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TITLE:

ATMOSPHERIC TRITIUM

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PROGRESS REPORT

1979-1980

A. FIELD SAMPLING DEVICES

The sampling network presently consists of slow-flow-rate samplers for tritiated water vapor (HTO) and tritium gas (HT) at Miami, Florida; Fairbanks, Alaska; and Baring Head, New Zealand. The Miami sampler has the additional capability of sampling atmospheric tritiated hydrocarbons (CH₃T).

The technique does not distinguish among the isotopic configurations; hence "HTO" includes DTO and T₂O, "HT" includes DT and T₂, and "CH₃T" includes all volatile tritiated hydrocarbons, aldehydes, and alcohols. Data from the stations for the year 1979 are contained in Tables 1-3, and shown in Figures 1-3.

Performance of the sampler at Fairbanks was below its usual standard due to mechanical failures. Sampling was unsuccessful between 79/07/12 and 79/08/25 and again between 79/09/20 and 79/12/30. Satisfactory operation has been restored; however, replacement of the sampler is planned for mid-summer 1980.

Sampling was interrupted at Baring Head by failure of the mechanical gas-meter on 79/08/06. An electronic mass flowmeter and integrator, which had originally been procured for installation at Fairbanks, was dispatched to Baring Head. Operation was restored on 79/09/06.

The Miami sampler yielded possibly erratic hydrocarbon tritium data between 79/01/22 and 79/02/19 because of undetected interruption of the methane carrier gas.

B. TRITIUM IN RAINS

We have continued our participation in the International Atomic Energy Agency's environmental isotope data network by analysis of tritium in rainfall at Miami and Barbados, West Indies. The 1979 results are contained in Tables 4-5.

C. AIRBORNE TRITIUM SAMPLING

Atmospheric HTO and HT were sampled as part of Project Airstream. Deployments were made in 1979 July and October-November, and 1980 April-May. The deployments provided samples of the stratosphere at four levels between latitudes 0° and 75°N, as well as vertical profiles between the middle troposphere and the middle stratosphere over Houston, Texas, and Fairbanks, Alaska, on some deployments.

Data from all Airstream deployments through 1979 July were the subject of a report published in the *Environmental Quarterly*, EML-371, which is reprinted as Appendix 1 to this report. Tables of the HT and HTO data, and figures of the HTO data, are contained in the appendix. Table 6 and Figure 4 of this report contain the data from the 1979 October-November deployment. The HT data continue to indicate a largely well-mixed distribution throughout the troposphere and lower stratosphere, and are not depicted. Data from 1980 April-May are not complete as of the time of writing of this report.

The 1979 April deployment was highly successful. The 1979 July deployment achieved the majority of its objectives despite cancellation of the Panama segment due to the political situation in Nicaragua. The 1979 October-November deployment also achieved the majority of its objectives, although an aircraft malfunction in Alaska forced a long delay in completion of the return flights to McChord AFB, Washington, and Ellington AFB, Texas, and omission of samples above 13.7 km altitude on those flights.

A modification to the sampler for improved hydrogen flow control was completed prior to the 1980 April-May deployment, and was satisfactory.

D. STRATOSPHERIC BALLOON SAMPLER

The sampler was tested in the Environmental Measurements Laboratory altitude chamber on 80/02/05, and performed in a highly satisfactory manner. It was then flown as a piggyback payload on Ashcan flights on 80/04/04 and 80/04/10 at Holloman AFB, New Mexico. Results of these flights are contained in Table 7, and are consistent with previous inferences that a reservoir of HTO remains in the middle stratosphere from the very large atmospheric nuclear tests of the 1960s.

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H. STRATOSPHERIC ¹⁴CO₂ COLLECTION

In cooperation with the Environmental Measurements Laboratory, an attempt was made to utilize the WB-57F tritium sampler for collection of stratospheric CO₂ simultaneously with H₂O. For this purpose, two traps containing 300g of type 4A molecular sieve (Linde) were connected in series, and flown on an Airstream vertical profile on 80/05/13.

Samples were taken at six altitudes from 30000 ft (9.2 km) to 63000 (19.2 km). Apparently quantitative recovery of CO₂ was achieved in the laboratory. Collection of more than 1 g of water in the first trap coincided with collection of an appreciable portion of the CO₂ in the second trap, indicating displacement of the CO₂ by the more strongly adsorbed water.

Results of the extraction are shown in Table 8. Samples were shipped to the Argonne National Laboratory for ¹⁴C determination.

I. DISCUSSION OF SIGNIFICANT FINDINGS

1. Tropospheric HT: The inventory appears to have stabilized since early 1978. Although fluctuations appear in the quarterly means, the annual mean has decreased only 8% since 1978.

2. Stratospheric HTO: A detailed discussion appears in Appendix 1. The most significant finding is that the e-folding time of the HTO removal from the lower stratosphere is 14 months, and agrees well with that for ⁹⁵Zr.

3. Global atmospheric tritium inventory: The global inventories of HTO and HT were estimated under the following assumptions:

a. The Fairbanks data are representative of the northern troposphere from latitudes 45° to 90°.

b. The Miami data similarly represent latitudes 5° to 45°, with a factor of 0.95 used to compensate for the usual decreasing gradient southward.

c. The Baring Head data represent the southern troposphere, from the ITCZ (5°N) southward.

d. The inventories calculated in Appendix 1 for the July deployments represent the northern stratosphere.

e. The southern stratosphere, in steady state, has 10% lower mixing ratios of both HTO and HT than the northern stratosphere.

f. For 1977 only, the southern stratosphere had a 25% lower HTO mixing ratio than the northern stratosphere, due to the limited time for the 1976 PRC input to be transported across the equator.

The estimates are listed in Tables 9 and 10. Quarterly means are shown in Figure 5.

J. PUBLICATIONS AND SYMPOSIA

A report based on the 1975-1979 Airstream data was published in the *Environmental Quarterly*. It is attached as Appendix 1 of this report. A paper was given at the American Nuclear Society's National Topical Meeting on Tritium Technology in Fission, Fusion, and Isotopic Applications. The paper, entitled "Environmental Tritium Applications to Atmospheric and Oceanographic Research", was based partly on this work. A preprint is attached as Appendix 2 of this report.

K. ACKNOWLEDGMENTS

The analysis of Airstream data contained in Appendix 1 was made possible by the cooperation of Dr. Gert Hut of the Isotope Physics Laboratory, State University of Groningen, The Netherlands, who was in our laboratory under a Fulbright scholarship, and of Mr. Kosta Telegadas of the Air Resources Laboratory, National Oceanic and Atmospheric Administration, who gave us much time and guidance as well as many hours of analysis. Dr. Hut also participated in the $^{14}\text{CH}_4$ work.

TABLE 1 ATMOSPHERIC HT, HTO, HYDROCARBON TRITIUM, MIAMI 1979

Location: Virginia Key, Miami FL
 Elevation 12 m; 25 47 N 80 11 W

Explanation of heading: Sample is an arbitrary serial number, except that when a date is shown, it is the start of a combined sample; Date is the starting date of the sample run which is normally of 48 hours. It is the starting date of the last component of a combined sample; Temp is the mean of starting and ending temperatures at the station in Centigrade; RH is the percentage relative humidity based upon Temp., mean barometric pressure, and the weight of the water vapor sample obtained. It may exceed 100% under conditions of widely-varying temperature. AH is the absolute humidity (grams water per cubic meter of air) based upon the above parameters; Vap TU is the activity of the water vapor sample; HTO AT and HT AT are the mixing ratios of HTO and HT in units of T-atoms per mg air; HC AT is the mixing ratio of tritium other than HTO and HT in the same units, and sig is the one-sigma error of the quantity preceding it. Water vapor samples were combined in the ratio of the water mixing ratios (grams water per kg air) for a monthly composite.

Sample	Date	Temp	RH	AH	Vap TU sig	HTO AT sig	HT AT sig	HC AT sig
1104	790101	17.8	89	13.7			39.6 1.6	3.9 0.2
1105	790104	21.6	55	10.7			41.2 1.7	4.2 0.2
1106	790108	17.9	82	12.7			38.3 1.7	4.5 0.2
1107	790111	21.4	86	16.3				9.2 0.4
1108	790115	22.5	63	12.9			40.5 1.2	3.8 0.2
1109	790118	22.0	65	12.9				3.7 0.3
1110	790122	18.2	77	12.1			41.7 1.8	4.1 0.7
1111	790125	17.8	44	6.8				3.8 1.1
1112	790129	19.7	45	7.8				
790101	790129			11.8	7.3 0.2	4.8 0.2		

TABLE 1 MIAMI (Cont.)

Sample	Date	Temp	RH	AH	Vap TU sig	HTO AT sig	HT AT sig	HC AT sig
1113	790201	12.5	45	5.1			58.1 2.2	3.8 1.6
1114	790205	21.9	76	14.9			41.0 2.2	3.8 1.9
1115	790208	13.6	10	1.2				4.1 1.1
1116	790212	21.1	44	8.3			40.0 1.6	4.1 1.1
1117	790215	21.7	68	13.1				3.9 1.9
1118	790219	23.9	64	14.2			42.2 1.2	4.3 2.1
1119	790222	23.6	95	20.4				4.3 0.3
1120	790226	20.0	48	8.4			40.5 1.5	4.1 0.2
790201	790226			11.6	8.2 0.3	5.4 0.2		
1121	790301	22.5	60	12.2				4.6 0.2
1122	790305	21.1	73	13.8			38.6 1.5	4.0 0.3
1123	790308	20.5	66	12.0				4.7 0.2
1124	790312	23.3	57	12.1			43.1 2.0	9.0 0.3
1125	790315	20.0	57	10.1				6.7 0.3
1126	790319	21.6	56	10.9			43.0 1.7	
1127	790322	23.9	72	15.9				3.9 0.2
1128	790326	18.3	64	10.2			40.5 1.6	4.0 0.2
1129	790329	22.8	43	8.9				4.3 0.2
790301	790329			11.8	9.0 0.3	5.9 0.2		
1130	790402	24.5	70	16.1			39.5 1.6	3.8 0.2
1132	790407	25.0	42	9.9			57.7 1.9	4.0 0.2
1133	790409	25.2	76	18.0				3.6 0.2
1134	790412	25.5	72	17.4			36.8 1.6	4.1 0.2
1135	790416	25.4	56	13.6				4.1 0.2
1136	790419	25.3	55	13.1			45.0 1.8	3.7 0.2
1137	790423	24.1	74	16.5				3.8 0.2
1138	790425	24.9	75	17.4			36.5 1.5	3.9 0.2
1139	790430	22.6	90	18.3				4.0 0.3
790407	790430			15.5	9.9 0.3	8.8 0.3		

TABLE 1, MIAMI (Cont.)

Sample	Date	Temp	RH	AH	Vap TU sig	HTO AT sig	HT AT sig	HC AT sig
1140	790503	25.5	71	17.1			36.9 1.3	3.6 0.2
1141	790507	26.1	81	20.3				4.1 0.2
1142	790510	25.8	74	18.1			37.9 1.5	4.2 0.2
1143	790514	25.5	75	18.1				4.8 0.3
1144	790517	23.2	72	15.3			40.1 1.2	4.9 0.3
1145	790520	25.8	60	14.7				4.6 0.2
1146	790524	23.0	85	17.8				3.9 0.2
1147	790528	26.7	80	20.6			35.5 1.5	4.0 0.2
1148	790531	27.1	76	20.0			38.9 1.7	4.2 0.2
790503	790531			18.0	7.4 0.4	7.7 0.4		
1149	790604	27.4	76	20.4				5.9 0.3
1150	790607	27.7	76	20.9			40.2 1.8	5.7 0.3
1151	790611	26.6	59	15.2			39.7 1.7	
1152	790614	26.0	86	21.3				
1153	790618	27.7	73	20.0				6.0 0.3
1154	790621	28.2	69	19.3			36.1 1.5	4.1 0.3
1155	790625	28.8	69	20.2				7.4 0.3
1156	790628	27.7	37	10.1			37.5 1.4	4.0 0.2
790604	790628			19.6	7.4 0.4	8.4 0.4		
1157	790702	29.4	72	21.7				4.2 0.2
1158	790705	28.2	77	21.5			36.5 1.6	4.0 0.2
1159	790709	29.7	69	21.0				3.9 0.3
1160	790712	28.2	80	22.4			32.6 1.5	3.4 0.2
1161	790716	27.4	77	20.7				4.1 0.2
1162	790719	28.1	74	20.6			35.0 1.1	3.8 0.2
1163	790723	26.9	81	21.0				3.7 0.2
1164	790726	28.0	73	20.2			34.9 1.3	3.7 0.2
1165	790730	27.4	78	21.0				3.8 0.2
790702	790730			21.1	5.9 0.2	7.2 0.2		

TABLE 1 MIAMI (Cont.)

Sample	Date	Temp	RH	AH	Vap TU sig	HTO AT sig	HT AT sig	HC AT sig		
1166	790802	28.3	72	20.3			33.9	1.4	5.3	0.3
1167	790806	26.3	83	21.0					5.0	0.3
1168	790809	27.4	77	20.7			28.9	1.5	13.8	0.6
1169	790813	29.4	70	21.2					5.9	0.3
1170	790816	28.6	73	21.1			26.8	1.2	12.7	0.6
1171	790820	28.0	77	21.2					5.2	0.3
1172	790823	27.2	77	20.4			35.7	1.9		
1173	790827	26.9	80	20.8					3.4	0.2
1174	790830	26.4	86	21.8			30.8	2.3	2.4	0.2
790802	790830			20.9	4.0	0.2	4.9	0.2		
1175	790904	26.6	84	21.4					4.0	0.2
1176	790906	27.7	74	20.2			37.7	1.4	3.6	0.2
1177	790910	28.5	78	22.3					3.2	0.2
1178	790913	27.1	84	22.2			33.1	1.4	3.2	0.2
1179	790917	26.6	83	21.2					2.8	0.2
1180	790920	26.9	87	22.7			31.7	1.3	2.9	0.2
1181	790924	26.9	80	20.9					2.5	0.2
1182	790927	25.5	79	19.1			33.3	1.2	2.8	0.2
790904	790927			21.2	4.6	0.2	5.7	0.3		
1183	791001	24.6	89	20.3					3.0	0.2
1184	791003	24.9	87	20.3			32.5	1.3	3.1	0.2
1185	791008	24.7	71	16.4					2.6	0.2
1186	791011	24.4	79	18.0						
1187	791015	25.8	84	20.6					2.7	0.2
1188	791018	26.0	80	19.8			34.2	1.3	2.9	0.2
1189	791025	24.4	73	16.6						
1190	791029	25.2	67	16.0			34.6	1.3	2.8	0.2
791001	791029			18.4	6.3	0.3	6.7	0.3		

TABLE 1 MIAMI (Cont.)

Sample	Date	Temp	RH	AH	Vap TU sig	HTO AT sig	HT AT sig	HC AT sig
1191	791101	25.0	95	22.2				2.7 0.2
1192	791105	23.8	77	16.8			36.8 1.7	3.3 0.2
1193	791108	22.5	94	19.1				3.0 0.2
1194	791112	21.6	104	20.0			43.9 1.6	3.4 0.2
1195	791115	20.0	66	11.7				2.8 0.2
1196	791119	24.5	66	15.0			36.9 1.6	2.7 0.2
1197	791121	24.7	66	15.2				2.5 0.2
1198	791126	23.8	92	20.1			32.5 1.5	2.3 0.2
1199	791129	19.1	54	9.0				
791101	791129			16.4	5.1 0.2	4.7 0.2		
1200	791203	21.0	72	13.4			32.6 1.6	2.6 0.2
1201	791206	22.7	97	19.8				2.1 0.2
1202	791210	23.5	74	15.9			32.7 1.3	2.8 0.2
1203	791213	22.1	91	18.0				3.6 0.3
1204	791217	18.8	67	11.0			34.9 1.5	2.1 0.2
1205	791220	22.0	61	12.1				2.5 0.2
1206	791224	19.5	53	9.1			31.0 1.0	2.9 0.2
1207	791227	19.5	67	11.4				2.3 0.2
1208	791231	14.8	80	10.3			33.8 1.1	2.4 0.2
791203	791231			13.4	7.0 0.3	5.3 0.2		

TABLE 2 ATMOSPHERIC HT AND HTO ALASKA 1979

Location: Fairbanks, Alaska

Elevation 300 m; 64 55 N 147 45 W

Explanation of heading: See Table 1. Sample run was generally 27-28 hours.

Sample	Date	Temp	RH	AH	Vap	TU	sig	HTO AT	sig	HT AT	sig
2100	790102	-19.0	103	1.2	36	4		2.1	0.3	45.9	2.2
2102	790111	-16.0	91	1.3	37	4		2.4	0.2	41.3	1.9
2104	790118	-18.0	97	1.2	45	4		2.8	0.3	40.1	1.8
2106	790125	-18.0	83	1.0	31	5		1.6	0.2	39.1	1.6
2108	790201	-24.0	73	0.6	48	11		1.3	0.3	39.6	1.9
2110	790208	-36.0	129	0.3						39.4	1.8
2112	790215	-27.0	72	0.4	91	19		1.8	0.3	43.9	2.0
2114	790222	-22.0	86	0.8	60	8		2.2	0.3	39.8	1.8
2115	790229	-18.5	163	2.0	50	4		4.8	0.4	39.4	1.5
2116	790301	-16.0	51	0.8	61	8		2.3	0.3	40.5	1.8
2118	790308	-12.5	116	2.3	54	3		6.1	0.4	44.9	1.9
2120	790315	-12.5	98	1.9	64	5		6.1	0.5	44.3	1.7
2122	790319	-5.0	133	4.5	41	2		9.4	0.5	43.8	1.9
2124	790404	-2.5	117	4.8	49	4		12.3	1.0	42.6	2.0
2126	790409	-4.5	45	1.6	76	5		6.3	0.4	42.8	2.1
2128	790416	-6.0	61	2.0	109	6		10.9	0.6	42.4	1.8
2130	790423	6.0	57	4.2	46	3		10.2	0.8	59.9	2.4
2132	790430	19.0	24	4.1	70	3		15.9	0.6	65.4	2.5
2136	790514	13.0	34	4.0	56	4		12.2	1.0	51.8	2.3
2138	790521	20.0								67.6	3.1
2140	790529	18.0	50	7.9	92	3		40.9	1.3	40.2	1.9
2144	790613	12.0	58	6.2	58	4		19.9	1.3	34.8	2.1
2146	790618	16.0	60	8.3	55	5		25.6	2.2	38.2	1.3
2148	790625	13.0	62	7.1	83	3		32.7	1.2	36.4	1.5
2151	790702	20.0	49	8.6	77	3		37.4	1.4	36.4	1.4
2152	790712	18.0	47	7.4	86	3		33.3	1.1	31.9	2.7
2153	790825	22.0	56	11.1	73	3		45.8	2.1	33.2	1.5
2154	790830	9.0	92	8.2	62	3		28.0	1.3	32.8	2.6
2156	790906	11.0	70	7.1	71	2		27.3	1.0		
2158	790913	5.0	63	4.3	53	3		12.3	0.7	116.3	4.3
2160	790920	4.0	108	6.9	32	2		11.9	0.7	29.5	0.8
2166	791230	-36.0	99	0.3	85	22		1.0	0.2	36.1	1.7

TABLE 3 ATMOSPHERIC HT AND HTO NEW ZEALAND 1979

Location: Baring Head Lighthouse, New Zealand
Elevation 74 m; 41 24 S 174 52 E

Explanation of heading: See Table 1. Sample run was generally 24 hours.

Sample	Date	Temp	RH	AH	Vap	TU	sig	HTO	AT	sig	HT	AT	sig
4079	790104	15.5	54	7.2							28.6		1.1
4080	790111	20.2	65	11.6							26.3		1.2
4081	790118	20.5	52	9.4							25.9		1.0
4082	790126	19.6	67	11.5							43.3		1.5
790104	790126			10.0	4.5	0.5		2.5	0.3				
4083	790202	19.9	57	10.0							26.3		0.9
4084	790207	19.9	52	9.1							27.0		1.1
4085	790212	17.4	64	9.7							27.6		1.0
4086	790215	17.4	64	9.6							29.8		1.2
4087	790222	14.5	65	8.2							53.8		2.2
790202	790222			9.3	5.6	0.7		2.9	0.4				
4088	790301	11.5	72	7.5							28.8		1.2
4089	790308	21.5	62	11.9							42.6		1.7
4090	790316	13.3	70	8.2							28.1		1.2
4091	790322	17.4	72	10.9							26.8		1.3
4092	790330	11.5	71	7.4							30.3		1.3
790301	790330			9.2	5.0	0.3		2.6	0.2				
4093	790406	17.6	71	10.9							31.9		1.3
4094	790409	17.8	64	9.9							29.0		1.2
4095	790417	10.8	75	7.5							25.9		1.2
4096	790423	15.9	66	9.1							27.9		1.2
4097	790430	13.4	48	5.7							28.8		1.2
790406	790430			8.6	6.1	0.4		2.9	0.2				
4098	790503	17.1	51	7.6							37.1		1.5
4099	790509	13.7	63	7.6							25.5		1.4
4100	790514	12.8	52	5.9									
4101	790523	9.2	65	5.9							26.6		1.1
790503	790523			6.7	5.3	0.4		2.0	0.1				
4102	790605	8.8	66	5.8	5.7	1.4		1.8	0.4		24.6		1.1
4103	790706	8.0	61	5.1							25.9		1.1
4104	790715	8.9	57	5.1							27.3		1.2
4105	790719	10.6	63	6.2							26.3		1.1
4106	790723	8.6	80	6.9							25.3		1.0
4107	790726	10.1	69	6.6							25.9		1.0
4108	790730	9.8	73	6.9							26.2		1.1
790706	790730			6.1	4.9	0.4		1.6	0.1				
4109	790802	11.8	74	7.9	3.1	1.3		1.5	0.5		25.6		1.1
4111	790906	13.5	62	7.4									
4112	790912	15.0	69	9.0							28.1		1.1
4113	790919	12.2	53	5.8							27.6		1.1
4114	790926	10.5	51	5.0							25.5		1.0
790906	790926			6.8	7.1	0.4		2.7	0.1				

TABLE 3 NEW ZEALAND (Cont.)

Sample	Date	Temp	RH	AH	Vap TU sig	HTO AT sig	HT AT sig
4115	791001	12.5	78	8.6			27.8 1.1
4116	791004	9.7	71	6.7			27.3 1.1
4117	791008	12.5	79	8.8			27.7 1.2
4118	791011	12.5	69	7.7			27.3 1.2
4119	791018	14.7	54	6.9			27.4 1.1
4120	791026	14.4	59	7.4			26.5 1.1
791001	791026			7.7	5.8 0.3	2.4 0.1	
4121	791101	16.5	55	7.8			26.8 1.2
4122	791108	13.6	61	7.3			26.1 1.1
4123	791115	10.0	63	6.0			27.7 1.1
4124	791122	16.6	72	10.3			26.9 1.3
4125	791126	17.4	58	8.7			24.5 1.2
4126	791129	17.1	65	9.6			
791101	791129			8.3	4.4 0.5	2.0 0.2	
4127	791206	16.9	71	10.4			27.6 0.9
4128	791213	16.2	61	8.6			27.9 1.1
4129	791221	16.3	58	8.1			24.8 1.2
4130	791227	14.5	69	8.8			31.6 1.0
791206	791227			9.0	5.1 0.6	2.6 0.3	

TABLE 4 TRITIUM IN RAINS MIAMI 1979

Station Location: Rosenstiel School of Marine and Atmospheric Science
Virginia Key, Miami, Florida
Rooftop of TRITIUM LABORATORY
Elevation 12 m: 25°47'N 80°11'W

Month	mm	TU \pm σ	GR #
January	34.16	4.75 \pm 0.17	79-2069
February	17.33	8.10 \pm 0.28	79-2117
March	5.41	8.77 \pm 0.55	79-8167
April	130.66	6.12 \pm 0.18	79-2218
May	131.25	7.72 \pm 0.23	79-8267
June	74.58	8.46 \pm 0.39	79-2316
July	81.20	5.60 \pm 0.18	79-7324
August	123.33	4.76 \pm 0.18	79-7457
September	173.75	6.70 \pm 0.23	79-7465
October	190.83	4.59 \pm 0.20	79-8522
November	89.0	6.22 \pm 0.25	79-8565
December	81.66	4.50 \pm 0.17	80-4025

Measured at the Tritium Laboratory
Rosenstiel School of Marine & Atmospheric Science
University of Miami, Miami, Florida 33149

TABLE 5 TRITIUM IN RAINS BARBADOS 1979

Station Location: Grantley Adams International Airport
 Seawell, Christ Church, Barbados
 Elevation 50 m: 13°04'N 59°29'W

Month	mm	TU \pm σ	GR #
January	21.8	2.96 \pm 0.17	79-4123
February	8.7	*5.31 \pm 2.00	79-1121
March	68.3	4.00 \pm 0.17	79-7209
April	42.9	3.60 \pm 0.15	79-4248
May	13.9	4.27 \pm 0.56	79-8272
June	149.9	7.31 \pm 0.89	79-8353
July	153.9	6.16 \pm 0.48	79-7463
August		NO SAMPLE RECEIVED	
September		NO SAMPLE RECEIVED	
October	129.1	4.01 \pm 0.28	79-7559
November		NO SAMPLE RECEIVED	
December		NO SAMPLE RECEIVED	

* Very small sample quantity.

Measured at the Tritium Laboratory
 Rosenstiel School of Marine & Atmospheric Science
 University of Miami, Miami, Florida 33149

TABLE 6

Stratospheric HT and HTO from Project Airstream
Mission A-17, 1979 October-November

MSN denotes the mission number, FLT is the flight number from the Project Support Plan, SX# is the sequential sample number on each flight. LAT and LON are the mid-latitude and mid-longitude of the sample, ALT is the pressure altitude, PRES is the pressure level, TEMP is the ambient temperature, PT is the potential temperature, and T-ATOMS/MG AIR are the mixing ratios of HTO and HT together with their one-sigma error estimates.

MSN	FLT	SX#	LAT	LON	ALT km	PRES mb	TEMP (C)	PT (K)	T-ATOMS/MG AIR			
			--S	--E					HTO	sig	HT	sig
17	01	001	28.3	95.1	9.1	302	-38.4	330	57.5	1.4		
17	01	002	28.5	95.3	12.2	188	-56.6	349	6.9	0.6		
17	01	003	28.5	95.3	13.7	148	-63.8	362	6.9	0.6		
17	01	004	27.7	95.8	15.2	116	-69.1	377	12.0	0.6		
17	01	005	28.8	95.1	16.8	92	-69.2	404	5.4	0.6		
17	01	006	27.5	96.2	19.2	63	-56.6	478	30.9	1.3		
17	03	001	18.0	84.3	15.2	116	-71.4	373	19.9	1.1	29.6	1.4
17	05	001	1.0	79.6	15.2	116	-77.9	361	53.3	3.2	33.7	1.0
17	05	002	1.0	79.6	16.8	92	-77.9	386	16.5	0.8	31.5	1.5
17	05	003	1.5	79.6	18.3	72	-75.8	418	34.4	2.4	38.4	1.7
17	05	004	1.5	79.8	19.2	63	-70.9	447	99.4	4.1	42.9	2.5
17	08	001	39.0	92.9	16.8	92	-61.5	419	20.5	1.0	32.7	1.5
17	08	002	39.0	92.5	19.2	63	-53.9	484	71.0	2.8	31.4	1.5
17	09	001	39.1	109.1	15.3	116	-65.4	385	63.0	2.1	27.6	0.9
17	09	002	49.0	125.9	15.1	118	-56.8	398	90.2	1.8	30.2	1.2
17	10	001	49.0	126.2	13.7	148	-51.4	383	2.6	0.4	30.7	1.1
17	11	001	66.1	148.4	12.2	189	-53.3	354	22.8	1.1	27.6	1.2
17	11	002	74.0	147.1	12.2	188	-52.8	355	28.0	1.3	24.1	1.1
17	11	003	74.0	148.2	16.9	90	-48.9	446	122.4	4.4	32.7	1.5
17	11	004	66.0	148.2	16.8	92	-49.0	444	167.2	5.9	30.6	1.5
17	12	001	66.0	149.8	15.2	116	-48.7	415	67.4	2.8	27.4	1.2
17	12	002	74.0	149.5	15.2	116	-50.9	411	78.3	3.2	32.5	1.1
17	12	003	74.0	149.6	18.8	67	-48.3	486	379.8	15.5	38.2	1.7
17	14	001	49.2	125.9	13.7	148	-65.5	358	97.2	3.7	31.7	1.3
17	15	001	39.0	107.2	13.7	148	-63.5	362			30.6	0.8

TABLE 7 STRATOSPHERIC HTO

Location: Holloman AFB, NM vicinity
33°N 107°W

Explanation of heading: Flight No. is assigned by the launching agency; Date is the launch date; Altitude is as reported by the launching agency; T-Atoms/mg air is the mixing ratio of HTO computed from the activity and quantity of the water sample flushed from the trap, and the quantity of air sampled as measured after the flight. The error is based on the 1- σ counting error and an estimated 5% error in air quantity measurement.

<u>Flight No.</u>	<u>Date</u>	<u>Altitude,km</u>	<u>T-Atoms/mg air</u>
H80-15/H-159	800404	26.1	699 \pm 51
H80-16/H-160	800410	23.0	1055 \pm 67

TABLE 8 STRATOSPHERIC CO₂ COLLECTIONLocation: Ellington AFB, TX vicinity
29°N 95°W

Date: 800513

Sample No.	1	2	3	4	5	6
Altitude, ft.	30000	40000	45000	50000	55000	63000
First sieve						
H ₂ O, g	11.79	1.48	0.44	0.48	1.14	2.00
CO ₂ , g	1.40	1.84	1.54	1.84	1.66	1.46
Second sieve						
H ₂ O, g	0.00	0.05	0.01	0.05	0.18	0.14
CO ₂ , g	0.46	0.39	0.08	0.10	0.15	0.27
Total						
H ₂ O, g	11.79	1.53	0.45	0.53	1.32	2.14
CO ₂ , g	1.86	2.23	1.62	1.94	1.81	1.73
Air, liters STP*	2933	3281	2355	3044	2801	2846
CO ₂ , liters STP*	1.01	1.22	0.88	1.06	0.99	0.94
Apparent CO ₂ v/v x 10 ⁶	346	371	375	348	353	332

Note: Air volume based on preflight calibration only.
Postflight calibration may result in changes in
air volume and CO₂ mixing ratio.

*STP = 20°C, 1013 mb

TABLE 9 GLOBAL HTO INVENTORY

Year	Northern Hemisphere				Southern Hemisphere			
	Troposphere		Stratosphere††		Troposphere		Stratosphere	
	$\frac{\text{T-atoms}}{\text{mg air}}$	grams T	$\frac{\text{T-atoms}}{\text{mg air}}$	grams T	$\frac{\text{T-atoms}}{\text{mg air}}$	grams T	$\frac{\text{T-atoms}}{\text{mg air}}$	grams T
1976	7.6	70	68	210	2.5	26	61†	203†
1977	13.5*	120*	636*	930*	4.4*	50*	477†*	600†*
1978	12.5*	110*	135*	410*	2.1	22	122†*	407†*
1979	9.2	82	85*	260*	2.3	23	77†*	257†*

†Estimate from northern hemisphere data.

*Affected by PRC input.

††Note change in method of calculation from previous reports, see text.

TABLE 10 GLOBAL HT INVENTORY

Year	Northern Hemisphere				Southern Hemisphere			
	Troposphere		Stratosphere		Troposphere		Stratosphere	
	$\frac{\text{T-atoms}}{\text{mg air}}$	grams T	$\frac{\text{T-atoms}}{\text{mg air}}$	grams T	$\frac{\text{T-atoms}}{\text{mg air}}$	grams T	$\frac{\text{T-atoms}}{\text{mg air}}$	grams T
1976	48.8	435	45.0	137	40.0	415	40.5†	135†
1977	41.4	370	43.0	131	36.5	380	38.7†	129†
1978	40.0	357	37.0	113	31.8	330	33.3†	111†
1979	38.5	342	36.3	111	28.5	290	32.7†	109†

†Estimate from northern hemisphere data.

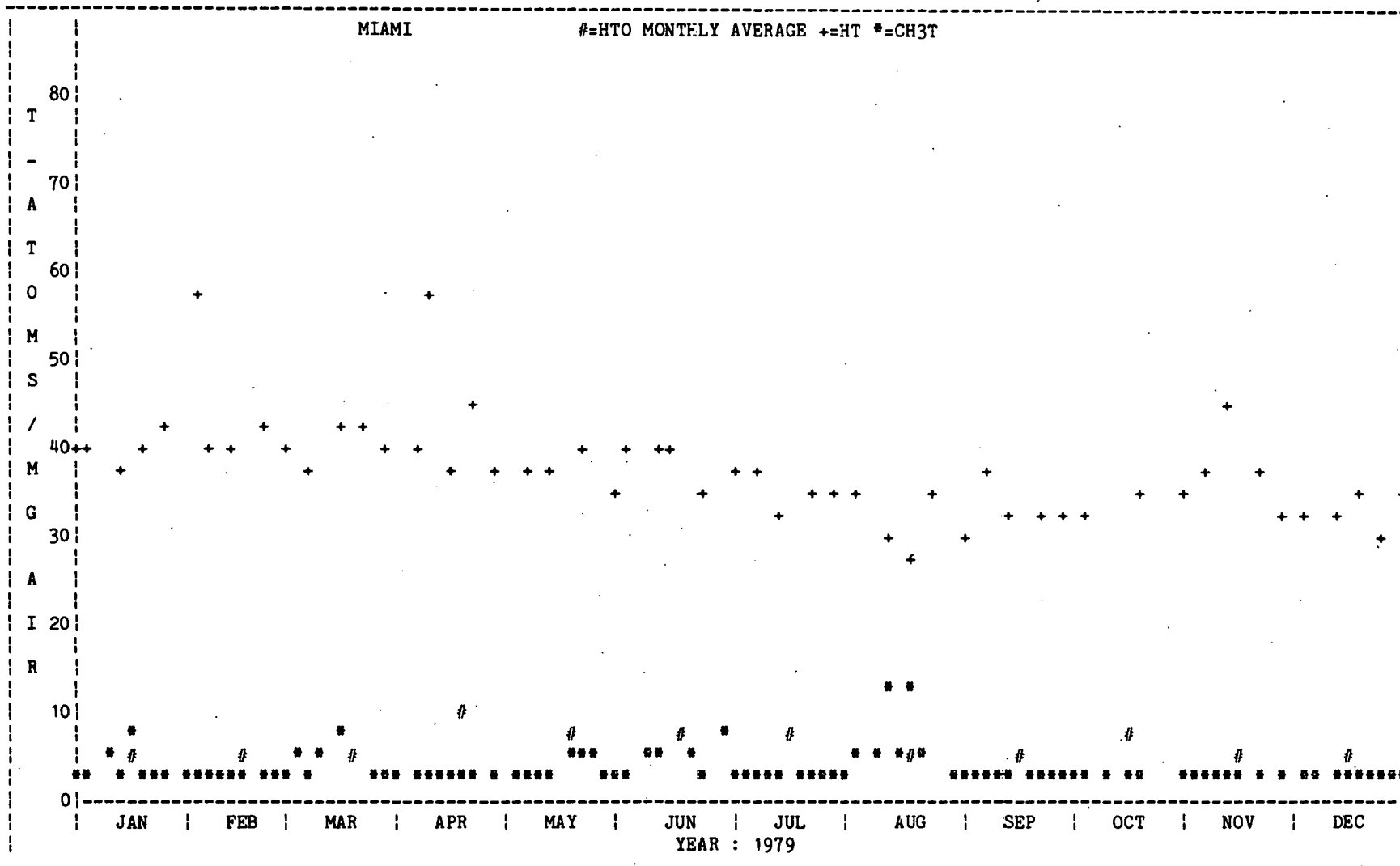


Fig. 1 Atmospheric tritium mixing ratios at Miami, FL 1979.

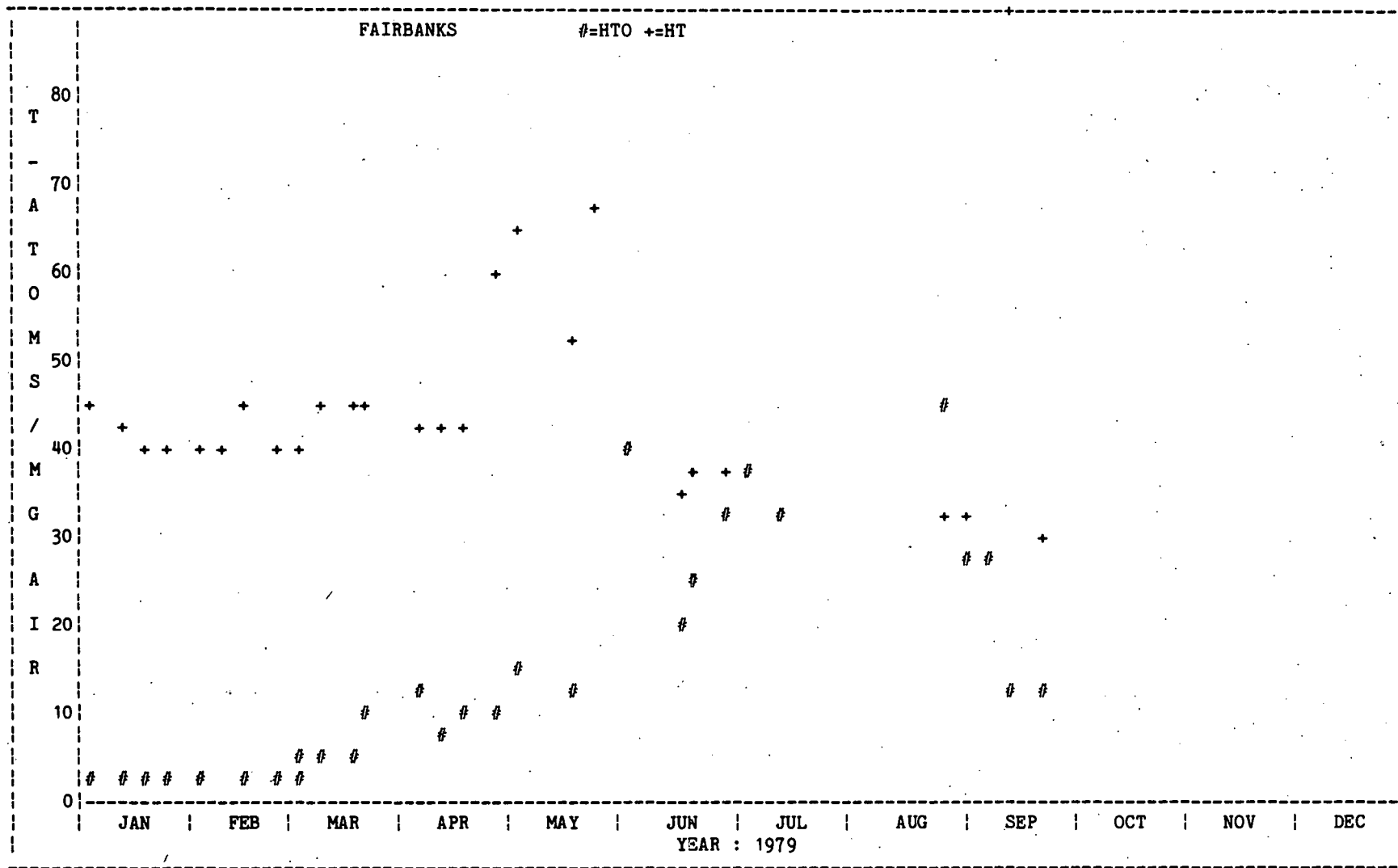


Fig. 2 Atmospheric tritium mixing ratios at Fairbanks, AK. 1979.

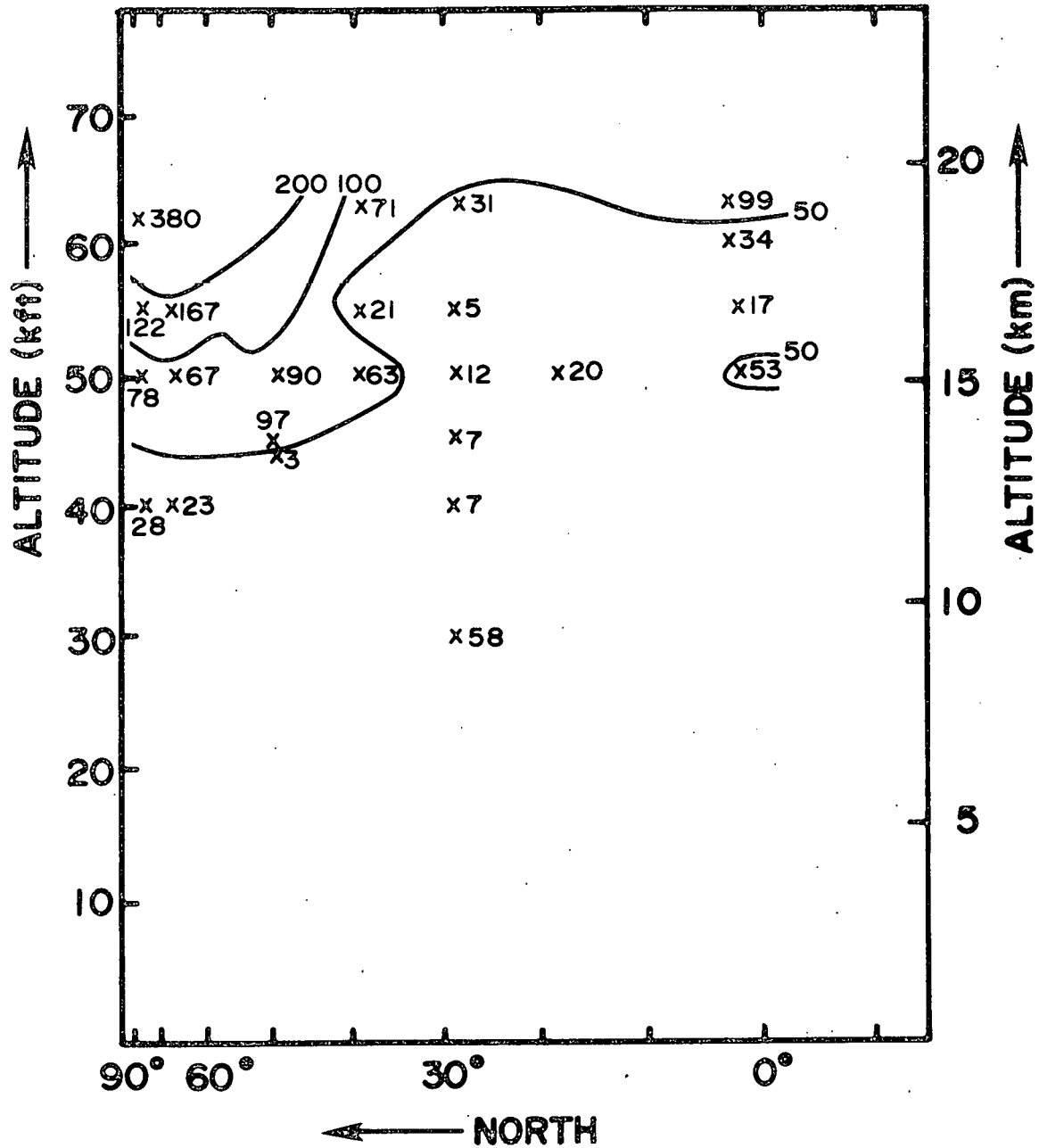


Fig. 4 Atmospheric HTO mixing ratios in T-atoms per mg air, from Project Airstream deployment 1979 Oct 10 - Nov 18.

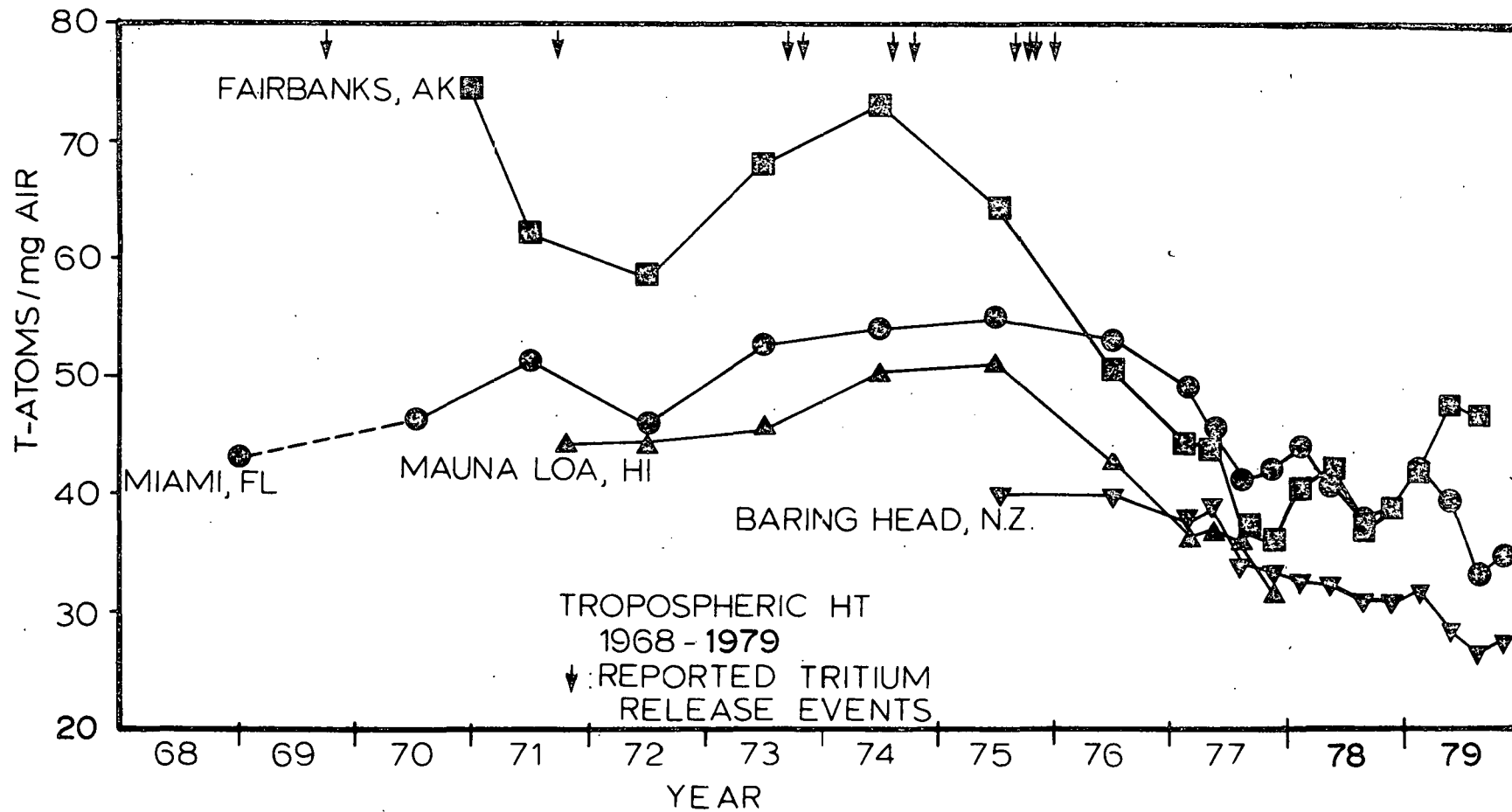


Fig. 5 Quarterly mean tropospheric HT mixing ratios, 1968-1979.

COMPARISON OF STRATOSPHERIC TRITIUM (AS HTO) AND ZIRCONIUM-95
BURDENS FROM THE HIGH YIELD CHINESE NUCLEAR TESTS
OF JUNE 27, 1973 AND NOVEMBER 17, 1976

by

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ABSTRACT

The depletion of the stratospheric burdens of particulate Zr-95 and gaseous HTO attributed to the November 17, 1976 Chinese high yield test indicates both have about the same residence half time (10 months) for up to 3 years after input. This indicates that gravitational settling of particles in the lower stratosphere can be considered to be negligible in studying transport processes.

The rate of depletion of the stratospheric burden of HTO from the high yield Chinese test of June 27, 1973 is not as conclusive, in part, due to greater uncertainties in calculating the stratospheric burdens.

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1. INTRODUCTION

The Tritium Laboratory of the University of Miami began programs of measurement of tritiated water vapor (HTO) and tritium gas (HT and T₂) collected at ground level in 1968, and from aircraft in 1971. Previous publications have described the techniques and interpreted the data (Östlund and Mason, 1974; Mason and Östlund, 1976, 1979; Mason, 1977). Data through 1976 are available in unpublished reports (Östlund, Mason and Ydfalk, 1972; Mason and Östlund, 1977).

Project Airstream (sponsored by the Department of Energy) is a long-term study of stratospheric radioactivity and chemistry, carried out by three series of flights annually, covering the latitude range from the equator to 75° north. Four flight levels above the tropopause are sampled over that span; in addition, vertical profiles have been flown in the vicinity of Panama, Republic of Panama; Houston, Texas, and Anchorage, Alaska, for each flight series since July 1977. The vertical profiles take samples from 3 to 19 km altitude, and the transects are made between 14 and 19 km.

Tritium measurements have been made as part of Project Airstream since 1975, however, the project is older than that, having begun in 1965 with sampling of particulate and noble gas radioactivity.

Zirconium-95 is a particulate fission product with a 65-day half-life, produced by all nuclear tests, while tritium is a gas, with a half-life of 12.26 years, produced by the fusion reaction in a thermonuclear device.

Measurements of Zr-95 have been used extensively to estimate stratospheric residence times, initial vertical activity distributions and atmospheric transport from the high yield nuclear tests conducted by China

between 1967 and 1976 (Telegadas, 1974; 1976; 1979). These data can now be compared with the tritium (as HTO) data to try to resolve the question of the significance of particle settling in studying transport processes in the upper atmosphere.

2. EXPERIMENTAL TECHNIQUES

The gas sampler flow diagram is shown in Fig. 1 (Mason and Östlund, 1976). Bleed air from the WB-57F airplane passes through a pressure regulator and a mass flowmeter into a manifold. Sampling is controlled from the cockpit by opening one of six pairs of solenoid valves. The air, to which the sampler adds 1 0/00 by volume of tritium-free hydrogen, passes first through a molecular sieve trap where H₂O and HTO are adsorbed, and then through a trap of palladium-coated molecular sieve, where the carrier H₂, plus ambient H₂, HT, and T₂ are oxidized and the resulting water adsorbed.

The mass of air sampled, the location of sampling, and the ambient pressure altitude and temperature are noted by the equipment operator. The sample traps are returned to Miami, where the sample water is extracted by techniques described by Östlund and Mason (1974), and the tritium determined by low-level proportional counting (Östlund and Dorsey, 1977).

The data take the form of mixing ratios of HTO and HT, i.e., tritium atoms per mg of air. These units may be converted to picocuries per standard cubic meter of air (pCi/SCM) by multiplication by 0.0625 for SCM defined at 1013 mb and 0°C. Specific activities of atmospheric water vapor and hydrogen cannot be determined accurately due to the very small samples obtained, and the consequent use of tritium-free water to flush the samples from the traps.

3. TRITIUM DATA AND OPERATIONAL COMMENTS

Tables 1 through 14 present the tritium data obtained between April 1975 and July 1979. The Airstream mission numbers begin with A-4, which was the first on which tritium sampling was conducted.

No fall deployment was made in 1975 due to the need for extensive aircraft maintenance. A special mission was flown on November 24, 1976 to assess the environmental impact of the high yield Chinese atmospheric nuclear test of November 17, 1976. Tritium sampling on mission A-9 (March-April 1977) was halted by a malfunction of an aircraft temperature controller, which damaged the sampler.

The vertical profiles commenced with mission A-10 (July 1977). Tritium sampling on the Panama vertical profile was discontinued after A-13 (July 1978) due to the low HTO concentrations encountered.

Tritium sampling on mission A-14 (October-November 1978) was halted by a mechanical failure of the sampler canister, brought on by an excessive pressurization air supply.

Figures 2 through 13 show the HTO data obtained from each Airstream deployment, except A-9, which were too few for detailed analysis. No HT figures are presented as an essentially well-mixed distribution has been found on all deployments.

Frequent observations of high HTO mixing ratios during the tropospheric portions of vertical profiles at 29° and 61°N are attributed to re-evaporation of precipitation as described by Ehhalt (1971), and also seen during previous flights near 40°N (Mason and Östlund, 1976). This effect is also seen at

Fairbanks, Alaska, in the form of a mid-summer peak of HTO mixing ratio (Mason and Östlund, 1977).

4. THE STRATOSPHERIC TRITIUM AND ZIRCONIUM-95 INVENTORY

The data presented in Tables 1 through 14 provide information about the tritium burden of the lower stratosphere (up to about 70,000 ft) in the Northern Hemisphere.

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

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

Two possible problems exist with using these tritium data for a direct comparison with fission product data: 1) the calculated stratospheric tritium burdens may contain a background from past high yield tests (attempts will be made to resolve this problem); and 2) although the production of fission products from nuclear tests is fairly well known (Harley, 1965), the tritium

production from thermonuclear tests has a much larger uncertainty. It has been reported to range from 7 to 50 megacuries per megaton fusion, (Mci/MT (fusion)), with a suggested average value of 20 Mci/MT (fusion) (NCRP, 1979). It is therefore difficult to know with certainty the amount of tritium injected into the atmosphere even if the total yield and fission yield of an event are known.

There were no simultaneous measurements of Zr-95 and HTO following the June 27, 1973 high yield test, whereas there were for three sampling series for the November 17, 1976 test. This test will therefore be discussed first followed by the analysis of the June 27, 1973 test.

The latitudinal distribution of the observed HTO concentrations between July 1977 to July 1979 is shown in Figures 7-13. The observed average tropopause height along the sampling corridor during these sampling missions is also shown. For Figures 10-13 meteorological data are not available, at this time, to calculate an average tropopause, therefore an assumed tropopause was used, based on earlier sampling periods.

The Northern Hemisphere stratospheric HTO burdens for the seven sampling periods between July 1977 and July 1979 are given in Table 15 together with the Zr-95 burdens calculated by Telegadas (1979). The first two columns show the Zr-95 burden to about 20 km (based on aircraft sampling) and to about 30 km (based on additional balloon sampling from 20 to 30 km). The first line under these two columns shows the Zr-95 burden prior to the November 17, 1976 test. The last significant test prior to this event occurred on June 17, 1974 (reported total yield of between 0.2-1 MT) and estimated by Leifer (1976) to have a fission yield of 0.4 MT. By the time of the November 17, 1976 test, due to stratospheric depletion and radioactive decay, the Zr-95 created by this test decayed below detection limits. The tritium col-

lected between October 24 and November 17, 1976 (Figure 6) indicated a background of 3100 kCi of HTO residing in the stratosphere prior to the November 17, 1976 test. This is shown in column A, line 1. Column B shows the observed burden (column A) decay corrected to the November 17, 1976 test. The burden for the October 13 - November 6, 1978 sampling period listed in column A is questionable due to limited data. To illustrate this, Figure 14 shows a reanalysis of the July 1978 distribution (Figure 10) with only those data points corresponding to the data points in Figure 11 (Oct.-Nov. 1978 sampling period). The stratospheric burden calculated from these limited data is 5,200 kCi of HTO, an increase of about 30% from the 4,000 kCi derived for Figure 10 which was based on much more data. The Oct. 13-Nov. 6, 1978 calculated burden should therefore be used with caution in any stratospheric burden interpretation.

One could also question the HTO burden calculated for the July 3-26, 1979 period (Figure 13) since no samples were collected in the lower equatorial stratosphere. A $\pm 50\%$ difference in the average concentration calculated from the subjective analysis in this region would only produce about a $\pm 10\%$ difference in the total N.H. burden (2500 kCi) reported in Table 15.

A line of regression through the decay corrected HTO burdens given in column B (from July 1977 to July 1979) would indicate a residence half-time of about 10 months. It was therefore assumed that the background HTO burden prior to the November 17, 1976 test would be depleted with this same residence time. It should be noted that the presumably constant natural background due to cosmic radiation is estimated to be at most 1,000 kCi, using the production rate, stratosphere-troposphere production distribution, and stratospheric residence time of Craig and Lal (1961). The natural

background has been neglected in the calculation of column C due to the uncertainties of the estimate and the absence of asymptotic behavior in the data acquired to date. The background burdens at later times are listed in column C. Column D shows the residual burden attributed to the November 17, 1976 test, that is, column B minus column C. The residual burdens are shown in Figure 15 together with the assumed depletion of the background listed in column C.

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

The line of regression through the tritium burden attributed to the November 17, 1976 test (Figure 15) indicates a residence half time of about 10 months with an input into the stratosphere of 16,500 kCi of HT0 to about 20 km. Increasing this amount by 15% (assuming the same percentage of tritium above the aircraft altitudes as was determined for the Zr-95 burden) would indicate an input of 19,000 kCi of HT0. Since the fusion yield was estimated to be 1.3 MT, the assumed production of HT0 from the November 17, 1976 event would be about 15 MCi/MT (fusion). This seems

reasonable, since the range has been reported as 7 to 50 MCi/MT with a suggested average value of 20 MCi/MT for thermonuclear devices (NCRP, 1979).

The observed latitudinal stratospheric distributions of HT0 following the June 27, 1973 high yield Chinese test are shown in Figures 2 through 6. The average observed tropopause along the sampling corridor during the collection period is also given. The first stratospheric sampling where a reliable stratospheric inventory could be performed was not until April 18-May 6, 1975 (Figure 2), nearly two years after the June 27, 1973 test.

Shown in Table 16 are the computed Northern Hemispheric stratospheric HT0 burdens to approximately 20 km based on the analysis shown in Figures 2-6. Column 1 gives the observed inventory. Decay correcting these inventories to June 27, 1973, the Chinese high yield test, is shown in column 2. The burdens listed in column 2 are shown in Figure 16 together with the Zr-95 inventories to 30 km attributed to the June 27, 1973 test, reported by Telegadas (1976). A line of regression through the decay corrected HT0 burdens indicate a residence half time of about 13 months. Extrapolating this regression line back to December 15, 1973, when it was estimated that significant fallout of Zr-95 began (Telegadas, 1976), indicates a production of 18,400 kCi of HT0. The stratospheric inventory of Zr-95 showed about 5% above the sampling altitude of the aircraft. The HT0 burden (to 20 km) at time of significant fallout was therefore increased by 5% for a total input of 19,300 kCi of HT0 into the stratosphere.

The Zr-95 burden to about 30 km indicated a 34,000 kCi input which is equivalent to a fission yield of 1.4 MT. The reported total yield for the June 27, 1973 event was 2 to 3 MT. Assuming a total yield of 2.5 MT

would mean that this event had a fusion yield of about 1.1 MT. Dividing the HTO stratospheric input of 19.3 MCi by 1.1 MT indicates a production of 18 MCi/MT (fusion), not too different from that calculated for the November 17, 1976 test.

The stratospheric residence half time of the Zr-95 burdens, Figure 16, based on only 2 sampling periods, was 5 months. This residence time is shorter than that for the November 17, 1976 test but about the same as Telegadas (1974) found for the June 17, 1967 high yield Chinese test between the sampling periods of February 1968 and April 1968 (6 months). The Zr-95 burden attributed to the June 27, 1973 test could not be determined beyond April 1974 due to the fact that the next Chinese test of June 17, 1974 (total yield 0.2-1 MT) dominated the lower stratosphere of the Northern Hemisphere during the next sampling period in October 1974.

As mentioned previously, Leifer (1976) estimated the fission yield of the June 17, 1974 test to be about 0.4 MT. When this event occurred, no mention was made whether it was an all fission or thermonuclear test. We will assume that this test was thermonuclear and it had a fission-fusion ratio of 1; that is, 0.4 MT (fusion). Assuming a HTO production of 15 MCi/MT (fusion), that was calculated for the November 17, 1976 test, this test would have injected 6,000 kCi of HTO into the lower stratosphere of the Northern Hemisphere. It is further assumed that this HTO input had a stratospheric residence half time of 10 months. This input was then decay corrected to the 1975 and 1976 sampling periods listed in Table 16 and subtracted from column 1. Column 3, Table 16, is therefore the residual

burden attributed to the June 27, 1973 high yield test if the June 17, 1974 test was thermonuclear and had a fission-fusion ratio of 1.

As can be seen from Figure 16 the residual burden attributed to the June 27, 1973 test had a stratospheric residence half time of 16 months. Extrapolating back to the start of significant fallout and assuming 5% of the HTO burden resided above the aircraft sampling altitude yields a HTO production of 9,600 kCi or an HTO production of 9 MCi/MT (fusion), half that estimated if the June 27, 1974 test were all fission.

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

There is more confidence in the estimated HTO burdens attributed to the November 17, 1976 test (Figure 15) than the estimated HTO burdens from the June 27, 1973 test (Figure 16). This is in part due to the fact that there was a partial overlap of the Zr-95 and HTO burdens starting 8 months after input. Further the background HTO inventory based on measurements taken shortly before the November 17, 1976 event could be accounted for and subtracted from the observed inventories at later times with some confidence.

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

5. CONCLUSIONS

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

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

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TABLE 1 STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM
Mission A-4 1975 Apr-May

The first element of the sample number is the flight number from the Project Support Plan (NASA, 1974), and the second element is the sequential number of the sample on that flight. Samples 39-1 through -4 were taken on a CIAP* flight, Prime 39, during the same deployment. Height is computed from the NACA standard atmosphere. Mid-latitude is calculated from the flight log. θ (potential temperature) is calculated from recorded altitude and temperature.

No.	Temp. (°C)	Pres. (mb)	θ (°K)	HTO Conc. (T-atoms/mg air)	HT Conc. (T-atoms/mg air)	Height (km)	Mid-latitude
1-1	-68	94	404	32 ± 2	48 ± 1	16.6	50N
1-2	-49	64	492	1471 ± 48	62 ± 2	19.1	50N
2-1	-48	173	372	694 ± 23	47 ± 2	13.7	50N
2-2	-44	175	377	1081 ± 32	46 ± 2	13.8	60N
4-1	-42	98	449	996 ± 39	59 ± 2	16.3	73N
4-2	-44	70	490	1713 ± 64	73 ± 3	18.5	73N
5-1	-48	69	484	1431 ± 57	66 ± 2	18.6	60N
9-1	-77	91	389	30 ± 3	58 ± 3	16.8	28N
10-1	-74	116	369	12 ± 1	37 ± 1	15.3	28N
10-2	-75	73	418	393 ± 16	41 ± 1	18.2	11N
11-1	-91	91	362	86 ± 4	54 ± 3	16.9	11N
11-2	-70	66	442	421 ± 15	39 ± 3	18.8	11N
12-1	-82	114	356	56 ± 3	40 ± 4	15.3	9S
12-2	-75	115	368	37 ± 2	61 ± 3	15.3	4N
13-1	-87	98	362	18 ± 1	49 ± 1	16.3	9S
13-2	-66	66	451	162 ± 6	44 ± 2	18.9	9S
14-1	-75	75	416	510 ± 14	73 ± 3	18.0	11N
14-2	-64	80	431	618 ± 17	50 ± 2	17.6	28N
39-1	-38	289	335	357 ± 14	46 ± 2	9.2	60N
39-2	-38	263	345	330 ± 13	43 ± 1	10.0	60N
39-3	-37	245	353	372 ± 14	45 ± 2	10.5	60N
39-4	-41	153	397	641 ± 27	53 ± 2	13.5	60N

*CIAP: Climatic Impact of Air Pollution, sponsored by the U.S. Department of Transportation.

TABLE 2. STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM.
Mission A-5 1975 Jul-Aug

These data are obtained from the same sources as that of Table 1, except for temperature, pressure, and potential temperature. Since no data recorder was carried on this deployment, the flight log was used for the air data.

No.	Temp. (°C)	Pres. (mb)	θ (°K)	HTO Conc. (T-atoms/mg air)	HT Conc. (T-atoms/mg air)	Height (km)	Mid-latitude
1-1	-65	92	418	381 ± 13	52 ± 2	16.8	41N
2-1	-46	148	391	451 ± 16	48 ± 2	13.7	56N
3-2	-47	116	423	460 ± 18	53 ± 2	15.2	74N
4-1	-46	92	455	456 ± 17	56 ± 2	16.8	74N
4-2	-44	69	490	732 ± 28	60 ± 2	18.6	74N
5-1	-49	92	452	213 ± 9	50 ± 2	16.8	56N
5-2	-46	63	503	343 ± 13	60 ± 2	19.1	56N
6-1	-48	118	423	699 ± 20	49 ± 2	15.1	56N
7-1	-67	148	356	123 ± 6	47 ± 2	13.7	41N
8-1	-66	116	389	154 ± 6	48 ± 2	15.2	41N
8-2	-54	62	490	524 ± 20	50 ± 2	19.2	41N
9-1	-62	91	418	163 ± 7	48 ± 4	16.8	25N
9-2	-59	63	474	177 ± 9	34 ± 3	19.1	25N
10-1	-72	116	373	100 ± 4	56 ± 2	15.2	25N
13-1	-73	92	396	14 ± 1	63 ± 4	16.8	7.5N
13-2	-58	63	474	431 ± 17	59 ± 2	19.2	6.5N
14-1	-76	116	367	9 ± 1	53 ± 3	15.2	10N
14-2	-62	72	444	532 ± 4	49 ± 2	18.3	25N

TABLE 3

STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM
Mission A-6, 1976 May-June

The first element of the sample number is the flight number from the applicable Project Support Plan, and the second element is the sequential number of the sample on that flight. Temperature, pressure, θ (potential temperature), height, and mid-latitude are calculated from the flight log.

NO.	TEMP. (°C)	PRES. (MB)	θ (°K)	T-ATOMS/MG		AIR		HEIGHT (KM)	MID- LAT.
				AS	HTO	AS	HT		
1-1	-60	92	422	327	+ 9	46	+ 1	16.7	42 N
2-1	-50	148	385	480	+ 17	43	+ 1	13.7	59 N
3-1	-49	179	367	455	+ 17	73	+ 2	12.5	74 N
3-2	-47	92	448	392	+ 16	49	+ 2	16.7	74 N
4-1	-48	116	417	144	+ 6	43	+ 1	15.3	74 N
4-2	-44	70	490	570	+ 22	47	+ 2	18.5	74 N
5-1	-54	92	434	526	+ 21	40	+ 1	16.7	59 N
5-2	-50	64	490	697	+ 28	50	+ 2	19.1	59 N
6-1	-52	116	410	144	+ 5	45	+ 1	15.3	59 N
7-1	-59	148	370	39	+ 2	44	+ 1	13.7	42 N
8-1	-62	116	391	179	+ 8	50	+ 2	15.3	42 N
8-2	-59	63	472	817	+ 28	50	+ 2	19.2	42 N
9-1	-75	92	392	25	+ 1	50	+ 2	16.7	25 N
9-2	-68	68	443	412	+ 16	49	+ 1	18.7	25 N
10-1	-72	116	373	17	+ 1	44	+ 1	15.3	22 N
10-2	-73	72	425	183	+ 6	45	+ 2	18.3	13 N
11-1	-78	92	386	18	+ 2	45	+ 2	16.7	8 N
11-2	-65	63	459	430	+ 14	40	+ 1	19.2	8 N
12-1	-78	116	361	5	+ 1	45	+ 2	15.3	9 S
12-2	-72	72	427	33	+ 2	42	+ 2	18.3	9 S
13-2	-67	63	455	107	+ 4	40	+ 1	19.2	9 S
14-1	-79	116	360	8	+ 1	49	+ 2	15.3	8 N
14-2	-65	72	442	287	+ 9			18.3	25 N

TABLE 4
 STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM
 Mission A-7, 1976 August

See Table 3 for explanation of data.

NO.	TEMP. (°C)	PRES. (MB)	θ (°K)	T-ATOMS/MG AIR			HEIGHT (KM)	MID- LAT.
				AS	HTO	AS HT		
1-1	-67	92	408	112	± 5	45 ± 2	16.7	32 N
1-2	-65	92	412	58	± 2	39 ± 1	16.7	42 N
2-1	-57	148	373	53	± 2	45 ± 1	13.7	53 N
3-1	-49	179	367	118	± 5		12.5	64 N
3-2	-54	179	359	70	± 3	42 ± 2	12.5	74 N
3-3	-51	92	440	119	± 5	42 ± 2	16.7	74 N
4-1	-49	116	415	63	± 3	42 ± 2	15.3	64 N
4-2	-51	116	411	127	± 5	51 ± 2	15.3	74 N
4-3	-48	71	480	213	± 9	45 ± 2	18.4	74 N
4-4	-48	63	497	330	± 14	51 ± 2	19.2	64 N
5-1	-48	92	446	121	± 5	46 ± 2	16.7	53 N
5-2	-47	68	488	326	± 13	46 ± 2	18.7	53 N
6-1	-48	116	417	184	± 8	45 ± 1	15.3	53 N
7-1	-59	148	370	36	± 2	42 ± 1	13.7	42 N
7-2	-65	148	360	25	± 1	40 ± 1	13.7	32 N
8-1	-65	116	386	49	± 2	45 ± 2	15.3	32 N
8-2	-66	116	384	62	± 3	39 ± 1	15.3	42 N
8-3	-58	63	474	359	± 14	44 ± 2	19.2	42 N
8-4	-61	63	468	343	± 13	42 ± 2	19.2	32 N
9-1	-70	92	402	221	± 9	40 ± 1	16.7	21 N
9-2	-66	64	455	98	± 4	40 ± 2	19.1	21 N
10-1	-75	117	366	13	± 1	39 ± 1	15.2	21 N
10-2	-66	73	438	180	± 7	40 ± 1	18.2	11 N
11-1	-71	92	400	26	± 1	43 ± 1	16.7	11 N
11-2	-59	63	472	176	± 7	41 ± 2	19.2	11 N
12-1	-74	116	369	10	± 1	45 ± 2	15.3	1 N
12-2	-75	116	367	51	± 2	43 ± 2	15.3	9 S
12-3	-68	72	435	90	± 4	45 ± 2	18.3	9 S
12-4	-65	72	442	164	± 7	48 ± 2	18.3	1 N
13-1	-72	92	398	12	± 1	43 ± 2	16.7	1 N
13-2	-70	92	402	75	± 3	46 ± 2	16.7	9 S
13-3	-68	64	450	134	± 6	47 ± 2	19.1	9 S
13-4	-67	63	455	71	± 3	46 ± 2	19.2	1 N
14-1	-74	116	369	15	± 1	47 ± 2	15.3	11 N
14-2	-68	72	435	90	± 4	42 ± 2	18.3	21 N

TABLE 5
 STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM
 Mission A-8, 1976 Oct-Nov

See Table 3 for explanation of data.

NO.	TEMP. (°C)	PRES. (MB)	e (°K)	T-ATOMS/MG		AIR		HEIGHT (KM)	MID- LAT.
				AS	HTO	AS	HT		
1-1	-69	92	404	97	± 4	42	± 1	16.7	32 N
1-2	-64	92	414	217	± 9	47	± 1	16.7	42 N
2-1	-51	150	382	62	± 3	48	± 1	13.6	53 N
3-1	-46	188	366	74	± 3	41	± 1	12.2	64 N
3-2	-50	188	360	68	± 3	41	± 1	12.2	74 N
3-3	-50	92	442	228	± 9	44	± 1	16.7	74 N
3-4	-49	92	444	265	± 11	46	± 2	16.7	64 N
4-1	-49	116	415	63	± 3	45	± 2	15.3	64 N
4-2	-52	116	410	133	± 6	47	± 2	15.3	74 N
4-3	-54	69	471	275	± 12	45	± 2	18.6	74 N
4-4	-50	63	492	449	± 18	46	± 2	19.2	64 N
5-1	-49	92	444	555	± 19	45	± 2	16.7	53 N
5-2	-46	65	494	890	± 36	47	± 2	18.9	53 N
6-1	-54	116	406	231	± 10	46	± 1	15.3	53 N
7-1	-65	148	360	129	± 3	43	± 1	13.7	42 N
7-2	-64	148	361	81	± 3	40	± 1	13.7	32 N
8-1	-68	116	380	60	± 3	47	± 2	15.3	32 N
8-2	-66	117	383	122	± 5	45	± 2	15.2	42 N
8-3	-53	63	486	1185	± 43	50	± 2	19.2	42 N
8-4	-61	63	468	391	± 16	46	± 2	19.2	32 N
9-2	-69	64	448	243	± 9	53	± 2	19.1	21 N
10-1	-77	116	363	25	± 1	42	± 1	15.3	21 N
10-2	-76	72	419	97	± 4	45	± 2	18.3	11 N
11-1	-73	62	443	68	± 3	42	± 2	19.3	11 N
11-2	-85	91	374	25	± 1	35	± 1	16.8	11 N
12-1	-80	116	358	11	± 1	41	± 1	15.3	1 N
12-2	-78	116	361	10	± 1	36	± 1	15.3	9 S
12-3	-72	72	427	39	± 2	40	± 1	18.3	9 S
12-4	-77	72	416	82	± 4	41	± 1	18.3	1 N
13-1	-82	90	381	24	± 1	41	± 1	16.9	1 N
13-2	-80	93	381	100	± 4	49	± 2	16.7	9 S
14-1	-78	116	361	76	± 3	43	± 1	15.3	11 N

TABLE 6

CLOUD SAMPLE HTO AND HT
1976 Nov. 24

GMT Start Finish	Location Start Finish	Temp. °C	Pres. mb	θ °K	HTO Conc. T-atoms/mg air (pCi/SCM)	HT Conc. T-atoms/mg air (pCi/SCM)
1548	34°55'N 85°45'W*	-62	72	448	693 ± 15	46 ± 2
1609	36°19'N 83°29'W	-61	72	450	(40.5 ± 0.9)	(2.7 ± 0.1)
1614	36°35'N 83°36'W	-61	72	450	745 ± 16	45 ± 2
1647	36°35'N 87°08'W	-62	71	450	(43.6 ± 0.9)	(2.6 ± 0.1)
1646	36°27'N 87°32'W	-61	71	452	780 ± 16	42 ± 2
1717	34°32'N 83°37'W	-63	70	449	(45.6 ± 0.9)	(2.4 ± 0.1)
1725	34°14'N 82°40'W	-61	69	455	780 ± 16	46 ± 2
1759	37°58'N 82°28'W	-59	66	466	(45.6 ± 0.9)	(2.7 ± 0.1)
1808	38°36'N 82°04'W	-61	66	461	973 ± 22	46 ± 2
1825	37°11'N 80°10'W	-59	66	466	(56.9 ± 1.3)	(2.7 ± 0.1)
1828	37°05'N 80°01'W	-59	63	472	1194 ± 29	51 ± 2
1855	40°01'N 79°52'W	-58	63	474	(69.8 ± 1.7)	(3.0 ± 0.1)

*Position from scientific equipment operator's log. Recorded inertial navigation system data not received for this time.

TABLE 7
 STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM
 Mission A-9, 1977 Mar-Apr

See Table 3 for explanation of data.

NO.	TEMP. (°C)	PRES. (MB)	e (°K)	T-ATOMS/MG		AIR		HEIGHT (KM)	MID- LAT.
				AS	HTO	AS	HT		
1-1	-65	91	413	328	+ 11	42	+ 1	16.8	32 N
1-2	-63	92	416	1064	+ 45	41	+ 2	16.7	42 N
2-1	-59	148	370	579	+ 17	37	+ 1	13.7	53 N
3-1	-52	188	357	137	+ 6	54	+ 2	12.2	64 N
3-2	-50	188	360	1200	+ 51	62	+ 3	12.2	74 N
3-3	-39	94	461	10475	+425	65	+ 3	16.6	74 N
9-2	-75	63	437	172	+ 7	33	+ 1	19.1	21 N
10-1	-76	116	365	14	+ 1	53	+ 2	15.3	21 N
10-2	-75	72	421	86	+ 4	40	+ 1	18.3	11 N
11-1	-81	92	380	9	+ 1	42	+ 1	16.7	11 N
11-2	-68	64	450	2383?	+ 85	38	+ 1	19.1	11 N
12-1	-77	116	363	10	+ 1	43	+ 1	15.3	1 N
12-2	-79	116	360	11	+ 1	40	+ 1	15.3	9 S
12-3	-71	65	442	25	+ 1	53	+ 2	19.0	9 S
12-4	-70	63	448	51	+ 2	43	+ 2	19.1	1 N
13-1	-88	92	366	13	+ 1	42	+ 2	16.7	1 N
13-2	-83	92	376	8	+ 1	41	+ 1	16.7	9 S
13-3	-75	74	417	27	+ 1	44	+ 2	18.1	9 S
13-4	-77	72	416	82	+ 4	43	+ 2	18.3	1 N
14-1	-77	116	363	24	+ 1	45	+ 2	15.3	11 N
14-2	-74	72	423	242	+ 10	38	+ 2	18.3	21 N

TABLE 8
STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM
Mission A-10, 1977 July

See Table 3 for explanation of data

NO.	TEMP. (°C)	PRES. (MB)	θ (°K)	T-ATOMS/MG		AIR AS HT	HEIGHT (KM)	MID- LAT.
				AS	HTO			
1-1	-59	92	424	1455	± 45		16.7	41
2-1	-51	148	384	189	± 6		13.7	52
3-1	44	188	370	858	± 30	38 + 1	12.2	68
3-2	-46	188	366	1322	± 47	51 ± 1	12.2	74
3-3	-44	92	453	1821	± 58	44 ± 2	16.7	74
3-4	-42	92	457	1761	± 67	48 ± 1	16.7	68
4-1	-45	116	423	493	± 15		15.3	68
4-2	-44	116	424	614	± 25	43 ± 2	15.3	74
4-3	-38	75	493	1580	± 62	52 ± 2	18.0	74
4-4	-38	68	507	1641	± 65	51 ± 2	18.7	68
5-1	-45	92	452	1957	± 47	45 ± 2	16.7	60
5-2	-48	92	446	3040	± 106	45 ± 2	16.7	52
5-3	-45	69	490	4386	± 150	48 ± 2	18.6	52
5-4	-41	67	503	6269	± 217	49 ± 2	18.8	60
6-1	-47	119	419	1042	± 40	41 ± 1	15.1	58
6-2	-49	119	415	1714	± 47	40 ± 1	15.1	52
7-1	-69	148	353	82	± 3	36 ± 1	13.7	41
8-1	-67	116	382	276	± 10	41 ± 2	15.3	41
8-2	-51	64	488	4277	± 119	45 ± 2	19.1	41
9-1	-65	92	412	275	± 7	38 ± 2	16.7	29
9-2	-70	92	402	461	± 18	41 ± 2	16.7	21
9-3	-59	64	470	1871	± 67	46 ± 2	19.1	21
9-4	-58	64	472	1137	± 46	36 ± 2	19.1	29
10-1	-68	114	382	135	± 5	49 ± 2	15.4	21
10-2	-67	73	436	622	± 19	40 ± 1	18.2	14
11-1	-74	92	394	140	± 5	43 ± 2	16.7	14
11-2	-62	63	466	1273	± 47	59 ± 2	19.2	14
12-1	-74	115	370	96	± 4		15.3	5
12-2	-76	116	365	51	± 2		15.3	1
12-3	-78	92	386	43	± 2	43 ± 2	16.7	1
12-4	-72	73	425	203	± 8	47 ± 2	18.2	5
12-5	-70	73	430	617	± 17	45 ± 2	18.2	1
12-6	-67	63	455	580	± 22	48 ± 2	19.2	1
14-1	-71	117	373	113	± 4	39 ± 1	15.2	14
14-2	-63	74	443	1323	± 33	51 ± 2	18.1	21
15-1	- 2	697	301	487	± 9		3.8	61
15-2	-24	466	310	842	± 22		6.4	61
15-3	-49	301	316	66	± 3		9.2	61
15-4	-48	238	340	465	± 17		10.7	61
15-5	-44	188	370	680	± 21		12.2	61

TABLE 8 STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM (Cont.)

NO.	TEMP. (°C)	PRES. (MB)	ρ (°K)	T-ATOMS/MG AIR		HEIGHT (KM)	MID- LAT.
				AS HTO	AS HT		
15-6	-45	148	394	742	± 22	13.7	61
16-1	- 9	466	329	30	± 1	6.4	27
16-2	-33	301	339	17	± 1	9.2	27
16-3	-57	188	349	17	± 1	12.2	27
16-4	-67	148	356	18	± 1	13.7	27
16-5	-68	116	380	92	± 4	15.3	27
16-6	-66	92	410	273	± 10	16.7	27
17-1	- 8	466	330	36	± 1	6.4	6
17-2	-31	301	341	13	± 1	9.2	6
17-3	-57	188	349	12	± 1	12.2	6
17-4	-68	148	354	6	± 1	13.7	6
17-5	-75	92	392	11	± 1	16.7	6
17-6	-63	65	459	266	± 9	19.0	6

TABLE 9

 STRATOSPHERIC HT AND HTO FROM PROJECT AIRSTREAM
 Mission A-11, 1977 October

See TABLE 3 for explanation of data.

NO.	TEMP. (°C)	PRES. (MB)	e (°K)	T-ATOMS/MG		AIR		HEIGHT (KM)	MID- LAT.
				AS	HTO	AS	HT		
3-1	-79	116	360	25	± 1	31	± 1	15.3	18
4-1	-75	93	391	62	± 3	34	± 1	16.7	18
4-2	-62	65	462	588	± 23	35	± 1	19.0	18
5-1	-70	116	365	53	± 2	35	± 1	15.3	1
5-2	-78	92	386	24	± 1	34	± 1	16.7	1
5-3	-70	72	431	131	± 6	38	± 1	18.3	1
5-4	-67	63	455	155	± 6	36	± 1	19.2	1
7-1	-69	148	353	11	± 1	38	± 1	13.7	18
8-1	-65	92	412	621	± 24	40	± 1	16.7	39
8-2	-60	63	470	967	± 41	46	± 2	19.2	39
9-1	-63	116	389	90	± 4	38	± 1	15.3	39
9-2	-59	116	397	592	± 23	37	± 1	15.3	49
10-1	-62	148	365	143	± 6	40	± 1	13.7	49
11-1	-50	188	360	229	± 9	41	± 1	12.2	66
11-2	-51	188	358	435	± 18	42	± 2	12.2	74
11-3	-50	92	442	1368	± 55	37	± 1	16.7	74
11-4	-47	92	448	2072	± 82	41	± 2	16.7	66
12-1	-47	116	419	704	± 29	35	± 1	15.3	66
12-2	-49	116	415	717	± 31			15.3	74
12-3	-48	66	490	1993	± 80	45	± 2	18.9	74
14-1	-53	92	436	992	± 42	28	± 1	16.7	49
14-2	-49	63	494	1478	± 61	44	± 2	19.2	49
15-1	-64	148	361	359	± 15	35	± 1	13.7	39
1-1	4	697	307	133	± 3			3.8	29
1-2	-14	466	322	25	± 1			6.4	29
1-3	-33	301	339	26	± 1			9.2	29
1-4	-57	188	349	6	± 1			12.2	29
1-5	-70	148	351	5	± 1			13.7	29
6-1	-31	294	344	22	± 1			9.3	7
6-2	-57	178	354	53	± 2			12.5	7
6-3	-70	140	357	18	± 1			14.1	7
6-4	-77	111	368	62	± 2			15.5	7
6-5	-73	88	401	51	± 2			17.0	7
6-6	-66	62	459	116	± 5			19.3	7
13-1	-14	697	287	346	± 10			3.8	62
13-2	-35	466	296	106	± 4			6.4	62
13-3	-53	301	310	35	± 2			9.2	62
13-4	-52	238	333	104	± 5			10.7	62
13-5	-51	188	358	380	± 13			12.2	62
13-6	-47	148	391	971	± 34			13.7	62

TABLE 10

Stratospheric HT and HTO from Project Airstream
Mission A-12, 1978 April

The first element of the sample number is the flight number from the applicable Project Support Plan, and the second element is the sequential number of the sample on that flight. Temperature, pressure, θ (potential temperature), height, mid-latitude and mid-longitude are calculated from the flight log.

NO.	TEMP. (°C)	PRES. (MB)	θ (°K)	T-ATOMS/MG AIR		HEIGHT (KM)	MID- LAT.	MID- LONG.		
				AS	HTO					
1-1	7	697	310	31.1	± 1.7	3.0	29.0	94.5		
1-2	-14	466	322	20.4	± 0.7	6.1	29.0	94.5		
1-3	-39	301	330	13.4	± 1.0	9.1	29.0	94.5		
1-4	-57	188	348	19.5	± 1.0	12.2	29.0	94.5		
1-5	-59	148	370	34.0	± 1.5	13.7	29.0	94.5		
1-6	-63	116	389	35.5	± 1.7	15.3	29.0	94.5		
3-1	-73	116	370	12.9	± 0.7	34.8	± 1.3	15.3	18.0	84.1
4-1	-77	93	387	16.9	± 1.1	36.0	± 1.5	16.7	18.0	79.1
4-2	-67	63	454	251.8	± 9.6	39.7	± 1.5	19.2	18.0	79.4
5-1	-78	116	361	23.1	± 1.0	37.4	± 1.5	15.3	1.0	79.6
5-2	-83	92	376	12.9	± 0.8	37.5	± 1.4	16.7	1.0	79.9
5-3	-72	72	427	128.7	± 5.2	37.6	± 1.4	18.3	1.0	79.7
5-4	-65	63	459	221.4	± 8.4	36.6	± 1.4	19.2	1.0	79.7
6-1	-31	301	341	15.8	± 0.7	9.1		6.9		80.2
6-2	-7	466	331	26.5	± 1.6	6.1		6.8		80.3
6-3	-56	188	350	8.0	± 0.7	12.2		6.9		80.2
6-4	-76	116	365	4.1	± 0.3	15.3		6.6		80.6
6-5	-81	92	380	12.1	± 0.6	16.7		7.0		81.1
6-6	-60	65	465	118.6	± 4.2	19.0		7.5		80.6
7-1	-68	148	354	28.6	± 1.2	38.4	± 1.3	13.7	18.0	84.1
8-1	-60	92	421	247.0	± 9.6	36.2	± 1.5	16.7	39.0	93.0
8-2	-57	63	476	737.0	± 27.0	54.2	± 2.0	19.2	39.0	93.4
9-1	-56	116	402	103.8	± 4.3	33.9	± 1.0	15.3	39.0	108.8
9-2	-48	116	417	290.0	± 11.0	35.2	± 1.1	15.3	49.0	125.2
9-3	-49	92	443	458.0	± 19.0	40.1	± 1.3	16.7	49.0	125.8
10-1	-46	148	392	750.0	± 28.5	38.4	± 1.3	13.7	49.0	128.1
11-1	-46	188	366	189.0	± 8.0	35.3	± 1.2	12.2	66.0	150.0
11-2	-48	188	363	251.0	± 11.0	34.7	± 1.2	12.2	74.0	149.9
11-3	-45	92	451	1096.0	± 44.0	33.6	± 1.4	16.7	74.0	150.5
11-4	-45	92	451	551.0	± 22.0	30.8	± 1.2	16.7	66.0	150.0
12-1	-51	116	411	491.0	± 21.0	38.4	± 1.5	15.3	66.0	152.1
12-2	-46	91	451	1275.0	± 52.0	39.0	± 1.2	16.8	74.0	151.0
12-4	-44	63	505	1118.0	± 44.0	40.8	± 1.5	19.2	66.0	151.6
13-1	-11	697	290	366.0	± 9.0	3.0		61.8		150.6
13-2	-28	466	305	103.9	± 4.0	6.1		61.7		151.0
13-3	-51	301	313	24.3	± 1.1	9.1		62.0		151.0
13-4	-54	238	330	32.8	± 1.4	10.7		62.1		150.3
13-5	-52	188	356	129.2	± 5.6	12.2		62.4		151.5
13-6	-51	148	383	304.0	± 13.0	13.7		62.0		150.9
14-1	-50	92	441	327.0	± 13.0	43.6	± 1.5	16.7	49.0	128.1
15-1	-55	148	376	570.0	± 22.0	39.2	± 1.4	13.7	39.0	114.1

TABLE 11

Stratospheric HT and HTO from Project Airstream
Mission A-13, 1978 July

See TABLE 10 for explanation of data.

NO.	TEMP. (°C)	PRES. (MB)	Θ (°K)	T-ATOMS/MG AIR		HEIGHT (KM)	MID- LAT.	MID- LONG.
				AS	HTO			
1-1	10	697	314	198.0	± 4.0	3.0	29.1	94.8
1-2	-8	466	329	51.0	± 2.0	6.1	29.1	94.6
1-4	-62	167	351	12.6	± 0.6	12.9	29.1	94.9
1-5	-75	116	366	6.4	± 0.4	15.3	29.2	94.6
1-6	-68	92	406	30.0	± 1.2	16.7	28.8	94.6
3-1	-70	116	377	299.0	± 10.0	30.5 ± 1.3	15.3	18.0 87.8
4-1	-71	92	400	181.0	± 7.0	39.2 ± 1.2	16.7	18.0 79.1
4-2	-61	63	467	585.0	± 21.0		19.2	18.0 78.8
5-1	-73	116	371	144.0	± 6.0	41.1 ± 1.6	15.3	1.0 79.6
5-2	-68	92	405	37.9	± 1.7		16.7	1.0 79.6
5-3	-63	72	447	273.0	± 12.0	34.4 ± 1.5	18.3	1.0 79.6
5-4	-63	63	464	395.0	± 16.0	37.2 ± 1.2	19.2	1.0 79.6
6-3	-57	188	349	17.1	± 1.0		12.2	8.0 79.1
6-4	-68	148	354	4.4	± 0.3		13.7	8.0 78.6
6-5	-71	92	399	12.8	± 0.6		16.7	8.0 78.9
6-6	-58	63	474	125.0	± 4.0		19.2	8.0 78.3
7-1	-67	148	355	44.1	± 1.9	37.8 ± 1.3	13.7	18.0 84.2
8-1	-64	92	413	275.0	± 11.0	37.6 ± 1.4	16.7	39.0 93.0
8-2	-54	63	483	1187.0	± 51.0	40.0 ± 1.5	19.2	39.0 93.0
9-1	-69	116	378	56.4	± 1.8	35.4 ± 1.1	15.3	39.0 107.6
9-2	-55	116	404	543.0	± 12.0	33.4 ± 1.1	15.3	49.0 125.7
10-1	-54	148	379	74.1	± 2.8	36.3 ± 0.9	13.7	49.0 126.0
11-1	-44	188	369	176.0	± 7.0	36.8 ± 1.2	12.2	66.0 149.8
11-2	-44	188	370	194.0	± 8.0	38.4 ± 1.2	12.2	74.0 149.5
11-3	-42	92	458	835.0	± 32.0	39.1 ± 1.5	16.7	74.0 149.1
11-4	-42	92	456	415.0	± 16.0	38.1 ± 1.4	16.7	66.0 149.7
12-1	-44	116	423	130.0	± 5.0	35.1 ± 1.4	15.3	66.0 149.5
12-2	-45	116	422	163.0	± 7.0	35.7 ± 1.4	15.3	74.0 148.5
12-3	-39	66	509	527.0	± 21.0	39.6 ± 1.4	18.9	74.0 149.3
12-4	-39	63	516	549.0	± 22.0	36.5 ± 1.3	19.2	66.0 149.2
13-1	-41	148	400	289.0	± 11.0		13.7	64.0 146.3
13-2	-41	188	375	64.8	± 2.7		12.2	65.0 144.3
13-3	-45	239	344	256.0	± 11.0		10.6	66.0 143.4
13-4	-43	302	324	1230.0	± 40.0		9.1	66.0 145.0
13-5	-22	466	312	352.0	± 12.0		6.1	66.0 144.6
13-6	-4	697	298	259.0	± 7.0		3.0	66.0 144.0
14-1	-53	92	435	96.0	± 4.0		16.7	49.0 135.4
14-2	-48	63	497	277.0	± 12.0	41.5 ± 1.5	19.2	49.0 129.7
15-1	-63	148	363	136.0	± 6.0	38.0 ± 1.2	13.7	39.0 107.7

TABLE 12

Stratospheric HT and HTO from Project Airstream
Mission A-14, 1978 October-November

See TABLE 10 for explanation of data.

NO.	TEMP. (°C)	PRES. (MB)	☉ (°K)	T-ATOMS/MG AIR		AS HT	HEIGHT (KM)	MID- LAT.	MID- LONG.
				AS HTO					
1-2	-54	188	353	60.6	± 1.8		12.2	28.8	94.7
1-4	-65	116	386	12.7	± 0.7		15.3	29.1	94.2
1-5	-63	92	416	65.0	± 2.9		16.7	29.0	94.3
3-1	-79	116	360	208.0	± 6.0	32.4 ± 1.4	15.3	18.0	84.3
4-1	-78	92	387	42.4	± 1.8	33.4 ± 1.4	16.7	18.0	84.3
4-2	-62	63	465	468.0	± 17.0	41.3 ± 1.7	19.2	18.0	84.3
5-1	-76	116	364	30.1	± 1.3	35.0 ± 1.5	15.3	1.0	79.7
5-2	-73	92	396	41.6	± 1.8	46.4 ± 1.9	16.7	1.0	79.7
7-1	-69	148	352	27.2	± 1.2	41.1 ± 1.6	13.7	18.0	84.2
8-1	-53	93	434	678.0	± 13.0	36.8 ± 1.5	16.7	39.0	93.2
8-2	-50	65	488	1592.0	± 28.0	33.7 ± 1.5	19.0	39.0	93.0
9-1	-65	116	385	37.7	± 1.7	35.3 ± 1.3	15.3	39.0	108.3
9-2	-65	116	386	68.6	± 2.8		15.3	49.0	125.9
10-1	-50	148	386	159.9	± 6.7	31.2 ± 0.8	13.7	49.0	128.7
11-1	-53	188	355	149.0	± 6.0	30.2 ± 1.1	12.2	66.0	149.8
11-2	-54	188	353	238.0	± 9.0	30.5 ± 1.1	12.2	74.0	149.7
11-3	-56	92	429	958.0	± 31.0	38.1 ± 1.5	16.7	74.0	150.8
11-4	-53	92	436	291.0	± 12.0	37.0 ± 2.0	16.7	66.0	150.0
13-1	-23	697	277	405.0	± 13.0		3.0	60.8	157.6
13-2	-41	466	289	51.6	± 2.0		6.1	60.8	156.2
13-3	-53	302	309	51.2	± 2.1		9.1	60.7	157.5

TABLE 13

Stratospheric HT and HTO from Project Airstream
Mission A-15, 1979 April

MSN denotes the mission number, FLT is the flight number from the Project Support Plan, SX is the sequential sample number on each flight. Other flight parameters are calculated from the flight log as explained in Table 10.

MSN	FLT	SX	LAT	LON	ALT	PRES	TEMP	θ	T-ATOMS		PER MG AIR	
					KM	MB	(C)	(K)	HTO	±	HT	±
15	01	001	29.3	94.0	9.1	302	-39	330	10.6	0.6		
15	01	002	29.4	94.1	12.2	188	-56	351	4.1	0.4		
15	01	003	29.1	94.2	13.7	140	-63	364	7.4	0.5		
15	01	004	29.4	94.0	15.2	116	-69	378	5.6	0.4		
15	01	005	29.2	94.1	16.8	92	-70	402	8.9	0.7		
15	01	006	29.3	94.1	19.2	63	-60	470	31.0	1.5		
15	03	001	18.0	84.3	15.2	116	-74	367	44.3	2.1	31.4	1.5
15	04	001	18.0	79.1	16.7	92	-76	390	26.3	1.3	33.8	1.4
15	04	002	18.0	79.1	19.0	64	-67	451	68.4	2.6	38.4	1.6
15	05	001	1.0	79.6	15.2	116	-78	361	29.6	1.4	73.8	2.6
15	05	002	1.0	79.7	16.8	92	-85	372	14.7	1.0	53.6	2.2
15	05	003	1.0	79.6	18.3	72	-77	415	39.2	2.0	55.0	2.3
15	05	004	1.0	79.7	19.2	63	-62	465	165.6	5.7	36.3	1.7
15	06	001	6.1	81.1	6.0	471	-6	331	52.3	4.9	34.3	1.3
15	08	001	39.0	93.2	16.8	92	-62	419	28.3	1.5	35.5	1.5
15	08	002	39.0	93.4	19.1	63	-55	480	56.5	2.3	83.1	3.0
15	09	001	39.0	109.1	15.2	116	-57	400	631.5	12.1	28.3	1.1
15	09	002	49.0	125.8	15.2	116	-52	408	286.3	4.9	32.2	1.2
15	10	001	49.0	126.6	13.7	148	-51	384	56.9	1.7	38.1	1.4
15	11	001	66.0	148.4	12.2	188	-50	360	460.8	16.6	34.1	1.4
15	11	002	74.0	147.3	12.2	188	-46	367	783.3	25.8	34.9	1.6
15	11	003	74.0	147.1	16.8	92	-50	442	1221.7	38.7	36.5	1.8
15	11	004	66.0	148.5	16.8	92	-53	436	552.3	19.2	37.8	1.8
15	12	001	66.0	148.8	15.2	116	-51	411	233.8	7.4	26.3	1.2
15	12	002	74.0	147.4	15.2	116	-47	417	447.0	15.6	42.4	1.9
15	12	003	74.0	148.4	18.8	66	-45	495	820.0	29.9	41.7	2.2
15	12	004	66.0	149.0	19.2	63	-47	499	375.0	14.5	40.1	1.9
15	13	001	62.0	145.8	3.1	697	-3	299	15.7	0.9		
15	13	002	61.9	145.1	6.1	467	-23	311	6.4	0.4		
15	13	003	62.2	144.9	10.7	239	-58	323	3.7	0.4		
15	13	004	62.1	144.8	12.2	188	-66	334	52.3	1.8		
15	13	005	62.0	145.1	13.7	148	-60	368	288.1	8.0		
15	13	006	61.5	147.4	16.8	92	-55	433	187.0	5.0		
15	14	001	49.0	127.3	16.8	92	-57	427	318.0	10.4	45.2	1.8
15	14	002	49.0	126.9	19.2	63	-55	482	275.2	10.4	32.9	1.5
15	15	001	39.0	106.7	13.7	148	-55	376	960.0	31.1	49.1	1.8

TABLE 14

Stratospheric HT and HTO from Project Airstream
Mission A-16, 1979 July-August

MSN denotes the mission number, FLT is the flight number from the Project Support Plan, SX is the sequential sample number on each flight. Other flight parameters are calculated from the flight log as explained in Table 10.

MSN	FLT	SX	LAT	LON	ALT	PRES	TEMP	Θ	T-ATOMS PER MG AIR			
					KM	MB	(C)	(K)	HTO	±	HT	±
16	01	001	28.8	94.1	9.1	302	-34	337	9.3	0.6		
16	01	002	28.8	94.4	12.2	188	-56	351	14.0	0.7		
16	01	003	28.6	94.4	13.7	148	-66	357	10.5	0.6		
16	01	004	28.5	94.4	15.2	116	-71	374	37.0	1.5		
16	01	005	28.8	94.0	16.8	92	-69	404	22.3	1.0		
16	01	006	28.3	94.7	19.2	63	-56	480	328.6	11.5		
16	03	001	18.0	85.6	18.3	72	-63	446	244.6	4.9	27.2	1.2
16	08	001	39.0	94.4	16.8	92	-64	415	134.8	4.7	37.1	1.6
16	08	002	39.0	96.0	19.2	63	-55	482	220.5	10.6	29.9	1.5
16	09	001	39.0	107.7	15.2	116	-63	388	58.4	2.4	32.8	1.2
16	09	002	49.0	125.7	15.2	116	-58	398	89.1	3.4	35.0	1.3
16	10	001	49.0	126.2	13.7	148	-57	373	37.6	1.5	29.7	1.1
16	11	001	66.0	148.5	12.2	188	-48	363			33.5	1.3
16	11	002	74.0	148.2	12.2	183	-47	365	204.2	7.3	43.9	1.7
16	11	003	74.0	148.9	16.8	92	-45	453	461.8	14.4	31.6	1.4
16	11	004	66.0	149.1	16.8	92	-45	452	144.0	5.5	35.3	1.8
16	12	001	66.1	148.8	15.2	116	-45	421	438.6	9.6	37.2	1.6
16	12	002	74.0	148.4	15.2	116	-46	419	410.1	10.0	35.9	1.6
16	12	003	74.0	148.1	17.8	78	-40	484	639.7	14.8	39.6	1.8
16	12	004	66.0	148.9	18.6	69	-40	501	243.5	8.6	28.8	1.5
16	13	001	63.2	151.8	9.1	302	-43	323	14.8	0.7		
16	13	002	63.4	151.9	12.2	188	-43	372	191.8	5.7		
16	13	003	62.5	151.4	13.7	148	-41	400	193.4	6.1		
16	13	004	62.2	151.5	16.8	92	-46	450	311.6	6.1		
16	13	005	62.1	151.1	18.8	66	-37	512	573.6	11.9		
16	14	001	48.9	125.9	16.8	92	-53	435	60.0	2.4	31.7	1.3
16	14	002	49.1	124.5	19.2	63	-47	499	114.5	4.5	40.9	1.9
16	15	001	39.0	107.1	13.7	148	-63	363	92.3	3.8	28.7	1.1

Table 15. N.H. Stratospheric Burden
(Kilocuries)

Sampling Period	Zr-95 ¹		Tritium (as HTO)			
	to ~20 km	to ~30 km	to ~20 km			
			A	B	C	D
Oct. 24-Nov. 17, 1976	0 ²	0 ²	3,100	3,100	3,100	
Mar. 22-Apr. 10, 1977	50,900	58,100	-----No Data-----			
Jul. 6-22, 1977	38,400	45,500	18,800	19,500	2,100	17,000
Oct. 12-29, 1977	30,600	36,100	8,700	9,200	1,700	7,500
Apr. 6-21, 1978	21,000	³	5,900	6,300	1,100	5,200
Jul. 12-31, 1978			4,000	4,400	950	3,450
Oct. 13-Nov. 6, 1978			5,300 ⁴	5,900 ⁴	750	5,150 ⁴
Apr. 6-24, 1979			3,400	3,900	500	3,450
Jul. 3-26, 1979			2,500	2,900	400	2,500

¹Decay corrected to Chinese test of November 17, 1976.

²Background (last significant test, June 17, 1974; total yield 0.2-1 MT)

³No data available above 20 km at this time.

⁴Questionable - see text.

A. Burdens based on observed data.

B. Burdens from column A decay-corrected to November 17, 1976.

C. Background at time of Chinese 11/17/76 test. Background burdens at later times assumed using a residence half time of 10 months.

D. Residual burden attributed to 11/17/76 test (B minus C).

Table 16. N.H. Tritium (as HTO) Stratospheric Burden to
Approximately 20 km. (Kilocuries)

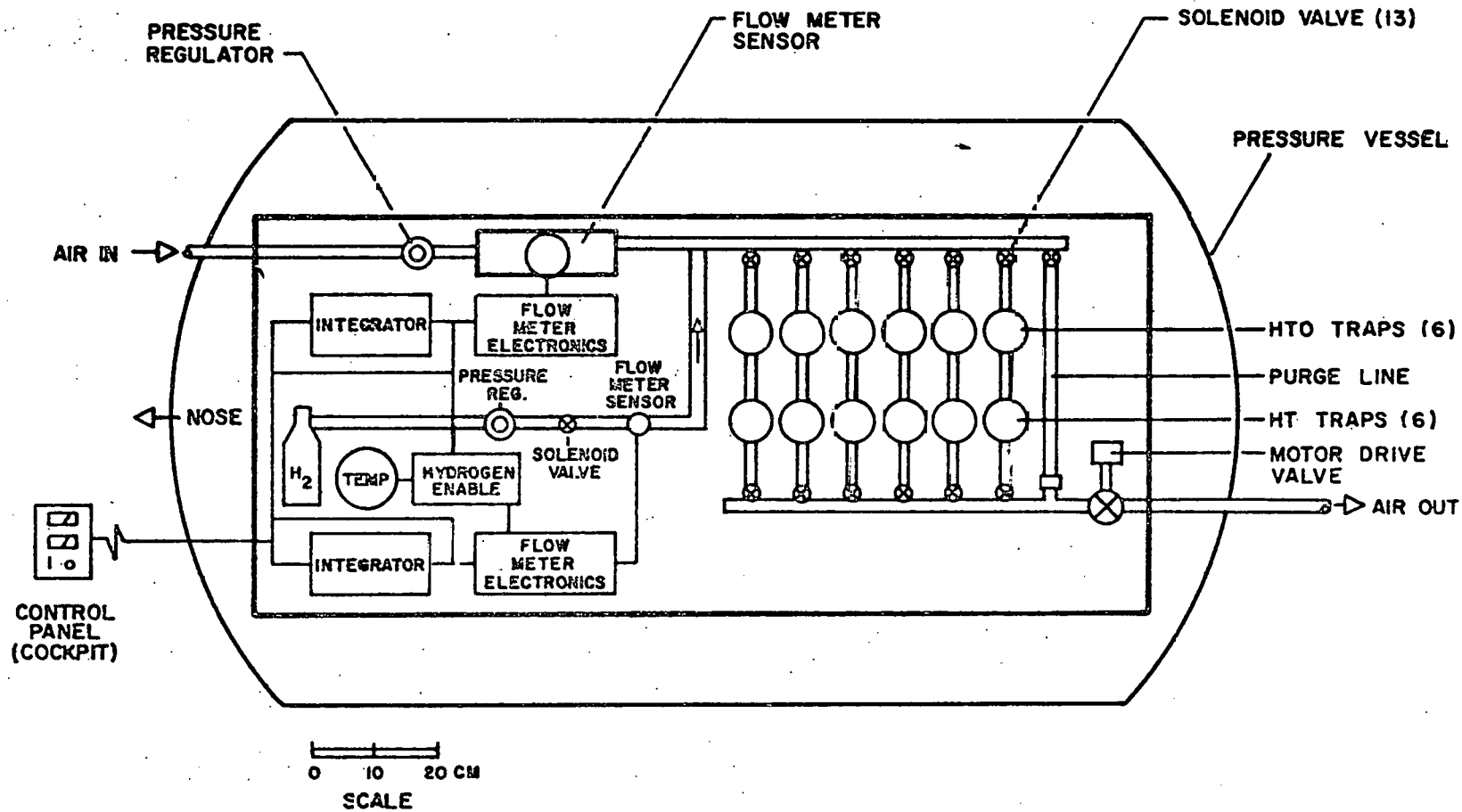
<u>Sampling Period</u>	<u>Observed¹</u>	<u>Decay Corrected² (to 6/27/73)</u>	<u>Obser.-Background³ (Decay Corrected to 6/27/73)</u>
April 18-May 6, 1975	9,400	10,500	7,300
July 14-Aug. 5, 1975	4,300	4,800	2,200
May 22-June 9, 1976	4,900	5,800	4,300
Aug. 12-30, 1976	2,000	2,400	1,300
Oct. 24-Nov. 17, 1976	3,100	3,800	2,800

¹Burden based on observed data (not decay corrected)

²Observed burden decay corrected to 6/27/73

³Observed burden ¹ minus background (decay corrected to 6/27/73)
where:

Background = Assumed input of 6,000 kCi from the June 17, 1974
test had a stratospheric half residence time of 10 months,
decay corrected to time of measurements in 1975 and 1976.



WB-57F SAMPLER
(PLAN VIEW)

Fig. 1. Plan view of the WB-57F aircraft tritium sampler.

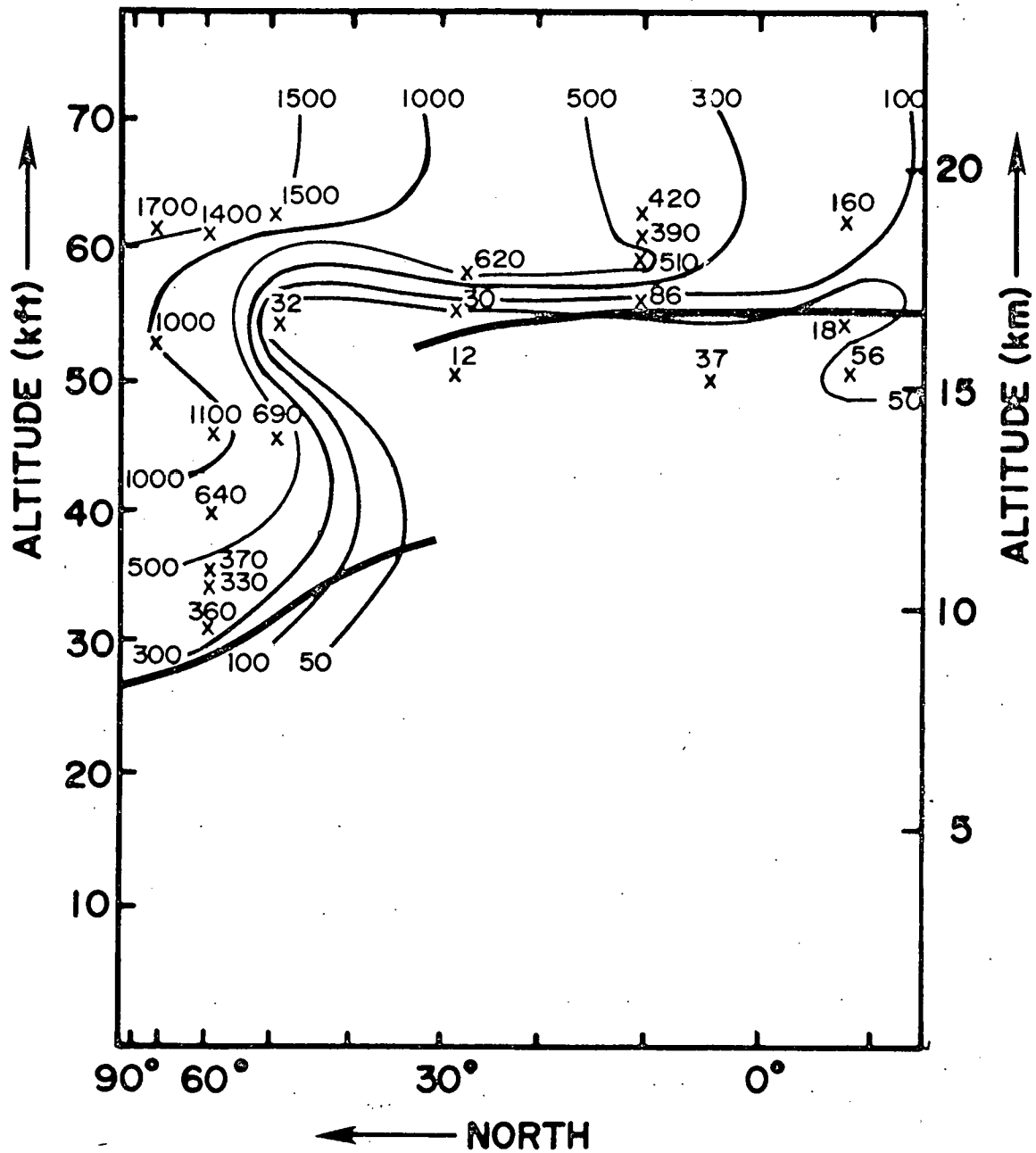


Fig. 2. Distribution of tritium (as HT0) for April 18-May 6, 1975, approximately 22 months after the Chinese test of June 27, 1973. The numbers represent the observed HT0 concentration (T-atoms/mg air). The heavy lines represent the mean tropopause along the sampling corridor.

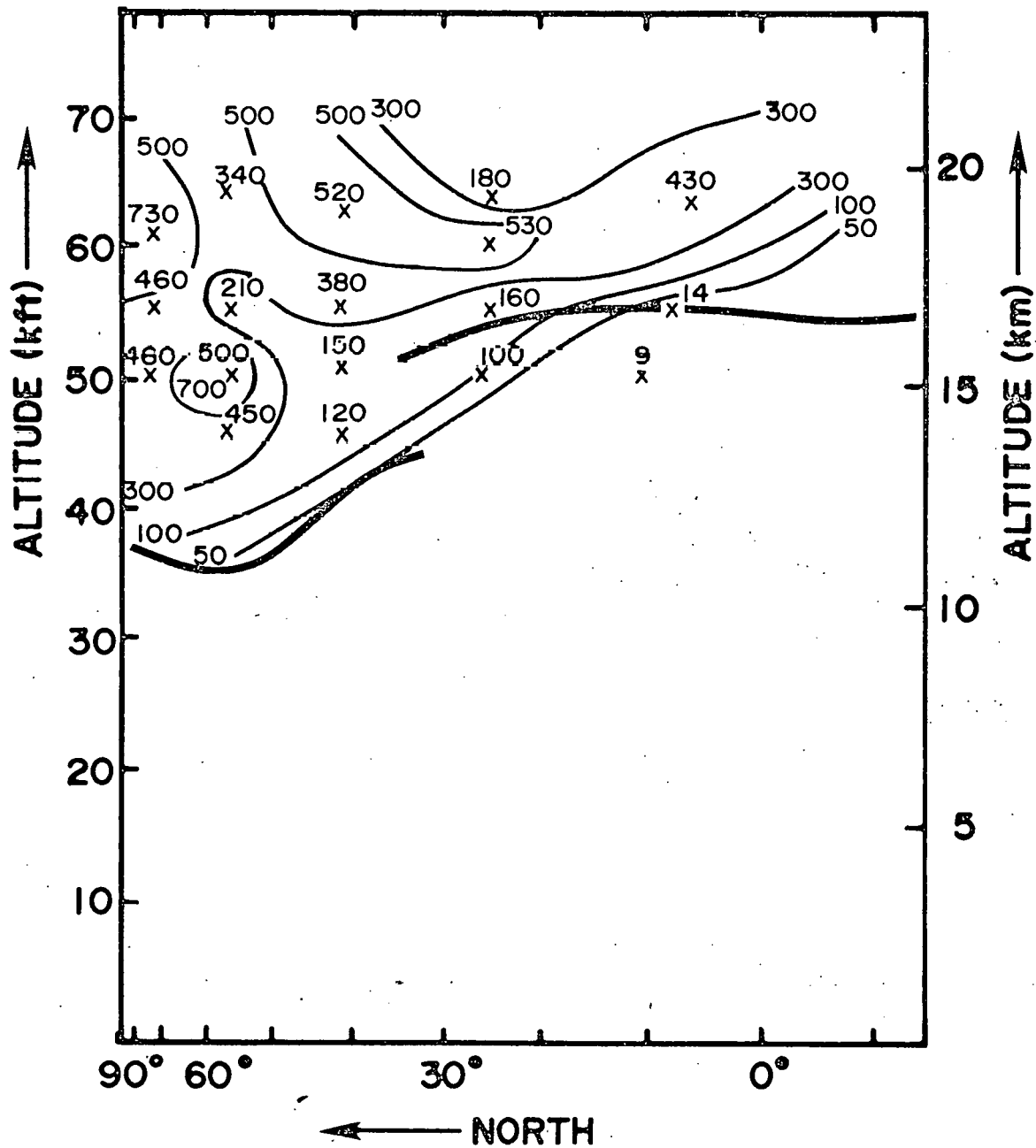


Fig. 3. Distribution of tritium (as HT0) for July 14-August 5, 1975, approximately 25 months after detonation. See legend to Fig. 2 for other identification.

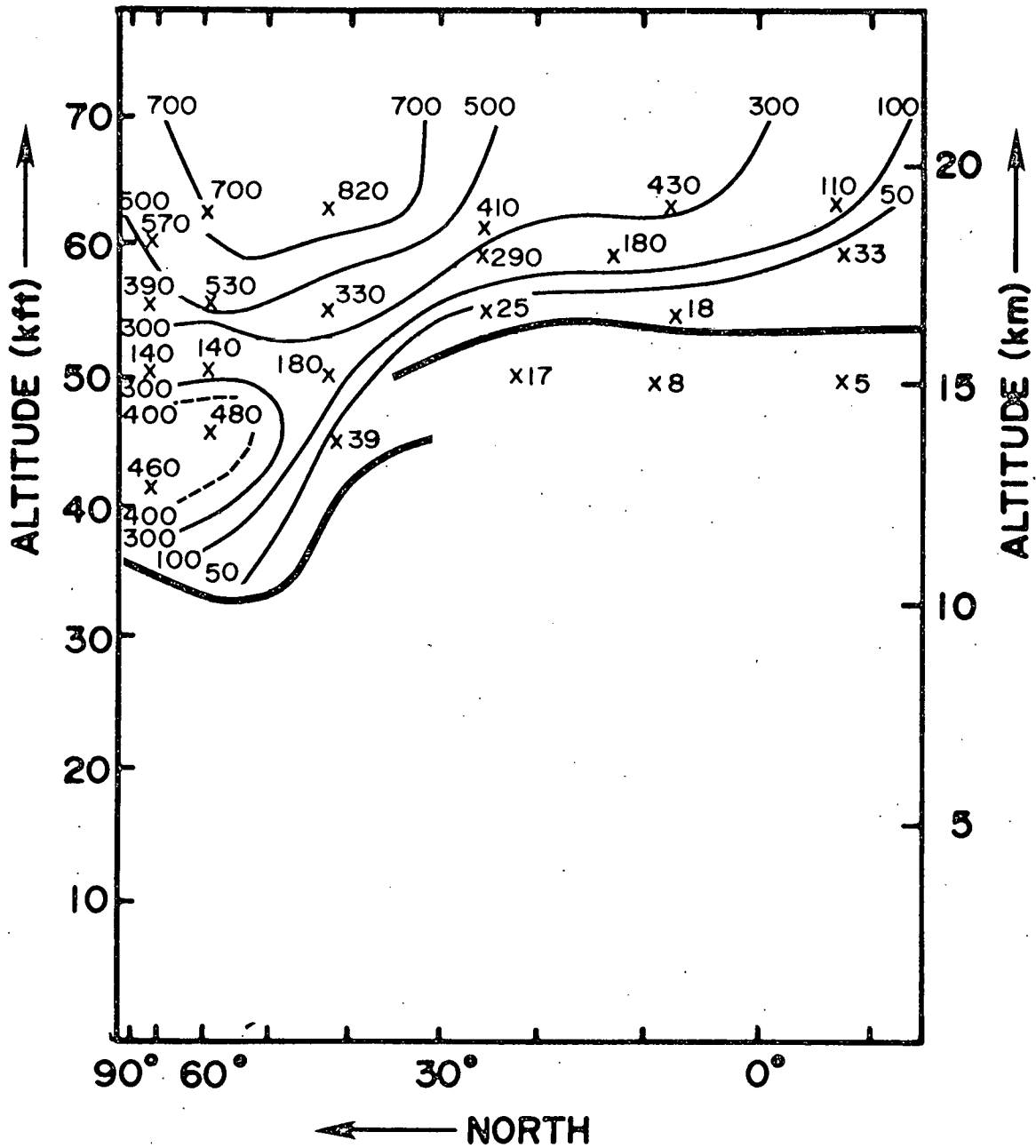


Fig. 4. Distribution of tritium (as HT0) for May 22-June 9, 1976, approximately 35 months after detonation. See legend to Fig. 2 for other identification.

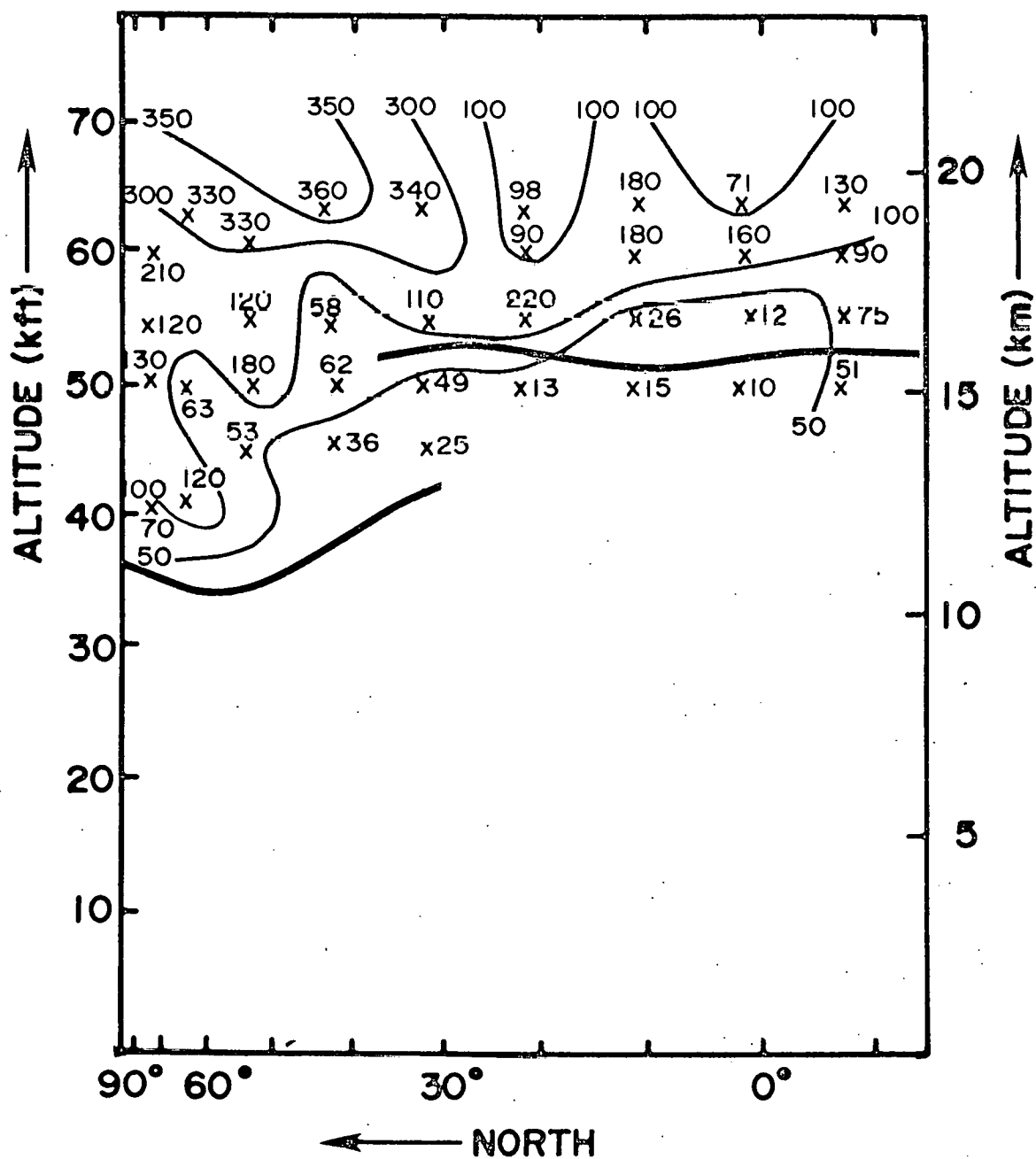


Fig. 5. Distribution of tritium (as HT0) for August 12-30, 1976, approximately 37 months after detonation. See legend to Fig. 2 for other identification.

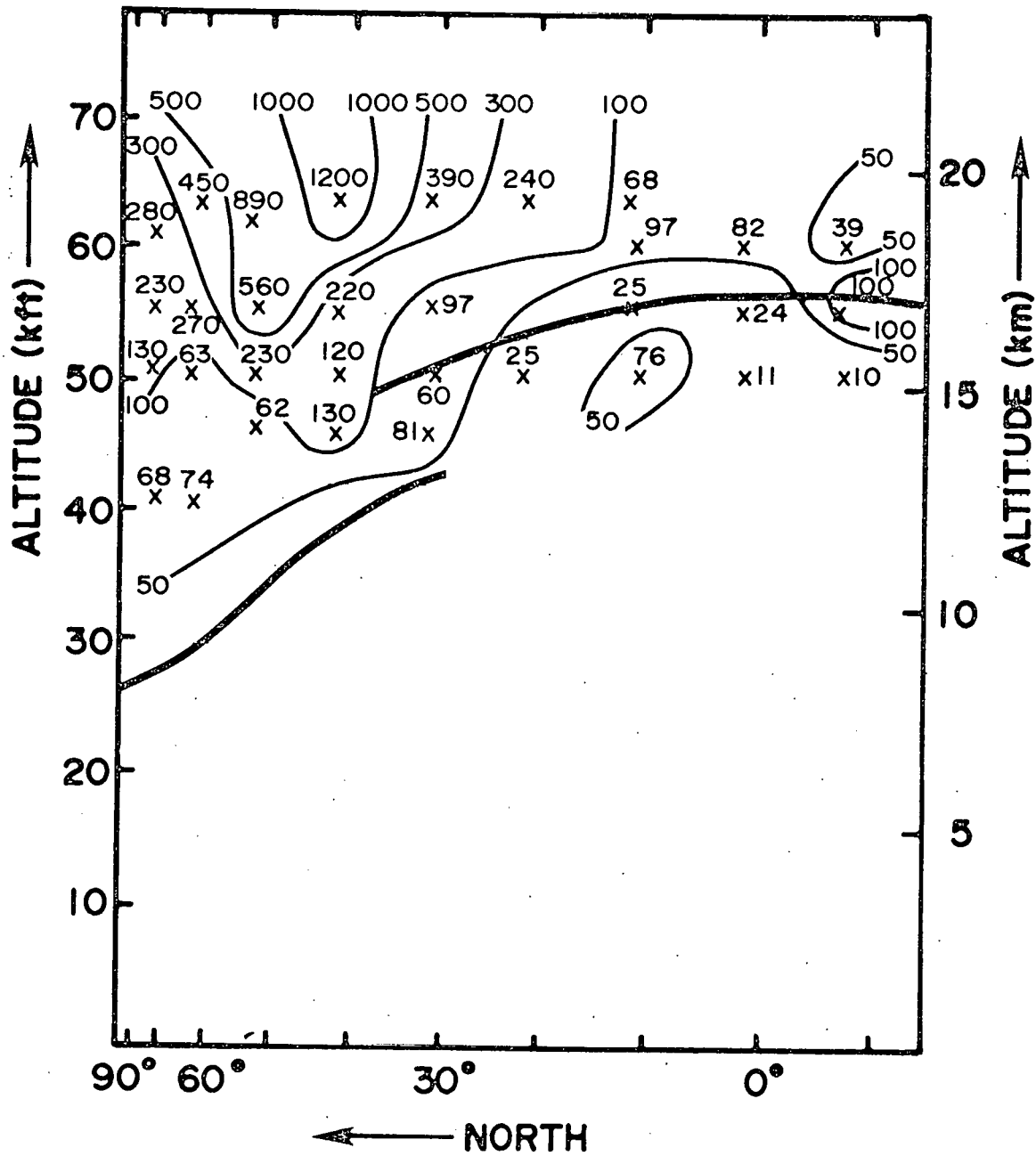


Fig. 6. Distribution of tritium (as HTO) for October 24-November 17, 1976, approximately 40 months after the Chinese test of June 27, 1973, and approximately 1/2 month before the Chinese test of November 17, 1976. See legend to Fig. 2 for other identification.

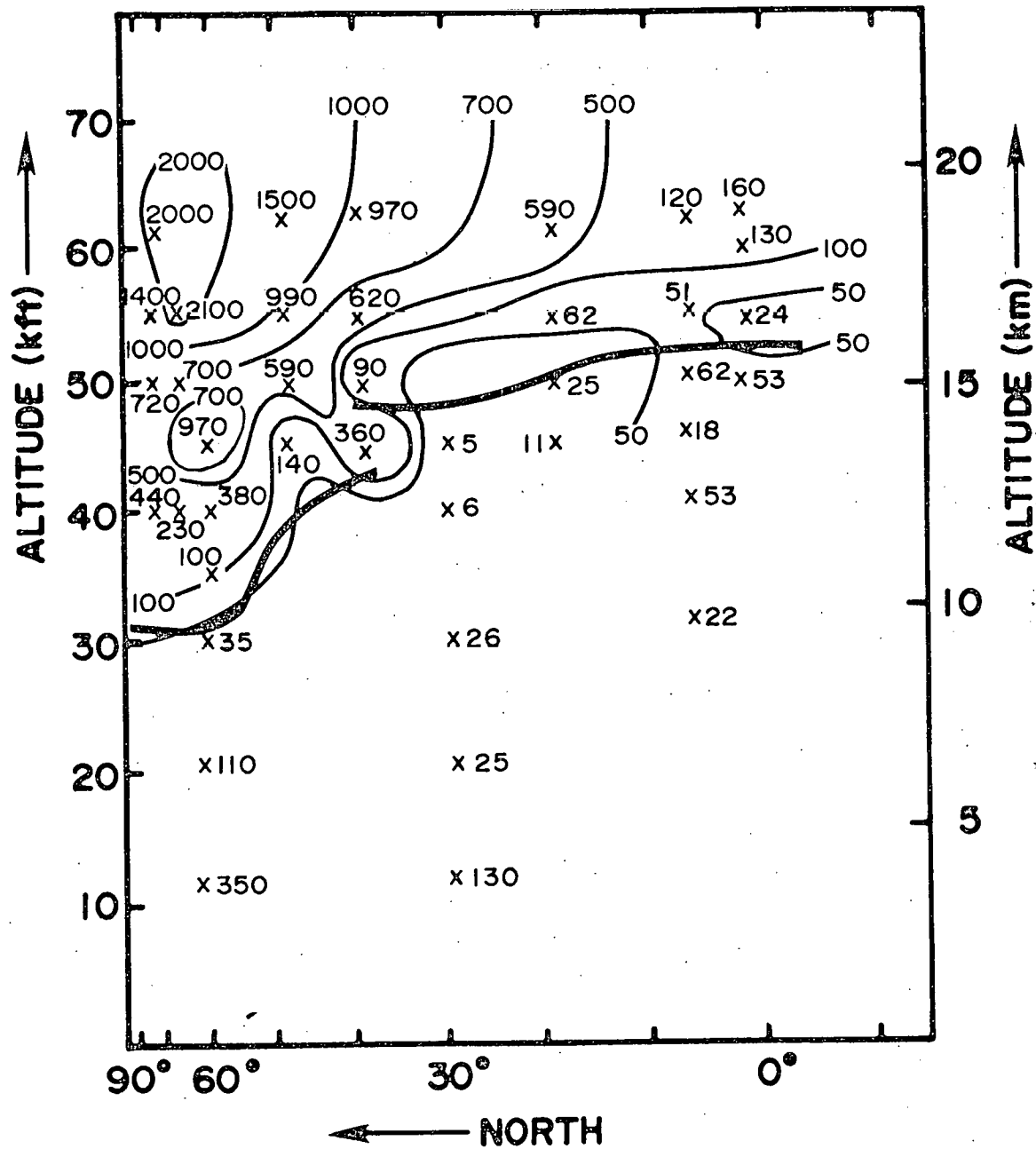


Fig. 8. Distribution of tritium (as HT0) for October 12-29, 1977, approximately 11 months after detonation. See legend to Fig. 7 for other identification.

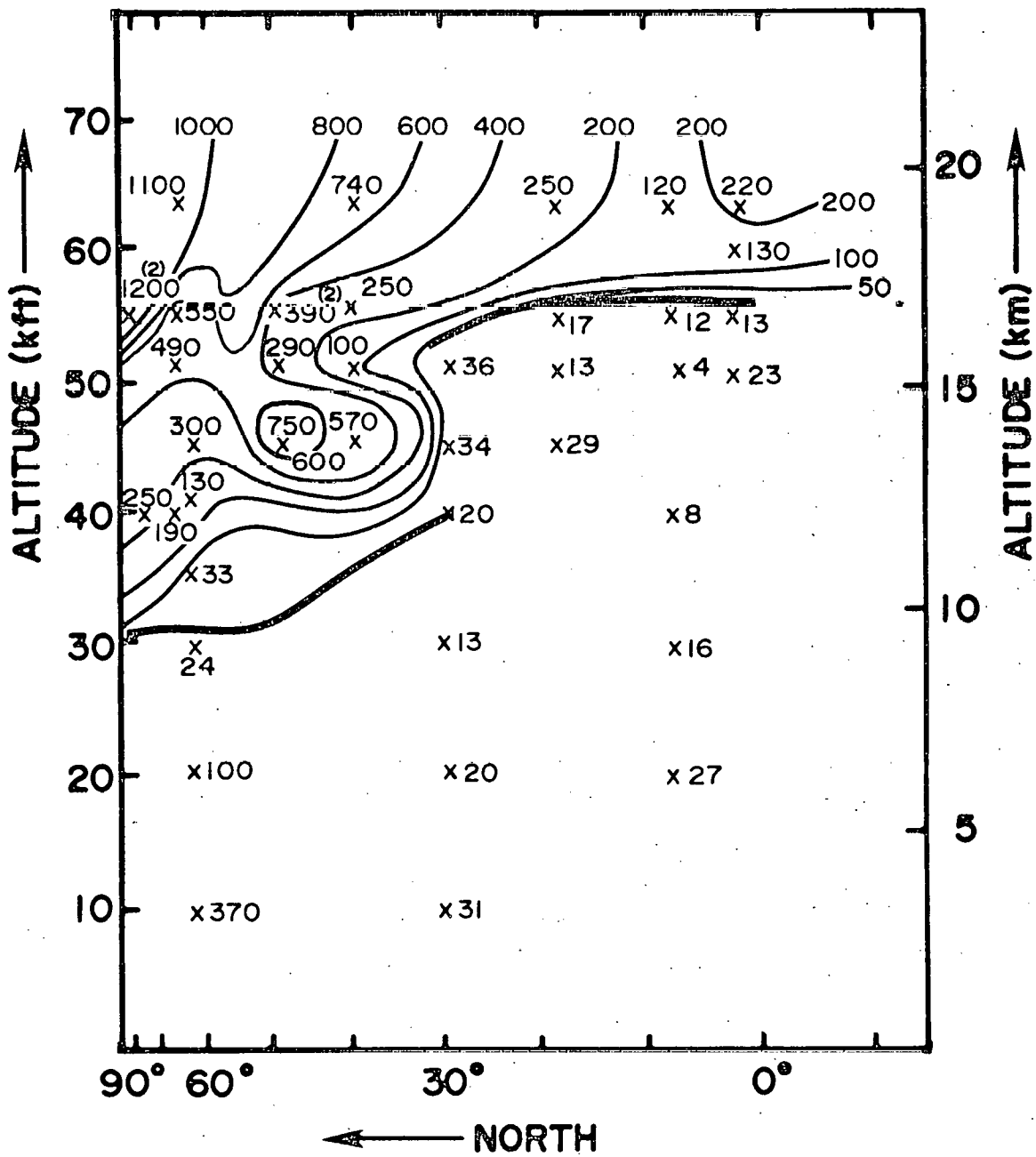


Fig. 9. Distribution of tritium (as HT0) for April 6-21, 1978, approximately 17 months after detonation. See legend to Fig. 7 for other identification. (2) indicates average of two samples in close proximity.

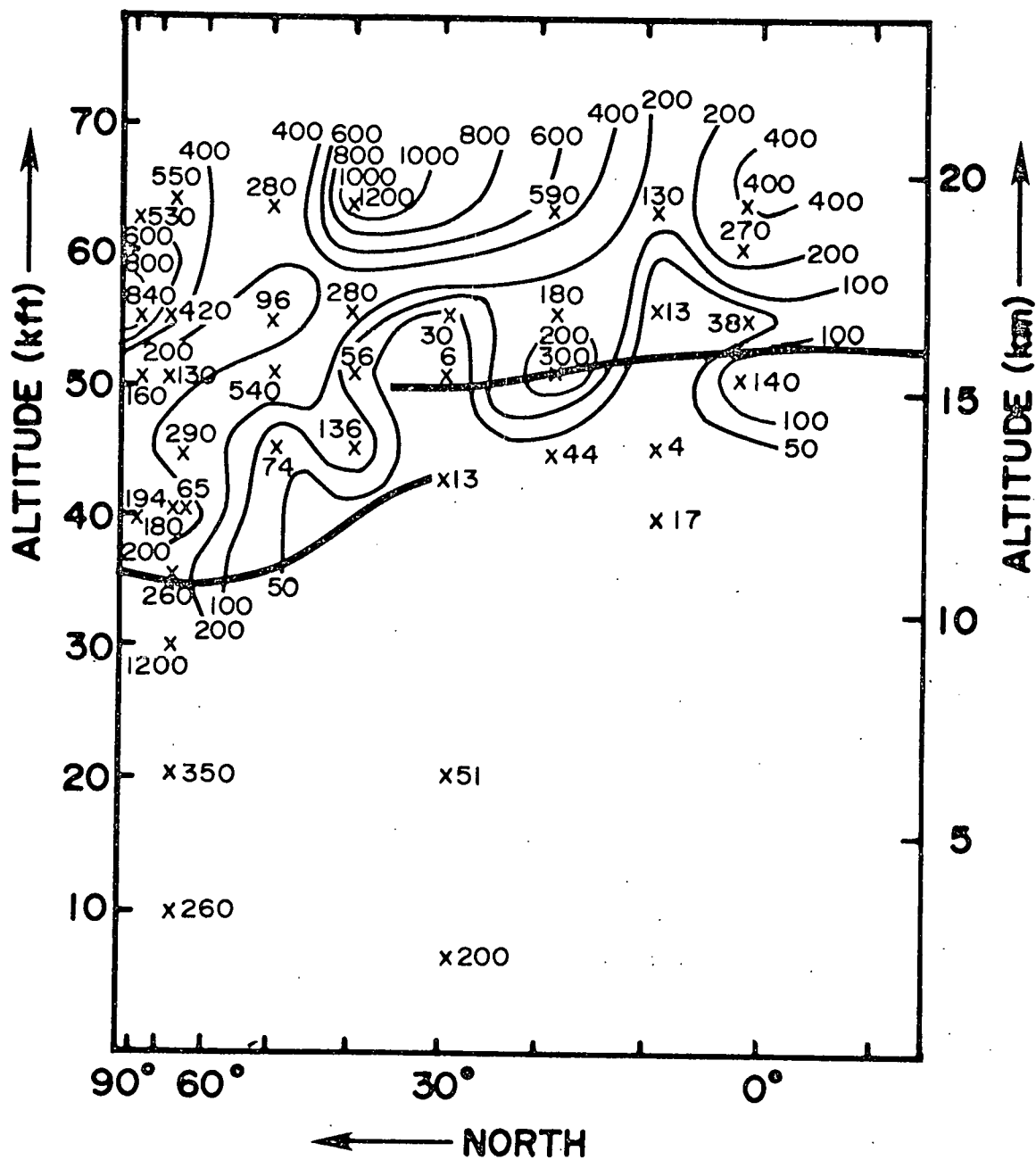


Fig. 10. Distribution of tritium (as HT0) for July 12-31, 1978, approximately 20 months after detonation. See legend to Fig. 7 for other identification. Average tropopause along sampling corridor was assumed (see text).

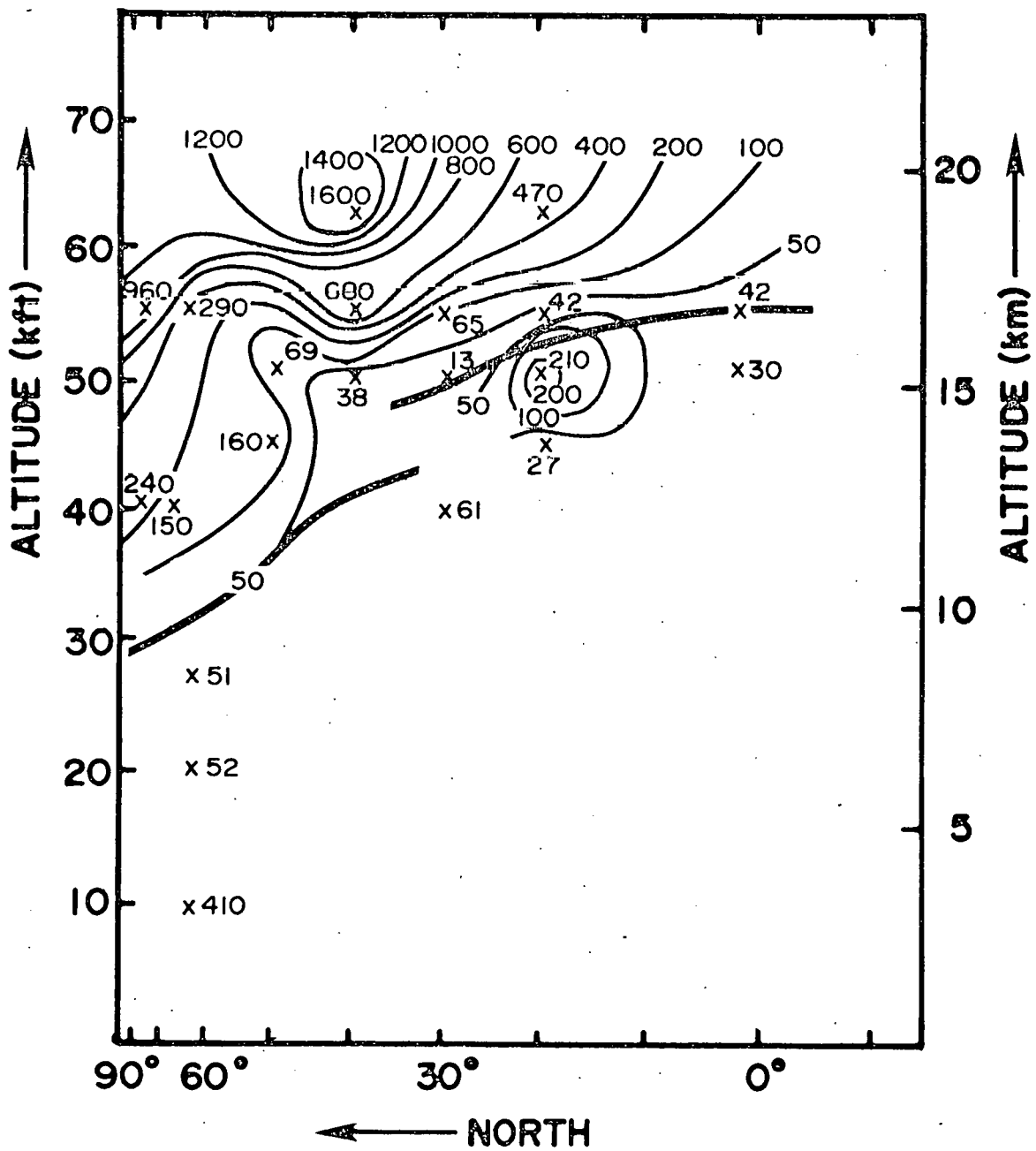


Fig. 11. Distribution of tritium (as HT0) for October 13-November 6, 1978, approximately 23 months after detonation. See legend to Fig. 7 for other identification. Average tropopause along sampling corridor was assumed (see text).

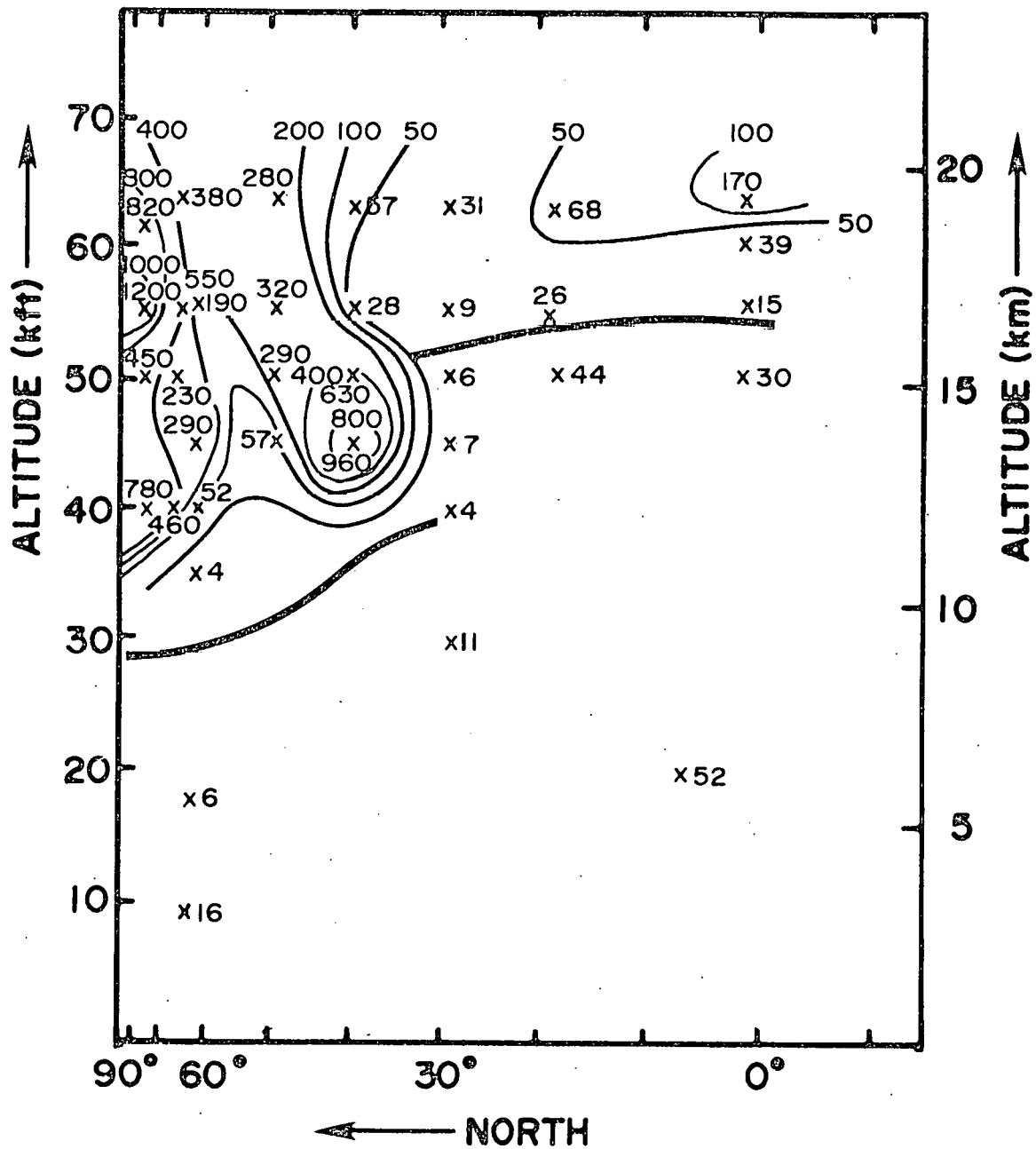


Fig. 12. Distribution of tritium (as HT0) for April 6-24, 1979, approximately 29 months after detonation. See legend to Fig. 7 for other identification. Average tropopause along sampling corridor was assumed (see text).

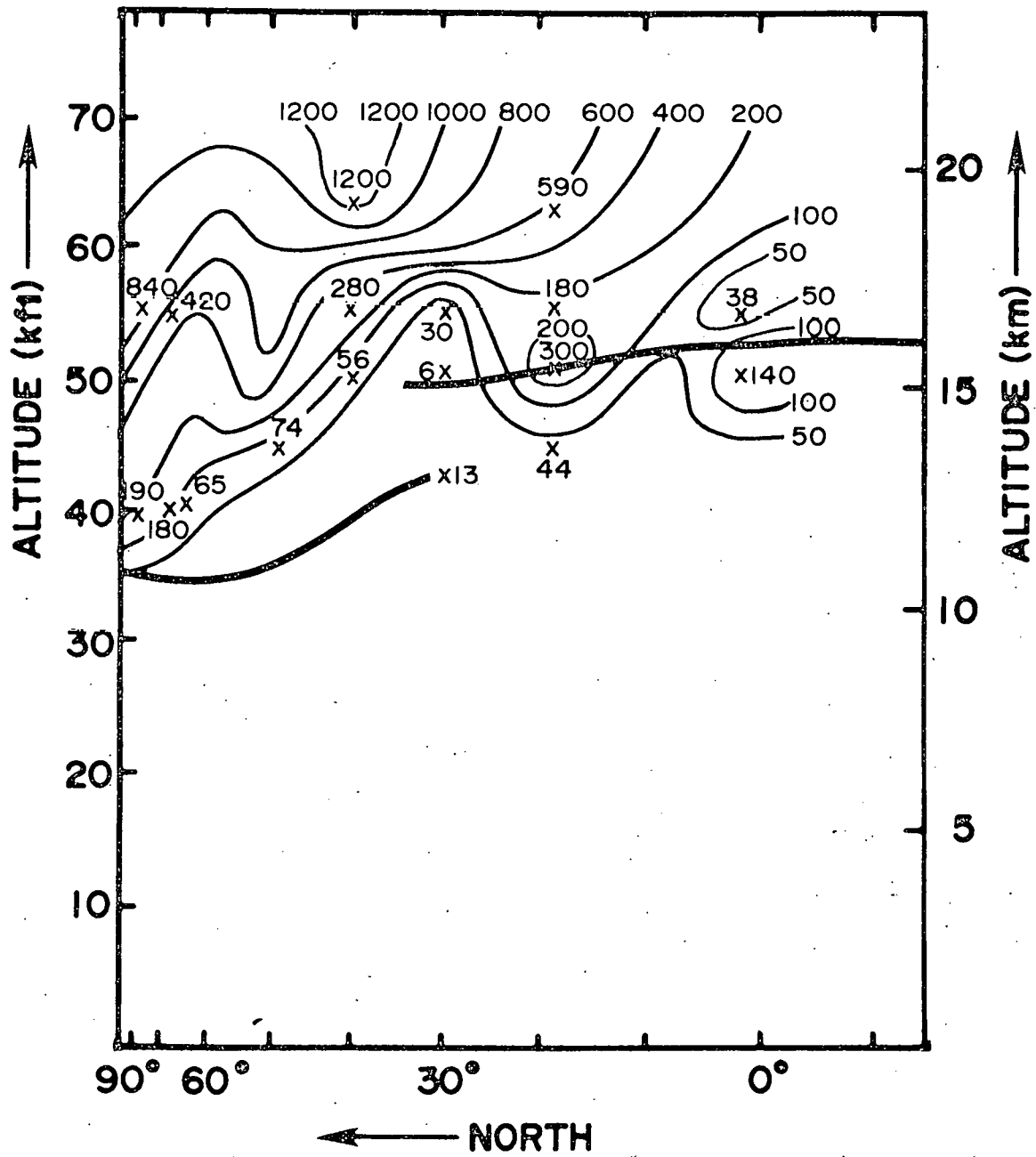


Fig. 14. Reanalysis of the distribution of tritium (as HT0) for July 12-31, 1978, (Figure 10) using limited data. See text.

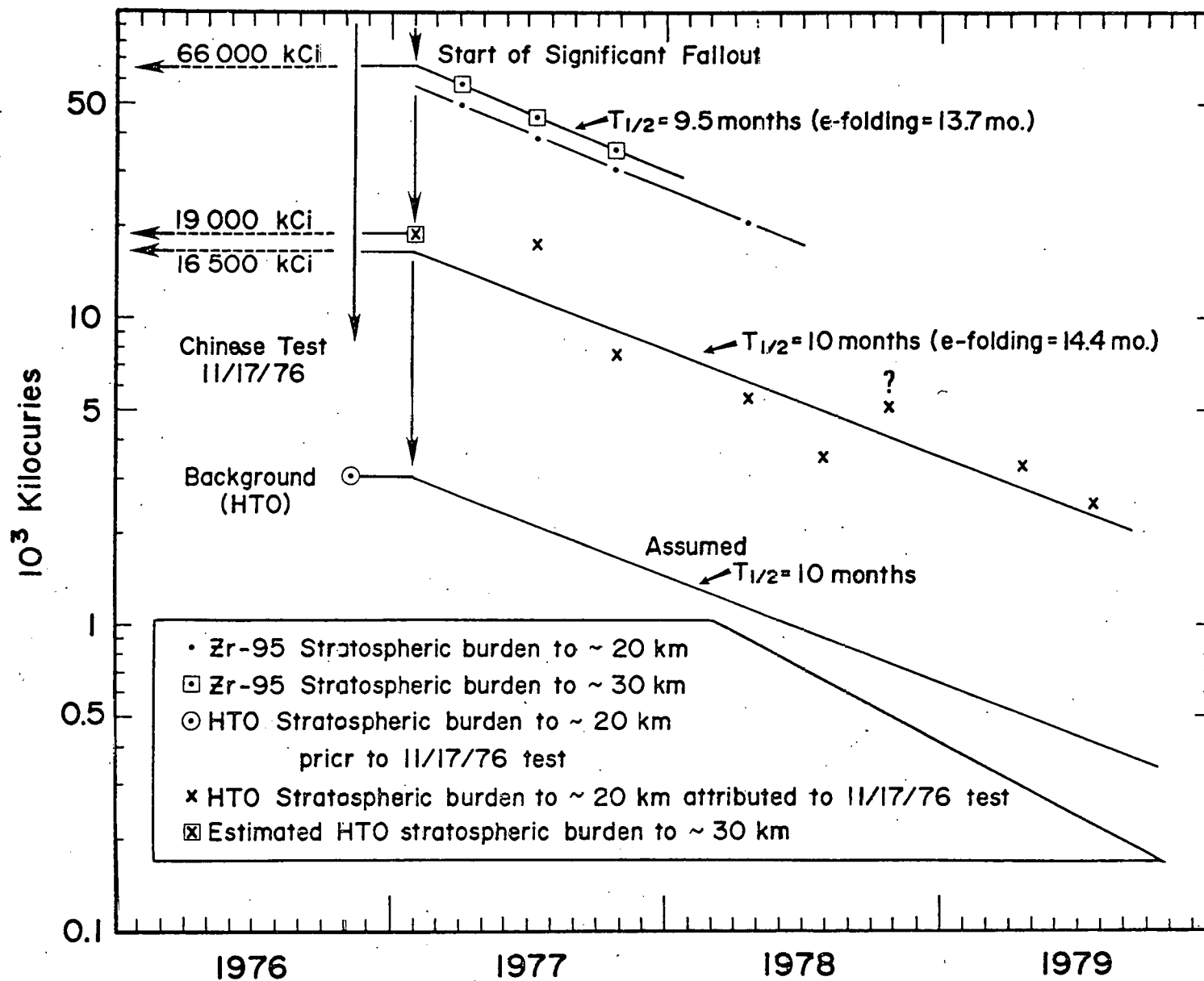


Fig. 15. Stratospheric half residence time of the Zr-95 and HTO burden for the Chinese test of November 17, 1976. All burdens decay corrected to time of test. See text for a more complete description.

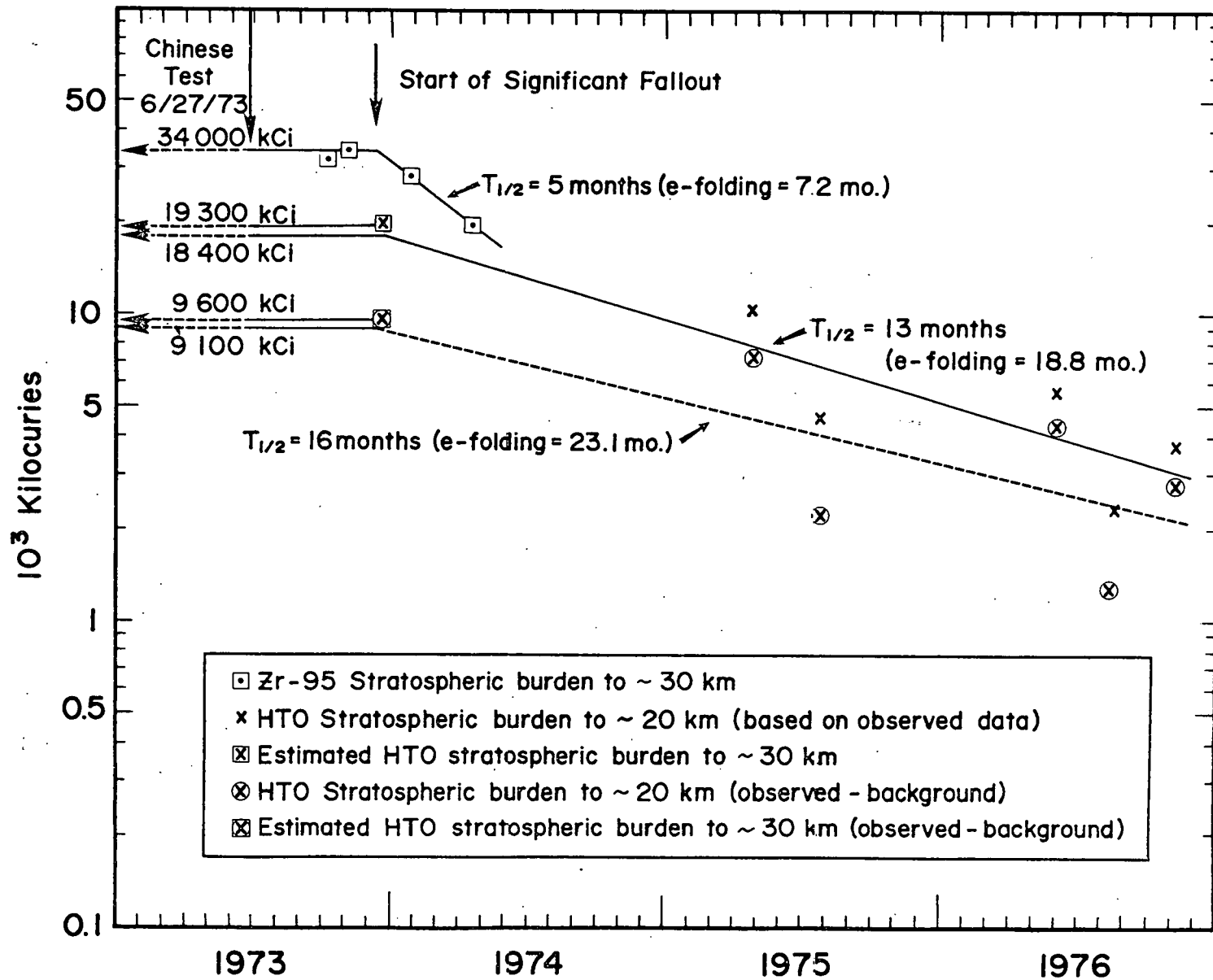


Fig. 16. Stratospheric half residence time of the Zr-95 and HTO burden for the Chinese test of June 27, 1973. All burdens decay corrected to time of test. See text for a more complete description.

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ENVIRONMENTAL TRITIUM APPLICATIONS
TO ATMOSPHERIC AND OCEANOGRAPHIC RESEARCH

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ABSTRACT

Hundreds of kg of tritium have been released to the environment during the nuclear era. Two chemical forms predominate, tritiated water (HTO) and tritium gas (HT and T₂). Scientific utility has been gained from these releases in the fields of atmospheric chemistry, meteorology, oceanography and hydrology. The results are improved estimates of global tritium burdens, of atmospheric and oceanic circulation and mixing processes, and of hydrogen chemistry in the atmosphere.

The inventory of HTO in the oceans was estimated to be 1.6 GCi at the end of 1972. The lower stratosphere of the Northern Hemisphere contained 2.5 MCi of HTO in mid-1979, with probably a similar amount in the Southern Hemisphere. The global HT inventory is ca. 9 MCi.

Utilization of the present environmental levels for tracer studies requires analytical sensitivities below 0.1 pCi/g.

Quantity distribution of T₂-lighted watches is expected to complicate environmental sampling operations, since they typically emit tens of nCi of HTO and lesser amounts of HT daily.

SOURCES OF TRITIUM

Prior to the nuclear era, a small tritium burden, in the form of HTO, was maintained by cosmic ray interactions with the atmosphere. The production rate and distribution were estimated by Craig and Lal (1961). The general features of their estimate were that the tritium is produced at a mean column rate of 0.5 atoms cm⁻²sec⁻¹, that the peak of the production occurs in the vicinities of the geomagnetic poles, and that 2/3 of the production occurs in the stratosphere. Using their estimates, one can further estimate a global stratospheric burden of ca. 2 MCi due to the cosmic source. The tropospheric burden would be very much smaller due to the rapid water turnover time. The majority of HTO would then reside in surface water and ice.

By contrast, atmospheric testing of thermonuclear devices released from 7 to 50 MCi per megaton of fusion (National Council on Radiation Protection and Measurements, 1979). The input during

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the era of large-scale testing was reported by Miskel (1973) to have totalled 8 GCi. Subsequently, six Chinese and five French large thermonuclear tests have released HTO to the stratosphere. The inputs from the two most recent tests have been reported recently (Mason *et al.*, 1980).

Additional releases of HTO occur from nuclear reactors and fuel reprocessing plants, much in the form of liquid water discharges; see for example, Hetherington and Robson (1979). A new source of HTO, and of lesser amounts of HT, is the distribution of T₂ gas-illuminated digital watches. The results of an experiment to measure their leakage will be reported in a following section.

The second major form of tritium input to the environment is in the form of hydrogen gas, HT or T₂, referred to henceforth as HT. Prior to the Threshold Test Ban Treaty implementation in 1974, underground thermonuclear tests frequently released significant amounts of HT to the atmosphere. Two underground tests by the USSR in the fall of 1973 were estimated to have released 80g, or roughly 800 kCi, of HT (Mason and Östlund, 1974). Most HT presumably is released from production, handling, and fuel reprocessing activities.

A minor tritium input is the release of tritiated hydrocarbons, mostly methane, presumably from industrial sources. Some atmospheric concentration measurements have appeared (Haines and Musgrave, 1968).

TRITIUM SAMPLING AND MEASUREMENT

Sampling of open bodies of water for HTO determination is usually done by means of wire-borne multiple samplers such as the rosette apparatus, which takes samples at multiple depths. Vertical profiles of other parameters, such as salinity, temperature, and stable chemical constituents, are normally taken simultaneously.

Atmospheric HTO sampling was first done by collection of rainfall. Our work, however, is based upon adsorption of water vapor, and is combined with collection of HT and tritiated hydrocarbons by catalytic oxidation and adsorption. A description of the technique has been published (Mason and Östlund, 1979).

The sensitivity required for tracer use of tritium at present environmental levels has been achieved by low-level gas proportional counting,

preceded by electrolytic enrichment for highest sensitivity when sufficient sample quantities are available. The detection limit without enrichment is 6 TU or 0.02 pCi/g. Enrichment lowers this to 0.1 TU or 0.0003 pCi/g. Details of the techniques were reported by Östlund and Dorsey (1977).

GLOBAL BURDENS

The most recent comprehensive oceanic tritium survey was made as part of the Geochemical Ocean Section Study (GEOSECS) program in the early 1970s. Those results, supplemented by other cruises in the same era, and decay-corrected to 1 January 1972, indicated an oceanic burden of 164 kg or 1600 MCi of tritium in water form (Östlund and Fine, 1979).

Additional aqueous tritium is present in lakes, ground water, and the Greenland and Antarctic ice caps. The latter may contain a significant fraction of the atmospheric input of the 1960s, based on analyses by Jouzel *et al.* (1979).

The tropospheric HTO and HT burdens have been established by our sampling program, which presently includes stations at Fairbanks, AK; Miami, FL; and Baring Head, New Zealand. Samplers have also been located at Mauna Loa, HI, and the South Pole at various times. Stratospheric sampling is conducted regularly as part of Project Airstream, which provides a comprehensive survey from the tropopause to 19 km altitude, over the latitude range between the equator and 75° north. Stratospheric balloons are now being utilized to extend the altitude coverage to 27 km. As of mid-July 1979, the Northern Hemispheric stratospheric burden was estimated at 2.5 MCi of HTO (Mason *et al.*, 1980). The tropospheric burden is very small by comparison, roughly 2 kCi (Mason and Östlund, 1979), due to the rapid scavenging of water vapor from the troposphere.

The global HT burden had declined to 0.93 kg (9 MCi) at mid-1978, from a high of 1.24 kg (12 MCi) in 1974. Tropospheric and stratospheric mixing ratios are normally equal, due to the long atmospheric chemical residence time of hydrogen gas (Mason and Östlund, 1979).

ATMOSPHERIC APPLICATIONS

Stratospheric HTO is a conservative tracer for water transport. Early studies such as those reported by Mason and Östlund (1976) used vertical profiles to estimate the vertical flux and stratospheric residence time. These studies were an extension of the tropospheric work of Ehhalt (1971, 1973). The acquisition of a far larger data set by means of Project Airstream has enabled calculation of burdens over a four-year time span, and a better estimate of residence time. Using the Chinese thermonuclear test of late 1976 as a source, Mason *et al.* (1980) reported a 10-month half-time for HTO injected in the 19 km height region. The complementary ⁹⁵Zr and HTO burdens are shown in Figure 1 taken from that report. The observed HTO burdens are denoted by x's, and the regression lines were obtained by least-square

fits. The late 1978 data, shown with a question mark, were sparse due to an aircraft problem during the Alaskan portion of the deployment, and are too few to be weighted heavily in the analysis. It is encouraging that the residence times of ⁹⁵Zr and HTO are in very good agreement, indicating that particle settling in the size range typical of ⁹⁵Zr can be disregarded. More detailed examination of the Airstream data suggests that it may be useful for study of latitudinal and vertical water transport; however, at least one or two years' additional data are needed. The Project Airstream instrumentation ensemble is currently being expanded to include continuous recording of water vapor, ozone, NO_x, and condensation nuclei concentrations.

Measurements of HT mixing ratios in the troposphere have been applied to study of the chemical residence time of hydrogen gas. Interhemispheric flights have shown lower mixing ratios in the Southern Hemisphere, which were interpreted to indicate a hydrogen chemical residence time of 6.5 years. Vertical HT profiles show no decreases through the tropopause and up to 19 km, supporting the long residence time estimate (Mason, 1977).

OCEANOGRAPHIC APPLICATIONS

Analysis of the Atlantic deep water formation by means of the GEOSECS tritium profiles has been reported by Östlund and Fine (1979), as has the use of earlier Atlantic tritium profiles for study of mixing through the thermocline (Rooth and Östlund, 1972).

Recent Pacific applications include study of the exchange times in the Pacific equatorial system (Fine and Östlund, 1980). In brief, a portion of the westward-flowing North Equatorial Current turns back eastward and recirculates as the North Equatorial Counter Current. Classical oceanographic parameters of salinity, temperature, and oxygen were not usable to define the exchange time scale of this process. Availability of tritium data permitted formulation of a two-box model which resulted in findings of no net flux between the two currents, and a lower bound of five years for exchange in the thermocline.

TRITIUM GAS-ILLUMINATED WATCHES

Large numbers of digital watches which include encapsulated tritium gas light sources are entering the market. Typically, two small gas capsules are embedded in a liquid crystal display. They contain an interior surface coating of phosphor, and are filled with T₂ gas at low pressure. The total allowable quantity of tritium is 200 mCi per watch, and the assumed leakage rate is 50 nCi per day. In order to explore the potential contamination problem which these watches might pose for our global monitoring task, we obtained 33 watches which had already been subjected to durability testing. These watches were separated into three groups according to appearance and functional condition. Two groups of nearly-new appearance contained 10 and 11 watches, respectively, while the third

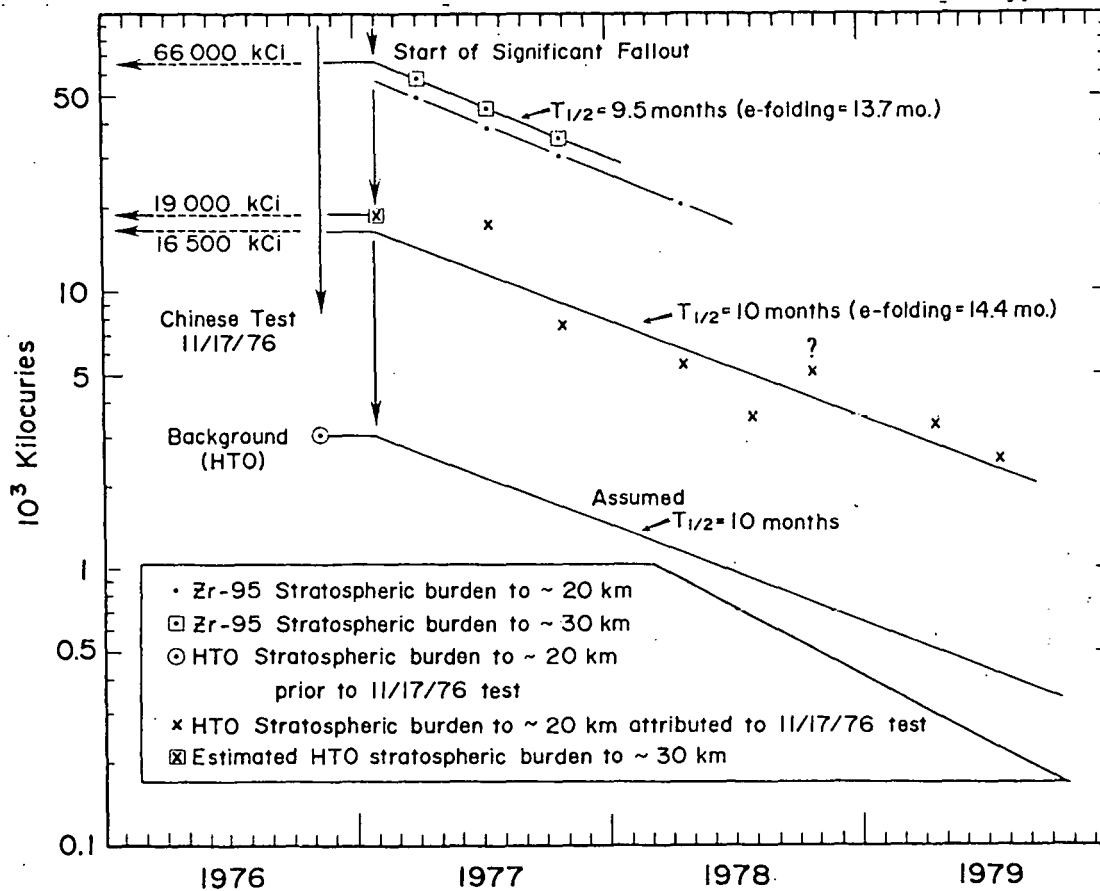


Fig. 1 Stratospheric HTO and ^{95}Zr burdens 1976-1979.

group of 12 watches included mostly worn-appearing or inoperative units. All the display elements were intact, and all were of equal brilliance to the observer's eye. The first test made was to place each group in the inlet of a high-flow-rate sampler of the configuration shown in Figure 2. This sampler, normally used aboard aircraft, operates by first adsorbing all atmospheric moisture in a trap of molecular sieve, then oxidizing atmospheric hydrogen plus carrier H_2 on a palladium-coated molecular sieve. The sampling and analysis techniques have been reported by Östlund and Mason (1974). Air was drawn over each group of watches by a pump attached to the sampler outlet. The samples were extracted from the traps and analyzed, and the apparent leakage rates calculated. Table 1 shows the results, which indicate that group 1, which consisted of functioning watches of good appearance, leaked well in excess of the allowable rate. Also noteworthy is the finding that the leakage was predominately in the form of HTO.

Following this experiment, the watches of group 1 were sampled individually using the simpler setup shown in Figure 3 for collecting HTO only. The collected moisture was analyzed by liquid scintillation. The results identified one watch of the

TABLE 1
Initial Watch Evaluation

Group	# of watches	Sampling time, min	Activity found, nCi		Leak rate, nCi/day	
			HTO	HT	HTO	HT
Blank	0	10	7.4×10^{-5}	1.9×10^{-3}	---	---
1	10	10	10.6	0.1*	1530	14*
2	11	10	1.2	0.24	170	35
3	12	10	0.9	0.22	130	32

*Lower limits, portion of sample lost in extraction.

group as the source of the high level of leakage. It was then run alone in the aircraft sampler. An HTO leakage of 2500 nCi/day was found, or 50 times the allowable amount. This watch was then disassembled, and the individual sub-assemblies sampled. The findings were that all of the parts emitted HTO. Subjecting them to vacuum for an hour suppressed the emission from all parts except the tritium capsule-containing display. This expected result confirmed that this particular display unit leaked tritium in the form of HTO, and that the other components had acquired an HTO burden by adsorption. The leakage rate found could be sustained for the probable life of the watch by the allowed filling of 200 mCi. The discrepancy between the findings in Table 1 and the later experiment (1500 vs. 2500 nCi/day) is attributed to the handling of the watch between experiments. It

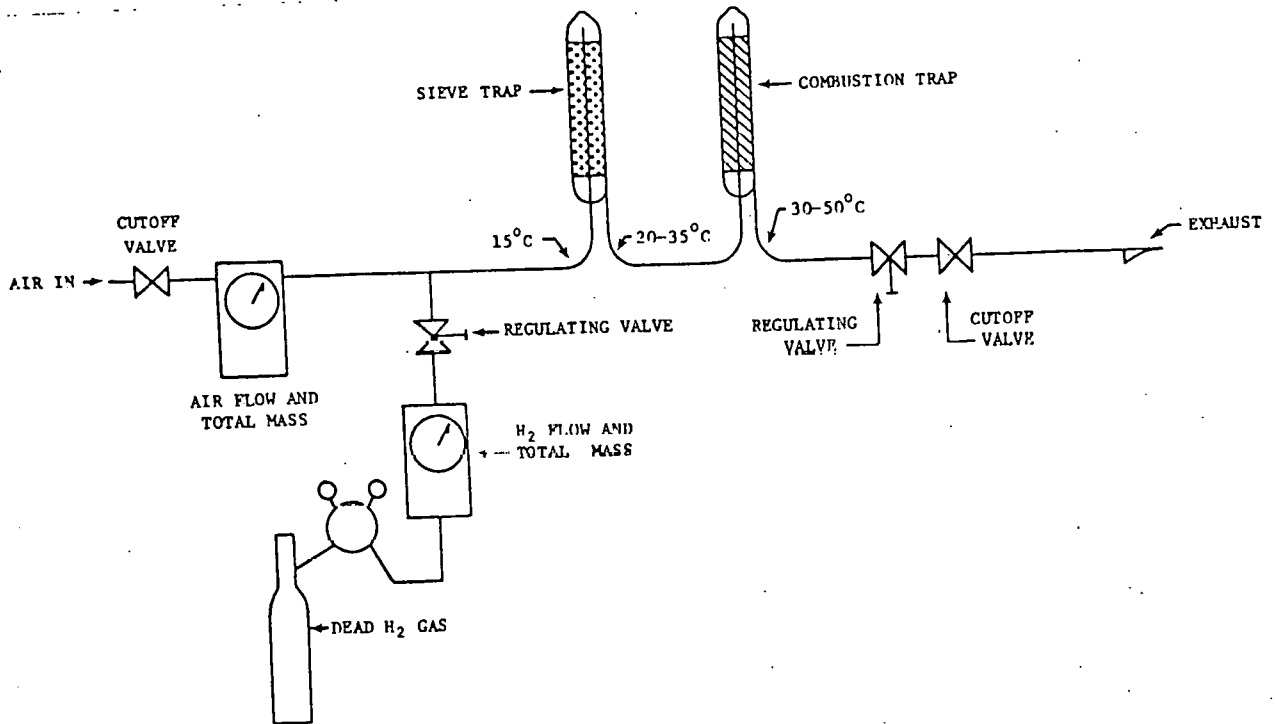


Fig. 2 High-flow-rate tritium sampler.

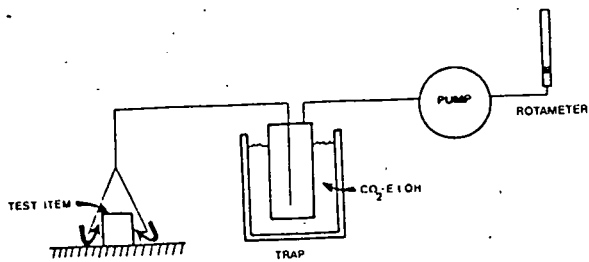


Fig. 3 HTO collection set-up.

is planned to further disassemble the display in an attempt to localize the site of T₂-HTO conversion.

Conversion of T₂ to HTO most probably occurs through radiation-induced reactions of tritium with the glass envelope or plastic encapsulating material. Based upon a study by Combs and Doda (1979), tritium gas-lighted watches could represent a source of 1 MCi of tritium, mostly in HTO form, to the troposphere annually. A 10-day residence time would lead to an increase of the present tropospheric burden by nearly 30 kCi.

The impact of these devices to background monitoring operations is obvious, and will require such operations to be conducted away from locations of their use, storage, or disposal.

CONCLUSIONS

The global environment contains measurable quantities of anthropogenic tritium which have been used as tracers for a variety of meteorological, oceanographic, and geochemical studies. The increasingly widespread distribution of tritium sources will complicate the environmental monitoring task, and require care in both sampling operations and interpretation of data.

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ERRATA

Progress Report

to

DEPARTMENT OF ENERGY

Division of Carbon Dioxide and Climate Research
Office of Health and Environmental Research

DE-AS05-76EV03944

For the Time Period
1979 July 1 - 1980 June 30

TITLE:

ATMOSPHERIC TRITIUM

DOE/EU/03944--17

Principal Investigator: Dr. H. Göte Östlund
Professor of Marine and
Atmospheric Chemistry

Co-Principal Investigator: Dr. Allen S. Mason
Research Associate Professor

APPROVED FOR RELEASE OR
PUBLICATION - O.R. OF TENT GROUP
BY SDH DATE 5-19-81

80 Oct. 22

CORRECTIONS

A. Pages to be replaced:

1. The attached pages replace Table 3 of the original report. Corrections in the range of 5-10% have been made to data from sample no. 4111 onward.
2. Table 7 is to be replaced, due to re-determination of the mass of air sampled.
3. Table 8 is to be replaced, due to re-calibration of the air mass-flowmeter and consequent changes in air quantities and CO₂ mixing ratios.

B. Annotations:

1. In Table 6, the HTO mixing ratio of sample 17 15 001, missing in the original publication, has been determined to be 77.5 ± 3.2 T-atoms/mg air. Please annotate the table accordingly.
2. In Figure 4, the value for sample 17 15 001 may be added and a small correction to the 50 T-atoms/mg isopleth made.

TABLE 3 ATMOSPHERIC HT AND HTO NEW ZEALAND 1979

Location: Baring Head Lighthouse, New Zealand
Elevation 74 m; 41 24 S 174 52 E

Explanation of heading: See Table 1. Sample run was generally 24 hours.

Sample	Date	Temp	RH	AH	Vap TU sig	HTO AT sig	HT AT sig
4079	790104	15.5	54	7.2			28.6 1.1
4080	790111	20.2	65	11.6			26.3 1.2
4081	790118	20.5	52	9.4			25.9 1.0
4082	790126	19.6	67	11.5			43.3 1.5
790104	790126			10.0	4.5 0.5	2.5 0.3	
4083	790202	19.9	57	10.0			26.3 0.9
4084	790207	19.9	52	9.1			27.0 1.1
4085	790212	17.4	64	9.7			27.6 1.0
4086	790215	17.4	64	9.6			29.8 1.2
4087	790222	14.5	65	8.2			53.8 2.2
790202	790222			9.3	5.6 0.7	2.9 0.4	
4088	790301	11.5	72	7.5			28.8 1.2
4089	790308	21.5	62	11.9			42.6 1.7
4090	790316	13.3	70	8.2			28.1 1.2
4091	790322	17.4	72	10.9			26.8 1.3
4092	790330	11.5	71	7.4			30.3 1.3
790301	790330			9.2	5.0 0.3	2.6 0.2	
4093	790406	17.6	71	10.9			31.9 1.3
4094	790409	17.8	64	9.9			29.0 1.2
4095	790417	10.8	75	7.5			25.9 1.2
4096	790423	15.9	66	9.1			27.9 1.2
4097	790430	13.4	48	5.7			28.8 1.2
790406	790430			8.6	6.1 0.4	2.9 0.2	
4098	790503	17.1	51	7.6			37.1 1.5
4099	790509	13.7	63	7.6			25.5 1.4
4100	790514	12.8	52	5.9			
4101	790523	9.2	65	5.9			26.6 1.1
790503	790523			6.7	5.3 0.4	2.0 0.1	
4102	790605	8.8	66	5.8	5.7 1.4	1.8 0.4	24.6 1.1
4103	790706	8.0	61	5.1			25.9 1.1
4104	790715	8.9	57	5.1			27.3 1.2
4105	790719	10.6	63	6.2			26.3 1.1
4106	790723	8.6	80	6.9			25.3 1.0
4107	790726	10.1	69	6.6			25.9 1.0
4108	790730	9.8	73	6.9			26.2 1.1
790706	790730			6.1	4.9 0.4	1.6 0.1	
4109	790802	11.8	74	7.9	3.1 1.3	1.5 0.5	25.6 1.1

CORRECTED

TABLE 3 NEW ZEALAND (Cont.)

Sample	Date	Temp	RH	AH	Vap TU sig	HTO AT sig	HT AT sig
4111	790906	13.5	66	7.9			
4112	790912	15.0	75	9.8			30.4 1.1
4113	790919	12.2	56	6.2			29.6 1.2
4114	790926	10.5	55	5.5			27.8 1.1
790906	790926			7.3	7.1 0.4	2.9 0.2	
4115	791001	12.5	84	9.4			30.1 1.2
4116	791004	9.7	77	7.2			29.5 1.2
4117	791008	12.5	85	9.5			29.8 1.2
4118	791011	12.5	74	8.3			29.6 1.3
4119	791018	14.7	59	7.5			29.9 1.2
4120	791026	14.4	64	8.1			28.8 1.2
791001	791026			8.3	5.8 0.3	2.6 0.1	
4121	791101	16.5	59	8.5			29.0 1.3
4122	791108	13.6	66	7.9			28.4 1.2
4123	791115	10.0	69	6.5			30.0 1.2
4124	791122	16.6	78	11.1			29.1 1.4
4125	791126	17.4	63	9.5			26.7 1.3
4126	791129	17.1	70	10.4			
791101	791129			9.0	4.4 0.5	2.2 0.3	
4127	791206	16.9	76	11.1			29.7 1.0
4128	791213	16.2	65	9.2			29.8 1.2
4129	791221	16.3	62	8.8			26.9 1.3
4130	791227	14.5	75	9.5			34.2 1.1
791206	791227			9.6	5.1 0.6	2.8 0.3	

TABLE 7 STRATOSPHERIC HTO

Location: Holloman AFB, NM vicinity
33°N 107°W

Explanation of heading: Flight No. is assigned by the launching agency; Date is the launch date; Altitude is as reported by the launching agency; T-Atoms/mg air is the mixing ratio of HTO computed from the activity and quantity of the water sample flushed from the trap, and the quantity of air sampled as measured after the flight. The error is based on the 1- σ counting error and an estimated 5% error in air quantity measurement.

<u>Flight No.</u>	<u>Date</u>	<u>Altitude,km</u>	<u>T-Atoms/mg air</u>
H80-15/H-159	800404	26.1	629 \pm 46
H80-16/H-160	800410	23.0	947 \pm 60

TABLE 8 STRATOSPHERIC CO₂ COLLECTION

Location: Ellington AFB, TX vicinity
29°N 95°W

Date: 800513

Sample No.	1	2	3	4	5	6
Altitude, ft.	30000	40000	45000	50000	55000	63000
First sieve						
H ₂ O, g	11.79	1.48	0.44	0.48	1.14	2.00
CO ₂ , g	1.40	1.84	1.54	1.84	1.66	1.46
Second sieve						
H ₂ O, g	0.00	0.05	0.01	0.05	0.18	0.14
CO ₂ , g	0.46	0.39	0.08	0.10	0.15	0.27
Total						
H ₂ O, g	11.79	1.53	0.45	0.53	1.32	2.14
CO ₂ , g	1.86	2.23	1.62	1.94	1.81	1.73
Air, liters STP*	3041	3402	2442	3157	2905	2952
CO ₂ , liters STP*	1.01	1.22	0.88	1.06	0.99	0.94
Apparent CO ₂ v/v x 10 ⁶	332	357	360	336	341	318

Note: Apparently high CO₂ mixing ratios may result from carry over of H₂O into CO₂ sample containers, as the amount was determined by weight.

*STP = 20°C, 1013 mb