



Supplement of

Stratospheric $\delta^{13}\text{CO}_2$ observed over Japan and its governing processes

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Scientific balloons with cryogenic air samplers were launched from the Sanriku Balloon Center (SBC; 39° 10' N, 141° 50' E) and Taiki Aerospace Research Field (TARF; 42° 30' N 143° 26' E), Hokkaido, during 1985–2020. The balloon observations conducted over Japan are summarized in Table S1, including sampling altitude, mole fractions of CO₂ and CH₄, $\delta^{13}\text{CO}_2$ and $\delta^{13}\text{CH}_4$, and $\langle\delta_G\rangle$ for each air sample.

Tables S1. Stratospheric CO₂ and CH₄ mole fractions, their $\delta^{13}\text{C}$ values, and GS values observed over Japan. Because CH₄ mole fractions were not measured in 1985 and 1986, they were estimated from the sampling altitude using a linear function fitted to the relationship between altitude and CH₄ mole fraction observed in 1988 and are shown in parentheses. $\delta^{13}\text{CH}_4$ values in parentheses were estimated from the CH₄ mole fractions using the average relationship between CH₄ mole fraction and $\delta^{13}\text{CH}_4$, and $\langle\delta_G\rangle$ values in parentheses were estimated from the average vertical profile (see text). The $\delta^{13}\text{CO}_2$ values listed for 1985 and 1986 are from Gamo et al. (1995), and the values for 1997 are from Aoki et al. (2003); in both cases, the values were corrected for MIE.

Date/Location	Altitude (km)	CO ₂ mole fraction ($\mu\text{mol mol}^{-1}$)	$\delta^{13}\text{CO}_2$ (‰-VPDB)	CH ₄ mole fraction (nmol mol^{-1})	$\delta^{13}\text{CH}_4$ (‰-VPDB)	$\langle\delta_G\rangle$ (per meg)
1985/09/03 SBC	19.5	343.84	-7.81	(1500)	(-45.6)	(-9.9)
	21.1	341.30	-7.66	(1407)	(-44.7)	(-15.5)
	23.0	340.55	-7.60	(1293)	(-43.7)	(-22.4)
	24.8	340.41	-7.59	(1171)	(-42.4)	(-29.0)
1986/05/28 SBC	13.5	346.74				(11.9)
	18.1	343.06	-7.78	(1587)	(-46.3)	(-4.7)
	21.4	341.08	-7.56	(1389)	(-44.6)	(-16.6)
	24.1	341.44	-7.69	(1227)	(-43.0)	(-26.4)
	27.4	342.12	-7.73	(1027)	(-40.7)	(-38.6)
	30.3	342.40	-7.75	(853)	(-38.3)	(-49.1)
1988/05/21 SBC	33.1	341.58	-7.71	(688)	(-35.5)	(-59.1)
	19.3	348.38	-7.81	1543	(-45.9)	(-9.0)
	21.1	346.32	-7.73	1408	(-44.7)	(-15.5)
	23.0	345.31	-7.73	1307	(-43.8)	(-22.6)
	25.1	343.87	-7.68	1101	(-41.6)	(-30.2)
	27.2	344.01	-7.69	961	(-39.8)	(-37.7)
1989/09/01 SBC	28.8	344.63	-7.67	1010	(-40.5)	(-43.7)
	31.1	344.45	-7.71	825	(-37.8)	(-51.8)
	18.8	350.32	-7.90	1601	(-46.4)	(-7.4)
	20.0	348.05	-7.82	1427	(-44.9)	(-11.5)
	21.2	346.94	-7.77	1298	(-43.7)	(-16.1)
	22.5	345.45	-7.74	1135	(-42.0)	(-20.6)
	23.9	345.57	-7.77	1135	(-42.0)	(-25.9)
	25.6	345.51	-7.75	1077	(-41.3)	(-32.1)
1990/06/04 SBC	27.0	345.52	-7.75	1077	(-41.3)	(-37.0)
	28.6	346.02	-7.78	1014	(-40.5)	(-43.0)
	30.3	345.68	-7.78	936	(-39.5)	(-49.1)
	30.4	345.66				
	16.1	353.94		1662	(-46.9)	(2.4)
	16.2	353.61		1651	(-46.8)	(2.1)
	18.1	351.63	-7.85	1587	(-46.3)	(-4.8)

	20.8			1104	(-41.6)	(-14.6)
	24.5	347.28	-7.76	1187	(-42.5)	(-28.1)
	26.5	347.56	-7.77	1171	(-42.4)	(-35.3)
	29.0	346.70	-7.78	1029	(-40.7)	(-44.4)
	31.0	347.14	-7.79	873	(-38.6)	(-51.7)
	33.4	346.99	-7.81	630	(-34.4)	(-60.4)
	34.7	346.83	-7.87	543	(-32.4)	(-65.1)
1991/08/29 SBC	13.3	355.20		1729	(-47.4)	(12.6)
	14.0	355.16	-7.80	1731	(-47.4)	(10.1)
	18.3			1743	(-47.5)	(-5.6)
	20.2	351.70	-7.93	1460	(-45.2)	(-12.5)
	22.1			1307	(-43.8)	(-19.4)
	24.6			1156	(-42.2)	(-28.4)
	26.6	347.85		1091	(-41.5)	(-35.7)
	28.7	348.96	-7.85	946	(-39.6)	(-43.3)
	30.7	349.13	-7.87	940	(-39.5)	(-50.6)
	33.1	348.79	-7.92	706	(-35.8)	(-59.3)
	35.2	349.31	-7.92	753	(-36.7)	(-66.9)
1994/08/31 SBC	14.0	356.92	-7.72	1813	-46.9	(10.1)
	16.4	358.08	-7.88	1794	(-47.8)	(1.3)
	18.2	357.66	-7.92	1625	-46.2	(-5.2)
	20.4	356.56	-7.88	1535	-45.9	(-13.2)
	22.3	354.52	-7.83	1253	-43.5	(-20.1)
	24.7	353.87	-7.75	1117	-41.8	(-28.8)
	26.8	353.73	-7.89	1060	-41.2	(-36.4)
	29.2	354.42	-7.86	1077	-41.2	(-45.1)
	31.1	354.77	-7.93	954	-39.4	(-52.0)
	32.8	354.24	-7.93	941	-39.2	(-58.2)
	34.7	354.24	-7.95	894	-38.0	(-65.1)
1995/06/08 SBC	13.6	363.38	-8.21	1751	-46.2	-8.1
	16.6	361.09	-8.17	1669	(-46.9)	-6.3
	18.0	356.09	-7.97	1544	-45.5	12.8
	20.3	355.40	-7.94	1310	-44.0	-8.3
	22.3	355.81	-7.96	1371	-44.3	-2.0
	24.5	355.17	-7.95	992	-41.5	-24.8
	26.7	354.80	-7.94	981	-40.4	-43.6
	28.8	355.33	-7.94	846	-38.4	-28.1
	30.6	354.96	-7.88	776	-37.9	-27.3
	32.4	354.75	-7.98	749	-36.0	-38.9
	33.8	355.03	-7.97	707	-35.1	-50.3
1997/05/30 SBC	14.5	362.70	-8.08	1729	-46.2	(8.2)
	16.3	361.80	-8.05	1710	-46.0	(1.7)
	18.4	359.56	-7.92	1509	-45.6	(-5.9)
	19.9	358.21	-7.90	1344	-44.2	(-11.4)
	21.8	355.68	-7.95	982	-41.6	(-18.3)
	23.7	356.36	-7.96	1286	-43.0	(-25.2)
	25.7	354.99		1100	-40.3	(-32.4)
	27.3	356.05	-7.94	1042	-42.5	(-38.2)
	29.6	356.04	-7.98	881	-40.5	(-46.6)
	31.8	355.97	-8.01	643	(-34.6)	(-54.6)
	33.8	357.38	-7.97	581	-38.1	(-61.8)
1998/09/03 SBC	14.9	365.54	-8.12	1800	-46.6	(6.8)
	16.5	366.37	-8.20	1707	-46.5	(1.0)
	18.0	364.30	-8.15	1646	-46.4	(-4.5)
	19.3	362.61	-8.08	1570	-45.8	(-9.2)
	20.6	360.85	-8.05	1411	-44.7	(-13.9)
	21.9	359.19	-8.00	1166	-43.3	(-18.6)
	23.4	359.19	-7.98	1104	-42.2	(-24.1)
	24.9	358.92	-8.04	1061	-41.7	(-29.5)

	26.4	359.27	-7.98	1096	-41.8	(-35.0)
	27.3	358.99	-7.99	1066	-41.5	(-38.2)
	29.5	359.71	-7.95	1042	-41.3	(-46.2)
1999/05/31 SBC	14.7	356.19		1697	-47.0	(7.5)
	17.0	355.37	-8.09	1681	-46.8	(-.8)
	18.7	362.66	-8.07	1422	-45.3	-29.8
	20.8	363.16	-8.03	1486	-45.4	-27.6
	23.1	359.88		1400	(-44.7)	-17.5
	24.1		-7.92	1349	-44.6	(-26.6)
	25.6	359.46	-8.06	1188	-43.4	-35.2
	27.3	358.87	-7.98	1052	-41.8	-77.9
	29.3	357.08	-8.03	782	-38.3	(-45.5)
	31.8					(-54.6)
	34.5	359.67	-8.14	560	-35.1	-91.9
2000/08/28 SBC	15.0	366.93	-7.91	1856	(-48.3)	3.9
	17.1	367.16	-8.08	1807	(-47.9)	-4.0
	18.7	366.29	-8.04	1614	(-46.5)	14.0
	20.9	363.00	-8.00	1307	(-43.8)	-7.6
	22.7	363.36	-8.04	1242	(-43.1)	-14.9
	24.4	361.02	-7.97	1149	(-42.1)	(-27.7)
	25.8	362.29	-8.05	1081	(-41.3)	-62.8
	27.7	362.35	-8.03	1053	(-41.0)	-49.0
	29.3	362.82	-8.07	1042	(-40.9)	-60.3
	31.3		-8.02	958	(-39.8)	(-52.8)
2001/05/30 SBC	14.9	369.03	-8.12	1676	-46.4	7.8
	16.8	367.20	-8.16	1667	-46.3	-11.2
	18.8	367.52	-8.08	1613	-45.9	-28.2
	20.8	363.88	-8.08	1190	-42.2	-48.3
	23.1	364.04	-8.04	1241	-42.6	-33.0
	23.8	364.10	-8.02	1235	-42.3	-9.4
	25.7	364.51	-8.02	1211	-42.2	-7.0
	27.6	364.83	-7.98	1173	-41.8	-33.1
	29.4	364.37	-8.09	986	-39.9	-35.5
	31.7		-8.13	720	(-36.1)	(-54.2)
2002/09/04 SBC	33.8	364.46	-8.24	699	-34.4	-32.1
	15.9	370.37	-8.15	1774	-46.8	9.8
	16.4	370.55	-8.19	1795	-46.9	12.5
	18.7	371.21	-8.25	1672	-46.2	-9.1
	20.6	366.92	-8.03	1389	-44.2	-17.9
	22.9		-8.10	1292	-43.4	(-22.3)
	24.0	364.72	-8.13	1320	-43.4	-24.0
	25.4		-8.12	1216	-42.2	(-31.3)
	27.7	363.56	-8.13	940	-38.3	-31.1
	30.3		-8.13	764	-34.7	(-49.1)
2004/09/06 SBC	31.8	364.10	-8.17	706	-33.7	-42.2
	34.0	365.05	-8.08	589	-30.5	-73.8
	14.7	375.78	-8.23	1786	(-47.8)	0.9
	16.5	376.67	-8.31	1772	(-47.7)	7.4
	18.7	374.65	-8.26	1637	(-46.7)	-4.1
	21.0	371.42	-8.24	1432	(-45.0)	-18.6
	23.0	369.63	-8.19	1217	(-42.9)	-33.3
	24.8	370.11	-8.21	1216	(-42.9)	-36.0
	27.0	368.98	-8.17	1064	(-41.1)	-53.1
	29.5	368.38	-8.23	775	(-37.0)	-99.0
2006/06/03 SBC	31.5	368.48	-8.21	643	(-34.6)	-43.5
	32.9	368.48	-8.21	643	(-34.6)	-34.9
	35.7	368.58	-8.27	564	(-32.9)	-74.2
	14.9		-8.23	1606	(-46.4)	(6.8)
	16.6		-8.25	1588	(-46.3)	(0.6)

	18.8		-8.21	1570	(-46.1)	(-7.4)
	20.8	374.06	-8.21	1346	(-44.2)	-15.7
	23.0	373.26	-8.17	1319	(-43.9)	-22.4
	24.8	373.64		1229	(-43.0)	-24.5
	27.2	373.34		1093	(-41.5)	-40.7
	31.0	373.34		897	(-38.9)	-48.9
	32.9	372.37	-8.23	725	(-36.2)	-59.9
	34.4	371.95	-8.22	667	(-35.1)	-36.1
2007/06/04 SBC	14.5	382.55	-8.36	1744	-46.8	-1.8
	17.0	380.76	-8.23	1699	-46.5	5.4
	18.5	377.92	-8.23	1521	-45.3	-10.8
	21.0	375.21	-8.25	1201	-42.8	-23.7
	23.0	375.56	-8.16	1292	-43.2	-10.9
	26.0	375.45	-8.20	1224	-42.4	-16.8
	28.3	374.68	-8.30	978	-40.0	-35.6
	29.9	374.84	-8.21	932	-39.1	-26.4
	31.3	374.35	-8.23	748	-36.1	-44.1
2010/08/22 TARF	32.8	374.33	-8.25	663	-34.5	-48.0
	15.2	387.89	-8.32	1818	-47.1	17.8
	16.8	388.43	-8.31	1744	-46.7	11.7
	18.7	385.74	-8.27	1619	-46.0	2.4
	20.8			1378	-43.9	(-14.7)
	22.9	381.55	-8.25	1275	-43.1	-2.2
	25.1		-8.24	1046	-41.1	(-30.4)
	28.5	381.15	-8.24	1091	-41.2	-33.8
	29.2	381.02	-8.26	1054	-40.6	-26.5
	31.0		-8.28	1060	-40.7	(-51.6)
2015/08/06 TARF	33.2	380.56	-8.26	826	-37.2	-28.4
	34.3	380.60	-8.32	694	-35.0	-61.0
	14.9	400.30	-8.47	1914	-47.2	(6.8)
	16.7	394.72	-8.54	1830	-47.0	(0.2)
	18.8	391.80	-8.42	1659	-45.9	(-7.4)
	20.9			1348	-43.4	(-15.0)
	22.8	390.78	-8.35	1218	-42.0	(-21.9)
	24.7			1175	-41.7	(-28.8)
	26.8	390.59		1161	-41.3	(-36.4)
	28.8	389.55		1040	-40.1	(-43.7)
2020/07/25 TARF	30.9			908	-33.8	(-51.3)
	33.0	390.16		790	-36.5	(-58.9)
	34.8	390.93	-8.44	739	-35.2	(-65.5)
	14.8	409.92	-8.56	1802	(-47.9)	(7.2)
	16.9	404.48	-8.51	1816	(-48.0)	(-0.3)
	18.6	407.73	-8.49	1658	(-46.8)	(-6.7)
	20.9	405.10		1301	(-43.7)	(-14.8)
	22.7	402.91	-8.42	1290	(-43.6)	(-21.5)
	24.8	401.93	-8.51	1083	(-41.4)	(-29.2)
	26.9	400.02	-8.52	1121	(-41.8)	(-36.6)
	28.8			1047	(-40.9)	(-43.8)
	30.9			886	(-38.8)	(-51.4)
	33.3	400.23		826	(-37.9)	(-59.9)
	35.4	400.21	-8.63	783	(-37.2)	(-67.5)