AIP Advances

ARTICLE COMMENTARY | DECEMBER 08 2022

Comment on "Observation of large and all-season ozone losses over the tropics" [AIP Adv. 12, 075006 (2022)]

Martyn P. Chipperfield ; Andreas Chrysanthou ; Robert Damadeo; Martin Dameris; Sandip S. Dhomse ; Vitali Fioletov ; Stacey M. Frith ; Sophie Godin-Beekmann ; Birgit Hassler; Jane Liu; Rolf Müller ; Irina Petropavlovskikh ; Michelle L. Santee ; Ryan M. Stauffer ; David Tarasick ; Anne M. Thompson ; Mark Weber ; Paul J. Young

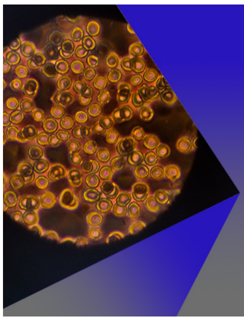
Check for updates

AIP Advances 12, 129102 (2022)
https://doi.org/10.1063/5.0121723

View Online
Export Citation
CrossMark

AIP Publishing

19 July 2023 20:52:45



AIP Advances
Special Topic: Medical Applications of Nanoscience and Nanotechnology
Submit Today!
AIP Publishing

Comment on "Observation of large and all-season ozone losses over the tropics" [AIP Adv. 12, 075006 (2022)]

Cite as: AIP Advances 12, 129102 (2022); doi: 10.1063/5.0121723
Submitted: 19 August 2022 • Accepted: 15 November 2022 •
Published Online: 8 December 2022

View Online Export Citation CrossMark

Martyn P. Chipperfield,[1,2,a)] Andreas Chrysanthou,[1] Robert Damadeo,[3] Martin Dameris,[4] Sandip S. Dhomse,[1,2] Vitali Fioletov,[5] Stacey M. Frith,[6] Sophie Godin-Beekmann,[7] Birgit Hassler,[4] Jane Liu,[8] Rolf Müller,[9] Irina Petropavlovskikh,[10,11] Michelle L. Santee,[12] Ryan M. Stauffer,[13] David Tarasick,[5] Anne M. Thompson,[13,14] Mark Weber,[15] and Paul J. Young[16]

AFFILIATIONS

[1]School of Earth and Environment, University of Leeds, Leeds LS2 9JT, United Kingdom
[2]National Centre for Earth Observation, University of Leeds, Leeds LS2 9JT, United Kingdom
[3]NASA Langley Research Center, Hampton, Virginia 23681, USA
[4]Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre, Oberpfaffenhofen, 82230 Wessling, Germany
[5]Environment and Climate Change Canada, Toronto, Ontario M3H 5T4, Canada
[6]Science Systems and Applications, Inc., Lanham, Maryland 20706, USA
[7]LATMOS, Sorbonne Université, UVSQ, CNRS, 75005 Paris, France
[8]Department of Geography and Planning, University of Toronto, Toronto, Ontario M5S 3G3, Canada
[9]IEK-7, Forschungszentrum Jülich, 52425 Jülich, Germany
[10]Cooperative Institute for Research in Environmental Sciences, CU Boulder, Boulder, Colorado 80309, USA
[11]NOAA GML, Boulder, Colorado 80305, USA
[12]Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California 91109, USA
[13]NASA Goddard Space Flight Center, Greenbelt, Maryland 20771, USA
[14]University of Maryland Baltimore County, Baltimore, Maryland 21228, USA
[15]Institut für Umweltphysik, University of Bremen, 28344 Bremen, Germany
[16]Lancaster Environment Centre, Lancaster University, Lancaster LA1 4YQ, United Kingdom

[a)]Author to whom correspondence should be addressed: M.Chipperfield@leeds.ac.uk

https://doi.org/10.1063/5.0121723

I. INTRODUCTION

Lu (2022) (hereafter L2022) used the Trajectory-mapped Ozonesonde dataset for the Stratosphere and Troposphere (TOST) to argue that there has been very substantial ozone depletion (>80%) in the tropical (30°S–30°N) lower stratosphere (LS) since the 1960s. This was labeled a "*large and all-season ozone hole*." Here, we show that this claim is false due to erroneously large tropical ozone values in the interpolated sparse historical TOST data. In addition, L2022 repeats the suggestion made in a number of the author's earlier papers that cosmic rays are involved in stratospheric ozone depletion. This claim is also not valid; a huge body of work has explained the observed stratospheric ozone depletion through a well-established gas phase and heterogeneous chemistry following the emission of ozone-depleting substances (ODSs) through human activities.

We expand on these points below. In particular, we present a simple analysis of the TOST dataset used by L2022 and show its unsuitability for the application performed. In contrast, we then summarize the much smaller observed variations in ozone in the tropical LS based on many international efforts of data validation and quality assurance, which are not cited by L2022. We then

19 July 2023 20:52:45

© Author(s) 2022

discuss flaws in the cosmic-ray electron-induced mechanism proposed by L2022 as being the main driver of stratospheric ozone losses.

II. OZONE IN THE TROPICAL LOWER STRATOSPHERE

A. TOST data used by L2022

The World Ozone and Ultraviolet Radiation Data Center (WOUDC) website (https://woudc.org/data.php) hosts the TOST data in two formats: (1) ozonesonde data at 1 km intervals in altitude expanded with 4-day forward and backward trajectories to increase spatial sampling (hereafter labeled TOST_RAW) and (2) a smoothed, gap-filled version of TOST_RAW using linear interpolation of maps (hereafter labeled TOST_SM). Methodologies used to construct these two datasets are described by Liu *et al.* (2013a; 2013b). In brief, profile data from about 50 000 ozonesonde profiles (1965–2012) are spatially extended using backward and forward trajectories from meteorological re-analyses to construct a global ozone climatology. Both TOST_RAW and TOST_SM datasets are provided in monthly, annual, and decadal means. L2022 does not specify which TOST dataset is used in that study, but we infer below (see Fig. 3 discussion) that it is TOST_SM. Thus, the ozone depletion diagnosed by L2022 clearly depends entirely on the accuracy and representativeness of the smoothed, gap-filled TOST_SM data in the tropics, especially in the 1960s–1980s.

Figures 1 and 2 show the coverage of the raw trajectory-extended ozonesonde data used to create the TOST datasets. The observed data coverage in the tropics is poor from the 1960s to the 1980s [see also Table I, Fig. 8 of the study by Liu *et al.* (2013a), and Figs. 1 and 12 of the study by Liu *et al.* (2013b)], which affects the validity of the data created by the TOST_SM gap-filling algorithm. Moreover, in the southern tropics (20°S–0°), where L2022 diagnosed the largest ozone depletion, the only observations in

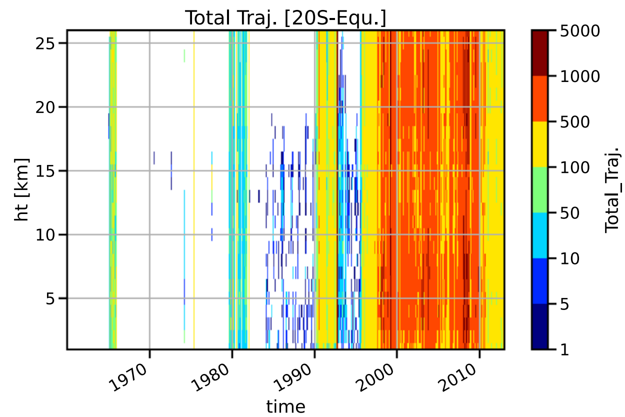


FIG. 2. Number of trajectory-extended ozonesonde observations (per km per month) in the TOST dataset in the latitude range 20°S–0°. Note the very sparse sampling in the 1960s (except 1965), 1970s, and 1980s.

the 1960s are in 1965, and the data coverage is very sparse until the late 1990s when a dedicated tropical ozonesonde network, SHADOZ (Southern Hemisphere Additional Ozonesondes), was initiated (Thompson *et al.*, 2003). While the lack of tropical ozonesonde observations is one severe limitation to diagnosing ozone changes in this region, the problem is further compounded by variable quality control and the instrument response at the different sonde stations between the 1960s and 1980s [e.g., at Pune in the 1980s—see the study by Rohtash *et al.* (2016)]. Reprocessing of most tropical ozonesonde data has greatly enhanced the quality of profiles (Thompson *et al.*, 2017; Sterling *et al.*, 2018), but those profiles were not used in TOST. The limitations imposed by sparse data and large data gaps are discussed in the TOST papers; for each altitude,

19 July 2023 20:52:45

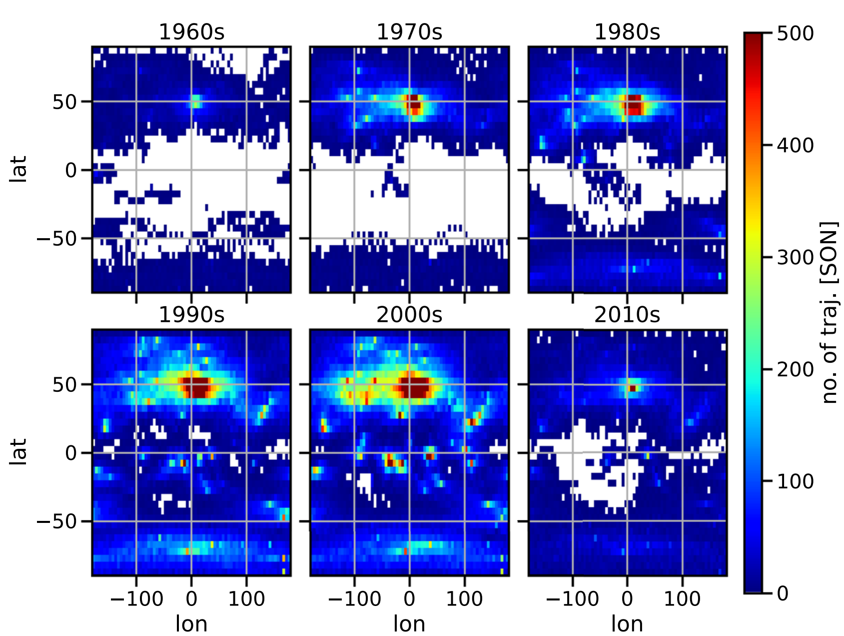


FIG. 1. Number of trajectory-extended ozonesonde observations in the TOST dataset for the September–October–November (SON) season as a function of longitude (°E) and latitude (°N) at a grid resolution of 5° × 5° for six different decades at 15 km. White indicates no observations. Note the lack of tropical observations in the 1960s–1980s, especially south of the equator. Sampling patterns for other seasons are similar.

© Author(s) 2022

TABLE I. Number of ozonesonde profiles in the inner tropics (20° S–20° N) used in the study by Liu *et al.* (2013b) to construct TOST datasets for the 1960s and 1970s.

Station ID	Station name	Latitude (°N)	Longitude (°E)	Period of observation	Number of profiles
108	Canton Island	−2.8	−171.7	1965	31
109	Hilo	19.6	−155.1	1964–1965	17
149	La Paz	−16.5	−68	1965	10
187	Pune	18.5	73.8	1966–1976	135
205	Trivandrum	8.5	76.97	1969–1979	32
203	Ft. Sherman	9.3	−80	1977	16
206	Bombay	19.1	72.8	1968–1969	7
219	Natal	−5.8	−35.2	1979	7
224	Chilca	−12.5	−76.8	1975	3
225	Kourou	5.3	−52.7	1974	3
236	Coolidge Field	17.3	−61.8	1976	7

maps are provided, with the data showing the standard error and the number of observations entering each TOST_RAW mean value. Using this information, the user can judge the quality of individual TOST_RAW averages and filter as required for appropriately robust data.

Figure 3 shows the differences in the TOST_SM and TOST_RAW decadal means for the September–October–November (SON) season, both between 2000 and 1960 and between 2000 and 1980. In addition to the simple difference between the TOST_SM decadal means [Figs. 3(a) and 3(b)], we also calculate decadal mean differences with the TOST_RAW data but filtered to only include grid cells with a minimum number of data points in the calculation of the decadal mean: 10 [Figs. 3(c) and 3(d)] and 100 [Figs. 3(e) and 3(f)]. Comparison of our Fig. 3 with the corresponding SON panels in Figs. 1–3 of L2022 confirms that he analyzed the seasonal mean decadal mean smoothed, gap-filled TOST_SM dataset [i.e., our Figs. 3(a) and 3(b)]. These data show an apparent large depletion of ozone in the TOST_SM data of up to 80% since the 1960s at around 17.5 km altitude and in the latitude range 20°S–0°. However, this change in the smoothed, gap-filled TOST_SM data is an artifact of the sparsely sampled tropical region, as noted above, and there is no robust link to the available observations. In Figs. 3(c)–3(f), the TOST_RAW data do not show such a large ozone decline, and in Figs. 3(c) and 3(e), ozone even increases below 18 km.

The impact of smoothing on the sparse observations in TOST_SM can be seen from the zonal mean climatology. Figure 4 compares decadal mean TOST data, as used by L2022, for both the wider tropics (30°S–30°N) and the 20°S–0° sub-region. The smoothing in TOST_SM not only fills gaps but has also artificially increased the mean ozone values in the tropical lower stratosphere. In the absence of observations in the tropical region, ozone values are interpolated from higher latitudes where ozone concentrations are often higher than those in the tropics. This will lead to an erroneously large diagnosed ozone change. This source of bias is discussed in the first TOST paper (Tarasick *et al.*, 2010). Even small biases, if not addressed, can invalidate trend analyses. In the study by Liu *et al.* (2013b), some simple trend calculations

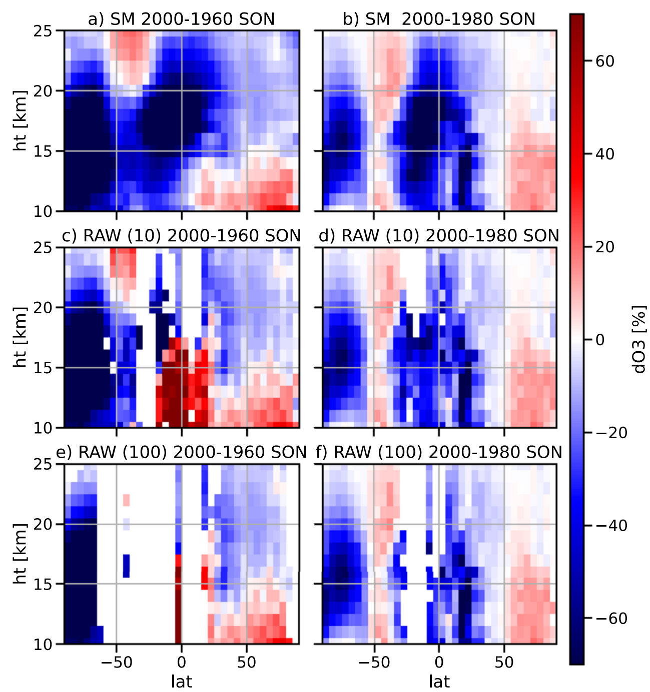


FIG. 3. Difference in decadal mean zonal mean ozone for the SON season from TOST datasets. (a) TOST_SM 2000 minus 1960, (b) TOST_SM 2000 minus 1980, (c) TOST_RAW 2000 minus 1960 restricted to having at least ten trajectory points in each grid cell (1 km × 5° latitude) in the zonal mean decadal mean, (d) TOST_RAW 2000 minus 1980 restricted to having at least ten TOST data points per grid cell, (e) TOST_RAW 2000 minus 1960 restricted to having at least 100 TOST data points per grid cell, and (f) TOST_RAW 2000 minus 1980 restricted to having at least 100 TOST data points. Panel (a) agrees well with the corresponding panels in Figs. 1 and 2 of the study by L2022. Panel (b) agrees well with the corresponding panel in Fig. 3 of the study by L2022.

19 July 2023 20:52:45

© Author(s) 2022

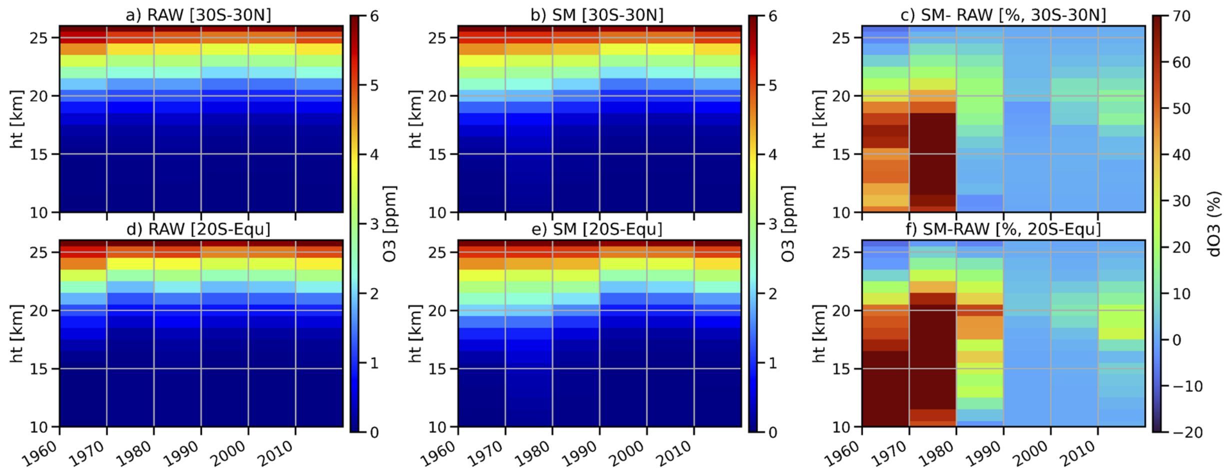


FIG. 4. Seasonal mean (SON) decadal mean O_3 volume mixing ratio (parts per million) from TOST datasets for (top row) 30°S–30°N and (bottom row) 20°S–0° and for (a) and (d) TOST_RAW (using all datapoints without filtering), (b) and (e) smoothed, gap-filled TOST_SM, and (c) and (f) percentage difference TOST_SM minus TOST_RAW. Note how the smoothing increases the ozone mixing ratio in these tropical averages during 1960–1980 compared to the raw data. Note that some decadal means, especially for 20°S–0° before the 1980s, are based on very few data points (see Figs. 1 and 2 and Table I).

are presented using the TOST_RAW dataset and after careful filtering for adequate data density. These, unsurprisingly, agree well with trends calculated from satellite and ground-based total ozone measurements.

L2022 (his Fig. 5) showed that the TOST_SM data strongly overestimate the independent data from GOZCARDS (Global OZone Chemistry And Related trace gas Data records for the Stratosphere; Froidevaux *et al.*, 2015) and BDBP (Binary DataBase of Profiles; Hassler *et al.*, 2008) until after the 1990s (plotted as anomalies on different altitude levels). Given that both of these datasets have more data points contributing to the monthly zonal mean ozone in the 1980s and 1990s (from satellite data), this is further evidence of bias in the TOST_SM data over the tropics for the 1990s, 1980s, and earlier.

B. Observed changes in tropical ozone

In addition to the limitations of the L2022 TOST analysis described above, the results presented by that study are in strong contradiction with the very large amount of research performed over the last several decades. Changes and trends in both tropical total column ozone and vertically resolved tropical LS ozone have been shown and discussed in every Scientific Assessment of Ozone Depletion since the early 1990s (WMO, 1992). L2022 states that "*no O_3 hole over the tropics has been reported*" and cites the most recent assessment (WMO, 2018). However, the reason for this is simple: even with the modified definition of an ozone "hole" as created by L2022 (i.e., a decrease in O_3 by more than 25% relative to levels in the 1960s), there is no evidence for such a decrease in the tropical (20°S–20°N) lower stratosphere (100–70 hPa) from observational records. Measurements of ozone in the tropical lower stratosphere are derived from a multitude of ground- and space-based instruments. Although the number of long-term records in the tropics prior to 2000 is limited, particularly for vertically resolved profiles covering the tropical upper troposphere-lower stratosphere (UTLS) where natural variability is comparatively large, the analyses of these data over the last few decades have painted a consistent picture of how ozone has changed in this region.

Total column measurements from both ground stations and satellites have routinely shown values that are mostly constant, aside from variability (e.g., due to interannual dynamical variations). Tropical trends, using data records starting as far back as the 1970s (Sahai *et al.*, 2000), were first reported as negative with a magnitude less than 2% per decade and having generally larger uncertainties than their determined trend (e.g., Stolarski *et al.*, 1991; WMO, 1992 and references therein). These analyses have been updated and refined over the years with similar results (e.g., Reinsel *et al.*, 1994; 2005; Harris *et al.*, 1997; Fioletov *et al.*, 2002; WMO, 2011; Chehade *et al.*, 2014; Weber *et al.*, 2018 and 2022). While total column ozone may not be the best metric by which changes in the tropical lowermost stratosphere could be assessed, it is a reliable metric for assessing potential changes in surface UV exposure. The total decrease in tropical total column ozone is only about 1% between the 1964–1980 and 2017–2020 averages (Weber *et al.*, 2022), meaning that harmful UV surface exposure remains mostly unchanged in the tropics until today, contrary to the concerns brought forth by L2022. This small change in total ozone is expected from positive trends in the tropical troposphere and negative trends in the tropical LS [see WMO (2018) and references therein].

Vertically resolved observations of the tropical UTLS prior to 2000 came from a limited number of ground-based (mostly ozonesondes) and satellite-based instruments. Trends in this region are difficult to determine because of the large natural variability that can complicate trend analyses (SPARC/IO3C/GAW, 2019 and references therein), the limited spatial coverage of ground stations with well-calibrated ozonesonde records, and the reduced vertical resolution and increased measurement uncertainty of satellite data

19 July 2023 20:52:45

© Author(s) 2022

in the UTLS, depending on the measurement system (Hassler *et al.*, 2014). While there have been some variations in derived trends over the years (e.g., McCormick *et al.*, 1992; Wang *et al.*, 1996; 2002; and Randel and Thompson, 2011), more recent results that take advantage of longer records that have been largely consistent as refinements to past datasets have become increasingly minor, and analysis techniques have evolved (e.g., Harris *et al.*, 2015 and references therein; Steinbrecht *et al.*, 2017; Ball *et al.*, 2018; and SPARC/IO3C/GAW, 2019 and references therein). Although uncertainties are large, tropical lower stratospheric ozone trends derived from both sondes and satellites are negative, with a magnitude of roughly 3%–5% per decade for the period typically ranging from 1984 to 1997, and these trends remain negative thereafter, with a magnitude of roughly 2% per decade for the period 2000–2020 (Thompson *et al.*, 2021; Godin-Beekmann *et al.*, 2022). These values are significantly smaller than the trends that are suggested by Fig. 3 of L2022 (about −20% per decade). The limited amount of measurement data prior to 1980 precludes the evaluation of trends back to 1960, but chemistry–climate models have advanced to replicate the long-term changes seen by observations reasonably well. Figures 3–16 of WMO (2018) show both the observational and model data at 70 hPa/19 km in the tropics, and model simulations show an average total decrease in ozone at this level from 1970 to 2000 of ~6%, where Fig. 2 of L2022 suggests a total decrease of nearly 60%.

III. MECHANISM OF OZONE LOSS

Independent of our assertion that there has not been large ozone depletion over the tropics (Sec. II), many aspects of stratospheric chemistry and dynamics presented by L2022 to explain this apparent depletion are incorrect and require some comment. L2022 did not consider any of the other mechanisms for ozone changes in the tropical UTLS that have been widely discussed in the ozone community (e.g., WMO, 2018; Dietmüller *et al.*, 2021). In particular, circulation changes [strengthening of the Brewer–Dobson circulation (BDC), i.e., increases in tropical upwelling and enhanced mixing between tropics and subtropics (Ball *et al.*, 2020)] related to increasing greenhouse gases (GHGs) are the main drivers of the small ozone decreases in this region (e.g., Eyring *et al.*, 2010; Dhomse *et al.*, 2018; and Dietmüller *et al.*, 2021). It is worth noting that recent analyses of observations support such an explanation for tropical lower stratospheric ozone loss. When a coordinate transformation is performed to look at trends relative to the tropopause height for either ground- (Thompson *et al.*, 2021) or space-based (Bognar *et al.*, 2022) observations, the negative trends just above the tropopause largely disappear, showing how dynamically driven trends in the tropopause region (Pisoft *et al.*, 2021) are primarily responsible for these ozone trends.

A. CRE model

The cosmic-ray-driven electron-induced (CRE) mechanism is not the cause of stratospheric ozone depletion in the polar regions or elsewhere. This mechanism has been thoroughly rebutted in previous comments on the author's papers (e.g., Harris *et al.*, 2002; Patra and Santhanam, 2002; Müller, 2003; Müller and Grooß, 2009 and 2014; Grooß and Müller, 2011 and 2013; and Nuccitelli *et al.*, 2014).

L2022 correlates ozone time series with a proxy representing cosmic-ray-driven ozone losses ("CRE model"), through his Figs. 6(a) and 6(b). The CRE model proxy time series is not clearly distinguishable from the superposition of ODS (ozone-depleting substance) changes [given by effective equivalent stratospheric chlorine (EESC)] and solar irradiance changes (solar activity). Figure 5(a) shows the CRE model proxy from the study by L2022 (digitized from his figure) along with the best fit of the EESC and Mg II UV solar irradiance activity index. The correlation of the SH polar CRE model proxy with the combined polar EESC and solar proxy is 0.96. The correlation is lower (0.74) in the tropical region.

Both EESC and solar activity are well-known drivers of ozone changes (e.g., WMO, 2018). It is known that the cosmic ray flux is modulated by solar activity, meaning that strong solar winds during solar maximum activity shield the earth from cosmic rays such that the cosmic ray flux is anticorrelated with solar flux variations (e.g., Usoskin *et al.*, 2005). This means that the decadal variation in total ozone can be explained by solar irradiance variation. The solar radiative effect on total ozone is well-established and in agreement with model simulations (e.g., Labitzke and van Loon, 1988; Maycock *et al.*, 2018; and Dhomse *et al.*, 2022).

Figures 6(a) and 6(b) in the study by L2022 used total ozone data from different sources (Total Ozone Mapping Spectrometer, Ozone Monitoring Instrument, and Ozone Mapping and Profiler Suite) without removing potential biases and drifts between the datasets, a step that is essential for accurate diagnosis of trends. Figure 5(b) shows the tropical total ozone time series, which is a median of five merged long-term total ozone datasets from the study by Weber *et al.* (2022). The median total ozone is considerably lower in the 1980s than what is shown in Fig. 6(b) of the study by L2022. The combination of EESC and the Mg II index can be reasonably fitted to the median total ozone, and additional factors such as the CRE mechanism are not needed to explain the variability in tropical total ozone. Some additional variability in tropical ozone is related to volcanic eruptions (Agung in 1963; El Chichón in 1982; and Mt. Pinatubo in 1991) that lead to stratospheric ozone depletion in the tropics due to heating and heterogeneous reactions on aerosol particles (e.g., Schoeberl *et al.*, 1993; Kilian *et al.*, 2020). These volcanic influences also coincided with the maximum phases in the solar cycle. Statistical analyses of ozone trends have found aliasing of the solar cycle and volcanic impacts (Chiodo *et al.*, 2014; Damadeo *et al.*, 2014; Dhomse *et al.*, 2016; and Kuchar *et al.*, 2017), and therefore, attribution of ozone trends has to be performed carefully.

For the above-mentioned reasons, a simple correlation of ozone with the CRE model proxy cannot be used as any proof of the cosmic-ray-driven electron-induced (CRE) mechanism.

B. Observations of CFC-12 and the tropopause

To support the explanation of chemical tropical ozone depletion via the CRE mechanism, L2022 used measurements of CFC-12 [his Fig. 4(d)] from the Cryogenic Limb Array Etalon Spectrometer (CLAES, incorrectly referred to as CLEAS throughout L2022) onboard the Upper Atmosphere Research Satellite (UARS). L2022 states that "*the CFC-12 concentration was depleted in the lower stratosphere below 25 km over the tropics (at latitudes 30°S–30°N),*

19 July 2023 20:52:45

© Author(s) 2022

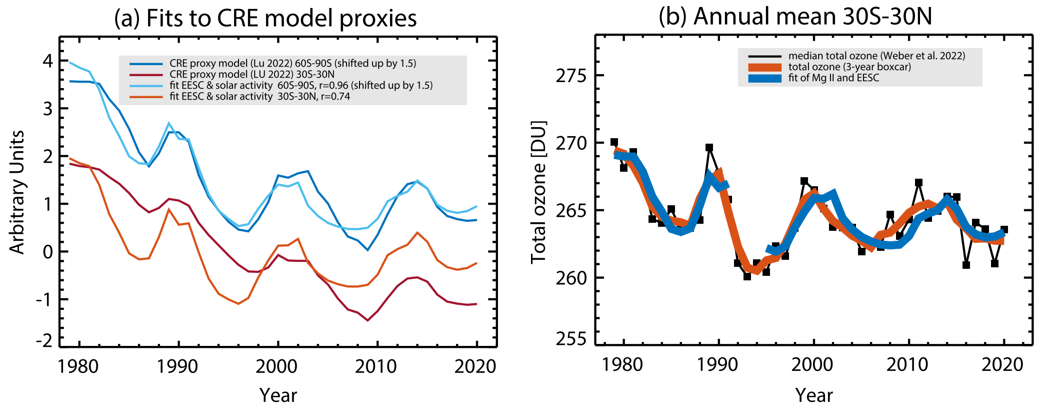


FIG. 5. (a) Normalized CRE model proxy, digitized from Figs. 6(a) and 6(b) in the study by L2022, in the tropics and SH polar region along with the best fits of EESC and solar UV activity proxy (Mg II index). EESC is obtained from the NASA automailer (https://acd-ext.gsfc.nasa.gov/Data_services/automailer/index.html) and assumes a mean age-of-air of 5.5 years and a width of 2.75 years in the polar region and 3.5 and 1.5 years outside the polar regions, e.g., tropics. The Mg II index is obtained from https://www.iup.uni-bremen.de/UVSAT/Datasets/mgii. (b) Median total ozone time series from five merged total ozone datasets (Weber *et al.*, 2022) and the best fit of EESC and solar activity proxy (Mg II index). Due to the major impact of the Mt. Pinatubo volcanic eruption in 1991, the period 1991–1994 was excluded from the fit.

most significant in the zone at 16–20 km and at 20°S–20°N, in which correspondingly the circularly symmetric annual mean tropical O_3 hole is centrally located." This statement is wrong on two counts. First, the tropical tropopause is located at about 17–18 km (e.g., Hoffmann and Spang, 2022), so measurements below the tropopause (i.e., in the troposphere) are not relevant for the stratospheric CRE mechanism. Above 18 km, in the inner tropics (10°S–10°N), CFC-12 is rather constant [about 472 ppt, L2022 Fig. 4(d)], as expected based on its atmospheric lifetime of many decades in this region (Chipperfield *et al.*, 2014). Lower values of CFC-12 toward the mid-latitudes are caused by in-mixing of mid-latitude air into the tropics (e.g., Butchart *et al.*, 2010; Abalos *et al.*, 2015 and 2021; Ploeger *et al.*, 2021; and Poshyvailo-Strube *et al.*, 2022). Second, the CRE mechanism is based on the destruction of CFC-12 (and other species) on atmospheric cloud particles by dissociative electron attachment (DEA) (L2022; Lu and Sanche, 2001). According to the CRE mechanism, the lifetime of CFC-12 in the presence of particles is hours (Lu and Sanche, 2001; Müller, 2003), so the action of the CRE mechanism in the tropics should lead to (patchy) areas with very low CFC-12 presence in association with clouds. Such low presence of CFC-12 in the tropical lower stratosphere is not obvious in either Fig. 4(d) of the study by L2022 or in any other CFC-12 measurement datasets that we are aware of (e.g., Tegtmeier *et al.*, 2016).

L2022 also states that "*significant decompositions of CFCs and N_2O but not CH_4 occur in the lower Antarctic stratosphere during winter*." This statement is in contradiction with the observation that N_2O and CH_4 are well correlated throughout the stratosphere, consistent with a similar (photochemical) loss mechanism for both species in the stratosphere (e.g., Michelsen *et al.*, 1998). Mixing ratios of N_2O are indeed particularly low in the polar regions (especially in winter), which is an effect caused by transport through the Brewer–Dobson circulation (BDC) and the descent of stratospheric air in polar regions. This polar descent is obvious in measurements of both N_2O and CH_4 (e.g., Müller *et al.*, 1999; Ray *et al.*, 2002; and Strahan *et al.*, 2015).

C. Stratospheric circulation

L2022 repeats a statement made similarly in previous papers by the author (e.g., Lu, 2013), namely, "*the transport lag times of CFCs from the troposphere to the lower stratosphere over the Antarctic and the tropics, [. . .] are about 1 year and 10 years, respectively*." This issue has already been debated and shown to be flawed (Müller and Grooß, 2014 and references therein). The erroneous information repeated by L2022 is in contrast to many observations and the theoretical understanding that the stratospheric BDC constitutes young air entering the stratosphere in the tropics. The stratospheric air is transported upward in the (leaky) tropical pipe (e.g., Neu and Plumb, 1999; Butchart, 2014) and then poleward in the lower and upper branches of the BDC, leading to the largest mean ages (of the order of four years) in the polar regions (e.g., Bönisch *et al.*, 2011; Ploeger *et al.*, 2021; and Poshyvailo-Strube *et al.*, 2022). These misconceptions of L2022 might contribute to his incorrect interpretation of tropical lower stratospheric chlorine and ozone chemistry. A full summary and explanations of the atmospheric processes responsible are provided in the scientific ozone assessments (WMO, 2014, Chap. 2; WMO, 2018, Chap. 3).

D. Tropical stratospheric clouds (TSCs)

The stratospheric CRE mechanism relies on the presence of particles in the stratosphere, referred to as TSCs by L2022. However, there is little observational evidence for clouds in the tropical lower stratosphere; while temperatures there are low, the abundance of condensable material (in particular water vapor) is also very low (e.g., Brewer, 1949; Lu *et al.*, 2020). Thus, ice clouds are not frequently observed in the tropics. Peter *et al.* (2003) observed subvisible, large-scale cirrus clouds, referred to as Ultrathin Tropical Tropopause Clouds (UTTCs), a few hundred meters below the tropical cold point tropopause. Zou *et al.* (2022) report that observations indicate the occurrence of ice clouds with cloud-top heights of only 250 m above the first lapse rate tropopause in the tropics. Therefore, any processes related to a tropical CRE mechanism (which is

19 July 2023 20:52:45

© Author(s) 2022

assumed by L2022 to be relevant for altitudes in the tropical stratosphere up to 25 km) could only occur infrequently and close to the tropopause because of the lack of particle surface area density in the tropical lower stratosphere.

IV. SUMMARY

As discussed above, and supported by extensive literature, there is no robust, credible observational evidence for substantial ozone depletion (i.e., an "ozone hole") in the tropics. It is well known that climatological total ozone in the tropics is much lower than that in the mid-latitudes (e.g., Sahai *et al.*, 2000; Weber *et al.*, 2022). Satellite and ozonesonde measurements indicate a 3%–5% per decade decline of tropical lower stratosphere ozone prior to 2000, far smaller than that reported by L2022. The stronger decline reported by L2022 is caused by inappropriate use of the gap-filled version of the TOST ozone dataset, which is based on sparse tropical ozone sondes before the 1990s. This misuse of data (TOST and total column ozone) shows the importance of collaboratively engaging with groups who obtain the measurements and create climatological datasets before performing such analyses.

Furthermore, the study by L2022 has multiple flaws in its discussion of atmospheric chemistry and dynamics, particularly in the proposed, and previously refuted (see Sec. III A), cosmic-ray-driven electron induced (CRE) mechanism. Evidence for the occurrence of tropical stratospheric clouds, as needed for the tropical CRE mechanism, is lacking, nor do CFC-12 observations show signatures of depletion in the tropical lower stratosphere, which could be associated with dissociative electron attachment-induced loss of CFC-12 on particulate matter (i.e., the CRE mechanism). Finally, it is worth reiterating that the CRE mechanism is also not responsible for polar LS ozone depletion. Polar ozone loss can be well explained by the gas phase and heterogeneous chemistry, based on extensive observations and modeling studies documented in many thousands of scientific papers on the topic [e.g., see WMO (2018) and references therein], which is not acknowledged by L2022.

L2022's research paper is a severely flawed one. There is no tropical ozone hole, and the CRE mechanism does not explain observed changes in stratospheric ozone either in the polar regions or in the tropics.

ACKNOWLEDGMENTS

We thank Wolfgang Steinbrecht (DWD) and Paul Newman (NASA) for helpful comments. Work at the Jet Propulsion Laboratory, California Institute of Technology, was carried out under a contract with the National Aeronautics and Space Administration (80NM0018D0004). The work at Leeds and Bremen was supported by the ESA OREGANO project (contract 4000137112/22/I-AG). The work at Leeds was also supported by NERC grant NE/V01163/1.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

All authors contributed to writing this document.

Martyn P. Chipperfield: Conceptualization (lead); Writing – original draft (equal); Writing – review & editing (equal). **Andreas Chrysanthou**: Writing – original draft (equal); Writing – review & editing (equal). **Robert Damadeo**: Writing – original draft (equal); Writing – review & editing (equal). **Martin Dameris**: Writing – original draft (equal); Writing – review & editing (equal). **Sandip S. Dhomse**: Conceptualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Vitali Fioletov**: Writing – original draft (equal); Writing – review & editing (equal). **Stacey M. Frith**: Writing – original draft (equal); Writing – review & editing (equal). **Sophie Godin-Beekmann**: Writing – original draft (equal); Writing – review & editing (equal). **Birgit Hassler**: Writing – original draft (equal); Writing – review & editing (equal). **Jane Liu**: Writing – original draft (equal); Writing – review & editing (equal). **Rolf Müller**: Writing – original draft (equal); Writing – review & editing (equal). **Irina Petropavlovskikh**: Writing – original draft (equal); Writing – review & editing (equal). **Michelle L. Santee**: Writing – original draft (equal); Writing – review & editing (equal). **Ryan M. Stauffer**: Writing – original draft (equal); Writing – review & editing (equal). **David Tarasick**: Writing – original draft (equal); Writing – review & editing (equal). **Anne M. Thompson**: Writing – original draft (equal); Writing – review & editing (equal). **Mark Weber**: Conceptualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Paul J. Young**: Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The TOST data were obtained from https://woudc.org/archive/products/ozone/vertical-ozone-profile/ozonesonde/1.0/tost/ (date of last access: August 16, 2022).

REFERENCES

Abalos, M., Calvo, N., Benito-Barca, S., Garny, H., Hardiman, S. C., Lin, P., Andrews, M. B., Butchart, N., Garcia, R., Orbe, C., Saint-Martin, D., Watanabe, S., and Yoshida, K., "The Brewer–Dobson circulation in CMIP6," Atmos. Chem. Phys. **21**, 13571–13591 (2021).

Abalos, M., Legras, B., Ploeger, F., and Randel, W. J., "Evaluating the advective Brewer-Dobson circulation in three reanalyses for the period 1979–2012," J. Geophys. Res. **120**, 7354–7554, https://doi.org/10.1002/2015jd023182 (2015).

Ball, W. T., Alsing, J., Mortlock, D. J., Staehelin, J., Haigh, J. D., Peter, T., Tummon, F., Stübi, R., Stenke, A., Anderson, J., Bourassa, A., Davis, S. M., Degenstein, D., Frith, S., Froidevaux, L., Roth, C., Sofieva, V., Wang, R., Wild, J., Yu, P., Ziemke, J. R., and Rozanov, E. V., "Evidence for a continuous decline in lower stratospheric ozone offsetting ozone layer recovery," Atmos. Chem. Phys. **18**, 1379–1394 (2018).

Ball, W. T., Chiodo, G., Abalos, M., Alsing, J., and Stenke, A., "Inconsistencies between chemistry–climate models and observed lower stratospheric ozone trends since 1998," Atmos. Chem. Phys. **20**, 9737–9752 (2020).

Bognar, K., Tegtmeier, S., Bourassa, A., Roth, C., Warnock, T., Zawada, D., and Degenstein, D., "Stratospheric ozone trends for 1984–2021 in the SAGE II–OSIRIS–SAGE III/ISS composite dataset," Atmos. Chem. Phys. **22**, 9553–9569 (2022).

19 July 2023 20:52:45

© Author(s) 2022

Bönisch, H., Engel, A., Birner, T., Hoor, P., Tarasick, D. W., and Ray, E. A., "On the structural changes in the Brewer-Dobson circulation after 2000," Atmos. Chem. Phys. **11**, 3937–3948 (2011).

Brewer, A. W., "Evidence for a world circulation provided by the measurements of helium and water vapour distribution in the stratosphere," Q. J. R. Meteorol. Soc. **75**, 351–363 (1949).

Butchart, N., "The Brewer-Dobson circulation," Rev. Geophys. **52**, 157–184, https://doi.org/10.1002/2013rg000448 (2014).

Butchart, N., Cionni, I., Eyring, V., Shepherd, T. G., Waugh, D. W., Akiyoshi, H., Austin, J., Brühl, C., Chipperfield, M. P., Cordero, E., Dameris, M., Deckert, R., Dhomse, S., Frith, S. M., Garcia, R. R., Gettelman, A., Giorgetta, M. A., Kinnison, D. E., Li, F., Mancini, E., McLandress, C., Pawson, S., Pitari, G., Plummer, D. A., Rozanov, E., Sassi, F., Scinocca, J. F., Shibata, K., Steil, B., and Tian, W., "Chemistry–climate model simulations of twenty-first century stratospheric climate and circulation changes," J. Clim. **23**, 5349–5374 (2010).

Chehade, W., Weber, M., and Burrows, J. P., "Total ozone trends and variability during 1979–2012 from merged data sets of various satellites," Atmos. Chem. Phys. **14**, 7059–7074 (2014).

Chiodo, G., Marsh, D. R., Garcia-Herrera, R., Calvo, N., and García, J. A., "On the detection of the solar signal in the tropical stratosphere," Atmos. Chem. Phys. **14**, 5251–5269 (2014).

Chipperfield, M. P., Liang, Q., Strahan, S. E., Morgenstern, O., Dhomse, S. S., Abraham, N. L., Archibald, A. T., Bekki, S., Braesicke, P., Di Genova, G., Fleming, E. L., Hardiman, S. C., Iachetti, D., Jackman, C. H., Kinnison, D. E., Marchand, M., Pitari, G., Pyle, J. A., Rozanov, E., Stenke, A., and Tummon, F., "Multimodel estimates of atmospheric lifetimes of long-lived ozone-depleting substances: Present and future," J. Geophys. Res. **119**, 2555–2573, https://doi.org/10.1002/2013jd021097 (2014).

Damadeo, R. P., Zawodny, J. M., and Thomason, L. W., "Reevaluation of stratospheric ozone trends from SAGE II data using a simultaneous temporal and spatial analysis," Atmos. Chem. Phys. **14**, 13455–13470 (2014).

Dhomse, S. S., Chipperfield, M. P., Damadeo, R. P., Zawodny, J. M., Ball, W. T., Feng, W., Hossaini, R., Mann, G. W., and Haigh, J. D., "On the ambiguous nature of the 11 year solar cycle signal in upper stratospheric ozone," Geophys. Res. Lett. **43**, 7241–7249, https://doi.org/10.1002/2016gl069958 (2016).

Dhomse, S. S., Chipperfield, M. P., Feng, W., Hossaini, R., Mann, G. W., Santee, M. L., and Weber, M., "A single-peak-structured solar cycle signal in stratospheric ozone based on Microwave Limb Sounder observations and model simulations," Atmos. Chem. Phys. **22**, 903–916 (2022).

Dhomse, S. S., Kinnison, D., Chipperfield, M. P., Salawitch, R. J., Cionni, I., Hegglin, M. I., Abraham, N. L., Akiyoshi, H., Archibald, A. T., Bednarz, E. M., Bekki, S., Braesicke, P., Butchart, N., Dameris, M., Deushi, M., Frith, S., Hardiman, S. C., Hassler, B., Horowitz, L. W., Hu, R.-M., Jöckel, P., Josse, B., Kirner, O., Kremser, S., Langematz, U., Lewis, J., Marchand, M., Lin, M., Mancini, E., Marécal, V., Michou, M., Morgenstern, O., O'Connor, F. M., Oman, L., Pitari, G., Plummer, D. A., Pyle, J. A., Revell, L. E., Rozanov, E., Schofield, R., Stenke, A., Stone, K., Sudo, K., Tilmes, S., Visioni, D., Yamashita, Y., and Zeng, G., "Estimates of ozone return dates from Chemistry-Climate Model Initiative simulations," Atmos. Chem. Phys. **18**, 8409–8438 (2018).

Dietmüller, S., Garny, H., Eichinger, R., and Ball, W. T., "Analysis of recent lower-stratospheric ozone trends in chemistry climate models," Atmos. Chem. Phys. **21**, 6811–6837 (2021).

Eyring, V., Cionni, I., Bodeker, G. E., Charlton-Perez, A. J., Kinnison, D. E., Scinocca, J. F., Waugh, D. W., Akiyoshi, H., Bekki, S., Chipperfield, M. P., Dameris, M., Dhomse, S., Frith, S. M., Garny, H., Gettelman, A., Kubin, A., Langematz, U., Mancini, E., Marchand, M., Nakamura, T., Oman, L. D., Pawson, S., Pitari, G., Plummer, D. A., Rozanov, E., Shepherd, T. G., Shibata, K., Tian, W., Braesicke, P., Hardiman, S. C., Lamarque, J. F., Morgenstern, O., Pyle, J. A., Smale, D., and Yamashita, Y., "Multi-model assessment of stratospheric ozone return dates and ozone recovery in CCMVal-2 models," Atmos. Chem. Phys. **10**, 9451–9472 (2010).

Fioletov, V. E., Bodeker, G. E., Miller, A. J., McPeters, R. D., and Stolarski, R., "Global and zonal total ozone variations estimated from ground-based and satellite measurements: 1964–2000," J. Geophys. Res. **107**, 4647, https://doi.org/10.1029/2001jd001350 (2002).

Froidevaux, L., Anderson, J., Wang, H.-J., Fuller, R. A., Schwartz, M. J., Santee, M. L., Livesey, N. J., Pumphrey, H. C., Bernath, P. F., Russell III, J. M., and McCormick, M. P., "Global OZone Chemistry And Related trace gas Data records for the Stratosphere (GOZCARDS): Methodology and sample results with a focus on HCl, H_2O, and O_3," Atmos. Chem. Phys. **15**, 10471–10507 (2015).

Godin-Beekmann, S., Azouz, N., Sofieva, V. F., Hubert, D., Petropavlovskikh, I., Effertz, P., Ancellet, G., Degenstein, D. A., Zawada, D., Froidevaux, L., Frith, S., Wild, J., Davis, S., Steinbrecht, W., Leblanc, T., Querel, R., Tourpali, K., Damadeo, R., Maillard Barras, E., Stübi, R., Vigouroux, C., Arosio, C., Nedoluha, G., Boyd, I., Van Malderen, R., Mahieu, E., Smale, D., and Sussmann, R., "Updated trends of the stratospheric ozone vertical distribution in the 60° S–60° N latitude range based on the LOTUS regression model," Atmos. Chem. Phys. **22**, 11657–11673 (2022).

Grooß, J.-U. and Müller, R., "Do cosmic-ray-driven electron-induced reactions impact stratospheric ozone depletion and global climate change?," Atmos. Environ. **45**, 3508–3514 (2011).

Grooß, J.-U. and Müller, R., "Corrigendum to 'Do cosmic-ray-driven electron-induced reactions impact stratospheric ozone depletion and global climate change?' [Atmos. Environ. 45 (2011), 3408–3514]," Atmos. Environ. **68**, 350 (2013).

Harris, N. R. P., Ancellet, G., Bishop, L., Hofmann, D. J., Kerr, J. B., McPeters, R. D., Prendez, M., Randel, W. J., Staehelin, J., Subbaraya, B. H., Volz-Thomas, A., Zawodny, J., and Zerefos, C. S., "Trends in stratospheric and free tropospheric ozone," J. Geophys. Res. **102**, 1571–1590, https://doi.org/10.1029/96jd02440 (1997).

Harris, N. R. P., Farman, J. C., and Fahey, D. W., "Comment on 'Effects of cosmic rays on atmospheric chlorofluorocarbon dissociation and ozone depletion,'" Phys. Rev. Lett. **89**, 219801 (2002).

Harris, N. R. P., Hassler, B., Tummon, F., Bodeker, G. E., Hubert, D., Petropavlovskikh, I., Steinbrecht, W., Anderson, J., Bhartia, P. K., Boone, C. D., Bourassa, A., Davis, S. M., Degenstein, D., Delcloo, A., Frith, S. M., Froidevaux, L., Godin-Beekmann, S., Jones, N., Kurylo, M. J., Kyrölä, E., Laine, M., Leblanc, S. T., Lambert, J.-C., Liley, B., Mahieu, E., Maycock, A., de Mazière, M., Parrish, A., Querel, R., Rosenlof, K. H., Roth, C., Sioris, C., Staehelin, J., Stolarski, R. S., Stübi, R., Tamminen, J., Vigouroux, C., Walker, K. A., Wang, H. J., Wild, J., and Zawodny, J. M., "Past changes in the vertical distribution of ozone—Part 3: Analysis and interpretation of trends," Atmos. Chem. Phys. **15**, 9965–9982 (2015).

Hassler, B., Bodeker, G. E., and Dameris, M., "Technical Note: A new global database of trace gases and aerosols from multiple sources of high vertical resolution measurements," Atmos. Chem. Phys. **8**, 5403–5421 (2008).

Hassler, B., Petropavlovskikh, I., Staehelin, J., August, T., Bhartia, P. K., Clerbaux, C., Degenstein, D., Mazière, M. D., Dinelli, B. M., Dudhia, A., Dufour, G., Frith, S. M., Froidevaux, L., Godin-Beekmann, S., Granville, J., Harris, N. R. P., Hoppel, K., Hubert, D., Kasai, Y., Kurylo, M. J., Kyrölä, E., Lambert, J.-C., Levelt, P. F., McElroy, C. T., McPeters, R. D., Munro, R., Nakajima, H., Parrish, A., Raspollini, P., Remsberg, E. E., Rosenlof, K. H., Rozanov, A., Sano, T., Sasano, Y., Shiotani, M., Smit, H. G. J., Stiller, G., Tamminen, J., Tarasick, D. W., Urban, J., van der A, R. J., Veefkind, J. P., Vigouroux, C., von Clarmann, T., von Savigny, C., Walker, K. A., Weber, M., Wild, J., and Zawodny, J. M., "Past changes in the vertical distribution of ozone—Part 1: Measurement techniques, uncertainties and availability," Atmos. Meas. Tech. **7**, 1395–1427 (2014).

Hoffmann, L. and Spang, R., "An assessment of tropopause characteristics of the ERA5 and ERA-Interim meteorological reanalyses," Atmos. Chem. Phys. **22**, 4019–4046 (2022).

Kilian, M., Brinkop, S., and Jöckel, P., "Impact of the eruption of Mt Pinatubo on the chemical composition of the stratosphere," Atmos. Chem. Phys. **20**, 11697–11715 (2020).

Kuchar, A., Ball, W. T., Rozanov, E. V., Stenke, A., Revell, L., Miksovsky, J., Pisoft, P., and Peter, T., "On the aliasing of the solar cycle in the

19 July 2023 20:52:45

© Author(s) 2022

lower stratospheric tropical temperature," J. Geophys. Res. **122**, 9076–9093, https://doi.org/10.1002/2017jd026948 (2017).

Labitzke, K. and Van Loon, H., "Associations between the 11-year solar cycle, the QBO and the atmosphere. Part I: The troposphere and stratosphere in the northern hemisphere in winter," J. Atmos. Terr. Phys. **50**, 197–206 (1988).

Liu, G., Liu, J., Tarasick, D. W., Fioletov, V. E., Jin, J. J., Moeini, O., Liu, X., Sioris, C. E., and Osman, M., "A global tropospheric ozone climatology from trajectory-mapped ozone soundings," Atmos. Chem. Phys. **13**, 10659–10675 (2013a).

Liu, J., Tarasick, D. W., Fioletov, V. E., McLinden, C., Zhao, T., Gong, S., Sioris, C., Jin, J. J., Liu, G., and Moeini, O., "A global ozone climatology from ozone soundings via trajectory mapping: A stratospheric perspective," Atmos. Chem. Phys. **13**, 11441–11464 (2013b).

Lu, J., Xie, F., Sun, C., Luo, J., Cai, Q., Zhang, J., Li, J., and Tian, H., "Analysis of factors influencing tropical lower stratospheric water vapor during 1980–2017," npj Clim. Atmos. Sci. **3**, 35 (2020).

Lu, Q.-B., "Cosmic-ray-driven reaction and greenhouse effect of halogenated molecules: Culprits for atmospheric ozone depletion and global climate change," Int. J. Mod. Phys. B **27**, 1350073 (2013).

Lu, Q.-B., "Observation of large and all-season ozone losses over the tropics," AIP Adv. **12**, 075006 (2022).

Lu, Q.-B. and Sanche, L., "Effects of cosmic rays on atmospheric chlorofluorocarbon dissociation and ozone depletion," Phys. Rev. Lett. **87**, 078501 (2001).

Maycock, A. C., Matthes, K., Tegtmeier, S., Schmidt, H., Thiéblemont, R., Hood, L., Akiyoshi, H., Bekki, S., Deushi, M., Jöckel, P., Kirner, O., Kunze, M., Marchand, M., Marsh, D. R., Michou, M., Plummer, D., Revell, L. E., Rozanov, E., Stenke, A., Yamashita, Y., and Yoshida, K., "The representation of solar cycle signals in stratospheric ozone—Part 2: Analysis of global models," Atmos. Chem. Phys. **18**, 11323–11343 (2018).

McCormick, M. P., Veiga, R. E., and Chu, W. P., "Stratospheric ozone profile and total ozone trends derived from the SAGE I and SAGE II data," Geophys. Res. Lett. **19**, 269–272, https://doi.org/10.1029/92gl00187 (1992).

Michelsen, H. A., Manney, G. L., Gunson, M. R., and Zander, R., "Correlations of stratospheric abundances of NO_y, O_3, N_2O, and CH_4 derived from ATMOS measurements," J. Geophys. Res. **103**, 28347–28359, https://doi.org/10.1029/98jd02850 (1998).

Müller, R., "Impact of cosmic rays on stratospheric chlorine chemistry and ozone depletion," Phys. Rev. Lett. **91**, 058502 (2003).

Müller, R. and Grooß, J.-U., "Does cosmic-ray-induced heterogeneous chemistry influence stratospheric polar ozone loss?," Phys. Rev. Lett. **103**, 228501 (2009).

Müller, R. and Grooß, J.-U., "Comment on 'Cosmic-ray-driven reaction and greenhouse effect of halogenated molecules: Culprits for atmospheric ozone depletion and global climate change,'" Int. J. Mod. Phys. B **28**, 1482001 (2014).

Müller, R., Grooß, J.-U., McKenna, D. S., Crutzen, P. J., Brühl, C., Russell III, J. M., Gordley, L. L., Burrows, J. P., and Tuck, A. F., "Chemical ozone loss in the Arctic vortex in the winter 1995–96: HALOE measurements in conjunction with other observations," Ann. Geophys. **17**, 101–114 (1999).

Neu, J. L. and Plumb, R. A., "Age of air in a 'leaky pipe' model of stratospheric transport," J. Geophys. Res. **104**, 19243–19255, https://doi.org/10.1029/1999jd900251 (1999).

Nuccitelli, D., Cowtan, K., Jacobs, P., Richardson, M., Way, R. G., Blackburn, A.-M., Stolpe, M. B., and Cook, J., "Comment on 'Cosmic-ray-driven reaction and greenhouse effect of halogenated molecules: Culprits for atmospheric ozone depletion and global climate change,'" Int. J. Mod. Phys. B **28**, 1482003 (2014).

Patra, P. K. and Santhanam, M. S., "Comment on 'Effects of cosmic rays on atmospheric chlorofluorocarbon dissociation and ozone depletion,'" Phys. Rev. Lett. **89**, 219803 (2002).

Peter, T., Luo, B. P., Wirth, M., Kiemle, C., Flentje, H., Yushkov, V. A., Khattatov, V., Rudakov, V., Thomas, A., Borrmann, S., Toci, G., Mazzinghi, P., Beuermann, J., Schiller, C., Cairo, F., Di Donfrancesco, G., Adriani, A., Volk, C. M., Strom, J., Noone, K., Mitev, V., MacKenzie, R. A., Carslaw, K. S., Trautmann, T., Santacesaria, V., and Stefanutti, L., "Ultrathin Tropical Tropopause Clouds (UTTCs): I. Cloud morphology and occurrence," Atmos. Chem. Phys. **3**, 1083–1091 (2003).

Pisoft, P., Sacha, P., Polvani, L. M., Añel, J. A., de la Torre, L., Eichinger, R., Foelsche, U., Huszar, P., Jacobi, C., Karlicky, J., Kuchar, A., Miksovsky, J., Zak, M., and Rieder, H. E., "Stratospheric contraction caused by increasing greenhouse gases," Environ. Res. Lett. **16**, 064038 (2021).

Ploeger, F., Diallo, M., Charlesworth, E., Konopka, P., Legras, B., Laube, J. C., Grooß, J.-U., Günther, G., Engel, A., and Riese, M., "The stratospheric Brewer–Dobson circulation inferred from age of air in the ERA5 reanalysis," Atmos. Chem. Phys. **21**, 8393–8412 (2021).

Poshyvailo-Strube, L., Müller, R., Fueglistaler, S., Hegglin, M. I., Laube, J. C., Volk, C. M., and Ploeger, F., "How can Brewer–Dobson circulation trends be estimated from changes in stratospheric water vapour and methane?," Atmos. Chem. Phys. **22**, 9895–9914 (2022).

Randel, W. J. and Thompson, A. M., "Interannual variability and trends in tropical ozone derived from SAGE II satellite data and SHADOZ ozonesondes," J. Geophys. Res. **116**, D07303, https://doi.org/10.1029/2010jd015195 (2011).

Ray, E. A., Moore, F., Elkins, J., Hurst, D., Romashkin, P., Dutton, G., and Fahey, D., "Descent and mixing in the 1999–2000 northern polar vortex inferred from in situ tracer measurements," J. Geophys. Res. **107**, 8285, https://doi.org/10.1029/2001jd000961 (2002).

Reinsel, G. C., Miller, A. J., Weatherhead, E. C., Flynn, L. E., Nagatani, R. M., Tiao, G. C., and Wuebbles, D. J., "Trend analysis of total ozone data for turnaround and dynamical contributions," J. Geophys. Res. **110**, D16306, https://doi.org/10.1029/2004jd004662 (2005).

Reinsel, G. C., Tiao, G. C., Wuebbles, D. J., Kerr, J. B., Miller, A. J., Nagatani, R. M., Bishop, L., and Ying, L. H., "Seasonal trend analysis of published ground-based and TOMS total ozone data through 1991," J. Geophys. Res. **99**, 5449–5464, https://doi.org/10.1029/93jd03517 (1994).

Rohtash, Mandal, T. K., Peshin, S. K., and Sharma, S. K., "Study on comparison of Indian ozonesonde data with satellite data," MAPAN **31**, 197–217 (2016).

Sahai, Y., Kirchhoff, V. W. J. H., Lerne, N. M. P., and Casiccia, C., "Total ozone trends in the tropics," J. Geophys. Res. **105**, 19823–19828, https://doi.org/10.1029/2000jd900001 (2000).

Schoeberl, M. R., Bhartia, P. K., Hilsenrath, E., and Torres, O., "Tropical ozone loss following the eruption of Mt. Pinatubo," Geophys. Res. Lett. **20**, 29–32, https://doi.org/10.1029/92gl02637 (1993).

SPARC/IO3C/GAW, SPARC/IO3C/GAW Report on Long-Term Ozone Trends and Uncertainties in the Stratosphere, Petropavlovskikh, I., Godin-Beekmann, S., Hubert, D., Damadeo, R., Hassler, B., and Sofieva, V., SPARC Report No. 9, GAW Report No. 241, WCRP-17/2018, available at www.sparc-climate.org/publications/sparc-reports, 2019.

Steinbrecht, W., Froidevaux, L., Fuller, R., Wang, R., Anderson, J., Roth, C., Bourassa, A., Degenstein, D., Damadeo, R., Zawodny, J., Frith, S., McPeters, R., Bhartia, P., Wild, J., Long, C., Davis, S., Rosenlof, K., Sofieva, V., Walker, K., Rahpoe, N., Rozanov, A., Weber, M., Laeng, A., von Clarmann, T., Stiller, G., Kramarova, N., Godin-Beekmann, S., Leblanc, T., Querel, R., Swart, D., Boyd, I., Hocke, K., Kämpfer, N., Maillard Barras, E., Moreira, L., Nedoluha, G., Vigouroux, C., Blumenstock, T., Schneider, M., García, O., Jones, N., Mahieu, E., Smale, D., Kotkamp, M., Robinson, J., Petropavlovskikh, I., Harris, N., Hassler, B., Hubert, D., and Tummon, F., "An update on ozone profile trends for the period 2000 to 2016," Atmos. Chem. Phys. **17**, 10675–10690 (2017).

Sterling, C. W., Johnson, B. J., Oltmans, S. J., Smit, H. G. J., Jordan, A. F., Cullis, P. D., Hall, E. G., Thompson, A. M., and Witte, J. C., "Homogenizing and estimating the uncertainty in NOAA's long-term vertical ozone profile records measured with the electrochemical concentration cell ozonesonde," Atmos. Meas. Tech. **11**, 3661–3687 (2018).

Stolarski, R. S., Bloomfield, P., McPeters, R. D., and Herman, J. R., "Total Ozone trends deduced from Nimbus 7 Toms data," Geophys. Res. Lett. **18**, 1015–1018, https://doi.org/10.1029/91gl01302 (1991).

Strahan, S. E., Oman, L. D., Douglass, A. R., and Coy, L., "Modulation of Antarctic vortex composition by the quasi-biennial oscillation," Geophys. Res. Lett. **42**, 4216–4223, https://doi.org/10.1002/2015gl063759 (2015).

19 July 2023 20:52:45

© Author(s) 2022

Tarasick, D. W., Jin, J. J., Fioletov, V. E., Liu, G., Thompson, A. M., Oltmans, S. J., Liu, J., Sioris, C. E., Liu, X., Cooper, O. R., Dann, T., and Thouret, V., "High-resolution tropospheric ozone fields for INTEX and ARCTAS from IONS ozonesondes," J. Geophys. Res. **115**, D20301, https://doi.org/10.1029/2009jd012918 (2010).

Tegtmeier, S., Hegglin, M. I., Anderson, J., Funke, B., Gille, J., Jones, A., Smith, L., von Clarmann, T., and Walker, K. A., "The SPARC data initiative: Comparisons of CFC-11, CFC-12, HF and SF6 climatologies from international satellite limb sounders," Earth Syst. Sci. Data **8**, 61–78 (2016).

Thompson, A. M., Stauffer, R. M., Wargan, K., Witte, J. C., Kollonige, D. E., and Ziemke, J. R., "Regional and seasonal trends in tropical ozone from SHADOZ profiles: Reference for models and satellite products," J. Geophys. Res. **126**, e2021JD034691, https://doi.org/10.1029/2021jd034691 (2021).

Thompson, A. M., Witte, J. C., McPeters, R. D., Oltmans, S. J., Schmidlin, F. J., Logan, J. A., Fujiwara, M., Kirchhoff, V. W. J. H., Posny, F., Coetzee, G. J. R., Hoegger, B., Kawakami, S., Ogawa, T., Johnson, B. J., Vömel, H., and Labow, G., "Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology 1. Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements," J. Geophys. Res. **108**, 8238, https://doi.org/10.1029/2001jd000967 (2003).

Thompson, A. M., Witte, J. C., Sterling, C., Jordan, A., Johnson, B. J., Oltmans, S. J., Fujiwara, M., Vömel, H., Allaart, M., Piters, A., Coetzee, G. J. R., Posny, F., Corrales, E., Andres Diaz, J., Félix, C., Komala, N., Lai, N., Maata, M., Mani, F., Zainal, Z., Ogino, S.-Y., Paredes, F., Luiz Bezerra Penha, T., Raimundo da Silva, F., Sallons-Mitro, S., Selkirk, H. B., Schmidlin, F. J., Stuebi, R., and Thiongo, K., "First reprocessing of southern hemisphere additional ozonesondes (SHADOZ) ozone profiles (1998–2016): 2. Comparisons with satellites and ground-based instruments," J. Geophys. Res. **122**, 13000–13025, https://doi.org/10.1002/2017jd027406 (2017).

Usoskin, I. G., Schüssler, M., Solanki, S. K., and Mursula, K., "Solar activity, cosmic rays, and Earth's temperature: A millennium-scale comparison," J. Geophys. Res. **110**, A10102, https://doi.org/10.1029/2004ja010946 (2005).

Wang, H. J., Cunnold, D. M., and Bao, X., "A critical analysis of Stratospheric Aerosol and Gas Experiment ozone trends," J. Geophys. Res. **101**, 12495–12514, https://doi.org/10.1029/96jd00581 (1996).

Wang, H. J., Cunnold, D. M., Thomason, L. W., Zawodny, J. M., and Bodeker, G. E., "Assessment of SAGE version 6.1 ozone data quality," J. Geophys. Res. **107**, 4691, https://doi.org/10.1029/2002JD002418 (2002).

Weber, M., Arosio, C., Coldewey-Egbers, M., Fioletov, V. E., Frith, S. M., Wild, J. D., Tourpali, K., Burrows, J. P., and Loyola, D., "Global total ozone recovery trends attributed to ozone-depleting substance (ODS) changes derived from five merged ozone datasets," Atmos. Chem. Phys. **22**, 6843–6859 (2022).

Weber, M., Coldewey-Egbers, M., Fioletov, V. E., Frith, S. M., Wild, J. D., Burrows, J. P., Long, C. S., and Loyola, D., "Total ozone trends from 1979 to 2016 derived from five merged observational datasets—The emergence into ozone recovery," Atmos. Chem. Phys. **18**, 2097–2117 (2018).

WMO (World Meteorological Organization), "Scientific assessment of stratospheric ozone: 1991," Global Ozone Research and Monitoring Project–Report No. 25, WMO, Geneva, Switzerland, 1992, https://csl.noaa.gov/assessments/ozone/1991/reports.html.

WMO (World Meteorological Organization), "Scientific assessment of ozone depletion: 2010," Global Ozone Research and Monitoring Project–Report No. 52, WMO, Geneva, Switzerland, 2011, https://csl.noaa.gov/assessments/ozone/2010/.

WMO (World Meteorological Organization), "Scientific assessment of ozone depletion: 2014," Global Ozone Research and Monitoring Project–Report No. 55, WMO, Geneva, Switzerland, 2014, https://csl.noaa.gov/assessments/ozone/2014/.

WMO (World Meteorological Organization), "Scientific assessment of ozone depletion: 2018," Global Ozone Research and Monitoring Project–Report No. 58, WMO, Geneva, Switzerland, 2018, https://csl.noaa.gov/assessments/ozone/2018/.

Zou, L., Griessbach, S., Hoffmann, L., and Spang, R., "A global view on stratospheric ice clouds: Assessment of processes related to their occurrence based on satellite observations," Atmos. Chem. Phys. **22**, 6677–6702 (2022).

19 July 2023 20:52:45

© Author(s) 2022