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A review of Antarctic stratospheric ozone trends and variability and their impacts on surface climate

Eun-Pa Lim, Ghyslaine Boschat, Irina Rudeva and Chris Lucas

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EXECUTIVE SUMMARY

Ozone is an important atmospheric constituent that absorbs the solar ultraviolet radiation in the stratosphere, therefore keeping the stratosphere warm and protecting the biosphere from the damaging effects of ultraviolet radiation. Regarding the role of ozone in climate variability and long-term change, large variations in ozone concentration can result in significant changes in stratospheric temperatures and circulations. These stratospheric changes can significantly impact lower atmospheric temperatures and circulations when atmospheric mean conditions are favourable for stratosphere-troposphere interactions. In this report, we review some key research papers that showed 1) significant long-term trends in ozone in the last 50 years and the associated trends in climate conditions and 2) impacts of ozone on the interannual variability and predictability of the tropospheric circulations and surface climate. At the end of the report, we attempt to identify some research questions to address to improve our understanding of the role of the Antarctic ozone changes in the Australian climate.

1. INTRODUCTION

Ozone (O_3) is a trace atmospheric constituent that is well known for its absorption of solar ultraviolet (UV) radiation at wavelengths below 300 nm, thereby warming the stratosphere where ozone is rich (the altitude of 15-50 km) (Holton 1992; Peixoto and Oort 1992). The absorption of solar UV radiation by ozone protects the biosphere from the biologically harmful effects of UV radiation. Ozone is formed above 25 km primarily in tropical latitudes where the solar UV radiation is the most intense, because the solar UV radiation breaks apart one oxygen molecule (O_2) into two oxygen atoms (2O), each of which, then, combines with an O_2 to form an ozone molecule (O_3) (Fahey and Hegglin 2010a). Ozone over the tropics is then re-distributed by the atmospheric circulation: the maximum column amounts of ozone are found near the North Pole in April and near 60°S in October, and the minima are found in the equatorial region in all seasons and in the Antarctic polar region in spring. The Antarctic minimum is known as the Antarctic ozone hole, which had a significant growing trend due to anthropogenically produced ozone-depleting substances (ODSs) (Bowman and Krueger 1985; Holton 1992).

The major ozone destruction occurs through complex series of catalytic reaction cycles involving the odd hydrogen, odd nitrogen, and odd chlorine families (HOx, NOx, Clx) and solar UV radiation (Andrews et al. 1987; Peixoto and Oort 1992). This process is simplified as

$$X + O_3 \rightarrow XO + O_2$$

$$XO + O \rightarrow X + O_2$$

where X is a catalytic molecule such as HO, NO, and Cl that remains unchanged after depleting ozone. As explained in Fahey and Hegglin (2010b) in detail, extensive ozone depletion occurs in the polar region because of its extreme low temperatures in late winter to early spring when liquid and solid polar stratospheric clouds (PSCs) form and the polar region starts to receive the sunlight after the winter polar night period. Reactions on the surfaces of these PSCs initiate a significant increase in the most reactive chlorine gas, chlorine monoxide (ClO). Different types of liquid and solid PSC particles form when stratospheric temperatures fall below about −78°C. As Antarctic temperatures remain below this temperature in the stratosphere from late autumn to early spring, much ozone gets depleted, and therefore, the minimum ozone concentration is found over Antarctica in spring. As well as providing favourable surfaces for the chlorine reaction, the PSCs remove nitric acid (HNO_3) from the ozone layer as they descend due to gravity, which is known as denitrification of the stratosphere. Denitrification removes the NOx available for converting the highly reactive chlorine gas ClO back into the stable reservoir gas $ClONO_2$, thereby allowing ClO to stay for an extended period, destroying more ozone.

An anomalous reduction of Antarctic ozone cools the polar region, therefore increasing the meridional temperature gradient in the lower stratosphere, which leads to a strengthening and poleward shift of the stratospheric polar vortex and a substantial delay of its breakdown in late spring to early summer (Waugh et al. 1999; Randel and Wu 1999). Because this is the time of year when the stratospheric wind and temperature anomalies move downward to the surface with active wave-mean flow feedback (Kuroda and Kodera 1998; Hio and Yoden 2005; Lim et al. 2018), the stratospheric changes associated with anomalous Antarctic ozone depletion result in a poleward shift of the tropospheric midlatitude jet, which is known as the positive phase of the Southern Annular Mode, SAM (e.g., Thompson and Solomon 2002; Thompson et al. 2005; Fogt et al. 2009; Lim et al. 2018; Keeble et al. 2014; Son et al. 2018). This chain of processes of the stratospheric anomalies leading to the tropospheric anomalies applies to both the long-term trend and interannual variability. Conversely, an anomalous increase of Antarctic ozone leads to the opposite processes, resulting in the negative phase of the SAM at the surface (Hendon et al. 2020).

The SAM is the leading mode of the Southern Hemisphere (SH) extratropical circulation variability at weekly to centennial timescales, which features annular patterns of pressure anomalies with the opposite signs between the mid- and high latitudes of the SH (Trenberth 1979; Karoly et al. 1996; Gong and Wang 1999; Thompson and Wallace 2000; Marshall 2003; Arblaster and Meehl 2006; Yang et al. 2020). The positive phase of the SAM (pSAM) is characterised by higher pressure anomalies in the midlatitudes equatorward of ~50°S and lower pressure anomalies in the high latitudes poleward of ~50°S, which is caused by a poleward shift of the tropospheric midlatitude jet. The negative phase of the SAM (nSAM) is characterised by the oppositely signed anomalies in the pressure field with an equatorward shift of the jet. The SAM is an important driver of Australian weather and climate: pSAM generally brings more rainfall over the continent and eastern Tasmania in all seasons except for winter when pSAM brings less rainfall on the southwest corner of Western Australia, Victoria, and parts of Tasmania (Hendon et al. 2007; Purich et al. 2013; Lim et al. 2016). Furthermore, studies such as Min et al. (2013), Harris and Lucas (2019), Lim et al. (2019), and Marshall et al. (2022) showed that nSAM tends to induce warm and dry conditions over eastern Australia in austral spring and summer, and therefore, plays a key role in increasing chances of heat and dry extremes and bushfire incidents on weekly to seasonal timescales.

The Antarctic ozone hole became steadily larger from the 1970s to 90s, which has been widely accepted to have been driven by the anthropogenic emission of ODSs including chlorofluorocarbons (CFCs) (e.g., Farman et al. 1985). During this period, the SAM had a strong positive trend in austral summer, which was largely driven by the increasing Antarctic ozone hole trend (Thompson and Solomon 2002; Arblaster and Meehl 2006; Thompson et al. 2011). Kang et al. (2011) showed that this positive trend in the SAM in austral summer, which was driven by the decreasing Antarctic ozone, led to a wetting trend in the SH subtropical latitudes including over the Australian continent. Thanks to the implementation of a series of restrictions started by the Montreal Protocol signed in 1987 that aimed to phase out the production and consumption of ODSs, recent studies have been reporting a reversal of the Antarctic ozone-depleting trend since 2000 (Salby et al. 2011; Solomon et al. 2016) and an associated pause or reversal of the positive trends in the stratospheric polar vortex strength (Zambri et al. 2021) and the tropospheric SAM (Banerjee et al. 2020).

In addition to the reversing trend in the Antarctic ozone, its year-to-year variability and associated impact on the circulation and surface climate have also received much attention in recent years from the perspective of an untapped source of climate predictability. The current state-of-the-art dynamical sub-seasonal to seasonal forecast systems generally use the monthly mean ozone, which is zonally and long-term averaged with the implicit assumption that the role of asymmetric patterns of ozone and/or interannual variability of ozone is minimal for the forecast large-scale circulation anomalies. However, idealised dynamical forecast experiments have showed that using realistic ozone forcing (even though it is still zonally averaged) can significantly improve the forecast skill of the tropospheric circulation anomalies and Australian temperature and rainfall anomalies in austral spring (Hendon et al. 2020; Oh et al. 2022).

Given the importance of Antarctic stratospheric ozone trends and variability in modulating the tropospheric circulation and the SH surface climate in the present and future climate, in this report we review some research findings on its trend and variability. We will also attempt to identify areas of research that need to be pursued to improve our understanding of the role of the Antarctic ozone changes for the Australian climate.

2. LONG-TERM TRENDS

As described above, it is well established that anthropogenically driven ozone depletion was a major driver of a strong trend in the surface climate of the SH in austral summer. To understand how ozone depletion drives the stratospheric and tropospheric circulation changes, Keeble et al. (2014) conducted atmosphere-chemistry coupled model experiments, using the version 7.3 of the Met Office's Unified Model HadGEM 3-A configuration (Hewitt et al., 2011) coupled with the United Kingdom Chemistry and Aerosol module (hereafter referred to as UM-UKCA). For this study, two simulations were considered: the ozone depletion experiment was run for 30 years with the year 2000 boundary conditions, including sea surface temperatures, sea-ice extent, and the loadings of greenhouse gases (GHG), CFCs, and aerosols. The pre-ozone depletion experiment is identical to the ozone depletion experiment except that chlorine activation due to heterogeneous reactions on PSC particles was suppressed, preventing large spring-time ozone losses in the lower polar stratosphere. From the comparison of the two experiments, they found that the lower stratospheric ozone depletion anomalies starting in August persisted through summer and autumn in the ozone depletion experiment, which were accompanied by ozone enhancement anomalies in the mid-stratosphere likely caused by an increased downward motion (i.e., increased strength of the Brewer-Dobson circulation; BDC). They also saw that tropospheric ozone mixing ratios were greatly reduced over the Antarctic polar cap throughout the year due to a decrease in stratosphere-troposphere exchange. While the largest ozone decreases were found in the SH spring-time polar vortex, total column ozone over the entire SH including the equator decreased. The decrease of ozone in the tropics was attributed to an intensified BDC, transporting ozone away from the tropics.

Dennison et al. (2015) conducted similar ozone sensitivity experiments, using the National Institute of Water and Atmospheric Research-United Kingdom Chemistry and Aerosols (NIWA-UKCA) coupled atmosphere-ocean chemistry-climate model, which is similar to that of Keeble et al. (2014) but is coupled to the ocean as well. Multi-member ensemble simulations were run with chlorinated or brominated ODSs following the IPCC A1 scenario covering 1950-2100 as a reference experiment, and the second experiment was with fixed ODSs at their 1960 levels (i.e., before the ozone-depletion trend started). Both experiments were forced with GHG forcing following the RCP6.0 scenario. The comparison of their modelling experiments showed that ozone depletion of the 1970s-90s could have caused the lower stratospheric polar vortex to be more extreme and more persistent in austral spring and summer, which led to stronger downward coupling. This conclusion was consistent with that of Fogt et al. (2009) who showed that ozone depletion during the late 20th century increased the persistence and strength of the ozone-SAM relationships over the South Pole. An important implication of these findings is that the recovery of ozone in the future could bring the opposite changes, which may imply a reduction in the skill of sub-seasonal to seasonal forecasts for the surface SAM and associated climate anomalies.

Possible impacts of the ozone trend have also been studied in austral autumn and winter. Delworth and Zeng (2014) attributed the autumn and winter drying trend of 1981-2012 relative to the 1911-1970 climatology over south-western and south-eastern Australia to radiative forcing changes associated with GHGs and ozone. They did this by conducting a series of experiments with different forcings with a high-resolution global climate model, the GFDL Coupled Model 2.5 (50 km). 1000-year control simulations were run with fixed concentrations of GHGs and aerosols of pre-industrial conditions. Then, 3 to 5-member ensemble simulations were run with all forcings; anthropogenic forcings (all forcings minus solar, volcanic forcings); natural forcings (solar, volcanic); well-mixed GHG forcing; and ozone forcing (i.e., time-varying ozone with all other forcings held at 1860). They found that ozone forcing produced similar or drier conditions over south-western Australia than well-mixed GHGs. In their results, changes over south-eastern Australia were unusual but not as clearly distinguished from internal variability as the changes in southwest Australia.

While the impacts of the Antarctic ozone depletion on the stratospheric and tropospheric circulations and the surface climate were still vigorously studied, scientists started noticing a sign of a pause or reversal of the ozone-depleting trend since 2000. For instance, Salby et al. (2011) extracted the Antarctic ozone trend by removing the ozone component driven by the interannual variability of the stratospheric circulations associated with the Antarctic polar vortex and the Quasi-Biennial Oscillation (QBO) and showed a clear sign of the rebound of the Antarctic ozone. From the various observational data sets and model simulations using the Whole Atmosphere Community Climate Model (WACCM), which is a fully coupled state-of-the-art interactive chemistry-climate model, Solomon et al. (2016) confirmed an increasing trend of the Antarctic ozone and a decreasing trend of the ozone hole in austral spring since 2000 and linked the change to the reductions of the ODSs.

Being consistent with this recovering trend in ozone, Banerjee et al. (2020) analysed three reanalysis data sets (ERA-5, JRA-5, and MERRA-2) and station data and reported that the positive trends in the SAM and the poleward expansion of the Hadley cell had been showing a pause or a sign of reversal in austral summer since 2000. Their simulations with the Canadian Earth System Model version 2 and the ensemble of climate-chemistry coupled models with all forcings, the ozone forcing, and the GHGs forcing showed that the recent reversal of the trend in the Antarctic circulation was largely driven by the ozone recovery, offsetting the trend driven by the increasing GHGs. Likewise, Zambri et al. (2021) showed the Antarctic polar cap was warmer, the vortex circling around Antarctica was weaker, and the geopotential height over Antarctica was higher during the warm seasons from late spring to summer in the period of 2001-2018 relative to the period of 1979-2001 according to two different reanalysis data sets (ERA-5 and MERRA-2) and the WACCM simulations that were run with the forcings of the GHG and ODSs.

Mindlin et al. (2021) focused on changes in the stratospheric polar vortex breakdown date due to the GHG *vs.* ozone trends. The GHG increase and the Antarctic ozone depletion both act to cool the lower stratosphere and cause the polar vortex to strengthen and persist throughout austral spring, therefore leading to a delay of the vortex breakdown. In their study, CMIP6 models suggest that the delaying trend of the Antarctic vortex breakdown date was nearly entirely explained by the equivalent effective stratospheric chlorine trend (representing the trend in the ODSs) for 1950-2000, but after 2000 most models show plateauing of the vortex breakdown date trend due to the cancellation between the decreasing ODSs and the increasing GHGs. They showed a considerable spread in the vortex breakdown date response to the degrees of global warming measured by the annual global mean surface temperature change relative to the 1950–1969 climatology. For instance, in the stratosphere a low level of global warming can still have a large effect on maintaining a delayed vortex breakdown date if its response to global warming is high. Conversely, if the vortex breakdown date response to global warming is low, ozone recovery will drive the trend of the vortex breakdown date during the 21st century regardless of the degree of warming. On the other hand, summer precipitation changes in the land areas of the SH are more complex than their responses to the vortex breakdown date changes in relation to the degrees of global warming or ODSs concentration level, warranting further investigation.

3. VARIABILITY

Stratospheric ozone has large year-to-year variability particularly in the polar regions in the late winter to spring season because of the tight linkage between ozone and polar vortex variabilities (e.g., Salby et al. 2011; Yook et al. 2020). For example, the ozone anomalies driven by significantly anomalous Antarctic polar vortex weakening (e.g., 1988 and 2002) or strengthening (e.g., 2015) were comparable to the magnitude of the ozone-depleting trend over the 1980s-90s (Salby et al. 2011; Solomon et al. 2016; Garny et al. 2022). Fogt et al. (2009) found with de-trended observational data that spring decreases in total ozone were associated with the occurrence of pSAM anywhere from one to four months later, and the opposite was true for spring increases in ozone and nSAM. The observational analyses of Son et al. (2013) and Bandoro et al. (2014) also demonstrated a significant correlation between Antarctic polar cap ozone and the SAM, resulting in substantial anomalies in rainfall and surface air temperatures in the SH, especially over Australia. However, because of the tight ozone-vortex coupling in spring and summer, it was not clear in those studies how much of the estimated ozone impact on the tropospheric circulation and the surface climate anomalies was purely due to the ozone independent of the stratospheric circulation.

To better understand the role of the internally-driven ozone anomalies in inducing the stratospheric Antarctic polar vortex, tropospheric circulation, and surface climate anomalies, Hendon et al. (2020) conducted 11-member ensemble forecast sensitivity experiments for spring 2002 when massive sudden stratospheric warming over Antarctica with a reversal of the vortex wind direction occurred and was accompanied by a significant increase of the Antarctic polar cap ozone (Stolarski et al. 2005). For this study, the Bureau of Meteorology's dynamical sub-seasonal to seasonal climate forecast system, ACCESS-S1 (Hudson et al. 2017), was initialised on the 1st of August, 2002, with high-quality observed atmosphere and ocean conditions. The ozone forcing was prescribed with monthly mean climatological ozone of 1994-2005 (control run) *vs.* with the realistic monthly mean ozone of 2002 (experimental run). The forecasts were verified in the following four months till the end of December 2002. The comparison of the control and experimental forecasts showed that the realistic ozone of 2002 characterised by its significantly higher-than-normal concentration over the Antarctic polar cap region significantly contributed to improving the prediction skill of the earlier-than-normal breakdown of the stratospheric polar vortex, the record strength of nSAM (by nearly doubling the strength), and the associated heat and dry extremes over Australia including parts of the state of Victoria in October 2002. These results suggest that polar stratospheric ozone variability is an important driver of the climate variability in the SH, which can be exploited to improve monthly to seasonal climate forecast skill at lead times beyond a month.

Using the GloSea-5 seasonal forecast system, which is the same system as ACCESS-S1 but initialised with different conditions and uses a different ensemble generation method, Oh et al. (2022) compared two sets of 18-member ensemble retrospective forecasts prescribed with monthly mean climatological ozone *vs.* realistic time-varying ozone above the 261 hPa level for the period of 2004-2020. The forecasts were initialised on the 1st of September, and two-week running averages of the forecasts were verified for the September-October period for the 17 years. Their forecast experiment results were consistent with those of Hendon et al. (2020) by demonstrating substantial reduction of forecast error in September and significant forecast skill improvement in October for the Antarctic tropospheric circulation, which represents the SAM, when realistic ozone concentrations were prescribed. Furthermore, the two-week mean surface air temperature over Australia (including Victoria) in the first half of October was remarkably better predicted at a lead time of up to 6 weeks with realistic ozone concentrations, which highlights a potential benefit of implementing an interactive chemistry scheme to incorporate realistic ozone information for skilful forecasts of sub-seasonal to seasonal climate anomalies, which has been under consideration for the future development of the ACCESS-S system in the Bureau of Meteorology.

Another recent topic of interest regarding ozone variability was the role of the Australian Black Summer bushfires (also called the Australian New Year's wildfires; ANY) in 2019-2020 in the ozone anomalies of 2020 and a subsequent occurrence of anomalous stratospheric polar vortex strengthening in late 2020. Yu et al. (2021) reported that from December 29, 2019, to January 4, 2020, Australian wildfires injected about 0.9 Tg of smoke into the stratosphere, which was the largest stratospheric smoke injection observed by satellites to date. The smoke was observed to spread over the entire SH, and the large-scale smoke (not individual plumes) climbed to 22 km in the first 3 months. Yu et al. (2021) conducted sensitivity simulations with and without the 2019-2020 Australian bushfire smoke with an assumption that smoke particles provide a surface reaction rate equal to that of volcanic sulfuric acid particles. The modelling results showed that the Black Summer smoke caused persistent negative ozone anomalies of 10–20 Dobson units in the SH mid- and high latitudes (40°S–90°S).

With satellite observation data of stratospheric aerosols, temperatures, and ozone, Rieger et al. (2021) confirmed both the magnitude as well as the latitudinal and seasonal patterns of the negative SH midlatitude ozone anomaly modelled by Yu et al. (2021), which, again, confirmed that these anomalies were induced by a chemical cause. The anomalously low ozone values were shown to be accompanied by record low polar stratospheric temperatures and a massive strengthening of the Antarctic polar vortex during November-December 2020, which were likely to occur in association with the reduction in radiative heating from the reduced ozone, at least in part. Further, Yook et al. (2022) documented a series of physically coherent linkages among the Australian bushfires in early 2020/the eruption of La Soufriere on Saint Vincent in April 2021, the ozone holes, and the subsequent pSAM and cooler-than-normal Australian summer temperatures, which were consistent with our descriptions of the mechanisms in this report. While these studies demonstrate that the ozone anomalies of 2020 were an important condition leading up to the massive strengthening of the Antarctic polar vortex and consequent strong pSAM that could contribute to the cooler climate over Australia, it is still unknown if the ozone anomaly was the trigger of the vortex strengthening in 2020.

4. SUMMARY & DISCUSSION

In this report, we have reviewed some papers that explain 1) the mechanisms of ozone anomalies to impact the SH stratospheric and tropospheric circulations, which ultimately result in anomalies in the Australian climate in spring and summer; 2) the depleting trend in the Antarctic ozone and its recovering trend since 2000 and associated trend reversals in the Antarctic polar vortex strength and temperature and the summer season SAM; and 3) the ozone's role in the variability and predictability of the SAM and Australian climate in spring-summer. It was clear through this review that whether it is the long-term trend of ozone or its year-to-year variability, ozone variation has a substantial impact on Australian spring to summer season climate anomalies. However, we have not found any detailed mechanism explained for the pathway of ozone to influence Australian temperature and rainfall other than that via modulating the location and intensity of the tropospheric midlatitude jet, which is depicted by the SAM. As mentioned in Section 2, Salby et al. (2011) noted the ozone components associated with not only the stratospheric polar vortex variability but also the QBO, which is the dominant mode of equatorial stratospheric wind variability with a time-scale of 2-3 years and has been reported to impact the Madden-Julian Oscillation (MJO) (e.g., Hendon and

Abhik 2018). Thus, the ozone-QBO relationship could be another pathway for the ozone anomalies to impact Australian climate if the ozone-driven temperature changes could modulate the tropical stratospheric winds of the QBO, which will be worth exploring in the future.

Regarding the ozone anomalies of the year 2020, which have been reported to be induced by the Australian Black Summer bushfires in the late December 2019 to early January 2020 period, we have pointed out that the studies in our review have not shown if the smoke-driven ozone depletion triggered the stratospheric circulation anomaly of late spring 2020 or only amplified it. This is an important question, especially for extended predictability of the stratospheric polar vortex variability and associated surface climate anomalies. To get an insight into this question, we have been conducting a forecast sensitivity experiment with realistic ozone concentrations of 2020 versus the climatological ozone, similar to the experiment of Hendon et al. (2020). The results of these experiments will be presented in our future work.

Another interesting fact we have become aware of from this literature review is that the Antarctic stratospheric polar vortex strengthening and cooling are driven not only by dynamical perturbations but also by chemical perturbations caused by volcanic eruptions and large-scale forest fire smoke that induce ozone anomalies, whereas the vortex weakening and warming are driven by dynamical perturbations. It will be useful to find out if this difference in causality would result in any difference in the predictability of the vortex strengthening versus weakening, which will affect the predictability of the SAM and Australian climate.

Finally, we have noted that in sub-seasonal to seasonal climate prediction systems and CMIP models, it is a common practice to have zonally averaged monthly climatology of ozone be prescribed rather than using 3-dimensional prognostic ozone in an interactive chemistry-climate scheme for the benefit of simplicity in the radiative and dynamical balance in climate simulations and for cheaper computational cost (e.g., Dennison et al. 2017; Johnson et al. 2018; Hendon et al. 2020; Oh et al. 2022). However, Crook et al. (2008) and Waugh et al. (2009) reported that the asymmetric ozone structure could cause a significant cooling in the stratosphere and upper troposphere with a magnitude comparable to that associated with the ozone-depleting trend in the 1980s-90s, and this impact of the zonal asymmetry could be greater when an Antarctic ozone hole was already present. In addition, Yook et al. (2020) found that the tropical lower stratospheric temperature could have twice greater variance with freely evolving ozone than with prescribed ozone. All these results point to the fact that the modelling studies that use zonally averaged ozone and/or prescribed ozone could have somewhat underestimated the role of ozone. We will have to bear this caveat in mind for our future modelling experiments and investigations on climate model projections to understand the role of ozone in climate variability or long-term changes.

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