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The Conversion of Tritiated Hydrogen to Water in the Atmosphere

by
L. L. Burger

September 1976

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NOTICE

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SUMMARY

The report summarizes present information on the atmospheric reactions of tritium. The global distribution of hydrogen and of water are first considered. Data on tritium distribution are then compared and, finally, known reactions which may convert tritiated hydrogen-containing molecules are discussed.

Approximately 99% of the world's inventory of tritium exists as HTO. Although most of it is in the ocean, a significant portion still resides in the stratosphere. However, in the troposphere, which is the primary concern of this review, most of the tritium is in the form of HT, a smaller amount as HTO, and a much smaller but still significant amount as CH_3T. Further, the tritium-to-hydrogen ratio in the troposphere is higher in hydrogen and in methane than it is in water vapor.

The formation of HTO by exchange of HT or T_2 with water or by direct oxidation with oxygen, in the absence of catalysts, is extremely slow at concentrations in the atmosphere that might exist a few minutes after a tritium release. Photochemical oxidation may be the predominant conversion mechanism and over larger periods of time may combine with bacterial action to serve as the principal pathways of conversion of HT (or T_2) and CH_3T to HTO or other more reactive forms of tritium. The net conversion rate following a tritium release to the atmosphere would be expected to be less than 1% in 24 hr. This greatly lowers the immediate hazard of tritium releases over that based on the often used assumption that all releases soon become HTO. However, it does raise the question of a possible build-up of atmospheric HT and CH_3T in a fusion power economy which could utilize two or three orders of magnitude more tritium than the natural level.

The significance of the relatively high tritium content in atmospheric methane needs evaluation. Monitoring of CH_3T has been largely neglected in the past.

Considerable uncertainty exists in some of the data on which these conclusions are based and recommendations are made for further work.

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INTRODUCTION

The radioactive isotope of hydrogen (tritium-^{3}H or T) occurs naturally in the atmosphere as a product of cosmic radiation and, perhaps to a small extent, of solar fluxes. Before man-made tritium was introduced, its production rate and decay half-life ($t_{1/2}$) of 12.3 yr maintained an equilibrium world inventory of about 1.1×10^8 Ci (11 kg), 90% of which was in the oceans as HTO.

Tritium is also produced in the nuclear industry by fission and by neutron reaction with light atoms. The contribution from power reactors will equal the natural inventory by 1990 or 2000, perhaps leveling off at a factor of two or three times greater by the year 2010 or 2020 (Jacobs, 1968; Burger, 1976). Thermal weapons testing has dumped a huge amount of tritium into the reservoir, estimated by Eriksson (1965) at 1.7×10^9 Ci through 1962. Although most of this has been transferred to the oceans, the net effect is that tritium levels throughout the world are an order of magnitude greater than before weapons testing began. Even so, tritium is not considered a hazard at the present time. In spite of the ease of transfer of tritium to biologically important compounds, the half-life in life systems is short ($\sim$9 days in man). Further, there appears to be no evidence of any specific DNA effect, and the maximum relative biological effect (RBE) is taken as 2, and in fact may be smaller (Young, 1975; Hansborough and Draper, 1973). However, there are efforts to lower the acceptable limiting concentrations and the present regulations which permit environmental dispersal of tritium may be changed (USEPA, 1975).

The development of fusion power places tritium in a different light. The 1000 MWth conceptual fusion reactor of Hansborough and Draper will have an inventory of 3.2 kg of tritium and will consume about 0.15 kg or 1.5×10^6 Ci/day. It will breed about 10% more than it burns. Translated

to curies produced per GWe-yr, this is compared to tritium production in other reactors in Table 1. Conversion efficiency of 0.4 is assumed for the fusion reactor, LMFBR, and HTGR, and 0.3 for the LWRs. Thus, the management of $>10^6$ Ci/day through the complex cycle of the fusion power reactor may well be one of the more difficult problems.

TABLE 1. Tritium Production in Nuclear Reactors, Ci/GWe-yr

BWR	2.5×10^4
PWR	2.7×10^4
LMFBR	3.4×10^4
HTGR	2.4×10^4
HWR	6.0×10^5
Fusion Reactor	1.4×10^9

Most of the global transport studies of tritium and nearly all the environmental studies have been concerned with HTO. In fact, the assumption is nearly always made that if HT or T_2 is released, it is rapidly converted to HTO. The strength of this assumption is very doubtful and, in fact, the basic chemistry of hydrogen would predict the opposite. This is clearly an important question since the biological effectiveness of tritium in its elemental form is some 200 times less than as HTO, and since T_2 is probably the most likely release form. If the conversion time is $>>t_{1/2}$, then there are two aspects that may be considered: 1) the hazard is lower because of the relative biological inertness of HT compared to HTO, and 2) the ocean is not the effective sink for HT that it is for HTO; thus, very large releases of tritium gas could appreciably raise the atmospheric radioactivity.

Many available reviews consider tritium technology and problems. The first comprehensive treatment is that of Jacobs (1968). Others have been cited in a recent shorter summary by the author (Burger, 1972). The 1971

Tritium Symposium at Las Vegas covered a wide cross-section of tritium topics (Moghissi, 1973). With regard to the fusion reactor, the excellent work of Hansborough and Draper (1973) must be cited. Earlier papers which surveyed tritium problems in fusion reactors include those by Watson (1972) and Hickman (1972). More recently tritium technology as related to fusion was reviewed at Symposia held at the Mound Laboratories in 1974 (Smith, 1975) and at Gatlinburg in 1975 (Watson, 1976). The tritium hazard problem from a large fusion power plant is reviewed in a LASL-ANL environmental impact study (Draley, 1975). Further general information can be found in a fusion reactor information requirements study by Young et al. (1975).

The purpose of this study is to review and assess the information available on reactions of tritium in the atmosphere. Since fusion reactors employ tritium in elemental form the basic question is the rate of oxidation to water or exchange with water. Other reactions will be considered as well as other tritium-containing molecules.

DISTRIBUTION OF HYDROGEN

Before considering the reactions of tritium, it is important to consider the global distribution of tritium and hydrogen. Table 2 shows the distribution of water throughout the world. The annual turnover of water is about 3.5×10^{14} m^3, of which about 3.4×10^{13} m^3 represents water returned to the oceans by rivers. Thus, approximately 2 to 3% of the world's water is in circulation within a time frame significant with respect to the half-life of tritium (12.3 yr). The great bulk of the water lies below the thermocline and is effectively fixed for thousands of years.

Most of the water in the atmosphere exists in the troposphere. As will be seen, however, the stratosphere also plays an important role in the tritium distribution. Table 2 lists the water inventory and distribution. The amount listed for the stratosphere is computed on the basis that the water concentration is about 3 volume parts per million (vppm) and that the pressure equals 1/4 atmosphere at the tropopause. The value for the stratosphere may be low, since many measurements indicate an increasing H_2O content with increasing altitude in the stratosphere (Machta, 1971; Brown, 1961).

TABLE 2. Distribution of Water[a]

	Volume, m^3	
	Exchangeable	Fixed
Oceans	top 100 m,[b] 3.6×10^{16}	Total ocean - 1.3×10^{18}
Lakes	1.2×10^{14}	--
Rivers	1.2×10^{12}	--
Ice Caps	--	$2.5\text{-}2.9 \times 10^{16}$
Soil Moisture	2.5×10^{13}	--
Lower Ground Water Table	--	4.1×10^{15}
Atmosphere below 11 km	1.3×10^{13}	--
Atmosphere above 11 km	5×10^{6}[c]	--

(a) Data from Franks (1972).
(b) Mixing time $< t_{1/2}$ ^3H. The thermocline varies from 50 to 200 m; arbitrarily set at 100 m for this calculation.
(c) See text.

The hydrogen distribution is quite different, and its behavior in the atmosphere is more controversial. The atmospheric concentration is normally placed at about 0.5 vppm. The most recent measurements (Ehhalt, 1975) up to 30 km indicate that the volume mixing ratio is fairly constant at about that value but increasing very slightly as the tropopause is reached. The concentration in the stratosphere is less certain. Scholz (1970) found 0.40 ± 0.01 vppm in a sample collected between 44 and 62 km. Small but statistically significant variations between the concentrations found in the northern and southern hemispheres, and between surface air and upper troposphere air indicate considerable contributions from industrial hydrogen (Schmidt, 1974). Schmidt's values are slightly higher (0.575 vppm) for the northern hemisphere. Fixed sources of hydrogen include decay of organic matter by anaerobic bacteria, photo dissociation of H_2O, photolysis of CH_4, and solar protons. Hydrogen is removed by photo oxidation, bacteria (protoplasm synthesis, and nitrite and amino acid reduction), and by escape. The rate of planetary escape is small (Hunter 1973, 1974); the planetary half-life of hydrogen atoms is estimated at 10^4 yr. However, it does place hydrogen in a unique position among atmospheric gases. Careful analysis of the deuterium and tritium content of hydrogen from air samples and consideration of the upward flux has within the last few years done much to clarify the behavior of hydrogen in the atmosphere.

The molecular hydrogen content of surface waters of the North and South Atlantic was found by Schmidt to vary from (0.8-5.0) x 10^{-5} ml/ℓ H_2O. The latter value is in excess of saturation and supports H_2 production by bacterial action. The measurements and budget calculations made by Schmidt (1974, 1975) indicate the present state of knowledge. His calculations lead to a production rate of 3.65 x 10^{13} g H_2/yr, and measurements suggest an atmospheric reservoir of 2.04 x 10^{14} g. (Assuming a saturation value of 1.0 x 10^{-5} ml/ℓ for H_2 in water, one calculates the top 100 m of ocean would contain 3 x 10^{14} g of dissolved H_2). Schmidt also calculates that up to 1.2 x 10^{13} g H_2/yr may be removed from the troposphere by soil bacteria.

The only other hydrogen-containing molecule in significant quantity in the atmosphere is methane. It is also produced by bacterial action. Schlegel (1974) has examined the CH_4 production and the relation to H_2 production and utilization. Scholz (1970) found that its concentration varied from about 1 vppm in surface air to ≤0.05 at the stratopause. Later measurements at NCAR by Ehhalt and coworkers (1975) indicated a constant level at 1.5 vppm through the troposphere, and a rapidly decreasing amount with height through the stratosphere. Its decreasing concentration with height in the stratosphere presumably results from oxidation to water. Scholz considers that it is virtually completely oxidized at 44 km. Ehhalt (1974a) has recently discussed the methane cycle and found a mixing ratio of 1.4 vppm in the troposphere and confirmed the decreasing stratospheric concentration with altitude. He suggests, however, that only 10% is destroyed in the stratosphere, most of it being destroyed in the troposphere through the reaction $CH_4 + OH \rightarrow CH_3 + H_2O$. His postulated turnover time for methane is 4 to 7 yr compared to approximately 100 yr originally listed by Israël (1973).

Table 3 shows the computed distribution of hydrogen in water, hydrogen, and methane. All three compounds will be of interest in studying tritium behavior. The hydrogen in organic compounds (other than methane) including life systems is ignored in this distribution. This should not imply that all such compounds are irrelevant to the HT-HTO cycle.

TABLE 3. Distribution of Hydrogen in Compounds Occurring in the Atmosphere

	As H_2O		As H_2		As CH_4	
	Amount of H_2, g	Res Time yr, (f)[a]	Amount of H_2, g	Res Time yr, (f)	Amount of H_2, g	Res Time yr, (f)
Troposphere	1.4×10^{18}	0.022 (1.3)	1.5×10^{14}	6.5 (2)[b]	5×10^{14}	4 (3),[b] 5.3 (1.3)[c]
Stratosphere	3×10^{14}	--	5×10^{13}	--	$\sim10^{14}$	--
Rivers	1.3×10^{17}	--	--	--	--	--
Soil Moisture	2.8×10^{13}	--	--	--	--	--
Top 100 m Ocean	4.0×10^{21}	--	3×10^{14}(est)		--	--

(a) (f) = uncertainty factor
(b) Junge (1974)
(c) Ehhalt (1974a, 1975)

TRITIUM DISTRIBUTION

Having examined water and the hydrogen distributions throughout the world, it is now of interest to consider the tritium concentrations. The naturally occurring tritium leads to a H/T ratio of about 10^{18} or one TU.[a] This amount, about 10^8 curies, or $\sim$10 kg, is swamped by the tritium added by fusion bomb tests which amounted to about 1.7×10^9 Ci by 1962. (Tritium from underground tests is not included.) Additional sources are leakage from military and industrial applications, and fission tritium produced in the nuclear power industry. The latter amounts to about 2.5×10^4 Ci/GWe-yr for LWRs. Thus, the tritium inventory has been greatly perturbed over the last two decades, and some uncertainties still exist as to the proper description of "natural" and "man-made" tritium distributions. It has been the subject of extensive study, however, and the distribution and behavior of tritiated water are reasonably well understood. Information on other hydrogen-containing species such as H_2, (HT) or CH_4, (CH_3T) is not as complete.

The natural tritium cycle can be described as: formation in the upper troposphere and stratosphere, photo-oxidation to HTO, exchange with surface water and removal by rainfall, final deposition in the ocean. Some is returned to the atmosphere by evaporation, and the decay half-life of 12.3 yr establishes the equilibrium inventory. Thus, water plays the major role in the tritium inventory turnover and, of course, is the major mode of entry into biological systems. However, other hydrogen compounds are important in assessing tritium behavior. Jacobs (1968) indicates the uncertainty that existed in the natural tritium distribution and tabulates Begemann's (1963) values as shown in Table 4.

Most of the bomb tritium was apparently deposited in the stratosphere as HTO. Extensive studies have been made of the fallout as related to latitude, seasons of the year, wind and rainfall patterns, and environmental

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(a) 1 tritium atom/10^{18} H atoms is referred to as the tritium unit or TU.

TABLE 4. Distribution of Natural Tritium (Begemann, 1963)

		%T	TU
Hydrosphere		90	≤10
Troposphere	H_2O	0.1	≤80
	H_2	0.02-0.2	4×10^3 - 3.5×10^4
	CH_4	≤0.04	$\leq10^3$
Stratosphere	H_2O	∿10	6×10^5
	H_2	0.004-0.007	3×10^3 - 6×10^4

holdup. Again, most of the studies have been restricted to HTO, although in recent years accurate measurements have been made on hydrogen and methane. The HTO remains in the stratosphere for an appreciable period. Brown and coworkers (1961) assumed a residence time of 5 yr, while Eriksson (1965) estimated the average residence time at 3 yr. HTO leaks out of the stratosphere at a variable rate, being much greater at the tropopause discontinuities and in the Spring. There is also some evidence of backmixing into the stratosphere (Smith, 1968). A discussion of these measurements is beyond the scope of this paper, but it is worth noting that they have been of great value in meteorological studies. The high concentration of HTO still in the stratosphere (Table 5) would seem to argue in favor of a longer residence time than 3 to 5 yr. Recent reviews may be represented by papers of Ehhalt (1971), Östlund (1974a), Michel (1975), (1975), Koranda (1972), and Schell (1970), as well as by earlier work of Begemann and Friedman (1959).

The HTO content of water is highly variable ranging from <1 TU in vapor above the ocean to $>10^6$ TU in the stratosphere. In the troposphere, where the range of water content is from 0.1% to several percent at the surface of the earth, another variable (the humidity) is introduced. Although details of the HTO distribution are not important to the present discussion, it is necessary to consider briefly the HTO concentrations in both the stratosphere and troposphere.

As mentioned earlier, most of the weapons-test tritium was deposited in the stratosphere as HTO. The amount remaining still greatly exceeds that produced by cosmic rays. Scholz (1970) examined the H_2O and HTO content near the stratosphere, and found HTO in the range from 22 to 80 x 10^6 TU corresponding to 3 to 10 vppm H_2O. This calculates to be about 8.9 x 10^3 HTO molecules/mg air. Brown (1961) found 9.0 x 10^3 HTO molecules/mg air at 80 to 90,000 ft (27.4 km). Both measurements were at medium north latitudes. Östlund's measurements in the arctic stratosphere show a much lower concentration (Östlund, 1974a), an average of about 200 HTO molecules/mg air with a marked increase as the altitude increased. This variation with altitude and latitude is well known, as are the higher tritium levels in the northern hemisphere. The mean value for stratospheric air is probably between 1 x 10^3 and 8 x 10^3 HTO molecules/mg air. A "guess" of 2 x 10^3 would place the stratospheric HTO inventory at the present time at 2.5 x 10^{27} HTO molecules, 12 kg tritium, or 1.2 x 10^8 Ci.

Data for tropospheric HTO are more extensive. Detailed monitoring by Ehhalt and coworkers (1971, 1973, 1974), and Östlund and coworkers [1973, 1974b, 1976[(a)]] show a wide range of concentrations from a few molecules/mg air to several hundred. The T/H ratio varies over four orders of magnitude because of changing humidity. Location of the sampling stations is also very important. Östlund's data, which appear to be generally lower than Ehhalt's, include both absolute humidity and absolute tritium data so that both TUs and molecules/mg air can be compared. It is difficult, however, to estimate a mean level for the troposphere. A concentration of 20 molecules/mg air would correspond to a troposphere inventory of 4 x 10^6 Ci. This number is used in Table 5 which summarizes the tritium partition.

Analytical data on isotopic composition of hydrogen gas were confusing until it was recognized that surface atmospheric samples were contaminated by industrial hydrogen (depleted in deuterium) and had to be corrected to the standard mean deuterium content (Begemann, 1959). The second bit of

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(a) The 1976 paper is representative of the Progress Reports of the Rosenstiel School of Marine and Atmospheric Science, University of Miami. The reports contain detailed monitoring data of atmospheric tritium.

TABLE 5. Approximate Tritium Partition

	Air Mass, g	HTO, Ci	HT, Ci	CH_3T, Ci
Stratosphere	1.3×10^{21}	1.2×10^{8}	3×10^{6}	$<10^{5}$
Troposphere	3.8×10^{21}	4×10^{6}	1×10^{7}	3×10^{5}
Ocean (surface)	--	9×10^{8}	See Text	

confusion resulted from many observations of a nonuniform tritium content (after the above correction) of tropospheric H_2. Variations occurred both with time and with geographical location. Initial assumptions appear doubtful that the changes, which were accompanied by an overall increase [a doubling time of about 2 yr (Gonsior, 1966)], were the result of atmospheric nuclear explosion tests. There was no direct correlation between test times and tritium increases in H_2 samples. (Such a correlation has been demonstrated for tritium in HTO). Ehhalt (1966) summarized these measurements and concluded that the source was nuclear testing, but that the HT was injected into the upper levels of the stratosphere. This resulted in a long mixing time into the troposphere compared with the intervals between nuclear tests, but less than 10 years. Östlund (1974a) discounts atmospheric testing and places the source at ground level. Tropospheric samples isolated from industrial activity (Friedman, 1974) suggested that by 1967 the concentration had reached a steady value, being constant at 2×10^{6} TU over a 2-yr period.

It is pertinent to note that even before atmospheric testing the T/H ratio in hydrogen was observed to be higher than in water. Harteck's (1954) classic paper explained this on the basis of competitive dissociation of TO_2 radicals compared with other reactions which form HTO.

Other fluctuations have been recorded by many workers. Layton (1969), Östlund and coworkers, and Martin and Hackett (1974) have made careful measurements over extended periods of time. Short time variations of an order of magnitude have been observed which are apparently the result of other anthropogenic activity (both military and industrial). Martin and Hackett found that after each excursion the level returned to 80 T atoms/mg

air ($\sim 4 \times 10^6$ TU). This is somewhat higher than indicated by the extensive monitoring data of Östlund which seems to suggest a level of around 40 to 50 HT molecules/mg air. Using Östlund's (1973) mean concentration of 44.6 tritium atoms/mg air in the atmosphere the amount of tritium as HT calculates to be 1.3 kg (1.3×10^7 Ci) of which 75% is in the troposphere.[(a)] Since this appears to be reasonably constant at the present time the decay rate of 5.5%/yr plus a roughly equal amount removed by microbial conversion suggests that over 100 g, 10^6 Ci, of HT are injected into the atmosphere each year.

As noted previously, the methane concentration is relatively constant throughout the troposphere at about 1.5 vppm, then decreases rapidly with altitude in the stratosphere, presumably the result of oxidation. Methane is produced by biological action with small contributions from industry. Thus, it was somewhat surprising to find that the tritium content of atmospheric methane was higher than could be explained by the tritium content of the natural methane precursors. It is known that the T/H ratio increased at about the same rate as for the early increases in tritium content of H_2. Speculation has included: direct tritiated methane production in a reducing core of a thermonuclear explosion (Wolfgang, 1961) and exchange of HT with CH_4 (Martell, 1963). Cadle (1967) investigated Martell's suggested mechanism which involved CH_2 and concluded that this was not a likely production path. Begemann and Friedman (1968), and Haines and Musgrave (1968) concluded that the best explanation was leakage of tritiated methane from industrial sources. Begemann and Friedmann found relatively constant concentrations of $\sim 2 \times 10^4$ TU from 1958 to 1963, preceded by the aforementioned exponential increase from about 1953 to 1958.

Using 2×10^4 TU and the data from Table 3, it can be seen that the amount of tritium as triated methane is approximately 30 g or $\sim 3 \times 10^5$ Ci. The high tritium content of methane is important with respect to the atmospheric chemistry of hydrocarbons. Also, it could well turn out to be

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(a) This number is possibly high since concentrations in the southern hemisphere are appreciably lower.

significant with respect to the tritium release-tritium conversion problem. If CH_3T release or leakage accompanies HT release or leakage in any appreciable fraction, then it must be considered a potentially significant source of HTO and perhaps a source of other important tritiated species including HT (see sections on biochemical and photochemical reactions).

It is important to note here that the usual methods of monitoring for tritium will not detect CH_3T. The usual and very reliable method involves: 1) removal of water for counting, and 2) catalytic oxidation of HT (usually after adding a small amount of H_2) and again counting the water removal. The mild oxidation at ambient temperature does not convert the CH_3T to water. It is, however, a relatively simple matter to add a third high temperature oxidation step for the methane and other hydrocarbons.

No data have been found for the HT content of ocean water. If it is produced by bacterial action, the tritium ratio of the dissolved hydrogen may be roughly the same as in water. Atmospheric moisture presumably exchanges rapidly with the ocean surface. The T/H ratio in dissolved hydrogen should be an important number for the study of hydrogen sources and sinks.

The rough distribution estimates made in Table 5 are in fair agreement with the earlier calculations of Begemann (Table 4). However, the T/H ratios obtained from the present estimates are an order of magnitude higher. The approximate percentage distribution is listed in Table 6. The relative

TABLE 6. Approximate Percentage Distribution of Tritium

Location	%
HTO in ocean (~top 100 m)	90
HTO in stratosphere	10
HT in troposphere	1
HTO in troposphere	0.3
HT in stratosphere	0.3
HT as CH_3T	0.03

values of the last five numbers in the table are the most significant since the amount attributed to the ocean in the present calculation is obtained by difference, using the estimated present tritium inventory.

With the above approximate picture of the present tritium distribution, which is neither equilibrium[a] nor steady-state, and the observation that the T_2 inventory in a single 1 GWe fusion reactor is greater than the world HT inventory, we now consider the tritium compounds that may be released and their reactions. Of primary interest are the reactions of T_2 or HT leading to HTO. However, other reactions may be significant.

(a) It should be emphasized that neither are the <u>total</u> hydrogen and water in equilibrium in the atmosphere.

Studies made on both HT and T_2 decay indicate that the HHe^+ and THe^+ species are by far the most abundant (Wexler, 1959). The other fragments indicate that the subsequent dissociation goes by both routes:

$$H^3He^+ \rightarrow {}^3He^+ + H \quad (3)$$

and

$$H^3He^+ \rightarrow {}^3He + H^+. \quad (4)$$

The former predominates at least in the case of HT in spite of the fact that it is more endothermic (Cacace, 1970a). The THe^+ ion appears to be more stable than the HHe^+. Cacace (1970b) has published a comprehensive review of the decay products and reactions of tritiated molecules including hydrocarbons. His review also includes the properties and reactions of the THe^+ ion. The heat from THe^+ formation is estimated to be 320 kcal/mole. Careful studies of the reaction with methane were made showing that CH_3T is the principal product.

Reactions of THe^+ with H_2 have not been reported. It seems reasonable to expect that reactions such as

$$THe^+ + H_2 \rightarrow TH + H^+ + He \quad (5)$$

or

$$THe^+ + H_2 \rightarrow TH + H + He^+ \quad (6)$$

should lead to appreciable HT formation due to the lower zero point energy of HT compared to H_2.

Information on the reaction with liquid water was obtained by Stöcklin and Cacace (1976) in experiments designed to study the tritiation of thymadine and deoxyridine in aqueous solution. T_2 was compared with HT with reaction times long enough to produce measurable amounts of THe^+ decay. It was found that T_2 was more effective than HT by a factor of about 12 at concentrations low enough that radiation induced mechanisms are not important. With a 4.5 wt% thymadine solution the yield of HTO was 0.6 per THe^+ ion formed. Ht was also formed.

The recoil energy of the $^3HeT^+$ ion was first employed by Wilzbach (1957) for tritium labeling of organic compounds.

No information on gas phase reactions between THe^+ and H_2O is available. It is clear, however, that at concentrations below those where

radiation is no longer the dominant factor, the reactions of THe^+ can contribute significantly to formation of other tritiated molecules.

T_2(HT) + H_2O EXCHANGE

The water-hydrogen exchange reaction has been utilized for isotopic enrichment of deuterium. Equilibrium favors the heavy isotope in the water phase. For the gaseous reaction

$$HD + H_2O \rightleftarrows HDO + H_2 \quad (D) \tag{7}$$

and

$$HT + H_2O \rightleftarrows HTO + H_2. \quad (T) \tag{8}$$

The equilibrium constants are

$$K_D = 3.87 \text{ at } 298°K; \ 2.47 \text{ at } 398°K \tag{9}$$

and

$$K_T = 6.25 \text{ at } 298°K; \ 3.40 \text{ at } 398°K. \tag{10}$$

In the absence of catalysts, the reaction does not proceed (Horiuti, 1933). However, a variety of materials, many metals and metal oxides in particular, permit more rapid equilibration especially at the higher temperatures. These reactions were utilized at the Trail, B.C. plant until 1956 to separate deuterium and the process is described by Murphy (1955). The method was reviewed by Levins (1970) and is still under development for both deuterium and tritium separation (Léger, 1970), (Damiani, 1972), (Stevens, 1973), (Bixel, 1976). Rolston and Goodale (1972) recently examined the mechanism for exchange using supported platinum catalysts. Above 100°C the reaction was free of mass transfer effects, but below 100°C the rates suggested that the adsorption of water was the rate limiting step.

Radiation is an efficient catalyst and the early work of Yang and Gevantman (1960) appears to adequately define the mechanism. Using tritium diluted with helium, they found a rate of formation of tritiated water,

$$\frac{dC}{dt} = 1.7 \times 10^{-3} \ (Co)^2, \ Ci \ \ell^{-1} hr^{-1} \tag{11}$$

where Co is the initial T_2 concentration in Ci/ℓ over the range 0.05 to 0.7 Ci/ℓ. The rate was independent of the water content. The radiation

interpretation was supported by X-ray irradiation tests which showed that the exchange was proportional to energy absorption.

Because of the low β energy, the equation cannot be used with certainty at concentrations much below those of the experiment. Assuming that it would hold at 10^{-3} Ci/ℓ (energy deposition $\sim 10^{15}$ ev $\ell^{-1}hr^{-1}$) about 0.004%/day conversion to water would be predicted.

At higher concentrations the above kinetic equation can be used to approximate the HT or $T_2 + H_2O$ exchange rate. The second order tritium dependence is a result of the first power radiation energy dependence. If other radiation sources are present, these can be added to the energy factor in the equation. For large tritium releases the exchange reaction could be significant for the first few minutes. However, in practice rapid dilution may occur as a simple result of plant design. For the accidental release of 479,000 Ci of T_2 from the Savannah River Plant in 1974 (Marter, 1974) the dilution in the stack, which had a discharge rate of 135,000 ft^3 min^{-1}, gives a concentration of 0.03 Ci/ℓ. For this, Equation (11) predicts about 0.005% conversion/hr. (The dilution factor after about 1 hr was estimated to be 10^6 and measured at 5 hr was about 10^{10}. Analytical samples indicated <0.2% of the tritium to be HTO).

The experimental data of Yang and Gevantman utilized tritium concentrations in the mole fraction range of 10^{-5} to 10^{-3}. Further information on the mechanism for the radiation induced exchange has been provided by the work of Baxendale and Gilbert (1964), by Firestone and coworkers (1957, 1972), and Bibler and Firestone (1972) who studied the deuterium-water exchange at much higher hydrogen (or deuterium) concentrations.[a] Briefly, Bibler and Firestone found that the G value for the exchanged hydrogen (e.g., HD for the $D_2 + H_2O$ reaction) was constant over an appreciable temperature range but increased rapidly above a certain temperature, and that the rate for the latter condition depended upon the water vapor concentration. The transition point is below 200°C. The chain mechanism

(a) "Hydrogen" is used here as a generic term to include any of the diatomic isotopic species.

which is involved at the higher temperatures was shown to involve hydrogen atoms. Radiation induced reactions are in fact similar to the atmospheric photochemical reactions which are discussed in a later section.

While the experiments do not relate directly to reactions of T_2 or HT in the atmosphere, knowledge of the mechanisms is useful for extrapolation to other conditions. The hydrogen-water reaction is important because of the great excess of atmospheric moisture over hydrogen. Thus, at equilibrium, essentially all the tritium will exist as HTO. However, it is seen that in the absence of very effective catalysts or radiation, this would require several years. The fact that most of the tritium in the troposphere is in the form of HT and not as HTO indicates the absence of such a catalyst.

It is often difficult to separate exchange with water from oxidation reactions. This topic is considered in the next section.

OXIDATION OF T_2

Like the previous reactions, the reaction between hydrogen and oxygen is negligible at ordinary temperatures unless catalyzed. The same catalysts, as employed above, are generally effective, in particular the noble metals of group VIII.

Eakins and Hutchinson (1973) carried out a study of metal surfaces on the oxidation rate. Elemental tritium was mixed with wet or dry air, and the effects of mild steel, brass, platinum, and aluminum surfaces noted. The reaction vessel employed a catalyst area of 90 cm^2 in a 250 ml bulb. The rate constants are shown in Table 7.

The surface-to-volume ratio is roughly that of a 20 x 20 x 10 ft laboratory. They calculate that half the tritium at an initial concentration of 3.4×10^{-2} Ci/m^3 in such a room lined with platinum would be converted to water in 4 yr. Again, dilution is very important. The conversion half-life for a second-order reaction is $1/(kCo)$, and for the SRL release discussed above (before dilution) the half-time for conversion would be about 20 hr. However, even under these highly unlikely conditions the conversion rate is small, and only under very unusual circumstances could catalyzed oxidation contribute much to HTO formation from a T_2 release.

TABLE 7. Effects of Metal Surfaces on Tritium Oxidation

Metal	Rate Constant, $\ell\ Ci^{-1}hr^{-1}$	
	Dry Air	Humid Air
None	5.6×10^{-4}	2.9×10^{-3}
Brass	6.0×10^{-4}	8.6×10^{-2}
Steel	4.2×10^{-3}	6.1×10^{-2}
Aluminum	3.9×10^{-3}	9.6×10^{-3}
Platinum	2.4×10^{-2}	8.6×10^{-1}
Brass, oxidized		6.5×10^{-2}
Steel, oxidized		7.4×10^{-2}

The radiation-induced reaction has been studied by Dorfman and Hemmer (1954), by Casellato (1962), and by Belovodskii (1975). Dorfman and Hemmer studied the reaction between T_2 and O_2 by direct pressure measurement, and found first-order kinetics with $k = 1.19 \times 10^{-4}$ min^{-1} over a concentration range of 94 to 324 Ci/ℓ. Casellato and coworkers examined the reaction at lower concentrations. Below 1 Ci/ℓ, they observed second-order kinetics with the rate constants $k = 1.2 \times 10^{-3}$ $\ell\ Ci^{-1}hr^{-1}$ in O_2 and 0.62×10^{-3} $\ell\ Ci^{-1}hr^{-1}$ in dry air. The presence of water vapor in O_2 increased the rate by a factor of 3. The latter rate constant is seen to be in general agreement with that of Eakins and Hutchinson (1973).

Belovodskii and coworkers (1975) repeated some of the experiments with the system tritium - dry air - water vapor, and using a wider concentration range, 10^{-3} Ci/ℓ to 600 Ci/ℓ. They found no difference between moist air and experiments repeated in dry argon and fitted the data by a 5/3 concentration dependence.

$$\frac{d(HTO)}{dt} = 10^{-6} Co^{5/3},\ Ci\ \ell^{-1} min^{-1} \qquad (12)$$

From the lack of moisture effect, they further concluded that the oxidation mechanism rather than exchange was dominant. The data presented earlier from exchange and oxidation studies lead to the opposite conclusion, and it is likely that exchange is generally a greater contributor than is oxidation.

Further experimental data under a variety of specific conditions covering a wider concentration range may be necessary to fix the importance of the two processes and to better define the mechanisms.

REACTIONS OF METHANE

The general reactions and abundance of methane are discussed in a previous section as was the reaction with T_2 via the THe^+ mechanism. However, the surprisingly high T/H ratio in atmospheric methane raises questions concerning the source as well as the ultimate fate of tritiated methane that must be considered.

The studies of Cacace (1970b) indicated the efficacy of the THe^+ mechanism. Earlier work of Kandel (1964) on the radiation-induced reaction showed that the direct combination produced a maximum CH_3T concentration in about 12 hr with peaks in the CH_2T_2 and CHT_3 concentrations at 25 and 50 hr, respectively.

It appears likely that tritiated methane is a by-product of any laboratory or industrial work involving T_2 or HT, and in fact cannot be ruled out as being formed in some situations with the genesis of the T atom. The situation is in many ways reminiscent of the methyl iodide problem--whenever studies of radioactive iodine are carried out, methyl iodide is formed. In almost any real situation there is sufficient organic matter present (grease, paint, plastic, etc.) that CH_3I (and CH_3T) are formed. The question of how extensive this reaction is in the case of tritium is yet to be answered.

The sequence of reactions and products involved in its oxidation are also of interest. Aside from the biological reactions considered in the next section, the principal fact that is apparently established at the present time is that methane is oxidized in the high stratosphere to water. According to Ehhalt (1974a) this involves 10% of the methane. He postulates that the most probable destruction mechanism in the troposphere is the reaction $CH_4 + OH \rightarrow CH_3 + H_2O$ which is tied to the CO and H_2 cycles via peroxide and aldehyde intermediates. Ehhalt's CH_4 turnover time is

4 to 7 yr (Table 3). Koyama (1963, 1964) who carried out very extensive investigations of methane production in agricultural areas estimates the lifetime of CH_4 as 16 yr. Tropospheric reactions such as those suggested by Ehhalt are obviously very important to fixing the residence time of CH_3T. It would appear that CH_3T studies may offer considerable information on the global methane pathways.

Reference should be made to the tests made on nuclear stimulation of natural gas wells. The chemical reactions occurring have been studied in some detail for two events: 1) the Gasbuggy test which was a 29-kton explosion at a depth of 1292 m, and 2) the Rulison test which was a 40-kton explosion at 2573 m. Most of the tritium produced was as HTO with small amounts as HT and CH_3T. Smith (1970) and Taylor (1970) have reviewed the chemical results. While those results do not bear directly on the problem of CH_3T reaction in the atmosphere, a further study of methane reactions should include an evaluation of those experimental data. Weed (1972) has compiled an excellent literature summary of methane reactions with tritiated water.

The water-CH_3T exchange reaction has been considered for purification of the released gas (Frink and Wethington, 1971). No observable reaction was observed in the absence of catalysts.

BIOCHEMICAL REACTIONS

Water, THO, is readily assimilated by biological systems and will not be discussed here. The remaining compounds of interest in the atmosphere are hydrogen and methane. Hydrogen is both consumed and produced by soil- and water-contained bacteria. The rates suggested by Schmidt (1974) were mentioned in the section on hydrogen. The reactions were first discussed by Smith and Marshall (1952) who indicated that low concentrations of an enzyme hydrogenase could cause oxidation of tritium. Smith and coworkers (1953) found that intestinal bacteria from rats were effective in oxidation of tritium in the animal body.

Schlegel's review (1974) indicates the paths that involve hydrogen, methane and other key trace gases. Under aerobic conditions hydrogen and methane are quickly oxidized by microorganisms. However, under anaerobic conditions methane is the principal product (besides CO_2), others being N_2O from NO_3^- reduction and H_2S from SO_4^{-2} reduction. As was indicated earlier such processes should (assuming no highly specific isotopic selectivity by bacteria) produce CH_3T with the same T/H ratio as the water environment. The higher value found suggests a direct anthropogenic source of CH_3T and a considerable stability.

Under aerobic conditions bacteria can either oxidize methane or utilize it as a growth substrate. Schlegel points out that these bacteria are highly specific in the sense that they do not use the methyl group on long chain hydrocarbons.

Thus, bacterial action can oxidize both HT and CH_3T to water and in addition may produce growth substrates containing tritium. One of the questions of interest concerns the possible isotopic selectivity of bacteria for both hydrogen and methane production and destruction.

PHOTOCHEMICAL REACTIONS

These are probably the most complex of the atmospheric reactions because they involve a myriad of chain reactions which, in turn, involve free radicals and tie hydrogen and tritium to the very important carbon, nitrogen, and sulfur cycles. No attempt will be made here to survey this field but several of the important reactions need to be discussed. The nitrogen and sulfur reactions will be omitted except as they become directly involved in the CH_4-H_2-H_2O cycles.

The principal primary photochemical steps involve oxygen:

	Reaction	Min. Energy[a]
p 1	$O_2 + h\nu \rightarrow O(^3P) + O(^1D)$	175.0 nm (7.05 ev)
p 2	$O_2 + h\nu \rightarrow 2\ O(^3P)$	242.4

(a) Hunt, 1966.

p 3	$O_3 + h\nu \rightarrow O_2 + O(^1D)$	310.0
p 4	$O_3 + h\nu \rightarrow O_2 + O(^3P)$	1,180.0
p 5	$H_2O_2 + h\nu \rightarrow 2\ OH$	565.0
p 6	$H_2O + h\nu \rightarrow OH + H$	239.0
p 7	$H_2CO + h\nu \rightarrow H_2 + CO$	360.0
p 8	$H_2CO + h\nu \rightarrow H + CHO$	
p 9	$CH_4 + h\nu \rightarrow CH_2 + H_2$	
p 10	$CH_4 + h\nu \rightarrow CH_3 + H$	

These lead to a variety of other reactions involving free radicals and virtually all the reactive trace gases in the atmosphere. Some of the important ones are:

R1	$OH + O_3 \rightarrow HO_2 + O_2$
R2	$OH + CO \rightarrow CO_2 + H$
R3	$H + O_2 + M \rightarrow HO_2 + M$
R4	$HO_2 + NO \rightarrow NO_2 + OH$

R5	$OH + CH_4 \rightarrow CH_3 + H_2O$
R6	$CH_3 + O_2 + M \rightarrow CH_3O_2 + M$
R7	$CH_3O_2 + NO \rightarrow NO_2 + CH_3O$
R8	$CH_3O + O_2 \rightarrow CH_2O + HO_2$
R4	$HO_2 + NO \rightarrow NO_2 + OH$

R9	$CH_4 + O(^1D) \rightarrow CH_3 + OH$
R10	$CH_3O_2 + NO_2 \rightarrow CH_2O + HNO_3$

R11 $CH_3O_2 + NO \rightarrow CH_2O + HNO_2$

R12 $CH_2O + OH \rightarrow HCO + H_2O$

R13 $CHO + O_2 \rightarrow CO + HO_2$

R14 $CO + HO_2 \rightarrow CO_2 + OH$

R15 $H_2 + OH \rightarrow H + H_2O$

R16 $O(^1D) + H_2O \rightarrow 2\ OH$

R17 $O(^1D) + H_2 \rightarrow H + OH$

R18 $HO_2 + OH \rightarrow H_2O + O_2$

R19 $OH + OH \rightarrow H_2O + O$

R20 $HO_2 + HO_2 \rightarrow H_2O_2 + O_2$

R21 $H_2O_2 + OH \rightarrow HO_2 + H_2O$

R22 $OH + CO \rightarrow CO_2 + H$

R23 $OH + O_3 \rightarrow HO_2 + O_2$

R24 $OH + NO + M \rightarrow HNO_2 + M$

R25 $H + HO_2 \rightarrow H_2 + O_2$

The list is incomplete but serves to illustrate both the complexity and importance of the photochemical reactions. Reactions R1 through R4 make up a small chain which oxidizes NO and CO. Reactions R5 through R8 plus R4 (R9 will supplement R5) comprise a chain which appears to have considerable significance. Formaldehyde eventually ends up as CO and water. Hence, the chain is a mechanism for oxidation of methane. The other product of importance to the present discussion is H_2O. Levy (1971) considers these reactions to be of significance near ground level and that photodissociation of ozone to be the principal initiating reaction. The concentration of OH in the atmosphere is a subject of controversy. Levy (1971) lists the total (unpolluted atmosphere) $OH + OH_2 + CH_3O_2$ at 5×10^8 molecule/cm^3 with the OH apparently about 10^7. McConnell (1971) used a value of 2.5×10^6. Warneck (1974) considers these values too high and

suggests 5 x 10^5 as an upper limit. Both McConnell and Levy consider these reactions to be the major source of CO. McConnell calculates that 2 x 10^{14} g/yr of CO is produced. The chain also produces H_2, reaction p-7, and Levy (1973) calculates 4.6 x 10^{13} g/yr is produced. However, as noted above this may be an order of magnitude high. Thus, CH_4 may be decomposed at a rate of about 10^{14} g/yr and H_2 produced at 10^{12} to 10^{13} g/yr. A greater uncertainty in the H_2 production would seem reasonable since p-7 is not the only reaction which decomposes formaldehyde.

However, an equally important point involves the oxidation of H_2. The rate constants for R5 and R6 are compared with R15 and R17 in Table 8. It is seen that they are roughly equivalent. The radicals formed are converted to water through other chains or by the chain stopping reactions R18, R19 and R20. Thus, photochemical oxidation of H_2 would be expected at about the same rate as for CH_4 leading to about 10^{13} g H_2/yr oxidized.

TABLE 8. Rate Constants for Oxidation[a]

Reaction	k, cm^3 $particle^{-1}$ sec^{-1}
$CH_4 + OH$	3.83×10^{-12} exp(-1830/T)
$CH_4 + O(^1D)$	3.6×10^{-10}
$H_2 + OH$	6.8×10^{-12} exp(-2020/T)
$H_2 + O(^1D)$	2.9×10^{-10}

(a) Reference, Chameides (1975). Rate constants are listed for 51 reactions of interest. Whitaker (1975) has recently compiled rate constant data for 93 reactions involving oxygen and isotopic hydrogen species.

Consideration of the tropospheric inventory of H_2 and CH_4 (Table 3) would seem to place photochemical oxidation as a major mechanism for the conversion of both HT and CH_3T to HTO in the troposphere. The present crude figures would place the oxidation turnover time at about 10 yr. Since large uncertainties exist in both the concentrations of reactive

species such as OH and in rate constants for many radical reactions, considerable effort needs to be expended in this area.

REACTION SUMMARY

The potential mechanisms for atmospheric conversion of T_2, HT (and CH_3T) to H_2O include: 1) exchange with H_2O, 2) oxidation by O_2, 3) reactions of the metastable THe^+ ion following tritium decay, 4) bacteriological reactions, and 5) photochemical oxidation.

If we define a significant conversion rate to be one that is equal to or greater than the radioactive decay rate, then water exchange and oxidation by air are eliminated at concentrations below about 10^{-4} Ci/ℓ. Reaction 3 is significant in the sense that the THe^+ species will react with many different molecules. However, on a rate basis there are two important limitations: it applies only to T_2 (not HT) and the rate cannot be greater than twice the natural decay rate.

Thus, bacterial action and photochemical oxidation remain as the more important reactions. Bacteria may play a significant part in the overall tritium cycle especially when considered on a global basis. It does not appear possible to quantify this contribution at the present time. With respect to the photochemical reactions, information on the concentrations of reactive species involved, or rate constants, and on average lifetimes of various reactants and products of the chain reactions is uncertain enough that the rates cannot be determined with any precision. It is not likely that these reactions are more than an order of magnitude faster than the decay rate.

TRITIUM RELEASE EVENTS

Measurements of HT and HTO ratios following known tritium releases offers at least qualitative information on the HT conversion reaction. At least two such events have been followed in some detail.

In 1970 289 kCi of T_2 was accidentally released through an exhaust stack at the Lawrence Livermore Laboratory (Myers, 1971). The effective release height was about 30 m but the dilution at the time of release was not indicated. Monitoring of air and airborne water vapor was immediately started using information on the topography and surface winds. All samples of water, milk and urine contained normal background levels of tritium. Tritium was detected in water vapor in the air but the maximum was at two orders of magnitude below the mpc for HTO in air.

In 1974 479 kCi of T_2 was released at the Savannah River Plant; this event was briefly mentioned earlier (Marter, 1974). The release over a 40 min interval produced a concentration of 3×10^{-2} Ci/ℓ at the 60 m stack top. The exhaust, having a linear velocity of 14 m/sec was essentially pure air, and the T/H ratio assuming normal hydrogen content of air would be about 30. A detailed sampling program along with mixing calculations was carried out. The highest HTO conversion was found at the center of the release plume about 5 hr after the release. The total tritium concentration at this point (about 40 km from the release point) was 3.9×10^{-10} Ci/ℓ of which 0.2% was HTO.

The behavior of tritium in these two cases argues for the low conversion rate.

ATMOSPHERIC MIXING

As indicated from the kinetic data the rate of dilution of released tritium is very important, for only at high concentrations are the exchange and oxidation reactions significant. Extensive dilution normally occurs at the release point due to normal air flow through the exhaust system. The remaining dilution is meteorological. This subject will not be reviewed here. However, it has received extensive study and testing and the models being developed are increasingly sophisticated. For mixing at short distances the Gaussian plume formula is normally employed. The equation gives the concentration χ at coordinates x, y, z from a point source of strength Q as a function of various experimental parameters.

The reader is referred to lectures on Air Pollution and Environmental Impact Analysis (Haugen, 1975) for concise reviews. Hanna (1976) has discussed recent work on the effects of topography and the removal and transformation mechanisms. Hales' (1975) atmospheric studies have included precipitation scavenging which is particularly important to the tritium problem.

Mixing on a local scale is important because of the dilution effect on reaction rates. Mixing on a global scale concerns the overall tritium distribution since many of the specific problems are global in nature: diffusion of HTO from the stratosphere, lattitude effects, mixing between hemispheres, the distribution of molecular species as a function of altitude and the residence or turnover time of these species. Although these topics may not directly concern the HT $\rightarrow$ HTO or $CH_3T \rightarrow$ HTO conversion they do determine what limits may eventually be placed on radioactive constituents. Renne and coworkers (1975) have carried out an analysis of a hypothetical tritium release (as HTO) from a fusion reactor and computed the environmental concentrations. Machta (1973) has carried out an analysis of global distribution of effluents released to the atmosphere.

EVALUATION AND RECOMMENDATIONS

At the present time most of the tritium in the troposphere is in the form of HT. The remainder, somewhat less than half, is largely HTO whose source is the stratospheric reservoir. A portion exists as CH_3T and the ratio of T/H in methane is surprisingly high but somewhat less than in H_2. Local variation of the T/H ratio in H_2 suggests anthropogenic sources, which are also suggested for CH_3T. It is quite likely that tritiated methane is constantly being released from sites where tritium work is being carried out.

These observations together with low conversion rates observed from known major T_2 releases suggest a very slow conversion of HT or T_2 to HTO in the atmosphere. Consideration of the various chemical and biological reactions that can occur in the atmosphere or at the surface of the earth support this conclusion. Only biological (bacterial action) reactions and photochemical oxidation appear to contribute extensively to the oxidation of tritium. The lifetime of HT (or T_2) or CH_3T in the atmosphere cannot be fixed at present but is certainly long compared to times involved in accidental releases and probably can be measured in years rather than hours or days. For a given HT or T_2 release to the atmosphere the conversion to HTO in 24 hr should be well under 1%. There is certainly much more HT than HTO in the troposphere and the amount of HT and of CH_3T may in fact be increasing.

Many of the conclusions stated in this paper are highly tentative and areas of study are suggested to provide a better basis.

Monitoring of HT should be continued on a world wide basis and at different altitudes through the stratosphere. Deuterium content should also be obtained and the data could be further improved by the analysis for a second reference trace gas. The techniques developed by Östlund and by Ehhalt and others seem perfectly adequate.

Tritiated methane seems to have been overlooked in most of the studies and should be added to the monitoring systems. A search for CH_3T sources from various nuclear facilities should be made to determine the extent of its release from different activities such as fuel reprocessing, and especially from processes where highly concentrated tritium is involved. The occurrence of tritiated methane is not only a potential problem but also offers opportunity to contribute information on the carbon cycle.

Many theoretical problems exist. They include the nature of the HeT^+ ion and its reactivity and the determination of rate constants for a host of free radical reactions involving H_2 and CH_4. Various models of the hydrogen-oxygen atmosphere are not in good agreement. The discrepancies show up not only in the rate constants but in the predicted concentrations as a function of altitude for the key reactive species. Orders of magnitude differences exist in some cases. This is clearly a complex problem and requires a combination of theoretical and laboratory studies together with increasingly sophisticated atmospheric analyses.

One is tempted to write off those chemical reactions that have extremely low reaction rates in the absence of catalysts such as $HT + O_2$, $HT + H_2O$ $CH_3T + H_2O$, etc. However, one must be sure that catalytic agents are not present in the vicinity of a tritium release. The study of these catalytic reactions can have a dual purpose: the further elucidation of reaction mechanisms and the potential application to separation processes.

The bacteriological reactions involving both hydrogen and methane are extremely interesting and may be important in the tritium cycle. The need for further study is obvious from the large uncertainties that exist in the formation and uptake rates of H_2 and CH_4. The isotopic specificity of aerobic and anaerobic bacteria should also be examined.

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