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Vertical distributions of N_2O isotopocules in the equatorial stratosphere

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Surface emission and atmospheric trend of N_2O isotopocules in the chemical transport model

Global mean atmospheric N_2O isotopic values and estimated global total emissions in model are shown in Figure S1. The emissions of four N_2O isotopocules ($^{14}N^{14}N^{16}O$, $^{14}N^{15}N^{16}O$, $^{15}N^{14}N^{16}O$, and $^{14}N^{14}N^{18}O$) were estimated by a method used in Ishijima et al. (2015), but the photolytic fractionation was not tuned in this study. Thus estimated surface emissions were used to simulate atmospheric N_2O isotopocules, which were used for stratospheric analysis of this study. The isotopocule ratios of N_2O emitted from surface sources in model were temporally constant ($\delta^{15}N^{bulk}$ = -8.4 ‰, $\delta^{18}O$ = 32.4 ‰, SP = 14.0 ‰). Although the model was optimized against relatively old measurement data mainly for 1990s, simulated atmospheric values in recent years at the surface were very reasonable (Table S1), considering necessary order of precision for analysis of the large vertical profiles in the stratosphere in this study.

Supplementary Tables and Figures

Table S1. Annual mean mixing ratio, $\delta^{15}N^{bulk}$, $\delta^{18}O$, and SP of atmospheric N_2O for 2010 observed and simulated at Hateruma station.

Method	N_2O mixing ratio (nmol mol^{-1})	$\delta^{15}N^{bulk}$ (‰)	$\delta^{18}O$ (‰)	SP (‰)	Reference
Observation	323.3	6.6	44.2	18.3	This study (top-down by the ACTM)
Model	324.0	6.3	44.5	18.7	Toyoda et al. (2013)

Table S2: Sampling location, date, altitude range, and tropopause height of previous observations and this study plotted in Figs. 2, 3, and 5.

Location	Latitude/ Longitude	Sampling Date	Altitude (km)	Number of samples	Tropopause height (km)	Reference
Sanriku Balloon Center, Japan (SBC)	39 °N/142 °E	4 June 1990	16.1–34.7	9	13.6	Toyoda et al. (2004)
		3 September 1998	14.9–29.5	11	15.8	Yoshida and Toyoda (2000); Toyoda et al. (2004)
		31 May 1999	14.7–34.5	11	12.1	Toyoda et al. (2001)
		28 August 2000	15.0–31.3	10	15.0	Toyoda et al. (2004)
		30 May 2001	14.9–33.8	11	13.8	Toyoda et al. (2004)
Kiruna, Sweden (ESR)	68 °N/20 °E	22 February 1997	10.2–25.6	10	10.2	Toyoda et al. (2004)
Syowa station, Antarctica (SYO)	69 °S/40 °E	3 January 1998	10.4–29.8	9	9.0	Toyoda et al. (2004)
Hyderabad, India (HDB)	18 °N/79 °E	26 March 1987	17–26	5	NA	Kaiser et al. (2006)
		29 Apr 1999	10–28	10	NA	Röckmann et al. (2001)
Eastern equatorial Pacific (EQP)	0 °N/105–115 °W	4–8 February 2012	20–29	4	17.0–18.2[a]	This work
Biak, Indonesia (BIK)	1 °S/136 °E	22–28 February 2015	17.2–27.4	6	17.4–19.3[a]	This work

[a] Cold point tropopause. NA: not available.

Table S3: Raw data of N_2O mixing ratios and isotopocule ratios, and values normalized with respect to tropospheric values that were accounted for secular trends.

Altitude (km)	$[N_2O]$ (nmol mol^{-1}) [a]	$10^3\delta^{15}N^{bulk}$	$10^3\delta^{18}O$	$10^3\delta^{15}N^{\alpha}$	$10^3\delta^{15}N^{\beta}$	$-\ln\{[N_2O]/[N_2O]_{trp}\}$ [b]	$10^3\ln[(1+\delta^{15}N^{bulk})/\{1+\delta^{15}N^{bulk}{}_{trp}\}]$ [b]	$10^3\ln[(1+\delta^{18}O)/\{1+\delta^{18}O_{trp}\}]$ [b]	$10^3\ln[(1+\delta^{15}N^{\alpha})/\{1+\delta^{15}N^{\alpha}{}_{trp}\}]$ [b]	$10^3\ln[(1+\delta^{15}N^{\beta})/\{1+\delta^{15}N^{\beta}{}_{trp}\}]$ [b]	$[N_2O](t_0)$ (nmol mol^{-1}) [c]	$10^3\delta^{15}N^{bulk}(t_0)$ [c]	$10^3\delta^{18}O(t_0)$ [c]	$10^3\delta^{15}N^{\alpha}(t_0)$ [c]	$10^3\delta^{15}N^{\beta}(t_0)$ [c]	$-\ln\{[N_2O]/[N_2O](t_0)\}$	$10^3\ln[(1+\delta^{15}N^{bulk})/\{1+\delta^{15}N^{bulk}(t_0)\}]$	$10^3\ln[(1+\delta^{18}O)/\{1+\delta^{18}O(t_0)\}]$	$10^3\ln[(1+\delta^{15}N^{\alpha})/\{1+\delta^{15}N^{\alpha}(t_0)\}]$	$10^3\ln[(1+\delta^{15}N^{\beta})/\{1+\delta^{15}N^{\beta}(t_0)\}]$
Date of observation	1990/6/4	1990.42																		
Location	Sanriku Balloon Center																			
13.8 [d]	310.5	7.08	43.91	16.76	-2.61															
16.1	300.9	8.16	45.81	16.89	-0.56	0.0313	1.08	1.81	0.13	2.05	307.9	7.11	43.89	16.84	-2.61	0.0231	1.04	1.84	0.05	2.05
18.1	282.9	9.25	46.97	19.24	-0.75	0.0930	2.15	2.93	2.43	1.87	306.3	7.14	43.87	16.88	-2.61	0.0794	2.09	2.96	2.31	1.87
20.8	178.4	17.16	53.37	28.85	5.47	0.5541	9.96	9.02	11.82	8.06	305.5	7.16	43.85	16.94	-2.61	0.5378	9.88	9.07	11.64	8.07
24.6	175.1	20.29	55.58	36.67	3.90	0.5727	13.03	11.11	19.39	6.51	305.0	7.20	43.83	17.02	-2.62	0.5551	12.91	11.20	19.14	6.52
26.6	163.7	22.47	57.44	37.36	7.58	0.6400	15.17	12.87	20.06	10.16	305.1	7.20	43.83	17.01	-2.62	0.6225	15.05	12.95	19.81	10.17
29.1	117.0	33.23	67.63	53.88	12.58	0.9759	25.64	22.47	35.86	15.12	305.0	7.21	43.82	17.04	-2.62	0.9582	25.51	22.56	35.58	15.13
31.1	71.8	46.03	78.41	72.04	20.03	1.4642	37.95	32.51	52.94	22.44	305.0	7.21	43.82	17.03	-2.62	1.4466	37.82	32.60	52.67	22.46
33.5	28.7	74.22	101.92	111.64	36.79	2.3812	64.54	54.08	89.21	38.74	305.0	7.21	43.82	17.04	-2.62	2.3635	64.40	54.17	88.94	38.75
34.7	18.7	86.48	114.44	130.62	42.34	2.8096	75.89	65.37	106.14	44.08	305.0	7.22	43.82	17.05	-2.62	2.7918	75.75	65.46	105.86	44.09
Date of observation	1998/9/3	1998.67																		
Location	Sanriku Balloon Center																			
15.8	314.6	6.89	44.04	16.37	-2.59															
16.4	300.9	8.38	46.14	17.55	-0.78	0.0446	1.48	2.00	1.16	1.81	314.1	6.91	44.03	16.41	-2.59	0.0431	1.46	2.02	1.12	1.81
18.0	287.7	8.90	46.35	18.37	-0.58	0.0894	1.99	2.20	1.97	2.01	313.1	6.94	44.01	16.48	-2.60	0.0845	1.94	2.24	1.87	2.02
19.3	271.9	9.31	47.11	18.89	-0.27	0.1459	2.40	2.93	2.48	2.32	312.4	6.96	43.99	16.52	-2.60	0.1387	2.33	2.98	2.33	2.33
20.6	234.3	11.88	49.27	22.92	0.84	0.2948	4.94	4.99	6.43	3.43	311.6	6.99	43.97	16.58	-2.60	0.2851	4.84	5.06	6.22	3.44
21.9	176.9	16.58	53.86	30.89	2.27	0.5758	9.58	9.36	14.19	4.86	311.6	7.02	43.95	16.64	-2.60	0.5661	9.45	9.44	13.92	4.87
23.4	154.3	20.70	56.44	35.26	6.15	0.7125	13.63	11.80	18.42	8.72	311.6	7.02	43.95	16.64	-2.60	0.7028	13.50	11.89	18.14	8.73
24.9	142.3	22.43	57.10	37.82	7.05	0.7934	15.32	12.43	20.89	9.62	311.4	7.02	43.95	16.65	-2.60	0.7833	15.19	12.52	20.61	9.63
26.4	144.4	24.69	59.01	40.12	9.25	0.7788	17.52	14.23	23.10	11.80	311.6	7.02	43.95	16.64	-2.60	0.7692	17.39	14.32	22.83	11.81
27.3	132.0	28.12	63.10	45.46	10.77	0.8686	20.86	18.09	28.23	13.31	311.5	7.02	43.95	16.65	-2.60	0.8586	20.73	18.18	27.95	13.32
29.5	117.8	33.12	67.61	50.67	15.57	0.9824	25.72	22.32	33.19	18.05	311.7	7.01	43.96	16.63	-2.60	0.9730	25.60	22.41	32.94	18.06
Date of observation	1999/5/31	1999.41																		
Location	Sanriku Balloon Center																			
12.1	315.0	6.87	44.06	16.33	-2.59															
14.7	291.6	8.13	45.81	17.16	-0.90	0.0772	1.25	1.68	0.81	1.69	313.8	6.92	44.02	16.44	-2.59	0.0735	1.20	1.71	0.71	1.69
17.0	282.5	8.31	46.43	18.00	-1.39	0.1089	1.42	2.27	1.64	1.21	313.6	6.93	44.02	16.45	-2.59	0.1043	1.37	2.31	1.52	1.21
18.7	234.6	11.85	47.92	22.59	1.12	0.2947	4.94	3.69	6.14	3.71	312.2	6.96	43.99	16.53	-2.60	0.2857	4.84	3.75	5.94	3.72
20.8	248.7	10.81	48.72	21.79	-0.16	0.2363	3.91	4.45	5.36	2.43	312.5	6.96	44.00	16.51	-2.60	0.2282	3.82	4.51	5.18	2.44
23.0	234.3	12.51	49.35	23.55	1.47	0.2960	5.59	5.06	7.08	4.07	311.6	7.00	43.97	16.60	-2.60	0.2852	5.46	5.14	6.82	4.08
24.1	182.5	15.10	50.77	27.98	2.23	0.5458	8.14	6.41	11.40	4.82	311.6	7.00	43.97	16.59	-2.60	0.5350	8.02	6.50	11.14	4.83
25.6	168.3	19.59	55.60	34.57	4.61	0.6268	12.55	11.00	17.79	7.19	311.7	7.01	43.96	16.63	-2.60	0.6162	12.41	11.09	17.49	7.20
27.3	122.5	28.62	62.92	48.80	8.44	0.9444	21.37	17.91	31.45	10.99	311.4	7.03	43.95	16.65	-2.60	0.9330	21.22	18.01	31.13	11.01
29.3	53.3	53.34	86.68	86.00	20.68	1.7766	45.12	40.01	66.31	23.07	311.6	7.02	43.95	16.64	-2.60	1.7656	44.97	40.11	66.00	23.08
31.8	24.7	73.76	104.40	112.60	34.92	2.5457	64.32	56.18	90.50	36.92	311.6	7.02	43.95	16.64	-2.60	2.5348	64.17	56.28	90.19	36.93
34.5	14.5	92.77	119.01	143.63	41.92	3.0784	81.87	69.33	118.00	43.66	311.5	7.02	43.95	16.64	-2.60	3.0674	81.72	69.43	117.70	43.67
Date of observation	2000/8/28	2000.66																		
Location	Sanriku Balloon Center																			
15.0	316.5	6.84	44.08	16.27	-2.59															
17.1	314.1	7.32	44.77	16.81	-2.18	0.0078	0.47	0.66	0.54	0.41	314.3	6.90	44.03	16.40	-2.59	0.0007	0.41	0.70	0.41	0.41
18.7	281.3	8.63	46.47	18.56	-1.30	0.1180	1.77	2.29	2.25	1.29	314.0	6.92	44.02	16.43	-2.59	0.1098	1.70	2.34	2.10	1.29
20.9	208.7	13.50	49.61	24.95	2.04	0.4166	6.59	5.28	8.50	4.63	312.2	6.96	43.99	16.52	-2.60	0.4029	6.47	5.37	8.25	4.64
22.7	191.8	14.39	51.66	25.78	3.01	0.5010	7.47	7.24	9.31	5.59	312.4	6.96	43.99	16.52	-2.60	0.4877	7.36	7.32	9.07	5.60
24.4	164.2	18.82	54.40	34.35	3.30	0.6564	11.83	9.84	17.63	5.88	312.0	6.97	43.99	16.54	-2.60	0.6421	11.70	9.93	17.37	5.89
25.8	143.4	23.25	58.46	39.25	7.25	0.7918	16.17	13.69	22.36	9.82	311.8	6.98	43.98	16.55	-2.60	0.7768	16.03	13.78	22.08	9.83
27.7	129.7	27.98	63.05	44.03	11.92	0.8922	20.77	18.01	26.95	14.44	311.8	6.98	43.98	16.55	-2.60	0.8772	20.64	18.09	26.67	14.45
29.3	116.7	32.57	66.95	51.39	13.75	0.9979	25.23	21.68	33.97	16.25	312.0	6.97	43.99	16.54	-2.60	0.9835	25.11	21.76	33.71	16.26
31.3	86.6	42.29	75.73	66.13	18.46	1.2962	34.61	29.87	47.90	20.88	311.9	6.97	43.98	16.55	-2.60	1.2813	34.47	29.96	47.63	20.89

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Altitude (km)	$[N_2O]$ (nmol mol^{-1}) [a]	$10^3\delta^{15}N^{bulk}$	$10^3\delta^{18}O$	$10^3\delta^{15}N^{\alpha}$	$10^3\delta^{15}N^{\beta}$	$-\ln\{[N_2O]/[N_2O]_{trp}\}$ [b]	$10^3\ln[(1+\delta^{15}N^{bulk})/\{1+\delta^{15}N^{bulk}{}_{trp}\}]$ [b]	$10^3\ln[(1+\delta^{18}O)/\{1+\delta^{18}O_{trp}\}]$ [b]	$10^3\ln[(1+\delta^{15}N^{\alpha})/\{1+\delta^{15}N^{\alpha}{}_{trp}\}]$ [b]	$10^3\ln[(1+\delta^{15}N^{\beta})/\{1+\delta^{15}N^{\beta}{}_{trp}\}]$ [b]	$[N_2O](t_0)$ (nmol mol^{-1}) [c]	$10^3\delta^{15}N^{bulk}(t_0)$ [c]	$10^3\delta^{18}O(t_0)$ [c]	$10^3\delta^{15}N^{\alpha}(t_0)$ [c]	$10^3\delta^{15}N^{\beta}(t_0)$ [c]	$-\ln\{[N_2O]/[N_2O](t_0)\}$	$10^3\ln[(1+\delta^{15}N^{bulk})/\{1+\delta^{15}N^{bulk}(t_0)\}]$	$10^3\ln[(1+\delta^{18}O)/\{1+\delta^{18}O(t_0)\}]$	$10^3\ln[(1+\delta^{15}N^{\alpha})/\{1+\delta^{15}N^{\alpha}(t_0)\}]$	$10^3\ln[(1+\delta^{15}N^{\beta})/\{1+\delta^{15}N^{\beta}(t_0)\}]$
Date of observation	2001/5/30	2001.41																		
Location	Sanriku Balloon Center																			
13.8	317.1	6.82	44.09	16.23	-2.59															
15.0	299.5	8.06	45.87	16.85	-0.74	0.0570	1.22	1.70	0.61	1.85	314.9	6.88	44.05	16.34	-2.59	0.0500	1.17	1.73	0.50	1.86
17.1	291.0	8.36	45.47	17.51	-0.79	0.0858	1.52	1.32	1.26	1.80	314.6	6.89	44.04	16.37	-2.59	0.0780	1.46	1.36	1.12	1.80
18.9	279.3	8.94	45.93	18.74	-0.86	0.1268	2.09	1.76	2.46	1.72	314.4	6.90	44.04	16.39	-2.59	0.1183	2.02	1.81	2.30	1.73
20.8	173.4	17.17	53.83	32.16	2.17	0.6035	10.22	9.29	15.55	4.76	312.5	6.95	44.00	16.51	-2.60	0.5891	10.09	9.38	15.28	4.77
23.4	180.8	17.55	53.18	30.38	4.72	0.5617	10.60	8.67	13.83	7.30	312.7	6.95	44.00	16.50	-2.60	0.5477	10.47	8.76	13.57	7.31
23.8	179.5	18.36	54.18	32.90	3.81	0.5689	11.39	9.62	16.26	6.39	312.7	6.95	44.00	16.50	-2.60	0.5550	11.26	9.70	16.00	6.40
25.8	169.2	20.63	55.92	35.93	5.32	0.6280	13.61	11.26	19.20	7.89	312.9	6.95	44.00	16.49	-2.60	0.6147	13.49	11.34	18.95	7.90
27.6	153.8	23.47	58.64	38.67	8.27	0.7234	16.40	13.84	21.84	10.82	313.0	6.94	44.01	16.48	-2.60	0.7106	16.28	13.92	21.60	10.83
29.3	106.2	29.81	63.55	47.20	12.42	1.0938	22.57	18.47	30.02	14.93	312.7	6.95	44.00	16.50	-2.60	1.0799	22.45	18.55	29.76	14.94
31.9	49.8	48.59	80.63	73.94	23.24	1.8511	40.65	34.40	55.23	25.57	312.6	6.95	44.00	16.50	-2.60	1.8370	40.52	34.48	54.96	25.58
34.0	38.0	64.21	92.45	97.50	30.92	2.1215	55.43	45.28	76.93	33.04	312.6	6.95	44.00	16.50	-2.60	2.1073	55.31	45.36	76.67	33.05
Date of observation	1997/2/22	1997.14																		
Location	Esrange, Kiruna																			
10.18	313.8	6.92	44.02	16.44	-2.59															
10.21	299.3	8.24	44.95	18.70	-2.22	0.0472	1.30	0.89	2.22	0.37	311.7	7.02	43.96	16.63	-2.60	0.0403	1.21	0.95	2.03	0.38
13.0	272.0	9.32	47.27	17.43	1.21	0.1431	2.38	3.11	0.97	3.81	311.7	7.01	43.96	16.61	-2.60	0.1363	2.29	3.17	0.80	3.81
14.1	258.5	9.88	47.60	21.87	-2.12	0.1940	2.93	3.42	5.33	0.48	311.7	7.01	43.96	16.62	-2.60	0.1872	2.85	3.48	5.16	0.49
15.9	197.0	14.57	51.55	27.57	1.57	0.4655	7.57	7.18	10.89	4.17	310.9	7.05	43.94	16.69	-2.60	0.4562	7.44	7.26	10.64	4.18
16.4	174.1	17.77	52.91	31.79	3.74	0.5893	10.71	8.48	14.99	6.33	310.9	7.06	43.93	16.72	-2.61	0.5799	10.58	8.57	14.72	6.34
17.2	170.6	18.36	53.57	34.12	2.60	0.6093	11.30	9.11	17.25	5.19	310.9	7.06	43.93	16.72	-2.61	0.5999	11.16	9.19	16.97	5.21
18.2	163.8	19.32	55.88	34.42	4.21	0.6499	12.23	11.30	17.54	6.80	310.9	7.06	43.93	16.72	-2.61	0.6406	12.10	11.39	17.26	6.81
20.0	123.3	26.06	60.64	44.11	8.00	0.9344	18.82	15.80	26.86	10.57	309.8	7.09	43.91	16.78	-2.61	0.9214	18.66	15.91	26.52	10.58
21.8	75.0	37.32	71.41	62.20	12.44	1.4317	29.74	25.90	44.04	14.96	310.6	7.07	43.91	16.76	-2.61	1.4215	29.59	26.00	43.72	14.98
23.5	24.4	61.51	92.30	96.50	26.53	2.5528	52.80	45.20	75.82	28.78	310.2	7.08	43.91	16.77	-2.61	2.5414	52.64	45.31	75.49	28.79
25.6	8.8	82.25	108.78	115.08	49.42	3.5702	72.14	60.18	92.62	50.84	310.0	7.08	43.91	16.78	-2.61	3.5581	71.98	60.29	92.29	50.85
Date of observation	1998/1/3	1998.01																		
Location	Syowa station, Antarctica																			
9.0	313.0	7.10	44.53	16.26	-2.59															
10.4	296.2	8.10	45.59	17.69	-1.50	0.0551	0.98	1.01	1.41	1.10	310.5	7.21	44.61	16.35	-1.93	0.0470	0.88	0.94	1.32	0.43
12.4	264.8	10.01	47.36	17.80	2.22	0.1672	2.88	2.70	1.52	4.81	310.2	7.22	44.62	16.36	-1.91	0.1583	2.76	2.62	1.42	4.13
14.3	201.6	15.16	51.69	26.54	3.78	0.4398	7.97	6.83	10.07	6.37	310.2	7.27	44.65	16.40	-1.86	0.4309	7.81	6.72	9.93	5.64
16.7	201.0	14.79	51.72	25.73	3.85	0.4428	7.60	6.86	9.28	6.44	309.9	7.27	44.65	16.40	-1.86	0.4329	7.44	6.75	9.14	5.71
18.5	214.6	13.49	49.94	24.99	1.98	0.3773	6.32	5.17	8.55	4.58	309.9	7.26	44.64	16.39	-1.87	0.3675	6.16	5.06	8.42	3.85
20.4	181.7	17.01	53.24	28.53	5.49	0.5438	9.79	8.31	12.00	8.07	309.7	7.28	44.66	16.41	-1.85	0.5334	9.61	8.19	11.85	7.33
22.7	125.6	24.23	58.92	39.41	9.06	0.9130	16.86	13.68	22.52	11.61	309.4	7.30	44.67	16.42	-1.83	0.9014	16.67	13.55	22.36	10.84
26.9	95.8	37.06	71.62	57.90	16.23	1.1839	29.32	25.60	40.16	18.69	309.7	7.28	44.66	16.41	-1.84	1.1733	29.14	25.48	40.01	17.94
29.8	63.6	54.24	85.94	82.05	26.44	1.5935	45.75	38.88	62.73	28.70	309.4	7.30	44.67	16.42	-1.83	1.5819	45.55	38.74	62.56	27.93
Date of observation	2012/2/4	2012/2/5	2012/2/7	2012/2/8	(individual sampling date)															
	2012/2/6	2012.10	(average)																	
Location	Eastern Equatorial Pacific																			
17.4	325.0	6.58	44.26	15.72	-2.56															
20.0	307.0	7.37	45.08	16.75	-2.01	0.0572	0.79	0.79	1.01	0.56	323.5	6.61	44.24	15.79	-2.57	0.0526	0.76	0.81	0.94	0.56
24.0	286.8	9.05	46.33	19.16	-1.06	0.1250	2.45	1.98	3.38	1.51	322.8	6.64	44.22	15.85	-2.57	0.1181	2.39	2.02	3.26	1.51
27.0	248.5	13.38	50.04	25.34	1.43	0.2685	6.73	5.52	9.42	3.99	322.6	6.65	44.21	15.86	-2.57	0.2610	6.67	5.56	9.28	4.00
29.0	237.2	15.43	51.69	28.26	2.60	0.3150	8.75	7.09	12.27	5.16	321.9	6.67	44.20	15.91	-2.57	0.3055	8.66	7.15	12.09	5.17
Date of observation	2015/2/22	2015/2/24	2015/2/26	2015/2/28	(individual sampling date)															
	2015/2/25	2015.15	(average)																	
Location	Biak island, Indonesia																			
17.0	328.0	6.51	44.31	15.57	-2.56															
17.2	327.4	6.51	44.32	15.58	-2.58	0.0018	0.00	0.01	0.01	-0.02	326.8	6.54	44.29	15.63	-2.56	-0.0019	-0.03	0.03	-0.05	-0.02
18.5	324.5	6.57	44.27	15.54	-2.42	0.0107	0.06	-0.04	-0.03	0.14	326.6	6.54	44.29	15.64	-2.56	0.0066	0.03	-0.02	-0.10	0.14
22.0	308.2	7.40	44.74	16.87	-2.08	0.0622	0.89	0.41	1.27	0.48	326.1	6.55	44.28	15.66	-2.56	0.0565	0.85	0.44	1.19	0.48
23.9	297.7	8.55	45.77	18.47	-1.39	0.0969	2.03	1.40	2.85	1.17	325.3	6.57	44.27	15.70	-2.56	0.0888	1.97	1.44	2.72	1.18
25.2	284.0	9.41	46.86	19.53	-0.71	0.1440	2.88	2.44	3.89	1.85	324.9	6.58	44.26	15.73	-2.56	0.1346	2.81	2.49	3.74	1.85
27.4	244.8	14.31	50.70	26.65	1.96	0.2925	7.72	6.11	10.84	4.52	324.9	6.58	44.26	15.73	-2.56	0.2831	7.65	6.15	10.69	4.53

[a] Offset value of 0.5 was subtracted from the raw data in order to adjust the calibration scale to that of AGAGE measurements.
[b] Subscript "trp" means the tropospheric value on the date of observation (see the note d).
[c] t_0 denotes the date of stratospheric entry of the observed air mass estimated from the CO_2 mixing ratio.
[d] Gray-shaded line indicates the height of tropopause, and tropospheric mixing ratio and isotopocule ratios on the date of observation which are estimated from the secular trends reported in literature (see also text).

Table S4: ε values presented in Figure 4.

Location / processes	Date	$-\varepsilon(^{15}N^{bulk})$ (‰)	$-\varepsilon(^{15}N^{\alpha})$ (‰)	$-\varepsilon(^{15}N^{\beta})$ (‰)	$-\varepsilon(^{18}O)$ (‰)	Notes
Sanriku Balloon Center, Japan (SBC)	4 June 1990	19.9 ± 2.7 27.9 ± 0.3	29.0 ± 7.1 39.2 ± 0.6	10.6 ± 1.9 16.0 ± 0.4	15.7 ± 1.9 23.7 ± 0.5	Top, values in the lower stratosphere; bottom, values in the middle stratosphere; boundary is defined by $-\ln\{[N_2O]/[N_2O]_0\} = 0.6$
	3 September 1998	15.6 ± 0.7 45.5 ± 5.9	24.9 ± 1.4 56.3 ± 7.1	6.0 ± 0.3 34.3 ± 5.7	14.5 ± 0.5 41.3 ± 6.2	
	31 May 1999	15.4 ± 1.5 27.9 ± 0.6	22.7 ± 1.7 39.7 ± 1.5	8.0 ± 1.7 15.3 ± 0.5	10.1 ± 1.9 23.8 ± 0.5	
	28 August 2000	15.3 ± 0.9 37.5 ± 1.8	19.3 ± 1.5 50.3 ± 3.4	11.2 ± 0.2 24.3 ± 3.1	13.1 ± 1.3 33.0 ± 1.5	
	30 May 2001	19.8 ± 0.4 27.3 ± 1.6	28.7 ± 1.4 37.6 ± 3.1	10.6 ± 1.1 16.9 ± 1.3	16.1 ± 0.9 22.5 ± 1.4	
	Average	17.2 ± 2.4 33.2 ± 8.1	24.9 ± 4.1 44.6 ± 8.3	9.3 ± 2.2 21.4 ± 8.1	13.9 ± 2.4 28.9 ± 8.1	
Kiruna, Sweden (ESR)	22 February 1997	18.0 ± 1.0 20.5 ± 0.4	27.2 ± 3.1 26.1 ± 2.0	8.6 ± 2.7 14.6 ± 1.4	13.8 ± 0.7 16.9 ± 0.5	
Syowa station, Antarctica (SYO)	3 January 1998	17.8 ± 0.6 42.3 ± 1.6	23.8 ± 2.4 58.8 ± 2.7	11.6 ± 2.6 25.1 ± 0.5	14.8 ± 0.8 36.7 ± 3.1	
Eastern equatorial Pacific (EQP) and Biak, Indonesia (BIK)	4–8 February 2012 and 22–28 February 2015	21.6 ± 1.4 44.9 ± 0.3	29.6 ± 2.1 63.0 ± 0.4	13.4 ± 1.0 26.3 ± 1.4	18.3 ± 1.8 35.7 ± 5.1	Same as above, but boundary is defined by $-\ln\{[N_2O]/[N_2O]_0\} = 0.2$
Photolysis	Not applicable	44.9 ± 3.1	63.8 ± 4.6	25.9 ± 1.6	37.0 ± 2.1	Estimated for the region of 20–40 km and 190–240 K based on Kaiser et al. (2002 & 2003)
Photooxidation	Not applicable	5.2 ± 0.3	2.8 ± 0.6	7.5 ± 1.3	10.8 ± 1.4	Average of the values in Kaiser et al. (2002) and Toyoda et al. (2004)

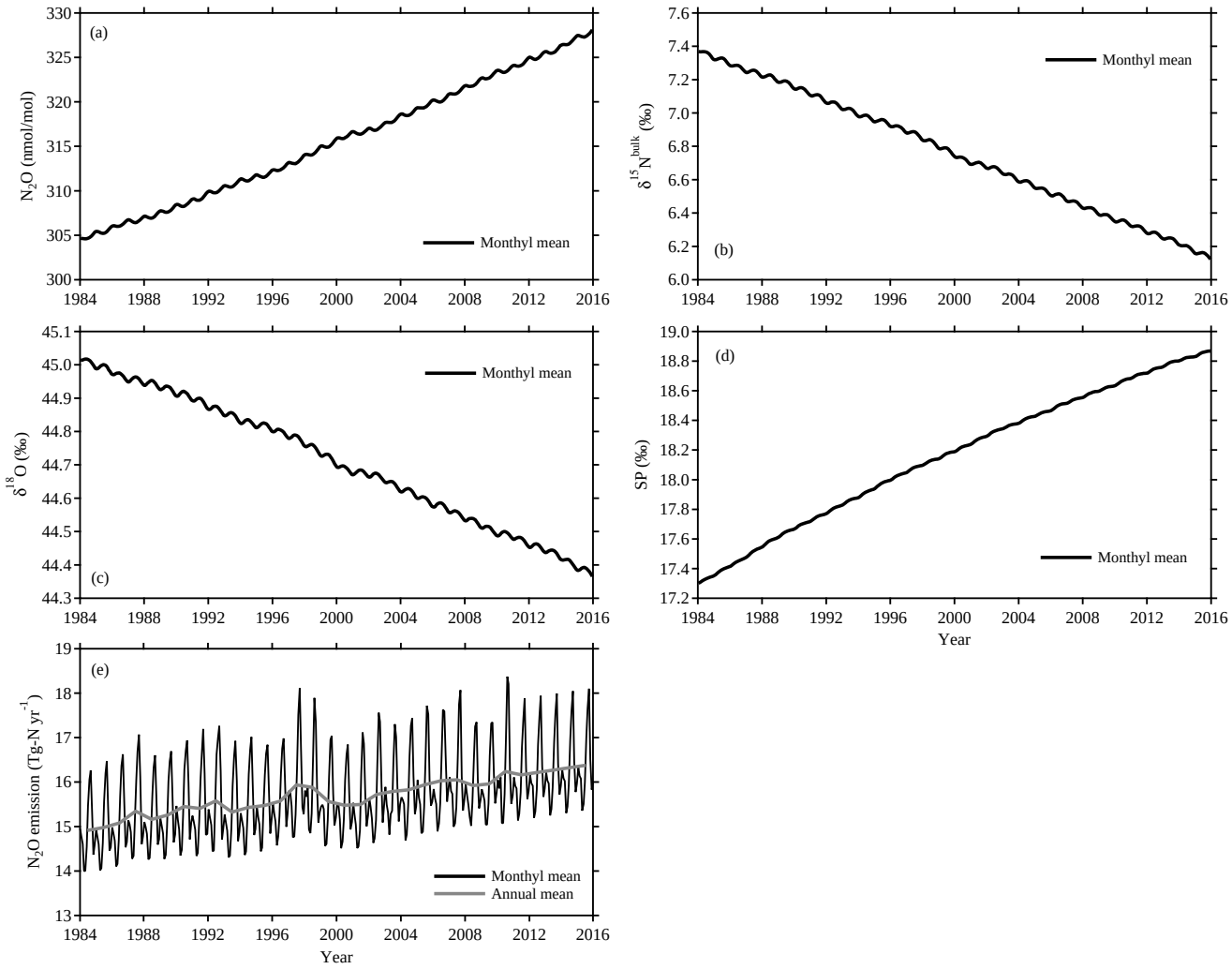

Figure S1: Global monthly mean mixing ratio (a), $\delta^{15}N^{bulk}$ (b), $\delta^{18}O$ (c), and SP (d) of atmospheric N_2O, and global total monthly and annual N_2O emissions (e) in model.

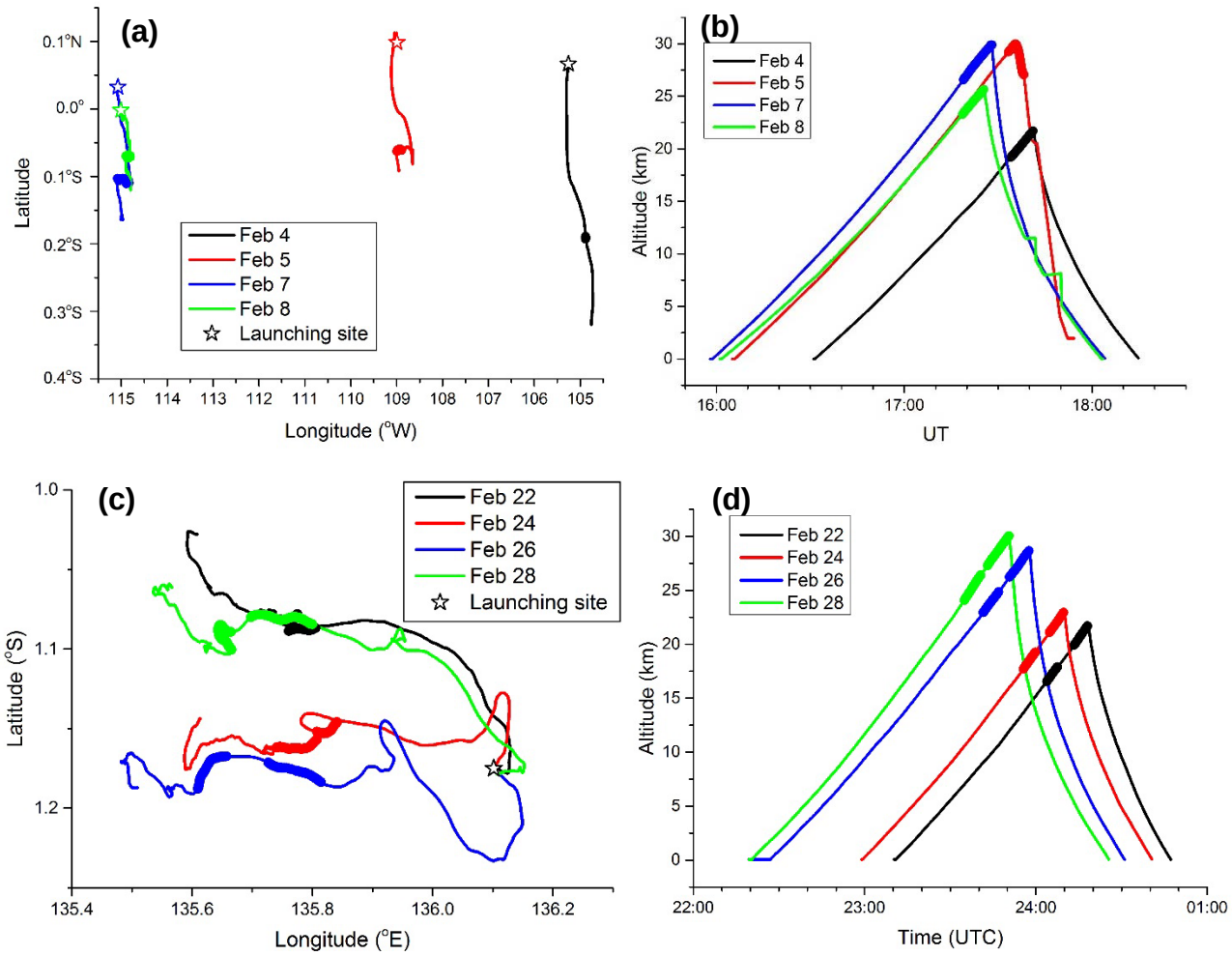

Figure S2: Trajectories of the balloons launched from Hakuho-maru in the eastern equatorial Pacific (a, b) and LAPAN observatory at Biak island, Indonesia (c, d). Panels (a) and (c) show horizontal trajectories; panels (b) and (d) show time-altitude trajectories. Sampling positions are shown by solid circles.

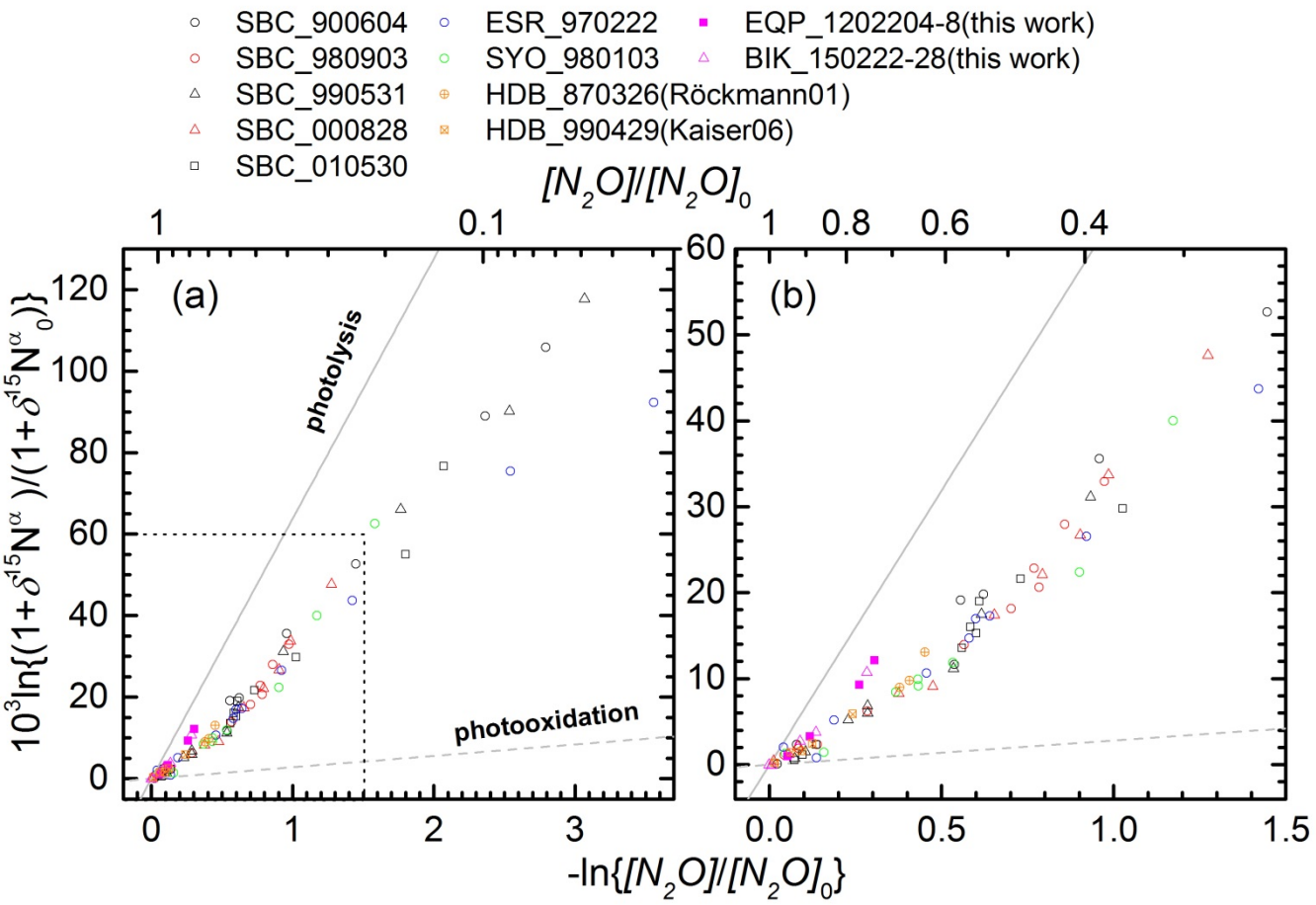

Figure S3: Correlation between mixing ratio and $\delta^{15}N^{\alpha}$ of N_2O (Rayleigh plot). The high mixing ratio range (> ca. 120 nmol mol^{-1}) in (a) is enlarged in (b). Both parameters are normalized to their values at the time when the corresponding air mass entered the stratosphere (see Eq. 6 in the text). Grey solid and broken lines show slopes obtained respectively from laboratory broadband photolysis experiments (Kaiser et al., 2002; 2003) and photooxidation experiments (Kaiser et al., 2002; Toyoda et al., 2004).

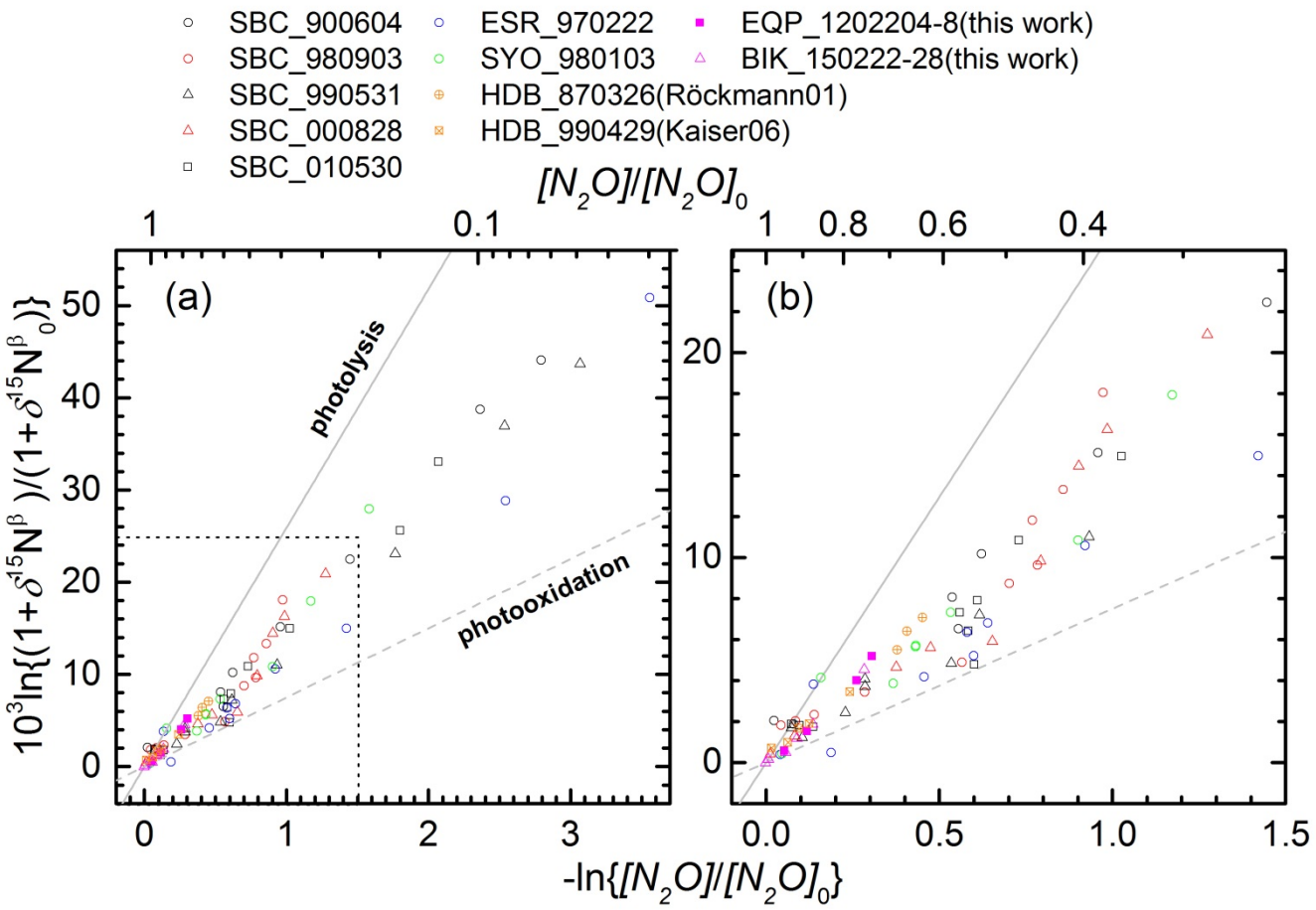

Figure S4: Same as Fig. S3, but for $\delta^{15}N^{\beta}$.

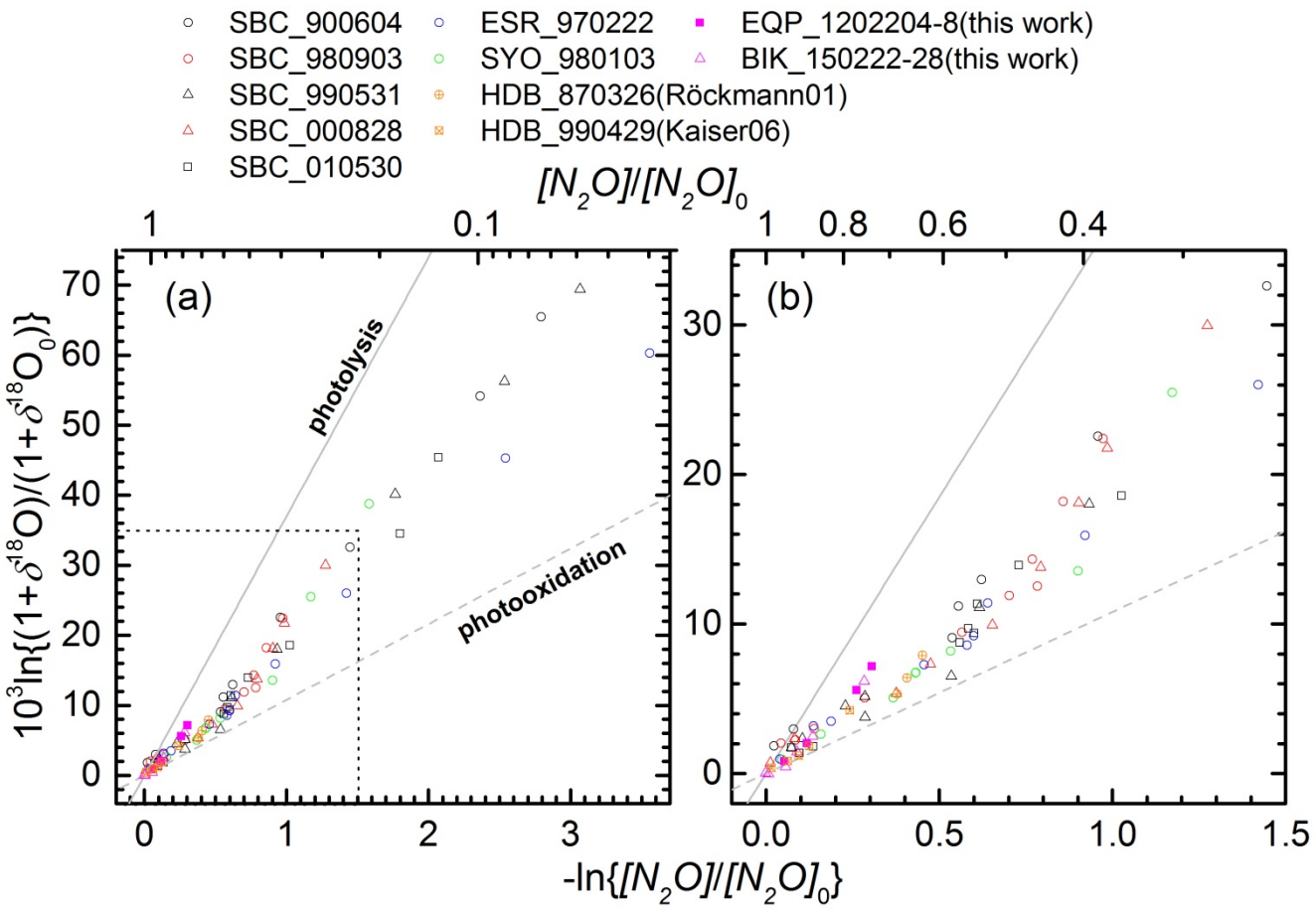

Figure S5: Same as Fig. S3, but for $\delta^{18}O$.

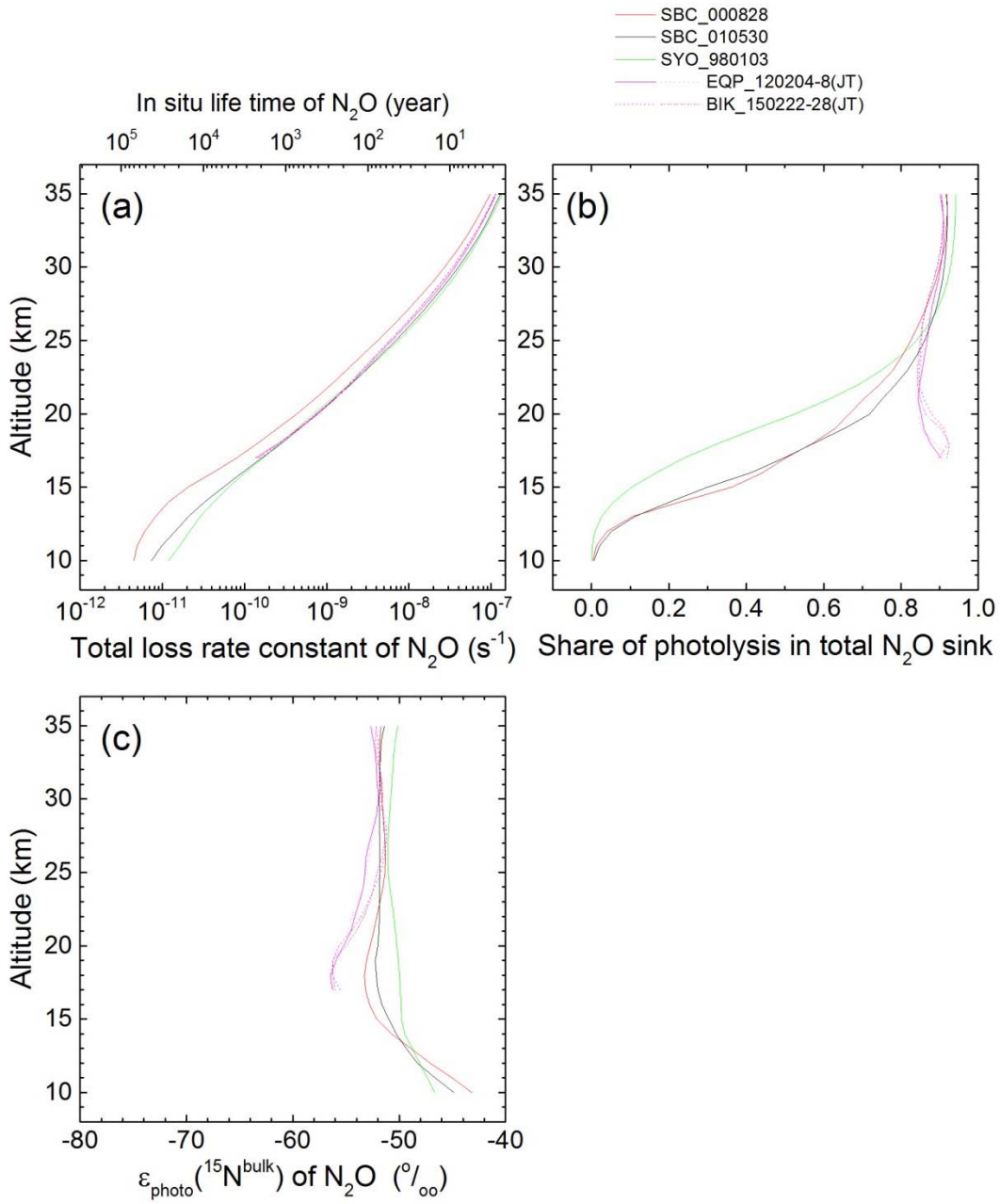

Figure S6: Vertical profiles of parameters derived from N_2O decomposition rates used in the ACTM. (a), total loss rate constant of N_2O; (b), share of photolysis in the total N_2O sink; (c), $\varepsilon(^{15}N^{bulk})$ for photolysis.