Tritium and deuterium in atmospheric hydrogen

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ABSTRACT

After correcting the tritium content of atmospheric molecular hydrogen samples for contamination by industrial hydrogen, the increases in the tritium content of atmospheric hydrogen over the past ten years are discussed.

It is found that the increases in the tritium concentration occur in steps about 2 to 2.5 years after major thermonuclear test series. It is concluded that most of the HT formed by thermonuclear explosions is injected into upper stratospheric levels and subsequently follows the mixing pattern observed for the high altitude tracer rhodium-102.

Introduction

The origin of atmospheric H_2 is still subject to speculation. However, studies using the T and D isotope content of H_2 as tracers may give some information especially on the turnover time of total atmospheric hydrogen. So far, only surface air samples have been available for isotopic studies. Furthermore, such samples were provided by air liquefaction plants mostly situated in heavily industrialized areas, subjecting them to contamination by locally produced industrial hydrogen.

Correction of industrial contamination

The industrial contamination of atmospheric hydrogen samples can be corrected for. Since the natural concentration of H_2 in the surface air is fairly well known (GLUECKAUF & KITT, 1957), all one needs to calculate the isotopic composition of the uncontaminated H_2 are the amount and the T content of the admixed H_2. The T content of the admixed H_2 is negligible, since industrial H_2 is produced from water, the T concentration of which is at least a factor of 10^3 lower than that of atmospheric molecular hydrogen.

The amount of H_2 present in the original air sample can be obtained from the composition of the sample of the crude-neon fraction, from which the H_2 is extracted for isotopic analysis. This fraction contains, besides a variable amount of N_2, the low boiling gases H_2, He and Ne which are enriched during the liquefaction process. Since the fractionation during this process must be small, for reasons discussed below, the original concentration ratio of H_2, He and Ne of the air sample is well preserved. Therefore, an increase of the measured H_2/He ratio above the "natural value" of 0.08 yields directly the amount of admixed industrial H_2, since the He content of the air remains fairly constant as indicated by the relatively constant He/Ne ratio measured. The natural value of 0.08 corresponds to the maximum of the distribution of the measured H_2/He ratios (EHHALT *et al.*, 1966). With a He concentration of 5.24 ppm (GLUECKAUF, 1952) it leads to a H_2 concentration of 0.42 ppm by volume (cf. GLUECKAUF & KITT, 1957). The fairly constant He/Ne ratio and the essentially complete recovery of atmospheric Ne in the low boiling fraction confirms the fact that fractionation during processing is small.

The local addition of industrial H_2 is by far the largest source of scatter in the T content. This is proved by the correlation of the measured T content with the H_2/He ratio (Fig. 1). Clearly the highest T concentrations are measured for the natural H_2/He ratio. Any addition of H_2, i.e., any increase in the H_2/He ratio decreases the T content measured. This decrease can be as large as a factor of 10. Since the T content of the admixed H_2 is negligible we can correct for the industrial contamination simply by multiplying the measured T values by the

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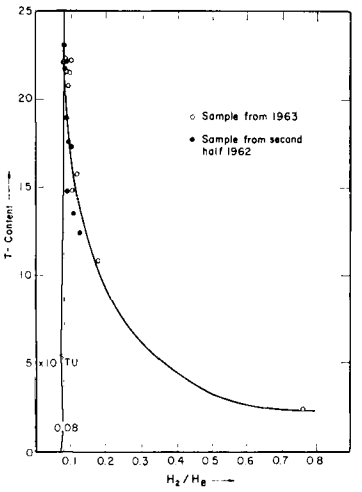

Fig. 1

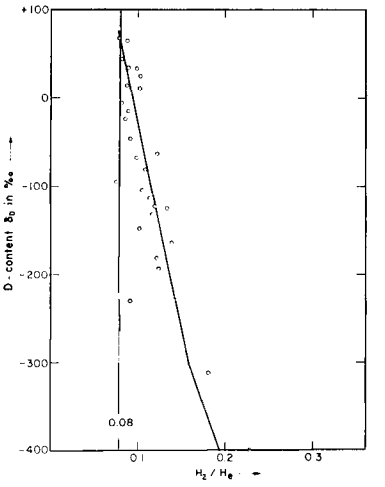

Fig. 2

FIG. 1. Relationship between T content and volume concentration of molecular hydrogen in air. The samples were taken at Lohhof during the second half of 1962 and 1963, when the average T content was relatively constant. The T content is given in TU (1 T atom/10^{18} H atoms). The hyperbola drawn is computed using the following values: the natural H_2/He volume concentration ratio is 0.08, the T content of the admixed H_2 is zero, and the average T content is 23×10^5 TU, obtained by averaging all corrected T measurements during 1962 and 1963.

FIG. 2. Relationship between relative D content and volume concentration of molecular hydrogen in air. The curve is calculated assuming the natural H_2/He ratio is 0.08, the contaminating H_2 has a D content of -700 ‰, and uncontaminated atmospheric hydrogen has a D content of $+80$ ‰.

quotient of H_2/He ratio of the sample divided by the natural H_2/He ratio.

The corresponding correlation for D (Fig. 2) shows a larger scatter. Again, the highest D content is observed for low H_2/He ratios. Extrapolation to the natural H_2/He ratio gives a value for the D content of natural H_2 of about $\delta = +80$ ‰.[1]

Furthermore, as first shown by BEGEMANN & FRIEDMAN there exists a linear correlation between D and T (Fig. 3). This indicates that the added H_2 has a constant or nearly constant D content. Its value is obtained by extrapolating to zero T content. For the Lohhof samples, our measurements exhibit two quite different correlations with different extrapolation values. The one with about $\delta = -250$ ‰ is in agreement with the values found earlier by BEGEMANN & FRIEDMAN (1959) and GONSIOR *et al.* (1963). It was ascribed to the admixture of H_2 produced by the water-gas reaction. The other is about -600 ‰, which may be caused by the admixture of electrolytical H_2 ($\delta = -700$ ‰). Both types of H_2 appear to have been processed in a nearby H_2 purification plant (at different times). This admixture of H_2 with different D content can explain part of the large scatter of the D vs H_2/He correlation in Fig. 2.

The older measurements used water samples (provided by Linde A. G.) which were obtained by oxidizing the H_2 in the low boiling fraction and no information on the H_2/He ratio was available. Assuming that the natural D content of $+80$ ‰ found in Fig. 2 is valid for other sampling sites and times we can correct these earlier measurements for industrial contamination in cases where D measurements have been made and a D–T correlation exists. Here the T correction is made by connecting the extrapolation point at $T = 0$ and $\delta = -250$ ‰ (resp. -600 ‰) with the point of measured D and T content and by extrapolation of the straight line to the natural D content. This method may be less accurate because two extrapolations are involved. However, using the raw neon samples of 1962 and 1963 for a comparison with the H_2/He correction method, a fairly good agreement is found (EHHALT *et al.*, 1966).

[1] The D content is given as relative deviation from standard mean ocean water (SMOW) (in ‰) $\delta = [(R - R_{st})/R_{st}] \cdot 100$; $R = [D]/[H]$, the isotope ratio.

Discussion

In Fig. 4 the T data of various authors have been combined after correction by the methods described above. Where no corrections were possible, the measured T values are given, as is the case for the four pre-bomb samples by FALTINGS & HARTECK (1950), GROSSE *et al.* (1953), ROWLAND & FIREMAN (1961) and BAINBRIDGE *et al.* (1961). There is considerable discrepancy between the sample measured by Bainbridge *et al.* and the others, leaving some doubt about the actual pre-bomb level of T in atmospheric H_2. However, the subsequent measurements by BEGEMANN & FRIEDMAN (1959),[1] GONSIOR *et al.* (1959, 1963), EHHALT *et al.* (1963, 1966) and GONSIOR (1964)[1] agree very well, in spite of the different sampling locations. The original scatter was largely removed by correction for locally admixed H_2.

Starting with the Ivy test, 1952, there is a more or less continuous increase of HT with time. But surprisingly there is no direct correlation between nuclear explosion series and the T radioactivity in H_2, as is the case for T in H_2O, for fission products and for C^{14} in CO_2 which has a tropospheric lifetime more comparable to that of HT. These latter all show a strong increase and seasonal variation during the year following nuclear tests due to the downward mixing of the debris injected into the lower stratosphere. This effect is lacking for HT. The discrepancy becomes especially evident in the years 1961 through 1963. After the large Russian test series in the fall of 1961 and the U.S. and U.S.S.R. tests of 1962, C^{14} in tropospheric CO_2, T in rain and fission products in air and rain increased considerably in 1962 and very strongly during 1963. However, the T content of H_2 exhibited a marked increase as early as January 1961 long before any shot had been fired and then remained nearly constant during 1962 and 1963.

On the other hand, neglecting the smaller variations, there exists a comparable trend between the T content of H_2 and the tropospheric Kr^{85} concentration. The Kr^{85} increase is indicated by the thin curve in Fig. 4. Since Kr^{85} is mainly produced by nuclear industry rather than by test explosions (EHHALT *et al.*, 1963, 1964) one is tempted to assume the same source for the HT also.

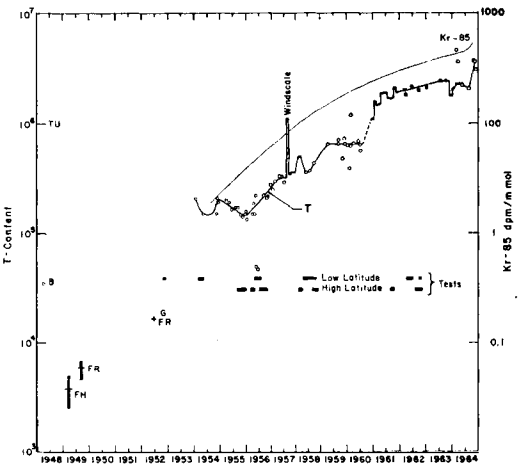

FIG. 4. The T content of H_2 in surface air during the years 1948 to 1964. Only the prebomb samples B (BAINBRIDGE *et al.*, 1961), FH (FALTINGS & HARTECK, 1958), G (GROSSE *et al.*, 1953), FR (FIREMAN & ROWLAND, 1961), are not corrected for industrial contamination. The samples for 1954, 1955 and some in 1956 were measured by BEGEMANN & FRIEDMAN (1959); those for 1956, 1957 and 1958 by GONSIOR *et al.* (1959, 1963). The samples for 1961 were also measured by GONSIOR (1964). Those for 1960, 1962, 1963, 1964 were measured by EHHALT *et al.* (1963, 1966). Circles indicate single measurements. Bars and squares represent average values for a month resp. a week.

[1] To correct BEGEMANN & FRIEDMAN's (1959) T measurements an extrapolation value of -300 ‰ was applied. For GONSIOR's (1964) measurements an extrapolation value $\delta = -250$ ‰ and a maximum D content of H_2 $\delta = 180$ ‰ were observed and had to be used for correction.

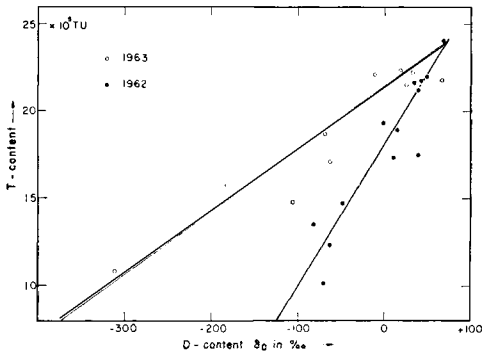

FIG. 3. Correlation between T and D content of samples from Lohhof taken in the second half of 1962 and during 1963 (EHHALT *et al.*, 1966). There are two extrapolated values. In 1962 samples H_2 with a D content of about -250 ‰ is admixed. In 1963 samples H_2 with about -600 ‰ is admixed.

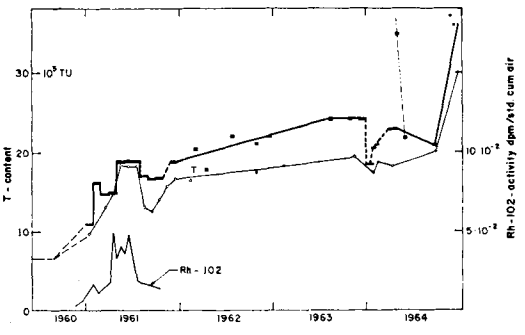

FIG. 5. The increase in HT during 1961–1964 following the series during 1958. *Upper curve:* average T content of samples from Lohhof near München and Ludwigshafen (Germany). *Middle curve:* minimum T content for the same samples. *Lower curve:* Rh^{102} in northern troposphere, at 5.5 km height, midlatitude, concentrations given as dpm per standard cubic meter of air (right hand scale).

Rh^{102} measurements are taken from KALKSTEIN (1963). The T measurements from 1961 (Ludwigshafen) are taken from GONSIOR (1964).

There exists one clear indication of industrially released HT. A very strong HT increase, starting on October 12, 1957 and reaching its peak on October 16 was observed for samples taken at Hamburg (GONSIOR *et al.*, 1963). This increase was originally attributed to direct admixture of HT produced by a Soviet thermonuclear explosion. However, a correlation with the Windscale accident of 10–11 October in England seems far more likely. The radioactive cloud released by Windscale arrived in the region of Hamburg about 12 October (CRABTREE, 1959). On subsequent days the cloud moved northeast, causing a further increase of I^{131} activity in air in the region around Hamburg, which agrees well with the observed time of increase in the HT concentration to its maximum.

The early 1961 increase in HT could, in principle, be attributed to industrial sources. However, it would require a very substantial release of tritiated H_2 in the northern hemisphere to explain the steep rise and marked peaking of the HT concentration at that time (Fig. 5). Assuming uniform distribution in the nothern hemisphere, the T input in the period from January to August 1961 amounts to ~ 350 g of T, an amount which seems high for an industrial release. Furthermore, for extremely strong and more or less local sources one would expect large fluctuations in the HT concentration like that for Windscale (Fig. 4) rather than those observed during early 1961.

The occurrence of a marked peak during late spring (Fig. 5) however, strongly suggests another explanation, namely, an influx of tritiated H_2 from the stratosphere. Since this HT could only have been injected during the 1958 tests, its appearance in the troposphere is apparently delayed by about 2.5 years. If this interpretation is correct we might expect a similarly delayed rise in the tropospheric HT concentration after other large nuclear test series also. Indeed, Fig. 4 indicates a more or less stepwise increase of HT concentration with steeper increases during the year following an interval of 2—2.5 years after each major test series. Thus the first observed increase in 1956 might be correlated with the Castle test of 1954; a less well marked second increase in 1958–1959 with the Redwing tests of 1956 and the Russian series at the end of 1955; and the third and most pronounced increase in 1961 with the 1958 tests, especially during October 1958 (the schedule of major nuclear tests is indicated in Fig. 4). Correspondingly, the HT produced during the Russian tests in late 1961 could explain the high values and steep rise during 1966.

MARTELL (1963) has estimated the amount of T produced during various nuclear tests, using the available data on thermonuclear yields and Leipunsky's value of 0.7 kg tritium per megaton of fusion energy. Table 1 compares his estimates with the corresponding T increase in tropospheric H_2 about two years later.

The height of each step increase corresponds roughly to the relative magnitude of the test giving further support to the suggested correlation of delayed increases and series. A more than qualitative agreement can hardly be expected, because the uncertainties of the fusion yields, especially for the Russian tests, subject the estimates for the T production to large uncertainties. Additionally, the surface explosions of the early series might yield different HT injection height and production amounts than those of Soviet air bursts during 1958.

Since there were nuclear test series concurrent with the 1956 and 1958 increases in HT (Fig. 4) it is possible that the direct admixture of bomb produced HT is at least partially responsible for those increases. However, there is little evidence of an increase in HT concentration during or

TABLE 1

Test series	Castle, 1954	USSR, late 1955 & Redwing, 1956	Tests 1958
Estimated T Production	10 kg	10 kg	25 kg
Increase in T content of tropospheric H_2 two years after test	$2\ 10^5$ TU	$3\ 10^5$ TU	$12\ 10^5$ TU[a]

[a] Assuming the average tropospheric T activity in H_2 to be represented by the minimum activities measured.

directly after the 1961 series. This indicates that such directly admixed HT plays only a small part in the overall increase. Additional evidence which also rules out a major direct contribution, is based on the high altitude injection of HT and is discussed below.

Without information on the HT concentration in the stratosphere, we cannot exclude the possibility that all or some of the step increases in the tropospheric HT could be caused by intermittent HT injections by nuclear industry. However, industrial HT should have given rise to stronger fluctuations during the increases than are indicated by the measurements. In addition, the assumption of a stepwise industrial release seems fairly arbitrary in view of the smaller fluctuations, more continuous increase of Kr^{85} with mainly industrial origin. There probably exists some continuous industrial release of HT. However, for the major increases the most probable explanation seems to be the downward mixing of HT into the troposphere about 2–2.5 years after thermonuclear tests. This time delay implies an injection of HT into high stratospheric altitudes. We might, therefore, compare the tropospheric increase of HT in 1960 and 1961 with that for the high altitude tracer Rh^{102} during the same period (Fig. 5). The heavy line curve in Fig. 5 shows the average T content and the thin line curve gives the minimum T values observed. The lower curve describes the Rh^{102} concentration in the northern troposphere at mid-latitudes as reported by Kalkstein. This tracer Rh^{102} was produced by the high altitude thermonuclear experiment Orange Shot, over Johnston Island on 11 August 1958. The Orange Shot cloud was estimated to have risen above 100 km altitude. Obviously the Rh^{102} mixed down into the troposphere with the same 2.5 year delay which seems to occur for the tritiated hydrogen. The Rh^{102} and HT furthermore, show similar spring peaks, indicating their common stratospheric origin. Due to the much shorter tropospheric residence time of the particulate Rh^{102} tracer its spring peak is, of course, more pronounced.

From the parallel behavior of Rh^{102} and HT it seems justified to conclude that the HT from the 1958 tests is injected to those altitudes which provide the critical delay in downward mixing for Rh^{102} and that its downward mixing follows the pattern observed for the Rh^{102} tracer (KALKSTEIN, 1962, 1963). This would mean that HT is injected into levels near the stratopause (50 km). However, the analogy with Rh^{102} cannot be carried so far as to assume uniform horizontal mixing at high altitude for HT, as is in the case for Rh^{102}. However, the step increase 2 years after the Castle tests of 1954 at low latitudes suggests mixing at high altitudes from equatorial to polar regions. In the absence of HT data for the southern hemisphere we cannot exclude the possibility that a large fraction of the HT injected by the 1958 tests was confined in the high atmosphere at northern latitudes. Therefore, the residence time of HT might be considerably shorter than the 10 years estimated for Rh^{102} (KALKSTEIN, 1962). Hence it is also not practical to calculate the fraction of total T injected in the form of tritiated molecular hydrogen. If tritiated H_2 were uniformly distributed in high altitudes and mixed down like Rh^{102} with a 10 year residence time, the 1961 increase in T content of about 6×10^5 TU would lead to 2.6 kg T originally injected in the form of H_2, i.e., 10 % of the total T production 1958. Compared to the 0.1 % of cosmic-ray produced T, which ends up as HT (HARTECK, 1954; ROWLAND, 1959), this figure seems very high. It might, therefore, indicate that the

residence time of HT is considerably shorter than 10 years and that HT is not uniformly mixed in high altitudes.

Because the 1954 and 1956 thermonuclear tests involved surface shots, it seems hard to conceive that the HT produced should be injected to the upper stratosphere. Such a process would imply that part of the T formed during the explosion escapes oxidation to water. This would presumably occur in those parts of the fireball which do not rapidly entrain air and thus allow for sufficient cooling before final mixing. Various possibilities for the HT formation during and after the explosion have been discussed (MARTELL, 1963; WOLFGANG, 1961). Because of this slow mixing one would also expect that these cloud portions remain at relatively higher temperature and, therefore, are carried to the highest altitudes reached by the explosion cloud. Although such a process could contribute during the 1958 and 1961 air burst tests the following explanation seems more reasonable for the earlier surface tests. Part of the tritiated water injected into the stratosphere is mixed upward above 50–60 km where photodissociation of the tritiated water takes place and forms HT. Both mechanisms would yield the same result in that the injection of HT would tend to be at greater altitudes than that of the main fractions of the other radionuclides formed and thus that the direct injection of HT into the troposphere or low stratosphere seems unlikely. Assuming the downward mixing of HT from above 50 km it appears that H_2 formed by photodissociation of water and subsequent recombination of the H atoms at levels around 80 km should partly escape oxidation by atomic oxygen at stratospheric levels in the process of mixing down into the troposphere.

If we assume that the main part of the measured HT is injected from the stratosphere down into the northern troposphere the input during a step increase is of the form:

$$\Phi = \Phi_0(1 + a \sin 2\pi\omega t + \alpha)\, e^{-t/\tau_s},$$

where τ_s is the time of depletion of the stratospheric reservoir by radioactive decay, chemical reaction and mixing. The term in the brackets describes the seasonal variation of the input. In view of the fact that the experimental data (Fig. 5), which, except for the first peak, are inadequate to resolve the seasonal variation, a two box model seems sufficient to describe the troposphere. The time of removal by radioactive decay, chemical reaction and mixing out of the troposphere is characterized by τ_t. The mixing time across the equator is assumed to be around one year (LAL, 1965).

The leading term in the calculated increase of the HT concentration of the northern troposphere is:

$$[\mathrm{HT}] = \frac{\Phi_0}{2} \cdot \frac{\tau_s \tau_t}{\tau_t - \tau_s} [e^{-t/\tau_t} - e^{-t/\tau_s}].$$

The other terms contribute significantly only during the first year. However, the form of the increase is changed only to a small extent by proper simultaneous variation of the parameters τ_t and τ_s. They are therefore inadequately determined by the present experimental data.

If we assume the tropospheric residence time τ_t to be maximum (i.e., equal to the radioactive decay time of about 18 years) we obtain a lower limit for $\tau_s = 1.5\ (+1.0/-0.5)$ years. This time is fairly short and if we could assume the same mixing time for HT as for Rh^{102}, it would indicate that the tritiated H_2 is partly destroyed by chemical reaction or isotopic exchange before reaching the troposphere. Any tropospheric residence time shorter than 18 years also implies chemical destruction in the troposphere or stratosphere.

Conclusions

From the qualitative discussion given above, the most probable explanation for the stepwise increases of tropospheric HT seems to be the injection of HT to high stratospheric altitudes and subsequent downward mixing following the pattern of the Rh^{102} high altitude tracer. Direct production by nuclear industry should represent a continuous minor source of HT. This means that for the larger explosions part of the radioactive cloud is carried to high stratospheric altitudes, with subsequent downward mixing, reaching the troposphere after a delay of about two years. That upper part of the cloud seems to contain most of the HT and relatively smaller fractions of the other nuclides produced.

Although no quantitative estimate of the atmospheric residence time of HT could be obtained, the results obtained encourage further study of the T content of atmospheric H_2. A

more comprehensive sampling program is needed to follow the current increase due to the 1961 and 1962 thermonuclear tests. Regular tropospheric samples from both the northern and southern hemispheres would provide the information necessary to make a good estimate of the tropospheric and stratospheric residence times, τ_s and τ_t. This result also should make it possible to estimate an independent value for the atmospheric residence time of molecular hydrogen.

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ТРИТИЙ И ДЕЙТЕРИЙ В АТМОСФЕРНОМ ВОДОРОДЕ

Рассматривается увеличение за последние десять лет содержания трития в атмосферном водороде с поправкой на загрязнение проб промышленным водородом.

Найдено, что увиличение трития происходит скачками следующими через 2–2,5 года после крупных термоядерных испытаний. Предполагается, что HT образовавшееся при термоядерных взрывах выносится на большие высоты и затем происходит перемешивание, наблюдаемое по следам родия на большой высоте.