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Stratospheric HTO perturbations 1980–1983

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ABSTRACT

Three perturbations of the stratospheric tritiated water burden have occurred. An atmospheric nuclear detonation in 1980 injected about 2.1 MCi. The massive eruptions of the volcano El Chichon may have contributed to a doubling of the removal rate in 1982. An unusually large wintertime exchange with the upper stratosphere may have occurred between 1982 and 1983.

1. Introduction

The data upon which this report is based were collected by Project Airstream, the aircraft component of the US Department of Energy's High Altitude Sampling Program. The project and the equipment used have been described in previous publications (Mroz et al., 1983; Mason and Östlund, 1976). Analysis for tritiated water vapor and tritium gas had been performed, prior to this work, on the contents of whole air samples collected by Airstream in high-pressure spheres, but no release of those data has been made. In 1973, the former US Atomic Energy Commission sponsored development of a catalytic-adsorptive tritium sampler by the University of Miami. It was based on technology developed for ground-level and transport-aircraft-borne samplers (Östlund and Mason, 1974). Beginning in 1975, this sampler has been used on all Airstream deployments. The sampler collects separately the two dominant tritium-containing chemical species, water vapor (HTO), and hydrogen gas (HT). Previous data have indicated that atmospheric thermonuclear detonations deposit HTO almost exclusively (Glasstone and Dolan, 1977; Mason et al., 1980), and this report is concerned with that species. A recent publication (Mason et al., 1982) used the Airstream HTO data to compare the stratospheric residence times of ^{95}Zr and HTO, and to estimate the injections of the two species by the large Chinese atmospheric tests of June 27, 1973 and Nov. 17, 1976. This is the first publication to estimate the injection by the Oct. 16, 1980 test, and is an expansion of an earlier unpublished report (Mason, 1982a). Measurements of the stratospheric sulfate aerosol burden are also a regular part of the project and have quantified the injections of recent volcanic eruptions, including those of El Chichon in 1982 (Sedlacek et al., 1983; Mroz et al., 1983).

2. Experimental results

Tritium data from Airstream deployments between April 1975 and March 1983 have been reported (Mason et al., 1980; Mason, 1982b; Mason and Östlund, 1983; Mason, 1984). The latitude spans of the October 1982 and March 1983 deployments were extended to 10 degrees south in order to assess the movement of the El Chichon aerosol across the inter-tropical convergence zone.

3. Interpretation

3.1. *Burden calculation*

The calculation of total hemispheric stratospheric burdens from the rather sparse Airstream data sets presents some difficulties. Previous estimates were made by hand-contouring and integration of the region from 15.2 km to 21.3 km between the equator and 30 degrees north, and from 9.1 km to 21.3 km between 30 and 90 degrees north. The tropopause was considered to be a zero isopleth in this technique. Some extrapolation was involved since the sampling flights provide coverage from 13.5 km to 19.2 km between the equator and 60 degrees north, and from 12.2 and 19.2 km between

60 and 75 degrees north. Vertical profiles are taken at about 7, 29, and 61 degrees north over an altitude range from 3 to 19.2 km.

Burdens for this report were calculated between the tropopause and 21.3 km for the entire hemisphere. Again, extrapolation was involved above the maximum flight altitude, and from 75 degrees to the North Pole. The impact of the extrapolation will be discussed below. Reported tropopause height data for the actual flight days were obtained for upper-air stations lying along the flight path, and smoothed to provide a continuous estimate of the tropopause over the latitude span. Tritium data below that tropopause were forced to zero in the contouring, and the region below was not included in the integration.

Calculations were done using matrix interpolation and contouring routines from the DISSPLA Version 9.0 (Integrated Software Systems Corporation) library. The interpolation consists of calculation of HTO mixing ratios at the points of a regular grid of atmospheric pressure and sine of latitude. This coordinate system has the property that each cell of the grid represents a constant zonal air mass. A 48 by 35 grid was used. The grid extends to 10 degrees south to permit use of data in that region, collected on the October 1982 and March 1983 deployments, in the contouring procedure. The contouring routine examines the neighbourhood of each grid point for data lying within a specified range, in this case 7 cells horizontally by 5 cells vertically, and calculates a value for the grid point by weighting the data according to a specified function of the distance from the grid point, here the inverse square. The integration is performed by taking the arithmetic mean of the four grid point values defining a cell as the mixing ratio within the cell, and multiplying that mixing ratio by the air mass of the cell. The resulting burdens are summed to calculate the hemispheric burden. Cells lying entirely below the tropopause or south of the equator are not included in the summation.

The northernmost cells, extending from 77.2° N to the pole, contain about 3% of the hemispheric atmosphere. The extrapolation done by the contouring routine has been found to assign no more than 85 kCi to those cells. The extrapolated burdens in those cells range from 3.4 to 7.1% of the reported total burden, and therefore cannot contribute a significant error to the following arguments.

3.2. *Injection in 1980*

Table 1 shows the measured burdens which have been decay-corrected, using the tritium half-life of 12.34 years, to October 16, 1980. This date was chosen as the common reference in order to exclude the irrelevant effect of radioactive decay from the injection and removal estimates. Also shown are calculated burdens obtained from least-squares regression of each year's data. This regression calculates the least-square fit of the logarithms of the decay-corrected burdens to the mid-dates of the deployments. The assumption of exponential removal process is implicit. Previous work (Mason et al., 1982) showed a characteristic residence time (e-fold), excluding radioactive decay, of 14.6 months with a 90% confidence range of 12.0 to 18.6 months for both ^{95}Zr and HTO in the lower stratosphere following the previous large atmospheric test. The 3 burdens preceding the October 16, 1980 test were found by the same least squares technique to have a 12.8 month characteristic residence time, and the 3 post-test burdens in 1981 were found to have a 14.7 month characteristic residence time. These regression lines were extrapolated to the test date. The discontinuity should then represent the injection estimate. This is shown in Table 1 and Fig. 1, and has a value of 2100 kCi. The uncertainty of the estimate was calculated by using the reported confidence level range (Mason et al., 1982) since

Table 1. *Stratospheric tritiated water burdens*

Mission number	Mid-date of mission	Measured burden (kCi)	Measured burden* (kCi)	Calculated burden† (kCi)
16	790715	2351	2191	2172
18	800426	1048	1020	1041
19	800802	836	826	809
‡	801016	—	—	667
§	801016	—	—	2768
21	810419	1871	1925	1829
22	810717	1302	1358	1498
23	811024	1187	1257	1200
24	820426	1256	1368	1467
25	820725	957	1057	949
26	821022	513	574	616
27	830318	891	1021	—

* Decay-corrected to October 16, 1980.

† Annual regression calculation of decay-corrected data by least-squares.

‡ Pre-test extrapolation.

§ Post-test back extrapolation.

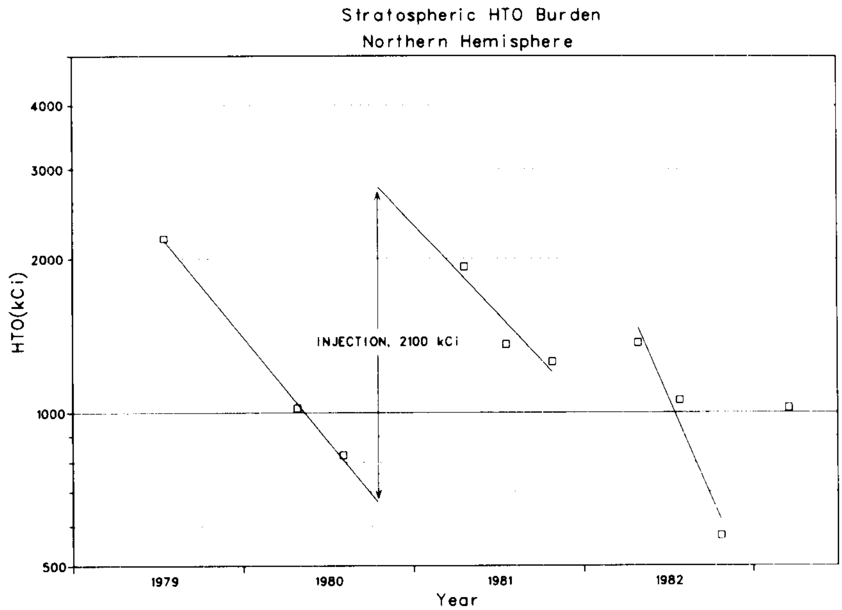

Fig. 1. Measured HTO burdens, regression lines, and injection. Burdens are decay-corrected to October 16, 1980.

three burdens are insufficient for calculation of a meaningful range. The uncertainty is +430 to −480 kCi. The estimated injection indicates a fusion yield of the device about of 0.1 MT, using a recommended value of 20 MCi/MT (NCRP, 1979). This is in reasonable agreement with the fusion yield of 0.15 MT inferred from a recent report which estimated the fission yield at 0.45 MT, and the total yield at 0.6 MT (UNSCEAR, 1982). Omission of the stratospheric burden above 21.3 km from the tritium injection calculation is believed to introduce little error, since the estimated magnitude of the test would indicate that the cloud rise would be limited to below that height (Glasstone and Dolan, 1977).

3.3. *Possible volcanic effect*

The further depletion of the burden in 1982 showed an anomalous rate by comparison to other years. The apparent characteristic time is 7.8 months for the three sampling periods in 1982, roughly half of the normal residence time.

Although the statistical uncertainty on the characteristic time is rather large, speculation as to a possible cause leads first to consideration of a direct chemical effect. Although hydration of the newly-formed aerosol consumes water, the mean mixing ratio of sulfate found by Mroz et al. (1983) after the El Chichon eruptions is only about 20 p.p.b.m. The aerosol itself is about 30% water by weight thus its formation could only affect the free water burden of the stratosphere in areas of locally high sulfur concentrations (Hofmann and Rosen, 1983). Fig 1 of Mason (1984) appears to show such an effect in the region between the equator and 20 degrees latitude. A selective removal of tritium throughout the entire stratosphere would require an isotopic effect of the order of 100 as discussed below. This line of argument further assumes that the aerosol is not collected by the tritium sampler; a reasonable assumption in view of the use of the aircraft bleed air as the supply for the sampler (Mason and Östlund, 1976).

The elapsed time between the spring and fall deployments in 1982 was 6.8 months. The required isotopic effect is estimated by first recognizing that a normal characteristic time of 14.6 months implies that exp(−6.8/14.6) or 63% of the beginning burden would remain after 6.8 months, vs exp(−6.8/7.8) or 42% in the accelerated case. The extra removal of HTO is then 21% of the beginning burden. The aerosol, being 70% H_2SO_4, is 43% H_2O. Using the 20 p.p.b.m. mean aerosol mixing ratio found by Mroz et al. (1983), the water bound in the aerosol is 0.86×10^{-8} stratospheric masses. In the absence of an isotopic effect, 37% of that water and HTO would be removed, or 0.32×10^{-8}

stratospheric masses. Assuming a mean stratospheric free water mixing ratio of 2 p.p.m.m., the aerosol-bound water is only 0.16% of the total. We have seen that 21% extra removal has occurred, which would require concentration of the tritiated water in the aerosol by a factor of order 100.

A more likely reason for the faster removal is alteration of the stratosphere-troposphere exchange rate by the radiative effect of the sulfate aerosol. The heating of the atmosphere in the 30mb region as a result of the additional volcanic aerosol has been reported by Quiroz (1983), and at 30 and 50 mb by Labitzke et al. (1983). Circulation effects from the Agung eruption of 1963 have been discussed by Dunkerton (1983).

3.4. *Winter 1982–83 exchange*

A further observation relating to possible abnormal depletion of the lower stratosphere has been made concerning the sulfate aerosol burden, which increased between November 1982 and March 1983 in the absence of significant new volcanic injections (Mroz et al., 1984). A similar observation can be made about the HTO data in this report. The burden increased by about 380 kCi in that interval, compared to 70 kCi during the previous winter. The assumed cause of any increase is the transport of air from higher altitudes bearing higher mixing ratios of the species. It is known that El Chichon injected large quantities of sulfate precursors above 20 km (Mroz et al., 1983). It has long been suspected that higher HTO mixing ratios are present above 20 km, since the highest values found in this work are located at the top of our altitude range. If the wintertime increase is the difference between the autumn and spring burdens, and the former is abnormally depleted but the latter is increased by enhanced transport from higher levels, then the difference will be larger than normal as appears to be the case here.

4. Conclusions

The tritiated water cycle of the lower stratosphere is subject to both anthropogenic and natural perturbations. The former are direct injections by atmospheric nuclear tests. Such tests provide a readily measureable tracer which appears to follow the water cycle of the region. Previous work has shown that the HTO is removed at a rate very similar to that of aerosol radioactivity. Extrapolation of measured burdens to dates of nuclear tests yields reasonable estimates of the magnitudes of fusion components of the tests.

A more speculative effect is that of natural events which alter the water circulation of the stratosphere. Tritiated water burdens measured in 1982 after the massive El Chichon volcanic eruptions decreased at a rate twice normal, and this unprecedented behavior is postulated to have been the result of alteration of the normal exchange processes between stratosphere and troposphere. A related effect may have appeared as enhanced exchange between layers of the stratosphere above and below 20 km.

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