Tellus (1982), *34*, 369–375

Stratospheric HTO and ^{95}Zr residence times

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(Manuscript received August 6; in final form November 19, 1981)

ABSTRACT

The depletion of the stratospheric burdens of particulate ^{95}Zr for times up to around 1 year, and gaseous HTO for times up to 3 years, attributed to the November 17, 1976 Chinese high-yield test indicates that both have about the same residence half-time (10 months). The indications are that gravitational settling of particles in the lower stratosphere can be considered to be negligible in studying transport processes. The rate of depletion of the stratospheric burden of HTO from the high-yield Chinese test of June 27, 1973 is not as well-defined in part due to greater uncertainties in calculating the stratospheric burdens.

1. Introduction

The Tritium Laboratory of the University of Miami began programs of measurement of tritiated water vapor (HTO) and tritium gas (HT and T_2) collected at ground level in 1968, and from aircraft in 1971. Previous publications have described the techniques and interpreted the data (Östlund and Mason, 1974; Mason and Östlund, 1976, 1979; Mason, 1977). Data through 1976 are available in unpublished reports (Östlund et al., 1972; Mason and Östlund, 1977). Project Airstream (sponsored by the U.S. Department of Energy) is a long-term study of stratospheric radioactivity and chemistry, carried out by three series of flights annually, covering the latitude range from the equator to 75° N. Four flight levels above and below the tropopause are sampled over that span; in addition, vertical profiles have been flown in the vicinity of Panama; Houston, Texas; and Anchorage, Alaska, for each flight series since July 1977. The vertical profile samples are obtained from 3 to 19 km altitude, and from transects between 14 and 19 km.

Tritium measurements have been made as part of Project Airstream since 1975; however, the project is older than that, having begun in 1965 with sampling of particulate and noble gas radioactivity. ^{95}Zr is a particulate fission product with a 65-day half-life, produced by all nuclear tests, while HTO is normally a gas, with a half-life of 12·26 years, produced by the fusion reaction in a thermonuclear device. Measurements of ^{95}Zr have been used extensively to estimate stratospheric residence times, initial vertical activity distributions, and atmospheric transport from the high-yield nuclear tests conducted by China between 1967 and 1976 (Telegadas, 1974, 1976, 1979). These data can now be compared with the HTO data to address the question of the significance of particle settling in studying transport processes in the lower stratosphere.

2. Tritium data

The HTO data used in this study were obtained during discrete sampling periods between April 1975 and August 1980. They are published elsewhere through July 1979 (Mason et al., 1980). Later data are available from the authors. The data

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† Mr. Kosta Telegadas died unexpectedly on Jan. 29, 1982. This paper is dedicated to his memory.

0040-2826/82/040369-07$02.50/0

take the form of mixing ratios of HTO, i.e., tritium atoms per mg of air. These units may be converted to picocuries per standard cubic meter of air (pCi/SCM) by multiplication by 0.0625 for SCM defined at 1013 mb and 0 °C. HT data for the same flights are also available but are not considered in this report. From the available data, the HTO burdens in kilocuries were calculated for the stratosphere up to about 20 km in the Northern Hemisphere.

The HTO burdens were calculated by the same graphical integration technique used by Telegadas (1979) for the ^{95}Zr burdens. The errors of the technique have been estimated at about $\pm 10\%$ by submission of identical data sets to independently working analysts and comparison of the results. This estimated error exceeds that calculated for the individual data points, which are typically $\pm 4\%$ (1σ) (Mason et al., 1980).

3. The stratospheric tritium and ^{95}Zr inventory

As was pointed out by Eriksson (1965), the source of stratospheric HTO is primarily the testing of thermonuclear (fusion) devices. One would expect that the six reported Chinese thermonuclear tests, all performed at Lop Nor (40° N 90° E) between 1967 and 1976, would be significant contributors to the stratospheric HTO burden. Changes in the stratospheric HTO burden should provide information about stratospheric-tropospheric exchange processes and transport in the stratosphere.

Telegadas (1976, 1979) has analyzed the fission product data (primarily ^{95}Zr and ^{144}Ce) following the June 27, 1973 and November 17, 1976 Chinese nuclear tests. Since ^{95}Zr has a relatively short half-life, due to radioactive decay and stratospheric depletion, the stratospheric input from these two events, as those from earlier high-yield Chinese tests, could be followed unequivocally for only about 1 year. The HTO stratospheric input could be followed for many years due primarily to its much longer radioactive half-life.

Two possible problems exist with using these HTO data for a direct comparison with fission product data: (1) the calculated stratospheric HTO burdens may contain a background from past high-yield tests (attempts will be made to resolve this problem); and (2) although the production of fission products from nuclear tests is fairly well known from the fission yield of the detonation (Harley et al., 1965), the HTO production from thermonuclear tests has a much larger uncertainty. The latter has been reported to range from 7 to 50 megacuries per megaton fusion (MCi/MT (fusion)), with a suggested average value of 20 MCi/MT (fusion) (National Council on Radiation Protection and Measurements, 1979). It is, therefore, difficult to know with certainty the amount of HTO injected into the atmosphere even if the total yield and fission of an event are known.

There were no simultaneous measurements of ^{95}Zr and HTO following the June 27, 1973 high-yield test, whereas there were for three sampling series after the November 17, 1976 test. This latter test will, therefore, be discussed first, followed by an analysis of the June 27, 1973 event. The Northern Hemisphere stratospheric HTO burdens for the ten sampling periods between July 1977 and August 1980 are given in Table 1 together with the ^{95}Zr burdens calculated by Telegadas (1979). The first two columns of numbers show the ^{95}Zr burden to about 20 km (based on aircraft sampling) and to about 30 km (based on additional balloon sampling from 20 to 30 km). The first line under these two columns shows the ^{95}Zr burden prior to the November 17, 1976 test. The last significant test prior to this event occurred on June 17, 1974 (reported total yield of between 0.2–1 MT) and estimated by Leifer et al. (1976) to have a fission yield of 0.4 MT. By the time of the November 17, 1976 test, due to depletion and radioactive decay, the stratospheric ^{95}Zr created by the earlier test had decayed below detection limits. The HTO collected between October 24 and November 17, 1976 indicated a background of 3100 kCi of HTO residing in the stratosphere prior to the November 17, 1976 test. This is shown in column A, line 1. Column B shows the observed burden (column A) decay-corrected to the November 17, 1976 test. The burden for the October 13–November 6, 1978 sampling period listed in column A is questionable due to limited data (Mason et al., 1980), as is that for October 18–November 11, 1979 for the same reason.

A line of regression through the decay-corrected HTO burdens given in column B (from July 1977 to August 1980) would indicate a residence

Table 1. *Northern hemisphere stratospheric burden (kilocuries)*

	^{95}Zr*		Tritium (as HTO)			
Sampling period	to 20 km	to 30 km	to 20 km			
			A	B	C	D
Oct. 24–Nov. 17, 1976	0†	0†	3100	3100	3100	
Mar. 22–Apr. 10, 1977	50900	58100		No data		
July 6–22, 1977	38400	45500	18800	19500	2100	17400
Oct. 12–29, 1977	30600	36100	8700	9200	1700	7500
Apr. 6–21, 1978	21000	‡	5900	6300	1100	5200
July 12–31, 1978			4000	4400	950	3450
Oct. 13–Nov. 6, 1978			5300§	5900§	750	5150§
Apr. 6–24, 1979			3400	3900	500	3400
July 3–26, 1979			2500	2900	400	2500
Oct. 18–Nov. 11, 1979			1140§	1350§	270	1080§
Apr. 9–May 21, 1980			1090	1320	220	1100
July 23–Aug. 11, 1980			920	1130	180	950

* Decay-corrected to Chinese test of November 17, 1976.
† Background (last significant test June 17, 1974; total yield 0.2–1 MT).
‡ No data available above 20 km at this time.
§ Questionable—see text.
A. Burdens based on observed data.
B. Burdens from column A decay-corrected to November 17, 1976.
C. Background at time of Chinese November 17, 1976 test. Background burdens at later times assumed using a residence half-time of 10 months.
D. Residual burden attributed to November 17, 1976 test (B minus C).

half-time of about 10 months. It was, therefore, assumed that the background HTO burden prior to the November 17, 1976 test would be depleted with this same residence time. The natural background is assumed to be small and has been neglected. If it were appreciable, it would cause the HTO residence time to be shorter than calculated, as well as shorter than that of ^{95}Zr. The background burdens at later times are listed in column C. Column D shows the residual burden attributed to the November 17, 1976 test, that is, column B minus column C. The residual burdens are shown in Fig. 1 together with the assumed depletion of the background listed in column C.

The Northern Hemisphere ^{95}Zr stratospheric burdens to 20 and 30 km shown in Fig. 1 are extrapolated back to February 1, 1977, when it was estimated that significant fallout started following the November 17, 1976 event (Telegadas, 1979). It can be seen from either Fig. 1 or Table 1 that about 15% of the ^{95}Zr burden resided above the aircraft altitude of approximately 20 km. The ^{95}Zr burden to 30 km is determined to be 66,000 kCi which is equivalent to a fission yield of 2.7 MT. The reported total yield for this event was 4 MT; therefore, the fusion yield is estimated to have been 1.3 MT.

Similar back-extrapolation of the HTO burdens above background (Table 1, Column D) indicates an input into the stratosphere of 16,600 kCi of HTO to 20 km. Increasing this amount by 15% (assuming the same % of tritium above the aircraft altitudes as was determined for the ^{95}Zr burden) would indicate an input of 19,000 kCi of HTO. Since the fusion yield was estimated to be 1.3 MT, the assumed production of HTO from the November 17, 1976 event would be about 15 MCi/MT (fusion). This falls within the range of production figures discussed above.

The error limits on the slope of the regression were calculated from the Student *t*-distribution at a 90% confidence level, and were found to be −0.083 to −0.054 per month, or a mean residence time of 14.6 months with upper and lower limits of 18.6 and 12.0 months, respectively. Adding the incomplete October–November 78 and October–

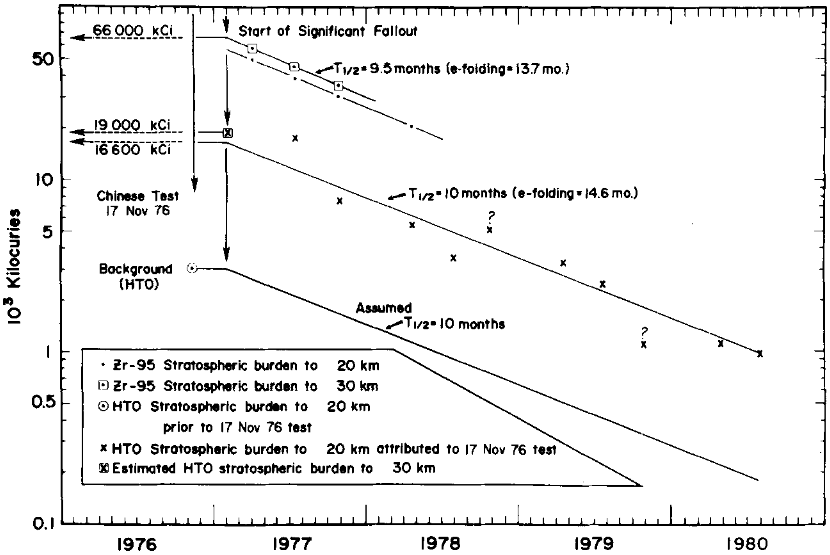

Fig. 1. Stratospheric half-residence-time of the ^{95}Zr and HTO burden for the Chinese tests of November 17, 1976. All burdens decay-corrected to time of test. See text for a more complete description.

November 79 data sets to the regression changes the residence time to 13.9 months with bounds of 17.6 and 11.4 months.

The first stratospheric sampling following the June 27, 1973 test for which a reliable stratospheric inventory could be calculated was not until April 18–May 6, 1975 (Table 2), nearly 2 years later. Shown in Table 2 are the computed Northern Hemispheric stratospheric HTO burdens to approximately 20 km. Column 1 gives the observed inventory. Decay-correcting these inventories to June 27, 1973, the Chinese high-yield test is shown in column 2. The burdens listed in column 2 are shown in Fig. 2, together with the ^{95}Zr inventories to 30 km attributed to the June 27, 1973 test, reported by Telegadas (1976). A line of regression through the decay-corrected HTO burdens indicates a residence half-time of about 13 months. Extrapolating this regression line back to December 15, 1973, when it was estimated that significant fallout of ^{95}Zr began (Telegadas, 1976), indicates a production of 18,400 kCi of HTO. The stratospheric inventory of ^{95}Zr showed about 5% above the sampling altitude of the aircraft. The HTO burden (to 20 km) at time of significant fallout was therefore increased by 5% for a total

Table 2. *Northern hemisphere tritium (as HTO) stratospheric burden to 20 km (kilocuries)*

Sampling period	Observed*	Decay-corrected† (to June 27, 1973)	Observed – background‡ (decay-corrected to June 27, 1973)
Apr. 18–May 6, 1975	9400	10500	7300
July 14–Aug. 5, 1975	4300	4800	2200
May 22–June 9, 1976	4900	5800	4300
Aug. 12–30, 1976	2200	2400	1300
Oct. 24–Nov. 17, 1976	3100	3800	2800

* Burden based on observed data (not decay-corrected).
† Observed burden decay-corrected to June 27, 1973.
‡ Observed burden* minus background (decay-corrected to June 27, 1973) where: background = assumed input of 600 kCi from the June 17, 1974 test had a stratospheric half-residence-time of 10 months, decay-corrected to time of measurements in 1975 and 1976.

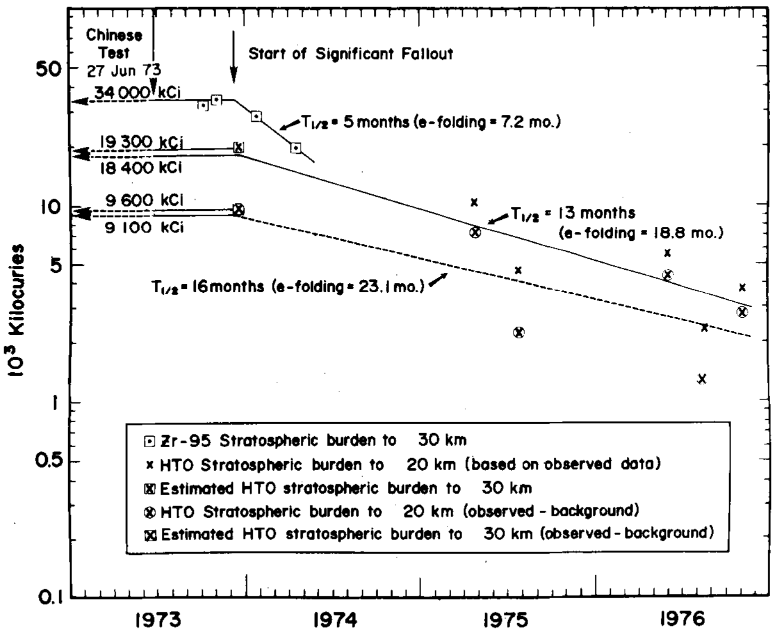

Fig. 2. Stratospheric half-residence-time of the ^{95}Zr and HTO burden for the Chinese test of June 27, 1973. All burdens decay-corrected to time of test. See text for a more complete description.

input of 19,300 kCi of HTO into the stratosphere. The ^{95}Zr burden to about 30 km indicated a 34,000 kCi input which is equivalent to a fission yield of 1.4 MT. The reported total yield for the June 27, 1973 event was 2 to 3 MT. Assuming a total yield of 2.5 MT would mean that this event had a fusion yield of about 1.1 MT. Dividing the HTO stratospheric input of 19.3 MCi by 1.1 MT indicates a production of 18 MCi/MT (fusion), not too different from that calculated for the November 17, 1976 test.

The stratospheric residence half-time of the ^{95}Zr burdens, Fig. 2, based on only two sampling periods, was 5 months. This residence time is shorter than that for the November 17, 1976 test but about the same as Telegadas (1974) found for the June 17, 1967 high-yield Chinese test between the sampling periods of February 1968 and April 1968. The ^{95}Zr burden attributed to the June 27, 1973 test could not be determined beyond April 1974 due to the fact that the next Chinese test of June 17, 1974 (total yield 0.2–1 MT) dominated the lower stratosphere of the Northern Hemisphere during the next sampling period in October 1974.

As mentioned previously, Leifer et al. (1976) estimated the fission yield of the June 17, 1974 test to be about 0.4 MT. When this event occurred, no mention was made of whether it was an all-fission or thermonuclear test. We will assume that this test was thermonuclear and it had a fission–fusion ratio of 1, that is, 0.4 MT (fusion). Assuming the HTO production of 15 MCi/MT (fusion) as was calculated for the November 17, 1976 test, this test would have injected 6000 kCi of HTO into the lower stratosphere of the Northern Hemisphere. It is further assumed that this HTO input had a stratospheric residence half-time of 10 months. This input was then decay-corrected to the 1975 and 1976 sampling periods listed in Table 2 and subtracted from column 1. Column 3, Table 2, is, therefore, the residual burden attributed to the June 27, 1973 high-yield test if the June 17, 1974 test were thermonuclear and had a fission–fusion ratio of 1.

The effect of assuming a fusion yield of 0.4 MT from the June 17, 1974 test upon the production estimate of the June 27, 1973 test is as follows: Fig. 2 indicates a stratospheric half-residence time of 16 months for the burden from the 1973 test. Extrapolating the burden back to the start of significant fallout and adding 5% to account for burden above the aircraft ceiling indicate an HTO production from the 1974 test of 9600 kCi, or 9 MCi/MT (fusion). This is half of the previous estimate which assumed no fusion yield from the 1974 test.

There are many uncertainties in estimating the stratospheric HTO inventories following the June 27, 1973 and November 17, 1976 events. One cannot determine unequivocally how much HTO was above the aircraft sampling altitudes or how much was transported into the Southern Hemisphere. The HTO inventories attributed to the June 27, 1973 test also had further uncertainties. Sampling did not start until about 2 years after the event; there was no overlap between the ^{95}Zr and HTO data, and there is the possibility that the June 17, 1974 test could have contributed substantially to the observed inventories calculated during 1975 and 1976.

There is more confidence in the estimated HTO burdens attributed to the November 17, 1976 test (Fig. 1) than the estimated HTO burdens from the June 27, 1973 test (Fig. 2). This is in part due to the fact that there was a partial overlap of the ^{95}Zr and HTO burdens starting 8 months after input. Further, the background HTO inventory based on measurements taken shortly before the November 17, 1976 event could be accounted for and subtracted from the observed inventories at later times.

The calculated residence half-times for particulate ^{95}Zr and gaseous HTO are approximately equal for the November 17, 1976 test, indicating that gravitational settling of particles in the lower stratosphere, to at least 20 km, possibly higher, for all practical purposes is negligible. A note of caution is in order—the HTO regression line in Fig. 1 is weighted toward the first calculated burden based on observations taken between July 6–22, 1977. If these samples were not taken and the questionable burden of October 1978 is not considered, a residence half-time of 15 months would have been calculated. One would then come up with the determination that particle settling is significant in transport processes assuming the initial distributions of particles and gases were the same. This points out that measurements at early times after a nuclear test are important in the determination of residence times, even though there is large spatial inhomogeneity.

4. Conclusions

The available HTO and ^{95}Zr data attributed to the November 17, 1976 Chinese high-yield test indicate that both the particulate ^{95}Zr and the gaseous HTO burdens were depleted from the lower stratosphere of the Northern Hemisphere at about the same rate. This would indicate that the input of particulates and gases from this event had about the same initial vertical distribution and that particle settling is negligible.

The burdens following the June 27, 1973 high-yield Chinese test have more uncertainties and should therefore be used with more discretion.

5. Acknowledgements

The support of this work by the Office of Health and Environmental Research, U.S. Department of Energy, is gratefully acknowledged. The University of Miami was supported under contract no. DE-AS05-76EVO3944. Dr. Robert Leifer of the Environmental Measurements Laboratory, U.S. Department of Energy, is Scientific Director of the High Altitude Sampling Program; Mr. Paul Guthals of Los Alamos National Laboratory, University of California, is Task Manager for Project Airstream. Mr. Thomas L. Barrow of the Lyndon B. Johnson Space Center, National Aeronautics and Space Administration, is the Aircraft Project Manager. Their efforts have made this work possible. The authors are grateful to Drs. H. G. Östlund, C. Rooth, L. Machta and E. Bauer for helpful discussions.

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