

An account of the major pulses of tritium and their effects in the atmosphere

By ERIK ERIKSSON, *International Meteorological Institute,¹ Stockholm, Sweden*

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ABSTRACT

Bomb tritium produced since 1954 during bomb tests was by and large injected into the stratosphere. The exact amounts are not known but various estimates agree reasonably well. The fate of the bomb produced tritium is discussed in some detail and it appears that ocean surfaces are very effective for withdrawing atmospheric tritium. Available data on rainfall concentrations are also reviewed as well as various models put forth to explain the transport of tritium in nature.

1. Introduction

Tritium is one of the isotopes of hydrogen which is normally produced in the atmosphere primarily by neutrons generated in cosmic radiation reacting with especially nitrogen in the atmosphere (LAL & PETERS, 1962). Carbon-14 is also produced in a similar fashion. When nuclear weapons testing started considerable amounts of tritium were added to the atmosphere allegedly by fusion bombs although it is probable that the neutrons released in fission bomb explosions also contribute.

Nuclear testing, which started on a larger scale in 1952, has not been continuous but has occurred in periods mostly rather well separated. The tritium produced in these experiments has thus been added to the atmosphere in pulses, especially to the stratosphere. Because of this and the fact that most of the bomb produced tritium is found in water as HTO it can be expected to be of great help in the study of the circulation of water in nature. Isolated studies have also shown that bomb tritium can be used for that purpose although it is also evident that auxiliary data are needed for proper interpretation.

To promote the use of tritium for the study of the circulation of water in nature the International Atomic Energy Agency and the World Meteorological Organization jointly organized a global network of stations for monthly collection of precipitation, which started operating in late 1960. At the same time a few laboratories took on the analytical part (which is rather

expensive and time consuming). This part of the programme has not been as successful as the sampling and consequently there are up to now relatively scanty data available. There are a few stations, however, where sampling has been going on for several years and although they are very few, they provide some information on the period from 1954 onwards. For the present purpose these series are most valuable although they are, as will be seen later, inadequate for some purposes. For the pre-bomb era there are insufficient data to form a satisfactory picture of the distribution of naturally produced tritium.

2. Bomb produced tritium since 1952

MARTELL (1963) has recently published estimates of the cumulative bomb produced tritium inventory using available data on thermonuclear energy yields of the different explosions and a tritium yield of 0.7 kg tritium per megaton (MT) of fusion energy released. In this way any contribution by neutrons released during bomb explosions is neglected. However, because carbon-14 is produced by these neutrons one would also expect a simultaneous production of tritium and the rate of production can be assumed to be related to carbon-14 production as in cosmic produced carbon-14 and tritium. According to the computations of LAL & PETERS (1962) the average cosmic ray production rate

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TABLE 1. Bomb produced tritium inventory.

Year	Prod. in kg	Inventory in kg	Stratospheric inventory in kg ^a	Time and location of major production
1952	0.5			4th quarter, low latitude
		0.5		
1953	0			
		0.4		
1954	13			2nd quarter, low latitude
		12.9	12	
1955	3		13	4th quarter, high latitude
		15.2	12	
1956	6		9	2nd quarter, low latitude
		20.2	11.5	
1957	8		14	2nd quarter, high latitude
		26.9	12	
1958	21		17	2nd quarter, low latitude, 4th quarter, high latitude
		46.0		
1959	0			
		43.5		
1960	0			
		41.0		
1961	56			3rd quarter high latitude
		95		
1962	89			
		176		2nd quarter, low latitude, 4th quarter, high latitude

^a Based on computations of HAGEMANN *et al.* (1959) and a ratio of tritium to carbon-14 in the stratosphere increasing linearly from 0.3 on 1st January 1956 to 0.4 on 1st January 1958. The values on the years refer to 1st July and between the years to 1st January.

of carbon-14 is 1.8 atoms per cm³ and second and that of tritium 0.25 atoms per cm³ and second. The production rate of tritium may therefore be about 15 % of the production rate of carbon-14.

MACHTA *et al.* (1963) estimated the carbon-14 production to $2 \cdot 10^{16}$ atoms/MT total yield for explosions in the air and half as much for explosions at ground level, where half of the released neutrons may disappear into the ground or water. With this figure one would expect a tritium production of $0.3 \cdot 10^{16}$ atoms/MT total yield in addition to that released in fusion bombs. HAGEMANN *et al.* (1959) also observed that there is a correlation between tritium and carbon-14 in the stratosphere and on the basis of their data they find on an average 0.4 tritium atoms per carbon-14 atom.

If the figure $0.3 \cdot 10^{16}$ tritium atoms/MT total yield is used, with due consideration of surface explosions, it is necessary to decrease the fusion production of tritium to get the proper ratio of tritium to carbon-14 produced. In fact, with

available data on bomb types, yields (TOMPKINS, 1963) and time of explosions (MARTELL, 1963) where the fusion yield is taken to be 0.4 kg/MT, we get the inventory shown in Table 1. The production is on an average about 10 % lower than computed by Martell. In 1957–1958 the ratio of total bomb produced tritium to total bomb produced carbon-14 becomes 0.4. In the table the stratospheric inventory of tritium has also been listed, based on computed inventories of carbon-14 (HAGEMANN *et al.*, 1959) and ratios of tritium to carbon-14 increasing linearly during the period, from 0.3 on January 1, 1956, to 0.4 on January 1, 1958, and 0.4 also on July 1, 1958. This gradual increase in the ratio is clearly indicated in Fig. 5 in HAGEMANN's *et al.* paper (1959) and may be due to preferential losses of tritium in the earlier experiments especially in those conducted in ocean areas at surface level.

The ratio of the 1954 production to the 1952 production has been taken as 2:1 by MARTELL

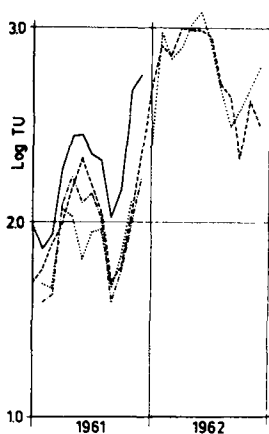


FIG. 1. Tritium concentrations in precipitation at four continental stations during 1961 and 1962. Ordinate in log TU, TU in 10^{18} T/H. —, Ottawa (Canada); — — —, Huddinge (Stockholm, Sweden); — · — · —, Vienna (Austria); · · · · ·, Stuttgart (Germany).

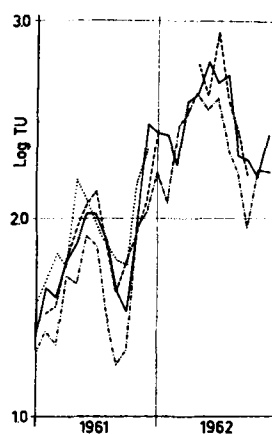


FIG. 2. Tritium concentrations in precipitation at two coastal stations (Lista and Vancouver) and two island stations (Reykjavik and Valentia). —, Reykjavik (Iceland); — — —, Lista (Norway); — · — · —, Valentia (Ireland); · · · · ·, Vancouver (Canada).

(1963). This, however, leads to great inconsistencies in the stratospheric inventory of tritium, hence in Table 1 the major part has been carried over to 1954. The total inventory is still too low compared to the stratospheric inventory and the same is true for carbon-14, examining the data by HAGEMANN *et al.* (1959). This may be due to an underestimate of the total yield of the 1954 explosions. The production of tritium in 1954 might as well have been twice as high as listed considering the rather high concentrations in precipitation observed this year. BEGEMANN & LIBBY (1956) estimated the global fall-out in 1954 from these test series to more than 20 kg tritium, an estimate which, however, may be too high, perhaps by a factor of two.

The production figures in Table 1 are of course rather tentative. The same can perhaps also be said about the stratosphere inventory which, however, is founded on admittedly few, but nevertheless experimental data.

3. Tritium in precipitation

Tritium shows a similar seasonal variation as Strontium-90 and hence it can be concluded that the same mechanism releases tritium and Strontium-90 from the stratosphere into the troposphere. Some typical patterns for northern hemisphere stations are shown in Figs. 1 and 2

for 1961 and 1962, where the logarithm of the concentration is plotted for each month. It is interesting to note that the curves in Figs. 1 and 2 run more or less parallel, especially for continental stations. This suggests that the precipitation pattern of tritium is more or less fixed, the concentration at any time and point being determined by the rate at which tritium enters from the stratosphere. There are, of course, deviations from this rule due to fluctuations which one always finds superimposed on the mean motion of the atmosphere. The scale of these is of such a magnitude that they are also noticeable in yearly averages. Hence, it is rather difficult to explain any global phenomena on monthly averages when the number of observation points is limited as in the present case. The variation on a percentage basis can be inferred from Fig. 3, which gives average relative values and the standard deviation for 8 months' data from Scandinavia (ODÉN, 1964, in press). There seems to be an inverse relation between the standard variation and the monthly rainfall as noted by Odén. This is probably due to more frequent rains where the rainfall is higher, i.e. the air masses are so to speak sampled more frequently.

There seems to be no correlation between concentration of tritium and monthly precipitation, hence when averaging tritium concentrations it does not seem necessary to weigh

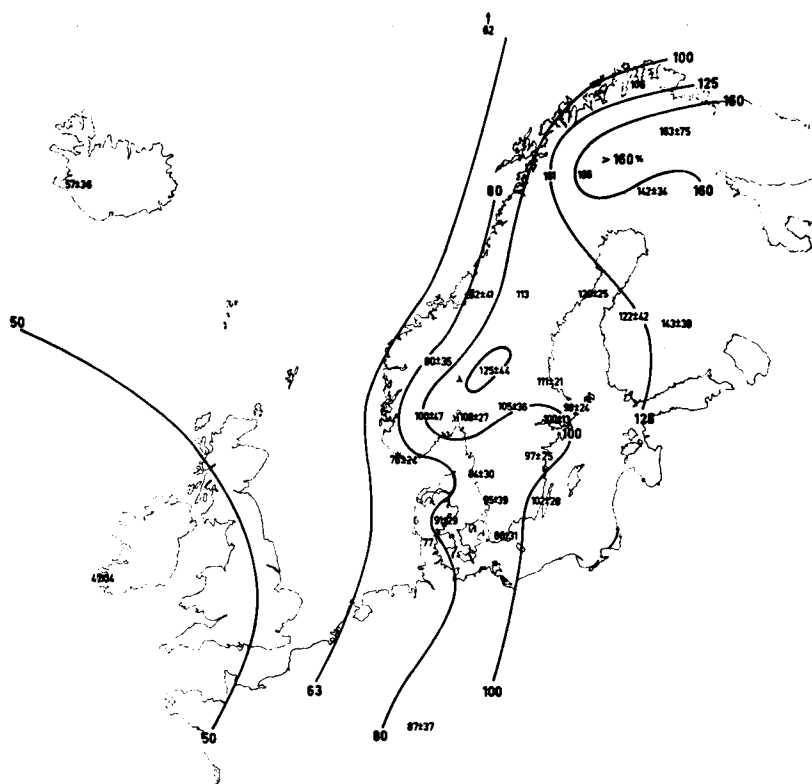


FIG. 3. The average pattern of tritium concentrations in precipitation in north-western Europe. Averages for eight months, Huddinge (Stockholm) set arbitrarily to 100. Standard deviations in the same units. (From ODÉN, 1964.)

the concentration according to rainfall unless one is interested in the deposition at a certain location.

Yearly averages for some stations with observations covering the major part of the year are assembled in Table 2, the data being taken from the lists of data distributed by the International Atomic Energy Agency (1962-63). Some selection of the data has been made in so far that as a rule only data from those laboratories which participated in the inter-comparison of standards and submitted their results have been considered (cf. IAEA, list No. 3). The 1961 data were investigated carefully before the yearly averages were worked out in order to remove any influence of the test series which started in September, hence they should reflect as closely as possible the downward trend in the average concentrations from 1959 onwards. A few of the northern hemisphere stations show the influence of these test series

already in October 1961 but most of them not until November 1961.

Looking at Table 2 the meridional variation is clearly indicated. However, to get a better picture of this variation the few data available for the years 1959 to 1962 are shown in Figs. 4, 5, 6 and 7 as functions of the latitude. The nature of the stations are also indicated, island stations being represented by open circles, coastal stations on large continental masses by half-filled circles and continental stations by filled circles. Island stations show the lowest concentrations while continental stations show the highest, but the latitudinal variation is very similar even if the levels are different. The equatorial stations are unfortunately few.

A lower limit seems to be represented by the island stations and has been tentatively suggested in the figures. This can be taken to represent the average concentration in precipitation over the sea. An upper limit can also be

TABLE 2. Yearly unweighted averages of tritium in precipitation at some stations (in TU, i.e. 10^{18} T/H).

Station and country	Geographical position		Concentration in TU									
	Lat.	Long.	1953	1954	1955	1956	1957	1958	1959	1960	1961	1962
Isfjord, Spitsbergen	78° N	14° E										570
Resolute Bay, Canada	75° N	95° W					182	460				
Reykjavik, Iceland	69° N	22° W								63	55	340
Bethel, Alaska, USA	61° N	162° W										590
Huddinge, Sweden	59° N	18° E						290	476	140	89	650
Lista, Norway	58° N	7° E									59	380
Edmonton, Canada	54° N	114° W										1270
Valentia, Ireland	51° N	14° W						147	128	55	34	250
Stuttgart, Germany	49° N	9° E									75	670
Vancouver, Canada	49° N	123° W		77	—	76	71	254	242	73	64	
Vienna, Austria	48° N	17° E										97
Ottawa, Canada	45° N	76° W	27	444	43	148	122	515	542	161	143	
Hong Kong	22° N	114° E										24
Hilo, Hawaii	20° N	155° W										55
Manila, Philippines	15° N	121° E									20	
Barbados	16° N	60° W							13			
Entebbe, Uganda	0° N	32° E									10	
Hollandia, New Guinea	3° S	41° E					9	21	20	8		
Malan, Union of S. Africa	44° S	18° E									11	11
Pretoria, Union of S. Africa	26° S	28° E						23	21			30
Melbourne, Australia	38° S	145° E								16		

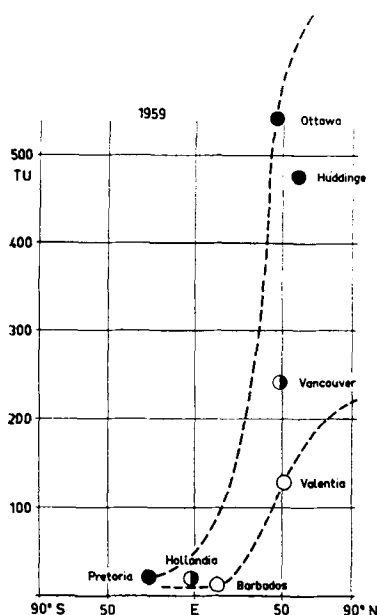


FIG. 4. The meridional distribution of yearly average tritium concentrations in precipitation for 1959. Open circles, island stations; half-filled circles, coastal stations; filled circles, continental stations.

drawn in the figures though the position of this is much more uncertain. This could at best represent the average continental concentration in precipitation. Thus, for the years 1959 to 1962 it is possible to make some estimates of the amount of tritium precipitated, at least over the oceans, although these estimates must

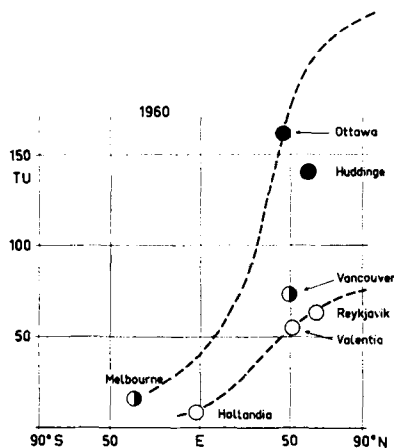


FIG. 5. Same as Fig. 4 for 1960.

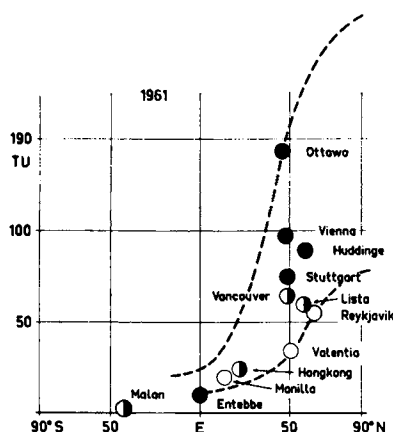


FIG. 6. Same as Fig. 4 for 1961.

be extremely tentative. What is further needed is the meridional distribution of precipitation over the sea. However, before this is done, it is perhaps better to discuss the difference in conditions of deposition of tritium over land and over the sea.

4. Transport of moisture and tritium in the atmosphere

The concentration of tritium in precipitation depends not only on the rate of addition from the stratosphere but also on the precipitable water in the atmosphere, as LIBBY (1963) recently pointed out, as well as on other meteorological factors. For a full understanding of the complexity of this problem it is worth while to consider the concentration of tritium in precipitation in terms of meteorological parameters, especially the flux of water vapor in the atmosphere, as ERIKSSON & ODÉN (1963) have done recently, and ERIKSSON (1964) has done in the case of stable isotopes in the atmosphere.

The water vapor content in the atmosphere is mostly expressed by the specific humidity, q , which is grams water vapor per kg of air. With this definition of q one defines the precipitable water, w , by

$$w = \frac{1}{g} \int_0^{P_0} q dp = -\frac{1}{g} \int_{P_0}^0 q dp,$$

where g is the acceleration of gravity and p is the pressure. The integration can for practical

purposes be extended to about 500 mb instead of zero, since the water vapor density above is rather unimportant for the precipitable water. Thus the lower part of the troposphere contains practically all the moisture and is often called the moist layer. This is a useful distinction in the present context as addition of tritium can be regarded as taking place on the top of the moist layer, thus not at the tropopause level. If the stratosphere addition takes place in the jetstream region, which has often been claimed, it has time to spread over a much wider region before it is added to the moist layer.

The transport or flux of water vapor in the atmosphere can be written

$$\mathbf{f} = q\mathbf{v}, \quad (1)$$

where \mathbf{v} is the wind, \mathbf{f} and \mathbf{v} being vectors. If the tritium is expressed in the same way as in precipitation, i.e. as concentration c , the transport of tritium can be written

$$\mathbf{f}^* = qc\mathbf{v}. \quad (2)$$

The fluxes can be integrated vertically and if the wind does not change appreciably within the moist layer we get

$$\mathbf{F} = w\mathbf{v}_a \quad (3)$$

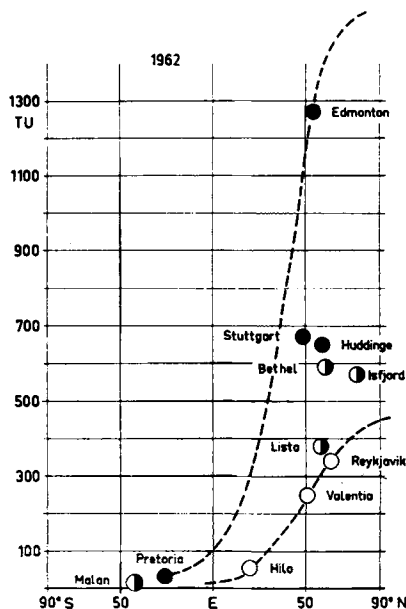


FIG. 7. Same as Fig. 4 for 1962.

$$\text{and } \mathbf{F}^* = c_a w \mathbf{v}_a, \quad (4)$$

where c_a is now the vertical average of the tritium concentration, c , weighted with respect to the specific humidity, q . At least in temperate latitudes this seems to hold with the 850 mb wind representing \mathbf{v}_a , but there are areas where this simple formulation cannot be used. As an example, Central Africa can be mentioned, where at low levels water vapor is transformed from west to east but in the upper part of the moist layer the moisture transport is directed from east to west. This peculiar water flux pattern is noticeable as far east as Lake Victoria (PEIXOTO, 1958).

With the restriction of uniform winds throughout the moist layer we can proceed. The vertical distribution of water vapor is normally rather irregular, with horizontal strata of varying specific humidity. The same can be expected for tritium as well as for other constituents in the atmosphere. The layering changes, of course, irregularly, and this is perhaps the main reason why it is so difficult to interpret instantaneous data, like tritium in showers. On the other hand, this irregularity is a consequence of the fact that the vertical transport of water vapor takes place through eddy transfer on a fairly large scale, the corresponding diffusion coefficient being of the order $10^8 \text{ cm}^2/\text{sec}$.

Thus, tritium data on precipitation have to be averaged over some time to bring out features which are expressible in terms of average meteorological parameters. This is true in general for all properties in the atmosphere.

Averaging the horizontal fluxes can be done using the usual connotation with bars for averages and primes for deviations from averages. Thus for water vapor we write, dropping the index "a" of the wind vector

$$\bar{\mathbf{F}} = \overline{w\mathbf{v}} \equiv \bar{w}\bar{\mathbf{v}} + \overline{w'\mathbf{v}'}, \quad (5)$$

where $\overline{w'\mathbf{v}'}$ is termed the eddy flux. From this it is seen that the flux of water vapor is not necessarily parallel to the average wind. The direction of $\overline{w'\mathbf{v}'}$ is often assumed to be against the direction of the gradient of precipitable water and this seems to be a reasonable assumption at least as an approximation. In fact the eddy flux is often expressed as $-K\nabla\bar{w}$ where K is the horizontal eddy diffusion coefficient

and has a magnitude of $10^{10} \text{ cm}^2/\text{sec}$ when averaging over a month. Hence we can write

$$\bar{\mathbf{F}} = \bar{w}\bar{\mathbf{v}} - K\nabla\bar{w}, \quad (6)$$

where ∇ refers to a two-dimensional co-ordinate system.

For tritium we can define a quantity $\bar{w}^* = \overline{c_a w}$ and call it precipitable tritium. Consequently, we get for the flux of tritium

$$\bar{\mathbf{F}}^* = \bar{w}^*\bar{\mathbf{v}} - K\nabla\bar{w}^*. \quad (7)$$

Comparing the two expressions for the fluxes, i.e. that of water vapor with that of tritium we find that with our restriction of a uniform wind in the moist layer the advective parts of the fluxes have the same directions. If the wind should change direction with altitude they would not necessarily have the same directions unless the moist layer is so well mixed that the vertical distributions of water vapor and tritium are identical.

The directions of the eddy fluxes depend on the directions of the gradients of precipitable tritium and water. Since these are determined mainly by the distribution of sources and sinks for water vapor and for tritium, it is obvious that it is almost fortuitous if they should happen to be parallel. In fact, in coastal regions over continents they may even have opposite directions. This fact, which can be readily demonstrated from the data given by ERIKSSON & ODÉN (1963), makes it most difficult to relate the two fluxes in a simple fashion.

We can thus conclude that the flux of water vapor and that of tritium in the atmosphere are not necessarily parallel. This is a significant observation in view of the computations of CRAIG & LAL (1961). They tried to establish the tritium balance for North America by using the water vapor flux data computed from meteorological data by BENTON & ESTOQUE (1954) multiplying these with pre-bomb concentrations of tritium in rain. Their assumption is thus that $\bar{\mathbf{F}}^* = \bar{c}_a \bar{\mathbf{F}}$ which is unlikely to hold and can lead to great errors especially in regions where the main transport of water takes place through eddy diffusion.

The divergence of the fluxes assuming stationary conditions has to be balanced by additions or subtractions of the amounts present in the moist layer. Thus for water vapor we have simply

$$\nabla \cdot \bar{\mathbf{F}} = -(\bar{P} - \bar{E}), \quad (8)$$

but for tritium we get a more complicated expression

$$\nabla \cdot \bar{\mathbf{F}}^* = \bar{Q} - \bar{c}_p \bar{P} - \overline{(c_a - c_w) M_w}, \quad (9)$$

where \bar{Q} is the rate of addition to tritium to the top of the moist layer, $\bar{c}_p \bar{P}$ is the rate of precipitation of tritium, and $\overline{(c_a - c_w) M_w}$ is transfer to the earth's surface due to water vapor exchange between the air and the surface. In this expression the radioactive decay of tritium in the moist layer has been neglected since the turnover time of water in the moist layer is a matter of only a few weeks.

Over land precipitation is mainly stored in the soils before part of it sinks and adds to the ground water and the rest evaporates. The soil as a reservoir for water is rather small, a figure of about 10 cm storage can be taken as a rough guide. Further, because it is a capillary system with but small possibilities of mixing, no effective fractionation can take place. Hence, the tritium concentration in evaporated water will be the same as that in precipitation. Therefore, one can assume that \hat{c}_w and \hat{c}_p are equal, \hat{c}_w and \hat{c}_p being weighted averages of concentrations of tritium in the soil or plant water and in precipitation. There may be seasonal unbalances in \hat{c}_w and \hat{c}_p but they should, on the whole, have but small effects. As no effective eddy exchange of tritium between air and surface can take place the last term reduces to $-\hat{c}_p \bar{E}$. Hence the divergence over land can be written

$$\nabla \cdot \bar{\mathbf{F}}^* = \bar{Q} - \hat{c}_p (\bar{P} - \bar{E}). \quad (10)$$

Over the sea the concentration of tritium in the sea surface water is rather low compared to the concentration in precipitation. Hence the last term will depend on the rate of exchange of water vapor between air and sea water. The mass exchange rate over the sea can be expressed as a transport velocity which tells at what rate an air parcel from a given level approaches the surface and comes into contact with it. An air parcel at the surface will of course reach the same level at the same average velocity. It is thus more properly an eddy diffusion velocity and has been estimated by ERIKSSON (1961) to about 1 cm/sec over the ocean areas close to the sea surface, the estimate

being based on rate of evaporation and relative humidity of the air. In fact the total water vapor exchange over the oceans in the lowest part of the moist layer is about three to four times as large as the evaporation. Between average cloud base level and the sea surface the exchange rate is somewhat less. Assuming that the concentration of tritium in atmospheric moisture at cloud level is that found in precipitation, the mass exchange transfer of tritium may be about twice as big as that transferred by precipitation. Approximately one can write over the sea

$$\nabla \cdot \bar{\mathbf{F}}^* = \bar{Q} - \alpha \hat{c}_p \bar{P}, \quad (11)$$

where α has a value of about 3 in temperate latitudes. In low latitudes because of low tritium concentrations in precipitation, addition of tritium by evaporation of sea water can no longer be neglected. To account for this a somewhat lower value for α could be chosen. BOLIN (1958) has also stressed the importance of the eddy transfer of tritium to the oceans using similar arguments as presented here.

LIBBY (1963) has recently set up a simplified model for the transport and balance of tritium in the atmosphere, a model which can be derived from the equations here by averaging meridionally. In this way the eddy diffusion terms in the meridional direction disappear. Neglecting the longitudinal eddy diffusion terms will give in principle the equations derived by Libby, with some important exceptions due to different assumptions regarding removal and additions over land and over the sea. Firstly, Libby assumes the presence of a "surface groundwater" reservoir which seems to correspond to the soil storage referred to here. He also uses a volume of 50 cm for this reservoir which seems to be too high. Because of this large size he finds that it will cause secondary maxima in the tritium concentrations of precipitation in summer. This is doubtful since these maxima normally occur in spring whereas the maximum evaporation takes place in the summer. Hence, there is no available support for his assumption about the size of the land reservoir (soil storage).

Libby's assumptions on the rate of removal over the sea are still more questionable because he neglects completely the possibility of eddy transfer to the ocean surface of tritium in the atmosphere. The result of this neglect is

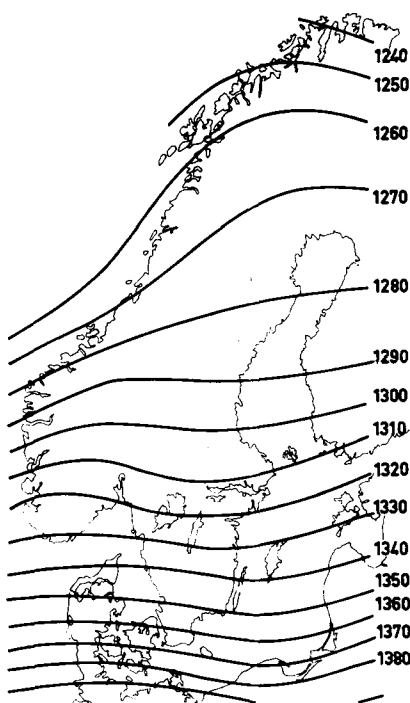


FIG. 8. The average height in meters of the 850 mb level during January, 1962, over Scandinavia. From ERIKSSON & ODÉN (1963).

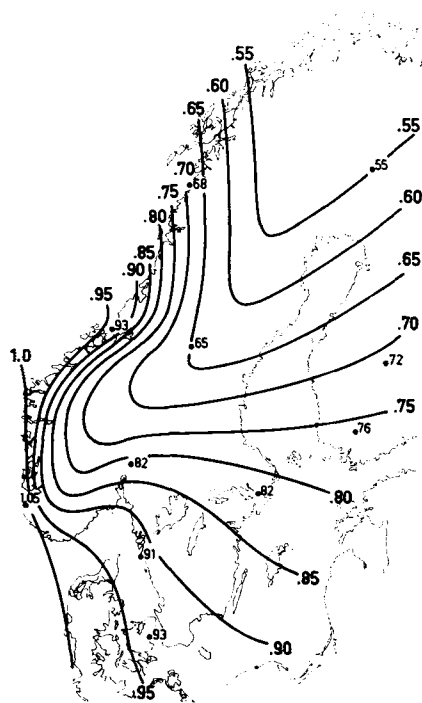


FIG. 9. The average precipitable water in g/cm^2 during January, 1962, over Scandinavia. From ERIKSSON & ODÉN (1963).

almost catastrophic. He assumes that the tritium balance over the oceans is such that the longitudinal gradient of precipitable tritium is zero on the west coasts in temperate latitudes, which seems reasonable. Hence, \bar{Q} must be balanced completely by the removal of tritium by precipitation (i.e. $\nabla \cdot \mathbf{F}^* = 0$). He then arrives at a precipitation rate of 2.5 m/year over the oceans, a rate which cannot be accepted since thousands of various measurements indicate a much lower figure, about 0.9 m/year (WÜST, 1954). The discrepancy is easily removed if the total rate of removal of tritium is set equal to three times the rate of removal by precipitation which means that eddy transfer accounts for 2/3 of the total removal. Libby thinks the spray production is responsible for the extra transfer and cites a figure of 1.7 m/year of spray production to cover the difference. Also this figure is far too high. If the total salt production over the sea is about 1000 million tons (ERIKSSON, 1959) this corresponds to 30 km^3 of sea water, which is extremely small compared to the total precipitation over the sea which is estimated

to more than 300 000 km^3/year (WÜST, 1954). There are other data which support the indication that the removal mechanism for tritium over oceans is much more effective than over land. Figs. 8, 9 and 10 show the average height of the 850 mb level as indicative of the mean flow, the average precipitable water, and the tritium concentration in precipitation respectively, for January 1962 over Scandinavia (from ERIKSSON & ODÉN, 1963). The 850 mb level was chosen as representative of the moist layer and from the height contours of this level the average winds can be computed, using the usual geostrophic approximation of the equations of motion, a procedure normally used for computing winds. It is seen that the winds were westerly over most of the area and rather strong in the southern part of the region. Using approximations of the balance equations given earlier they estimated \bar{Q} to be about 8000 TU cm per month in the southern part of Sweden. As the same value is likely also for the coastal stations (including a station on Iceland), they concluded that the total rate of removal

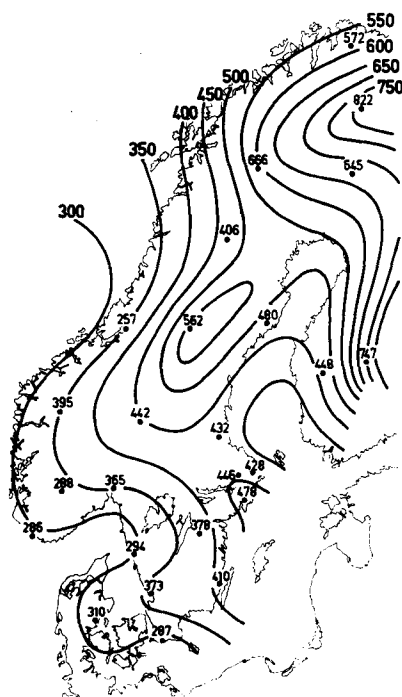


FIG. 10. The tritium concentration in precipitation during January, 1962, over Scandinavia. Concentration in TU (10^{18} T/H). From ERIKSSON & ODÉN (1963).

at these stations representing oceanic conditions must have been about three times that in precipitation.

5. Deposition of tritium over land and over the sea

From Figs. 4, 5, 6, and 7 it is possible to estimate at least the amount of tritium precipitated on the northern hemisphere oceans, using Wüstr's (1954) data on the total oceanic precipitation on different latitude belts. The average tritium concentrations are taken from the lower curves in the figures. The result of these computations is shown in Table 3 being expressed in units of 10^8 TU km², which is equal to $0.67 \cdot 10^{23}$ T atoms, i.e. 0.33 g of tritium. In the table the region 90–80° N has been taken as land because it is covered by ice. It is seen that the uncertainties in concentrations at high latitudes have very little influence on the results because the amount of water precipitated is small. The maximum precipitation of tritium takes place in mid-latitudes.

The total amount of tritium precipitated over oceans in the northern hemisphere is also shown, converted also into kg of tritium. If these are proportional to the stratospheric inventory they indicate a turnover time of 3 to 4 years of stratospheric tritium, which at the beginning of 1959 may have amounted to about 25 kg. The total rate of deposition should thus be somewhat more than 3 times the deposition by precipitation over the northern hemisphere oceans. Deposition over the southern hemisphere may amount to about 15 % of total deposition in this period (LIBBY, 1959), which would make the northern hemisphere deposi-

TABLE 3. *Precipitation of tritium over the N. hemisphere oceans 1959 to 1962.*

Latitude belt	Precipitation 10^8 km ²	Concentration in TU				Precipitated in 10^8 TU km ²			
		1959	1960	1961	1962	1959	1960	1961	1962
90–80	0.0								
80–70	1.3	200	70	67	400	260	90	90	520
70–60	2.8	175	65	56	360	490	180	160	1010
60–50	10.0	140	58	39	280	1400	580	390	2800
50–40	16.0	100	50	28	200	1600	800	450	3200
40–30	16.6	60	40	21	120	1000	660	350	2000
30–20	15.6	30	29	17	75	470	450	270	1170
20–10	27.5	15	20	15	40	410	550	410	1100
10–0	56.5	10	13	13	25	570	730	730	1400
Total 90–0						6200	4040	2850	13200
Total 90–0 in kg ^a						2.1	1.3	0.9	4.4

^a 10^8 TU km² = 0.33 g.

tion about 3 times the amount removed by oceanic precipitation. The major part of this deposition would be over ocean areas. Table 2 indicates that continental precipitation may have a concentration of tritium at most 5 times higher than oceanic precipitation. As the continental runoff is only 10–15 % of the oceanic precipitation (Wüstr, 1954) the removal by precipitation over land can hardly be more than 50–70 % of the removal by precipitation over oceans. Of the total deposition only about 15 % could be deposited over continental areas, the rest over the sea making the total deposition of tritium over the sea about 2.5 times the deposition by precipitation, a figure consistent with the estimate discussed earlier.

6. Stratospheric turnover of tritium

Although the stratosphere is no well mixed reservoir with a well-defined residence time it seems likely that on an average the turnover time, i.e. mass divided by flux, is of the order of 3 to 4 years. Looking at Table 1, comparing the stratospheric inventory figures with the yearly production in the 3 year period 1955–1957, the total production is 17 kg and the stratospheric inventory remained rather constant around 12 kg. This indicates a somewhat shorter turnover time than 3 years, but on the other hand it is likely that the removal rate the first year after an injection is greater than during following years. For Strontium-90 it has been suggested (MINX, 1963) that the stratosphere should be divided into five parts, each with different residence times ranging from half a year to 5 years. Again this suggests an average turnover time of about 3 years.

There are, of course, other possibilities to describe the removal of tritium from the stratosphere. If the physical behavior of the stratosphere is known in enough details with respect to average winds and eddy diffusion, the removal of any debris from the stratosphere could be predicted if the origin of release was known (BOLIN, in press). Another possibility is to follow the deposition in detail and compute empirically the fractions removed during successive years, but this requires accurate estimates of the deposition rates. To make any further conclusions on the basis of tritium in precipitation is hardly possible at present, but the 1961–1962 additions are of such a magnitude that

with the present global network of sampling stations much better deductions can be made. However, it is not likely that they can furnish better information than the Strontium-90 data complemented by a high-altitude sampling programme.

7. Deposition of tritium over oceans and the turnover of sea surface water

BAINBRIDGE (1963) carried out a number of measurements over the North Pacific Ocean during the period 1960 to 1961. If the deposition rate of bomb tritium is known accurately enough from 1954 then it is possible to deduce the turnover time of sea surface water from observations on the increase of tritium in sea surface water. Some assumptions have to be made for instance about the depth of rapid mixing in the oceans, i.e. the thickness of the so-called mixed layer. If this is known the inventory of tritium in the sea surface can be computed from analyses of sea surface water. When the inventory and the deposition rate are known the turnover time of the surface water can be computed. The balance of tritium in the sea surface can be formulated (BAINBRIDGE, 1963):

$$\frac{dM}{dt} = I - (k + \lambda)M \quad (12)$$

where M is the amount in the mixed layer, I is the rate of deposition, λ is the decay coefficient of tritium and $1/k$ is the turnover time of the mixed layer water. Bainbridge estimates I for different years from available data on tritium in precipitation and computes that $1/k$ must be 3.5 years if the depth of the mixed layer is taken to be 75 m and half the precipitated tritium reaches the oceans.

Bainbridge's estimate of the tritium fallout is rather questionable as it gives too great weight to continental stations. The result is also that the fallout from 1958 onwards is greatly over-estimated. Looking at his figures he estimates that during the time period 1954–1961, 75 % of the deposition occurred in the period 1958–1961. From the figures in Table 1 this seems most unreasonable as the production in the period 1958–1961, discounting the 1961 production, which is but feebly reflected in precipitation, is only 40 % of the total produc-

tion in 1954–1961. This means that in the equation above k must be rather large to cope with 70 % of the total addition during the last period. Hence, the 3.5 years Bainbridge computes for the turnover time of the mixed layer is most likely far too small. A revision of his computations can be done as follows. The balance equation can be written in the following finite difference form

$$\left(1 + \frac{k + \lambda}{2}\right) \Delta M_n = I_n - (k + \lambda) M_n, \quad (13)$$

where ΔM_n is the increase in the mixed layer inventory during the n th year, I_n is the addition during the n th year, and M_n is the mixed layer inventory at the beginning of the n th year. When I_n is known successive computations can be done using selected values of $(k + \lambda)$.

As to I_n it seems most reasonable to estimate it from the production rates in Table 1 and the assumption that about 75 % of the production reaches the oceans on the northern hemisphere. Judging from Table 1 and previous results as to the total deposition for the years 1959 to 1961, we can use the following data

Year	Total dep. kg	75% of total kg
1954	10	7.5
1955	3	2.2
1956	5	3.8
1957	5	3.8
1958	10	7.5
1959	7	5.3
1960	4	3.0
1961	3	2.2
	47	35.2

With $k + \lambda = 0.2$ we get a mixed layer inventory at the end of 1961 amounting to 16.3 kg and with $k + \lambda = 0.1$ we get a figure of 23.4 kg. Since 1 TU in the northern hemisphere oceans down to 75 m depth is equivalent to 3.75 kg we find that $k + \lambda = 0.2$ gives a final concentration of 4.3 TU while with $k + \lambda = 0.1$ we get a concentration of 6.2 TU. To this should be added the natural "cosmic" level which may be around 0.5 TU, giving 4.8 and 6.7 TU respectively. The average for the Pacific surface water at the end of 1961 according to Bainbridge's averages is slightly below 7 TU, a figure indicating that the assumption of $k + \lambda = 0.1$ gives a better agreement. However, it must be noted

that if the average depth of the mixed layer is reduced to 50 m the assumption of $k + \lambda = 0.2$ gives a good agreement. Because of the uncertainty in the average depth of the mixing layer of the North Pacific it seems rather difficult to get a good precision for estimating k , but it seems unlikely to be less than 75 m. As $\lambda = 0.055$ it follows that for a 75 m depth of the mixed layer, $k = 0.045$, i.e. $1/k = 22$ years gives the best agreement.

One can also use the short term change in the concentration with time for computing k as Bainbridge demonstrates, which may be a good possibility at present. However, for this purpose a more representative net of sampling stations is needed, because the local variation is apparently rather great.

8. Concluding remarks

Although data at present are not too abundant to demonstrate all possible applications of the pulses of tritium in nature in recent years, those which exist indicate that much information both of meteorological and hydrological nature can be derived from a study of the history of tritium which has been injected into the stratosphere in recent years. Systematic collection of data on various kinds of waters will make it possible in future to gain more insight into the behaviour of the various reservoirs for water in nature.

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