The vertical distribution of tritium in water vapor in the lower troposphere

By WAYNE E. BRADLEY and GLENN E. STOUT, *Illinois State Water Survey, Urbana, Illinois*

(Manuscript received May 25, 1970)

ABSTRACT

A series of 16 aircraft soundings was made during the summers of 1966 and 1967 to measure the vertical distribution of tritium in water vapor from the surface to an altitude of 5 km. Three types of tritium distributions were found. The type I soundings showed a generally increasing tritium concentration with increasing height. Type II soundings were characterized by a constant tritium concentration with increasing altitude. On type III soundings the tritium concentration gradually decreased to approximately 2.4 km and then abruptly increased. These soundings are discussed in terms of the origin of the air masses, evapotranspiration, and the water vapor mixing ratio.

Introduction

Tritium, a heavy isotope of hydrogen, has been relatively abundant in the atmosphere since the advent of thermonuclear testing in 1954. Numerous measurements have been made of its worldwide distribution in precipitation, and in surface and ground water, but fewer measurements of it have been made in atmospheric water vapor. Most of these measurements were associated with the use of tritium as a tracer enabling hydrologists, meteorologists, and oceanographers to follow the movement of water through the hydrologic cycle or some portion thereof (Eriksson, 1958; Bolin, 1958; Giletti et al., 1958). Significant knowledge has been gained despite the complications which arise from fractionation during evaporation and condensation.

Information on the vertical distribution of tritium in the atmosphere is of interest for several reasons. To interpret the areal and temporal variation in the rainout of tritium, it is desirable to know the distribution and concentration of tritium (T) at its immediate source, that is, the atmosphere. Studies concerned with the movement of water or tritium through evapotranspiration, or with the molecular exchange of water vapor at the air–sea interface, also benefit from information on the vertical distribution of T.

A series of tritium soundings was therefore taken in central Illinois during the summers of 1966 and 1967 to measure the distribution of tritium in water vapor in the lower troposphere. Studies were conducted of the vertical distribution of tritium, its variation with time, and the meteorological factors influencing the distribution. The tritium soundings were compared with their associated temperature and water vapor mixing ratio soundings from the U.S. Weather Bureau in Peoria, Illinois, 120 km northwest of the soundings, and with the origin of the air masses sampled.

Sampling procedure

The tritium sampling altitudes were chosen prior to each sounding by examining the U.S. Weather Bureau rawinsonde data from Peoria, Illinois, and were selected to avoid measurements across temperature inversions or moisture discontinuities whenever possible. Calculations of the mixing ratio were made to determine the sampling time necessary to collect 25 ml of water, but, occasionally, extremely dry air aloft necessitated the collection of a smaller sample.

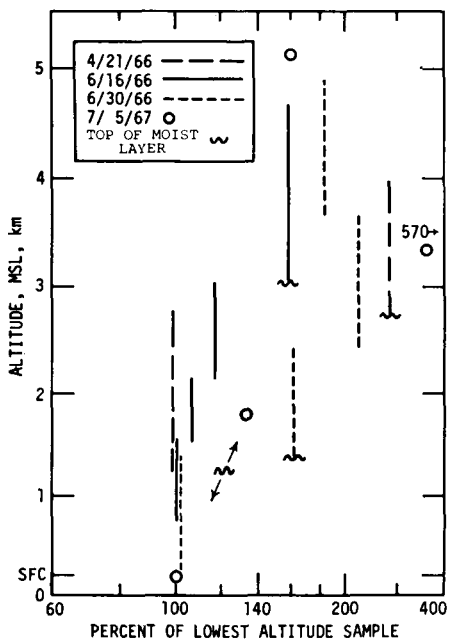

Fig. 1. Type I normalized tritium soundings over central Illinois.

During 1966 the samples were collected while the aircraft ascended through the sampling layer at a constant rate, but in 1967 the vertical sampling consisted of a series of samples, each taken at a different constant altitude to facilitate simultaneous radon measurements. The sampling flights were begun at approximately 1000 CST and the sampling layers were determined with the Peoria radiosonde data taken at 0600 CST.

Details of the tritium sampling system have been given by Bradley and Feteris (1966), but a brief description follows. The water vapor samples were collected for tritium analysis with an adsorption system carried aloft in a Twin-Beech aircraft. The vapor was adsorbed in traps containing 400–500 grams of Lindly molecular sieve No. 4 AXW. Air was sampled through a forward facing nozzle and drawn through the sieve traps with a 1 hp positive displacement blower at a sampling rate of approximately 0.5 m^3 min^{-1}.

After the sampling process, the water was removed from the traps by subjecting them to a temperature of 560°C and vacuum of 10^{-4} mm mercury. The released water vapor was collected in a cold trap at −72°C and later analyzed for tritium. Although the bulk of the water was removed from the traps in the first 2 or 3 hours of oven warmup, the traps were baked an additional 7 to 20 hours at maximum temperature to minimize the volume of water remaining in the trap.

Most of the samples were analyzed without enrichment. The accuracy of the 1966 data is ±20 to 35 TU (1 TU = 1 tritium atom/10^{18} hydrogen atoms). The 1967 data have an accuracy of ±10 TU for TU < 300 and ±3% for TU > 300. A small additional error results from ~0.3 ml water which remains in the trap after baking, and becomes mixed with the next sample. A 25 ml sample was usually collected to minimize the effect of the memory overlap.

Types of tritium soundings

The concentration of tritium in the lower troposphere varies considerably according to the season and the sampling altitude, as can be seen by examining the 16 tritium soundings listed in Table I. It is therefore useful in comparing the soundings with each other to normalize the tritium concentrations. This normalization was accomplished by dividing each observation of a sounding by the concentration at the lowest altitude. In this manner, soundings of differing concentrations were compared with respect to their relative variation with altitude, and three basic types of tritium soundings were then observed.

The tritium concentration of the type I soundings showed a general increasing trend from the surface to the top of the soundings. The type II soundings were characterized by a constant T with increasing altitude. On the type III soundings, T initially decreased in the first couple kilometers and then abruptly increased.

The primary factors expected to influence the concentration of tritium at a given elevation are (1) the stratospheric concentration and degree of stratospheric–tropospheric exchange, (2) the origin of the air mass with respect to the stratospheric and continental sources and the oceanic sink, (3) the degree of vertical exchange, (4) the water vapor mixing ratio (MR) of the environmental air, and (5) the amount of evapotranspiration. Since most of these factors are controlled by atmospheric circulations, the T soundings may be best explained by con-

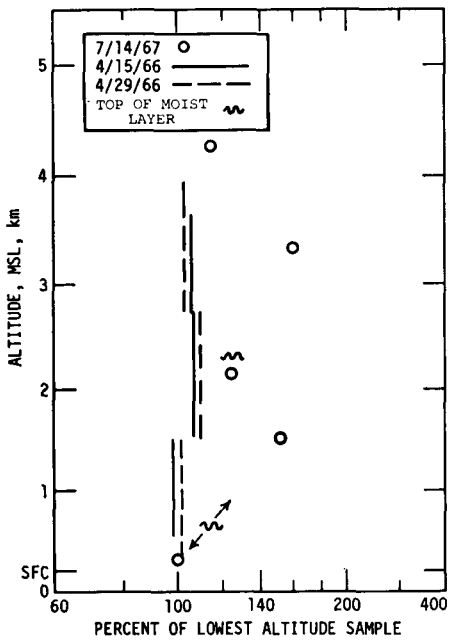

Fig. 2. Type II normalized tritium soundings over central Illinois.

sidering the associated weather along with the tritium sources and sinks.

Type I soundings shown in Fig. 1 are characterized by a general increase in T with increasing altitude, although two of the four decrease above 3 km. The length of the line reflects the thickness of the layer through which the sample was taken. The greatest change in T with altitude is concurrent with an abrupt decrease in the water vapor mixing ratio (see Fig. 1) which, in three out of four cases, resulted from a subsidence inversion. The four soundings were taken in continental polar (cP) air masses in which air trajectories at all sampling levels were either westerly or northerly.

The increase in T associated with the drying aloft occurs because air at those elevations contains a relatively high percentage of tritiated water vapor from the stratosphere. In contrast, the lower level air is originally filled with Pacific Ocean moisture having a tritium concentration of 15–20 TU (Houtermans, 1965). When the downward flux of tritiated water vapor mixes into this moist layer it becomes relatively diluted, resulting in a low tritium concentration. The most abrupt change in the T gradient thus occurs at the interface between the dry and moist layers.

On the type II soundings shown in Fig. 2 the tritium concentration is relatively constant with increasing altitude. As with the type I soundings, the measurements were taken in cP air masses with air trajectories out of the west or north. During the 15 April 1966 flight the aircraft was completely surrounded by towering cumulus indicating good vertical mixing resulting in a constant T with height. On the 29 April 1966 sounding, T was constant but only because of the moist layer associated with the highest altitude sample resulting in dilution and a relatively low T. For the sounding of 14 July 1967 an explanation of the relatively constant T is not apparent in the moisture or temperature data.

The type III soundings shown in Fig. 3 are characterized by T gradually decreasing with altitude from the surface to the 2 400 to 3 300 m MSL level. The decrease is relatively small, varying from 12–24% of the surface value, and is followed by an abrupt increase in T in the order of 100%. The sounding of 26 July 1966 included in this group does not have measurements above 2 400 m MSL, but does have the

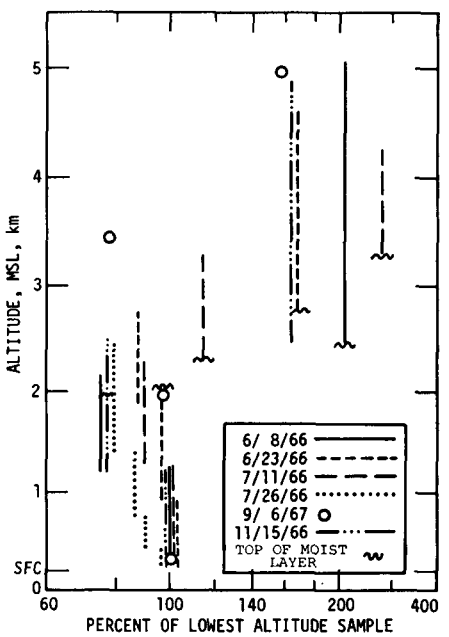

Fig. 3. Type III normalized tritium soundings over central Illinois.

Table 1. *Tritium soundings over east central Illinois and other locations*[a]

Date	Sampling altitude (m) MSL	T (TU)
4-15-66	610–1 520	812
	1 520–2 740	866
	2 740–3 660	862
4-21-66	1 220–2 740	864
	2 740–3 960	2 073
4-29-66	300–1 520	911
	1 520–2 740	1 000
	2 740–3 960	931
6-8-66	300–1 220	450
	1 220–2 130	342
	2 440–5 030	938
6-16-66	700–1 520	888
	1 520–2 130	945
	2 130–3 050	1 033
	3 050–4 660	1 405
6-23-66	230– 910	744
	910–1 830	726
	1 830–2 740	656
	2 740–4 570	1 270
6-30-66	300–1 220	476
	1 220–2 440	766
	2 440–3 660	1 007
	3 660–4 880	869
7-11-66	230–1 310	203
	1 310–2 290	186
	2 290–3 260	232
	3 260–4 270	480
7-26-66	*Control Sounding*	
	190– 400	496
	460– 760	445
	760–1 370	342
	1 370–2 440	396
7-26-66	*Windward sounding*	
	240– 400	399
	460– 760	414
	760–1 370	406
	1 370–2 440	352
7-26-66	*Leeward sounding*	
	240– 400	411
	400– 760	418
	760–1 370	358
	1 370–2 440	388
11-15-66	230–1 220	287
	1 220–2 440	225
	2 440–4 880	474
7-5-67	230	563
	1 750	752
	3 280	2 980
	5 110	896
7-14-67	380	710
	1 490	989
	2 240	808
	3 350	1 050
	4 280	754
7-20-67	*Gulf of Mexico*	
	250	37
	1 050	49
	2 100	58
	2 880	81
	3 810	200
9-6-67	400	411
	1 910	400
	3 430	320
	4 950	643

[a] Locations for soundings not taken in east central Illinois have been underscored.

decreasing T, or tritium inversion, in the lower layers.

The abrupt increase in T in the type III sounding, as in the type I sounding, was associated with the top of the moist layer in four out of five of the soundings, as shown in Fig. 3. The interesting difference between the type III and type I soundings, however, is the decrease in T in the lower layers on the type III soundings. This tritium inversion is related to air trajectories in the lower moist layer as follows.

Discussion of type III soundings. Four type III soundings were taken during June and July of 1966 either in maritime tropical air masses or in a warm frontal zone. During each of these soundings the isobaric air trajectories at the 850 mb level, and some times at the 700 mb level were from the Gulf of Mexico. The question arises as to why the air masses from the south exhibited the T inversion, whereas those from the Pacific coast or Canada usually did not.

Oceans are effective sinks (Bolin, 1958) for tritium so the concentration in oceanic air is several times lower than it is in the Midwest. Östlund (1968) reported the average T concentration of sea water in the Bahamian area as 13.5 TU, and as mentioned previously, the Pacific ocean measurements were from 15 to 20 TU. The low concentration of T in oceanic air is exhibited by the sounding taken at the Gulf coast on 20 July 1967 (Table I).

A trajectory analysis determined that the average over land travel times for the isobaric trajectories at the 500 mb (5.6 km MSL) and

700 mb (3.0 km MSL) levels were similar for Gulf, Pacific, and northern trajectories. In addition, the July evapotranspiration rate for the regions within 1 100 km south, west, and north of the sounding area are approximately the same (Visher, 1954). Two differences, however, were found to exist between the Gulf trajectories and the westerly or northerly trajectories and are as follows: (1) the Gulf trajectory travel times at the 850 mb (1.5 km MSL) level average one-half or less the overland travel time of the other trajectories, and (2) the Gulf air in moving northward crosses into regions of ever increasing T in rainfall because of latitudinal variation in the fallout of tritium (Thatcher, 1962). The higher T concentrations in the more northerly rain produces higher T's in the evapotranspirated water.

A possible explanation of the T inversions is as follows. As an air mass approaches the U.S. from the west coast or Canada, it enters the United Stated at a latitude where the stratospheric fallout is relatively high. The air in the lower levels (850 mb) has a long and circuitous route, perhaps over the mountains and across the plains before reaching Illinois. Therefore, the tritium input from evapotranspiration has ample time to become well mixed and a T inversion does not exist, at least by the time the air reaches the Midwest. In contrast, air trajectories from the Gulf at the 850 mb level moves rapidly into the Midwest and up the latitudinal tritium gradient. The rate of increase of T in evapotranspiration is rapid enough that the inversion is not destroyed by normal vertical mixing.

However, there were two type III soundings, those on 6 September 1967 and 15 November 1966, that were not in the Gulf air. Air trajectories on these days were out of the north and west, respectively. The decrease in tritium in the 1 220–2 440 m layer on 15 November was apparently the result of an intense moist layer in that region with a mixing ratio maximum at 1 800 m, but an explanation for the 6 September sounding is not apparent.

Tritium mixing ratio soundings

The discussion thus far has been concerned with the vertical variations in the tritium concentration. To examine the variation with altitude of the actual *quantity* of tritium in the atmosphere, the concentration is weighted by the amount of water vapor present. This is done by multiplying T by the average atmospheric water vapor mixing ratio (MR) in the layer, expressed in grams H_2O/kg dry air. The product TMR, the tritium mixing ratio, is proportional to the total number of tritiated water molecules in the layer per kg of dry air.

All the TMR soundings except two in April exhibited a general decreasing trend with increasing altitude. That is, while the tritium concentration in water vapor was often found to increase with elevation, the actual amount of tritium per kg of air usually decreased. This is because MR decreases more rapidly than T increases for reasons to be presented.

The TMR soundings were examined in much the same manner as the T soundings. The variations in TMR are not directly related to the air trajectories, air mass classification or to the type of tritium sounding. On the two days where the TMR increased with altitude, both the MR and TMR were very low. On 21 April 1966 the TMR increase with altitude was apparently the result of a very moist layer aloft. On the 29 April sounding it was related to a very strong subsidence inversion accompanied by an unusually high T. These TMR-increase days occurred during the annual spring–early summer maximum in tropospheric radioactivity, but prior to the summer maximum in evapotranspiration and MR.

As water vapor of high T descends into the lower tropopause, it mixes with water vapor of low T from the oceans or with moderate T water vapor from the continent. Thus, its tritium concentration is diluted. An inverse relationship should, therefore, exist between MR and T. A regression analysis between T and MR was conducted to determine this relationship and the results are shown in Fig. 4. For a given MR the fall measurements exhibit a T of approximately 500 tritium units (TU) lower than the spring and summer measurements. This difference is a reflection of the annual midsummer decrease in tropospheric radioactivity caused by a reduction in the stratospheric–tropospheric exchange (Junge, 1963). The regression analysis of T vs. MR for the data in Fig. 4 excluded the autumn data because they are of a different magnitude, and a 2 980 TU value which may be in error. The regression equation for

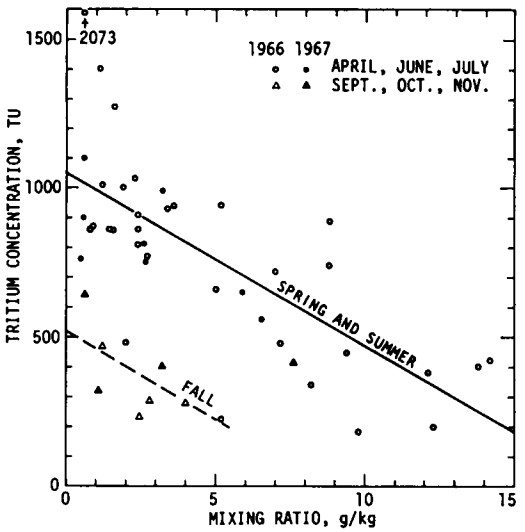

Fig. 4. Tritium concentration versus mixing ratios at all altitudes, for central Illinois.

the data is T = 1 046 − 58.2 MR and the resulting regression equation curve is plotted in Fig. 4. The correlation coefficient for T vs. MR is 0.65. A regression line is not given for the fall data because of their excessive scatter.

Considering only the spring and summer data in Fig. 4, the data with relatively high mixing ratios, that is MR > 4 g/kg, are composed of measurements in the lower portions of the summer soundings. The measurements with MR < 4 g/kg consist of high altitude summer data and both high and low altitude spring data. This illustrates that during the spring and summer, T is determined not so much by the altitude or season specifically, but by the MR, that is, the degree of dilution of the stratospheric tritium.

It may be further noted from Fig. 4 that the 1966 tritium concentrations were only slightly lower than those in 1967. This is interesting because the concentration of artificial radioactive material in the air has been decreasing since the start of the nuclear test moratorium in January 1963. An examination of tritium measurements from several different locations in the United States published by the International Atomic Energy Agency (1963–1967) indicated that the T in precipitation decreased by a factor of approximately two each year from 1963 through 1966. The data also show, however, a large variability in the tritium concentrations. With a limited number of samples it was apparently possible for samples taken in consecutive years to be similar in magnitude.

Lake Michigan soundings

Three soundings were made near Lake Michigan on 26 July 1966 to measure the horizontal variation of tritium and, if possible, the influence of Lake Michigan on the vertical distribution of tritium. A sounding was taken (1) several km inland on the windward (western) shore, (2) several km offshore on the leeward (eastern) shore, and (3) 70 km southwest of the lake (control sounding). The soundings were begun at 0953, 1055, and 1351 CST, respectively.

On 26 July the lake area was under the influence of an MT air mass with southwesterly winds at 6 m sec^{-1} at the flight altitudes, but calm at the surface. There was no lake breeze effect on either side of the lake (Lyons & Wilson, 1968). The conditions present were not optimum for a modification of T by the lake, but the sounding date was necessarily predetermined.

The results of the soundings are tabulated in Table I. The soundings show a general decrease of tritium with increasing altitude, and, except for the lowest control sample, the measurements at all altitudes agree with each other within counting error limits. The soundings thus indicate a fair degree of homogeneity of T in the atmosphere over distances of 250 km.

The exchange of tritium between the atmosphere and surface of a large body of water can be extensive (Bolin, 1958). Bolin predicted that with sufficient vertical mixing the molecular exchange of water vapor at the lake surface would cause 68 % of the tritium in the first kilometer of air above the lake to pass into the lake, assuming a wind speed of 3 m sec^{-1}. Yet the average concentration in the lowest leeward measurement taken from 67 to 280 m above the lake surface was three times greater than the average lake concentration (140 TU) and apparently was unaffected by the Lake. The lack of apparent exchange between the sampled air and the lake for the day investigated is attributed to a strong thermal inversion which existed throughout the depth of the lowest sample as measured by the aircraft

thermometer. The inversion prohibited the tritium concentrations present in the lowest levels from reaching the sampling altitude by vertical mixing. It would have been of great interest to sample the lowest 67 m, but safety factors made it prohibitive.

Summary and conclusions

It has been shown that there are three basic types of tritium soundings found in the lower troposphere over Illinois. The type I soundings, which showed a general increase in T with increasing altitude, were produced when the accompanying air mass had a long continental trajectory from the west or north. The type II soundings, characterized by a constant T with increasing altitude were caused, in one instance, by good vertical mixing, and in another by a high-level moist layer. An explanation of another type II sounding was not apparent. The type III sounding were characterized by a T which gradually decreased with altitude, followed by an abrupt increase between 2 and 3 km. Four out of six of the type III soundings were associated with warm air masses from the Gulf and the relative tritium maximum near the surface appeared to be caused by rapid tritium input through evapotranspiration. In most of the soundings of type I and III the abrupt increase in T aloft was associated with a subsidence inversion or the top of a moist layer.

In 8 out of 11 higher altitude soundings the tritium concentration in the lower troposphere over the Midwest increased by a factor of approximately two from the surface to 4 or 5 km MSL. This results from the stratosphere being the source of water vapor of high tritium concentration. However, the absolute amount of tritium, that is, the number of tritium atoms per kg of air, a number proportional to the TMR, usually decreases with increasing elevation. This is the consequence of two factors. First, the lowest levels of the troposphere are rich in moisture and act as a trap for the descending tritium. The tritium diffuses and mixes down to the moist layer at a high concentration and becomes diluted. Once diluted it can return upward by convection, but only at a reduced concentration. Thus the moist, lower atmosphere acts as a tritium sink in much the same manner as the oceans. The second factor responsible for the large amount of tritium in the moist layer is the evapotranspiration of water vapor that has a moderate tritium concentration. The evapotranspiration factor was apparent in the form of a tritium inversion in air masses from the Gulf of Mexico arriving in the Midwest.

The soundings demonstrate several problems that may arise if tritium were used to trace water vapor through a storm. Since the concentration of tritium does not vary greatly within the first 5 km, processes such as fractionation during condensation and the molecular exchange of water vapor between raindrops and their environment (Bolin, 1958; Booker, 1965) may cause changes in the precipitation concentration of the same order of magnitude as the variations of tritium with different elevations.

Acknowledgements

We are grateful to J. W. Wilson and N. G. Towery for assistance in sampling and trajectory analysis and S. A. Changnon, Jr, and R. G. Semonin for reviewing the manuscript. We thank L. B. Lundgren and Dr E. Eriksson of the International Meteorological Institute in Stockholm, Sweden, for analysis of the 1966 tritium samples under contract with the U.S. Atomic Energy Commission. We are grateful to Dr H. G. Östlund and R. D. Stearns of the Institute of Marine Sciences, University of Miami, for helpful discussions of sampling techniques and the analysis of several samples. The 1967 tritium samples were analyzed by Isotopes, Inc. of Westwood, New Jersey, under contract with the Water Survey.

This research was sponsored by the U.S. Atomic Energy Commission, Fallout Studies Branch, Division of Biology and Medicine, and published as appendix B of Report No. COO-1199-17.

REFERENCES

Bolin, B. 1958. On the use of tritium as a tracer for water in nature. *Proceedings of the Second United Nations Conference on the Peaceful Uses of Atomic Energy*, 18, Geneva, Switzerland, 336–343.

Booker, D. V. 1965. Exchange between water droplets and tritiated water vapor. *Quart. J. Roy. Meteorol. Soc. 91*, 73–79.

Bradley, W. E. & Feteris, P. J. 1966. *Study of rainout of radioactivity in Illinois.* Fifth Progress Report, AEC Contract AT(11-1)-1199, 27.

Eriksson, E. 1958. The possible use of tritium for estimating groundwater storage. *Tellus 10*, 472–478.

Giletti, J. B. Bazan, F. & Kulp, J. L. 1958. The geochemistry of tritium. *Trans. Amer. Geophys. U. 39*, 807–818.

Houtermans, J. 1965. Tritium in surface water of the North Pacific Ocean. *6th Carbon 14 and Tritium Dating Conf.*, CONF 650562, 565.

Junge, C. E. 1963. *Air chemistry and radioactivity* p. 261. Academic Press, New York.

Lyons, W. A. & Wilson, J. W. 1968. The control of summertime cumuli and thunderstorms by Lake Michigan during non-lake breeze conditions. *Satellite and Mesometeorology Research Project Paper No. 74*, University of Chicago, 32.

Östlund, H. G. 1968. Hurricane tritium II: Air–sea exchange of water in Betsy 1965. *Tellus 20*, 577–594.

Thatcher, L. L. 1962. The distribution of tritium fallout in precipitation over North America. *Bull. of the Int. Assoc. of Sci. Hydro. 7(2)*, 48–58.

Visher, S. S. 1954. *Climatic atlas of the United States.* Harvard University Press, Cambridge.

ВЕРТИКАЛЬНОЕ РАСПРЕДЕЛЕНИЕ ТРИТИЯ В ВОДЯНОМ ПАРЕ В НИЖНЕЙ ТРОПОСФЕРЕ

Для определения вертикального распределения трития в водяном паре от поверхности до высоты 5 км в летние периоды 1966 и 1967 гг. было проведена серия из 16 самолётных зондирований. Были найдены три типа распределений трития. Тип I проявляет общее увеличение концентрации трития с высотой. Тип II характеризуется постоянством концентрации с высотой. В типе III концентрация постепенно уменьшается приблизительно до 2,4 км, а затем резко возрастает. Результаты зондирований обсуждаются в терминах происхождения воздушных масс, эвапотранспирации и отношения смеси для водяного пара.