Supplement of

An improved Trajectory-mapped Ozonesonde dataset for the Stratosphere and Troposphere (TOST): update, validation and applications

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Table S1. Information on the ozonesonde stations used, including each station's ID number, name, locations, the number of profiles used, and measurement period. The stations in bold fonts are the stations showing a drop-off of 2-8% in the stratospheric ozone and total ozone column since 2013. The Beijing station is named the Nanjiao Meteorological Observatory.

ID	Station name	Longitude	Latitude	Altitude	Start-year	End-year	No. profiles
255	Ainsworth (Airport)	-100	42.6	789	1986	1986	7
494	Alajuela	-84.2	10	899	2005	2009	133
229	Albrook	-79.5	9	66	1980	1980	20
18	**Alert**	**-62.3**	**82.5**	**62**	**1987**	**2021**	**1650**
111	Amundsen-scott	-24.8	-90	2810	1970	1987	183
348	Ankara	32.9	40	891	1994	2012	340
328	**Ascension Island**	**-14.2**	**-7.6**	**91**	**1990**	**2021**	**905**
483	Barbados	-59.4	13.2	32	2006	2006	27
199	Barrow	-156.6	71.3	11	1974	2008	21
104	Bedford	-71.3	42.5	80	1970	1971	53
/	Beijing	116.5	39.8	30	2001	2019	902
/	Belgrano	-34.6	-77.9	250	2016	2021	119
420	Beltsville (md)	-76.5	39	72	2006	2006	12
181	Berlin/Templehof	13.4	52.5	50	1970	1973	134
197	Biscarrosse/Sms	-1.2	44.4	18	1976	1983	359
525	Bogota	-74.1	4.7	2541	1998	2008	64
67	Boulder Esrl Hq (co)	-105.2	39.9	1743	1970	2022	1976
338	Bratts Lake	-104.7	50.2	580	2003	2011	405
329	Brazzaville	15.2	-4.3	314	1990	1992	82
394	Broadmeadows	144.9	-37.7	110	1999	2021	1075
38	Cagliari/Elmas	9.1	39.2	4	1970	1980	379
20	Caribou	-68	46.9	192	1981	1981	1
444	Cheju	126.5	33.5	300	2001	2001	13
224	Chilca	-76.8	-12.5	-1	1975	1975	3
77	**Churchill**	**-94.1**	**58.8**	**35**	**1973**	**2021**	**1748**
198	Cold Lake	-110	54.8	702	1977	1981	66
236	Coolidge Field	-61.8	17.3	10	1976	1976	7
472	Cotonou	2.2	6.2	9.5	2005	2007	97
334	Cuiaba	-56.1	-15.6	990	1992	1992	21
450	Davis	78	-68.6	14	2003	2019	501
316	De Bilt	5.2	52.1	4	1992	2021	1528
238	Denver	-104.9	39.8	1611	1977	1977	1
/	Dumont	140	-66.4	20	1991	2019	704
441	Easter Island	-109.4	-27.2	69.2	1995	2021	323
456	Egbert	-79.8	44.2	251	2003	2011	372
213	El Arenosillo	-6.7	37.1	41	1977	1983	18
335	Etosha Pan	15.9	-19.2	1100	1992	1992	16
315	**Eureka**	**-86.4**	**80**	**10**	**1992**	**2021**	**1911**
203	Ft. Sherman	-80	9.3	57	1977	1977	16
228	Gimli	-97	50.6	228	1980	1985	31
76	Goose Bay	-60.3	53.3	44	1970	2021	2317
237	Great Falls	-111.3	47.5	1118	1977	1977	4
330	**Hanoi**	**105.8**	**21**	**6**	**2004**	**2021**	**350**
40	Haute provence	5.7	43.9	684	1981	1997	61
477	Heredia	-84.1	10	1176	2005	2011	127
109	**Hilo (hi)**	**-155.1**	**19.7**	**11**	**1982**	**2021**	**1742**
92	Hobart	147.5	-42.8	4	2021	2021	1
99	Hohenpeissenberg	11	47.8	976	1970	2021	6058
361	Holtville (ca)	-115.4	32.8	-19	2006	2006	13

484	Houston (tx)	-95.4	29.7	19	2004	2006	62
418	Huntsville	-86.6	35.3	196	1999	2007	575
303	Iqaluit	-68.5	63.8	20	1991	1992	30
265	Irene	28.2	-25.9	1524	1990	2021	540
336	Isfahan	51.7	32.5	1550	1995	2011	151
404	Jokioinen	23.5	60.8	103	1995	1998	99
439	Kaashidhoo	73.5	5	1	1999	1999	54
7	Kagoshima	130.6	31.6	283	1970	2005	816
457	**Kelowna**	**-119.4**	**49.9**	**456**	**2003**	**2017**	**700**
344	Kings park	114.2	22.3	66	2000	2021	976
225	Kourou	-52.6	5.3	4	1974	1974	3
436	La Reunion	55.5	-21.1	24	1998	2021	816
/	Laquila	13.3	42.4	683	1994	2021	309
256	**Lauder**	**169.7**	**-45**	**370**	**1986**	**2021**	**1966**
254	Laverton	144.8	-37.9	21	1984	1999	340
221	Legionowo	21	52.4	96	1979	2021	2170
43	Lerwick	-1.2	60.1	80	1992	2016	1223
174	Lindenberg	14.1	52.2	112	1975	2021	2727
235	Long View	-94.8	32.5	103	1976	1976	2
29	Macquarie Island	159	-54.5	6	1994	2021	1176
308	Madrid	-3.8	40.5	640	1994	2021	1132
400	Maitri	11.4	-70.5	330	1994	2008	141
448	Malindi	40.2	-3	-6	1999	2006	191
233	Marambio	-56.6	-64.2	198	1988	2019	1286
466	Maxaranguape (Natal)	-35.4	-5.4	42	2002	2016	355
/	Mcmurdo	166.7	-78.8	10	1986	2010	822
88	Mirny	93	-66.5	30	1989	1991	114
190	Naha	127.7	26.2	27	1989	2018	1107
175	**Nairobi**	**36.8**	**-1.3**	**1710**	**1996**	**2020**	**1001**
487	Narragansett	-71.4	41.5	21	2006	2008	51
219	Natal	-35.2	-5.8	32	1979	2021	866
323	Neumayer	-8.3	-70.7	42	1992	2017	1737
10	New Delhi	77.1	28.3	273	1984	2016	197
280	Novolasarevskaya/Forster	11.9	-70.8	110	1985	1991	393
89	Ny-aalesund	11.9	78.9	11	1992	2021	2562
/	OHP	5.7	43.9	777	1991	2021	1428
523	**Pago Pago/American Samoa**	**-170.6**	**-14.2**	**77**	**1998**	**2021**	**850**
210	Palestine	-95.7	31.8	121	1975	1985	163
432	Papeete (Tahiti)	-149	-18	2	1995	1999	168
488	Paradox	-73.6	43.9	284	2006	2006	8
435	Paramaribo	-55.2	5.8	7	1999	2021	834
156	Payerne	6.6	46.5	491	1970	2021	6693
360	Pellston (mi)	-84.7	45.6	235	2004	2004	38
322	Petaling Jaya	101.7	2.7	17	1998	2021	477
332	Pohang	129.4	36	2.5	1995	2020	1050
217	Poker Flat	-147.5	65.1	204	1979	1982	40
187	Poona	73.8	18.5	559	1984	2009	140
526	Port Hardy	-127.4	50.7	17	2018	2021	137
333	Porto Nacional	-48.4	-10.8	240	1992	1992	15
242	Praha	14.4	50	304	1979	2021	1873
440	r h Brown Research Ship	-65.6	30.6	2	1999	2006	89
212	r/v a.k.Shirshov	75	-15	-1	1977	1977	32
24	**Resolute**	**-95**	**74.7**	**64**	**1970**	**2021**	**2037**
489	Richland	-119.2	46.2	123	2006	2006	24
297	S.Pietro Capofiume	11.6	44.6	11	1984	1993	98
480	Sable Island	-60	43.9	4	2004	2006	61

434	**San Cristobal**	**-89.6**	**-0.9**	**8**	**1998**	**2021**	**445**
239	San Diego	-117.2	32.8	124	1977	1977	2
234	San Juan	-66.1	18.5	17	1976	1976	6
524	**San Pedro**	**-84.2**	**10**	**899**	**2005**	**2021**	**658**
401	Santa Cruz-Botanico (Tenerife)	-16.3	28.5	36	1995	2021	1362
12	Sapporo	141.3	43.1	19	1970	2018	1392
406	Scoresbysund	-21.9	70.5	9999	1989	2020	1494
443	Sepang Airport	101.7	2.7	17	1998	2017	372
214	Singapore	103.9	1.3	36	2012	2015	37
262	Sodankyla	26.6	67.4	179	1988	2021	1763
132	Sofia	23.4	42.8	588	1982	1991	239
997	South Pole	-169	-90	2834	1970	2021	2251
231	Spokane	-117.4	47.7	576	1976	1976	7
21	**Stonyplain/Edmonton**	**-114.1**	**53.5**	**766**	**1970**	**2021**	**2121**
491	Summit	-38.5	72.6	3202	2005	2015	551
438	**Suva/Fuji**	**178.2**	**-18.1**	**6**	**1997**	**2021**	**502**
101	Syowa	39.6	-69	21	1970	2021	1925
260	Table Mountain (ca)	-117.7	34.4	2285	2006	2006	35
95	Taipei	121.4	25	11	2000	2021	141
14	Tateno (Tsukuba)	140.1	36.1	31	1970	2021	1876
485	Tecamec (Unam)	-99.2	19.3	2272	2006	2006	35
205	Thiruvananthapuram	77	8.5	60	1984	2009	205
460	Thule	-68.7	76.5	57	1991	2003	249
65	Toronto	-79.5	43.8	198	1976	1994	15
445	Trinidad Head	-124.1	41.1	36	1997	2021	1286
53	Uccle	4.3	50.8	100	1970	2021	6352
339	Ushuaia	-68.3	-54.9	17	2008	2019	227
318	Valentia	-10.2	51.9	14	1994	2021	745
490	Valparaiso (in)	-87	41.5	240	2006	2006	18
257	Vanscoy	-107.3	52.2	510	1990	2004	57
55	Vigna Di Valle	12.2	42.1	260	2011	2021	172
107	Wallops Island	-75.5	37.9	13	1970	2020	2113
482	Walsingham	-80.6	42.6	200	2006	2006	43
437	Watukosek (Java)	112.7	-7.6	50	1998	2021	357
458	**Yarmouth**	**-66.1**	**43.9**	**9**	**2003**	**2021**	**794**
194	Yorkton	-102.5	51.3	504	1975	1978	72

Table S2. Validation of the trajectory-derived (TOST) against ozonesonde measurement in two seasons (JJA: June-July-August, DJF: December-January-February) by decade from the 1980s to 2010s (in red text). The validation of SAGE and MLS ozone in the corresponding decade is provided as a comparison (in blue text).

	1980s JJA		**1980s DJF**		**1990s JJA**		**1990s DJF**	
	TOST	**SAGE**	**TOST**	**SAGE**	**TOST**	**SAGE**	**TOST**	**SAGE**
R	0.97	0.97	0.97	0.95	0.97	0.98	0.96	0.96
RMS	359.79	357.71	436.99	569.58	381.08	335.68	522.18	509.68
Bias	-26.80	-47.01	-19.46	-115.93	1.09	11.74	58.95	3.33
RD	-1.16	-2.04	-0.64	-3.81	0.05	0.52	2.04	0.11
N	271	271	779	779	1494	1494	1920	1920
	2000s JJA		**2000s DJF**		**2010s JJA**		**2010s DJF**	
	TOST	**MLS**	**TOST**	**MLS**	**TOST**	**MLS**	**TOST**	**MLS**
R	0.96	0.96	0.96	0.96	0.96	0.95	0.97	0.97
RMS	470.80	566.49	516.80	495.71	474.20	575.47	446.90	449.48
Bias	10.23	249.18	1.68	-18.40	-7.92	258.47	0.96	-43.47
RD	0.44	10.93	0.06	-0.69	-0.34	11.33	0.03	-1.59
N	7460	7460	7661	7661	11432	11432	12133	12133

Table S3. Comparisons of data coverage, number of ozonesonde stations and ozonesonde profiles between TOST-v1 and TOST-v2.

Data coverage (%)	**90-60S**	**60-30S**	**30S-30N**	**30-60N**	**60-90N**
TOST-v1	53.57 ± 14.06	39.61 ± 15.75	24.26 ± 13.73	78.44 ± 18.75	62.61 ± 12.70
TOST-v2	57.40 ± 15.36	44.12 ± 17.42	28.00 ± 16.34	80.98 ± 18.10	64.24 ± 13.36
Ozonesonde stations					
TOST-v1	8	4	32	46	10
TOST-v2	12	6	48	64	13
Ozonesonde profiles					
TOST-v1	3764	2216	6363	32779	4765
TOST-v2	10176	4785	17934	54432	13630

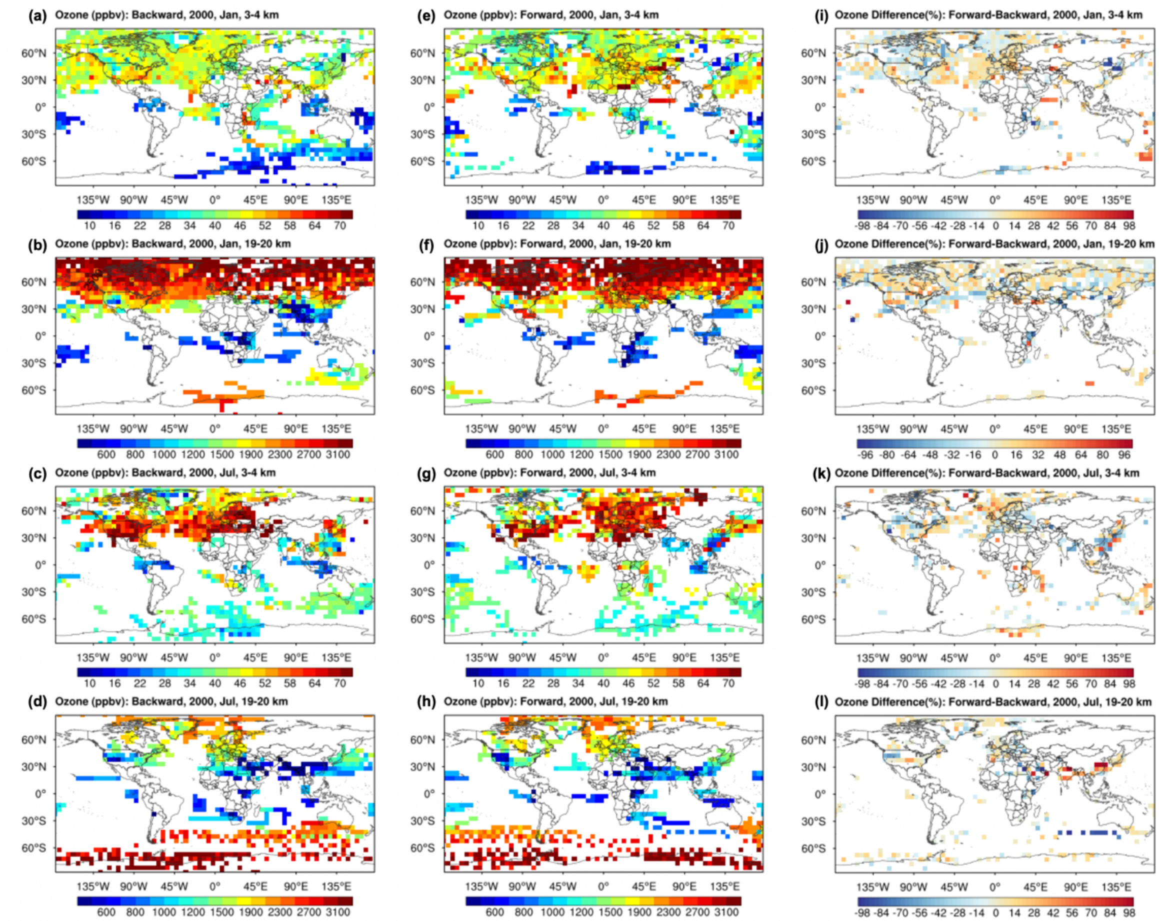

Figure S1. (a-d) Global distribution of monthly mean ozone at 3-4 km and 19-20 km in January and July 2020 from forward trajectories. (e-h) same as (a-d) but for backward trajectories. (i-l) the relative difference in monthly mean ozone between forward and backward trajectories [100 × (forward trajectories - backward trajectories) / (0.5 × forward trajectories + 0.5 × backward trajectories), in %].

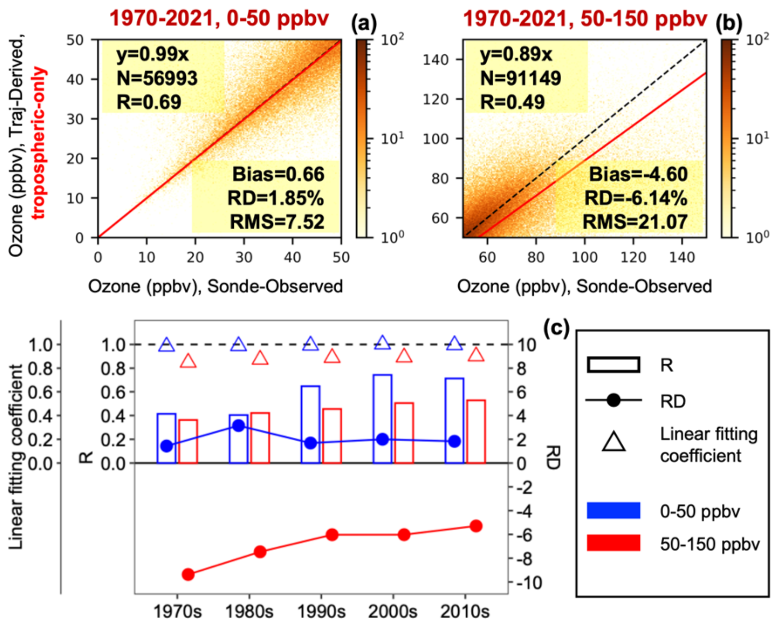

Figure S2. (a-c) Comparison of monthly average tropospheric ozone mixing ratios from ozonesondes (Sonde-Observed) and trajectory-derived TOST data with trajectories from observations only in the troposphere (**Traj-Derived, tropospheric-only**) for the entire study period of ozone concentration at 0-50 ppbv and 50-150 ppbv. Solid red lines represent the linear fitting line (with the intercept set to 0) and dashed black lines denote the 1:1 axis. N is the total number of data points, R is the correlation coefficient, Bias is the overall average difference in monthly mean values [Traj-Derived ozone - Sonde-Observed ozone, in ppbv], RD is the relative difference in % [100 × (Traj-Derived ozone - Sonde-Observed ozone)/ Sonde-Observed ozone)], and RMS is the root mean square difference in ppbv). Note that Traj-Derived ozone at each station is derived without input from the station itself; that is, Traj-Derived represents an ensemble of 141 separate computations of TOST, each one withholding a single validation station. (d) the R (bars), RD (dots and lines) and linear fitting coefficient (with the intercept set to 0; triangles) between the Traj-Derived ozone and Sonde-Observed ozone by decade. The dashed line denotes where the linear fitting coefficient is 1.

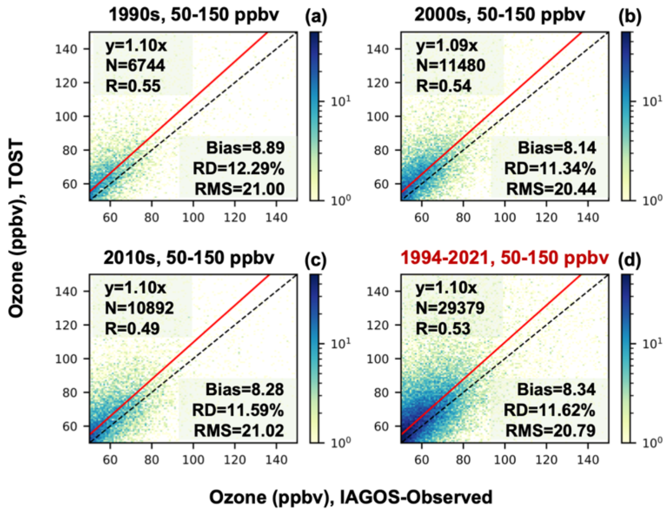

Figure S3. The comparison of monthly ozone mixing ratios between IAGOS-observed (x-axis labeled: IAGOS-Observed) and TOST data (y-axis labelled: TOST) by decade (a-c) and for the entire study period (d) of ozone concentration at 50-150 ppbv. Solid red lines represent the linear fitting line (with the intercept set to 0) and dashed black lines denote the 1:1 axis. N is the total number of data points, R is the correlation coefficient (unitless), Bias is the difference in monthly mean values [TOST ozone - IAGOS ozone, unit: ppbv], RD is the relative difference [100 × (TOST ozone - IAGOS ozone)/(0.5 × TOST ozone + 0.5 × IAGOS ozone)], and RMS the root mean square difference (unit: ppbv).

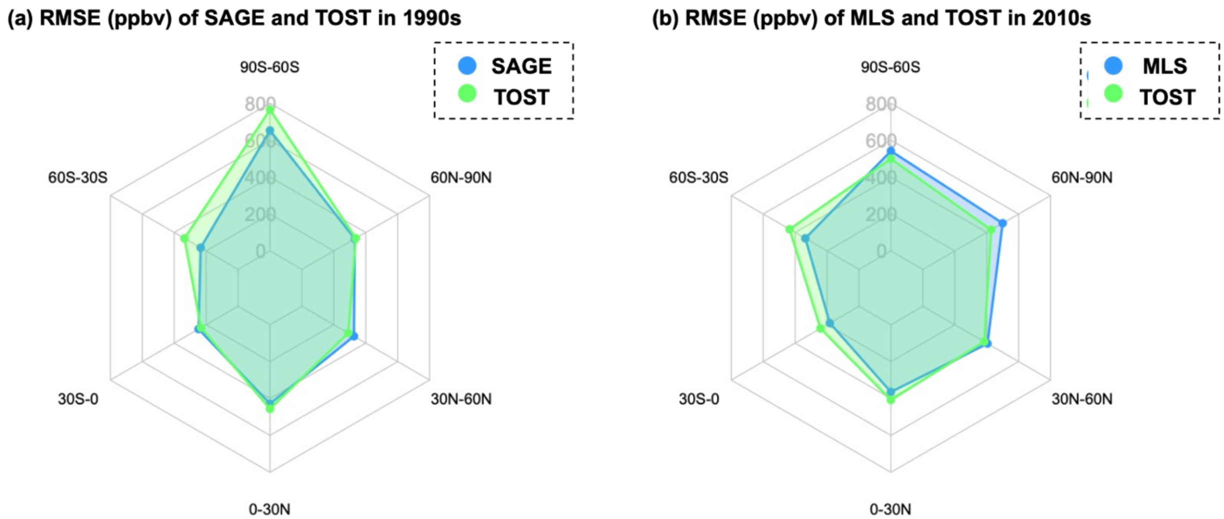

Figure S4. Radar plot for RMSE of TOST Traj-derived and satellite ozone data against ozonesonde measurements at different latitudinal zones. (a) the RMSE for Traj-derived and SAGE ozone in the 1990s; (b) the RMSE for Traj-derived and MLS ozone in the 2010s. The RMSE in a given latitudinal zone is from the monthly ozone mixing ratios between the Traj-derived (or satellite) ozone and ozonesonde measurement in 16-26 in that latitudinal zone. Traj-derived ozone is without the input of stations being tested.

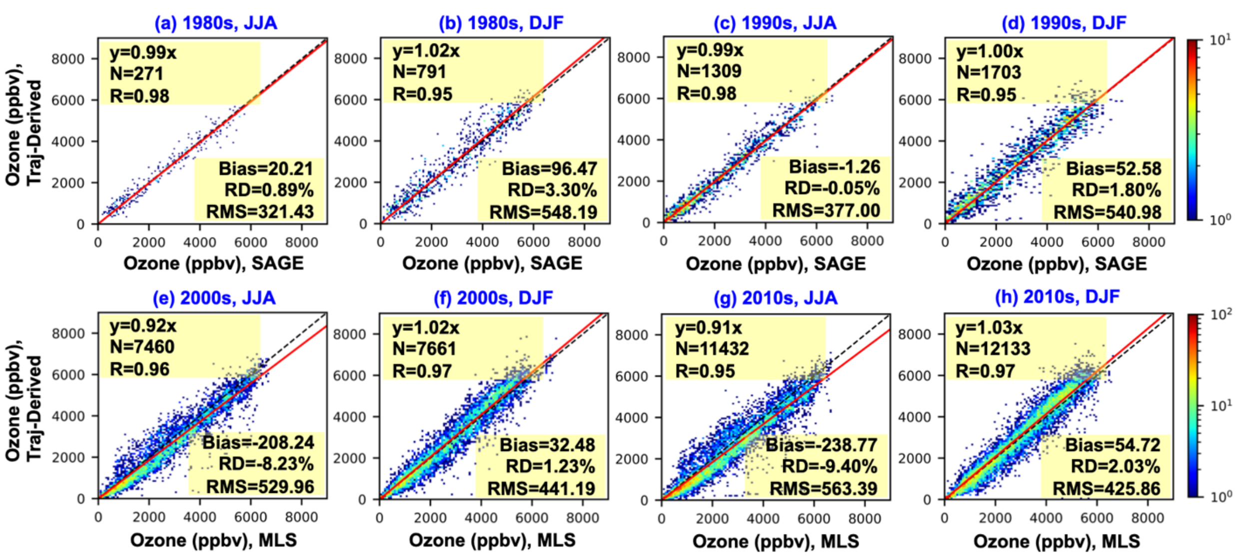

Figure S5. Similar to Figure 2a-c, comparisons in monthly ozone mixing ratios between trajectory-derived (Traj-Derived) and satellite-based (SAGE and MLS) ozone data in JJA and

DJF in the stratosphere from 16-26 km by decades, in the 1980s and 1990s for SAGE and in the 2000s and 2010s for MLS.

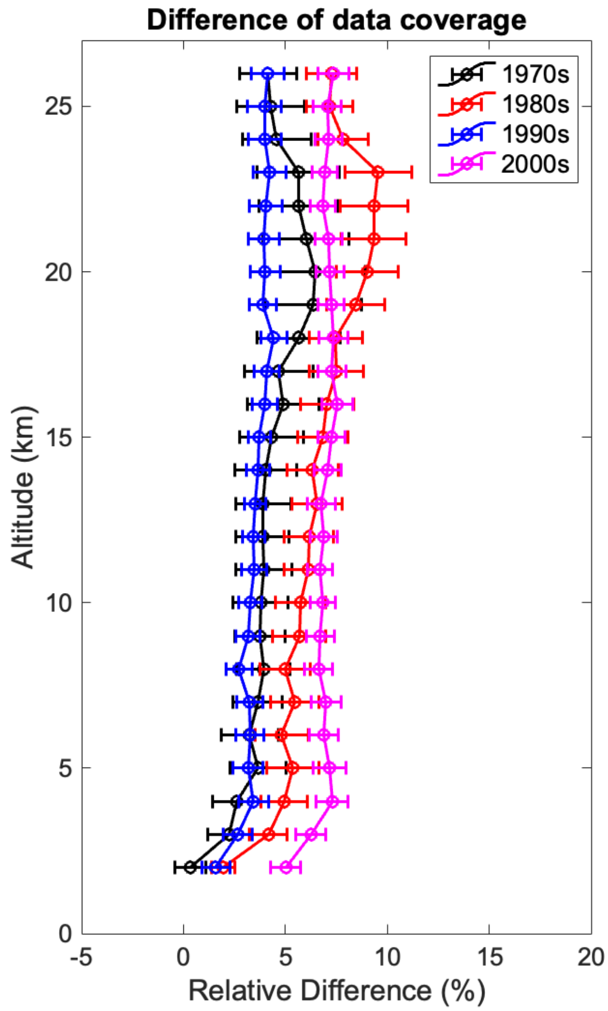

Figure S6. The mean relative difference in data coverage between TOST-v1 and TOST-v2 [100 × (TOST-v2 data coverage – TOST-v1 data coverage)/TOST-v1 data coverage (in %)] for four decades from 1970-2008. The suface layer (0-1 km) is not compared here due to the topography issue with TOST-v1 (see Figure 9 and Section 3.3 for details).

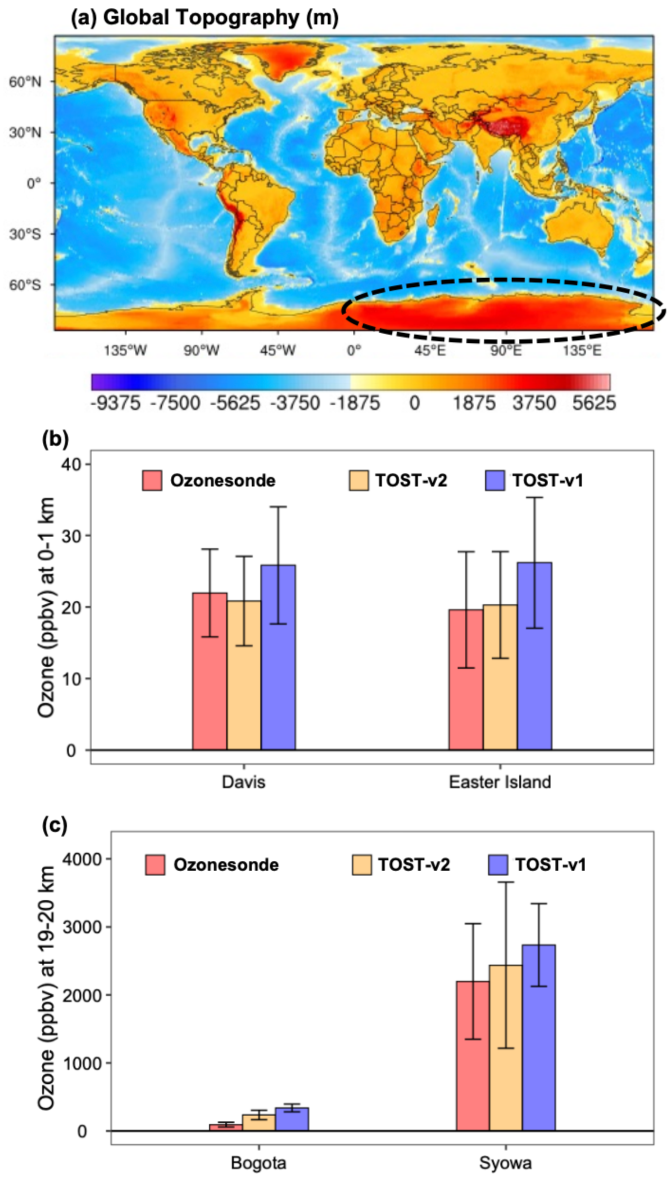

Figure S7. (a) The global topographical map. The dashed circles indicate the regions with large spatial differences between the two versions of TOST data (v1 and v2) at 0-1 km. (b, c) The mean ozone concentrations at 0-1 km and 19-20 km in the 2000s at two stations for ozonesonde, TOST-v2 and TOST-v1.