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Analysis of Antarctic ozone trends from 1979 to 2023

Haotian He[1], Martyn P. Chipperfield[2,3], Sandip S. Dhomse[2,3], Wuhu Feng[2,4], Shujie Chang[1,2], Yajuan Li[5,6], Mark Weber[7] and Saffron Heddell[2]

[1]College of Ocean and Meteorology, South China Sea Institute of Marine Meteorology, Laboratory for Coastal Ocean Variation and Disaster Prediction, Key Laboratory of Climate Resources and Environment in Continental Shelf Sea and Deep Ocean (LCRE), Key Laboratory of Space Ocean Remote Sensing and Application, Ministry of Natural Resources, Guangdong Ocean University, Zhanjiang, China

[2]School of Earth and Environment, University of Leeds, Leeds, UK

[3]National Centre for Earth Observation (NCEO), University of Leeds, Leeds, UK

[4]National Centre for Atmospheric Science (NCAS), University of Leeds, Leeds, UK

[5]School of Electronic Engineering, Nanjing Xiaozhuang University, Nanjing, China

[6]Key Laboratory of Middle Atmosphere and Global Environment Observation, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China

[7]Institute of Environmental Physics, University of Bremen, Bremen, Germany

Correspondence to: Shujie Chang (changsj@gdou.edu.cn)

Abstract. Antarctic ozone has shown a sustained recovery since 2000, but levels were distinctly low during 2020-2023, potentially affecting estimates of ozone recovery and long-term trends. To assess the impact of recent low ozone on long-term variability, we analyze total column ozone (TCO) data from World Ozone and Ultraviolet Radiation Centre, multi-sensor reanalysis, and Total Ozone Mapping Spectrometer/Ozone Monitoring Instrument. Ozone fields from TOMCAT, a 3-D chemical transport model, are also used to gain better insight ozone changes. Multiple linear regression (MLR) is applied to estimate ozone trends over Antarctica from 1979 to 2023, incorporating proxies representing key chemical and dynamical processes such as the El Niño-Southern Oscillation and the Brewer-Dobson circulation (BDC).

Our analysis suggests that before 2000, all datasets show significant declines in annual TCO of about 2 and 6 Dobson Units per year (DU/yr) for September and October, respectively. For the 2001-2023 period, the magnitude of the October trend shifted to -1.5 DU/yr. The MLR effectively captures long-term ozone changes as well as unusual dynamical events such as the sudden stratospheric warmings in 2002 and 2019. As dynamical proxies show the largest influence, we use TOMCAT simulations to illustrate the impact of the BDC on the Antarctic ozone. Two sensitivity simulations further demonstrate that the strengthening (weakening) of the circulation leads to high (low) ozone values in spring. These findings suggest that after

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ozone-depleting substances were strictly controlled, dynamical processes have played an increasingly important role in controlling the ozone recovery patterns in Antarctica.

1 Introduction

The discovery of the Antarctic ozone hole in 1985 sparked decades of intensive research on the causes of stratospheric ozone depletion and its broader climate implications (Farman et al., 1985; Solomon et al., 1986). Early scientific studies correctly established link between the decline in the Antarctic ozone and anthropogenic emissions of halogenated ozone-depleting substances, such as trichlorofluoromethane (CFC-11) and dichlorodifluoromethane (CFC-12) (e.g. WMO, 2014, 2018, 2022). These compounds historically contributed to a large portion of the stratospheric chlorine loading. In response to this environmental threat, the 1987 Montreal Protocol and its subsequent amendments were successfully implemented which led to reduction in stratospheric chlorine and bromine loading (WMO, 2022). Beyond their role in ozone depletion, these halogenated substances are also potent greenhouse gases with high global warming potentials, meaning their phase-out has provided substantial co-benefits for climate change mitigation (e.g. Ramanathan et al., 1985; Velders et al., 2007).

These regulatory measures led to stabilising in global ozone trends and initiated a gradual recovery toward pre-1980 conditions (e.g. Dhomse et al., 2018; WMO, 2022). Significant signs of recovery have been confirmed in the upper stratosphere, where ozone increases are attributed to both declining halogens and stratospheric cooling resulting from increased greenhouse gas abundances (Chipperfield et al., 2017; Steinbrecht et al., 2017; Godin-Beekmann et al., 2022). However, the evolution of the lower stratosphere remains a subject of ongoing debate and high uncertainty (Chipperfield et al., 2018). Several observation-based studies suggest a continued decline in lower-stratospheric ozone since 1998, which has been linked to changes in stratospheric dynamics and increased tropical upwelling (Ball et al., 2018; Wargan et al., 2018). In the Antarctic region specifically, while a sustained recovery has been observed since 2000, the period between 2020 and 2023 was characterized by exceptionally large and long-lasting ozone holes (Kessenich et al., 2023). The accurate quantification of how these recent perturbations affect long-term recovery trends remains unclear.

Antarctic ozone variability depends not only on declining halogens but also on a complex interplay of chemical and dynamical processes that vary across multiple timescales. External climate forcing, such as 11-year solar variability and sporadic volcanic eruptions, exerts a significant influence on polar ozone levels (Dhomse et al., 2016, 2022). Increased ultraviolet radiation during solar maxima enhances ozone production in the upper stratosphere (Gray et al., 2010). Major volcanic events, such as Mount Pinatubo in 1991, have caused significant mid-latitude ozone depletion through heterogeneous chemical processing on sulphate aerosols (Aquila et al., 2013; Dhomse et al., 2015). More recently, the 2022 eruption of the Hunga Tonga-Hunga Ha'apai volcano and major wildfires, such as the 2019-2020 Australian fires, have been

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identified as significant perturbations that altered stratospheric aerosol loading and water vapor, potentially delaying the expected recovery of the ozone hole (Khaykin et al., 2020; Peterson et al., 2021; Zhou et al., 2024).

The use of multiple linear regression (MLR) has greatly improved our understanding of these chemical and dynamical processes by allowing for the assessment of various proxies on ozone variability (Dhomse et al., 2006; Steinbrecht et al., 2017; Ball et al., 2019; Weber et al., 2022; Li et al., 2023). Key proxies utilised in such analyses include the quasi-biennial oscillation, El Niño-Southern Oscillation, and the Antarctic Oscillation (Chehade et al., 2014; Weber et al., 2018). Dynamical processes, particularly the Brewer-Dobson circulation, exert a dominant influence on the seasonal and interannual variability of Antarctic ozone (Weber et al., 2011; Butchart, 2014). As ozone-depleting substances are strictly controlled, the relative importance of these dynamical drivers in determining the recovery pattern has increased (Li et al., 2023). However, regression models can be prone to overfitting due to the complex coupling and correlation between different atmospheric proxies (Dhomse et al., 2022; Li et al., 2023).

The aim of this paper is to assess the latest long-term trends of total column ozone over Antarctica using updated observational data from the World Ozone and Ultraviolet Radiation Data Centre, multi-sensor reanalysis fields, and 3-D chemical transport model simulations up to the end of 2023. Given that Antarctic depletion is most pronounced during the Southern Hemisphere Spring, we focus on spring ozone changes to quantify the contributions of key factors to ozone variability. The structure of this manuscript is as follows: Section 2 introduces the ozone datasets and the TOMCAT model configuration, followed by MLR methodology in Sect. 3. Section 4 presents analyse long-term trends and proxy contributions, and Section 5 discusses the results of model sensitivity experiments, followed by a summary and conclusions (Section 6).

2 Ozone datasets

TCO data from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC), the Multi-sensor reanalysis (MSR-2) and Total Ozone Mapping Spectrometer/Ozone Monitoring Instrument (TOMS/OMI) are utilised in this study to assess long-term Antarctic variations. In addition to these observational products, ozone profile datasets simulated by the global three-dimensional chemical transport model, TOMCAT, is also used to provide consistancy for the analysis and to gain better insight into vertical changes. A detailed summary of the data sources and their respective spatio-temporal resolution are shown in Table 1.

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2.1 WOUDC data

The WOUDC ground-based dataset is generated by merging measurements from Dobson and Brewer spectrophotometers along with filtered ozonometers. Zonal mean ozone value is derived using the method of calculating the "climatological" ozone deviation of stations, followed by smoothing or approximation across different stations and months to reduce uncertainty, resulting in 5° zonal averages (Fioletov et al., 2002). To ensure high data quality, the WOUDC records undergo rigorous filtering to eliminate systematic errors or unreliable results. These ground-based observations typically show excellent agreement with satellite-derived data, usually within ±0.5 %, ensuring high consistency between the merged satellite records and the ground-based observations utilised here (Chiou et al., 2014).

2.2 MSR data

The MSR dataset is a comprehensive, revised ozone product constructed by merging measurements from 14 different satellite retrieval instruments. These include the TOMS series (Nimbus-7 and Earth Probe), SBUV (Nimbus-7, NOAA-9, NOAA-11 and NOAA-16), GOME (ERS-2), SCIAMACHY (Envisat), OMI (EOS-Aura), and GOME-2 (Metop-A). Systematic biases in all satellite records are first corrected using independent ground-based total ozone data from the WOUDC, accounting for factors such as solar zenith angle, viewing angle, trend, and effective ozone temperature. The final global ozone dataset is generated using data assimilation techniques based on a chemical transport model driven by meteorological fields from the European Centre for Medium-Range Weather Forecasts (ECMWF) (Van Der A et al., 2010).

2.3 TOMS/OMI data

TOMS continuously measures the sunlit regions of the Earth. The Version 8 total ozone column measurements from TOMS onboard Earth probe (TOMS-EP) were used in this study (Wellemeyer et al., 2004). OMI, onboard the Aura satellite, continues to monitor ozone columns in the atmosphere as a continuation of the TOMS series. OMI measurements provide extremely high spatial resolution and have made significant contributions to the study of stratospheric and tropospheric chemistry (Levelt et al., 2006, 2018).

2.4 TOMCAT model data

TOMCAT is a three-dimensional chemical transport model (CTM) that simulates global data for stratospheric chemical elements and substances such as ozone based on consistent chemical equations (Chipperfield, 2006). The model includes a comprehensive chemical scheme and is driven by ERA-5 reanalysis meteorological fields provided by the ECMWF. Model setup is similar to the one used in Zhou et al. (2024). Time varying solar spectral irradiances are from NRL v2 (Coddington et al., 2016) that are extended until December 2023. Variations in stratospheric aerosol resulting from volcanic eruptions are represented by stratospheric sulphate area density (SAD) fields. These fields are same as used in CMIP6 simulations (until December 2016) and for later period, we use SAGE III measurements based SAD data products (Knepp et al., 2024).

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Implementation of SAD and solar spectral irradiance (SSI) variations is described by Dhomse et al. (2015, 2016). TCO values from the model are calculated by vertical integration of these simulated ozone profiles.

Table 1. Sources and temporal coverage of ozone datasets.

Dataset	Start and end year	Spatio-temporal resolution	Source
WOUDC	1979-2023	Monthly; 5° zonal mean of TCO	http://woudc.org/archive/Projects-Campaigns/ZonalMeans
MSR-2	1979-2023	Monthly; lat * lon = 360 * 720 with TCO	https://www.temis.nl/protocols/O3global.php
TOMS/OMI	2001-2023	Monthly; lat * lon = 180 * 360 with TCO	https://disc.gsfc.nasa.gov/datasets/TOMSEPL3mtoz_008 and https://www.earthdata.nasa.gov/learn/find-data/near-real-time/omi
TOMCAT	1979-2023	Daily; T42 L32	Simulation of Global Ozone Data based on EAR 5 (Chipperfield, 2006).

3 Methods

3.1 Multiple linear regression (MLR)

Ozone trends are generally estimated using MLR, which uses linear trend terms to quantify long term changes due to unknown processes and also includes proxies for the known dynamical and chemical processes. Various methods have been applied to represent trend terms in MLR, such as the piecewise linear trends (PLT), the independent linear trends (ILT), and the equivalent effective stratospheric chlorine (EESC) to account for long term ozone changes due to changes in the ODS (Harris et al., 2008; Nair et al., 2013; Chehade et al., 2014). These trend terms represent the only non-periodic terms of MLR, and aside from ODS, any aperiodic term for ozone change is attributed to this trend term. Previous studies show that stratospheric cooling, due to increases in greenhouse gases, and ODS have comparable effects on ozone increase, while EESC only reflects the chemical influence of ODS. Not all aperiodic changes can be assumed to follow EESC, hence we employ ILT as the trend term of the MLR and ILT and all proxies match the entire period. Using factors such as QBO, 11-

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year solar cycle, ENSO, Antarctic Oscillation (AAO), BDC, stratospheric aerosol optical depth (AOD) (Weber et al., 2018, 2022), the MLR equation used here is shown in Eq. (1):

$$y(t)=a_1\cdot X_1(t_1)+a_2\cdot X_2(t_2)+\alpha_{QBO10}\cdot QBO_{10}(t)+\alpha_{QBO30}\cdot QBO_{30}(t)+\alpha_{AOD}\cdot AOD(t)+\alpha_{sun}\cdot S(t)+\alpha_{BDC}\cdot BDC(t)+\alpha_{ENSO}\cdot E(t)+\alpha_{AAO}\cdot AAO(t) \quad (1)$$

where y(t) is the ozone time series, with t representing the year of the observation, a_1 and a_2 are the linear trend before and after EESC reaches maximum over the Antarctic. t_1 and t_2 indicate that X_1 and X_2 are only different from zero for years t before (1979-2000) and after (2001-2023) the EESC peak, respectively. Analysis of ozone data shows a turning point in the continued decline of Antarctic ozone around 2000, consistent with the EESC calculations showing a maximum in the polar regions at that time (Newman et al., 2006, 2007).

3.2 Proxy for main impact factors

Sources of proxy data are shown in Table 2. To account for the effect of QBO phases and strength on ozone, equatorial zonal winds (10 hPa and 30 hPa) are commonly utilized as indices (Chehade et al., 2014; Li et al., 2020). AOD has been used to represent volcanic aerosol changes following eruptions such as those of El Chichón and Pinatubo volcanoes, which have been shown to affect ozone in the Southern Hemisphere (Sato et al., 1993; Aquila et al., 2013; Dhomse et al., 2015). The AOD proxies are provided as a function of latitude, while we utilised the SH average aerosol data. To account for solar variability, a driver of long-term ozone changes, we use Bremen composite Mg II index (Snow et al., 2014). The BDC is usually expressed as the eddy heat flux (EHF) at 100 hPa, a proxy widely used to access dynamical influence on the interannual ozone variability (e.g. Newman et al., 2001; Dhomse et al., 2006; Weber et al., 2011). ENSO variability is also known to have significant impact on the Southern Hemisphere stratosphere, leading to early or delayed break-up of the polar vortex (e.g. Randel et al., 2002; Camp and Tung, 2007). AAO is closely related to the Antarctic ozone hole (Thompson and Solomon, 2002). Thus, AAO and ENSO are included as proxies for long-term ozone change. In the MLR, AAO and EHF are represented by the mean of the autumn-to-spring accumulation, while other proxies use the monthly mean time series.

Table 2. Sources of impact proxies.

Proxy	Explanatory proxy	url / file
QBO 10 hPa, QBO 30 hPa	Singapore wind speed at 30 hPa and 10 hPa	https://www.iup.uni-bremen.de/OREGANO/proxy/qbo_era5_6S-6N_zonalwind_monthly_mean_1979-2024.txt
AOD(t)	Stratospheric aerosol optical depth at	https://asdc.larc.nasa.gov/project/GloSSAC

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	550 nm	
S(t)	Bremen composite Mg II index	https://www.iup.uni-bremen.de/UVSAT/data/
BDC(t)	Eddy heat flux (100 hPa, 45 S°-75 S°)	https://www.iup.uni-bremen.de/OREGANO/proxy/ehf_m_era5_100hPa_accumulated.txt
E(t)	Multivariate ENSO Index (MEI V2)	https://psl.noaa.gov/data/climateindices/list/
AAO(t)	Antarctic Oscillation (AAO)	https://www.cpc.ncep.noaa.gov/products/precip/CWlink/daily_ao_index/aao/aao.shtml

An important criterion of MLR is that the impact proxies should not be highly correlated with each other. As shown in Fig. 1, correlations among the proxies are minimal, with the highest coefficient at 0.3, satisfying the precondition for MLR analysis. Therefore, these proxies are suitable for analyzing long-term ozone changes.

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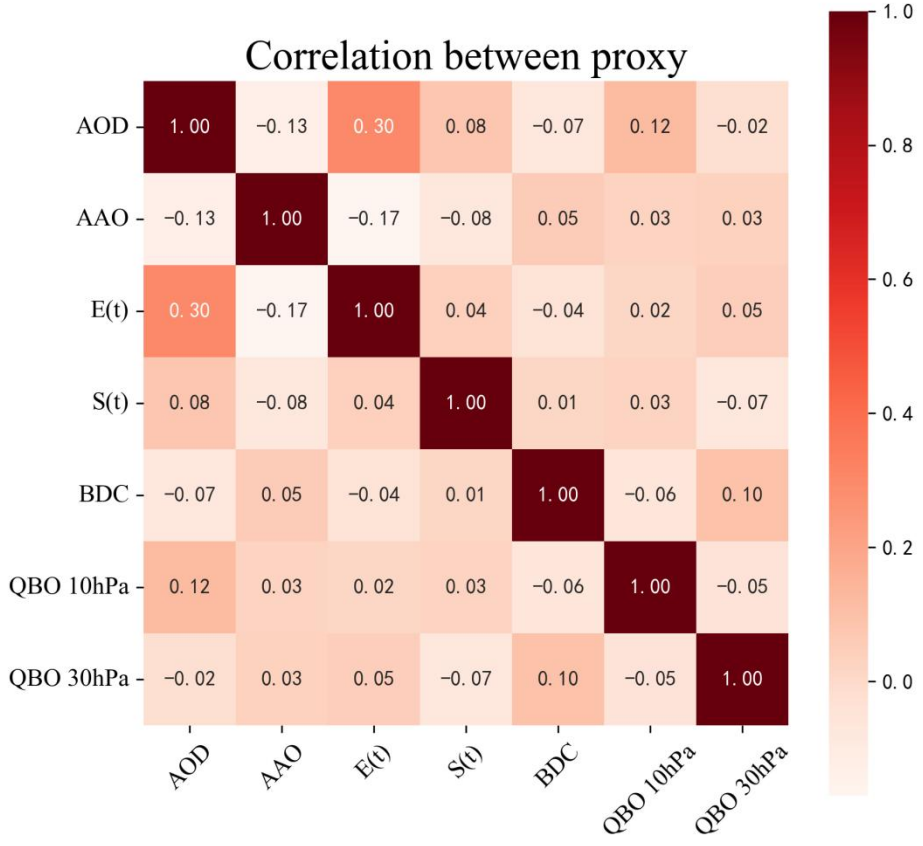

Figure 1. Correlation coefficients among the main impact proxies.

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4 Long-term trends in Antarctic ozone

Antarctic ozone recovery exhibits strong seasonal dependence, particularly the contrasting behavior in September and October. Figure 2 illustrates the long-term ozone trends in the Antarctic region (60°S-90°S) from four datasets, reflecting the persistence of the deep ozone hole and extended periods of low ozone over Antarctica in recent years. Among these,
175 WOUDC data indicate relatively small fluctuations in TCO values. In contrast, the TOMCAT and MSR2 exhibit more pronounced variations. To ensure consistency across datasets with different temporal coverage, trends were analyzed for 2001- 2023. During this period, September shows signs of recovery, whereas October exhibits a notable decline of approximately -1 DU/yr. Overall, the ozone variations among the datasets show good consistency, and we examined trends across different time spans to clarify these seasonal behaviors.

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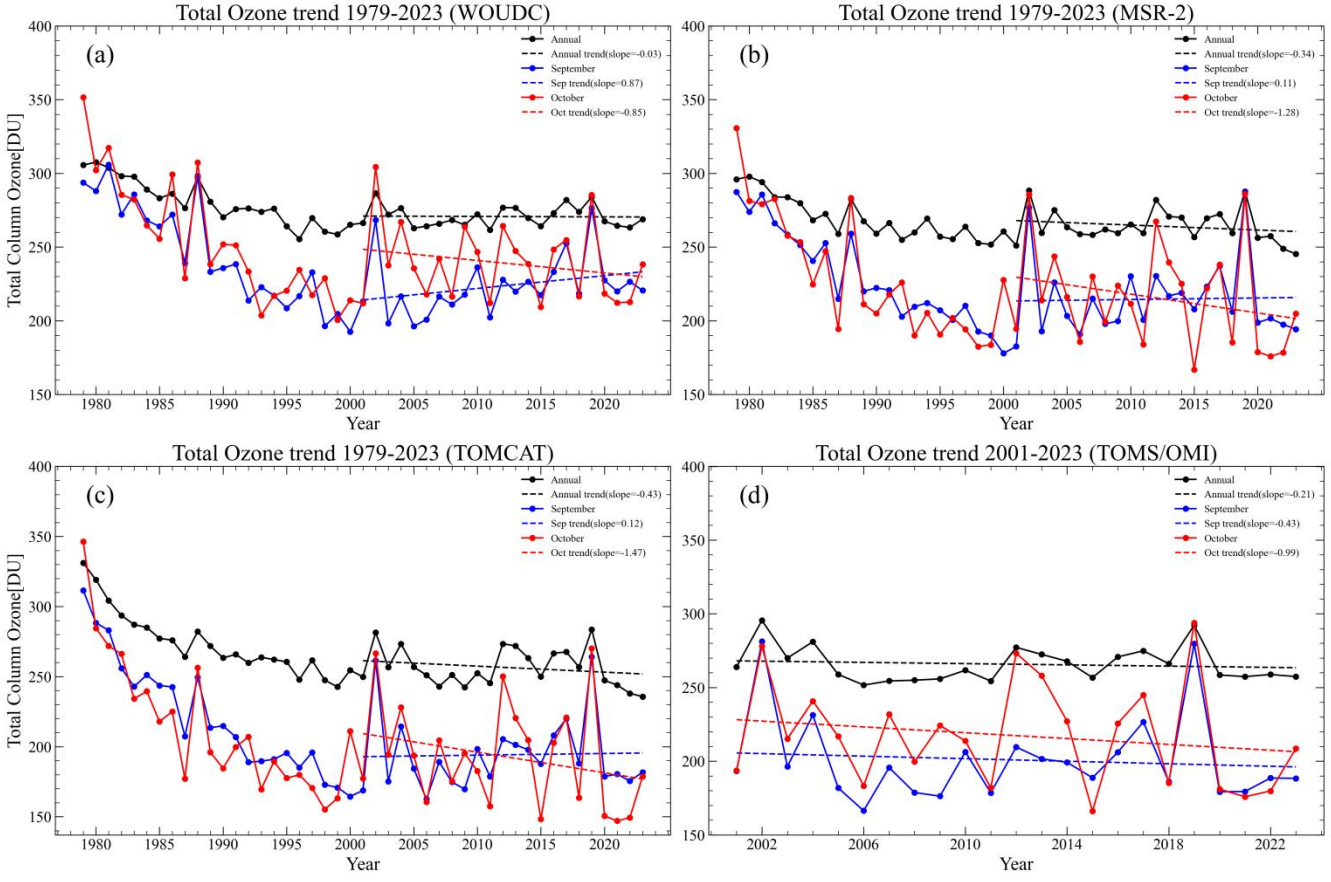

Figure 2. The TCO time series in the Antarctic (90°-60°S) from multiple datasets. (a) WOUDC, (b) MSR, (c) TOMCAT, and (d) TOMS/OMI. The black line represents the annual mean time series, the blue line represents the September time series, and the red line represents the October time series. The dotted lines show the linear trends from 2001 to 2023, corresponding to the annual mean (black), September (blue), and October (red).

Table 3 summarizes the trends from four datasets along with MLR correlation coefficients (R^2). Antarctic ozone declined steadily at an annual decline rate of 2-3 DU/yr, with a more pronounced rate of 5-6 DU/yr during September and October. The long-term annual trend from 2001 to 2019 was approximately 0.5 DU/yr across multiple datasets, whereas the TOMCAT simulation for 2001- 2023 showed a negative trend (0.4 DU/yr). After 2000, September consistently exhibits positive trends, while October trends shift from weakly positive (0.3 ± 3.2 DU yr^{-1} for 2001–2019) to significantly negative (–1.5 ± 2.4 DU yr^{-1} for 2001–2023). This divergence suggests that dynamical processes drive chemical depletion in October, and EESC might not accurately reflect the ozone changes in October.

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195 Table 3. Linear trends (DU/yr) of TCO in the annual mean, September, and October means with different time spans for each dataset, and the correlation between dataset and MLR (R^2).

	Dataset / Time span	TOMCAT	WOUDC	MSR-2	TOMS/OMI
Annual	1979-2000	-3.2 (0.7)	-2.3 (0.4)	-1.9 (0.5)	
	2001-2019	0.5 (1.1)	0.3 (0.6)	0.4 (0.9)	0.3 (1.1)
	2001-2023	-0.4 (0.9)	-0.03 (0.5)	-0.3 (0.7)	-0.2 (0.8)
	R^2	0.85	0.86	0.73	0.67
September	1979-2000	-5.9 (1)	-5 (1.1)	-4.7 (0.8)	
	2001-2019	1.4 (2.4)	1.5 (1.8)	1.5 (2.3)	0.6 (2.8)
	2001-2023	0.1 (1.8)	0.9 (1.3)	0.1 (1.7)	-0.4 (2)
	R^2	0.91	0.88	0.85	0.8
October	1979-2000	-6 (1.9)	-5.4 (1.5)	-5.1 (1.7)	
	2001-2019	0.3 (3.2)	-0.03 (2.4)	0.2 (3)	0.7 (3.1)
	2001-2023	-1.5 (2.4)	-0.8 (1.7)	-1.3 (2.2)	-1 (2.3)
	R^2	0.83	0.77	0.82	0.76

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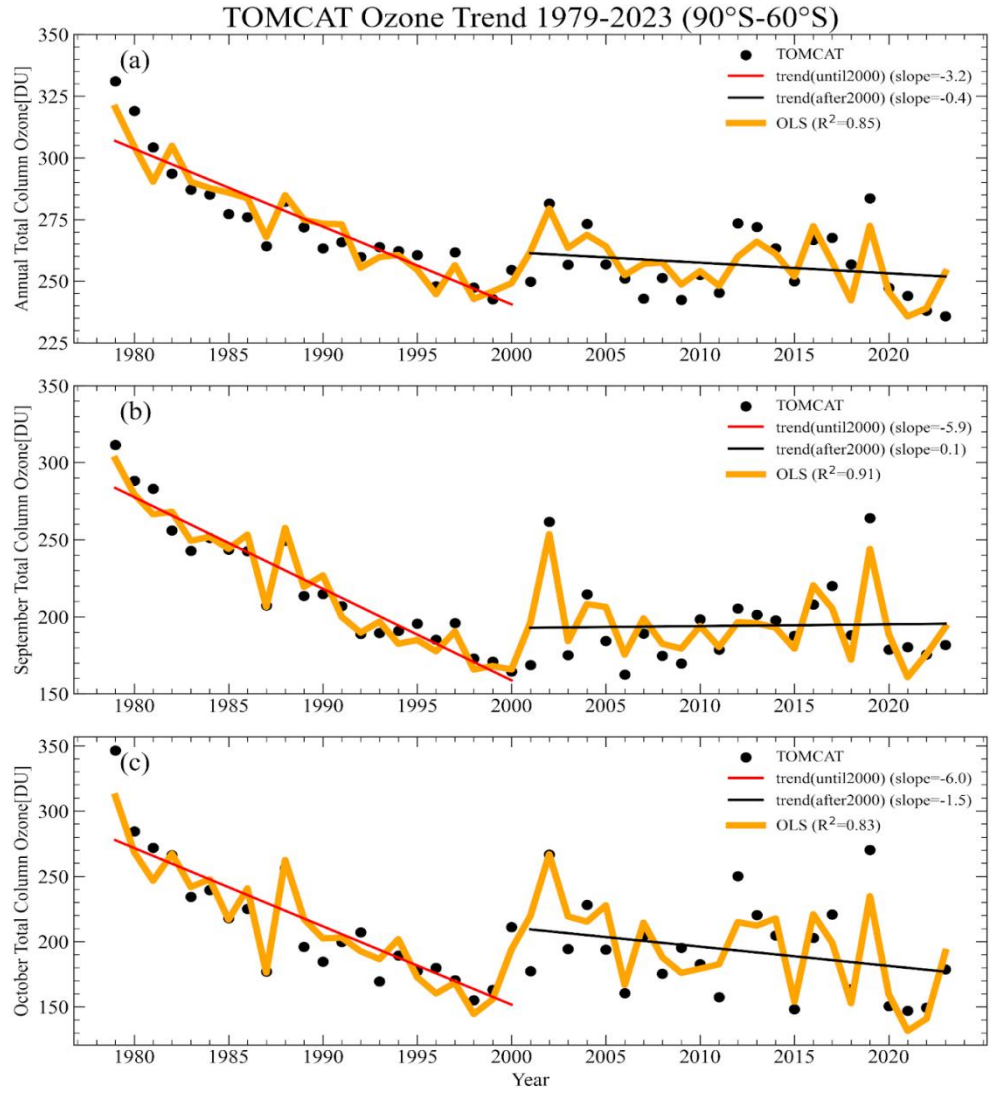

Figure 3. TCO time series of the TOMCAT dataset from 1979 to 2023. (a) Annual mean, (b) September, and (c) October. The black dots are TOMCAT, the orange thick line is time series based on MLR results, the red line is the linear trend of the TOMCAT from 1979 to 2000, and the black is the linear trend of the TOMCAT from 2001 to 2023.

To further evaluate the ability of the regression framework to reproduce observed variability, we examine the TOMCAT-based MLR results in detail. Fig. 3 presents the time series of TCO and the MLR based on the TOMCAT dataset. The results suggest a post-2000 decline, with an interannual trend of -0.4 (0.9) DU/yr and a stronger October trend of-1.5 (2.4) DU/yr. However, September shows a weak positive trend of 0.1 (1.8) DU/yr. Regression analysis across four datasets demonstrates good agreement in the long-term ozone changes. The long-term ozone changes in the MLR can explaining about 85 % of the

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variance in the interannual time series. Among them, the MLR of TOMCAT is the largest, explaining 91 % of the variance in the time series, indicating strong reproduction of observed long-term ozone variability.

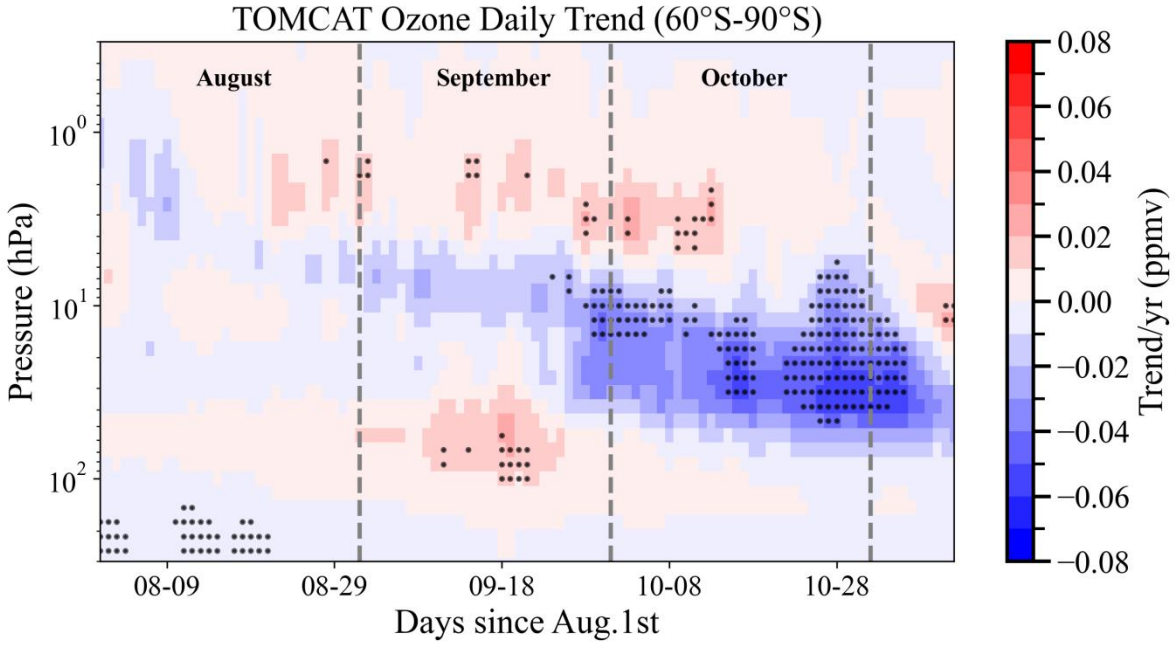

Figure 4. Vertical cross section of daily trends (2001-2023) in ozone volume mixing ratios from TOMCAT. Trends are shown for August 1 to November 10 (2001-2023). Stippled areas are statistically significant above the 95% confidence level.

To determine whether the September–October contrast arises from changes at specific altitudes, we examined vertical ozone trends in the austral winter/spring. Ozone mixing ratios from the TOMCAT dataset were analysed as a function of altitude and day of the year (1 August–10 November) for 2001-2023 (Fig. 4). In August, the ozone mixing ratio trend at 1 hPa showed a negative change and gradually extended downward. By September, this trend reached the mid-stratosphere, resulting in a negative anomaly, with a rate of change in the ozone mixing ratio of -0.03 parts per million per year (ppmv/year). However, positive trends dominate the middle and lower stratosphere, reaching ~0.04 ppmv/year, exceeding the magnitude of the negative changes and consistent with the recovery observed in September. In October, a broader negative region emerged (5-80 hPa), peaking at -0.07 ppmv/year and coinciding with the main Antarctic ozone layer (4-20 hPa). The persistent negative trend in ozone continued into early November, suggesting prolonged low Antarctic ozone values and requiring continued monitoring of dynamical and chemical processes driving these trend changes.

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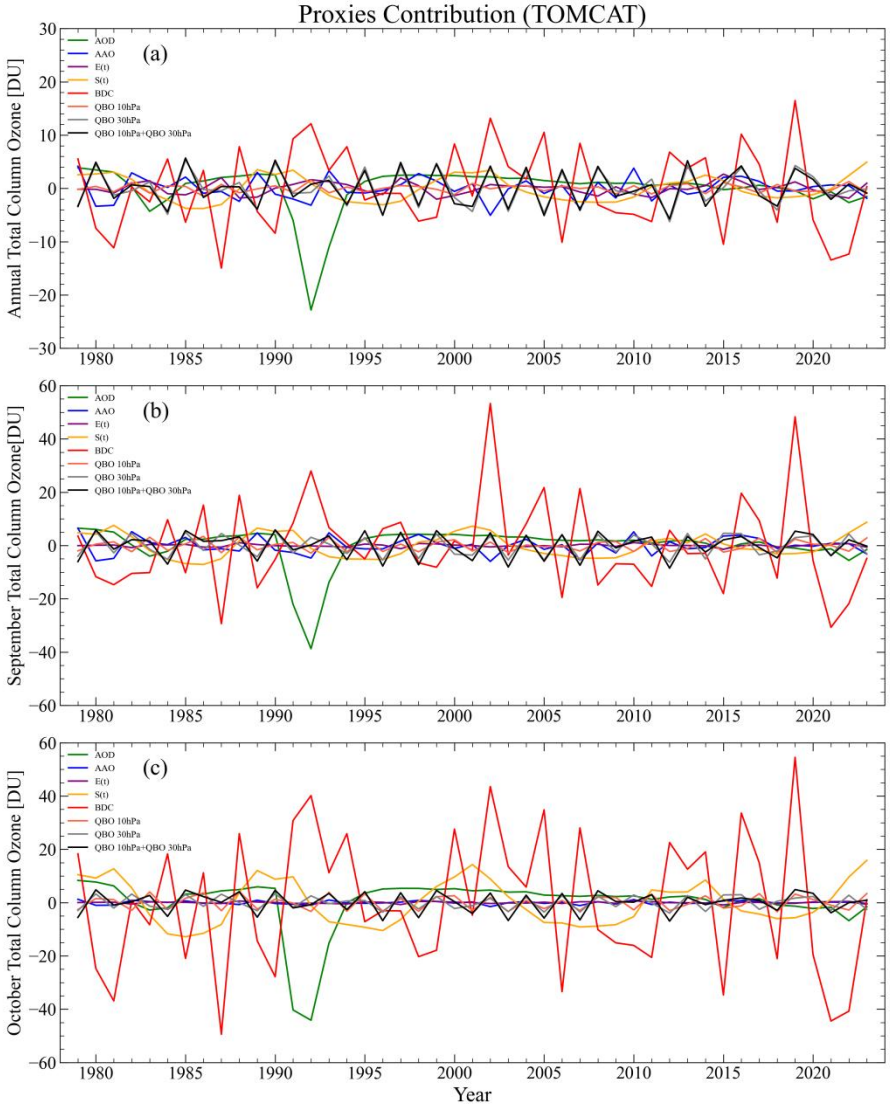

Figure 5. The contribution of each proxy to TCO from 1979 to 2023 based on TOMCAT data. (a) Annual mean, (b) September, and (c) October. Green line: AOD, blue line: AAO, purple line: ENSO, orange line: Solar cycle, red line: BDC, light red line: QBO 10 hPa, grey line: QBO 30 hPa, black line: Overall combined QBO.

To elucidate the impact of each proxy, we analyse their contributions to ozone variation using the MLR results of the TOMCAT dataset. BDC has a significant impact on the long-term ozone variation and recent interannual ozone variability, contributing up to –12 DU during 2020-2023 (Fig. 5a). AOD exerted a significant influence on Antarctic ozone following the El Chichón (1982) and Pinatubo (1991) volcanic eruptions, with a peak contribution of ~–25 DU. Other proxies contribute less than 6 DU but their influences are not negligible.

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During the austral winter/spring, the MLR well described the contribution of dynamic processes to stratospheric ozone, with BDC being the main driver of long-term ozone changes. Variations in planetary wave forcing, represented by EHF, modulate the BDC and influence polar vortex behavior. Concurrently, as a factor with periodic fluctuations, solar cycle contributes less than ±15 DU to ozone variability in September and October (Figs. 5b-c). After a volcanic eruption, AOD will remain high in the stratosphere for a limited period. For instance, the Pinatubo eruption resulted in the largest reduction of up to -40 DU for a few years, and smaller volcanic eruptions, such as the 2022 Hunga Tonga-Hunga Ha'apai volcano eruption in 2022, influenced ozone by up to -7 DU.

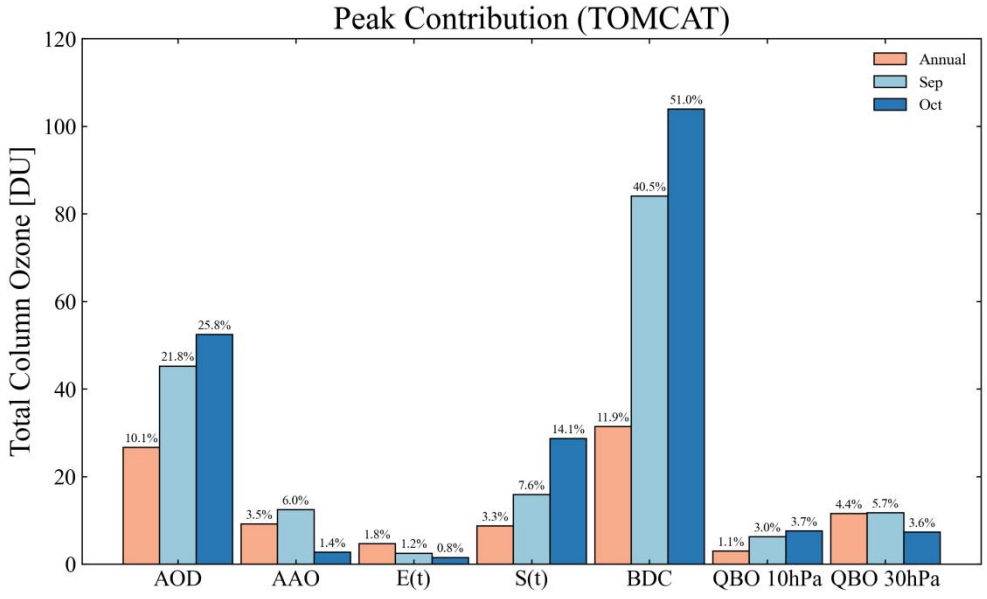

Figure 6. Peak contribution (unit: DU) of each proxy to TCO changes from 1979 to 2023 based on TOMCAT. Orange: Annual mean; light blue: September; blue: October. The rate of ozone change in percent is labelled above each bar.

Fig. 6 illustrates the peak influence of each proxy on the TCO changes. BDC and AOD dominate interannual variation of ozone, while the combined QBO accounts for up to 5.5% of the variance. The dominant role of BDC can be explained by its transport of ozone from the tropics to high latitudes, with ozone accumulation reaching a maximum at mid to high latitudes from May to September. During winter, ozone is transported from mid latitudes to the polar regions in spring, and the efficiency of this transport depends on the strength of BDC (Weber et al., 2011; Fioletov et al., 2023). These transport processes lead to a more pronounced contribution of BDC to the long-term ozone variability in September and October, with

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peak rate reaching 51%, indicating its significant impact on Antarctica TCO fluctuation. Comparatively, peak contributions of QBO and solar cycle are approximately 8% in September, whereas other proxies contribute less than 6%.

5 Model simulations

5.1 Setup of the model sensitivity experiment

Figures 5 and 6 clearly show that variations in BDC strength have a profound impact on the Antarctic ozone recovery. To investigate the role of BDC on Antarctic ozone, we performed two sensitivity simulations using TOMCAT to explore the modulation effect of BDC on ozone. The control experiment (CRL) uses the standard chemical and dynamical parameters spanning the period 2000-2009. To assess the impact of BDC intensity, two sensitivity experiments were conducted based on typical years of BDC anomalies: EXP1 represents a year with strong BDC (2002), while EXP2 represents a year with weak BDC (2006). In these experiments, wind forcing and temperature from 2000 to 2009 was altered to modify BDC intensity, while other parameters remain unchanged. The experimental design is summarized in Table 4.

The selection of 2002 and 2006 as typical years was guided by interannual variation of ozone and ODS changes to ensure that the BDC intensity is the dominant factor influencing the ozone variation. Previous studies have shown a weakening of ozone transport to the polar regions in 2006, accompanied by persistent cold temperatures and stable polar vortex in late winter and early spring (Peshin, 2008; Grytsai, 2011). In contrast, the typical strengthening of BDC occurred in 2002 as a result of unusually strong upward planetary wave propagation. Elevated stratospheric temperature in the SH, along with polar vortex splitting, created an unfavorable environment for polar ozone depletion (Allen et al., 2003; Sinnhuber et al., 2003). These marked differences in circulation intensity highlight the contrasting dynamical regimes of 2002 and 2006, making them ideal case studies for examining the role of BDC in Antarctic ozone variability.

Table 4. Experimental design.

Trial	Simulation Result	Simulation Process
Control experiment (CRL)	TOMCAT Dataset for 2000-2009	-

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Sensitivity experiment 1 (EXP1)	Simulated TOMCAT dataset for 2000-2009	The 2002 wind forcing and temperatures are applied to all years during 2000-2009 and other variables are unchanged from the CRL.
Sensitivity experiment 2 (EXP2)	Simulated TOMCAT dataset for 2000-2009	Same as EXP1, but wind forcing and temperatures of 2006 applied for all years.

5.2 Simulation result

280 An increase in TCO is observed in EXP1 (strong BDC; Fig. 7), with values from 2000 to 2009 approximating those in 2002. Conversely, EXP2 (weak BDC) reveals ozone reductions, with September and October ozone values resembling those in 2006. The sensitivity experiment results suggest that the peak contribution rates are consistent with the MLR results, with the annual mean and October peak contribution rates of ~15% and ~52%, respectively.

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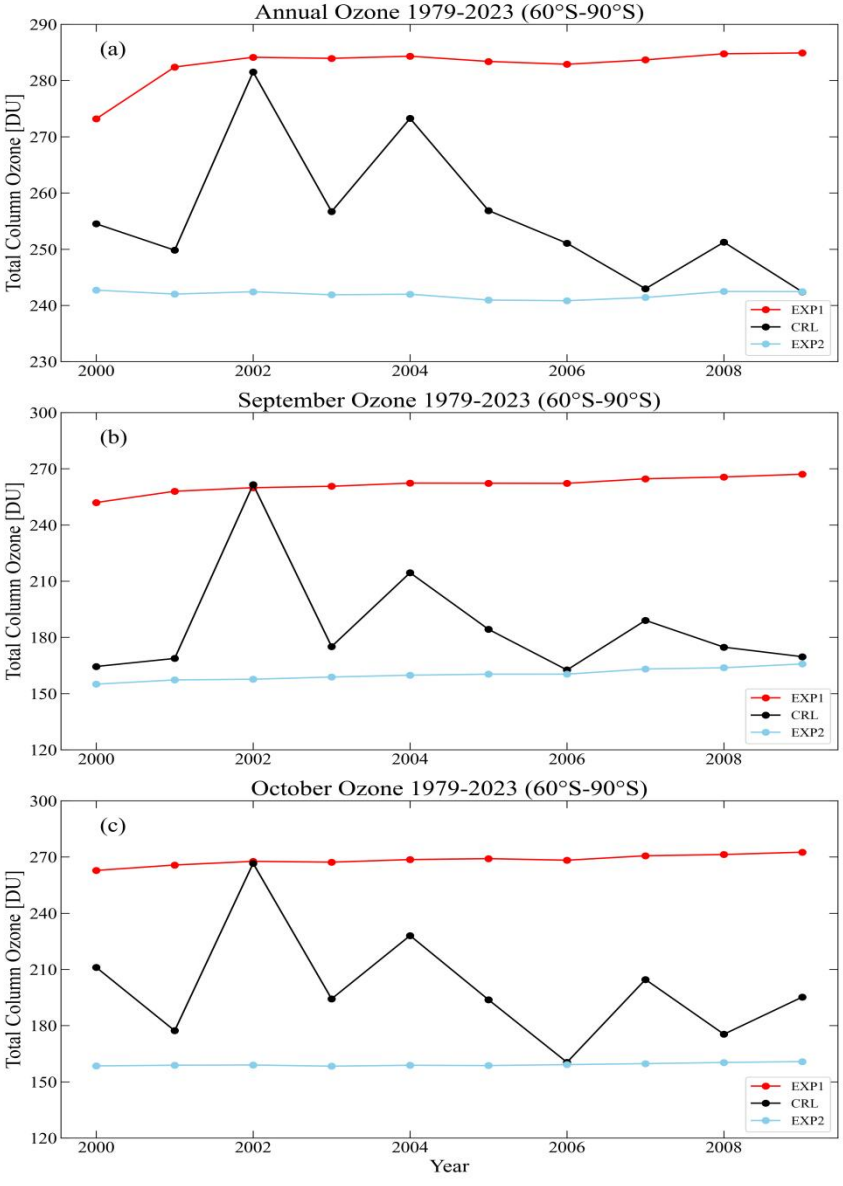

Figure. 7. TCO changes in Antarctica for control and sensitivity experiments from 2000-2009. (a) Annual mean, (b) September, and (c) October. Red: EXP1 (weak BDC), sky blue: EXP2 (strong BDC), black: CRL.

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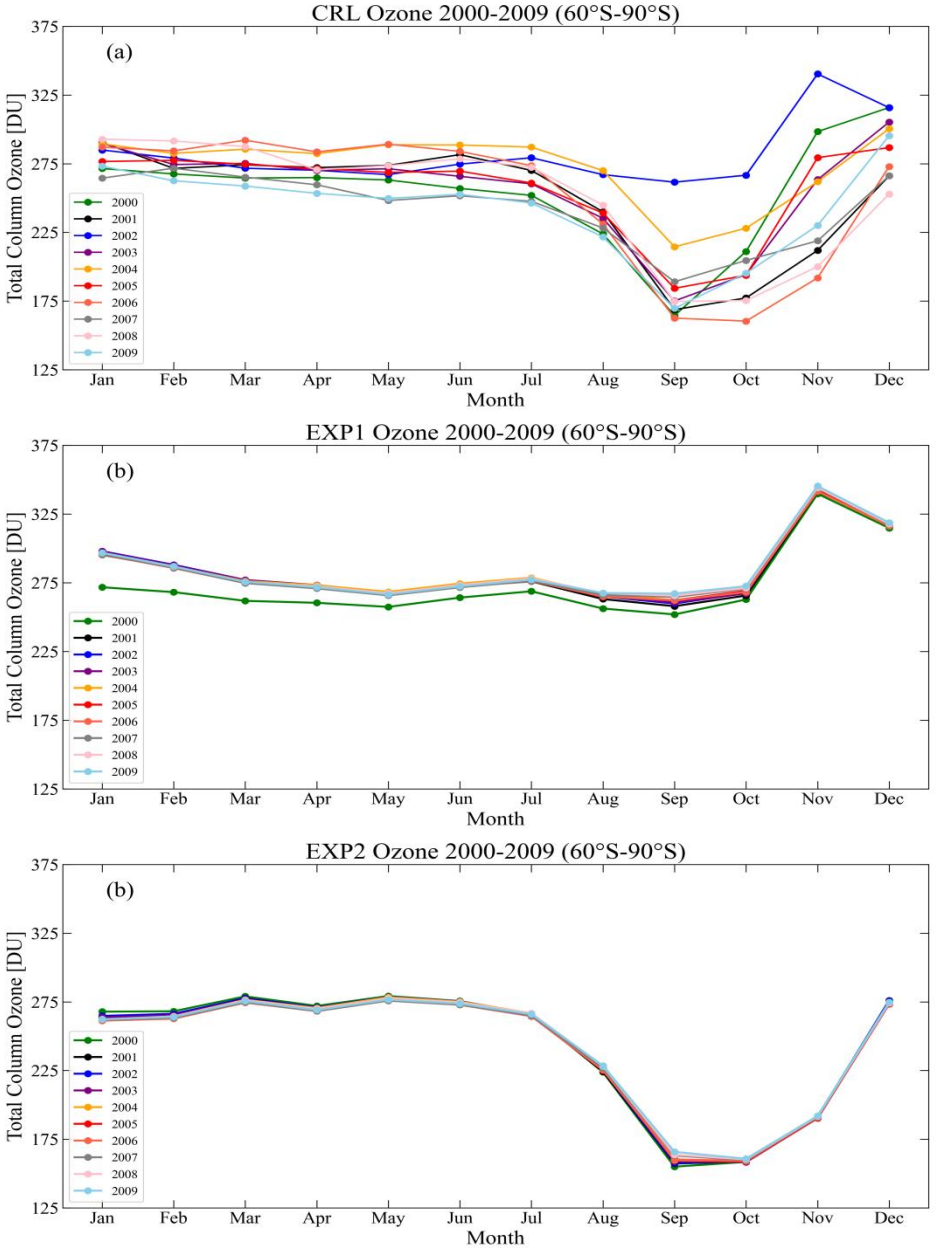

290 Figure. 8. Monthly TCO in the Antarctic for control and sensitivity experiments. (a) CRL, (b) EXP1, and (c) EXP2.

Monthly TCO changes for control and sensitivity experiments from 2000 to 2009 are illustrated in Figure 8. Ozone typically experiences pronounced depletion in September, with TCO dropping below 220 DU, a threshold associated with the formation of the ozone hole. In 2002, September and October showed significant anomalies, with TCO values reaching 250
295 DU. According to the EXP1, enhanced circulation during the winter and spring increased TCO towards those of 2002. In

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contrast, during spring of 2006, the lowest TCO of the decade occurred, with a substantial ozone hole persisting into October (TCO ~150 DU). Recovery signs emerged in November, delayed by persistent low temperatures and the protracted breakup of the polar vortex.

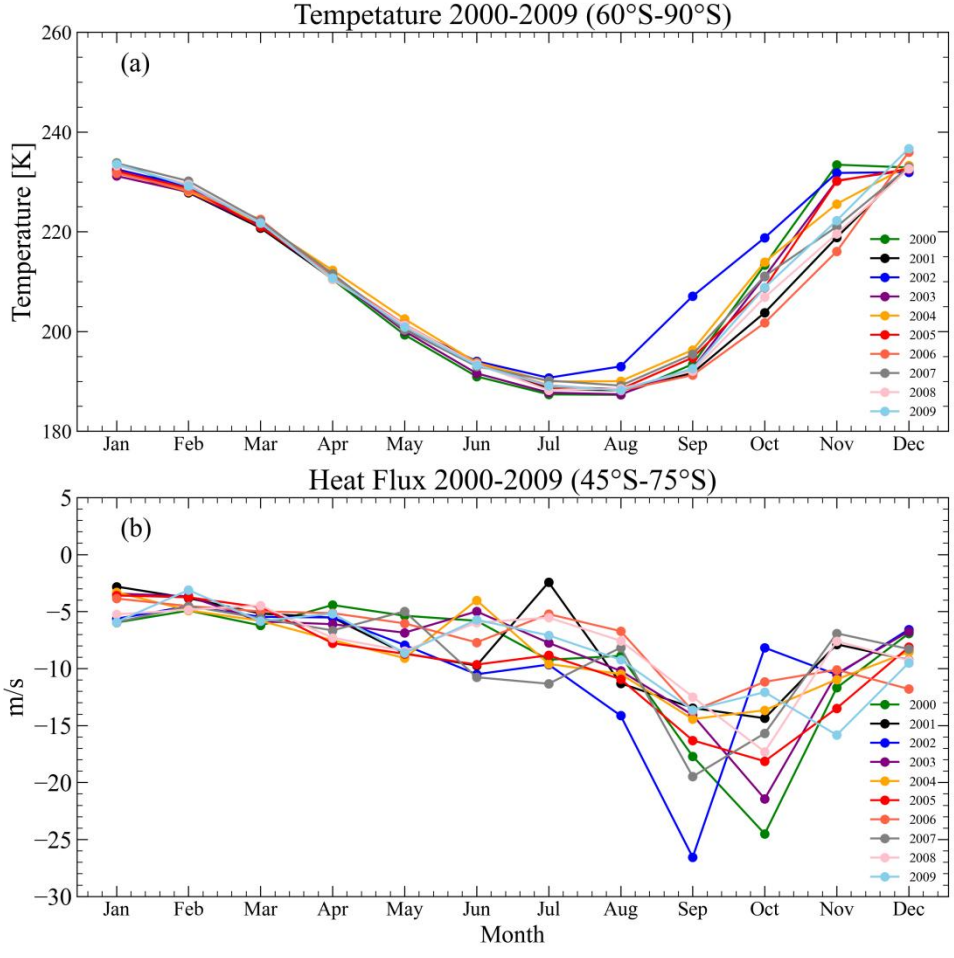

Figure 9. (a) Monthly mean temperature at 50 hPa in Antarctica. (b) Monthly mean eddy heat flux (EHF) at 100 hPa (45° S-75° S).

Figure 9a shows the monthly mean temperature at 50 hPa from 2000 to 2009. The abnormal increase in temperature in the winter and spring of 2002 coincided with higher TCO, while the cooler temperatures in the spring of 2006 were associated with the persistently low ozone values (Fig. 8 and Fig. 9a). These temperature variations are inextricably linked to BDC strength and the timing of vortex breakup (Weber et al., 2011; Butchart, 2014). As shown in Fig. 9b, EHF was high in the spring and winter of 2006, indicating the weakened circulation and reduced ozone transport from the tropics to the polar

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regions. Meanwhile, the persistent stratosphere cooling favored vortex survival and delayed breakup, resulting in a prolonged and extensive ozone hole in 2006.

The mechanistic link between circulation and ozone is clear. In 2002, enhanced planetary wave activity strengthened the BDC, promoting tropical-to-polar ozone transport. Elevated temperatures have suppressed the efficiency of stratospheric cloud formation, limiting ozone depletion. The polar vortex split over Antarctica, with one part moving equatorward in the South Pacific and the other remaining over Antarctica, but the anomalies triggered an earlier breakup than in previous years. The combination of strong BDC transport, favorable temperatures and split vortex dynamics led to exceptionally high TCO in 2002, primarily related to the SSW and enhanced transport mechanisms (Sinnhuber et al., 2003; Wang et al., 2005).

6 Summary and conclusions

This study combined satellite-based observations, reanalysis datasets as well as chemistry-climate model simulations to analyze the latest long term ozone trend over Antarctica during 1979-2023. Using MLR, we analyzed the contributions of dynamical and chemical proxies to ozone variability. Based on the TOMCAT 3-D model, we conducted sensitivity experiments to investigate the impact of the BDC on ozone. Important conclusions are:

(1) Multiple datasets can consistently well represent the long-term ozone changes over Antarctica. In 2001-2019, annual ozone showed signs of recovery, while the annual change in TCO shifted downward at -0.4 (1.1) DU/yr during 2001-2023.

(2) The MLR reproduces observed long-term ozone changes with high fidelity, explaining about 67-91 % of the variance in the time series over the Antarctic. The daily ozone trends based on TOMCAT dataset during the period 2001-2023 showed that the recovery of ozone in September is due to increasing ozone trends in the lowermost stratosphere. While in October, negative trends are observed in the entire lower stratosphere. This seasonal contrast explains why the TCO trends are negative in October but slightly positive in September.

(3) Proxy analysis highlights the dominant role of BDC in the Antarctic spring, with volcanic aerosol forcing, particularly following the Pinatubo eruption, producing substantial ozone losses. Other proxies also exert smaller but non-negligible contributions to the ozone change.

(4) Sensitivity experiments further reveal that the strengthening (weakening) of BDC led to an increase (decrease) in the transport of tropical ozone to polar regions. The BDC anomaly in the SH significantly affects the polar temperature, and thereby ozone depletion, with circulation anomalies capable of driving large monthly variability as high as 51% in October.

Overall, in the long-term, Antarctic ozone evolution reflects the interplay of multiple processes, with dynamical drivers holding a particularly strong influence on recovery patterns. BDC perturbations play a significant role in the long-term ozone trend, requiring more research and continued attention to the ozone hole and dynamic processes to improve our understanding of long-term ozone variability and predict future changes in the Antarctic ozone hole.

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Data availability

Observational and satellite data used are available in Sect. 2. The TOMCAT/SLIMCAT model data used is supported by the University of Leeds.

Author contributions

HH analysed the data and prepared the manuscript under the guidance of SC. MPC, SSD, WF, SC, YL, MW, SH supported the discussion, interpretation and analyse, and helped to write the paper. MPC provided support in model running and processing. All authors edited and contributed to the write of the manuscript.

Competing interests

The authors declare that they have no conflict of interest with other people.

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