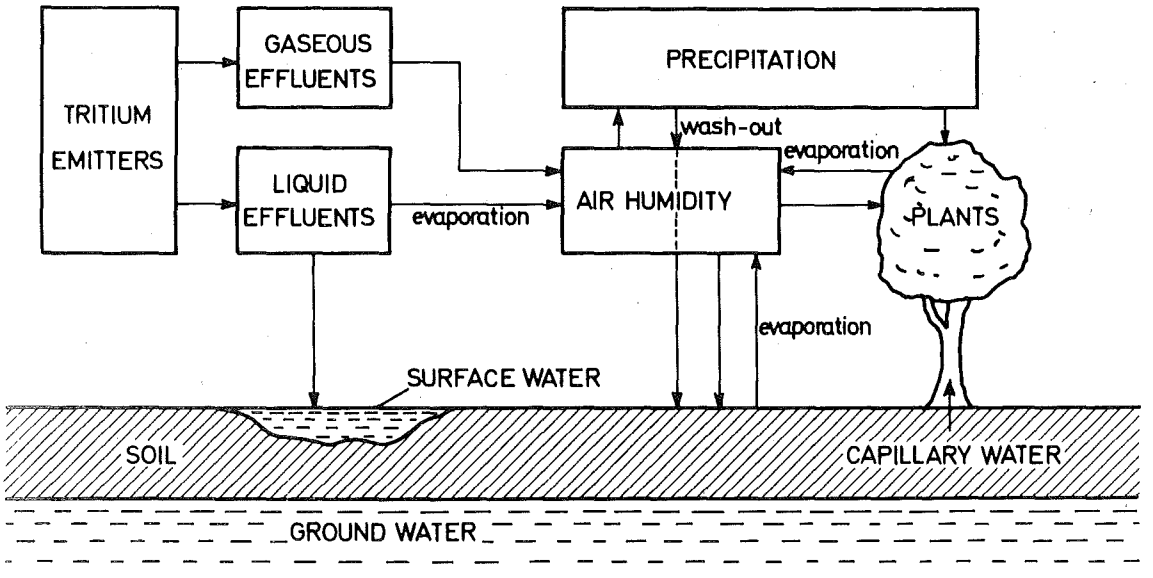

Fig. 20 Simplified model of the transport of tritium in the environment

5.1 Air Path

The diffusion of tritium in the atmosphere can be described with sufficient accuracy by familiar dispersion formulae (compare also [16, 17]). The equation for ground level air concentration values C (curies per cubic meter) is

$$C = Q(\pi\sigma_y\sigma_z u)^{-1}\exp[-(y^2/2\sigma_y^2+h^2/2\sigma_z^2)] \qquad \text{Eq. (1)}$$

where

Q = source strength in curies per second,
u = average wind speed in meter per second,
h = height of the source above ground in meter,
y = lateral (cross wind) distance from the plume axis in meter .

The horizontal and vertical dispersion parameters σ_y and σ_z, respectively, are functions of the downwind distance.

Eq. (1) is valid for release periods of 1/2 to 1 h and can be verified in the experiment. At the Karlsruhe Nuclear Research Center diffusion experiments have been performed to determine the parameters σ_y and σ_z. Initially, tritiated water vapor was used as a tracer. Now, non-radioactive organic compounds are used as tracers. A result of previous experiments was that the tracer substances used do not differ in their atmospheric diffusion behavior, which confirms the applicability only postulated originally of Eq. (1) to the dispersion of tritiated water vapor in the atmosphere [18-22]. A significant result is the shifting towards the source of the position of the radial concentration maximum as well as the increase in amount of this maximum relative to the values anticipated on the basis of the diagnosed Pasquill diffusion categories. Another result was the finding that σ_y and σ_z depend on the surface roughness caused by buildings and vegetation.

However, the impact on the environment due to tritium releases is not so much determined by temporary dispersion conditions, but rather by the long term diffusion factor. The latter can be calculated for the respective weather conditions from temporary dispersion factors, taking into account the meteorological data applicable to the area concerned (compare e.g. [23]). The experimental verification of the long term diffusion factor, based on available tritium measurements, is not possible because the prevailing conditions are not well defined.

5.2 Water Path

Tritium released via the waste water may result in a radiation impact on persons in several ways. The ways to be considered for drinking water and breathing air are:

seeping into the ground water from leakages in sewage treatment plants and in the sewer system as well as seeping from surface waters;

evaporation on uncovered water surfaces while contaminating the environment via the air path .

With respect to the influences of temperature, wind speed and the difference between the saturation pressure and pressure prevailing of the water vapor in the air the determination of the momentary evaporation rate is the most difficult problem. The diffusion via the air path has been treated in chapter 5.1.

By contrast, it is hardly possible to estimate with an acceptable accuracy the radiation impact due to seeping. Experimental investigations are necessary in each individual case. Generally, leakages occurring in sewer systems are not adequately taken into account. However, if such systems carry tritium bearing liquid effluents, contaminations get measurable. Tritium which has reached the ground water is diluted at a very slow rate only. Periods of months and years must be anticipated. Also the ground water flow velocity is low (order of magnitude 1 m/d) so that the diffusion of a possible contamination is very slow or the contamination is detected after a relatively long period only [10].

Surface water contaminated with tritium may entail radiation exposure also via the food chain on different exposure paths. The following paths have to be considered above all:

cattle - milk or meat
fodder crops - milk or meat
irrigation - vegetables .

A useful estimate of radiation exposure for all exposure paths combined is obtained if one assumes that all the water taken up has the mean tritium concentration C of the sewer system. The following formula applies for the radiation exposure D

$$D = 168 \frac{\text{rem}}{\text{a}} \frac{\text{ml}}{\mu\text{Ci}} \cdot C \; . \qquad \text{Eq. (2)}$$

In Eq. (2) the quality factor (QF) was set 1.7.

If one estimates the radiation exposure of the population on the assumption that the mean tritium concentration in the human body equals the mean tritium concentration in drinking water, a radiation exposure of 85 µrem is obtained for Kehl in 1976. The radiation exposure due to tritium is markedly lower for the majority of population living in the area monitored.

6. Concluding Remarks

Results available so far give rise to more questions elucidation of which will be attempted in the future. The questions still to be answered concern the level of tritium contamination in the root system and the branches and the relationship between the tritium contamination in different parts of the trees and in the air, soil and ground water. Above all, the attempt will be made to find out which percentage of tritium contained in the humidity of the air gets into leaves and needles directly and via the roots. However, it is not expected that the statement must be modified according to which the tritium concentration in the tissue water of leaves and needles, respectively, largely corresponds to that in the humidity of the air.

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