



Supplement of

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

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Section S1. The metrics for validation and comparison

Multiple metrics were used to indicate the agreement and differences between the TOST (use y here) and ozonesonde/aircraft data (use x here).

1. Correlation Coefficient (R, unitless):

$$R = \frac{\sum(x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum(x_i - \bar{x})^2 \sum(y_i - \bar{y})^2}}, \text{ where } \bar{x} \text{ and } \bar{y} \text{ is the mean of the } x \text{ and } y \text{ variables, respectively.}$$

2. Linear fitting coefficient (m , unitless), with the intercept set to 0:

$$m = \frac{\sum(x_i y_i)}{\sum(x_i^2)}$$

3. Bias (in ppb):

$$Bias = \frac{1}{n} \sum (x_i - y_i)$$

4. Relative Difference (RD, in %):

$$RD = 100 \times \frac{Bias}{\bar{x}}, \text{ if comparing with ozonesonde data } x$$

$$RD = 100 \times \frac{Bias}{\frac{1}{2n} \sum (x_i + y_i)}, \text{ if comparing with satellite data } x$$

5. Root Mean Square Difference (RMSD, in ppb):

$$RMSD = \sqrt{\frac{1}{n} \sum (x_i - y_i)^2}$$

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 those 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. Information on the airports from In-Service Aircraft for a Global Observing System (IAGOS) used (310 in total), including each airport's name, measurement period and the number of flights used.

Airport name	Start year	End year	Number of flights
ABIDJAN	1997	2017	145
ABU-DHABI	1999	2017	148
ABUJA	2003	2021	135
ACCRA	2000	2018	94
ADDIS-ABABA	2001	2019	206
ADELAIDE	2013	2015	9
AGADIR	2001	2001	1
AKITA	2013	2013	1
ALGHERO	2018	2018	1
ALMATY	1999	2020	169
ALMERIA	2002	2002	1
AMMAN	2006	2020	5
AMSTERDAM	1995	2016	175
ANGELES-CITY	2015	2015	1
ANKARA	1997	1997	1
ANTALYA	1996	2006	29
ANTANANARIVO	1997	2018	57
ANTIGUA	1994	1995	16
APIA	2018	2018	6
ASHGABAT	2001	2013	32
ASMARA	2002	2012	18
ASTANA	2006	2020	141
ATHENS	1997	2017	8
ATLANTA	1994	2020	1292
AUCKLAND	2016	2020	70
AUSTIN	2019	2020	16
BAHRAIN	2007	2021	183
BAKU	2001	2013	49
BALTIMORE	2006	2006	1
BAMAKO	2013	2017	89
BAMENDA	2018	2018	1
BANGALORE	2015	2021	24
BANGKOK	1994	2020	1108
BANJUL	1997	1997	1
BARCELONA	2016	2016	1
BEIJING	1995	2018	642
BEIRUT	1998	2018	74

BERLIN	1994	2018	30
BOGOTA	1994	2021	522
BORDEAUX	1998	2018	16
BOSTON	1994	2021	1029
BRATISLAVA	1996	2000	3
BRAZZAVILLE	1997	2018	26
BREMEN	1997	2009	3
BRISBANE	2013	2020	87
BRUSSELS	1997	2009	1389
BUCHAREST	2004	2006	2
BUDAPEST	2000	2000	1
BUENOS-AIRES	1994	2016	46
BUSAN	2016	2019	21
CAIRNS	2013	2014	19
CAIRO	1998	2019	240
CALDWELL	1995	1995	1
CALGARY	2009	2012	153
CALI	1995	1995	1
CAMPINAS	1996	1999	3
CANCUN	2001	2021	15
CAPE-TOWN	1996	2021	111
CARACAS	1994	2018	808
CAYENNE	1994	2013	140
CEBU	2013	2018	41
CHANGCHA	2015	2019	17
CHARLOTTE	2004	2019	22
CHENGDU	2013	2018	7
CHICAGO	1994	2019	896
CHONGJU	2017	2017	1
CHONGQING	2019	2019	3
CHRISTCHURCH	2016	2018	19
CINCINNATI	1997	2004	176
CLEVELAND	2017	2017	2
COLOGNE	1995	2018	12
COLOMBO	1994	2018	69
CONAKRY	1998	2018	111
COTONOU	1997	2018	128
DAKAR	1997	2018	58
DALAMAN	1995	2005	6
DALIAN	2015	2016	2
DALLAS	1995	2020	958
DAMMAM	2000	2020	152
DARWIN	2004	2004	2
DELHI	1995	2021	623
DENPASAR	2014	2019	61
DENVER	2001	2020	44
DETROIT	1999	2020	413
DHAHRAN	1999	1999	1
DJIBOUTI	2013	2018	12
DOHA	2011	2017	179
DONAUESCHINGEN	2018	2018	1
DOUALA	1997	2018	116
DRESDEN	1999	2018	3
DRUMMOND-ISLAND	2018	2018	1
DUBAI	1997	2020	537
DUBLIN	2019	2020	3

DUBROVNIK	1999	1999	2
DURBAN	2008	2008	1
DUSSELDORF	1995	2015	643
ENTEBBE	1997	1998	25
FAIRBANKS	2015	2016	7
FRANKFURT	1994	2021	8514
FREETOWN	2009	2018	41
FRESNO	2017	2017	1
FUJEIRAH	2015	2015	1
FUKUOKA	2012	2020	146
GANDER	2004	2004	1
GERONA	2018	2018	3
GIEBELSTADT	2011	2011	1
GOOSE-BAY	2000	2000	1
GRAN-CANARIA	2002	2016	4
GREENVILE	1997	1997	1
GUAM	2016	2019	7
GUANGZHOU	2004	2018	92
GUATEMALA-CITY	2014	2015	26
GUAYAQUIL	1997	2016	22
HAMBURG	1997	2019	20
HANGZHOU	2017	2017	1
HANNOVER	2000	2000	1
HANOI	1994	2019	145
HARARE	1998	2008	47
HAVANA	2003	2018	19
HEFEI	2016	2016	1
HELSINKI	1997	1997	1
HERAKLION	1996	1997	16
HIROSHIMA	2016	2018	10
HO-CHI-MINH-CITY	1994	2019	321
HOLGUIN	2001	2001	4
HONG-KONG	1999	2021	1580
HONOLULU	2015	2020	570
HOUSTON	1994	2021	474
HUESCA	1998	1998	1
HURGHADA	2000	2001	11
HYDERABAD	2005	2018	450
IBARAKI	2001	2001	1
ILORIN	2018	2018	1
ISLAMABAD	2001	2001	1
ISTANBUL	1995	1995	1
JAKARTA	1996	2018	157
JEDDAH	1999	2019	193
JOHANNESBURG	1995	2011	256
KAGOSHIMA	2012	2018	4
KAHULUI	2017	2018	73
KAOHSIUNG	2013	2018	25
KARLSRUHE-BADEN-BADEN	2018	2018	1
KATHMANDU	2002	2006	10
KHABAROVSK	1997	1997	1
KHARTOUM	2000	2014	102
KIGALI	1997	1998	32
KINSHASA	1998	2018	69
KOBE	1998	1998	4
KOBLENZ	1995	2001	2

KOCHI	2018	2018	1
KOLKATA	2009	2009	1
KONA	2017	2020	37
KUALA-LUMPUR	1998	2019	207
KUWAIT-CITY	2001	2020	241
LAAGE	2004	2004	2
LAGOS	1997	2020	310
LAJES-(TERCEIRA-ISLAND)	2016	2016	1
LARNACA	2000	2000	1
LAS-VEGAS	2017	2018	48
LEIPZIG	1999	1999	2
LIBREVILLE	1997	2018	25
LIMA	1994	2016	28
LINZ	2018	2018	1
LISBON	1999	2019	4
LOME	1997	2018	112
LONDON	1995	2016	244
LOS-ANGELES	1995	2020	317
LUANDA	1997	2020	190
LUSAKA	2012	2012	1
LUXOR	2001	2018	5
LYON	2017	2020	3
MADRAS	1994	2018	521
MADRID	2014	2016	255
MALABO	2014	2020	164
MALAGA	1998	2001	4
MALE	1997	2021	63
MALTA	2011	2019	15
MANCHESTER	2012	2012	1
MANILA	2005	2020	232
MAPUTO	1998	1999	6
MARACAY	1995	1995	1
MARSEILLE	1998	2017	5
MARTINSBURG	2000	2000	1
MELBOURNE	2013	2018	68
MEXICO-CITY	1994	2018	49
MIAMI	1994	2020	139
MILANO	2016	2018	2
MILWAUKEE	2003	2003	4
MINNEAPOLIS	2013	2019	124
MIYAZAKI	2014	2014	1
MONTEVIDEO	1994	2016	28
MONTREAL	1994	2019	331
MOSCOW	2011	2018	2
MUMBAI	1996	2021	211
MUNICH	1996	2020	1340
MUSCAT	2002	2015	50
NAGOYA	1997	2020	826
NAIROBI	1994	2020	97
NANAIMO	2014	2014	1
NANCHANG	2013	2018	5
NANJING	2009	2020	304
NDJAMENA	2013	2018	16
NEFTEYUGANSK	2015	2015	1
NEW-ORLEANS	1997	1997	1
NEW-YORK	1994	2021	1977

NIAMEY	2002	2018	66
NICE	1994	2018	6
NINBO	2012	2018	38
NOUAKCHOTT	2003	2018	70
NUREMBERG	2007	2018	2
OBIHIRO	2017	2018	3
OKINAWA	2012	2019	67
ORLANDO	2008	2013	10
OSAKA	1994	2020	1609
OSTEND	1994	2000	3
OUAGADOUGOU	2002	2018	100
PALMA-DE-MALLORCA	2001	2001	1
PANAMA-CITY	2014	2020	75
PAPEETE	2017	2018	2
PARCHIM	2011	2011	1
PARIS	1994	2018	3804
PAYA-LEBAR	1998	1998	1
PENANG	2013	2015	3
PERTH	2013	2017	32
PHILADELPHIA	1999	2019	619
PHNOM-PENH	2014	2014	1
PHOENIX	2001	2018	12
PISA	1998	1998	1
POINTE-A-PITRE	2013	2018	15
POINTE-NOIRE	2014	2018	16
PORLAMAR	2001	2001	2
PORT-HARCOURT	2002	2020	126
PORTLAND	2003	2018	310
PRAGUE	2016	2016	1
PUERTO-PLATA	1995	2002	9
PULA	2018	2018	1
PUNTA-CANA	1995	2021	10
QINGDAO	2012	2020	182
QUEBEC	2015	2015	1
QUITO	1994	2016	74
RECIFE	1994	1995	11
REYKJAVIK	2003	2013	8
RHODES	1996	2002	8
RIGA	2006	2006	1
RIO-DE-JANEIRO	1994	2016	402
RIYADH	1999	2020	254
ROME	2002	2016	8
SAINT-MARTIN	1994	2018	114
SALVADOR	2013	2015	26
SALZBURG	1996	1996	1
SAN-DIEGO	2017	2020	71
SAN-FRANCISCO	1994	2019	265
SAN-JOSE	2014	2020	45
SAN-JUAN	1994	1995	28
SAN-SALVADOR	2014	2015	23
SANAA	2000	2004	6
SANTA-MARIA	2004	2004	1
SANTIAGO	1994	2016	20
SANTO-DOMINGO	1996	2016	3
SANYA	2017	2017	2
SAO-PAULO	1994	2016	377

SAPPORO	2014	2020	45
SEATTLE	2013	2021	64
SEOUL	1994	2020	476
SHANGHAI	1997	2020	428
SHANNON	1999	1999	1
SHENYANG	2012	2020	151
SHENZHEN	2014	2020	4
SINGAPORE	1995	2021	325
SOESTERBERG	2014	2014	1
SOKOTO	2018	2018	1
SPANGDAHLEM	2016	2016	1
ST.-JOHN'S	2009	2009	1
STRASBOURG	1997	2001	3
SURABAYA	2013	2019	74
SYDNEY	2013	2020	169
TAHOUA	2018	2018	2
TAIPEI	2012	2019	1944
TAKAMATSU	1998	2018	4
TAMANRASSET	2018	2018	2
TAMPA	2018	2019	5
TASHKENT	2001	2001	1
TEHRAN	1997	2019	412
TEL-AVIV	1994	2018	275
TENERIFE	1995	1998	11
THESSALONIKI	1999	1999	2
TIANJIN	1995	2020	3
TIPPI	2018	2018	1
TOKYO	1995	2020	1577
TORONTO	1994	2021	821
TOULOUSE	1995	2015	18
UNKNOWN	1994	2020	905
URUMQI	2016	2017	9
VANCOUVER	1995	2020	771
VARADERO	2001	2002	6
VIENNA	1995	2017	3318
WAHIAWA	2017	2017	1
WASHINGTON	1994	2021	410
WEST-HAMPTON-BEACH	1998	1999	3
WINDHOEK	1997	2013	1234
WUHAN	2012	2019	52
WUXI	2016	2019	16
XI'AN	2016	2019	12
XIAMEN	2012	2019	133
YAOUNDE	1997	2017	26
YENBO	2018	2018	1
ZHENGZHOU	2012	2018	21
ZHEZKAZGAN	2018	2018	1
ZHUHAI	2014	2016	2
ZURICH	1997	2013	7

Table S3. Validation of the trajectory-derived (TOST) against ozonesonde measurement by decade from the 1980s to 2010s. The validation of SAGE and MLS ozone in the corresponding decade is provided as a comparison. Note that only ozone profiles from 16-26 km are compared in this table.

	1980s		1990s		2000s		2010s	
	TOST	SAGE	TOST	SAGE	TOST	MLS	TOST	MLS
R	0.96	0.96	0.96	0.96	0.96	0.96	0.97	0.97
RMSD (ppb)	449.47	469.31	486.00	455.40	485.18	494.21	448.89	461.02
Bias (ppb)	13.75	-39.73	41.82	27.73	12.37	107.47	0.15	98.77
RD (%)	0.51	-1.48	1.56	1.03	0.50	4.41	0.01	3.96
N	2629	2629	6717	6717	30366	30366	47904	47904

Table S4. Comparisons of data coverage, number of ozonesonde stations and ozonesonde profiles between TOST-v1 and TOST-v2.

Data coverage (%)	90°-60°S	60°-30°S	30°S-30°N	30°-60°N	60°-90°N
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

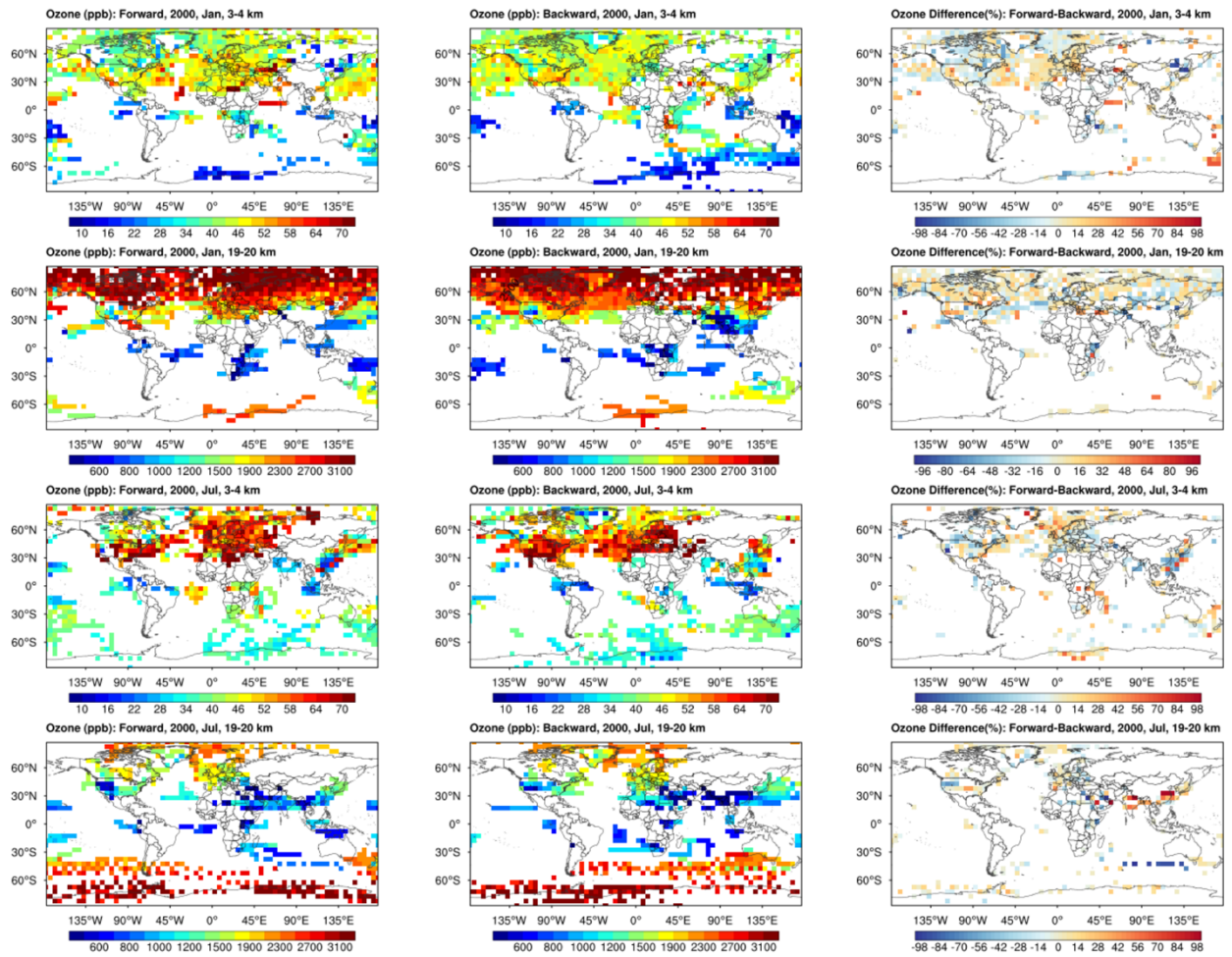


Figure S1. (a-d) Global distribution of monthly mean ozone at 3-4 km and 19-20 km in January and July 2020 from forward trajectories. (e-h) same as (a-d) but for backward trajectories. (i-l) the relative difference in monthly mean ozone between forward and backward trajectories [$100 \times (\text{forward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

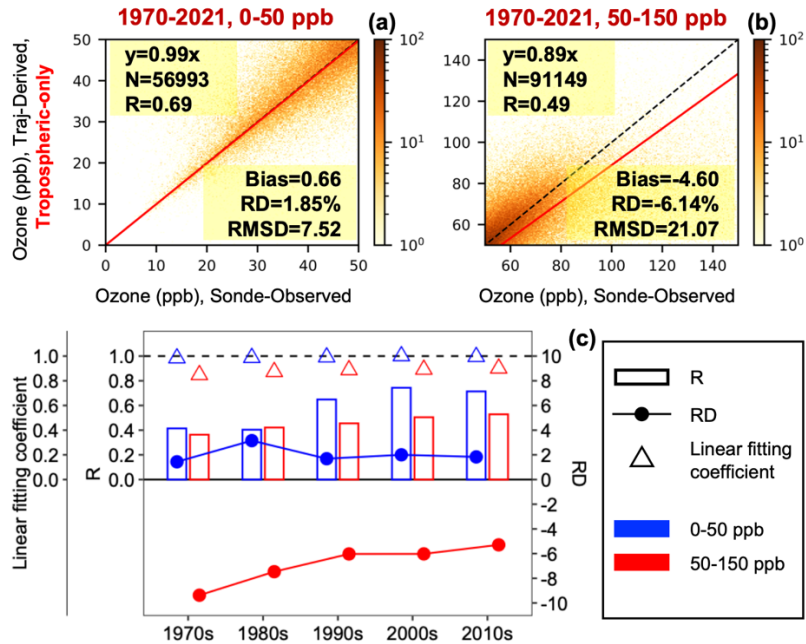


Figure S2. (a-c) Comparison of monthly average tropospheric ozone mixing ratios from ozonesondes (Sonde-Observed) and trajectory-derived TOST data with trajectories from observations only in the troposphere (Traj-Derived, tropospheric-only) for the entire study period of ozone concentration at 0-50 ppb and 50-150 ppb. 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 ppb], RD is the relative difference in % [$100 \times (\text{Traj-Derived ozone} - \text{Sonde-Observed ozone}) / \text{Sonde-Observed ozone}$], and RMSD is the root mean square difference in ppb). 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.

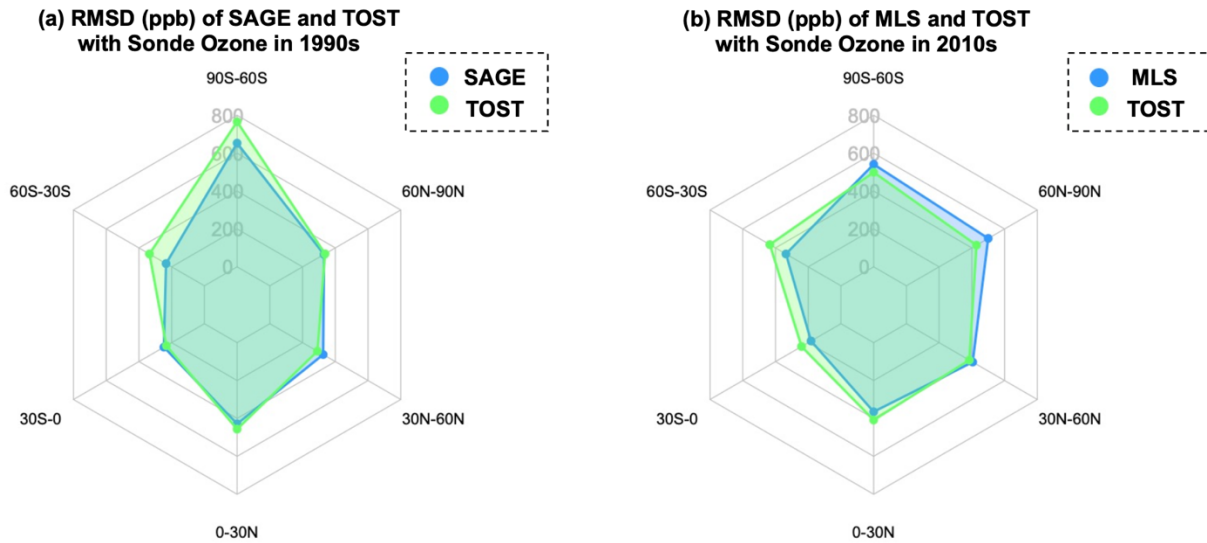


Figure S3. Radar plot for RMSD of TOST Traj-derived and satellite ozone data against ozonesonde measurements at different latitudinal zones. (a) the RMSD for Traj-derived and SAGE ozone in the 1990s; (b) the RMSD for Traj-derived and MLS ozone in the 2010s. The RMSD in a given latitudinal zone is from the monthly ozone mixing ratios between the Traj-derived (or satellite) ozone and ozonesonde measurement in 16-26 km in that latitudinal zone. Traj-derived ozone is without the input of stations being tested.

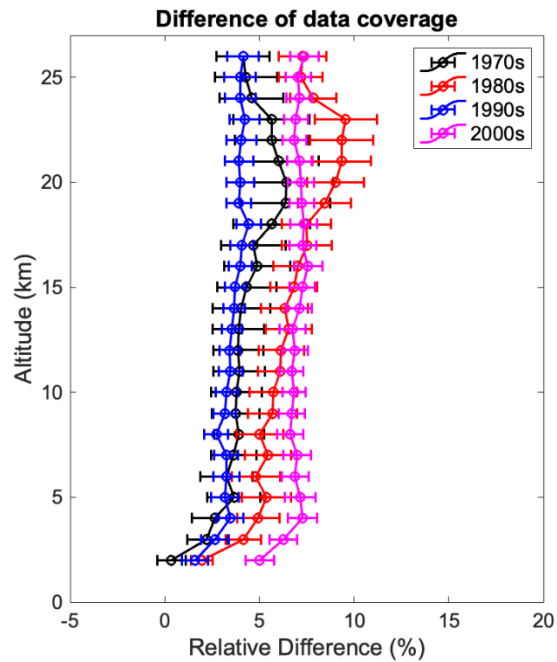


Figure S4. The mean relative difference in data coverage between TOST-v1 and TOST-v2 [$100 \times (\text{TOST-v2 data coverage} - \text{TOST-v1 data coverage}) / \text{TOST-v1 data coverage}$ (in %)] for four decades from 1970-2008. The surface layer (0-1 km) is not compared here due to the topography issue with TOST-v1 (see Figure 9 and Section 3.3 for details).

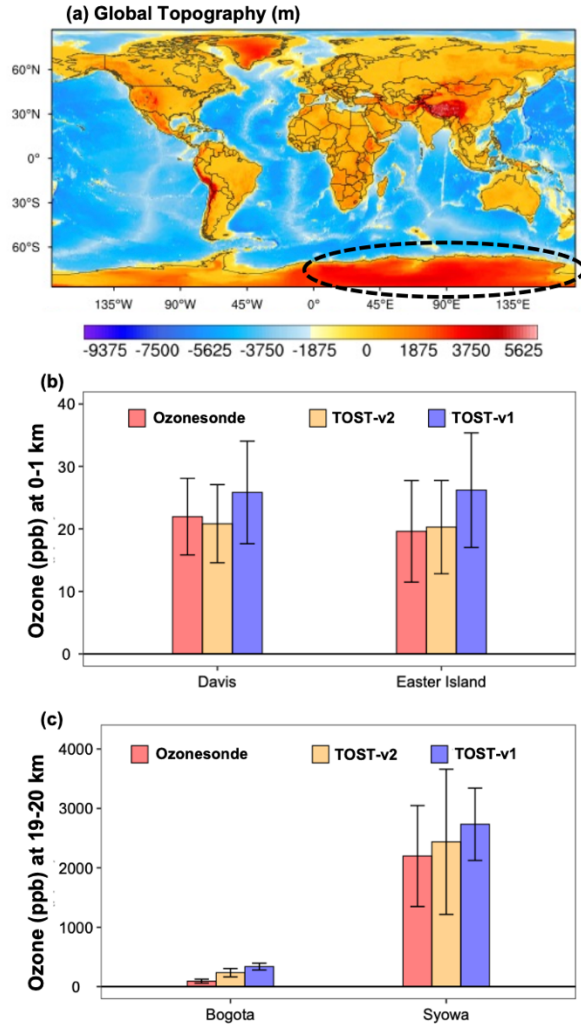


Figure S5. (a) The global topographical map. The dashed circles indicate the regions with large spatial differences between the two versions of TOST data (v1 and v2) at 0-1 km. (b, c) The mean ozone concentrations at 0-1 km and 19-20 km in the 2000s at two stations for ozonesonde, TOST-v2 and TOST-v1. Error bar represents the mean \pm standard deviation of the monthly mean ozone concentrations in this decade.