Measurements of the Water Vapour, Tritium and Carbon-14 Content of the Middle Stratosphere over Southern England

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Abstract

Measurements of the water vapour, tritium and carbon-14 content of the stratosphere at heights of between 80,000 and 100,000 feet, made over England during the years 1956 to 1960, are described.

The tritium and carbon-14 concentrations are greater than those expected from natural production due to the cosmic radiation. The bulk of these two isotopes, at present in the stratosphere, has been injected there during the course of thermonuclear explosions.

Mass spectrometric analyses show that the deuterium/hydrogen ratio of the water and the carbon-13/carbon-12 and oxygen-16/oxygen-18 ratios of the carbon dioxide collected at these heights were the same, within the accuracy of measurement, as those for samples collected near the ground.

The concentration of tritium at the heights of the measurements has not fallen since the termination of thermonuclear weapon testing in autumn 1958. There is poor correlation between the tritium concentration at 90,000 feet and that of particulate caesium-137 at 47,000 feet measured over the sampling period. The humidity above 80,000 feet is found to be greater than that in the lower stratosphere.

It is concluded from these humidity and tritium results that the large scale meridional circulation in the stratosphere, first postulated by Brewer and Dobson, does not reach heights of 90,000 feet over temperate latitudes.

1. Introduction

During the years 1956 to 1960, a series of balloon flights at altitudes of 80,000—100,000 feet were carried out over southern England. Samples of water vapour and carbon dioxide were collected by drawing stratospheric air through a vapour trap cooled in liquid nitrogen. A radio-command or clock-controlled system operated the equipment and subsequently released it from the balloon on a parachute.

The samples of water and carbon dioxide, so obtained, were separated, measured and then assayed by conventional low background counting techniques to give their tritium and carbon-14 contents respectively. Mass spectrometric analyses of the gases yielded data on the oxygen-18/oxygen-16, carbon-13/carbon-12 and deuterium/hydrogen ratios.

Since the carbon dioxide concentration of the atmosphere, remote from the ground, is constant (GLUECKAUF, 1944, HAGEMANN ET AL., 1959), the amount of carbon dioxide collected on a flight is used as a measure of the total quantity of air sampled. Thus, the tritium and carbon-14 concentrations of the stratospheric air are deduced, together with the humidity mixing ratio.

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Preliminary results on the 1956—1958 flights have already been reported (BARCLAY ET AL., 1960, GOLDSMITH ET AL., 1960). These, together with later, more extensive, results, are presented and discussed in this paper.

2. Experimental Techniques

2.1. *The Balloon Flights*

The sampler and associated equipment (basket, parachute, radar reflectors, radio transmitters, radio command receiver and helicopter homing equipment) weighed approximately 200 lbs and were carried on a polythene balloon of the type developed by the Cosmic Ray Research Group of the University of Bristol, England. These balloons were non-expansible, open necked and about 180 feet long. The rate of ascent was approximately 800 ft/min. and so ceiling (~ 90,000 feet) was reached in about 2.5 hours when the hydrogen gas had expanded to fill completely the polythene envelope. The balloon then floated and any further height changes were caused by gas leaks or fluctuations in the gas temperature. These changes were usually quite small over three to four hours and the sampling was carried out during most of this time. The load and parachute were released from the balloon either by a preset clockwork trigger or a radio command system operated from the ground.

All balloons were launched from Cardington, near Bedford, England. There were three essential meteorological conditions required on the day of a flight.

a) Light surface winds during launching operations, to facilitate balloon handling.
b) A vertical wind profile such that the balloon had not drifted over the sea by the end of the sampling period.
c) Clear skies so that the balloon did not rise through cloud and so introduce the possibility of tropospheric water being deposited on the equipment.

In general condition (b) can only be met if the stratospheric wind at the sampling height is easterly. This limits the period when flights of long duration can be made over England to the spring and summer months, and the number of days in this period which meet the other flight conditions are very limited.

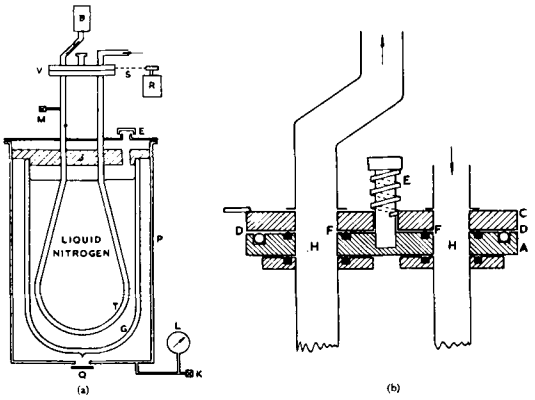

Fig. 1. (a) The vapour trap; (b) details of rotary valve V.

The balloons were tracked by Radar and recovery was by car, although on some occasions helicopters were used.

2.2 *Description and Operation of the Collection Apparatus*

The sampling apparatus, which has been fully described elsewhere (GOLDSMITH and PARHAM, 1960) is shown schematically in figure 1a. The essentials were a stainless-steel tubular vapour-trap T, a Dewar flask G containing liquid nitrogen, a centrifugal blower B and a disc-type rotary valve V which is detailed in figure 1b.

The air entered the sampler through the intake pipe connected to a port in the upper movable disc C of the rotary valve V. It was then drawn through the vapour-trap tube T, via the outlet port of the valve, by the blower B.

The valve V, consisted of two polished aluminium plates pressed together by a spring E and slightly separated by a ring of steel balls D located in a channel in the lower plate A. A vacuum seal was maintained by neoprene rubber-ring seals F, likewise located in channels in the lower plate A, one around each port H. The valve was then operated by rotating the upper plate C with respect to the lower fixed plate A by a rotary actuator R.

A wide-bore valve of this type was chosen to avoid constrictions, to minimise the use of rubber and grease, and to facilitate the baking-out of as much of the inner surfaces of the trap as possible.

Prior to the day of a flight, the vapour-trap was pumped out, through the all-metal valve M, with the valve V in the closed position, to a pressure of $\sim 10^{-5}$ mm Hg, whilst the tube was baked out in a special oven at a temperature of $\sim 400°$ C. The trap was then placed in the aluminium pressure vessel P which already contained the empty Dewar flask G. The non-conducting lid J of the flask was then carefully positioned and the stainless steel top of the pressure vessel, which was an integral part of the vapour-trap tube, bolted down. A rubber ring seal ensured that the vessel was gas-tight.

The vapour-trap was then pumped continuously until the day of the flight, when the trap was sealed at M. Only then was the liquid nitrogen loaded into the sampler through the filler plug E. Precautions were taken to avoid the collection of oxygen and to prevent the solidification of the liquid nitrogen when boiling under reduced pressure. This was achieved by maintaining the pressure inside P at about one atmosphere above that of the environment using the ball release valve K. K was adjusted before the flight, and so at full altitude, the absolute pressure inside P was about one atmosphere. This excess pressure was maintained by slow evaporation of the liquid nitrogen in the Dewar flask. A large-bore safety valve Q was also incorporated in case breakage of the Dewar flask, on launch or landing, resulted in rapid evaporation of the liquid nitrogen. As a further precaution, Q was opened permanently on closure of the valve V after sampling. As an additional safeguard Q could be tripped by a pressure capsule which was set to trigger at about 5,000 feet on the descent.

The sampler was powered by a nominal 24 volt silver-zinc accumulator, contained in a sealed housing. The opening and closing of the valve at altitude was determined by a preset clockwork mechanism or the radio command system.

The position of the valve, the performance of the blower and the pressure above the liquid nitrogen were all monitored by a modified radiosonde carried with the equipment. A Pressure-sonde, developed at the University of Bristol, was also flown and used to calculate the height of the balloon (an I.C.A.N. atmosphere was assumed).

2.3 *Precautions, Tests and Calibrations*

Every effort was made to minimize the amount of water carried up on the equipment. The external surfaces of the sampler were maintained at temperatures above the dew point of the surrounding air, by effective thermal insulation around the liquid nitrogen and because the upper parts of the sampler were black, so enhancing direct solar heating. These conditions did not apply on the first two flights in 1956 and 1957, which resulted in excessive water being collected.

Many other possible sources of spurious water were checked, the cane basket was replaced on later flights by a metal frame without affecting the measured humidity. Care was always taken that no components of the equipment were allowed to come into contact with grass of the launching field.

The most obvious sources of spurious water were the balloon and the hydrogen gas. Cylinders of dry gas were used to lessen the latter effect. The length of cord between sampler and balloon was varied between 200 and 500 feet without affecting the amount of water collected.

A more elegant check was made by measuring the deuterium/hydrogen ratio of the hydrogen of the collected water in 1959. On the 1960 flights several grams of heavy water (D_2O) were added to the balloon gas, this approximately doubled the amount of water in the gas and so made the deuterium/hydrogen ratio of this water about 2. The deuterium/hydrogen ratio of the water collected at altitude, however, remained unchanged at 2×10^{-4} which conclusively showed that diffusion of water from balloon to sampler was not important. These deuterium/hydrogen ratios are given with the other Mass Spectrometric results (Table 3).

The minimum length of the tube in the cold trap was calculated using the well established solutions to the analogous heat-conduction equations (CARSLAW and JÆGER, 1947). Uncertainty in the collection efficiency for molecules of carbon dioxide and water at the cold surface as well as the possibility of mist formation in the tube made it seem that a collection efficiency of less than 100 % for the two gases was possible. Measurements were therefore made of the flow rate through the sampler

Table 1

Flight Date	Height 10^3 feet	Volume of CO_2 ccs NTP	Sampling Time Mins.	Flow rate from CC_2 litres/sec	Calibrated Flow rate Litres/sec
13.5.59	91	95	160	1.6	2.3
5.6.59	80	150.1	110	2.2	2.7
15.6.59	93	69.0	51	3.8	2.3
18.6.59	90	157.9	150	2.9	2.4
7.7.60	89	139.6	125	2.7	2.4
11.9.60	99	76.5	133	2.2	< 2.3

in a large decompression chamber using a hot-wire anemometer. In Table 1, the results of these calibrations are compared with the flow rates on actual flights, as deduced from the amount of carbon dioxide collected, the duration and height of the flight and by assuming an air temperature of −53° C. The carbon dioxide concentration for this purpose is taken as 0.031 % by volume (HAGEMANN et al., 1959), not 0.025 % as found in earlier work (GLUECKAUF, 1944).

The flow rates found on the flight were more variable than those found on calibration. This is not unreasonable since: —

a) the load imposed on the blower motor by the monitoring reduction gear was probably variable.
b) different blowers of the same model were used on each flight.
c) the calibrated flows are appropriate to an accumulator voltage of 27, whilst in fact the potential applied to the blower during a flight was 30 volts falling to 24 volts after about one hour.

The mean flow rate on the first five of the above flights is 2.6 litres/sec, compared with a mean calibrated flow of 2.4 l/sec. This suggests that the vapour collection in the sampler was complete.

2.4 *Gas Analysis*

On recovery of the sampling equipment after a flight the contents which consisted of the collected carbon dioxide and water vapour together with air at the ambient pressure of the sampling altitude, were transferred to a storage tube to await detailed analysis.

The recovered sampler was connected to a vacuum apparatus and the gases drawn from the trap, via a needle valve, through two pyrex glass U-tubes connected in series and cooled in liquid nitrogen. The gas flow rate was adjusted so that the pressure of the non-condensable gases on the exit side of the U-tubes did not exceed 0.05 mm Hg. Under these conditions complete condensation of the carbon dioxide and water was achieved as verified by the fact that the second U-tube never contained any condensed vapours. Any vapour absorbed on the walls of the sampler tube was removed by raising the temperature to approximately 400° C and pumping until the pressure fell to the order of 10^{-5} mm Hg. The total transfer was completed in three hours, whereupon the U-tubes were sealed by melting constructions in the arms. Subsequent entry to the U-tubes was via break seals.

In the laboratory the U-tube was connected to a high vacuum apparatus and the break seal opened. The gas sample was passed slowly through a nitrogen cooled trap to remove any last traces of noncondensable gases.

Separation of the carbon dioxide from the water was achieved by repeated distillation from frozen pentane (MP ~ −130° C, V. P. $H_2O < 10^{-7}$ mm Hg. V. P. $CO_2 \sim 1$ mm. Hg). The carbon dioxide was then pumped into a calibrated volume and samples taken in small break seal tubes for mass spectrometric analysis. The water was condensed into a tube containing approximately 10 g of zinc, which had been outgassed by heating at 400° C under vacuum for several hours. This tube was then sealed and heated at 400° C for 36 hours to effect complete reduction of the water. On completion of reduction the tube was opened, the hydrogen transferred into a calibrated volume and samples taken for mass spectrometric analysis. The amount of water

originally collected was calculated from the volume of hydrogen obtained and calibration runs with known amounts of water indicated that the inaccuracies would not be > 1 % using this procedure. Known amounts of the hydrogen and the carbon dioxide were pumped into proportional counter for tritium and carbon-14 determinations respectively.

2.5 *Counting*

Tritium

The tritium was measured in a proportional counter. The counter, 3 inches in diameter and 12 inches long, was constructed of copper with a 1 mil tungsten anode centre wire and had a sensitive volume of approximately 1 litre. To reduce the natural background and so increase the sensitivity of the apparatus, a double layer of Geiger counters was arranged round the counter and operated in anti-coincidence. The whole counting assembly was mounted in a steel castle with 8 inch thick walls and 8 inch thick sliding doors. The counter was followed by conventional amplifier anticoincidence and scaler units. Backgrounds of the order of 10 counts/minute were obtained and were at least an order of magnitude less than the activity of the measured samples.

The hydrogen samples were introduced into the counter and the pressure raised to 1 atmosphere with the counting gas (argon/10 % methane). Provided that the partial pressure of hydrogen was less than 10 cm. Hg. the resolution and plateau characteristics of the counter were unimpaired. Samples were counted to 1 % statistics. Counter end effects had been determined experimentally and the necessary corrections were applied. A Fermi plot of the β spectrum was consistent with all counts being due to tritium.

The 1956 sample was also counted independently by Geiger Counters (GARD, 1958), and satisfactory agreement was found.

Radiocarbon

The radiocarbon was determined in a similar manner but the counter was smaller having a sensitive volume of approximately 250 cc. Backgrounds of approximately 2 counts/minute were obtained. Since the carbon dioxide samples were small, the radiocarbon activity was less than the background. Under these conditions the accuracy was poor and standard deviations of ± 8 %— ± 60 % were the best that could be obtained.

2.6 *Mass Spectrometric Analyses*

Analyses of the carbon dioxide and hydrogen samples yielded data on the carbon-13/carbon-12, oxygen-18/oxygen-16 and deuterium/hydrogen ratios respectively (Table 3).

A Metro-Vic Mass Spectrometer Type MS_2 was used which had a 90° tube, 15 cm. radius ion path and an acceleration voltage of 2KV. It was fitted with a model 51A Vibron MS vibrating reed type amplifier to increase the sensitivity.

To correct for instrumental variations over the period, samples of the same standard carbon dioxide or hydrogen were measured before and after each stratospheric sample.

3. Results

3.1 *Tritium, radiocarbon and humidity*

The results of the tritium, radiocarbon and humidity determinations are summarised in Table 2. Basic measurements are given in columns 1 to 6, while data in columns 7 to 15 have been derived from these measurements using the following factors: —

a) The density of air at N.T.P. is 1.292 gram per litre.
b) Air contains 0.031 % by volume of carbon dioxide and the natural radio-carbon background is 7.1×10^6 atoms per gram of air [HAGEMANN et al. 1959).
c) The half-lives of tritium and radiocarbon are 12.23 yrs and 5,600 yrs respectively.

The majority of the results are consistent but some variations are apparent and can be explained on known experimental observations.

On the 1956 and 1957 flights the liquid nitrogen caused cold spots on the sampler surface, which led to condensation of tropospheric water on the equipment. Humidity mixing ratios and tritium/hydrogen ratios were thus invalidated. This did not effect the actual tritium count since the tritium content of this tropospheric water was so low that its contribution could be neglected. If an average value of 0.041 g/Kg is taken for the humidity mixing

Table

Tritium, radiocarbon and humidity measurements

1	2	3	4	5	6	7	8
Date of Flight	Altitude (10^3 feet)	Collected CO_2 (cc. NTP)	Collected Water (mgm)	Tritium Activity (dpm)	C^{14} Activity (dpm)	Tritium atoms (10^9)	C^{14} atoms (10^{10})
20.9.56	93	33.2	800	252	—	2.33	—
10.5.57	92	20.4	180	53	—	0.49	—
2.5.58	89	118.3	21.3	239	2.4 ±1.0	2.21	1.0 ±0.4
13.5.59	91	95.0	17.8	223	9.9 ±0.8	2.06	4.2 ±0.3
5.6.59	80	150.1	5.6	338	3.0 ±1.7	3.13	1.3 ±0.7
15.6.59	93	69.0	14.1	283	7.0 ±1.8	2.62	3.0 ±0.8
18.6.59	90	157.9	50.6	397	2.8 ±0.4	3.68	1.2 ±0.2
7.7.60	89	139.6	19.6	451	2.2 ±0.5	4.18	0.9 ±0.2
11.9.60	99	76.5	11.1	203	0.65±0.21	1.88	0.3 ±0.1

ratio on these flights, the tritium/hydrogen ratios are 6×10^{-12} and 2×10^{-12} and thus fall within the range of the later results.

The highest humidity mixing ratio and correspondingly the lowest tritium/hydrogen ratio, were obtained on the flight of 18th June, 1959. Here the balloon was observed to rise through thin stratocumulus cloud and it is possible to attribute these results to the collection of tropospheric water on the surfaces of the sampling apparatus.

The lowest humidity mixing ratio and highest tritium/hydrogen ratio are both associated with the flight of 5th June, 1959, which was made at an altitude of 80,000 ft., i.e. at least 9,000 ft. lower than the other flights. This can be construed as evidence in favour of pick-up of tropospheric water on all the other flights, but this is considered unlikely since the flight was identical to the others except that the balloon was somewhat smaller.

Combining the sources of error associated with sample condensation, volume and counting measurements and calculations of the amount of air sampled it is estimated that the tritium concentrations, humidity mixing ratios and tritium/hydrogen ratios should be accurate to within 10 %.

In the case of radiocarbon concentrations, the errors in the activity determinations are the predominant factors. Since these factors vary from sample to sample the errors are quoted on the results in Table 2.

3.2 *Mass Spectrometric Determinations*

Results for the atom per cent carbon-13, oxygen-18 and deuterium in the carbon dioxide and hydrogen respectively are given in Table 3. Within the accuracy of the analyses, the atom percent deuterium was the same for high altitude moisture as for that at ground level. No increase was noted in the 1960 flights when heavy water (D_2O) was added to the balloon hydrogen. It was estimated that the balloon hydrogen contained approximately 6 g. of water and this amount of heavy water was added to give 50 atom percent deuterium, in the combined water. Since the deuterium figures on the water condensed in the sampling equipment showed no increase in these cases, moisture pick up from the balloon could be neglected.

The deuterium figures were in all cases higher than the value of 0.015 quoted for natural samples. The reason was that at the pressures pertaining in the mass spectrometer some $(H\,H\,H)^+$ ions were formed (BARNERD, 1953) and appeared at the HD peak so enhancing it. When a series of measurements were taken at various pressures, the plot of atom percent deuterium against pressure gave a value of 0.015 when extrapolated to zero pressure.

Carbon-13 and oxygen-18 results were the same for carbon dioxide from low level as for the high level samples and consistent figures were obtained over the whole series.

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on samples of stratospheric air.

9	10	11	12	13	14	15
Weight of air sampled (g)	Mixing Ratio (g. water per Kg air)	Tritium Atoms (10^6 per g. of air)	C^{14} atoms (10^7 per g. of air)	C^{14} atoms (10^7 per g. of air) corrected for natural background	Tritium to Hydrogen atom ratio (10^{-12})	Atoms of excess C^{14} per atom of tritium
138	—	16.9	—	—	—	—
85	—	5.78	—	—	—	—
493	0.044	4.48	2.1±0.9	1.4±0.9	1.52	3.1±2.2
396	0.045	5.20	10.6±0.9	9.9±0.9	1.73	19.0±2.5
626	0.009	5.00	2.0±1.2	1.3±1.2	8.39	2.6±2.4
288	0.049	9.10	10.3±2.7	9.6±2.7	2.77	10.5±3.0
658	0.077	5.59	1.8±0.3	1.1±0.3	1.12	2.0±0.6
582	0.034	7.18	1.6±0.3	0.9±0.3	3.26	1.3±0.5
319	0.035	5.89	0.9±0.3	0.2±0.3	2.60	0.3±0.5

4. Discussion

Fig. 2(a) shows the time variation of the tritium concentration over the sampling period. Also plotted are the only other published tritium measurements above 80,000 feet at comparable latitudes to England (HAGEMANN ET AL., 1959); these were obtained over Minneapolis, U.S.A., using a somewhat different technique. The figure near each point refers to the height of the sample in thousands of feet. It is seen the agreement between the two sets of measurements is reasonable. The fluctuations in the tritium concentration up to 1958 during a time of many thermonuclear explosions, are not surprising. However, the 1959 and 1960 results obtained after the termination of weapon testing in November 1958, demonstrate no decline in the tritium concentration.

It is not possible to give an accurate estimate of what proportion of this tritium is due to natural cosmic ray sources. However, an order of magnitude estimate, using the natural production rate of about 1 tritium atom /cm²/sec (WILSON & FERGUSSON, 1960), can be made if a very idealised stratosphere is considered. Assuming that two-thirds of the natural production takes place in a well mixed stratosphere

Table 3.

Mass spectrometric determinations of the atom % C^{13}, O^{18} and D on the stratospheric CO_2 and hydrogen and comparable results on samples collected at ground level.

Date of Flight	Carbon Dioxide		Hydrogen	Amount of D_2 added to balloon hydrogen (g)
	Atom % C^{13}	Atom % O^{18}	Atom % D	
13.5.59	1.165±0.001	0.208±0.001	0.0220±0.0005	Nil
5.6.59	1.159±0.001	0.209±0.001	0.0220±0.0010	Nil
15.6.59	1.167±0.001	0.209±0.001	0.0187±0.0007	Nil
18.6.59	1.167±0.007	0.209±0.002	0.0207±0.0006	Nil
7.7.60	1.163±0.004	0.210±0.001	0.0184±0.0010	6.0
11.9.60	1.168±0.004	0.210±0.001	0.0174±0.0010	6.5
Samples collected at ground level at the launching site				
1959	1.165±0.007	0.208±0.001	0.0220±0.0013	
1960	—	—	0.0194±0.0010	

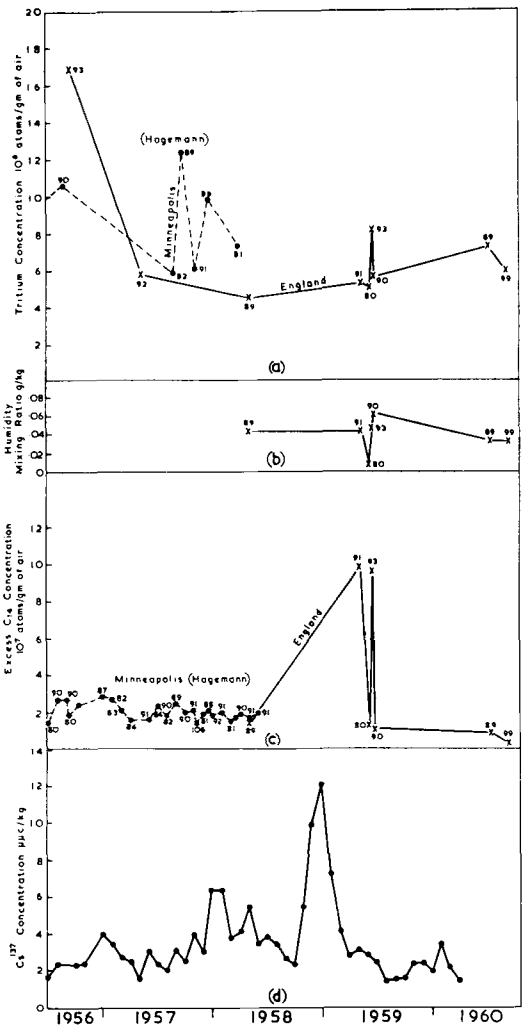

Fig. 2. (a) The tritium concentration, (b) the humidity mixing ratio, and (c) the excess carbon-14 concentration above 80 000 feet measured during the years 1956 to 1960. (d) The monthly mean concentrations of caesium-137, produced in nuclear tests, at a height of 47,000 feet over England during the same period.

in which the tritium has a residence time of 5 years, then the natural tritium concentration will be about 3×10^5 atoms/gm. This compares with the measured values of between 4×10^6 and 2×10^7 atoms/gm reported in this paper. It is probably fair to conclude that most of the tritium at present in the stratosphere is not of natural origin.

The humidity and excess carbon-14 variations are similarly presented in figs. 2(b) and 2(c). The more extensive carbon-14 results of HAGEMANN ET AL., (1959), are also plotted. The two peak carbon-14 concentrations found in 1959 are much greater than any variations reported in the American work, although the other results are in satisfactory agreement. Only the peak at 93,000 ft. on 15th June, 1959, is associated with a tritium peak. The correlation found between tritium and carbon-14 is not as good as that reported by Hagemann et al., but this may be due primarily to our carbon-14 counting being somewhat poor statistically, because of the small amounts of carbon dioxide collected.

The extensive measurements (BANNON ET AL., 1952, MURGATROYD ET AL., 1955, HELLIWELL ET AL., 1957), using aircraft fitted with the "Dobson-Brewer" frost-point hygrometer (BREWER ET AL., 1948), of the humidity up to 50,000 feet show that it is invariably dry there, with a humidity mixing ratio of about 0.002 g/Kg with no significant seasonal variations. The humidity data, presented in this paper, means that the air at heights of about 90,000 feet is very much moister with a mixing ratio of about 0.04 g/Kg.

The one result at 80,000 feet on 5.6.59 is the driest (.009 g/Kg) and may mean that the transition from moist to dry air on this occasion was in the region of this height.

The 1958 humidity result has already been compared with other data on the stratospheric humidity (BARCLAY ET AL., 1960). At that time the main evidence in support of a moist middle stratosphere was the occurrence of Mother-of-Pearl clouds at heights of about 80,000 feet over northern latitudes (HESSTVEDT, 1959). Automatic hygrometer balloon flights of BARRETT ET AL., (1950) also were in reasonable agreement, but this work also showed the lower stratosphere to be moist.

However, since the publication of the 1958 result, not only have the subsequent flights, presented here, confirmed the early result, but considerably more evidence in support of the presence of moist air above the dry lower stratosphere has been accumulating.

Recordings of the sun's infra-red spectrum by a spectrometer carried to heights of 92,000 feet by balloon (MURCRAY ET AL., 1960) over New Mexico, U.S.A., have been interpreted to point to this type of humidity profile in

the stratosphere. Similar work, using a spectrometer, mounted on a U.2 aircraft flown at heights of 65,000 feet, have yielded similar information on the vertical distribution of water in the stratosphere (Lab. of Astrophys., 1960). Recent soundings up to 100,000 feet, using a balloon borne automatic frost-point hydrometer, are again consistent with the concept of a dry lower stratosphere with moister air above (MASTENBROOK & DINGER, 1960).

BREWER (1949) explained the low humidities measured in the lower stratosphere over England by postulating a meridional circulation such that air rises through the equatorial tropopause, moves horizontally polewards and finally sinks in the middle and high latitudes. The cold regions near the equatorial tropopause act as a cold trap which dries this circulating air. DOBSON (1956) accounted for seasonal variations in the total ozone over temperate and polar regions, by suggesting that the cold pool over the winter pole stores ozone-enriched air which arrives by the meridional circulation from ozone producing regions. This air subsides into the lower stratosphere of the polar and temperate latitudes in the late winter and spring. This subsidence of the ozone rich air accounts for the spring peak in total ozone at these latitudes. The spring maximum of fission product fallout was first attributed to the same circulation by STEWART ET AL. (1957). The peaks in the radioactive debris of stratospheric origin falling out at temperate latitudes (STEWART ET AL., 1957, CROOKS ET AL., 1960) were explained by a combination of this meridional circulation and air transfer from stratosphere to troposphere of those latitudes.

The measurements presented in this paper are not in accordance with this "Dobson-Brewer" stratospheric model. The tritium concentration during the period 1959 to 1960 indicates that the air at 90,000 feet is not exchanging with the troposphere at any appreciable rate. This would be consistent with the "Dobson-Brewer" circulation only if that circulation went to great heights, and took a period of several years. However, the most recent estimates of a stratospheric half residence time for radioactive debris of 1—2 years, FEELY (1960), PEIRSON ET AL., (1960), are not in accordance with this latter requirement. The high humidities found at 90,000 feet are also quite inconsistent with the former requirement.

The source of the moist air above about 80,000 feet cannot be the tropical tropopause regions because of the low temperature there. The late winter and spring polar zones are also doubtful sources, since the ozone data is consistent with sinking air there. The most likely natural source, therefore, seems to be the summer polar upper troposphere which is comparatively warm. Humidity measurements above 40,000 feet at latitudes higher than 70° would be extremely interesting in this context.

Direct transfer of water into the stratosphere at 90,000 feet by thermonuclear explosions is not considered to be a likely cause of the high humidities. This would involve the injection of the order of 4×10^9 tons of water into the middle stratosphere, assuming the high humidities to be global in extent. An energy expenditure of about 2,000 Megatons TNT equivalent would be required to evaporate this quantity of water. This is an order of magnitude greater than the energy release of all weapon tests (U. K. *Min. of Defence*, 1959), only about a third of which was associated with water bursts.

In figure 2(d) the variations of the particulate caesium-137 concentration of the air at 47,000 feet over England, measured using aircraft fitted with filters, is reproduced from PEIRSON ET AL. (1960). There is little correlation between the caesium-137 variations at 47,000 feet and those of the tritium at 90,000 feet over the same period. It may be that the factors governing the source and transfer of air in the lower stratosphere over England are quite different from those pertaining at 90,000 feet. The humidity data are certainly very strong evidence in favour of this thesis.

5. Conclusions

The humidity and tritium measurements presented in this paper indicate that the large scale meridional circulation, suggested by Brewer, does not reach heights of 90,000 feet over England.

The world-wide distribution in the stratosphere of late 1959 of the particulate tracer tungsten-185, which was injected near the

equator in mid 1958, is also difficult to reconcile with the requirements of the Brewer circulation. FEELY and SPAR (1960) suggest that lateral mixing, with the possibility of some gravitational settling, within the stratosphere best explains the tungsten-185 distribution. However, it is difficult to use this explanation to account for the presence and maintenance of high humidities at 90,000 feet compared with 50,000 feet.

It would seem, therefore, that neither of these two mechanisms of meridional transfer within the stratosphere can fully account for all the information available on the distribution of the various tracers (water, ozone, radioactive debris) found at those heights. In a subsequent paper, two of us (P. G. and F. B.) will consider this aspect in more detail.

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