Stockholm natural tritium measurements. I

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ABSTRACT

After a brief description of our methods and standards for the determination of tritium in natural water, results are reported on the controversial rain water samples from Valentia, Ireland. The main part of this paper is a selection of tritium and deuterium data on samples, which may have general geophysical interest. Among them are long series of monthly precipitation from global and Scandinavian networks, and samples from rivers feeding into the Baltic.

Introduction

This institute was established in 1954 for C 14 dating service. In 1958 the erection of an apparatus for the determination of tritium in natural waters was begun. The improved counting techniques allowed an accuracy of ± 15 TU without enrichment, and we developed a periodic addition-type electrolysis enriching the sample water 100-fold in one cell. After an expansion completed in February 1963, enrichment facilities are in operation to keep the two low-level tritium counters processing about 50 samples a month. One of the authors and two women technicians handle all the work from receipt of samples to presentation of final results.

Some discrepancies have occurred between tritium data from different laboratories on the same or very similar waters. In order to standardize natural tritium measurements, samples with known, low tritium assay were prepared at our laboratory. A joint report on this set of tests is published in this journal (ÖSTLUND, BROWN & BAINBRIDGE, 1964). The result shows that the three laboratories produce data which are all on the same scale, namely the NBS-standard tritium scale.

Below will first be described some of the technical features of our method. As a proof for its reliability a series of double checks are also reported.

One of our series of monthly rain water data is from Valentia, Ireland. Some of these rains have also been measured in Los Angeles, with results widely differing from ours. This discrepancy is discussed below.

The last part of this paper consists of tables of tritium and deuterium assays in rain and other waters.

Counting techniques

Only a brief description of the counting system will be given here, since there are only a few changes compared with an earlier technical report (ÖSTLUND, 1962).

About 1 ml of water is admitted drop by drop into the warm zone (50–100°C) of a Pyrex tube in the evacuated reduction-filling system. Each drop evaporates almost instantaneously, and the water vapor passes through magnesium turnings at 600°C. The reduction yields hydrogen which is passed through a freezing trap at -80°C, through a bed of Molecular Sieves no. 4 and 13[2], a fine fritted glass filter, directly on to the 1-liter counter, which contains 10 mm Hg of propane. Water addition is continued until a total pressure of about 760 mm Hg is reached. In this way the evaporation is performed without fractionation; no condensation of hydrogen taking place, one can be sure that the counting gas has the same isotopic composition as the water. Normally no water is condensed in the freezing trap. By introduction of the molecular sieve filters a short-lived radioactive contamination that had spoiled some previous results was avoided.

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The tritium counters are shielded with iron, paraffin, boric acid, lead, and cosmic ray Geiger tubes, as has been described in the above-mentioned report and in the C14 paper by ÖSTLUND & ENGSTRAND (1963). The latter paper also gives constructional details of the counter itself. For tritium Geiger counting, the center wire has a diameter of 0.12 mm and the working voltage is about 3800 V. Electronic quenching is applied with a simple circuit. The hydrogen inside the efficient volume is equivalent to 0.7 g of water and backgrounds are about 2.0 cpm. If the amount of propane is increased to 150 mm or more, and a thinner center wire is used, the counters work nicely as proportional counters, and by rejecting pulses above the maximum energy of tritium, the background could be decreased some 0.1 cpm compared with the Geiger counting. So far we have not considered it worth the trouble.

BAINBRIDGE *et al.* (1961) have developed a method of synthesizing ethane from the sample hydrogen and acethylene. The ethane, containing 4 atoms of sample hydrogen, can be easily filled to a higher pressure thus giving much higher water equivalent. With a volume enrichment of 200-fold and starting with larger amounts of water than we do (cf. below) his method pushes the detection threshold further down. For samples with moderately low tritium assays our method should be faster, requiring only 15 minutes from start of water reduction to start of counting.

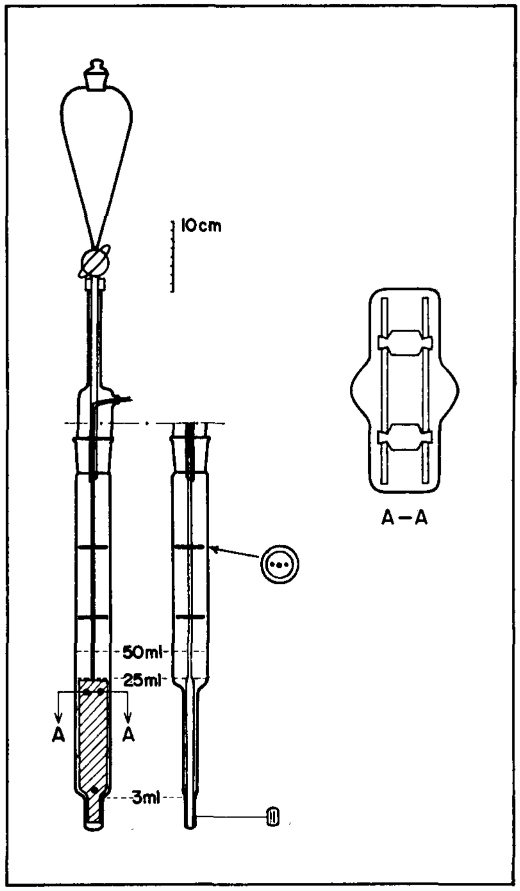

FIG. 1. Electrolysis cell for periodic addition. Section A–A shows the cross section and teflon spacers for the electrodes.

Enrichment procedure

Our electrolytic enrichment procedure has been described by ÖSTLUND & WERNER (1962), and only its major features will be mentioned here. Of 250 ml of water, 50 ml is charged in the electrolysis cell, Fig. 1, and about 0.7 g of NaOH is added. The remaining 200 ml is stored in the drop-funnel on top of the cell. The current, 3 A, is chosen so that 25 ml of water is disintegrated per 24 hours. The cells are kept at 10°C. Each day a portion of 25 ml is added to the electrolyzing solution, and after 9 days only 25 ml of solution remains. With lower current the electrolysis is now continued until about 3 ml of solution is left. After neutralization with CO_2, the water is vacuum-frozen into a weighing bottle and weighed. This quantity, about 2.5 g, is sufficient for two fillings of the counter.

There are several advantages with this periodic addition procedure compared with an equivalent two-step electrolysis. It reduces the amount of manual work spent on each sample, and it decreases the danger of contamination from the air. This system needs only 1/10 of the amount of electrolyte necessary for the conventional electrolysis, which makes it possible to use commercial NaOH. Even 100 TU in the NaOH adds less than 0.1 TU in the final measurement of a tritium-free water sample. The disadvantage with our system is that the recovery of tritium is about 15% lower than two-step batch electrolysis with the same enrichment factor. This is accounted for by using corrected enrichment factors.

Enrichment formulae

Electrolyzing V ml of water, containing n_P moles of protium, n_D moles of deuterium, and n_T moles of tritium, these quantities follow the equation:

$$\frac{dn_P}{n_P} = \alpha \frac{dn_D}{n_D} = \beta \frac{dn_T}{n_T}. \qquad (1)$$

Integrating this equation from the initial n_P^0, n_D^0 and n_T^0 to the final condition, considering that $n_P \gg n_D \gg n_T$, assuming the molar volume of H_2O and D_2O to be equal, and using α and β as the average values of these factors during the electrolysis, we get:

$$\frac{V}{V_0} = \left[\frac{n_D}{n_D^0}\right]^{\alpha} = \left[\frac{n_T}{n_T^0}\right]^{\beta}. \qquad (2)$$

If the fractional recoveries of protium (V/V_0), deuterium (n_D/n_D^0), and tritium (n_T/n_T^0) are denoted R_V, R_D and R_T respectively, formula (2) becomes:

$$R_V = R_D^{\alpha} = R_T^{\beta}. \qquad (3)$$

By using Xs for the molar fractions of the isotopes in question, (2) can be transformed to:

$$X_T^0 = X_T \cdot \frac{V}{V_0} \left[\frac{X_D^0 V_0}{X_D V}\right]^{\alpha/\beta}. \qquad (4)$$

Equation (4) shows which quantities have to be known to determine an unknown assay of tritium in a sample of natural water. BIEGELEISEN (1962) showed theoretically that α/β could *not* be expected to be constant but that $\ln\beta/\ln\alpha$ is fundamentally constant. Measurements by ÖSTLUND & WERNER (1962) showed that

$$\ln \beta/\ln \alpha = 1.337 \pm 0.007. \qquad (5)$$

This expression is to be used with (1) to (4) for the regular batch electrolysis at 10°C.

All the above formulae are valid for batch electrolysis, where the entire sample reacts. When we wish to apply them on our periodic addition electrolysis, we have to consider the fact that only part of the sample takes part in the reaction. Among the possible ways to make this adaption we prefer transforming the enrichment factors so that (3) becomes:

$$R_V = R_D^{\alpha'} = R_T^{\beta'}. \qquad (6)$$

If applied to (6), the factors for our particular enrichment system follow the formula:

$$\ln \beta'/\ln \alpha' = 1.369 \pm 0.006. \qquad (7)$$

Measurements and deductions for this case were reported by ÖSTLUND & WERNER (loc. cit.).

Standards and uncertainties

So far we have not been able to obtain sufficiently constant fractionation factors from run to run. This means that normally deuterium is measured in original water and in the enriched sample. From these values α' is calculated, and β' for the actual run can be obtained from (7). The D measurements were made mass-spectrometrically, with standard errors of ± 3 ppm for natural waters and $\pm 3\%$ (relative error) for the concentrates. The natural assays were standardized on the absolute scale (NIEF & BOTTER, 1959). In this scale, the NBS-1 Deuterium Standard Water has the absolute value of 154.2 ± 0.4 ppm (ROTH, 1963, personal communication) and subsequently Standard Mean Ocean Water, SMOW, (CRAIG, 1961) is 161.9 ± 0.5 ppm. The concentrates were standardized against dilutions of 99.9% heavy water. In the final X_T value the two errors contribute about $\pm 2\%$.

In many cases, only 10-fold enrichment was applied owing to high expected tritium assay or limited amount of sample. D measurements on a long series of such runs showed an average of $\beta = 19.5 \pm 2.5$. For the case $R_V > 5\%$ this allows us to omit D measurement and yet calculate R_T with $\pm 2.2\%$ relative error.

In a few cases we have reported values obtained after 1000-fold enrichment, performed by pre-enriching a 2500 ml sample to 250 ml in a separate cell. Experience with this procedure has shown it to be rather unsatisfactory, and such tritium values are reported only when completely independent double checks agreed.

All tritium figures are expressed in TU, tritium units (TU $= 10^{18} X_T^0$), and are standardized against the NBS standard tritium water of 1954. Careful cross checks with samples of known (by preparation) tritium assay have been run by us and some other laboratories to affix our values securely to this scale. If no error is given, the uncertainty (as 1 σ) is estimated to $\pm 3\%$. In this error is not included the $\pm 3\%$ adherent to the original NBS water stated by the Bureau.

Double checks and interlaboratory cross checks

In order to check the overall reliability and reproducibility of our system we have run several series of double checks, and some samples have allowed measurement to be made both before and after enrichment. One of these series consists of the Stockholm Reference and the IAEA Reference Samples. These results are reported in another paper in this issue (Östlund *et al.*, 1964) and they rigidly nail our tritium data down to the NBS scale. One series of double check samples was made soon after the routine work had begun. The results are reported in Table 1.

Recently Dr. Robert Brown at the Chalk River tritium laboratory kindly submitted a series of samples from 1952–1956, analyzed by his laboratory (Brown & Grummitt, 1956). Our measurements, with double and triple analyses, and direct counting checked well among themselves. By comparison with Brown's original values, however, the agreement was only within about 20%. Since our values were mostly on the high side, and the samples had been stored during many years, the small discrepancy could very well be due to contamination during storage.

In our normal procedure, starting with 250 ml or less, no difficulties have been experienced in obtaining zero tritium values. This is illustrated by the Sabi Valley series reported in Table 3*c*. Another series of zero samples is reported *among the measurements on the Stockholm* Reference Samples.

Among the monthly rain water series reported in this paper are samples from Valentia, Ireland. There, the collected amount of water each month has been divided into two parts, each about 1 liter. One portion has been sent to us in sealed tin containers, and the other to the Los Angeles laboratory. In spite of that, our values and the Los Angeles values (Libby, 1961 and 1963) on the same rains disagreed with factors between 0.3 and 15. To prove at least to ourselves that we have not been subjected to technical faults, we have made a considerable number of double checks and direct *measurements* (*without enrichment*) *in this* series. A comparison of our measurements on samples also measured in Los Angeles is given in Table 2. In Los Angeles nothing is left of these samples (Libby, 1961, personal communication) but in Stockholm 10–500 ml are available for any laboratory being able to analyze this small quantity.

Table 1. *Results on double checks and direct measurements.*

Sample	St No.	V_0/V	TU
Huddinge, snow	T33A	57	524 ± 16
580401–03	T33D	direct	510 ± 25
Huddinge, rain 580604	T43A	59	562 ± 17
	T43A	same	544 ± 17
	T43D	direct	536 ± 20
Murchison Bay, rain	T47A	80	925 ± 30
580710	T47D	direct	879 ± 35
Pykäjärvi, lake 580401	T48A	78	41.4 ± 1.6
	T48B	69	38.5 ± 1.2
Mälaren, lake 580323	T58A	66	49.1 ± 2.0
	T58B	85	49.0 ± 1.5
	T58B	same	48.2 ± 1.5
	T58C	75	46.4 ± 1.5
Huddinge, rains 5902	T102A	115	818 ± 25
	T102D	direct	764 ± 30
	T102E	direct	773 ± 30
Huddinge, rains 5903	T103A	96	728 ± 25
	T103D	direct	686 ± 20
Huddinge, rains 5904	T104A	105	765 ± 25
	T104D	direct	739 ± 20
Huddinge, rains 5905	T105A	81	889 ± 30
	T105D	direct	920 ± 30
Huddinge, rains 5907	T107A	90	686 ± 25
	T107D	direct	660 ± 20

Comments to Table 3

In Table 3 below are reported results of measurements on various water samples. The StT number is a laboratory serial number. In order *to save space in the* "*Date*" *columns a particular* day is written year (57), month (10), day (04), which easily may be extended to include hour and minute. Amount of precipitation is given when available, as is run-off for rivers. Deuterium figures are in ppm absolute scale (cf. above) with an uncertainty of ± 3 ppm (1σ). The standard error of the tritium figures is $\pm 3\%$ when not otherwise stated.

The precipitation stations, as well as the river samples, are arranged in the order of latitude from north to south.

Most of the precipitation samples have been submitted by Dr. Svante Odén at the International Institute of Meteorology of the University of Stockholm (Odén, 1964). They were

TABLE 2. *Double checks and comparison with Los Angeles results on Valentia rain samples.*

(Our entire Valentia series is reported in Table 3 *a* below).

Year/ Month	StT No.	V_0/V	TU	Sthlm. average TU	Los Angeles value
5801	157A	78	65 ± 2	63 ± 2	18 ± 3
	157B	65	58 ± 3		
	157D	Direct	88 ± 30		
	157E	Direct	63 ± 20		
	157F	Direct	63 ± 15		
5807	162A	74	273 ± 10	278 ± 10	53 ± 1.5
	162D	Direct	287 ± 20		
5904	171A	79	275 ± 10	270 ± 10	17 ± 1.5
	171B	66	271 ± 10		
	171D	Direct	270 ± 20		
	171E	Direct	258 ± 20		
5906	188	70	198 ± 6	198 ± 6	61 ± 2.0
5908	189	94	76 ± 3	76 ± 3	19 ± 1.4
5909	190	110	78 ± 3	78 ± 3	14 ± 3
5911	173A	88	18.3 ± 0.6	20.3 ± 0.3	8.1 ± 1.6
	173B	123	22.3 ± 0.3		
5912	174	77	27.7 ± 0.9	27.7 ± 0.9	14 ± 1.3
6001	176A	83	21.7 ± 0.8	21.7 ± 0.8	5 ± 0.7
6002	288	91	37.6 ± 1.5	37.6 ± 1.5	15 ± 1.0
6004	290	73	61.9 ± 2.0	61.9 ± 2.0	34 ± 2.1
6005	291A	76	94.3 ± 3.0	94 ± 3	23 ± 1.2
	291D	Direct	79 ± 18		
6006	292A	60	144 ± 5	144 ± 5	27 ± 1.2
	292D	Direct	144 ± 16		
6008	294A	52	48.4 ± 1.5	50.3 ± 1.0	12 ± 0.7
	294B	67	52.1 ± 1.5		
6009	295	78	39.0 ± 1.2	39.0 ± 1.2	11 ± 0.8
6010	296A	74	25.2 ± 0.8	26.4 ± 0.4	5.5 ± 0.4
	296B	79	27.5 ± 0.4		
6101	299	88	20.6 ± 0.7	20.6 ± 0.7	9.4 ± 1.2
6102	300A	92	27.2 ± 1.0	27.8 ± 1.0	20 ± 0.6
	300B	16	31.6 ± 2.5		
6103	301	79	23.1 ± 0.7	23.1 ± 0.7	48 ± 2.2
6106	398	75	82 ± 2.5	82 ± 2.5	49 ± 1.0
6107	399	89	72 ± 2.2	72 ± 2.2	43 ± 1.4
6108	400	79	32.8 ± 1.0	32.8 ± 1.0	30 ± 0.7
6111	510	67	89 ± 3	89 ± 3	306 ± 5
6112	512	66	108 ± 3	108 ± 3	106 ± 2.2
6202	515	59	121 ± 4	121 ± 4	147 ± 2.8
6204	657A	76	339 ± 10	342 ± 9	198 ± 2.8
	657D	Direct	354 ± 21		
6205	660A	59	441 ± 13	455 ± 12	
	660D	Direct	492 ± 20		
6206	661A	47	354 ± 11	348 ± 10	
	661D	Direct	325 ± 20		
6207	658A	57	406 ± 13	407 ± 12	269 ± 2.3
	658D	Direct	412 ± 25		
6208	659	64	230 ± 7	230 ± 7	132 ± 3.3
6209	662	59	172 ± 5	172 ± 5	114 ± 2.3
6210	746	71	92 ± 3	92 ± 3	64 ± 1.9

TABLE 3*a*. *Precipitation Samples.*

StT No.	Date	Amount mm	Deuterium X_D^0 ppm	Tritium X_T^0 TU
Kinnvika, 80°00′ N Lat., 18°50′ E Long., Spitsbergen[a]				
252	571004–580323		148	106
253	580330		149	107
254	580514–580530		151	295
255	580610–580628		154	480
256	580704–580710		140	836
257	580713–580806		146	642
Tana, 70°24′ N Lat., 28°12′ E Long., Norway				
532	1961 Sept.	51	152	77
526	1962 Jan.	24	147	572
547	1962 March	56	—	289
Nellim, 68°51′ N Lat., 28°17′ E Long., Finland				
560	1961 Nov.		141	242
561	1962 Jan.		145	822
571	1962 March		145	1048
603	1962 May		145	1429
597	1962 July		145	1647
Kiruna, 67°52′ N Lat., 20°14′ E Long., Sweden				
373	1961 July	107	150	257
419	1961 Sept.	43	147	64
437	1961 Nov.	43	142	138
525	1962 Jan.	40	139	666
711	1962 July	45	140	1387
Pelkosenniemi, 67°06′ N Lat., 27°30′ E Long., Finland				
568	1961 Nov.	60	145	181
569	1962 Jan.		141	645
570	1962 March		140	912
601	1962 May		143	1536
Arjeplog, 66°03′ N Lat., 17°54′ E Long., Sweden				
374	1961 July	140	150	305
421	1961 Sept.	36	146	69
439	1961 Nov.	48	137	163
522	1962 Jan.	46	142	406
706	1962 July	50	145	1320
Bredkälen, 63°54′ N Lat., 15°18′ E Long, Sweden				
629	1961 Nov.	19	146	116
627	1962 Jan.	47	142	562
Röbäcksdalen, 63°48′ N Lat., 20°12′ E Long., Sweden				
375	1961 July	81	150	231
422	1961 Sept.	26	153	55 ± 5
440	1961 Nov.	66	132	159
524	1962 Jan.	74	141	480
585	1962 May	42	146	1036
718	1962 July	65	152	931

StT No.	Date	Amount mm	Deuterium X_D^0 ppm	Tritium X_T^0 TU
Ytteröy, 63°48′ N Lat., 11°13′ E Long., Norway				
384	1961 July	38	152	194
431	1961 Sept.	85	146	45 ± 2
448	1961 Nov.	77	154	87
535	1962 Jan.	59	152	257
550	1962 March	31	151	376
719	1962 July	28	154	769
Gjermundnes, 62°37′ N Lat., 07°10′ E Long., Norway				
519	1961 July	107	152	121
Lauka, 62°29′ N Lat., 25°45′ E Long., Finland				
554	1961 Nov.	43	147	135
555	1962 Jan.		142	747
556	1962 March		141	1156
600	1962 May		149	1191
602	1962 July		150	1503
Jalasjärvi, 62°18′ N Lat., 22°55′ E Long., Finland				
557	1961 Nov.	43	148	96
558	1962 Jan.		145	488
559	1962 March		140	1092
596	1962 May		150	1216
599	1962 July		149	1309
Fanaråken, 61°31′ N Lat., 07°54′ E Long., Norway				
385	1961 July	102	149	135
432	1961 Sept.	146	139	41.6 ± 1.7
449	1961 Nov.	119	154	159
542	1962 Jan.	136	147	395
553	1962 March	61	146	430
589	1962 May	68	149	671
715	1962 July	43	159	562
Trysil, 61°20′ N Lat., 12°15′ E Long., Norway				
530	1961 July	76	151	222
531	1961 Sept.	80	151	63
553	1961 Nov.	78	150	119
544	1962 Jan.	57	147	442
626	1962 March	16	143	1235
578	1962 May	54	149	905
707	1962 July	85	152	1209
Åmot, 60°58′ N Lat., 16°25′ E Long., Sweden				
376	1961 July	96	153	203
423	1961 Sept.	34	153	61
441	1961 Nov.	52	144	94
521	1962 Jan.	61	—	432
497	1962 March	39	145	824
583	1962 May	61	150	1012
712	1962 July	59	147	1112
Værvaslingan, 60°23′ N Lat., 05°21′ E Long., Bergen, Norway[b]				
403	1958 Febr.	79	150	88
404	1958 March	64	157	101
405	1958 Apr.	101	161	209

Table 3 *a* (*continued*)

StT No.	Date	Amount mm	Deuterium X_D^0 ppm	Tritium X_T^0 TU
406	1958 May	142	158	238
407	1958 June	26	162	546
408	1958 July	185	152	258
409	1958 Aug.	191	157	357
410	1958 Sept.	72	154	119
411	1958 Oct.	295	155	79
412	1958 Nov.	291	158	55.2
355	1958 Dec.	118	155	184
356	1959 Jan.	90	160	283
360	1959 Febr.	176	157	191
361	1959 March	93	155	409
362	1959 Apr.	152	158	513
357	1959 May	72	162	630
354	1959 June	183	162	372
358	1959 July	119	166	412
363	1959 Aug.	124	159	126
Forshult, 60°10′ N Lat., 13°47′ E Long., Sweden				
378	1961 July	83	150	138
426	1961 Sept.	59	149	61.6
443	1961 Nov.	69	151	109
539	1962 Jan.	68	149	340
552	1962 March	37	144	1015
590	1962 May	82	150	874
714	1962 July	107	152	1067
Erken, 59°54′ N Lat., 18°42′ E Long., Sweden				
389	1961 July	119	148	172
425	1961 Sept.	41	155	59
442	1961 Nov.	29	150	89
517	1962 Jan.	64	147	428
549	1962 March	39	147	734
574	1962 May	39	146	822
Ultuna, 59°48′ N Lat., 17°42′ E Long., Sweden				
377	1961 July	142	148	168
424	1961 Sept.	46	154	50 ± 2
520	1962 Jan.	52	146	446
628	1962 June	50	153	974
716	1962 July	81	150	868
Ås, 59°40′ N Lat., 10°47′ E Long., Norway				
387	1961 July	78	149	163
434	1961 Sept.	115	153	60
451	1961 Nov.	103	153	132
540	1962 Jan.	74	149	365
546	1962 March	38	144	875
587	1962 May	99	150	900
709	1962 July	101	150	1030
Dalen, 59°27′ N Lat., 08°00′ E Long., Norway				
386	1961 July	56	147	186
433	1961 Sept.	126	146	58
450	1961 Nov.	96	146	154
518	1962 Jan.	72	147	288
551	1962 March	59	—	815
584	1962 May	75	153	698
Huddinge, 59°14′ N Lat., 17°59′ E Long., Stockholm, Sweden				
74	1958 March	14	144	217
75	1958 Apr.	48	143	401
85	1958 May	90	147	437
89	1958 June	47	153	468
90	1958 July	56	154	387
91	1958 Aug.	53	144	391
92	1958 Sept.	21	145	151
95	1958 Oct.	37	155	103
98	1958 Nov.	42	148	137
114	1958 Dec.	73	150	352
101	1959 Jan.	73	140	487
102	1959 Febr.	7	139	818
103	1959 March	29	141	728
104	1959 Apr.	75	145	765
105	1959 May	20	149	889
107	1959 June	18	154	686
108	1959 July	6	153	407
109	1959 Aug.	20	151	269
111	1959 Sept.	41	146	154
134	1959 Oct.	41	153	85
135	1959 Nov.	48	144	87
136	1959 Dec.	56	145	56.4
184	1960 Jan.	46	145	65
185	1960 Febr.	6	141	120
186	1960 March	17	147	98
212	1960 Apr.	20	149	209
191	1960 May	29	157	369
215	1960 June	13	152	166
216	1960 July	128	151	254
218	1960 Aug.	174	150	161
219	1960 Sept.	51	149	85
220	1960 Oct.	50	146	63
278	1960 Nov.	84	147	43.2
279	1960 Dec.	67	144	47.4
280	1961 Jan.	22	140	49.0
281	1961 Febr.	10	145	57.4
388	1961 March	21	147	78
390	1961 Apr.	13	142	97
391	1961 May	57	150	150
392	1961 June	77	155	212
393	1961 July	151	151	155
394	1961 Aug.	94	153	111
395	1961 Sept.	30	157	49.2
396	1961 Oct.	54	153	59
414	1961 Nov.	58	147	107
415	1961 Dec.	14	142	275
416	1962 Jan.	60	148	478
482	1962 Febr.	46	146	818
483	1962 March	29	147	721
484	1962 Apr.	33	140	1004
538	1962 May	48	151	973
537	1962 June	61	150	968
581	1962 July	63	150	904
582	1962 Aug.	116	150	529
592	1962 Sept.	96	150	448
622	1962 Oct.	41	150	225
623	1962 Nov.	31	137	424
664	1962 Dec.	28	139	313
Vreta Kloster, 58°36′ N Lat., 15°38′ E Long., Sweden				
379	1961 July	51	152	137
427	1961 Sept.	39	151	5.3 ± 1.9

Table 3 *a* (*continued*)

StT No.	Date	Amount mm	Deuterium X_D^0 ppm	Tritium X_T^0 TU
444	1961 Nov.	35	147	90
541	1962 Jan.	57	144	378
527	1962 March	24	145	859
591	1962 May	25	149	773
704	1962 July	58	148	901
849	1962 Sept.	66	147	420
Göteborgs frihamn, 57°42′ N Lat., 11°58′ E Long., Sweden				
383	1961 July	94	151	111
420	1961 Sept.	88	158	56.2
438	1961 Nov.	77	153	95
543	1962 Jan.	90	148	294
548	1962 March	32	150	696
579	1962 May	71	155	627
710	1962 July	87	150	920
840	1962 Sept.	100	152	362
Smedby, 56°42′ N Lat., 16°12′ E Long., Sweden				
381	1961 July	119	151	177
429	1961 Sept.	56	151	64
446	1961 Nov.	40	151	127
523	1962 Jan.	26	149	410
588	1962 May	40	153	862
713	1962 July	40	153	935
847	1962 Sept.	41	149	343
Plönning, 56°41′ N Lat., 12°45′ E Long., Sweden				
380	1961 July	159	153	145
428	1961 Sept.	95	153	65
445	1961 Nov.	39	146	146
536	1962 Jan.	63	151	373
573	1962 May	90	151	672
717	1962 July	85	154	728
841	1962 Sept.	76	156	306
Ödum, 56°12′ N Lat., 10°05′ E Long., Denmark				
624	1961 Nov.	56	143	102
630	1962 Jan.	63	152	310
625	1962 March	38	145	740
708	1962 July	139	149	1034
848	1962 Sept.		143	363
Askov, 55°30′ N Lat., 09°08′ E Long., Denmark				
577	1962 May	116	149	744
Skurup, 55°28′ N Lat., 13°30′ E Long., Sweden				
382	1961 July	98	153	131
430	1961 Sept.	69	157	63
447	1961 Nov.	62	149	101
534	1962 Jan.	76	152	287
528	1962 March	21	150	665
572	1962 May	76	155	691
705	1962 July	75	149	899
846	1962 Sept.	50	155	288
Valentia Observatory at Caherciveen, 51°06′ N Lat. 10°14′ W Long., Ireland (cf. Table 2)				
154	1957 Sept.	165	157	32.6 ± 1.2
155	1957 Oct.	145	154	21.0

StT No.	Date	Amount mm	Deuterium X_D^0 ppm	Tritium X_T^0 TU
156	1957 Dec.	131	156	28.7
157	1958 Jan.	157	153	65 ± 2
158	1958 Febr.	131	156	67
159	1958 March	133	153	117
187	1958 April	41	158	103
160	1958 May	127	153	250
161	1958 June	127	157	213
162	1958 July	112	160	273
163	1958 Aug.	139	156	286
164	1958 Sept.	201	153	63
166	1958 Oct.	102	157	55.8
168	1958 Dec.	150	151	166
169	1959 Jan.	120	155	177
170	1959 March	169	155	214
171	1959 Apr.	113	153	276
188	1959 June	68	160	198
189	1959 Aug.	63	160	76
190	1959 Sept.	61	158	78
172	1959 Oct.	209	160	30.6
173	1959 Nov.	196	158	18.3
174	1959 Dec.	319	154	27.7
176	1960 Jan.	164	158	21.7
288	1960 Febr.	135	149	37.6
289	1960 March	126	153	36.0
290	1960 Apr.	99	154	62
291	1960 May	85	157	94.3
292	1960 June	69	152	144
293	1960 July	175	157	90
294	1960 Aug.	60	157	50.3
295	1960 Sept.	163	154	39.0
296	1960 Oct.	123	152	25.2
297	1960 Nov.	274	154	17.3
298	1960 Dec.	224	156	16.3
299	1961 Jan.	267	148	20.6
300	1961 Febr.	123	152	27.8
301	1961 March	19	156	23.1
302	1961 Apr.	154	152	50.9
397	1961 May	39	157	46.6
398	1961 June	60	157	82
399	1961 July	93	154	72
400	1961 Aug.	47	157	32.8
402	1961 Sept.	207	163	17.8
401	1961 Oct.	172	153	21.6
510	1961 Nov.	100	154	89
512	1961 Dec.	100	153	108
514	1962 Jan.	167	153	173
515	1962 Febr.	36	159	121
516	1962 March	128	153	288
657	1962 Apr.	74	152	339
660	1962 May	95	153	441
661	1962 June	69	160	354
658	1962 July	69	156	406
659	1962 Aug.	131	155	230
662	1962 Sept.	140	150	172
746	1962 Oct.	87	157	92
747	1962 Nov.	105	154	179
748	1962 Dec.	132	154	172

Table 3 *a* (*continued*)

StT No.	Date	Amount mm	Deuterium X_D^0 ppm	Tritium X_T^0 TU
Seawell Airport, 13°05′ N Lat., 59°30′ W Long., Barbados				
121	1959 May		161[c]	26.9 ± 1.0
122	1959 June		161[c]	21.0 ± 1.0
123	1959 July		161[c]	3.6 ± 0.2
—	1959 Aug.[d]		—	—
124	1959 Sept.		161	14.4 ± 0.5
125	1959 Oct.		158	10.0 ± 0.5
126	1959 Nov.		162	7.0 ± 0.8
127	1959 Dec.		161	8.3 ± 0.7

StT No.	Date	Amount mm	Deuterium X_D^0 ppm	Tritium X_T^0 TU
Aspendale, 38°20′ S Lat., 145°00′ E Long., Melbourne, Australia.				
305	1960 Jan.	51	159	13.9
306	1960 Febr.	34	154	13.5
307	1960 March	9	162	21.5
308	1960 Apr.	112	153	8.2 ± 0.4
309	1960 May	92	151	9.0 ± 0.4
310	1960 June	31	159	5.9 ± 0.4
311	1960 July	92	154	11.2
312	1960 Aug.	67	159	28.2
313	1960 Sept.	92	155	32.3
314	1960 Oct.	—	163	20.9

[a] Samples collected during the IGY by K. Wärme.
[b] Samples collected during the IGY by Forsvarets forskningsinstitutt in Kellar.
[c] Not measured, average of samples StT 124–127.
[d] No rain.

TABLE 3*b*. *Lakes, Rivers, and Bays.*

StT No.	Date	Run-off $m^3 sec^{-1}$	Deuterium X_D^0 ppm	Tritium X_T^0 TU	Remarks
Inari, Lake, 68°51′ N Lat., 28°17′ E Long., Finland					
586	6111	164	147	120	
593	6202	64	145	150	
594	6205	458	146	293	
648	6208	181	153	207	
Abiskojokk, lake, 68°20′ N Lat., 18°49′ E Long., 200 m SW of the railway bridge, Sweden					
197	580319		144	58.5	
198	590629		145	330	
Torne älv, river, 67°51′ N Lat., 20°37′ E Long., Sweden[a]					
751	620702		140	173	
752	621002		141	234	
Stora Lule Älv, river, at Messaure, 66°40′ N Lat., 20°33′ E Long, Sweden					
193	580420		149	66.2	
194	590624		149	149	
Kemijärvi, lake, 66°27′ N Lat., 27°20′ E Long., Finland					
49	580402		143	74.3	
207	590610		146	243	
Hornavan, river, at Arjeplog, 66°03′ N Lat., 17°55′ E Long., Sweden					
203	580414		145	23.1	
204	590704		146	61.6	
Råne Älv, river, 66°01′ N Lat., 20°00′ E Long., Sweden[b]					
749	620705		144	400	
750	620930		144	501	

StT No.	Date	Run-off $m^3 sec^{-1}$	Deuterium X_D^0 ppm	Tritium X_T^0 TU	Remarks
Kemi, river, at Taivalkoski, 65°57′ N Lat., 24°42′ E Long., Finland					
621	6111	873	145	118	
620	6202	224	142	111	
631	6205	2101	144	337	
651	6208	425	147	466	
Skellefte Älv, 65°41′ N Lat., 18°08′ E Long., Sweden					
55	580327		146	44.5	
192	590628		148	193	
656	620704		144	189	
755	621001		150	261	
Pjorsa, river, at Urridafoss, 63°55′ N Lat., 20°41′ w Long., Iceland					
645	611101		150	26.4	
644	620215		152	71.5	
647	620501		146	273	
646	620801		146	153	
Åre, lake, 63°23′ N Lat., 27°20′ E Long., Sweden					
205	580330		149	55.5	
50	590406		146	160	
267	6008		150	176	
452	6108		149	125	
Kyrö, river, at Lansorsund, 63°09′ N Lat., 21°51′ E Long., Finland					
595	6111	71	149	113	
605	6202	17.5	146	172	
604	6205	109	145	430	
652	6208	26	146	489	
Ljungan, river, 62°30′ N Lat., 16°03′ E Long., Sweden					
206	580407		149	49.9	
61	590408		147	205	
270	6008		146	169	

Table 3 *b* (*continued*)

StT No.	Date	Run-off $m^3 sec^{-1}$	Deuterium X_D^0 ppm	Tritium X_T^0 TU	Remarks
Päijänne, lake, at Kalkinen, 61°17′ N Lat., 25°36′ E Long, Finland					
575	6111		148	123	
576	6202		148	129	
580	6205		151	143	
650	6208		150	189	
Ljusnan, river, 61°12′ N Lat., 17°08′ E Long., Sweden					
51	580407		145	71.5	
63	590408		144	145	
Pyhäjärvi, lake, at Pyynikki, 61°00′ N Lat., 22°17′ E Long., Finland					
48	580401		145	41.3	
Siljan, lake, at Tällberg, 60°50′ N Lat., 14°58′ E Long., Sweden					
53	580310		147	56.1	
Österdalälven, river, 60°37′ N Lat., 15°01′ E Long., Sweden					
756	620930		150	200	
Dalälven, river, at Älvkarleby, 60°34′ N Lat., 17°27′ E Long, Sweden					
52	580407		147	86.5	
60	590408		150	189.0	
268	6008		147	122	
753	621001		141	310	
Västerdalälven, river, 60°29′ N Lat., 14°47′ E Long Sweden[c]					
754	620930		142	353	
Indalsälven, river, 60°05′ N Lat., 16°22′ E Long., Sweden[d]					
731	620712		148	208	
732	621001		148	268	
Klarälven, river, 60°01′ N Lat., 13°40′ E Long., Sweden[e]					
722	620703		144	271	
723	621001		149	321	
Siksjödammen, lake, Bastkärn, 62°02′ N Lat., 14°59′ E Long., Sweden					
66	590624		151	151	
Forshult, powerstation, 59°58′ N Lat., 13°32′ E Long., Sweden					
195	580317		149	49.5	
196	590624		145	155	
Mälaren, lake, at Hjulsta, 59°32′ N Lat., 16°01′ E Long., Sweden					
58	580323		148	48.2	
Kyrkfjärden, bay, NE of Stockholm, 59°26′ N Lat., 18°11′ E Long., Sweden					
128	600221		151	47.0	Bottom
129	600221		151	47.1	14 m
130	600221		152	51.8	10 m
131	600221		152	94.9	Surface
132	600221		151	51.9	Bottom
133	600221		151	55.8	5 m
230	601226		152	53.4	Bottom
228	601226		149	81.2	Surface
232	601226		149	98.8	Surface
234	601226		152	50.7	Bottom
Helsundet, bay, 59°26′ N Lat., 18°11′ E Long., Sweden					
231	601226		150	75.6	Surface
Stora Värtan, bay, 59°25′ N Lat., 18°09′ E Long., Sweden					
227	601226		149	87.9	Surface
229	601226		154	41.1	Bottom
Fågelnäsviken, bay, 59°26′ N Lat., 18°10′ E Long., Sweden					
233	601226		151	112	
Norrström, lake, 59°15′ N Lat., 18°04′ E Long., Sweden[f]					
694	621001		148	186	
Glaningen, lake, 59°07′ N Lat., 15°03′ E Long., Sweden					
119	600128		150	147	
Gonäsån, river, at Blötberget, 59°07′ N Lat., 15°03′ E Long.,Sweden					
118	600128		149	146	
Vättern, lake, at Kärra, 58°45′ N Lat., 14°59′ E Long., Sweden					
56	580322		150	9.1 ± 0.4	
265	590625		153	39.5	
Göta kanal at Borensberg, 58°34′ N Lat., 15°17′ E Long., Sweden					
54	580322		153	13.9	
699	620701		157	71.8	
700	621001		155	289	
Vänern, lake, 58°16′ N Lat., 12°17′ E Long., Sweden[g]					
720	620701		151	81.4	
721	621001		151	106	
Göta älv, river, at Bohus, 57°52′ N Lat., 12°01′ E Long., Sweden					
263	580312		151	19.4	
Botorpsströmmen, river, at Brunsö, 57°42′ N Lat., 16°27′ E Long., Sweden[h]					
655	620630		150	176	
757	621001		152	213	
Ätran, river, 57°17′ N Lat., 13°01′ E Long., Sweden[i]					
744	620701		153	245	
745	621001		152	261	

Table 3*b* (*continued*)

StT No.	Date	Run-off m^3 sec^{-1}	Deuterium X_D^0 ppm	Tritium X_T^0 TU	Remarks
Nissan, river, at Fröslida, 66°55′ N Lat., 13°04′ E Long., Sweden					
199	580320		150	52.7	
200	590624		151	185	
Ljungbyån, river, 56°42′ N Lat., 16°08′ E Long., Sweden[k]					
201	580329		153	62.2	
202	590703		154	307.0	
702	620702		151	273	
703	621003		149	320	
Rönneån, river, 56°15′ N Lat., 12°54′ E Long., Sweden[l]					
654	620701		150	218	
Mörrumsån, river, 56°12′ N Lat., 14°46′ E Long., Sweden[m]					
698	620701	23	153	231	
700	621001		155	289	
Rhein, river, at Kehl, 48°26′ N Lat., 7°49′ E Long., Germany					
261	580625		149	100	
Züricher See, lake, at Horgen, 47°12′ N Lat., 8°40′ Switzerland, E Long.					
264	590325		149	83.2	
Vierwaldstättersee, lake, at Luzern, 47°03′ N Lat., 8°19′ E Long., Switzerland					
266	590325		149	65.5	
Sabi valley, river, at Musikivanthu, 20°30′ S Lat., 32°15′ E Long., South Rhodesia					
240	591127		160	6.7 ± 0.3	

[a] Samples collected by A. Henriksson.
[b] Samples collected by I. Engström.
[c] Samples collected by M. Hagström.
[d] Sample collected by S. Duvner.
[e] Samples collected by P. Andersson.
[f] Sample collected by Å .Mattsson.
[g] Samples collected by S. Svensson.
[h] Samples collected by E. Johansson.
[i] Samples collected by N. Persson.
[k] Samples collected by K. G. Nyrén.
[l] Sample collected by L. Nilsson.
[m] Samples collected by S. Samuelsson.

TABLE 3*c*. *Various samples.*

StT No.	Sampling at	Date	Deuterium X_D^0 ppm	Tritium X_T^0 TU	
Gotlandsdjupet, Baltic, 57°21′ N Lat., 19°57′ E Long.					
68	Surface	580215	152	8.0 ± 0.5	
110	200 m	580215	151	1.9 ± 0.1	
167	Surface	580725	150	13.1 ± 0.6	
177	50 m	580725	148	8.1 ± 0.4	
179	100 m	580725	157	3.8 ± 0.2	
182	200 m	580725	158	2.1 ± 0.1	
35	Surface	581108	153	2.3 ± 0.1	
821	Surface	590417	155	20.2	
822	50 m	590417	155	19.6	
823	100 m	590417	154	19.5	
824	200 m	590417	154	18.9	
273	Surface	610111	152	35.5	
274	50 m	610111	152	22.4	
275	100 m	610111	154	18.4	
276	200 m	610111	154	9.5	± 0.5
758	Surface	621025	153	45.7	
759	50 m	621025	154	49.1	
760	100 m	621025	154	17.9	
761	200 m	621025	154	17.3	
Landsortsdjupet, Baltic, 58°35′ N Lat., 18°14′ E Long.					
59	445 m	580216	153	8.3 ± 0.4	
65	Surface	580719	147	15.4 ± 0.7	

StT No.	Sampling at	Date	Deuterium X_D^0 ppm	Tritium X_T^0 TU	
Kristianstadsslätten, groundwater 56°02′ N Lat., 14°09′ E Long., from different wells in agricultural district, Sweden					
606	Vinnö	621116	153	3.6 ± 0.5	
607	Kristian-stad	621116	153	3.3 ± 0.5	
649	Vinslöv	621116	151	15.6 ± 0.6	
679	Vinslöv	630122	147	13.6 ± 0.7	
675	Gärds-köpinge	630123	150	0.6 ± 0.25	
676	Ljungby	630123	147	0.06 ± 0.42	
677	Vinnö	630123	159	4.62 ± 0.37	
678	Kristian-stad	630123	147	1.55 ± 0.38	
733	Åhus	630128	149	28.9 ± 1.1	
734	Yngsjö	630128	151	0.91 ± 0.18	
735	Maltes-holm	630131	151	3.06 ± 0.37	
767	Tollarp	630426	147	278	Rain
Ryholm, ground water, 58°52′ N Lat., 14°15′ E Long., Sweden					
792	Surface	630419	140	663	Peatbog
793	2 m	630419	141	33.1	Peatbog
794	3 m	630419	142	19.6	Peatbog
795	4 m	630419	143	61.5	Peatbog

Table 3 c (*continued*)

StT No.	Sampling at	Date	Deuterium X_D^0 ppm	Tritium X_T^0 TU	
Mörhult, ground water, 57°31′ N Lat., 14°15′ E Long., Sweden					
796	0.5 m	630418	142	96.7	Peatbog
797	1.5 m	630418	141	17.1	Peatbog
798	2.5 m	630418	142	23.7	Peatbog
799	3.5 m	630418	152	7.3 ± 1.2	Peatbog
800	4.5 m	630418	143	11.1	Peatbog
Sabi Valley, ground water, Musikivanthy reserve, 20°30′ S Lat., 27°30′ E Long., Southern Rhodesia[a]					
247		591125	157	−0.1 ± 0.2	
249		591125	157	0.8 ± 0.2	
244		591126	156	0 ± 0.2	
245		591126	157	0 ± 0.2	
246		591126	157	−0.2 ± 0.2	
248		591126	159	−0.5 ± 0.2	
250		591126	156	0 ± 0.2	
241		591127	155	0.6 ± 0.3	
242		591127	157	−0.2 ± 0.2	
243		591127	158	−0.2 ± 0.2	
251		591127	156	−0.4 ± 0.2	
(Comment (E. Eriksson): Above sampling was made for estimation of turn-over time of the ground water by age determination of the water.)					
Saigon, ground water, 11°00′ N Lat., 106°40′ E Long., South Vietnam					
237	44.8 m	600328	154	4.84 ± 0.3	
238	49.2 m	600328	156	4.93 ± 0.3	
239	204.57 m	600328	154	5.25 ± 0.3	
236	36.2 m	600328	154	6.44 ± 0.3	
235		600401	158	19.1	Rain
(Comment: Saigon ground water samples have been taken from wells at four different sites in order to determine the velocity of the water replacement in the well.)					
Västisen, glacial ice, 80°00′ N Lat., 20°00′ E Long., Spitsbergen[b]					
258	100–160 cm	580116	146	96.4	
259	firn 180 cm	580329	148	92.1	
260	200 cm	580416	145	91.9	
Gråsubreen, glacial ice,[c]					
499		620414	144	103	
500		620414	150	2.0 ± 0.16	
501		620414	145	40.9	
502		620414	152	1.9 ± 0.3	
503		620414	145	2.6 ± 0.45	
768		630405	141	26.4	
Ultuna, vegetation, 59°48′ N Lat., 17°42′ E Long., Sweden					
726	0–20 cm	620710	144	616	Soil
727	20–40 cm	620710	147	312	Soil
728	40–60 cm	620710	143	163	Soil
729	60–80 cm	620710	147	152	Soil
730	80–100 cm	620710	149	147	Soil
608	0–20 cm	620910	148	508	Soil
609	20–40 cm	620910	145	279	Soil
610	40–60 cm	620910	146	192	Soil
611	60–80 cm	620910	147	183	Soil
612	80–100 cm	620910	148	151	Soil
615		620702	152	598	Leaves
616		620720	150	590	Leaves
617		620810	148	647	Leaves
618		620830	145	492	Leaves
619		620920	149	182	Leaves
777		620702	145	429	Oat
613		620720	149	658	Oat
776		620810	143	500	Oat
614		620830	152	479	Oat
779		620702	152	473	Barley
725		620720	153	587	Barley
778		620810	150	565	Barley
724		620830	147	497	Barley
838		620920	146	424	Barley
773		620710	143	184	Ground-water
774		620810	145	156	Ground-water
775		620910	142	142	Ground-water
Skurup, vegetation, 55°25′ N Lat., 13°30′ E Long., Sweden					
789		620701	143	354	Leaves
790		620720	154	485	Leaves
769		620901	147	263	Leaves
770		620920	143	252	Leaves

[a] Samples collected by T. Shand. [b] Samples collected during the IGY by K. Wärme.
[c] Samples collected by G. Östrem, Stockholm University.

selected from the Scandinavian network and from the global network maintained by the International Atomic Energy Agency, Vienna, although 128 samples from Spitzbergen, Iceland, Norway, Stuttgart, Uganda, and South Africa are omitted, being published during 1964. In Huddinge each separate rain since March 1958 has been collected by Mr. Sven Östlund, and monthly samples prepared by mixing amounts proportional to the amount of each particular rain. All other rain samples were collected in containers changed monthly.

The vegetation samples at the end of Table 3c were submitted by Dr. Odén. Measurements are made on the water obtained by the combustion of dried samples.

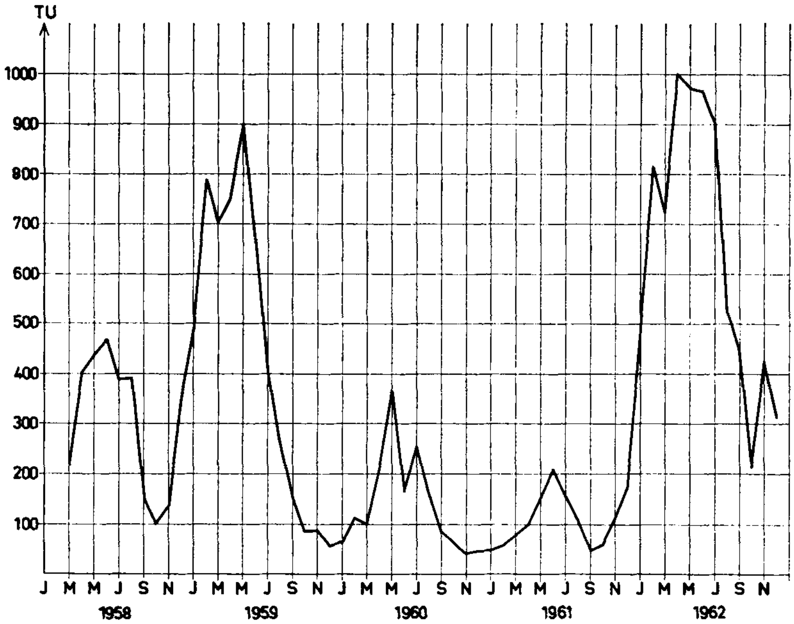

FIG. 2. Tritium contents of monthly samples of precipitation at Huddinge, Sweden.

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