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Qing-Bin Lu[a]

AFFILIATIONS

Department of Physics and Astronomy and Departments of Biology and Chemistry, University of Waterloo, 200 University Ave. West, Waterloo, Ontario N2L 3G1, Canada

[a]Author to whom correspondence should be addressed: qblu@uwaterloo.ca

ABSTRACT

This paper reveals a large and all-season ozone hole in the lower stratosphere over the tropics (30°N–30°S) existing since the 1980s, where an O_3 hole is defined as an area of O_3 loss larger than 25% compared with the undisturbed atmosphere. The depth of this tropical O_3 hole is comparable to that of the well-known springtime Antarctic O_3 hole, whereas its area is about seven times that of the latter. Similar to the Antarctic O_3 hole, approximately 80% of the normal O_3 value is depleted at the center of the tropical O_3 hole. The results strongly indicate that both Antarctic and tropical O_3 holes must arise from an identical physical mechanism, for which the cosmic-ray-driven electron reaction model shows good agreement with observations. The whole-year large tropical O_3 hole could cause a great global concern as it can lead to increases in ground-level ultraviolet radiation and affect 50% of the Earth's surface area, which is home to approximately 50% of the world's population. Moreover, the presence of the tropical and polar O_3 holes is equivalent to the formation of three "temperature holes" observed in the stratosphere. These findings will have significances in understanding planetary physics, ozone depletion, climate change, and human health.

I. INTRODUCTION

The successful execution of the Montreal Protocol and its revisions has led to the declining total level of tropospheric ozone-depleting substances (ODSs) (mainly chlorofluorocarbons—CFCs) since around 1994. Yet, the O_3 hole over the Arctic in 2020 set the biggest record,[1] while the O_3 holes over the Antarctic in 2020 and 2021 were among the largest, deepest, and most persistent ones. As a matter of fact, these observations were not expected from photochemical models[2] but precisely predicted by the cosmic-ray-driven electron-induced-reaction (CRE) model.[3–9] There is also significant research interest in electron-induced reactions of halogen-containing molecules, including CFCs in the gas phase and clusters and on ice surfaces,[10–15] and in the effects of cosmic rays (CRs) on Earth's clouds, climate, and space weather.[16–20]

This study was motivated by the following observations: first, the author[9] has recently shown that the CRE-initiated O_3-depleting reaction completely destroys or even overkills the O_3 layer in the Antarctic lower stratospheric region at altitudes of 13.5–17.5 km corresponding to the CR ionization maximum. Second, CRs have a latitude effect due to the geomagnetic field, which leads to a maximum intensity in the polar stratosphere and a lower intensity by about 50% over the tropics, whereas CFCs have a spatial distribution anti-correlated with the intensity of CRs.[4–8] Third, critically, there exist all-season low-temperature cyclones centered at the altitude of around 15 km over the tropics (30°N–30°S), in which the CR ionization intensity peaks and the temperatures are low at 190–200 K, as will be shown by observed data in this study. The latter are comparable to those in the polar stratospheric vortex over Antarctica in winter and cold enough for forming tropical stratospheric clouds (TSCs), similar to polar stratospheric clouds (PSCs) that are required for surface reactions leading to an O_3 hole.[21] It is important to note that the composition and climate of the lower stratosphere over the tropics are very different from those over the polar regions. This should give rise to a unique active halogen evolution for the tropical lower stratosphere: with the constant presence of TSCs, there are no stable chlorine/bromine reservoirs (HCl and $ClONO_2$) as they are rapidly destroyed on the surfaces of TSCs, while with the constant presence of intense sunlight, no ClO dimer (Cl_2O_2) can exist and O_3-depleting reaction cycles are much more effective. Therefore,

halogen-catalyzed reactions are much more efficient for tropical O_3 destruction, and even a low level of active halogen can cause significant ozone depletion over the tropics in all seasons. The O_3 holes over the Antarctica[22] and Arctic[23] have been well observed, whereas no O_3 hole over the tropics has been reported.[2] Nevertheless, there have been reports of significant ozone losses over the tropics, implying the possibility of a tropical ozone hole. For example, Bian *et al.*[24] and Chen *et al.*[25] reported observations of an "ozone mini hole" over the Tibetan Plateau (29.6° N, 91.1° E) at times with colder winters in the 1990s–2000s, which is located at the edge of the tropics. Polvani *et al.*[26] observed significant O_3 losses at the altitude of 18.5 km or 67/68 hPa over the tropics (30°S–30°N) from 1979 to 1997 from three independent datasets. Newton *et al.*[27] observed ozone-poor air (with very low O_3 concentrations) in the tropical tropopause layer over the west Pacific by aircraft measurements from the three experiment campaigns based in Guam in January–March 2014. Based on the above reasoning, the author hypothesized that there likely exists a large O_3 hole over the tropics, which should be comparable to the well-known Antarctic O_3 hole in depth and much larger than the latter in area. Therefore, this article is devoted to a search for the tropical O_3 hole.

Despite the rationales given above, the search for the tropical O_3 hole is challenging. This is due to some intrinsic challenges. First, unlike polar O_3 holes that are seasonal and mainly appear in spring, a tropical O_3 hole if exists is essentially unchanged across the seasons and is therefore invisible in original observed data. Second, unlike in polar regions, the tropical O_3 layer mainly lies in the middle stratosphere (≥25 km) and only a relatively small percentage (25%–30%) of total O_3 is distributed in the tropical lower stratosphere (at altitudes of 10–25 km). Thus, the amplitude of O_3 depletion in the tropical lower stratosphere is not explicitly reflected in measured total O_3 data. An associated fact that led to no previous observations of any O_3 hole over the tropics is the conventional definition of an O_3 hole, which is defined as an area in which total O_3 values drop below the historical threshold of 220 Dobson Units (DU) (the Antarctic total O_3 value measured by the NASA TOMS satellite in 1979). With this definition, no tropical O_3 holes could be seen in satellite images of total O_3 even if a large percentage up to ~65% of the O_3 amount in the tropical lower stratosphere is depleted. Moreover, much attention was drawn to the studies of O_3 depletion over the polar regions as no O_3 hole was expected to appear over the tropics from current photochemical models. Fortunately, ground-based measurements since the 1960s combined with satellite measurements of O_3, CFCs, and temperature in the stratosphere since 1979 still provide sufficient data to examine the above hypothesis. Here, an O_3 hole is defined as an area with O_3 loss in percent larger than 25%, with respect to the undisturbed O_3 value when there were no significant CFCs in the stratosphere (~ in the 1960s). Despite its difference from the conventional definition of an O_3 hole, this new definition is supported by the observed O_3 loss (~25% in spring) over the Arctic (shown later) where and when an O_3 hole was reported. In other words, an area with O_3 loss by 25% is approximately the threshold of observing an Arctic O_3 hole.

II. DATA AND METHODS

A standard troposphere–stratosphere O_3 climatology in altitude relative to the ground level in the Trajectory-mapped Ozonesonde dataset for the Stratosphere and Troposphere (TOST) obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC), latitude–altitude CFC distribution, time-series total ozone, Ozonesonde and Umkehr datasets, and lower stratospheric temperature climatology from multiple ground-based or satellite measurements are used for this study. The TOST is a global 3D (latitude, longitude, and altitude) climatology of tropospheric and stratospheric O_3, derived from the WOUDC global O_3 sounding record by trajectory mapping, and uses ~77 000 ozonesonde profiles from some 116 stations worldwide since 1965, with details given by Liu *et al.*[28] The long-term zonal mean latitude–altitude distribution of the O_3 climatology measured over the past five decades (1960s–2010s) is shown in Fig. S1 in the supplementary material.

To reveal the tropical O_3 hole, we show the differences in the O_3 climatology in 1970s, 1980s, 1990s, 2000s, and 2010s with respect to those in the 1960s. It is critical to note that the distribution of O_3 over the tropics is highly nonuniform in altitude. Thus, the differences in the zonal mean latitude–altitude distribution of the O_3 climatology do not simply reflect the value of O_3 depletion by a physical mechanism, and there are larger O_3 losses in absolute values at higher altitudes in the stratosphere. Thus, we plot the relative O_3 changes in percent (%) in 1970s–2010s with respect to those in the 1960s to reveal the tropical ozone hole. Furthermore, to confirm the robustness of the observation, we also show the seasonal O_3 changes in percent in 2000s *with respect to those in the 1980s* since the ozone data of the latter should be more reliably measured and more abundant than those in the 1960s. Moreover, the observed results of the present study are also compared with those of the study by Polvani *et al.*[26] using the TOST[28] and other datasets from combined ground-based and satellite measurements.[29,30]

In addition, the relative O_3 change in percent in an O_3 hole is given by the following equation derived from the CRE mechanism:[6–8]

$$\Delta[O_3]/[O_3]_0 = k[C]I^2, \tag{1}$$

where $[O_3]_0$ is the normal O_3 amount when there were no significant stratospheric effects of CFCs (i.e., $[O_3]_0 \approx [O_3]$ in the 1960s), $[C]$ is the equivalent effective chlorine (EECl) in the stratosphere, I is the CR intensity, and k is a fitting constant determined by the best fit to past measured O_3 data. Thus, we also use Eq. (1) to analyze the observed ozone and temperature data in the stratosphere.

III. RESULTS

A. Ozone climatology

The large Antarctic O_3 hole is well known to appear in the spring, while the hypothesized tropical O_3 hole is expected to appear in all four seasons. Thus, we first show the observed results for the Antarctic spring season (September, October, and November—SON) in order to compare the tropic O_3 hole if exists with the well-known Antarctic O_3 hole. As shown in Fig. S2, the thus plotted result shows that from the 1960s to the 1970s, there were almost no ozone holes, but small ozone decreases at 10–20 km over the Antarctica and at 16–25 km over the tropics, and there was a tropospheric–stratospheric region of significant ozone *increasing* at altitudes below 16 km and latitudes of 10°N–40°S, which was likely

caused by volcanic effects. These small O_3 decreases in the Antarctic and tropical lower stratospheres are expected, as it is known that the reaction of stratospheric CFCs remained insignificant up to the 1970s. Despite that there were arguably more sparse measured data available for constructing the TOST data for the 1960s, the result in Fig. S2 shows the validity of the 1960s' TOST data.

As plotted in Fig. 1, in contrast to Fig. S2, the observed results for the 1980s–2010s with respect to those of the 1960s show clearly a deep and large tropical O_3 hole with significant O_3 loss since the 1980s, along with the well-known Antarctic O_3 hole. The Antarctic O_3 hole became the largest and deepest in the 1990s, while the tropic hole continued to grow and deepen in the 1990s and reached its maximum in the 2000s. For both the largest and deepest Antarctic (1990s) and tropical (2000s) O_3 holes, the same maximum (decadal averaged) O_3 loss of ~80% at the hole centers was observed. However, it is worthwhile noting that the geometric area of the tropical O_3 hole at latitudes of 30°S–30°N is ~7.5 times that of the Antarctic hole at 60°–90°S. Both Antarctic and tropical holes in the 2010s were slightly smaller than those in the 2000s.

To show the relative O_3 changes in the Antarctic, tropical, and Arctic stratospheres over the seasons, the O_3 climatology differences in the 2000s with respect to those in the 1960s for SON, DJF (December, January, and February), MAM (March, April, and May), and JJA (June, July, and August) are plotted in Fig. 2. As expected, the polar O_3 holes have strong seasonal variations and mainly appear in their respective springs (SON for the Antarctica and MAM for the Arctic), whereas the tropical O_3 hole shows insensitivity to seasons, existing in all seasons. The Arctic O_3 hole in MAM with a maximum O_3 loss of ~25% is far less and smaller in both depth and area than the Antarctic hole in SON or the tropical hole in any season. This is not unexpected, as the winter stratosphere of the Arctic is well known to be much warmer than that of the Antarctic or tropics and the Arctic vortex is not as well formed and as persistent as in the Antarctic or tropics.

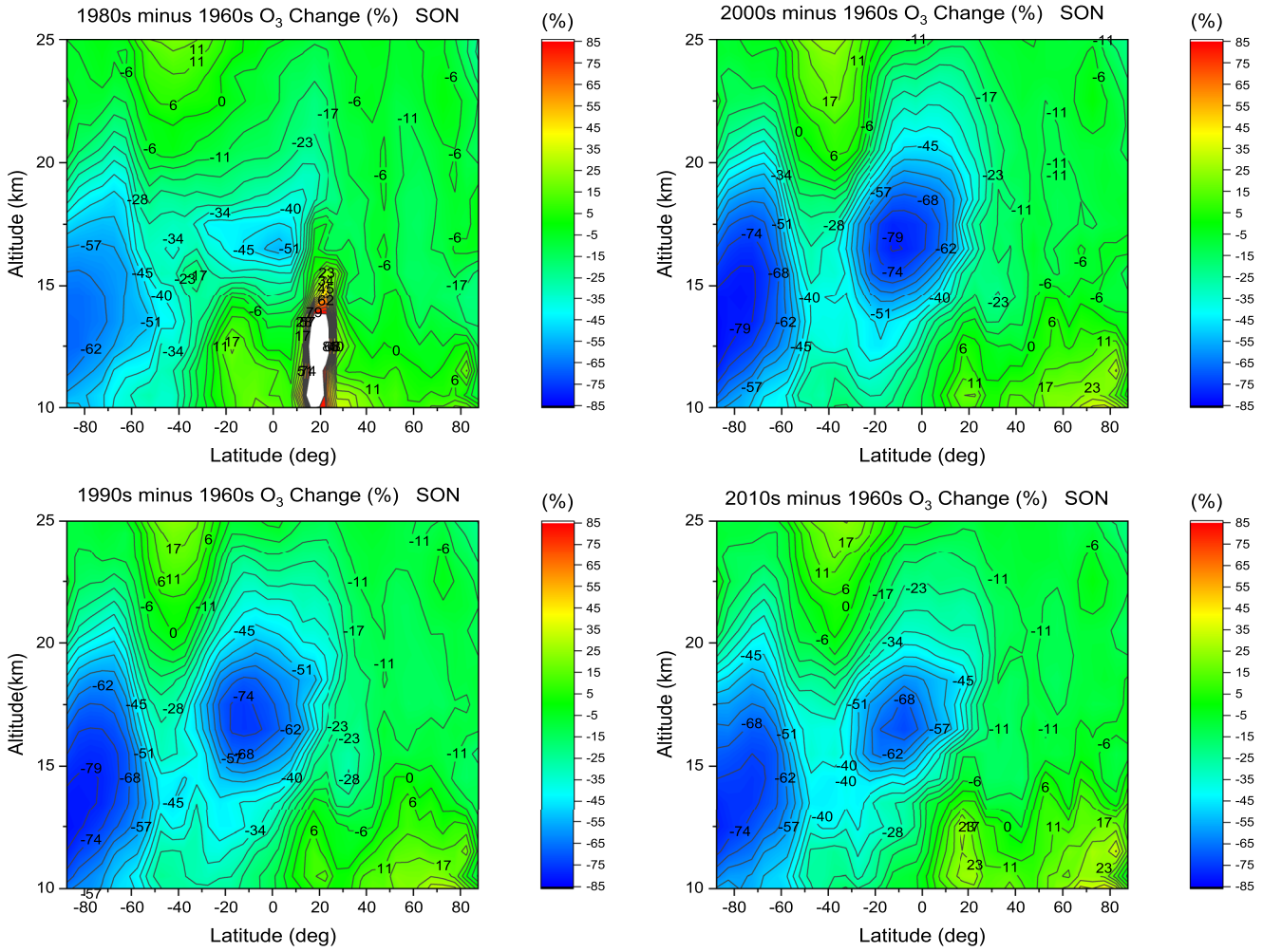

FIG. 1. Changes in % of the O_3 climatology in the season SON of the 1980s–2010s relative to those in the 1960s.

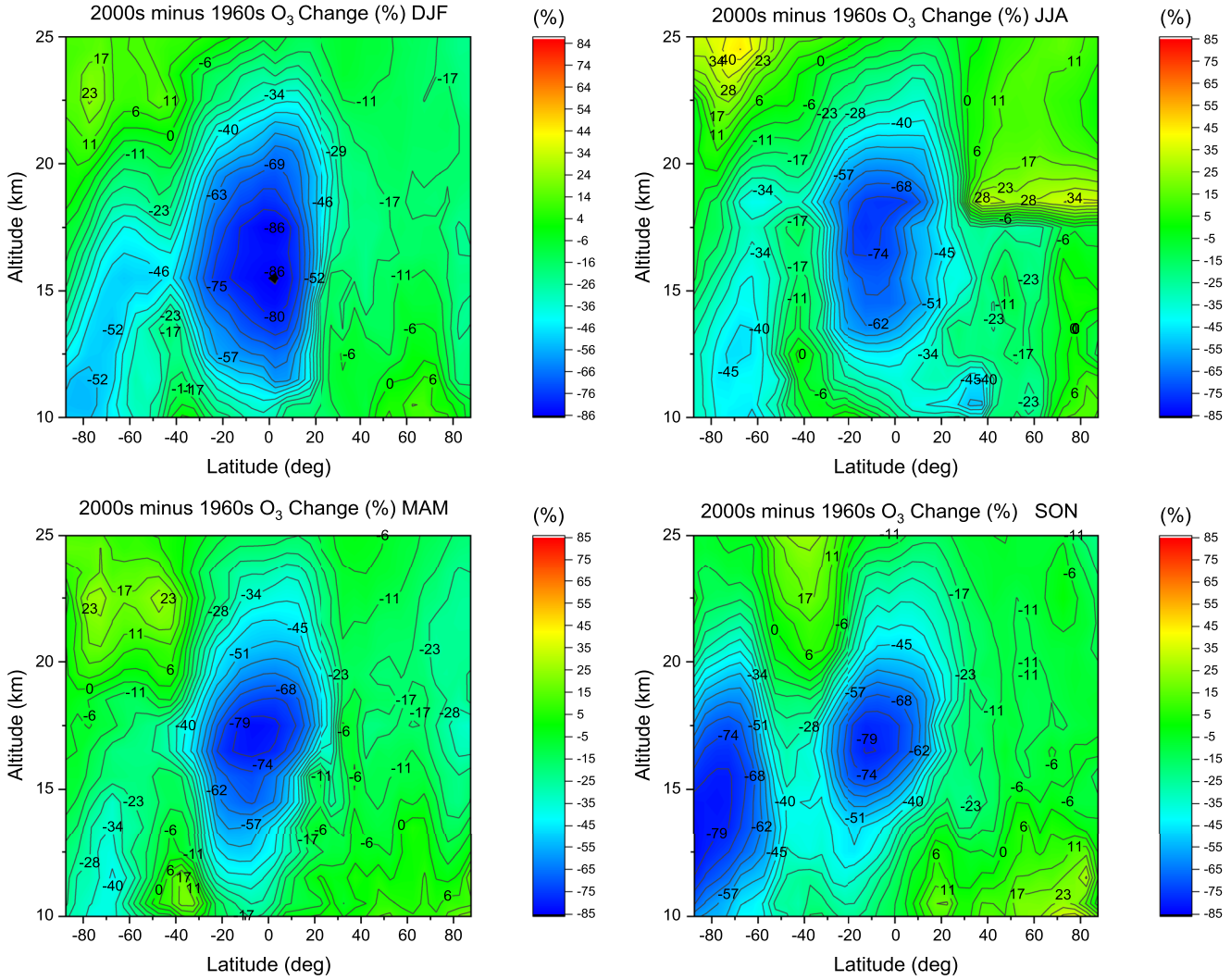

FIG. 2. Changes in % of the O_3 climatology in the seasons of the 2000s relative to those in the 1960s.

Moreover, to show the robustness of the observations in Figs. 1 and 2, the relative seasonal O_3 changes in percent in the 2000s are plotted with respect to those in the 1980s in Fig. 3. The latter solidly confirm the observation of the large and all-season ozone hole over the tropics, which was even slightly deeper than the springtime Antarctic ozone hole in SON (−62% vs −49%). In either the tropical or Antarctic ozone hole, ozone losses in Fig. 3 are smaller than those in Fig. 2. This is reasonable and expected, as ozone depletion was well known to become significant since 1980 (the beginning of the 1980s), and therefore, there was a significant loss of ozone in average from the 1960s to the 1980s. The plotted results in Fig. 3 clearly confirm the validity of the data presented in Figs. 1 and 2 using the 1960s' data as the undisturbed ozone climatology.

To show time-series depths of the decadal mean Antarctic, Arctic, and tropical O_3 holes in SON, MAM, and annual, respectively, averaged O_3 changes in % at the lower stratospheric altitudes of 14–21 km are shown in Figs. 4(a)–4(c), which also include the data for the sum of concentrations of main ODSs (CFCs and CCl_4) measured in the troposphere (no transport lag times considered). It is interesting to show quantitatively that the Antarctic O_3 hole reached its maximum O_3 loss by about 60% in the 1990s, whereas the tropical hole reached its maximum loss by about 56% in the 2000s. By contrast, the Arctic O_3 hole reached its maximum O_3 loss by 20%–25% in the 1990s, less than the half of the maximum O_3 loss in the Antarctic or tropical hole. Both tropical and polar ozone hole trends closely follow that of the total level of ODSs, having significant O_3 decreases in the 1980s and 1990s only.

Similar to our results shown in Fig. 4(b), Polvani *et al.*[26] made an interesting observation. They observed large tropical ozone loss by about 300 ppbv from 1979 to 1997 although they only showed ozone anomalies (not O_3 loss in percentage) relative to the

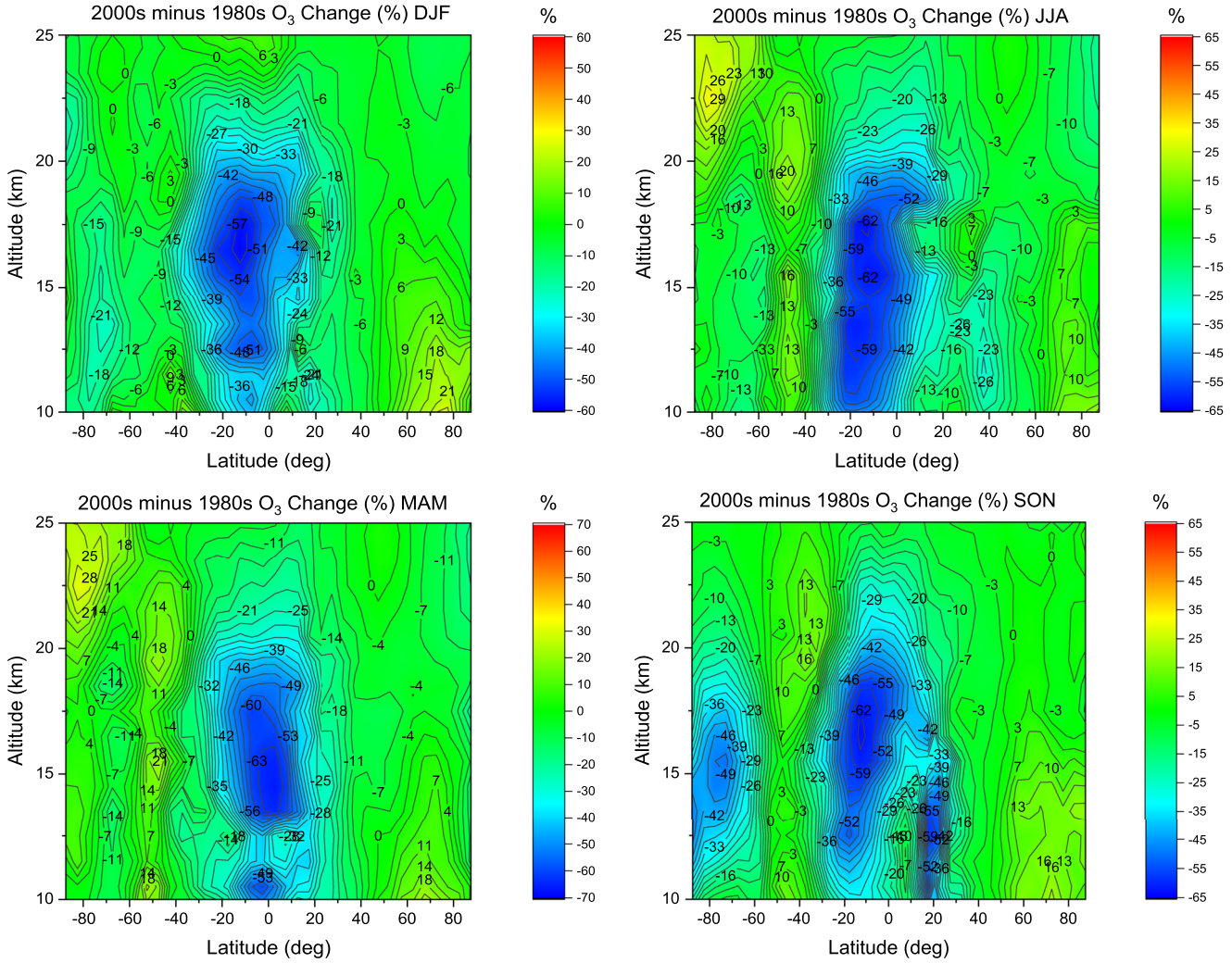

FIG. 3. Changes in % of the O_3 climatology in the seasons of the 2000s relative to those in the 1980s.

post-1998 mean at the single altitude of 18.5 km or at the pressure of 67 or 68 hPa over the tropics (30°S–30°N) from 1979 to 2013. They showed such a large tropical O_3 loss, consistently from three gridded datasets: the TOST for the period 1979–2013 (the same dataset as used for the present study),[28] the binary database of ozone profiles (BDBP) based on a compilation of observations obtained from several sources of high vertical resolution ozone profile measurements for the period 1979–2006,[29] and the NASA's global ozone chemistry and related trace gas data records for the stratosphere (GOZCARDS) merged satellite dataset based on a compilation of data from a number of satellite measurements for the period 1979–2012.[30] For a comparison, the results of the present study and the study by Polvani *et al.*[26] are co-plotted in Figs. 5(a) and 5(b). It is evidently shown that these observed data from multiple datasets of ground-based and combined ground-based and satellite measurements show excellent agreement.

It is also worthwhile noting that the results in Fig. 4(b) indicate that no ozone hole over the tropics would be observed by the conventional definition of an ozone hole, as the total ozone over the tropics was above 220 DU even with a loss by 56% of the column ozone at altitudes of 14–21 km, given that the undisturbed total ozone was about 270 DU over the tropics, of which only ~25% is distributed at 14–21 km. In contrast, with the new definition of an ozone hole in the present study, a pronounced tropical O_3 hole is observed, as clearly seen in Figs. 1–4.

B. CFC-12 distribution and tropical O_3 hole

The author[6–8] has previously shown that significant decompositions of CFCs and N_2O but not CH_4 occur in the lower Antarctic stratosphere during winter. The data provided solid evidence of the CRE reactions of CFCs and N_2O, but not CH_4, in the *winter* polar

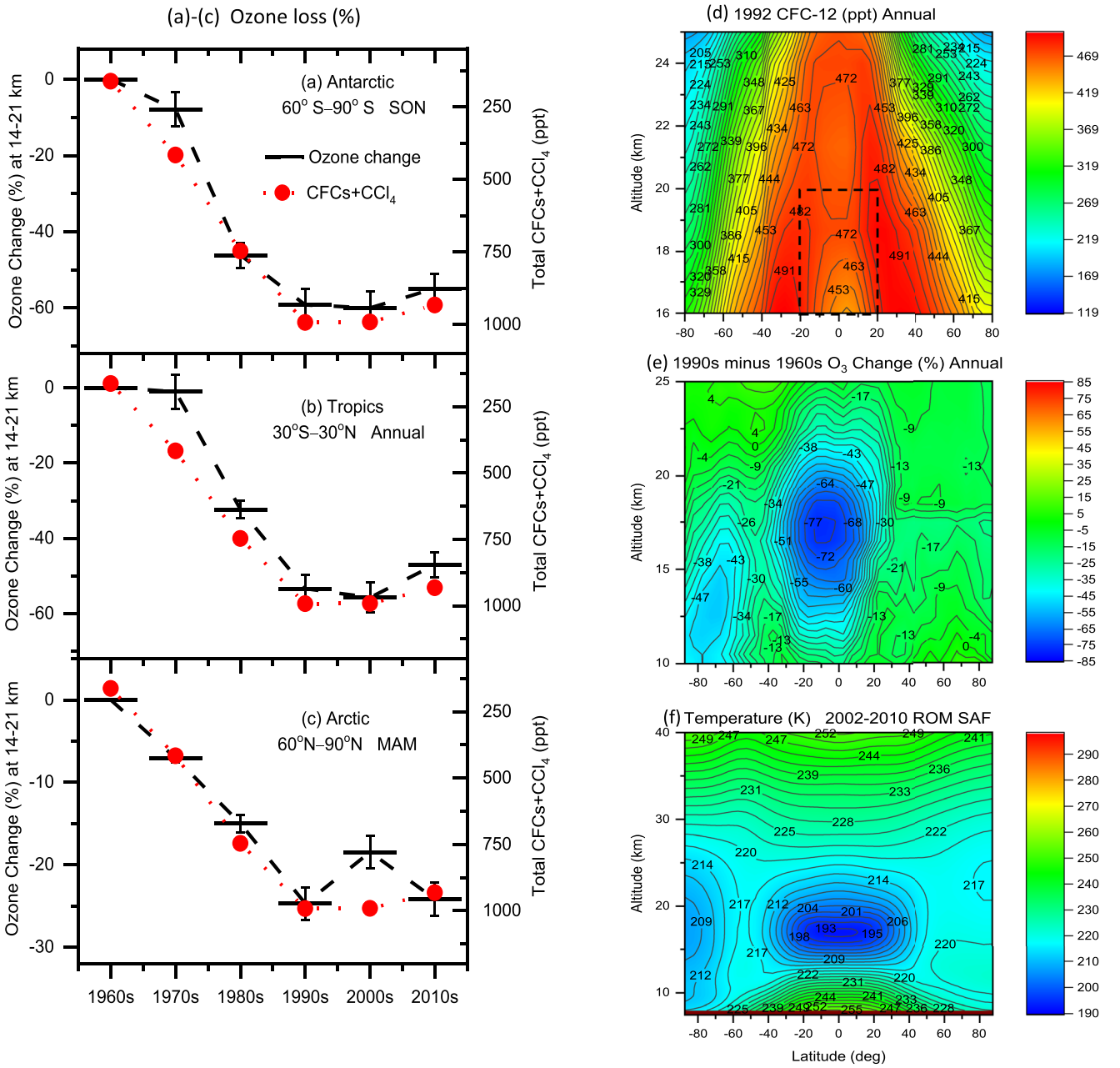

FIG. 4. (a)–(c) Time-series decadal mean O_3 changes in % with respect to those in the 1960s at the lower stratosphere of 14–21 km of the Antarctic in SON, the Arctic in MAM, and the annual tropical (solid lines in black) and sum of measured concentrations of main ODSs (CFCs and CCl_4) (solid circles in red) over the period 1960–2010. (d) Zonal mean latitude–altitude distribution of the CF_2Cl_2 (CFC-12) concentration in 1992 obtained from the NASA UARS's CLEAS dataset, where the dashed square shows the zone of most significant CFC destruction. (e) Difference in % of the annual O_3 climatology in the 1990s with respect to that in the 1960s. (f) Decadal mean zonal mean latitude–altitude distribution of the temperature climatology averaged over seasons in the 2000s.

stratosphere although CH_4 is not even a secure "inert trace gas" because it can also react with radicals generated from the reactions of CFCs or N_2O. Here, the zonal mean latitude–altitude distribution of CF_2Cl_2 (CFC-12) in 1992 from the dataset of the NASA UARS (CLEAS) and the relative difference averaged over the seasons of the O_3 climatology in the 1990s with respect to that in the 1960s are plotted in Figs. 4(d) and 4(e), respectively. The CFC-12 images over the four seasons are shown in Fig. S3. The data clearly show

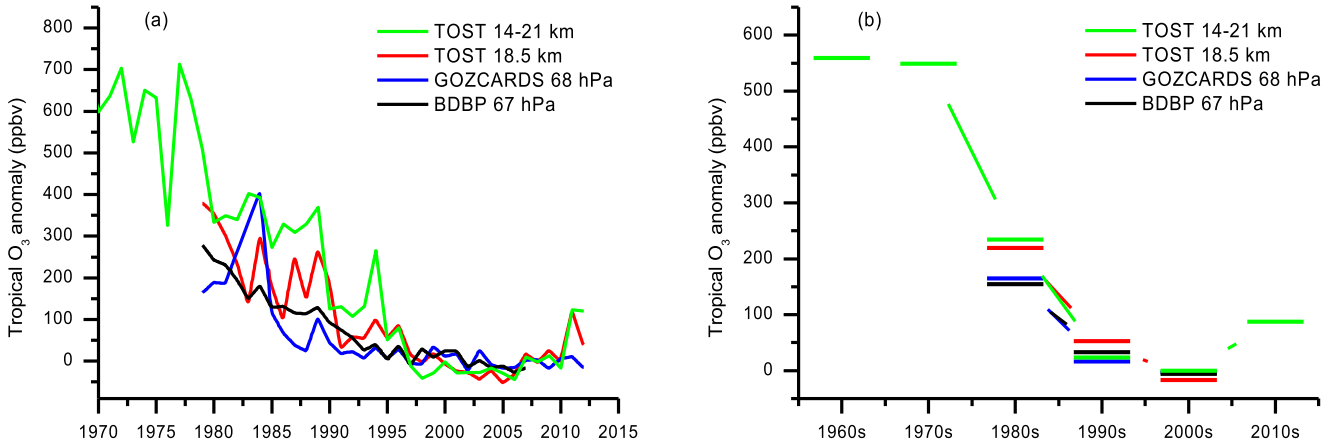

FIG. 5. (a) and (b) Comparison of time-series annual and decadal mean tropical (30°S–30°N) ozone anomalies at the lower stratosphere of 14–21 km relative to the 2000s mean from the TOST[28] (the present study) with those at the altitude of 18.5 km or 67/68 hPa relative to the post-1998 mean from three gridded datasets by Polvani *et al.*:[26] the TOST for the period 1979–2013,[28] the binary database of ozone profiles (BDBP) for the period 1979–2007,[29] and the NASA's global ozone chemistry and related trace gas data records for the stratosphere (GOZCARDS) for the period 1979–2012.[30]

the depletion of CFC-12 in the winter and early spring lower polar stratosphere, as observed previously.[6–8] Most importantly, the data in Fig. 4(d) and Fig. S3 show that in all seasons, the CFC-12 concentration was depleted in the lower stratosphere below 25 km over the tropics (at latitudes of 30°S–30°N), 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 [Fig. 4(e)].

C. Time-series total ozone and Umkehr datasets

Time-series total O_3 data at the springtime Antarctic (60°–90°S) and the annual tropics (30°S–30°N) measured by satellites since 1979 are shown in Figs. 6(a) and 6(b). It is seen that the decreases in absolute total O_3 values over the tropics are smaller than those over the Antarctic. This is expected due to the difference between the undisturbed O_3 distributions over the Antarctic and the tropics (as mentioned above). The 11-year cyclic variation in total O_3 over Antarctica has been well reproduced by the CRE equation previously.[6–8] Here, we use the CRE equation (1) to give a quantitative analysis of observed total O_3 data. Given the observed distributions of the CR ionization rate and CFCs vs latitude and the use of the CR intensity measured at the Antarctic and the concentrations measured at the lower troposphere of CFCs, which are well mixed globally, as inputs to calculate O_3 loss over both the Antarctic and tropics, the reaction constant k in Eq. (1) for the Antarctic is approximately double that for the tropics, i.e., $k_{Tro} \approx \frac{1}{2}k_{Ant}$. Another factor must also be considered: the transport lag times of CFCs from the troposphere to the lower stratosphere over the Antarctic and the tropics, which are about 1 year and 10 years, respectively, determined from the analyses of observed data in terms of the CRE mechanism.[6–9] Moreover, we make a rough assumption that only the O_3 layer in the lower tropical stratosphere below 25 km (i.e., ~25% of the total O_3) is affected by the CRE mechanism. Taking all these factors into account, the CRE-modeled results are shown in Figs. 6(a) and 6(b). It is seen that given the fluctuation level of observed data, the results fitted by Eq. (1) for the Antarctic O_3 hole are excellent. This gives rise to the best fitting constant k_{Ant}, half value of which ($k_{Tro} \approx \frac{1}{2}k_{Ant}$) is used to calculate the O_3 loss over the tropics *with no adjustments*. We notice that the modeled results for tropical O_3 loss are not as good as for the Antarctic. Given several rough approximations made in this modeling, however, the modeled results reproduce the observed O_3 trends surprisingly well and are able to capture the main characteristic of 11-year cyclic variations.

Moreover, the Umkehr dataset for the Mauna Loa (19.5°N, 155.6°W, HI, USA) is shown in Figs. 6(c) and 6(d). For the Antarctic O_3 hole, the 11-year cyclic oscillation of total O_3 was mainly due to the CRE-caused O_3 loss in the stratospheric layers at which the CR ionization intensity was strong, with the most pronounced effect at the Umkehr layer of 63–32 hPa (21–25 km) slightly above the CR-peak layer of 126–63 hPa (15–21 km) in which the CRE mechanism overkilled the O_3 layer.[9] In contrast, no 11-year cyclic variations were observed for O_3 in the upper stratospheric layer where the photochemical mechanism dominates.[9,31] A similar behavior is now observed for O_3 loss over the tropics, as shown in Figs. 6(c) and 6(d). The results in Figs. 4(a), 4(b), and 6(a)–6(d) indicate that O_3 depletion over the Antarctica and the tropics must arise from an identical physical mechanism.

D. Lower stratospheric temperature (LST)

It is well known that O_3 depletion causes stratospheric cooling; the temperature drop in the lower stratosphere is a direct measure of O_3 loss. The seasonal zonal mean latitude–altitude distribution of the temperature at altitudes of 8–40 km for 2002–2010, which is made from the zonally averaged monthly means on a latitude–altitude grid of radio occultation (RO) satellite datasets since 2002, obtained from the high-quality ROM SAF's Climate Data Record (CDR) datasets,[32] is shown in Fig. S4. Remarkably, the data

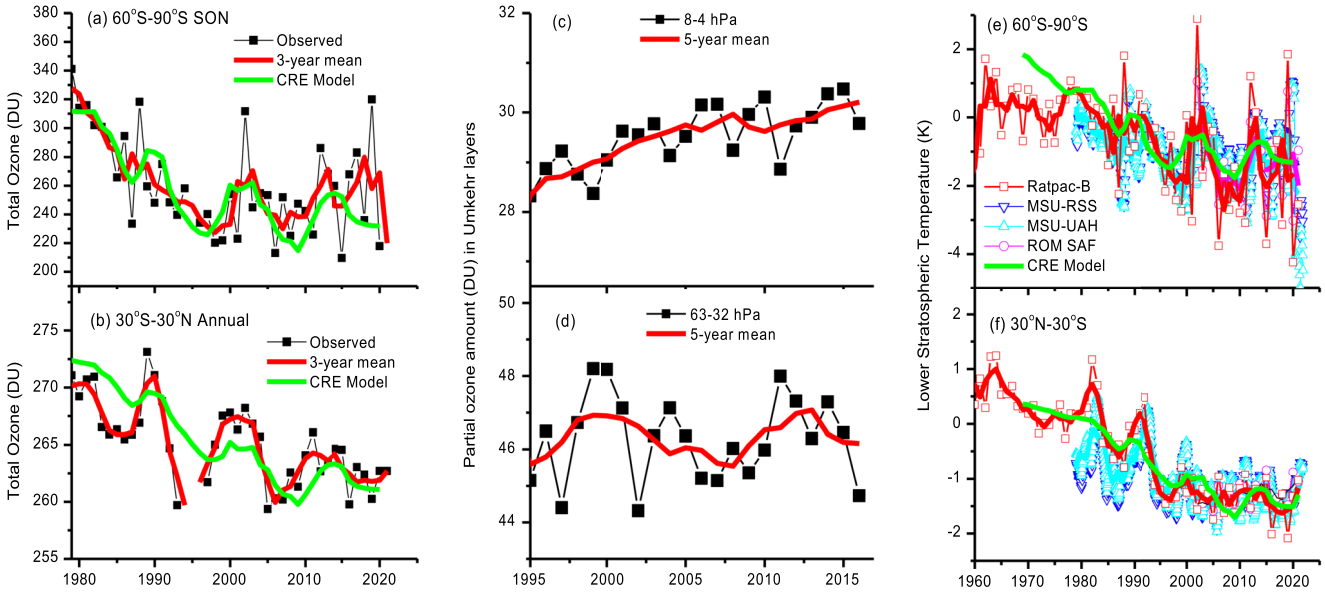

FIG. 6. (a) and (b) Time-series total ozone at the springtime Antarctic (60°–90°S) and the annual tropics (30°S–30°N) measured by NASA TOMS, OMI, and OMPS satellites since 1979. (c) and (d) Time-series Umkehr data of annual mean vertical distribution partial column ozone at the lower and upper stratospheric layers (63–32 and 8–4 hPa) at Mauna Loa (19.5°N, 155.6°W, HI, USA). (e)–(f) Time-series annual mean lower stratospheric temperature anomalies of the Antarctic and the tropics since 1960, obtained from multiple ground- and satellite-based data measurements (Ratpac, MSU-UAH, MSU-RSS, and ROM SAF) and offset to compare with each other. Also shown are the fitted total O_3 and temperature curves given by the CRE equation (see text) and 3(5)-year smoothing (thick solid lines in colors) to observed data (symbols).

show that the temperature in the low-temperature cyclones over the tropics over the seasons is 190–200 K, being only 5–8 K above that in the Antarctic polar vortex in winter (JJA) but 13–16 K below that in the Arctic polar vortex in winter (DJF). This can well explain why the O_3 hole over the Antarctic or tropics is much larger and deeper than the Arctic O_3 hole and why the tropical O_3 hole is constantly formed over the seasons. The annual mean zonal mean latitude–altitude distribution of the temperature averaged over the four seasons is shown in Fig. 4(f). The latter shows a remarkable phenomenon that the annual mean LST over the tropics is the lowest at 193–198 K, compared with 209–217 K over the Antarctic and 217–220 K over the Arctic. As shown in Fig. 4(e), correspondingly, the annual mean O_3 depletion is the largest in the tropical O_3 hole, which is 77% vs 47% for the Antarctic and vs ~10% for the Arctic in the 1990s when the polar holes reached their respective peaks. Consistent with the observation from ozone datasets shown in Figs. 1–5, the stratospheric temperature data in Fig. 4(f) and Fig. S4 exhibit a pronounced "temperature hole" over the tropics, providing strong evidence of a deep and large tropical O_3 hole, along with the well-known Antarctic and Arctic O_3 holes.

Furthermore, the author has well demonstrated that over the Antarctic, the LST regulated by stratospheric cooling due to O_3 loss has pronounced 11-year cyclic variations and can be well reproduced by the CRE equation.[6–9] Here, Figs. 6(e) and 6(f) show time-series temperature anomaly datasets at the lower stratospheres (100–30 hPa) over the Antarctic and the tropics from multiple satellite and ground-based measurements, including NOAA's MSU UAH[33] and RSS[34] satellite datasets, ROM SAF's RO satellite datasets,[32] and NOAA's radiosonde-based Ratpac-B time-series dataset.[35] Note that the LST anomalies in original datasets were normalized to various reference temperatures and had therefore to be offset to compare with each other, but this offsetting has no effects on their long-term trends. These datasets for the Antarctic solidly confirm 11-year cyclic LST variations reported previously,[6–9] while the LST over the tropics shows similar cyclic variations. Moreover, all the datasets show that *significant* O_3 *or LST reductions only occurred in the 1980s and 1990s with no significant trends over the past ~25 years*. This is similar to the observation by Polvani *et al.*[26] and that reported in the newest IPCC AR6.[36] The latter states "most datasets show that lower stratospheric temperatures have stabilized since the mid-1990s with no significant change over the last 20 years" (see p. 2–48, chap. 2 of Ref. 36). Using the same relationship of $k_{Tro} \approx \frac{1}{2} k_{Ant}$ as that used for modeling the O_3 data in Figs. 6(a) and 6(b), we now show that the CRE equation fits to the observed LST data in Figs. 6(e) and 6(f) reasonably well. Again, these results clearly confirm that the Antarctic and tropical O_3 holes must arise from an identical physical mechanism, for which the CRE mechanism has given the best agreement with observations. So far, the CRE mechanism is the only physical mechanism that has predicted and well reproduced the observed 11-year cyclic stratospheric O_3 and LST variations.[4–9]

IV. DISCUSSION

The present finding of a tropical ozone hole is closest to the observation by Polvani *et al.*[26] of large O_3 losses at the altitude of 18.5 km or 67/68 hPa over the tropics (30°S–30°N) from 1979 to 1997 with data from three independent datasets (TOST, BDBP,

and GOZCARDS)[28–30] although they did not reveal the tropical O_3 hole. Polvani *et al.*[26] also showed that global and tropical LST cooling had disappeared since 1997, while the tropical O_3 concentration reached the minimum around 2005. When explaining their observed results, however, Polvani *et al.* argued for the low abundance of active chlorine and hence no local chemical O_3 destruction in the tropical lower stratosphere. Instead, they argued that the observed tropical O_3 and cooling trends were primarily driven by *tropical upwelling* caused by ODSs rather than greenhouse gases (GHGs) (mainly non-halogenated GHGs, as widely believed). As we have noted in the Introduction and will further discuss later, however, even a low level of active halogen can cause significant ozone depletion in the tropical lower stratosphere.

Interestingly, Polvani *et al.*[26] also performed simulations from a chemistry–climate model (CCM) with incrementally added single forcings (sea surface temperatures—SSTs, GHGs, ODSs, volcanic eruptions, and solar variations) to detail the contribution of each forcing to tropical ozone and LSTs. Although their simulated results showed that ODSs were the dominant forcing of tropical ozone loss over GHGs, it must be pointed out that their simulated values of sum ozone loss (−28 ± 13 ppbv per decade; see their Table 1) were about *five times* smaller than their observed results (~−150 ppbv per decade for the 1980s and 1990s), even ignoring that not all individual ensemble members showed statistically significant trends (see their Table 2). Moreover, in contrast to their claim that tropical lower stratospheric ozone would be closely tied to tropical upwelling w^*, their simulated value of the w^* increase by ODSs is *not* dominant but very close to that by GHGs, each force contributing to an increment by ~0.1 km yr^{-1} $decade^{-1}$ at 85 hPa, namely, 0.04 ± 0.09 for SSTs, 0.10 ± 0.11 for SSTs + GHGs, and 0.21 ± 0.11 for SSTs + GHGs + ODSs (see their Table 1). The results of the latter were also consistent with their simulated results of tropical LST trends (see their Fig. 3).

The simulated results of CCMs by Randel and Thompson[37] and others[38] had some differences from but were overall similar to the above-mentioned CCM results by Polvani *et al.*[26] In most simulated results of CCMs, the strength of tropical upwelling was projected to increase from 1960 to 2100 by *~2% per decade* with the largest trends occurring in JJA, corresponding to tropical O_3 reductions at 50 hPa of 0.15–0.35 ppmv (11–25 ppbv *per decade*).[38] This ozone loss trend resulting from CCM simulations is *about* 10 *times less than* the observations by Polvani *et al.*[26] and the present observations shown in Figs. 1–5. More crucially, the observed data have robustly shown that significant tropical ozone loss and LST cooling occurred *in the 1980s and 1990s only*, which is in drastic discrepancy from the simulated results of CCMs. Polvani *et al.*[26] were then led to the open question: How could ODSs affect the stratospheric circulation? They conceded that the underlying mechanism for ODSs being a key forcing for tropical lower stratospheric O_3 and temperature trends remained largely unexplored. Knowing the wide belief in CCMs that the key drivers of tropical upwelling and thus tropical O_3 or LST trends since 1960 are non-halogenated GHGs (mainly CO_2), Polvani *et al.*[26] were forced to suggest that polar O_3 depletion caused by ODSs would cause tropical upwelling and hence large tropical O_3 losses. This explanation cannot be correct either, as the polar O_3 hole is seasonal and appears only in the springtime, whereas the tropical O_3 hole is all-season and has no changes in its central location over the seasons and over the decades since its appearance in the 1980s [Figs. 1–3 and 4(e)].

The present observed results in Figs. 1–6 and Figs. S1–S4 strongly indicate that, like the Antarctic O_3 hole that was once incorrectly explained by the misconceived air transport mechanism ("dynamical theory"), the tropical O_3 hole must not result from changes in normal atmospheric circulation patterns over the tropics since the 1960s or 1970s but result from an identical physical/chemical mechanism to that for the polar O_3 hole. Obviously, the tropical O_3 hole varies closely with the atmospheric level of CFCs [as seen in Fig. 4(b)], so it must originate from a CFC-related mechanism. The postulated stratospheric cooling and tropical upwelling effects of increasing non-halogenated GHGs have disappeared in observed O_3 and temperature data for the Antarctic lower stratosphere[6–8] and for the tropical lower stratosphere.[26] It is obvious that the simulated results from CCMs[26,37,38] do not agree with the observed results shown in Figs. 4(b), 5(a), 5(b), and 6(f), which show that the negative O_3 trends were about 10 times larger (−25 *to* −30% *per decade*) in the 1980s and 1990s and there have been *no significant* O_3 *or LST trends* in the tropical/Antarctic since the mid-1990s. The latter are actually consistent with the observations summarized in the newest IPCC report (Chap. 2).[36] Moreover, the proposed *enhanced* tropical upwelling directly contradicts with the observed CFC depletion in the lower tropical stratosphere [Fig. 4(d)], as increased upward motion would transport CFC-rich air from the troposphere. Indeed, the observed data robustly show no shifts in the positions of both Antarctic and tropical O_3 holes that have constantly been centered in the altitude region corresponding to the CR ionization peak since the 1960s/1980s and circularly symmetric O_3 depletion cyclones are formed with the largest depletion at the centers [Figs. 1–3 and 4(e)]. These major features cannot be explained by tropical upwelling due to non-halogenated GHGs (mainly CO_2) that have kept rising since the industrial revolution starting in 1760. All the observed data strongly indicate that tropical upwelling cannot be the major mechanism for the observed large, deep, and all-season tropical O_3 hole. The simultaneous depletions of both CFCs and O_3 in the lower tropical stratosphere are most likely due to a physical reaction mechanism that occurs locally. For the latter, the CRE mechanism, supported by the observed data in Figs. 1–6 and the substantial datasets obtained from both laboratory and atmospheric measurements,[3–9,39–41] has provided the best and predictive model.

It is well known that the presence of PSCs is crucial for the formation of the Antarctic O_3 hole.[42–45] It was proposed that on the surfaces of PSCs, chlorine reservoir molecules (HCl and $ClONO_2$) are converted into photoactive forms (Cl_2) that can then undergo photolysis to destroy O_3. There are two types of PSCs, namely, Type I and Type II PSC. The composition of Type II PSC is water ice, while Type I PSC is composed of mixtures of nitric acid (HNO_3), water vapor (H_2O), and sulfuric acid (H_2SO_4). The temperatures required for the formation of Type I and II PSCs are 195 and 188 K, respectively. Thus, it is very likely that TSCs, at least Type I PSC-like TSCs, can also form in the tropical lower stratosphere over the seasons due to the observed low temperatures of 190–200 K [Figs. S4 and Fig. 4(f)]. CRs may also play a certain role in forming PSCs and PSC-like TSCs.[16,19] Note that the tropical lower stratosphere is very different from the polar lower stratosphere in both composition and climate. The former is rich in CFCs and

other halogen-containing gases, whereas the latter are composed of inorganic chlorine species and lower-level CFCs. However, the CRE mechanism put forward two decades ago has proposed that O_3-depleting reactions of both CFCs and inorganic halogen species can effectively occur on the surfaces of PSCs.[3–9,39–41] Therefore, there are required and sufficient conditions for O_3-depleting reactions occurring on the surfaces of proposed PSC-like TSCs in the tropical lower stratosphere. As noted in the Introduction, the constant co-presence of low-temperature TSCs and intense sunlight should lead to a unique active halogen evolution in the tropical lower stratosphere, in which halogen-catalyzed reactions are much more efficient for O_3 destruction than those in the polar lower stratosphere.

The tropics (30°N–30°S) constitutes 50% of the Earth's surface area, which is home to about 50% of the world's population. O_3 depletion in the tropics could cause a great global concern. In areas where O_3 depletion is observed to be smaller in absolute O_3 value, UV-B increases are more difficult to detect as the detection can be complicated by changes in cloudiness, local pollution, and other difficulties. However, it is generally agreed that the depletion of the O_3 layer leads to an increase in ground-level UV radiation because ozone is an effective absorber of solar UV radiation. Exposure to enhanced UV-B levels could increase the incidence of skin cancer and cataracts in humans, weaken human immune systems, decrease agricultural productivity, and negatively affect sensitive aquatic organisms and ecosystems.[46] Indeed, there was a report called HIPERION published by the Ecuadorian Space Agency in 2008.[47] The study using ground measurements in Ecuador and satellite data for several countries over 28 years found that the UV radiation reaching equatorial latitudes was far greater than expected, with the UV index as high as 24 in Quito. This Ecuadorian report concluded that O_3 depletion levels over equatorial regions are already endangering large populations in the regions. Further delicate studies of O_3 depletion, UV radiation change, increased cancer risks, and other negative effects on health and ecosystems in the tropical regions will be of great interest and significance.

Another important result is that the global lower stratospheric temperature is essentially governed by the O_3 layer, which is expected as ozone is the main and dominant molecule that absorbs solar radiation in the stratosphere. As a result, the presence of the tropical and polar O_3 holes will play a major role in stratospheric cooling and regulating the global lower stratospheric temperature, as seen previously[6–9,26] and in the results shown in Fig. S4 and Figs. 4(f) and 6(e)–6(f). As seen in Fig. S4 and Fig. 4(f), this is equivalent to the formation of three "temperature holes" in the stratosphere, corresponding to the Antarctic, tropical, and Arctic O_3 holes, respectively. This interesting result will be further explored in a subsequent paper.

V. CONCLUSIONS

In summary, a critical review of the CRE mechanism led to a hypothesis of an ozone hole in the lower stratosphere over the tropics. Substantial datasets of troposphere–stratosphere ozone climatology since the 1960s, latitude–altitude CFC distribution, time-series total ozone, Ozonesonde and Umkehr datasets, and lower stratospheric temperature climatology from multiple ground-based or satellite measurements have provided strong evidence of the existing whole-year ozone hole over the tropics. At the hole center, up to ~80% of the ozone concentrations relative to the normal value in the 1960s is depleted. The averaged depth of the annual tropic ozone hole is comparable to that of the springtime Antarctic ozone hole, while the size of the tropic hole is about seven times that of the Antarctic hole. The annual mean ozone depletion in the lower stratosphere over the tropics is strikingly large, which is 77% vs 47% over the Antarctic and vs ~10% over the Arctic in the 1990s when the polar holes were in their peaks. The observed results strongly indicate that both the Antarctic ozone hole and the tropical ozone hole must arise from an identical physical mechanism and the CRE mechanism has provided the best predictive model that shows good agreement with observations. The tropical lower stratosphere is rich in CFCs and other halogenated ODSs and CR ionization events, and critically, its temperature for all seasons is sufficiently cold for the formation of PSC-like TSCs. Therefore, there is every reason to conclude that the CRE mechanism of ozone-depleting reactions have most likely taken place on the surfaces of TSCs present in the lower stratosphere over the tropics, leading to the formation of the tropical ozone hole in the whole year.

SUPPLEMENTARY MATERIAL

See the supplementary material for Figs. S1–S4.

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AUTHOR DECLARATIONS

Conflict of Interest

The author has no conflicts to disclose.

DATA AVAILABILITY

The data used for this study were obtained from the following sources: the TOST datasets were obtained from the WMO's World Ozone and Ultraviolet Radiation Data Centre (WOUDC) (https://woudc.org/data.php); zonal mean latitude–altitude distribution of CF_2Cl_2 (CFC-12) in 1992 was obtained from the NASA UARS (CLEAS) dataset (https://earthdata.nasa.gov/ or https://uars.gsfc.nasa.gov/Public/Analysis/UARS/urap/home.html); time-series total ozone data at the springtime Antarctic (60°–90°S) and the annual tropics (30°S–30°N) were obtained from NASA TOMS, OMI, and OMPS satellite datasets (https://ozonewatch.gsfc.nasa.gov/meteorology/SH.html or https://woudc.org/data.php); the Umkehr dataset of ozone for the Mauna Loa (19.5°N, 155.6°W, HI, USA) was obtained from NOAA's Global Monitoring

Laboratory (https://gml.noaa.gov/aftp/data/ozwv/) and WOUDC (https://woudc.org/data.php); lower stratospheric temperature data of NOAA's MSU UAH and RSS datasets and Ratpac-B time-series dataset were obtained from NOAA (https://www.ncdc.noaa.gov/climate-monitoring/) and of RO satellite datasets were obtained from the ROM SAF (https://www.romsaf.org/product_archive.php); cosmic ray data were obtained from Ref. 9; and tropospheric data of CFCs and CCl_4 were obtained from the 2021 IPCC AR6 Report.[36]

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