TRITIUM in the Physical and Biological Sciences

Volume I

PROCEEDINGS SERIES

TRITIUM IN THE PHYSICAL AND BIOLOGICAL SCIENCES

I

PROCEEDINGS OF THE SYMPOSIUM ON
THE DETECTION AND USE OF TRITIUM IN THE PHYSICAL
AND BIOLOGICAL SCIENCES
SPONSORED BY
THE INTERNATIONAL ATOMIC ENERGY AGENCY
IN CO-OPERATION WITH THE
JOINT COMMISSION ON APPLIED RADIOACTIVITY
AND HELD IN VIENNA, 3—10 MAY 1961

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA 1962

FOREWORD

The use of tritium for research in physics, chemistry, biology and hydrology has in recent years become increasingly important. It was for this reason that the first international conference to discuss the progress of new developments was organized by the IAEA in conjunction with the Joint Commission on Applied Radioactivity and held from 3–10 May 1961, in Vienna.

The first five sessions of the Symposium were devoted to the use of tritium in hydrology, physics and chemistry. Special emphasis was laid on the role of tritium as a tracer in hydrology, especially in the study of water movement. The establishment and improvement of counting and detection techniques to facilitate the application of tritium as a tracer was another aspect discussed in this part of the proceedings. Papers were read on the preparation of tritiated compounds and it was generally agreed that further clarification of the mechanism of various techniques, and of the Wilzbach gas exposure technique in particular, would lead to further developments in the synthesis of a number of tritium compounds important in biology. Other papers were concerned with tritium applications to studies of the mechanism of some chemical reactions together with the effects of tritium isotopes.

During the second part of the Symposium the biological applications of tritium and tritiated compounds were discussed. These included general problems connected with the biological uses of tritium and the radiation effects of tritium on living organisms such as viruses, bacteria and cancer cells. The value of tritium in biological studies became apparent because of the ease with which a large number of metabolically active compounds such as hormones, vitamins and other important constituents in the body can be labelled with tritium. Tritium is also a weak beta-emitter and autoradiographies of tissues and single cells containing tritium-labelled compounds allow an excellent localization of the tracer.

The Symposium was attended by some 290 scientists from 27 countries and five international organizations who altogether contributed a total of 67 papers.

The Agency believes that the publications of the proceedings will not only provide information for a wider public but will also help to stimulate further research in the use of tritium.

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IAEA, VIENNA, 1962
STI/PUB/39

CONTENTS OF VOLUME I

A. DISTRIBUTION OF TRITIUM IN NATURE, METHODS OF ENRICHMENT AND APPLICATIONS IN HYDROLOGY

B. TRITIUM IN CHEMISTRY AND PHYSICS

C. DETECTION AND COUNTING OF TRITIUM

A

DISTRIBUTION OF TRITIUM IN NATURE, METHODS OF ENRICHMENT AND APPLICATIONS IN HYDROLOGY

(Session I)

INTRODUCTION

Prof. W. F. LIBBY (United States of America), Chairman of the inaugral session, opened it with the following speech:

Tritium was first discovered in the Cavendish Laboratory by Lord Rutherford and his co-workers, who found that the (d,d) reaction produced tritium. The (d,d) reaction also produces helium-3. It was not known which of these two, helium-3 or tritium, was radioactive. Alvarez at Berkeley established that helium-3 occurred in nature and was stable. He later proved that tritium was radioactive. Lord Rutherford, in one of his last scientific researches, looked for tritium in natural water. He used the method of enrichment which most of us now studying tritium geophysics use, namely, electrolysis to produce heavy water. He then examined his enriched sample for tritium. However, not knowing that tritium was radioactive, he locked for natural tritium with a mass spectrometer and failed to find it. But such is the course of scientific research that the first tritium to be discovered in natural water was found some years later in Lord Rutherford's sample, when a Geiger counter was used instead of a mass spectrometer. Our research at the University of Chicago, which was concerned with the occurrence of tritium in water in nature, began with an examination of Lord Rutherford's sample, and it was found to be intensely radioactive with tritium. The occurrence of tritium in natural waters is now an old story. We are concerned with two epochs: the pre-thermonuclear explosion epoch and the post-thermonuclear explosion epoch or, described differently, the cosmic ray tritium era and the synthetic tritium era. In samples taken before March 1954, the tritium is very largely cosmic ray in origin. Samples taken after that the time are, by and large, synthetic, i. e. man-made, in origin, although there is of course some admixture, so that certain samples taken as early as in 1952 can show synthetic tritium while other samples taken now, for example, from 20-year old wine, can show only cosmic ray tritium.

TRITIUM GEOPHYSICS: RECENT DATA AND RESULTS*

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Abstract — Résumé — Аннотация — Resumen

Tritium geophysics: recent data and results. Rain waters contain radioactive hydrogen (tritium) because of the bombardment of the atmosphere by the cosmic rays and because of the synthetic production in the atmosphere by thermonuclear explosions. Prior to 1952, the only source of rain tritium was due to cosmic rays and early measurements established the level of the cosmic ray tritium in terrestrial waters.

Since 1952, and particularly since 1954 and up to 1959, the cosmic ray tritium in rain has been overshadowed by synthetic tritium. For example, California rain now averages about 15 to 20 Tritium Units (I.T.U. is 1 T-atom per 10^{18} H-atoms) and the Mississippi River runs at over 100 T.U., whereas it ran at 6 T.U. before. It is now possible to measure tritium more easily and the finer details of many problems which were not resolved by the cosmic ray tritium can now be established.

The turnover time between the oceans and continental water can now be measured more accurately. Comparisons can be made of the surface waters of the oceans in the northern and southern hemispheres which provide the mixing rate between the two hemispheres. It has also been discovered that tritium remains in the stratosphere as long as non-volatile radioactivity such as strontium-90. The measurement of surface ocean water has indicated that there are extensive areas of upwelling off the coast of Southern California.

Emploi du tritium en géophysique: données et résultats obtenus récemment. Les eaux de pluie contiennent de l'hydrogène radioactif (tritium) dont une partie provient du bombardement de l'atmosphère par les rayons cosmiques et l'autre s'est formée par synthèse dans l'atmosphère, sous l'effet des explosions thermonucléaires. Avant 1952, les eaux de pluie ne contenaient que du tritium provenant des rayons cosmiques, et les premières mesures qui ont été faites ont établi la concentration de ce tritium dans les eaux terrestres.

Depuis 1952, et notamment entre 1954 et 1959, la quantité de tritium synthétique contenue dans l'eau de pluie a considérablement augmenté par rapport à celle du tritium naturel. Ainsi, en Californie, l'eau de pluie contient maintenant en moyenne de 15 à 20 unités de tritium (1 unité de tritium = 1 atome de tritium pour 10^{18} atomes d'hydrogène) et la teneur en tritium du Mississipi atteint plus de 100 unités, contre 6 auparavant. Il est maintenant possible de mesurer plus facilement la concentration du tritium et de préciser les détails de nombreux problèmes que l'on n'avait pu résoudre à l'aide du tritium provenant des rayons cosmiques.

On peut maintenant mesurer exactement le cycle des échanges entre les eaux des océans et les eaux continentales. En comparant les eaux de surface des océans de l'hémisphère nord et de l'hémisphère sud, il est possible de calculer le taux de mélange des eaux des deux hémisphères. On a également découvert que le tritium séjourne dans la stratosphère aussi longtemps que les corps radioactifs non volatils, comme le strontium-90. La mesure de la teneur en tritium des eaux de surface de l'océan a mis évidence l'existence de mouvements verticaux en de vastes zones au large de la côte de la Californie méridionale.

Геофизика трития: последние данные и результаты. Дождевые воды содержат радиоактивный водород (тритий) в результате бомбардировки атомов атомсферы космическими лучами и в результате вызванного термоядерными взрывами

* Supported by the Water Resources Center of the University of California, the University of California Research Committee and the United States Air Force through the Air Force Office of Scientific Research of the Air Research and Development Command.

процесса синтетической генерации в атмосфере. До 1952 года единственным источником содержащегося в дождевой воде трития являлись космические лучи и согласно более ранним измерениям был установлен уровень появившегося таким образом трития в земных водах.

С 1952 года, и в особенности с 1954 года и вплоть до 1959 года, выпадение получающегося от бомбардировки атмосферы космическими лучами трития было мало по сравнению с выпадением синтетического трития. Например, в настоящее время во время дождя в Калифорнии выпадает в среднем от 15 до 20 единиц трития (1 единица трития равняется 1 атом трития на 10^{18} атомов водорода), и река Миссисипи несет всыше 100 единиц трития в то время как прежде в ней было до 6 единиц трития. В настоящее время является возможным более легко измерить содержание трития и могут быть установлены более тонкие детали многиц проблем, которые не были решены с помощью трития, получающегося от бомбардировки атмосферы космическими лучами.

В настоящее время может быть точно измерено время оборота между океанами и континентальными водами. Могут быть сделаны сравнения степени смешения поверхностных вод океанов в северном и южном полушариях. Было также открыто, что тритий остается в стратосфере также долго, как нелетучие радиоактивные вещества подобно стронцию-90. Измерение, произведенное в поверхностных слоях океанских вод, показало, что имеются обширные районы подъема уровня воды у побережья Южной Калифорнии.

Empleo del tritio en geofísica: datos y resultados recientes. El agua de lluvia contiene hidrógeno radiactivo (tritio) originado en el bombardeo de la atmósfera por los rayos cósmicos y producido también sintéticamente como consecuencia de las explosiones termonucleares. Antes del año 1952, el tritio presente en el agua de lluvia tenía como origen exclusivo la mencionada acción de los rayos cósmicos y las primeras mediciones permitieron determinar la cantidad de tritio, producida, por tales rayos, en las aguas del globo terrestre.

A partir de 1952 y especialmente entre los años 1954 y 1959, la proporción de tritio sintético en el agua de lluvia ha ido superando a la proporción debida a los rayos cósmicos. Así, por ejemplo, el contenido de tritio en las aguas pluviales de California oscila, en la actualidad, entre 15 y 20 unidades de tritio (1 unidad de tritio corresponde a 1 átomo de tritio por 10^{18} átomos de hidrógeno) y asciende a unas 100 unidades de tritio en las aguas del río Mississippi, en tanto que anteriormente contenía 6 de esas unidades. Hoy en día la determinación cuantitativa del tritio ofrece menos dificultades, siendo posible aclarar muchos aspectos de una serie de problemas que no era posible resolver partiendo de los datos obtenidos con ayuda del tritio originado por los rayos cósmicos.

Así, se puede ahora medir con exactitud el tiempo de intercambio entre las aguas oceánicas y las aguas continentales. También es posible comparar las aguas superficiales de los océanos del hemisferio Norte con las del hemisferio Sur y deducir la velocidad de mezcla entre las aguas de los dos hemisferios. Del mismo modo, se ha comprobado que el tritio permanece en la estratosfera tanto tiempo como las sustancias radiactivas no volátiles, tales como el estroncio-90. Por otra parte, las mediciones en las aguas superficiales del océano permitieron comprobar la existencia de vastas zonas de corrientes ascendentes a la altura de la costa de la California meridional.

Introduction

The method of analysis for natural tritium in water described earlier by our group (GROSSE, A. V., JOHNSTON, W. H., WOLFGANG, R. L. and LIBBY, W. F., 1951; KAUFMANN, S. and LIBBY, W. F., 1954; VON BUTTLAR, H. and LIBBY, W. F., 1955; BEGEMANN, F. and LIBBY, W. F., 1957) and by others (BROWN, R. M. and GRUMMITT, W. E., 1956; GILLETTI, B. J., BAZAN, F. and KULP, J. L., 1958; SUESS, H. E., 1958; BROWN, R. M., 1960; CARLSTON, W., THATCHER, L. and RHODEHAMEL, E. C., 1960) has been used to measure a selection of recent rain and surface water and ground water samples of geophysical interest — principally hydrological and oceanographic.

The results are given in Table I, and certain salient features are presented in Figs. 1-4.

Results

RELATIVE STRATOSPHERIC RESIDENCE TIMES FOR WATER VAPOUR AND SR^{90} PARTICLES (SAMPLES FROM E. A. MARTELL)

Fig. 1 shows for Bedford, Massachusetts, the variation in tritium content of the rains throughout 1959 and the spring, summer, and early fall of 1960, together with the average northern hemisphere fallout for Sr^{90} during 1959 (LIBBY and PALMER, 1960, Fig. 1). It is immediately clear that during the spring and summer

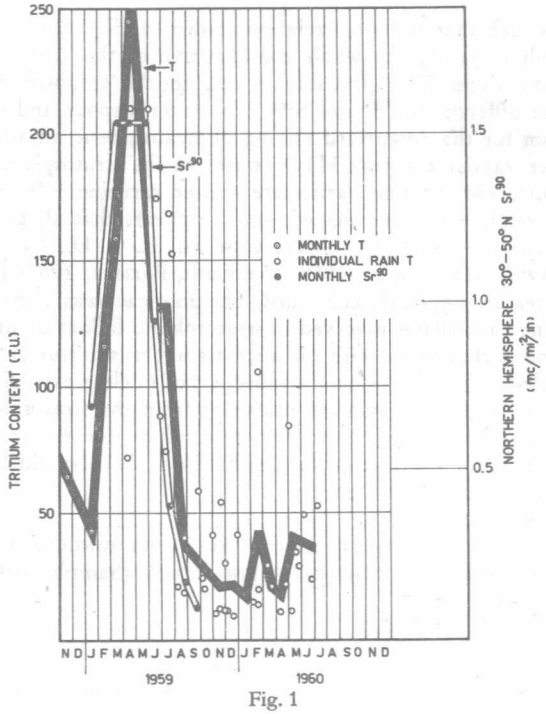

Fig. 1

Bedford tritium in rain vs. northern hemisphere average 30° – 50° N Strontium 90 in rain.

of 1959 the peaks attained by the T (Tritium) and Sr^{90} fallout rates were proportional to one another with an average proportionality constant of about 130 T.U. (1 Tritium Unit [T.U.] is 1 T-atom/10^{18} H-atoms) per mc of Sr^{90} per square mile per inch of rain or 13 T atoms per Sr^{90} atom. Considering that the fallout in the spring and summer of 1959 in the northern hemisphere (LIBBY, 1959; MARTELL, 1959; LIBBY and PALMER, 1960; MARTELL and DREVINSKY, 1960; LOCKHART, 1960) was due largely to the Russian October 1958 test series, this parallelism of the T and Sr^{90} fallout strongly suggests that both the T and Sr^{90} came from the same source — the island of Novaya Zemla in October 1958 — and that the mechanism of removal from the stratosphere into the troposphere was the same for both T and Sr^{90}. Some of the Bedford rains were measured for Sr^{90} as well as T, the results being given in Table II (the Sr^{90} data were furnished by Dr. E. A. MARTELL and P. J. DREVINSKY of the United States Air Force [Cambridge Research Laboratories]).

TABLE II

T/Sr^{90} RATIO DURING SPRING 1959 PERIODS

Period of Collection	Sr^{90} (mc/mi²/inch)	T. U.	T/Sr^{90} Atoms
Feb. 24 to Mar. 31	0.81 ± .08	159 ± 4	19
Apr. 1 to Apr. 3	0.83 ± .08	292 ± 15	33
May 25 to June 3	0.17 ± .01	220 ± 7	123
July 7 to July 13	0.06 ± .02	74 ± 7	117
			Average 73

The scatter is such that if the correlation shown in Fig. 1 were not so clear, it would be difficult to justify the whole consideration of the T/Sr^{90} atomic ratio on the Bedford data alone. This probably is due to the tropospheric precipitation mechanism being different for T and Sr^{90}, i. e. water vapour and dust.

The mechanism for the downward mixing of stratospheric air thus must be such as to treat water vapour carrying HTO (water in the stratosphere most probably is mainly vapour), and the fine particulate matter carrying Sr^{90} in essentially the same way. This result is not unexpected, and it appears that all present theories of the fallout mechanism (STEWART, N. G., OSMUND, R. G. D., CROOKS, R. N. and FISHER, E. N., 1957; MACHTA, L., 1959; LIBBY and PALMER, 1960; FEELY and SPAR, 1961) have this feature implicitly contained. The physical basis is that no appreciable settling of the fine particulates involved in stratospheric fallout occurs so that for all intents and purposes they move with the air mass just as the true gases do. Thus, the expectation would be that all radioactive porducts of nuclear explosions should show the same fractional rate of downward transport from the stratosphere, and except for the complication of re-entry into the stratosphere for the long-lived gaseous products which have no appreciable surface hold-up, there would be essentially no distinction between any of them, either.

GENERAL DEPENDENCE OF TRITIUM LEVELS ON TOTAL THERMONUCLEAR ENERGY RELEASE LARGE SCALE UPWELLING ALONG THE CALIFORNIA COAST (Samples from K. O. Emery and H. E. Suess)

The Pacific Ocean surface waters:

	Sample Date	T. U.
Santa Monica Beach	9 Mar. 1960	14 ± 2.3
Johnston Island	30 Apr. 1960	21 ± 2
Samoa, Lago Lago	1 Feb. 1960	6 + 1

show that, in general, the surface waters are much more active than they were before bomb testing started. (1 T. U. in 1952-53 and about 3 T. U. in late 1954 and 1955 after "Castle's" early fallout; Begemann and Libby, 1957; Gilletti, Bazan and Kulp, 1958.)

In fact, using the experience on the North American continent, where the diluting effects of very low T concentration water is very much smaller than in the open ocean, we can see in Fig. 2 that the rise in T content in post-early-fallout surface waters or land rains (three to six months after the tests) is proportional roughly to the estimated total number of megatons of ordinary explosive energy equivalent of thermonuclear fusion energy released (Libby, 1959) despite the wide range of firing conditions with consequent wide variations in local and quick fallout of tritium into the ocean in the case of some of the tests in the Pacific. Therefore, we can expect

that the surface waters of the oceans in general must rise in the manner indicated in Fig. 2 and that the average for the northern oceans now should be about 8 T. U.

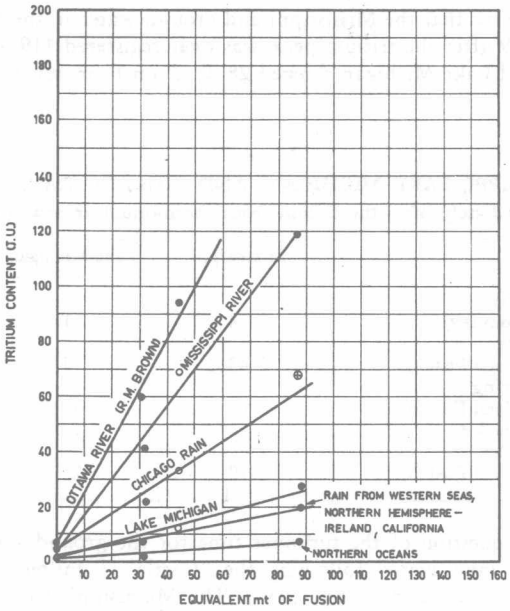

Fig. 2
Bomb Tritium Plot.
O Data of Begemann (unpublished)
X Data of Barrett (unpublished)
(Values are for post fallout; Aug. 1955; Sept. 1956; Aug. 1959)

Consequently, it is with extreme interest that we observe values of 2 T. U. or less in surface ocean water.

TABLE III
PACIFIC (SOUTH OF SANTA CRUZ ISLAND OFF CALIFORNIA COAST) SAMPLES FROM K. O. EMERY, U. S. C.

		T. U.
Lat. N. 32°50.2′	Long. W. 119°42.2′	1.8 ± .3 0.55 ± .4
Lat. N. 31°52′	Long. W. 120°26′	1.6 ± .2 0.9 ± .2

From these data it appears to be very probable that these two points over 75 miles apart, about 100 miles off the lower California coast, are in a large upwelling area such that the water is very largely from the depths with only a small component of recent surface water. Further measurements must be undertaken to elucidate the extent of this area. Professor Emery submitted these samples for the reason that he expected the first to show upwelling. The second was collected as a control point, but it, too, apparently lies within the area.

CONTINENTAL WATER BALANCE AND TURNOVER TIME FOR CONTINENTAL GROUND WATER
(Samples from A. C. Wahl and A. F. Hakanson and unpublished Chicago rain data of Begemann and of Barrett)

From Fig. 2 we see that the Mississippi and ground water in the Mississippi Valley in the fall of 1959 after the tritium peak was over registered 119 T. U. as compared to 68 in 1957, and Lake Michigan showed 28 T. U. on February 1, 1960, a rise from about 12 T. U.

TABLE IV

MISSISSIPPI, LAKE MICHIGAN, AND CHICAGO RAIN LEVELS
Immediately after the fallout from thermonuclear tests (T. U.)

	Mississippi	Lake Michigan	Chicago Rain
Cosmic Ray (? Mt from Ivy)	4	1.6	6
Post Castle (30 Mt Total Fusion)	43	7	22
Post Redwing (44 Mt Total Fusion)	68	12	33
Post Hardtack & Russian Oct. (87 Mt Total Fusion)	119	28	67

The important question of the turnover time for the ground water in the Mississippi Valley was investigated following the "Castle" T fallout (Von Buttlar and Libby, 1955; Begemann and Libby, 1957). The Mississippi was measured at Rock Island and the results in Table V obtained.

TABLE V

MISSISSIPPI RIVER AFTER „CASTLE"

Sample Date	T. U.
Jan. 3, 1955	55 ± 1
Feb. 3, 1955	44 ± 2
Mar. 1, 1955	40 ± 2
Apr. 1, 1955	43 ± 3
May 4, 1955	44 ± 1
June 2, 1955	42 ± 1
Aug. 1, 1955	44 ± 5
Sept. 3, 1955	41 ± 2

These data strongly indicate that the replacement time for Mississippi Valley ground water is longer than a few months and it was concluded (Begemann and Libby, 1957) that the mean lifetime was about ten years. Shortly after the last of the measurements listed in Table V, further Russian nuclear tests occurred, followed by the Pacific "Redwing" series in 1956 so the curve could not be followed further. At the present time, over two years having elapsed since the last substantial atmospheric injections, the Mississippi is showing 119 T. U. — a value on the curve (Fig. 2) for tritium concentration without turnover — again indicates a long time for turnover.

The hydrology of Lake Michigan is such (Kaufmann and Libby, 1954) (95 m depth