1	Total Column Ozone Trends from the NASA Merged Ozone Time Series 1979 to 2021 Showing Latitude			
2	Dependent Ozone Recovery Dates (1994 to 1998)			
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### 17 Abstract

- 18 Monthly averaged total column ozone data  $\Omega_{MOD}(t,\theta)$  from the NASA Merged Ozone Data set
- 19 (MOD) were examined to show that the latitude-dependent,  $\theta$ , ozone depletion turnaround dates T<sub>A</sub>( $\theta$ )
- range from 1994 to 1998.  $T_A(\theta)$  is defined as the approximate date when the zonally average ozone
- 21 ceased decreasing.  $\Omega_{MOD}$  data used in this study were created by combining data from Solar
- 22 Backscattered Ultraviolet instruments (SBUV/SBUV-2) and the Ozone Mapping and Profiler Suite (OMPS-
- 23 NP) from 1979 to 2021. The newly calculated systematic latitude-dependent hemispherically
- asymmetric  $T_A(\theta)$  shape that currently does not appear in the suite of chemistry-climate models that are
- 25 part of the Chemistry-Climate Model Validation Activity CCMVal, the Chemistry-Climate Model
- 26 Validation Activity, which combines the effects of photochemistry, volcanic eruptions, and dynamics in
- 27 their estimate of ozone recovery. Trends of zonally averaged total column ozone in percent per decade
- 28 were computed before and after  $T_A(\theta)$  using two different trend estimate methods that closely agree,
- 29 Fourier Series Multivariate Linear Regression and linear regression on annual averages. During the
- 30 period 1979 to  $T_A(\theta)$  the most dramatic rates of SH ozone loss were  $P_D = -10.9 \pm 3\%$  per decade at
- 31 77.5°S and -8.5±0.9% per decade at  $65^{\circ}$ S, which is about double the NH rate of loss of P<sub>D</sub> = -5.6±4
- 32 %/decade at 77.5°N and 4.4±1%/decade at 65°N for the period 1979 to  $T_A(\theta)$ . After  $T_A(\theta)$ , there has
- 33 been an increase at  $65^{\circ}S$  of  $P_{D} = 1.6 \pm 1.4\%$  per decade with smaller increases from  $55^{\circ}S$  to  $25^{\circ}S$  and a
- small decrease at 35<sup>o</sup>N of -0.4±0.3 %/decade. Except for the Antarctic region, there only has been a
- 35 small recovery in the Southern Hemisphere toward 1979 ozone values and almost none in the Northern
- 36 Hemisphere.
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#### 40 **1.0 Introduction**

- 41 Ozone is a photolytically produced, photochemically destroyed, and dynamically distributed
- 42 atmospheric gas that plays a crucial role in protecting the planet from harmful ultraviolet (UV) radiation
- 43 from the sun. The atmospheric presence of bromine and the release of chlorine from the UV
- 44 dissociation of man-made chemicals, such as chlorofluorocarbons (CFCs), can break down the ozone
- 45 layer at all latitudes. This is especially the case in the Antarctic region where heterogeneous chemistry
- 46 on and within ice crystals and liquid droplets (Tritscher, et al., 2021) in polar stratospheric clouds PSCs
- 47 have a strong effect on the destruction of ozone during September and October (WMO, 2022; Tritscher,
- 48 et al., 2021; Solomon et al., 1986; 1999; 2016; Crutzen and Arnold, 1986; Khosrawi et al., 2011). As the
- 49 sun rises in Spring, chemically active nitrogen oxides, chlorine and bromine are released causing the
- 50 ozone hole to develop within the region enclosed by the polar vortex winds. The weak levels of sunlight
- 51 are sufficient to initiate and maintain the catalytic ozone loss photochemistry. In November and
- 52 December, the isolating polar vortex winds break down and the Antarctic ozone hole region back fills by
- 53 air exchange from southern mid-latitudes causing  $T_A(35^\circ S 65^\circ S)$  to be delayed compared to the
- 54 Northern Hemisphere NH mid-latitudes. The recurring annual ozone hole event triggered international
- action to limit the production and use of ozone-depleting substances (ODS) under the Montreal
- 56 Protocol, which has been successful in reducing the emission of these substances, slowing down the
- 57 depletion of the ozone layer globally, leading to a partial recovery in the Antarctic ozone hole region
- 58 (Solomon et al., 2016; Strahan and Douglass 2018). After the mid-1990s, several studies have reported
- 59 an increase in total column ozone (TCO), particularly in the mid to high latitudes of the Southern
- 60 Hemisphere, as well as a reduction in the size and depth of the Antarctic ozone hole starting in the late
- 61 1990s (Solomon et al., 2016; Stone et al., 2018; 2021, Weber et al., 2022).
- 62 The cessation of ozone decrease was first observed in the mid-1990s when satellite data showed a
- 63 stabilization and slight increase in ozone concentrations in the Antarctic ozone hole region. However,
- 64 the recovery was not significant enough to be considered a trend at that time (Strahan and Douglass
- 65 2018). In the early 2000s, further analysis of satellite and ground-based data showed that the rate of
- ozone depletion had slowed down. After the mid-1990's, the cessation of ozone depletion has been
- 67 most evident in the Southern Hemisphere SH polar region, where ozone depletion had been most
- severe. Ozone recovery has been slow or non-existent at other latitudes. Recently, Weber et al. (2022)
   showed reduction in ozone at all latitudes prior to 1995 and reported positive statistically significant
- 70 TCO trends from 1996-2020 at southern middle and high latitudes, and over the SH polar cap in
- 71 September. When dynamical terms were included in the regression, small positive trends were near the
- 72 2-standard deviation  $2\sigma$  threshold at northern mid- and high-latitudes, with no trend detected in the
- 73 tropics or over the NH polar cap.
- 74 Despite the success of the Montreal Protocol (Velders and Andersen, 2018), ozone concentrations
- continue to fluctuate, driven by natural and anthropogenic factors, such as, changes in solar radiation,
- stratospheric circulation, global warming, volcanic activity, and changing emissions of ozone precursors
- 77 (Dameris and Baldwin, 2012; Weber et al., 2022). The discussion by Dameris and Baldwin (2012)
- 78 explored possible effects of climate change on the dynamics of the atmosphere affecting ozone as ODSs
- read change, and particularly the change in the Brewer-Dobson circulation (Brewer, 1949; Dobson et al.,

1926) that transports ozone from an upwelling in the equatorial region into the stratosphere and to
 downwelling into mid- and high-latitudes.

82 A comparison of several atmospheric chemistry and dynamics model studies as part of the Chemistry-83 Climate Model Validation (CCMVal) Activity (Eyring, et al. 2010a, their Fig. 1; Dhomse et al., 2018; 84 Robertson et al., 2023) generally predict an ozone turnaround date  $T_A$  in the year 2000 with no 85 systematic latitude dependence. In particular, Robertson et al. (2023) shows latitude dependence of 86 long-term ozone recovery, but  $T_A = 2000$  for all cases. Quoting from the Sparc Report No. 5 (Eyring, et al. 87 2010b), "Common systematic errors in CCM results include: tropical lower stratospheric temperature, 88 water vapor, and transport; response to volcanic eruptions", which may affect the determination of  $T_A$ 89 as a function of latitude and time. The results of this study may provide a convenient metric for model 90 validation compared to ozone data T<sub>A</sub>.

91 This study will estimate new latitude dependent ozone recovery dates, or more accurately the dates of 92 cessation of ozone decrease,  $T_A(\theta)$  ranging from 1994 (equatorial region and 60°N to 70°N) to 1998  $(60^{\circ}\text{S} - 80^{\circ}\text{S})$ . The calculated T<sub>A</sub>( $\theta$ ) and ozone trends (%/decade) include the effects of volcanic 93 94 eruptions such as Mt. Pinatubo in 1991, dynamics, and atmospheric temperature changes. Ozone data 95 used in this study are a subset of the Merged Ozone Data MOD set  $\Omega_{MOD}(t)$  (1970 – 2021) starting in 96 1979 with the Nimbus-7 SBUV (Solar Backscattered Ultraviolet) satellite instrument. From 1979 to 2021, 97 the MOD data set was created by combining data from Solar Backscattered Ultraviolet instruments 98 (SBUV/SBUV-2) and the Ozone Mapping and Profiler Suite (OMPS-NP). Methods of calculating trends 99 from time series data are essential in the analysis of environmental and climate-related data. Here, we 100 discuss two independent methods to estimate linear trends: 1) linear regression of annual averaged 101 data and 2) Fourier time series decomposition or multivariate linear regression MLR (Ziemke et al., 102 2019) are discussed below. The two methods are compared and shown to give nearly identical results 103 over their mutual latitude range of validity, 65°S to 65°N. The MLR method is not used in the regions 104 poleward of the Arctic and Antarctic circles that have latitude dependent extended winter polar night. The advantage of the MLR method (Eq. 1), or that in Weber et al, 2002, is that it can be used to estimate 105 106 the effects of its individual components, while the annual average method can be used in the polar

107 regions to estimate sun lit ozone trends where there is latitude dependent extended winter night.

## 108 1.1 The Merged Ozone Data Set MOD

- Figure 1 (left panel) shows the MOD zonally averaged  $\Omega_{MOD}$  TCO data (Frith et al., 2014; 2020) set as a
- 110 function of latitude (5<sup>o</sup> latitude bands from 77.5<sup>o</sup>S to 77.5<sup>o</sup>N) and time (January 1979 to December
- 111 2021). Part of the Antarctic ozone hole (75°S to 80°S) is shown (blue color), and the high latitude
- maxima, North and South, (red color), with low values in the equatorial region. Figure 1 (right panel)
- shows the 42-year zonal and time averaged ozone amounts and the maxima and minima annual
- 114 envelopes as a function of latitude. Figure 1 shows the asymmetry in the monthly and zonally averaged
- 115 ozone data between the hemispheres, with the Northern Hemisphere NH having more ozone than the
- Southern Hemisphere SH at corresponding latitudes. Part of the asymmetry is driven by the Spring
- 117 Antarctic ozone hole backfilling in the SH summer.

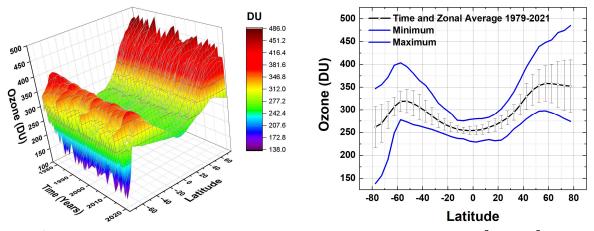


Fig. 1 Left: The zonally and monthly averaged  $\Omega_{MOD}$  data set 1979 – 2021 and -77.5° to 77.5°. Right: Time and zonal averaged ozone and its maxima and minima 1979 – 2021. Error bars are 1 standard deviation  $\pm 1\sigma$ .

- 118  $\Omega_{MOD}(t,\theta)$  provides a global view of ozone levels needed to track changes in ozone concentrations over
- time t for each latitude band  $\theta$ . The SBUV and OMPS-NP series of satellite instruments form the longest
- 120 (1979 to 2022) continuous global ozone  $\Omega_{MOD}(t,\theta)$  data record from a single instrument type. Merged
- ozone retrievals from the individual instruments use the version 8.7 retrieval algorithm (described by
- 122 Weber et al., 2022) as an extension of the version 8.6 algorithm (Bhartia et al., 2013; McPeters et al.,
- 123 2013; DeLand et al., 2012; Frith et al., 2017) specifically designed to improve cross calibrations between
- the later SBUV-type instruments in MOD starting from NOAA-16 in 2000. There were no external
- adjustments made to the ozone retrieval except for small high-altitude (> 35 km) diurnal corrections to
- account for different measurement times between satellites and varying measurement time of day as
- 127 individual satellite orbits slowly drift in equator crossing time. These adjustments are very minor in TCO
- 128 (Frith, personal communication). Data from each instrument are selected based on quality criteria
- outlined in Frith et al. (2014; 2020) and the data are averaged during periods when more than one
- 130 instrument was operational. The  $\Omega_{MOD}(t,\theta)$  are available as a function of latitude and month,
- 131 <u>https://acd-ext.gsfc.nasa.gov/Data\_services/merged/.</u>
- 132

133 Analysis of the long-term ozone time series has been looked at extensively with references given in Weber et al., 2022. Methods for estimating trends from an oscillating time series with several distinct 134 135 periodicities are well known (Ziemke et al., 2019; Stolarski et al. 1991;1992, Herman et al., 1993). For 136 ozone, one of the difficulties in trend estimation is that the early part of the time series shows a strong 137 ozone decrease at all latitudes that continued until the mid-1990s and then flattens out and shows 138 almost no recovery thereafter toward 1979 values. The  $\Omega_{MOD}$  time series has been used extensively in 139 ozone assessments and State of the Climate reports (e.g., WMO, 2022) and was recently compared to 140 several other merged total ozone records in Weber et al. (2022). The validity of the  $\Omega_{MOD}$  time series for 141 estimating ozone trends was further checked (See Appendix Figs. A1 to A3) in this study by showing 142 detailed comparisons between the deseasonalized  $\Omega_{\text{MOD}}$  time series with the deseasonalized MLS 143 (Microwave Limb Sounder) overlapping stratospheric ozone time series (2005 to 2023).

#### 144 **2.0 Trend Estimates from the MOD Ozone Data**

145 Multivariate Linear Regression MLR is a Fourier based method for analyzing atmospheric time series 146 data that decomposes the time series into its component parts, including trend, quasi-biennial

147 oscillation QBO, solar cycle, ENSO (El Nino Southern Oscillation), seasonality, and noise resulting in a

148 trend estimate and 2-standard deviation  $2\sigma$  uncertainty estimates (Ziemke et al., 2019). Calculated  $2\sigma$ 

149 uncertainties for the MLR trends include a first order autoregressive adjustment applied to the derived

150 residuals (Weatherhead et al., 1998).

151 Linear trend estimates for the long-term changes in  $\Omega_{MOD}(t, \theta_i)$  globally and as a function of latitude  $\theta_i$ 

- have been obtained using the multivariate linear regression (MLR) model (e.g., Randel and Cobb, 1994,
- and references therein). Trends B( $\theta_i$ ) were determined for  $\Omega_{MOD}(t, \theta_i)$  using Eqns. 1 and 2.
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$$\Omega_{MOD}(t, \theta_i) = A(\theta_i t) + B(\theta_i, t) \cdot t + C(\theta_i, t) \cdot QBO_1(t) + D(\theta_i, t) \cdot QBO_2(t) + E(\theta_i, t) \cdot SOlar(t) + R(t, \theta_i)$$
(1)

155

156 where t is the month index (t=1 to 516 months with data for 1979–2021),  $A(\theta_i t)$  is the seasonal cycle 157 coefficient,  $B(\theta_i,t)$  is the trend coefficient,  $C(\theta_i,t)$  is the first empirical orthogonal function (EOF) QBO 158 coefficient,  $D(\theta_i, t)$  is the second EOF QBO coefficient, both representing the major components of 159 the QBO variability,  $E(\theta_{i},t)$  is the ENSO coefficient,  $F(\theta_{i},t)$  is the solar cycle coefficient, and R(t) is the 160 residual error time series. The F10.7 cm solar flux monthly time series is used for the Solar(t) proxy, first 161 and second leading EOF QBO monthly time series proxies  $QBO_1(t)$  and  $QBO_2(t)$  are used for the QBO 162 component (Wallace et al., 1993), and Nino 3.4 (Oldenborgh et al 2021) is used for ENSO(t) (Nino 3.4: 163 https://www.ncei.noaa.gov/access/monitoring/enso/sst). QBO<sub>1</sub>(t) and QBO<sub>2</sub>(t) are nearly orthogonal 164 (correlation coefficient approximately zero) oscillating time series based on data with approximately a 165 2.3-year periodicity. A( $\theta_{l}$ ,t) involves 7 fixed constants while B( $\theta_{l}$ ,t) (and all other remaining coefficients) 166 involves 5 fixed constants for each  $\theta_{l}$ . The harmonic expansion for A(t) (similar for the other 167 coefficients) is (Eqn. 2).

168

$$A(t) = a(0) + \sum_{p=1}^{3} \left[ a(p) \cos(2\pi pt/365) + b(p) \sin(2\pi pt/365) \right]$$
(2)

169

where a(p) and b(p) are constants. Statistical uncertainties for A(t) and B( $\theta_i$ ) were derived from the calculated statistical covariance matrix involving the variances and cross-covariances of the constants (e.g., Guttman et al., 1982; Randel and Cobb, 1994).

173

174 In this study the Locally Weighted Scatterplot Smoothing Lowess(f) least-squares technique is used to 175 reduce oscillations in the time series data and to estimate  $T_A(\theta)$  where f = the fraction of data averaged 176 together (Cleveland, 1979 and Cleveland and Devlin, 1988).

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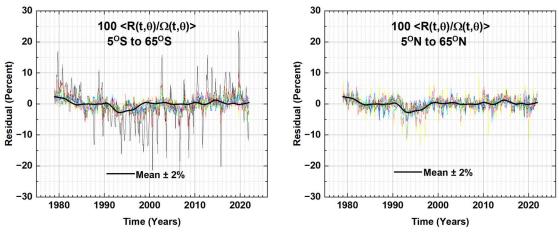


Fig. 2 The latitude average residual term from Eq. 1 in percent 100 <R(t,q<sub>i</sub>)/W<sub>MOD</sub>(t,q<sub>i</sub>)> The black line is the Lowess(0.1) fit (Cleveland, 1979) to the R(t,q) with an average error estimate of ±2%. The light-colored lines are each latitude's R(t,q) in a hemisphere  $0^{\circ} < \theta < 65^{\circ}$ .

180 The latitude average residual R(t) in percent of the MOD ozone amount  $(100 < R(t, \theta_i) / \Omega_{MOD}(t, \theta_i) >)$  is shown 181 in Fig. 2 for the SH and NH as an indication of how well Eq 1 is able to fit the  $\Omega_{MOD}(t, \theta_i)$  time series.

181 182

183 The SH R(t, $\theta$ ) is more variable than the NH with the largest variations arising in the 55°S and 65°S latitude 184 bands. On average Eq. 1 fits the original data  $\Omega_{MOD}(t,\theta_i)$  to within ±2%.

185

The linear deseasonalized trend results  $B(\theta_i)$  are obtained for 14 latitude bands  $\theta_i$  (centered on 65°S to 65°N). The latitudinal trends  $P_D(\theta_i)$  are expressed in %/Decade given by Eq. 3, where the denominator D is either the time average < $\Omega$ > of the area weighted global ozone average (Fig 1) or the time average  $D(\theta_i)$ = <  $\Omega_{MOD}(t, \theta_i)$ > for each latitude band over the considered period. The whole year period considered is 1979 – 2021.

191

$$P_{D}(\theta_{i}) = 1000 B(\theta_{i}) / D(\theta_{i}) \qquad (\% / \text{Decade})$$
<sup>(3)</sup>

192

193 In the second method, the trend is estimated using annual integrals (annual averages) that remove the 194 seasonality and other short-term oscillations but ignore longer term oscillations such as the 28-to-29-195 month QBO cycle and the average 11.3-year solar cycle. A comparison of the two trend estimating 196 methods is shown in Fig. 3 for the entire 1979 to 2021 period showing that they agree quite closely, but 197 that the annual average method has slightly larger two standard deviations  $2\sigma$  than the MLR method. 198 199 The MLR method (Eqns. 1 and 2) are not applied poleward of the Arctic and Antarctic circles where 200 latitude dependent extended winter night periods occur. Additional latitude dependent terms of varying 201 periods would be needed for latitudes greater than 70°. The annual average method does not have 202 these complications. 203

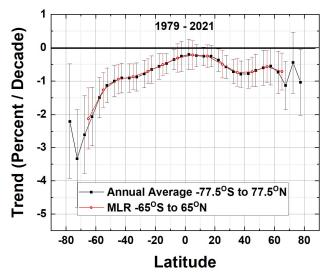


Fig. 3 The ozone trend  $P_D(\theta)$  for the entire period 1979 – 2021 for two methods, MLR and Annual Average. The latitude grids for the two methods are offset to show the agreement in the trends and  $2\sigma$  error bars.

The Fig. 3 estimation of linear long-term trends since 1979 is misleading, since ozone showed significant annual declines until the mid-1990s and then increased slightly thereafter, meaning the average longterm time series is non-linear. The usual procedure is to determine linear trends separately before and after the turnaround dates  $T_A$  (Weber et al., 2022). However, as is shown later, there is no single turnaround date applicable to all the latitudes between 80°S and 80°N. Instead, there is a range spanning 1994 to 1998.

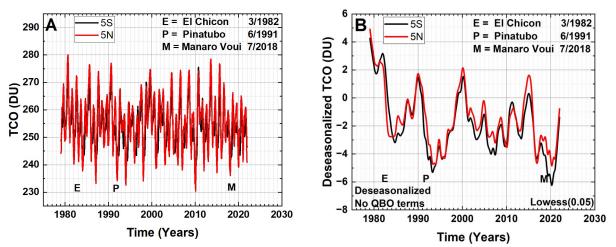


Fig. 4. A.  $\Omega_{MOD}$  time series for  $\theta = 5^{\circ}N$  and  $5^{\circ}S$ . B. The deseasonalized TCO time series for  $\theta = 5^{\circ}N$  and  $5^{\circ}S$  without removing QBO effects (Eq. 1). The approximate dates are shown of volcanic eruptions that injected large amounts of SO<sub>2</sub> into the stratosphere leading to minima approximately 1 year later.

- Figure 4A shows the  $\Omega_{MOD}$  time series for 5°S and 5°N, and Fig. 4B the deseasonalized and smoothed
- 213 (Lowess(0.05))  $\Omega_{MOD}$  time series. After deseasonalizing, but not removing QBO effects (Eq. 1), both the
- 214 2.3-year QBO oscillation and the reduced ozone effects from volcanic eruptions, are shown in Fig. 4B.
- Some volcanos (e.g., from El Chicon March 1982, Mt. Pinatubo June 1991, and Manaro Voui July 2018)
- 216 inject significant amounts of SO<sub>2</sub> into the lower stratosphere leading to the formation of aerosols that
- reduce UV light and the production of ozone, especially in the equatorial region.
- Figure 5 shows the Lowess(0.3) fits (black curves) to the  $\Omega_{MOD}$  data for four sample latitude bands 55°S,
- $45^{\circ}$ S,  $55^{\circ}$ N, and  $45^{\circ}$ N that tracks the longer-term changes in the  $\Omega_{MOD}$  time series. Also shown are
- examples of f = 0.1 (red) and f = 0.05 (blue dots). The Lowess(0.05) fit (blue dots) shows considerable
- structure with a minimum in 1993 that is likely related to the Mt. Pinatubo eruption and a modest El
- 222 Nino effect in 1991-1992. The estimated values of T<sub>A</sub> for f = 0.1 and 0.05 can differ by 6 months from that
- determined when f=0.3 because of short term oscillations. The Lowess(0.3) degree of smoothing
- 224 removes most of the short-term effects on ozone such as QBO and those from volcanic eruptions from
- 225 El Chichon (1982) and Mt. Pinatubo (1991), both well before the earliest estimated T<sub>A</sub> in 1994.

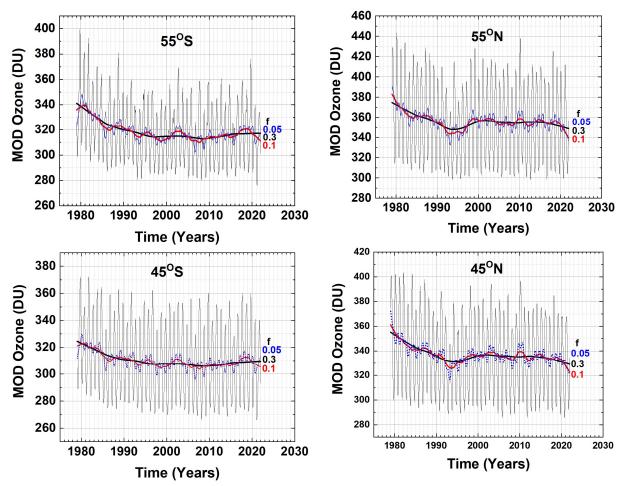


Fig. 5  $\Omega_{MOD}$  in four latitude bands and Lowess(0.3) fitting functions (f = 0.3, black lines). Examples of different f = 0.1 (Red) and 0.05 (blue dots) are shown at 45°S and 45°N. Note the slight downturn since 2010 in the Lowess(0.3) at 45°N and 55°N.

- Figure 6 shows the Lowess(0.3) fits to the  $\Omega_{MOD}$  data (1979 to 2021) for 16 latitude bands, -75° <  $\theta$  < 75°
- 227 on an expanded ozone scale. Each of the Lowess(0.3) plots for the various latitudes shows different
- periods of ozone decrease and subsequent turnaround  $T_A(\theta)$  after the mid-1990's. Use of expanded
- ozone scales appears to show a sharp downturn after 2010 at some latitudes (25°N to 75°N). As shown
- later, the apparent downturns in the Lowess(0.3) fit to  $\Omega_{MOD}$  after 2010 are not yet statistically
- 231 significant in trend estimates from  $\Omega_{MOD}$  as an indicator of long-term ozone decrease.

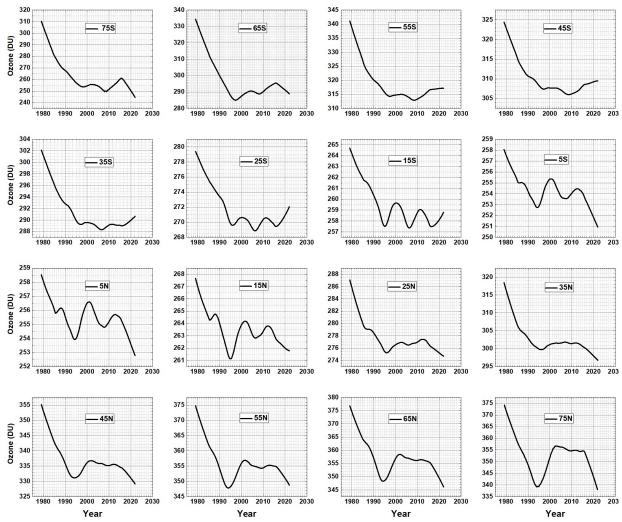


Fig. 6 Lowess(0.3) fits to the  $\Omega_{MOD}$  data for 16 latitude bands used to determine  $T_A(\theta)$ . Note that the ozone scale varies for each latitude.

- Figure 7 shows the turnaround dates  $T_A(\theta)$  that are obtained by taking the 1<sup>st</sup> derivatives of Fig. 6 data
- and finding the zero-crossing time corresponding to the appropriate minimum value in Fig. 6. The exact
- turnaround dates determined have a precision of ±0.1 years and an accuracy of ±0.5 years. The ±0.5
- uncertainty does not affect the calculation of trends before and after the estimated  $T_A(\theta)$ . What is
- interesting is that some of the turnaround dates in Fig. 7 are separated by over 4 years and are strongly
- asymmetric between the hemispheres. Figure 7 shows a near symmetry for early turnaround dates

- 238 1994-1996 for low latitudes between ±25° that corresponds to the Brewer-Dobson ozone upwelling
- region (Brewer et al., 1926; Dobson, 1949; Butchart, 2014) where most of the ozone is created by
- sunlight and then transported poleward. At poleward latitudes, the turnaround dates are quite
- different, with a delayed date, 1997, at high SH latitudes  $(35^{\circ}S 65^{\circ}S)$ , 1998 at 75°S compared to 1994
- at high NH latitudes (45°N to 75°N).
- 243 The  $T_A$  delay to 1997 for latitudes  $35^{\circ}S 65^{\circ}S$  follows the delayed recovery of ozone depletion within
- the Spring Antarctic Ozone Hole (Solomon, 1990; Stone et al., 2021, their Fig. 3; Bodeker and Kremser,
- 245 2021, their Figs. 6 and 9) and backfilling (air exchange with lower latitude ozone-rich air) during the
- summer months after the polar vortex winds break down in October November.
- 247

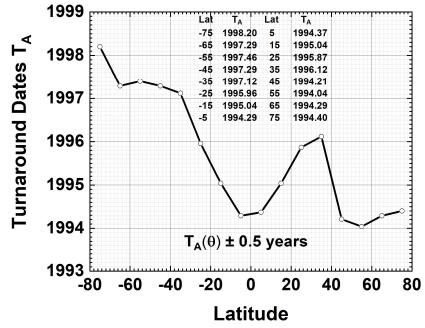


Fig 7 Turnaround dates  $T_A(\theta)$  as a function of latitude from Fig.6 with an estimated accuracy of ±0.5 years based on the analysis in Fig. 5.

249 The general  $T_A(\theta)$  pattern shown in Fig. 7 should appear in model calculations as a signature of the

combined effects of photochemistry, dynamics, and volcanic eruptions on the cessation of decreasingozone in the mid-1990s.



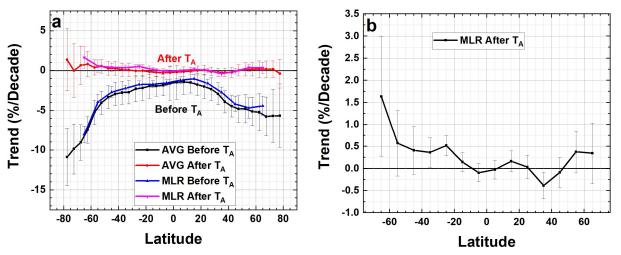


Fig. 8a Ozone trends  $P_D(\theta)$  (percent per decade) using the MLR and Annual Average methods before and after  $T_A(\theta)$ . 6b A magnified version of the MLR estimated trends after  $T_A$  with  $2\sigma$  uncertainties.

253	Trends (linear slopes)	$P_{\rm D}(\theta)$ in percent per de	ecade are estimated (Egn. )	3) for the separate periods before

and after  $T_A(\theta)$  in each latitude band (Fig. 8) and for the entire period (Fig. 3). The linear slopes obtained

by the two methods, MLR and annual average closely agree (Figs. 3 and 8) with the annual average

256 method extended to polar latitudes (Fig. 8a). Table 1 contains the data from Figs. 8a and 8b.

Table 1 MLR Trends (%/decade) $\pm 2\sigma$						
Latitude	$P_D$ Before $T_A$	$P_D$ After $T_A$				
-65	-8.04±1.1	1.64±1.4				
-55	-3.93±1.0	0.57±0.7				
-45	-2.69±0.7	0.41±0.5				
-35	-2.22±0.4	0.36±0.3				
-25	-1.75±0.5	0.52±0.2				
-15	-1.71±0.4	0.15±0.2				
-5	-1.54±0.4	-0.10±0.2				
5	-1.21±0.4	-0.03±0.2				
15	-1.01±0.6	0.16±0.2				
25	-1.61±0.5	0.03±0.3				
35	-2.71±0.6	-0.39±0.3				
45	-4.20±1.0	-0.09±0.3				
55	-4.67±1.3	0.38±0.5				
65	-4.43±1.2	0.35±0.7				

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The latitude dependent trends derived by Weber et al. (2022) using 1996.5 as the approximate  $T_A$  (their Fig. 3) agree within error bars with the trends shown in Fig. 8 for all latitudes but they suggest  $T_A = 2000$ for the polar regions. The trends also agree within error bars with those in WMO (2022). As mentioned

- 261 earlier, the trend estimates are not very sensitive to the exact  $T_A$ , but the shape of  $T_A(\theta)$  should be a 262 model validation marker contained in model calculations for all effects, not just ODSs.
- The delayed (1997) Southern Hemisphere mid and high latitude values of T<sub>A</sub> are caused by coupling to
  the increasing Antarctic spring ozone loss after 1979 until a recovery starting in about 1998-2000
  (Solomon et al., 2016). The mid and high latitude, from 35°S to 65°S, delay is caused by the summer
- 266 mixing of ozone poor air from the Antarctic region with SH midlatitude ozone-rich air once the polar
- 267 vortex winds break down in November-December.
- 268
- 269 The asymmetry between the Arctic and Antarctic is caused by the lower winter Antarctic temperatures
- 270 (-80<sup>o</sup>C) leading to the formation of low altitude clouds containing ice crystals along with the isolating
- 271 Antarctic polar vortex winds (Solomon et al., 2007; 2016). In the spring sunlight the ice and water
- droplets (Tritscher, et al., 2021) release ODS and depletes ozone to a monthly average of about 155 DU.
- 273 During the summer, air exchange with ozone rich air from lower latitudes comes into the polar latitudes and fill
- in the ozone layer above Antarctica (monthly average about 300 DU. Smaller but significant ozone losses
- 275 occurred in the Artic region caused by occasional low temperatures and ODSs. The Arctic does not
- 276 routinely have the low temperatures needed for winter ice clouds nor does it have the persistent
- isolating polar vortex winds because of wave action forced by the land topography The latitude band at
- 278 75<sup>o</sup>N (Fig.1) has the highest amount of monthly average winter ozone 450±25 DU that decreases to
- 279 290±20 DU monthly average during the summer that are comparable to mid-latitude values. The result
- is earlier values of T<sub>A</sub> in the NH compared to the SH. The NH T<sub>A</sub> is earlier than the 1997 minimum in
- 281 stratospheric halogens (Weber et al., 2022; Newman et al., 2007). Note that T<sub>A</sub> is not the time of the
- start of recovery, but rather the time for the end of rapid ozone decrease.
- 283
- 284 Before the SH T<sub>A</sub>, total column ozone decreased at a rate of P<sub>D</sub>= -10.9 $\pm$ 3.6% at 77.5°S and -8.0 $\pm$ 1.1% per 285 decade at 65°S, during the period from 1979 to 1997 with smaller decreases from 55°S to 25°S (Fig. 8a). 286 After the turnaround period T<sub>A</sub>, ozone at 65°S increased at P<sub>D</sub> = 1.6 $\pm$ 1.4%/decade based on the MLR 287 method. After T<sub>A</sub>, most other latitudes (Fig. 8b) show stationary ozone amounts within 2 $\sigma$ . In the NH the
- decreases were smaller than in the SH before  $T_A$  because of the absence of an Arctic ozone hole region.
- 289 At 77.5<sup>o</sup>N was  $P_D = -5.6 \pm 4\%$ /decade and at 65<sup>o</sup>N  $P_D = -4.4 \pm 0.35\%$ /decade.
- An analysis of ozone trends prior to the start of reliable satellite data in late 1978 showed that the annual rate of ozone loss (%/Year) increased after 1978 (Staehelin et al., 2001). Based on the first derivatives of the data in Fig. 6, the maximum annual rate of ozone reduction occurred in 1979 and 1980 in the NH and SH (Fig. 9) except for 65°N in 1992 where the rate of loss is -8.75%/Year. The loss rates range from -20.6 %/Year at 75°S to 2.39 %/Year at 5°N. A smaller loss rate occurred for 35°N to 75°N where the loss rate is almost constant between 8%/Year and 10%/Year compared to the larger SH loss rates caused by the presence of the springtime Antarctic ozone hole.

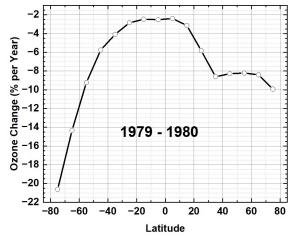


Fig. 9 The percent change in ozone per year in 1979 or 1980



298 The Lowess(0.3) plots in Fig. 6 suggest that  $\Omega_{MOD}$  has been declining since approximately 2010 from 5<sup>o</sup>S

299 to 65°N but still increasing from 45°S to 65°S (Fig. 6). However, computing the trends (Fig. 10) from

300  $\Omega_{MOD}(t,\theta)$  using either the MLR (Eq. 1) or annual average methods suggest that the declines in ozone

from  $25^{\circ}$ S to  $65^{\circ}$ N are not yet significant at the  $2\sigma$  level over the period 2010 - 2021.

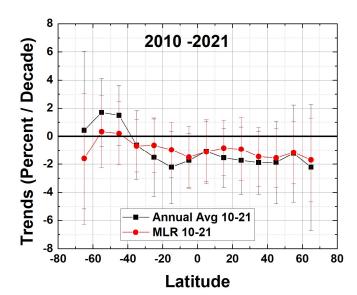


Fig. 10 Ozone trends  $P_D(\theta)$  (Percent per Decade) for the period 2010 – 2021 for the Annual Average and MLR methods applied to  $\Omega_{MOD}(t,\theta)$ .

- 303 Comparing deseasonalized  $\Omega_{MOD}(t,\theta)$  with deseasonalized Microwave Limb Sounder MLS (see Appendix
- Figs. A1, A2, and A3) Stratospheric Ozone from 2005 to 2021 shows small average (Lowess(0.3))
- differences that are within ±1DU except for 2021 when the differences at both 65°S and 65°N are about
- 306 -2.5DU. This suggests that the calibrations of the later SBUV-2 and OMPS-NP instruments are stable. For

- 307 2016 to 2018,  $\Omega_{MOD}$  is obtained from NOAA-19 SBUV plus OMPS-NP and from just OMPS-NP since 2018.
- Figure A3 suggests that there was a decrease in tropospheric ozone in 2020 that may correspond to reduced economic activity during the COVID-19 pandemic.
- Age of air AoA is a measure of how long a parcel of air resides in the stratosphere after it leaves the
- troposphere (Linz et al., 2016; Ploeger et al, 2021). A comparison of T<sub>A</sub> with AoA estimates from the
- relatively inert tracer gas CO<sub>2</sub> (Fig. 11) for the altitude range near the ozone maximum (approximately
- 20 km) vs latitude (based on Waugh and Hall, 2002, their Fig. 6a and Ploeger et al, 2021 their Fig. 10a)
- shows near symmetry between the hemispheres with the shortest AoA in the equatorial region. The
- turnaround dates T<sub>A</sub> in Fig. 6 are also symmetric in the equatorial zone corresponding the upwelling
   Brewer Dobson circulation and the smaller AoA. This suggests that the combined effects of chemistry
- and dynamics on ozone amounts are similar between ±25°. The precursors to ODS are also lifted into
- 318 the equatorial stratosphere and transported towards the polar regions (Newman et al., 2004; 2007)
- 319 where they can be photo-dissociated into ODS. Ozone at higher latitudes, NH and SH, with longer AoA,
- 320 will be dependent on transported ozone and ODS and their photochemistry, and especially the different
- 321 dynamics and chemistry in the Arctic and Antarctic regions.

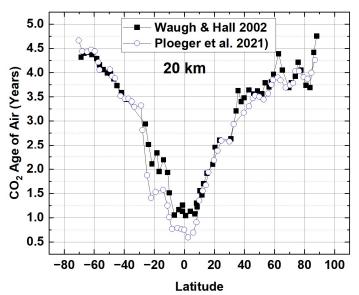


Fig. 11 Age of air derived from CO<sub>2</sub> data (Waugh and Hall, 2002; Ploeger et al., 2021)

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### 329 3.0 Summary

The monthly averaged Merged Ozone Data set  $\Omega_{MOD}$  (2.5° latitude bands, 77.5°S to 77.5°N) from 1979 330 to 2021 were averaged into 10<sup>o</sup> latitude bands 75<sup>o</sup>S <  $\theta$  < 75<sup>o</sup>N. A smoothed  $\Omega_{MOD}$  version based on 331 Lowess(0.3) was used to determine the approximate dates of the latitude dependent ozone end of 332 333 ozone decrease date  $T_A(\theta)$  ranging from 1994 to 1998 with an error estimate of ±0.5 years. The 334 systematic hemispherically asymmetric latitude dependent pattern  $T_A(\theta)$  should appear in atmospheric models that combine the effects of volcanic eruptions, photochemistry, and dynamics in their estimate 335 336 of the end of ozone decrease. An examination of model studies that are part of CCMVal shows a nearly 337 uniform  $T_A = 2000$ , suggesting that the several models' chemistry and dynamics including volcanic 338 effects are incomplete. The hemispheric asymmetry is caused by the formation of the annual Spring Antarctic ozone (monthly spring average about 155 DU) hole with persistent isolating polar vortex winds 339 340 followed by the summer mixing with mid-latitude ozone rich air (December average about 300 DU). The 341 Arctic region does not form a large spring ozone hole, nor does it have sustained isolating polar vortex winds. Instead at 75<sup>o</sup>N (Fig. 1) it has the highest amount of monthly average winter ozone 450±25 DU 342 343 that decreases to 290±20 DU monthly average during the summer. Trends of ozone  $P_D(\theta)$  in percent per decade were computed before and after the latitude dependent  $T_A(\theta)$  using two different methods, MLR 344 345 and annual averages, that closely agree over their mutual latitude range of validity, 65°S to 65°N. The annual average method can extend into polar latitudes. The most dramatic rates of ozone loss were  $P_{\rm D}$  = 346 -10.9 $\pm$ 3.6% decade at 77.5°S and -8.0 $\pm$ 1.1%/decade at 65°S, which is about double the rate of loss of P<sub>D</sub> 347 348 = -5.7 $\pm$ 4 %/decade at 77.5°N and -4.4 $\pm$ 1.2% per decade at 65°N. During the period after T<sub>A</sub> to 2021, there has been a small increase at latitudes in the SH from 25°S to 65°S with the largest value being 349 350 1.6±1.4% per decade at 65°S. Aside from the small increases in the SH region there has been no 351 statistically significant ozone recovery toward 1979 values, just an almost constant ozone amount after 352  $T_{A}(\theta)$ . The largest annual rate of ozone decrease occurred near the beginning of the SBUV data record, **1979**, showing large high latitude losses of -20.6 %/Year at 75°S caused by the springtime Antarctic 353 ozone hole compared to a smaller Arctic loss of -9.9%/Year at 75°N. During the period 2010 to 2021, 354 355 there has been a small apparent decrease in ozone amount in  $\Omega_{MOD}$  that is not yet statistically significant 356 at the 2-standard deviation level. A comparison between  $\Omega_{MOD}$  and MLS stratospheric column ozone 357 shows small systematic negative differences in 2020 that mostly recovered in 2021 except near the 358 equator. This suggests that there is no statistically significant instrumental calibration drift between 359  $\Omega_{MOD}$  TCO and MLS stratospheric ozone.

360

#### 362 Appendix

- 363 The MOD TCO data record since 2018 is obtained from OMPS-NP, which appears to show decreasing
- 364 TCO (Fig. 6). Because of this, the deseasonalized  $\Omega_{MOD}$  are compared with MLS (Microwave Limb
- Sounder) deseasonalized stratospheric column ozone for the period 2004 to 2021 to look for calibration
- 366 drifts in the  $\Omega_{MOD}$  time series. The question addressed here is not the absolute agreement between  $\Omega_{MOD}$
- 367 and the MLS mostly stratospheric ozone column, but rather if there is a systematic drift between the
- two data sets after 2016. Figures A1 and A2 show that the difference between the two deseasonalized
- time series for latitudes from  $65^{\circ}$ S to  $65^{\circ}$ N and for the entire period 2005 2021. Of interest is the
- period 2016 to 2021 when  $\Omega_{\text{MOD}}$  was derived using NOAA-19 SBUV plus OMPS-NP 2016 2018 and from
- 371 OMPS-NP since 2018.

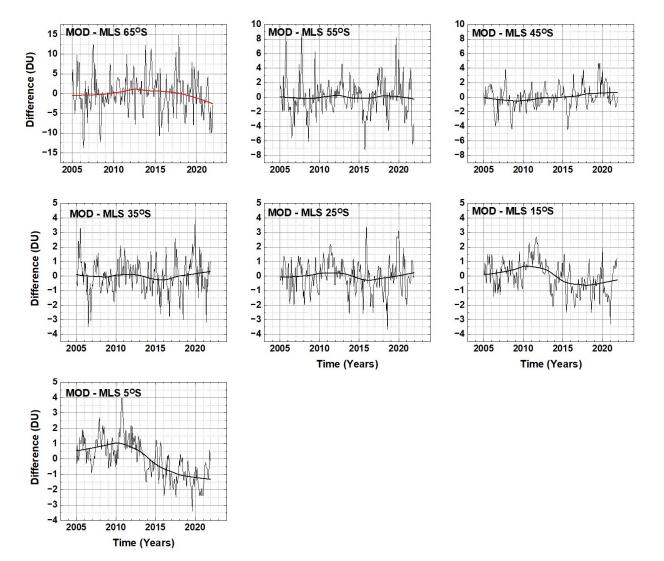


Fig. A1 A comparison of deseasonalized  $\Omega_{\rm MOD}$  with deseasonalized MLS stratospheric column ozone for 65°S to 5°S.



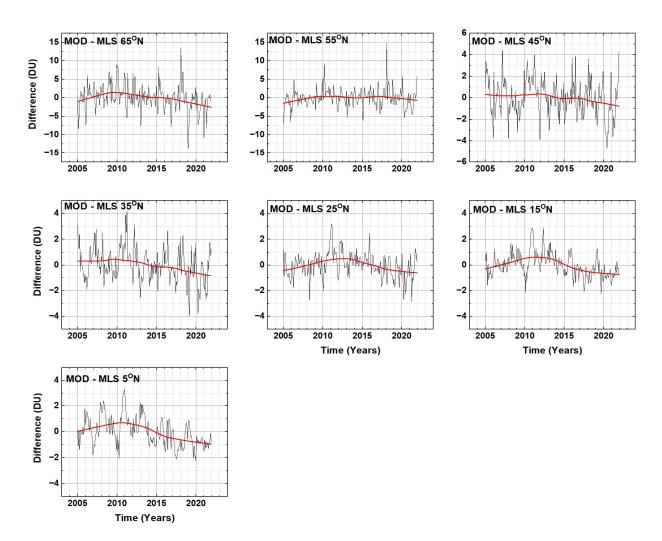


Fig. A2 A comparison of deseasonalized MOD total ozone with deseasonalized MLS stratospheric column ozone for 5°N to 65°N. Variations of ±3DU are within the MOD merged record uncertainties.

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The differences in Figs A1 and A2 between  $\Omega_{MOD}$  and MLS since 2016 are not statistically significant at the  $2\sigma$  level. Variations of ± 3DU are within the  $\Omega_{MOD}$  merged record uncertainties.

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378 Since both MOD and MLS time series were deseasonalized, the mean values would be zero unless there

379 were changes in tropospheric ozone or instrument calibration drift. The differences are summarized in

Fig. A3 along with the  $2\sigma'$ , ( $\sigma'$  = standard deviation from the mean) error bars estimated from the

381 average of each deseasonalized time series. In 2020 there appears to be a systematic change in <MOD –

382 MLS> that may be a reduction in tropospheric ozone amount of about 3 DU caused by the economic

slowdown associated with COVID-19 (Ziemke et al, 2022). The systematic change mostly recovered in

2021 (Fig. A3) except for -1DU near the equator (-5°S to 15°N).

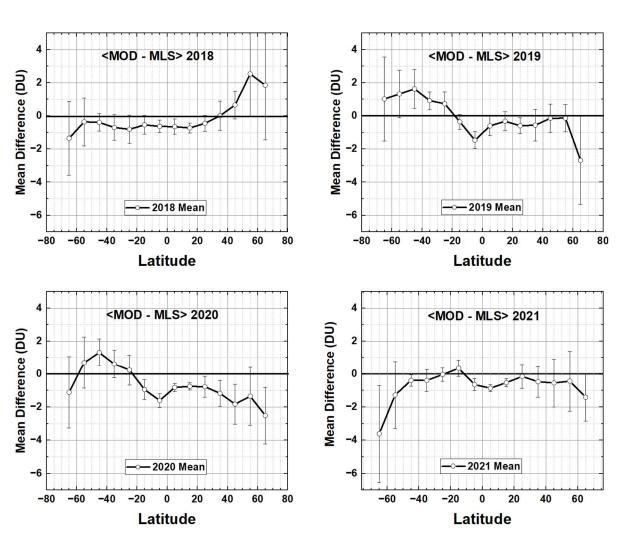


Fig. A3 Annual average <MOD – MLS> for the years 2018 to 2021. Error bars are  $2\sigma'$ , where  $\sigma'$  = standard error of the mean estimated from the average of the deseasonalized time series for each year shown in Figs. A1 and A2.

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537 Author contribution:

- 538 Jay Herman is responsible for writing the text, the annual integral trend calculations, and all the
- 539 figures. Jerald Ziemke supplied the MLR trend calculations and the comparison with MLS. Richard
- 540 McPeters supplied the MOD ozone as a continuous function of time from 1979 to 2021 for each
- 541 latitude band.
- 542 Data Availability
- 543 The original data used are publicly available in an ASCII format.
- 544 <u>https://acd-ext.gsfc.nasa.gov/Data\_services/merged/</u>
- 545 and processed data in Excel format
- 546 <u>https://avdc.gsfc.nasa.gov/pub/DSCOVR/JayHerman/MOD\_Ozone\_Trends/</u>
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# 549 **Competing interests**:

- 550 The authors declare that they have no conflict of interest.
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