

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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6 **Changes in response to Reviewer 1 are marked in **Green**.**

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21 **Abstract**

22 Monthly averaged total column ozone data $\Omega_{\text{MOD}}(t, \theta)$ from the NASA Merged Ozone Data set
23 (MOD) were examined to show that the latitude-dependent, θ , ozone depletion turnaround dates $T_A(\theta)$
24 range from 1994 to 1998. $T_A(\theta)$ is defined as the approximate date when the zonally average ozone
25 ceased decreasing. Ω_{MOD} data used in this study were created by combining data from Solar
26 Backscattered Ultraviolet instruments (SBUV/SBUV-2) and the Ozone Mapping and Profiler Suite (OMPS-
27 NP) from 1979 to 2021. The new calculated systematic latitude-dependent **hemispherically asymmetric**
28 $T_A(\theta)$ shape should appear in atmospheric models that combine the effects of photochemistry, **volcanic**
29 **eruptions**, and dynamics in their estimate of ozone recovery. Trends of zonally averaged total column
30 ozone in percent per decade were computed before and after $T_A(\theta)$ using two different trend estimate
31 methods that closely agree, Fourier Series Multivariate Linear Regression and linear regression on
32 annual averages. During the period 1979 to $T_A(\theta)$ the most dramatic rates of SH ozone loss were $P_D = -$
33 $10.9 \pm 3 \%$ per decade at 77.5°S and $-8.5 \pm 0.9\%$ per decade at 65°S , which is about double the NH rate of
34 loss of $P_D = -5.6 \pm 4 \%$ /decade at 77.5°N and $4.4 \pm 1\%$ /decade at 65°N for the period 1979 to $T_A(\theta)$. After
35 $T_A(\theta)$, there has been an increase at 65°S of $P_D = 1.6 \pm 1.4\%$ per decade with smaller increases from 55°S
36 to 25°S and a small decrease at 35°N of $-0.4 \pm 0.3 \%$ /decade. Except for the Antarctic region, there only
37 has been a small recovery in the Southern Hemisphere toward 1979 ozone values and almost none in
38 the Northern Hemisphere.

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42 1.0 Introduction

43 Ozone is a photolytically produced, photochemically destroyed, and dynamically distributed
44 atmospheric gas that plays a crucial role in protecting the planet from harmful ultraviolet (UV) radiation
45 from the sun. The atmospheric presence of bromine and the release of chlorine from the UV
46 dissociation of man-made chemicals, such as chlorofluorocarbons (CFCs), can break down the ozone
47 layer at all latitudes. This is especially the case in the Antarctic region where heterogeneous chemistry
48 on and within ice crystals and liquid droplets (Tritscher, et al., 2021) in polar stratospheric clouds PSCs
49 have a strong effect on the destruction of ozone during September and October (WMO, 2022; Tritscher,
50 et al., 2021; Solomon et al., 1986; 1999; 2016; Crutzen and Arnold, 1986; Khosrawi et al., 2011). As the
51 sun rises in Spring, chemically active nitrogen oxides, chlorine and bromine are released causing the
52 ozone hole to develop within the region enclosed by the polar vortex winds. The weak levels of sunlight
53 are sufficient to initiate and maintain the catalytic ozone loss photochemistry. In November and
54 December, the isolating polar vortex winds break down and the Antarctic ozone hole region back fills by
55 air exchange from southern mid-latitudes causing $T_A(35^{\circ}\text{S} - 65^{\circ}\text{S})$ to be delayed compared to the
56 Northern Hemisphere NH mid-latitudes. The recurring annual ozone hole event triggered international
57 action to limit the production and use of ozone-depleting substances (ODS) under the Montreal
58 Protocol, which has been successful in reducing the emission of these substances, slowing down the
59 depletion of the ozone layer globally, leading to a partial recovery in the Antarctic ozone hole region
60 (Solomon et al., 2016; Strahan and Douglass 2018). After the mid-1990s, several studies have reported
61 an increase in total column ozone (TCO), particularly in the mid to high latitudes of the Southern
62 Hemisphere, as well as a reduction in the size and depth of the Antarctic ozone hole starting in the late
63 1990s (Solomon et al., 2016; Stone et al., 2018; 2021, Weber et al., 2022).

64 The cessation of ozone decrease was first observed in the mid-1990s when satellite data showed a
65 stabilization and slight increase in ozone concentrations in the Antarctic ozone hole region. However,
66 the recovery was not significant enough to be considered a trend at that time (Strahan and Douglass
67 2018). In the early 2000s, further analysis of satellite and ground-based data showed that the rate of
68 ozone depletion had slowed down. After the mid-1990's, the cessation of ozone depletion has been
69 most evident in the Southern Hemisphere SH polar region, where ozone depletion had been most
70 severe. Ozone recovery has been slow or non-existent at other latitudes. Recently, Weber et al. (2022)
71 showed reduction in ozone at all latitudes prior to 1995 and reported positive statistically significant
72 TCO trends from 1996-2020 at southern middle and high latitudes, and over the SH polar cap in
73 September. When dynamical terms were included in the regression, small positive trends were near the
74 2-standard deviation 2σ threshold at northern mid- and high-latitudes, with no trend detected in the
75 tropics or over the NH polar cap.

76 Despite the success of the Montreal Protocol (Velders and Andersen, 2018), ozone concentrations
77 continue to fluctuate, driven by natural and anthropogenic factors, such as, changes in solar radiation,
78 stratospheric circulation, global warming, volcanic activity, and changing emissions of ozone precursors
79 (Dameris and Baldwin, 2012; Weber et al., 2022). The discussion by Dameris and Baldwin (2012)
80 explored possible effects of climate change on the dynamics of the atmosphere affecting ozone as ODSs
81 change, and particularly the change in the Brewer-Dobson circulation (Brewer, 1949; Dobson et al.,

82 1926) that transports ozone from an upwelling in the equatorial region into the stratosphere and to
 83 downwelling into mid- and high-latitudes.

84 This study will estimate new latitude dependent ozone recovery dates, or more accurately the dates of
 85 cessation of ozone decrease, $T_A(\theta)$ ranging from 1994 (equatorial region and 60°N to 70°N) to 1998
 86 (60°S – 80°S). The calculated $T_A(\theta)$ and ozone trends (%/decade) include the effects of volcanic
 87 eruptions such as Mt. Pinatubo in 1991, dynamics, and atmospheric temperature changes. Ozone data
 88 used in this study are a subset of the Merged Ozone Data MOD set $\Omega_{\text{MOD}}(t)$ (1970 – 2021) starting in
 89 1979 with the Nimbus-7 SBUV (Solar Backscattered Ultraviolet) satellite instrument. From 1979 to 2021,
 90 the MOD data set was created by combining data from Solar Backscattered Ultraviolet instruments
 91 (SBUV/SBUV-2) and the Ozone Mapping and Profiler Suite (OMPS-NP). Methods of calculating trends
 92 from time series data are essential in the analysis of environmental and climate-related data. Here, we
 93 discuss two independent methods to estimate linear trends: 1) linear regression of annual averaged
 94 data and 2) Fourier time series decomposition or multivariate linear regression MLR (Ziemke et al.,
 95 2019) are discussed below. The two methods are compared and shown to give nearly identical results
 96 over their mutual latitude range of validity, 65°S to 65°N . The MLR method is not used in the regions
 97 poleward of the Arctic and Antarctic circles that have latitude dependent extended winter polar night.
 98 The advantage of the MLR method (Eq. 1), or that in Weber et al, 2002, is that it can be used to estimate
 99 the effects of its individual components, while the annual average method can be used in the polar
 100 regions where there is latitude dependent extended winter night.

101 1.1 The Merged Ozone Data Set MOD

102 Figure 1 (left panel) shows the MOD zonally averaged Ω_{MOD} TCO data (Frith et al., 2014; 2020) set as a
 103 function of latitude (5° latitude bands from 77.5°S to 77.5°N) and time (January 1979 to December
 104 2021). Part of the Antarctic ozone hole (75°S to 80°S) is shown (blue color), and the high latitude
 105 maxima, North and South, (red color), with low values in the equatorial region. Figure 1 (right panel)
 106 shows the 42-year zonal and time averaged ozone amounts and the maxima and minima annual
 107 envelopes as a function of latitude. Figure 1 shows the asymmetry in the monthly and zonally averaged
 108 ozone data between the hemispheres, with the Northern Hemisphere NH having more ozone than the
 109 Southern Hemisphere SH at corresponding latitudes. Part of the asymmetry is driven by the Spring
 110 Antarctic ozone hole backfilling in the SH summer.

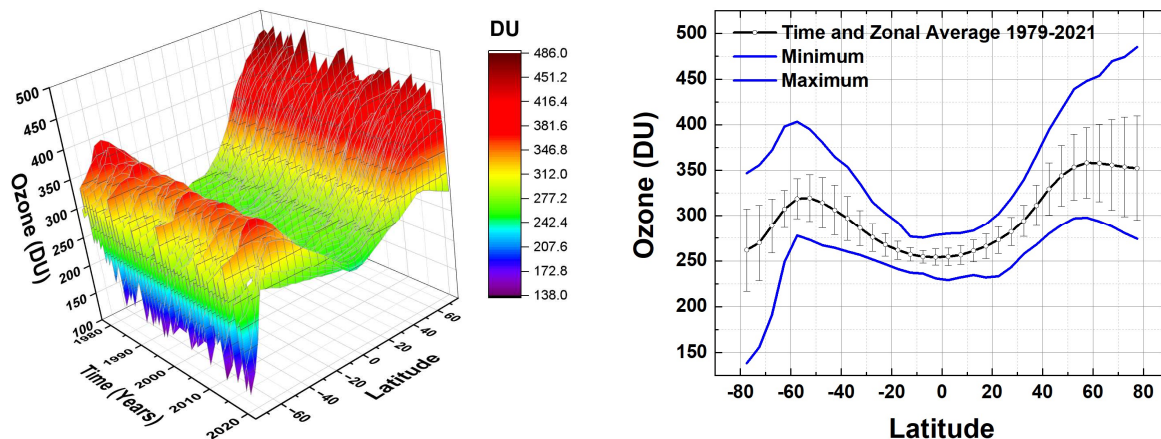


Fig. 1 Left: The zonally and monthly averaged Ω_{MOD} data set 1979 – 2021 and -77.5° to 77.5° . Right: Time and zonal averaged ozone and its maxima and minima 1979 – 2021. Error bars are 1 standard deviation $\pm 1\sigma$.

111 $\Omega_{\text{MOD}}(t, \theta)$ provides a global view of ozone levels needed to track changes in ozone concentrations over
 112 time t for each latitude band θ . The SBUV and OMPS-NP series of satellite instruments form the longest
 113 (1979 to 2022) continuous global ozone $\Omega_{\text{MOD}}(t, \theta)$ data record from a single instrument type. Merged
 114 ozone retrievals from the individual instruments use the version 8.7 retrieval algorithm (described by
 115 Weber et al., 2022) as an extension of the version 8.6 algorithm (Bhartia et al., 2013; McPeters et al.,
 116 2013; DeLand et al., 2012; Frith et al., 2017) specifically designed to improve cross calibrations between
 117 the later SBUV-type instruments in MOD starting from NOAA-16 in 2000. There were no external
 118 adjustments made to the ozone retrieval except for small high-altitude (> 35 km) diurnal corrections to
 119 account for different measurement times between satellites and varying measurement time of day as
 120 individual satellite orbits slowly drift in equator crossing time. These adjustments are very minor in TCO
 121 (Frith, personal communication). Data from each instrument are selected based on quality criteria
 122 outlined in Frith et al. (2014; 2020) and the data are averaged during periods when more than one
 123 instrument was operational. The $\Omega_{\text{MOD}}(t, \theta)$ are available as a function of latitude and month,
 124 https://acd-ext.gsfc.nasa.gov/Data_services/merged/.

125
 126 Analysis of the long-term ozone time series has been looked at extensively with references given in
 127 Weber et al., 2022. Methods for estimating trends from an oscillating time series with several distinct
 128 periodicities are well known (Ziemke et al., 2019; Stolarski et al. 1991;1992, Herman et al., 1993). For
 129 ozone, one of the difficulties in trend estimation is that the early part of the time series shows a strong
 130 ozone decrease at all latitudes that continued until the mid-1990s and then flattens out and shows
 131 almost no recovery thereafter toward 1979 values. The Ω_{MOD} time series has been used extensively in
 132 ozone assessments and State of the Climate reports (e.g., WMO, 2022) and was recently compared to
 133 several other merged total ozone records in Weber et al. (2022). The validity of the Ω_{MOD} time series for
 134 estimating ozone trends was further checked (See Appendix Figs. A1 to A3) in this study by showing
 135 detailed comparisons between the deseasonalized Ω_{MOD} time series with the deseasonalized MLS
 136 (Microwave Limb Sounder) overlapping stratospheric ozone time series (2005 to 2023).

137 2.0 Trend Estimates from the MOD Ozone Data

138 Multivariate Linear Regression MLR is a Fourier based method for analyzing atmospheric time series
 139 data that decomposes the time series into its component parts, including trend, quasi-biennial
 140 oscillation QBO, solar cycle, ENSO (El Nino Southern Oscillation), seasonality, and noise resulting in a
 141 trend estimate and 2-standard deviation 2σ uncertainty estimates (Ziemke et al., 2019). Calculated 2σ
 142 uncertainties for the MLR trends include a first order autoregressive adjustment applied to the derived
 143 residuals (Weatherhead et al., 1998).

144 Linear trend estimates for the long-term changes in $\Omega_{\text{MOD}}(t, \theta_i)$ globally and as a function of latitude θ_i
 145 have been obtained using the multivariate linear regression (MLR) model (e.g., Randel and Cobb, 1994,
 146 and references therein). Trends $B(\theta_i)$ were determined for $\Omega_{\text{MOD}}(t, \theta_i)$ using Eqns. 1 and 2.

147

$$\Omega_{\text{MOD}}(t, \theta_i) = A(\theta_i, t) + B(\theta_i, t) \cdot t + C(\theta_i, t) \cdot \text{QBO}_1(t) + D(\theta_i, t) \cdot \text{QBO}_2(t) + E(\theta_i, t) \cdot \text{ENSO}(t) + F(\theta_i, t) \cdot \text{Solar}(t) + R(t, \theta_i) \quad (1)$$

148

149 where t is the month index ($t=1$ to 516 months with data for 1979–2021), $A(\theta_i, t)$ is the seasonal cycle
 150 coefficient, $B(\theta_i, t)$ is the trend coefficient, $C(\theta_i, t)$ is the first empirical orthogonal function (EOF) QBO
 151 coefficient, $D(\theta_i, t)$ is the second EOF QBO coefficient, both representing the major components of
 152 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
 153 residual error time series. The F10.7 cm solar flux monthly time series is used for the Solar(t) proxy, first
 154 and second leading EOF QBO monthly time series proxies $\text{QBO}_1(t)$ and $\text{QBO}_2(t)$ are used for the QBO
 155 component (Wallace et al., 1993), and Nino 3.4 (Oldenborgh *et al* 2021) is used for ENSO(t) ([Nino 3.4: https://www.ncsl.noaa.gov/access/monitoring/enso/sst](https://www.ncsl.noaa.gov/access/monitoring/enso/sst)). $\text{QBO}_1(t)$ and $\text{QBO}_2(t)$ are nearly orthogonal
 156 (correlation coefficient approximately zero) oscillating time series based on data with approximately a
 157 2.3-year periodicity. $A(\theta_i, t)$ involves 7 fixed constants while $B(\theta_i, t)$ (and all other remaining coefficients)
 158 involves 5 fixed constants for each θ_i . The harmonic expansion for $A(t)$ (similar for the other
 159 coefficients) is (Eqn. 2).

160

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

162

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

166

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

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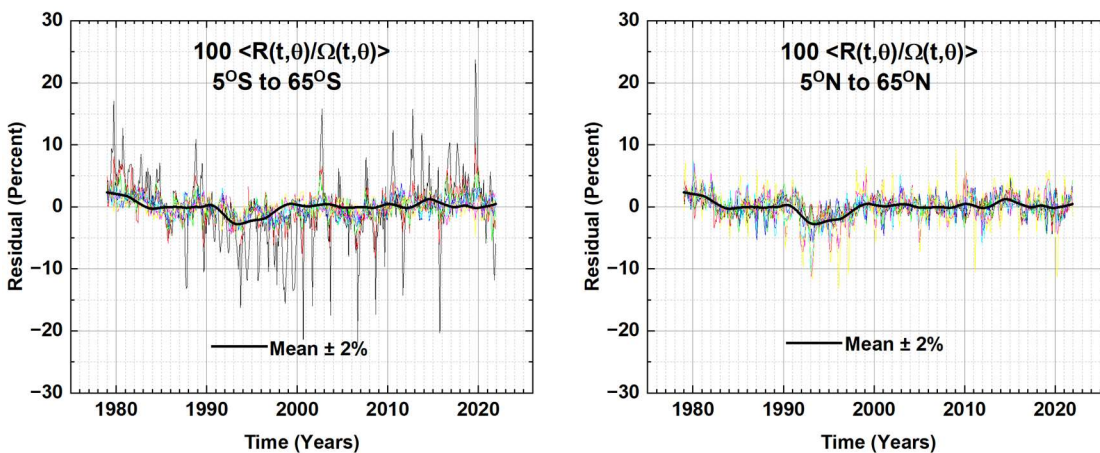


Fig. 2 The latitude average residual term from Eq. 1 in percent $100 \langle R(t, \theta) / \Omega(t, \theta) \rangle$. The black line is the Lowess(0.1) fit (Cleveland, 1979) to the $R(t, \theta)$ with an average error estimate of $\pm 2\%$. The light-colored lines are each latitude's $R(t, \theta)$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

172
 173 The latitude average residual $R(t)$ in percent of the MOD ozone amount ($100 \langle R(t, \theta_i) / \Omega_{\text{MOD}}(t, \theta_i) \rangle$) is shown
 174 in Fig. 2 for the SH and NH as an indication of how well Eq 1 is able to fit the $\Omega_{\text{MOD}}(t, \theta_i)$ time series.

175
 176 The SH $R(t, \theta)$ is more variable than the NH with the largest variations arising in the 55°S and 65°S latitude
 177 bands. On average Eq. 1 fits the original data $\Omega_{\text{MOD}}(t, \theta_i)$ to within $\pm 2\%$.

178
 179 The linear deseasonalized trend results $B(\theta_i)$ are obtained for 14 latitude bands θ_i (centered on 65°S to
 180 65°N). The latitudinal trends $P_D(\theta_i)$ are expressed in %/Decade given by Eq. 3, where the denominator D
 181 is either the time average $\langle \Omega \rangle$ of the area weighted global ozone average (Fig 1) or the time average $D(\theta_i)$
 182 $= \langle \Omega_{\text{MOD}}(t, \theta_i) \rangle$ for each latitude band over the considered period. The whole year period considered is
 183 1979 – 2021.

$$184 \quad P_D(\theta_i) = 1000 B(\theta_i) / D(\theta_i) \quad (\% / \text{Decade}) \quad (3)$$

185
 186 In the second method, the trend is estimated using annual integrals (annual averages) that remove the
 187 seasonality and other short-term oscillations but ignore longer term oscillations such as the 28-to-29-
 188 month QBO cycle and the average 11.3-year solar cycle. A comparison of the two trend estimating
 189 methods is shown in Fig. 3 for the entire 1979 to 2021 period showing that they agree quite closely, but
 190 that the annual average method has slightly larger two standard deviations 2σ than the MLR method.

191
 192 The MLR method (Eqns. 1 and 2) are not applied poleward of the Arctic and Antarctic circles where
 193 latitude dependent extended winter night periods occur. Additional latitude dependent terms of varying
 194 periods would be needed for latitudes greater than 70° . The annual average method does not have
 195 these complications.

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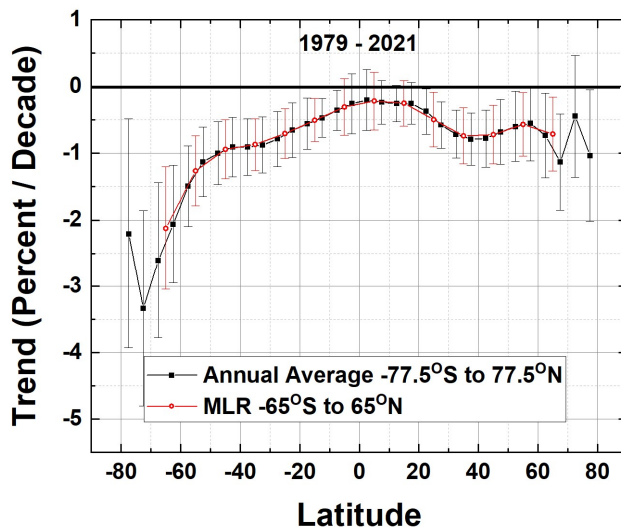


Fig. 3 The ozone trend $P_D(\theta)$ for the entire period 1979 – 2021 for two methods, MLR and Annual Average. The latitude grids for the two methods are offset to show the agreement in the trends and 2σ error bars.

197

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

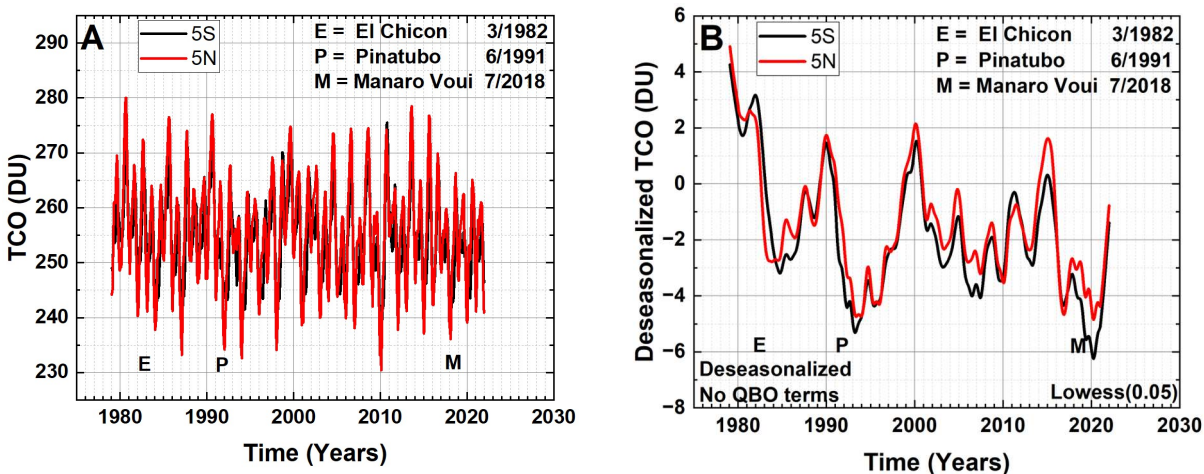


Fig. 4. A. Ω_{MOD} time series for $\theta = 5^\circ\text{N}$ and 5°S . B. The deseasonalized TOC time series for $\theta = 5^\circ\text{N}$ and 5°S without removing QBO effects (Eq. 1). The approximate dates are shown of volcanic eruptions that injected large amounts of SO_2 into the stratosphere leading to minima approximately 1 year later.

204

205 Figure 4A shows the Ω_{MOD} time series for 5°S and 5°N , and Fig. 4B the deseasonalized and smoothed
 206 (Lowess(0.05)) Ω_{MOD} time series. After deseasonalizing, but not removing QBO effects (Eq. 1), both the
 207 2.3-year QBO oscillation and the reduced ozone effects from volcanic eruptions, are shown in Fig. 4B.
 208 Some volcanos (e.g., from El Chicon March 1982, Mt. Pinatubo June 1991, and Manaro Voui July 2018)
 209 inject significant amounts of SO_2 into the lower stratosphere leading to the formation of aerosols that
 210 reduce UV light and the production of ozone, especially in the equatorial region.

211 Figure 5 shows the Lowess(0.3) fits (black curves) to the Ω_{MOD} data for four sample latitude bands 55°S ,
 212 45°S , 55°N , and 45°N that tracks the longer-term changes in the Ω_{MOD} time series. Also shown are
 213 examples of $f = 0.1$ (red) and $f = 0.05$ (blue dots). The Lowess(0.05) fit (blue dots) shows considerable
 214 structure with a minimum in 1993 that is likely related to the Mt. Pinatubo eruption and a modest El
 215 Nino effect in 1991-1992. The estimated values of T_A for $f = 0.1$ and 0.05 can differ by 6 months from that
 216 determined when $f=0.3$ because of short term oscillations. The Lowess(0.3) degree of smoothing
 217 removes most of the short-term effects on ozone such as QBO and those from volcanic eruptions from
 218 El Chichon (1982) and Mt. Pinatubo (1991), both well before the earliest estimated T_A in 1994.

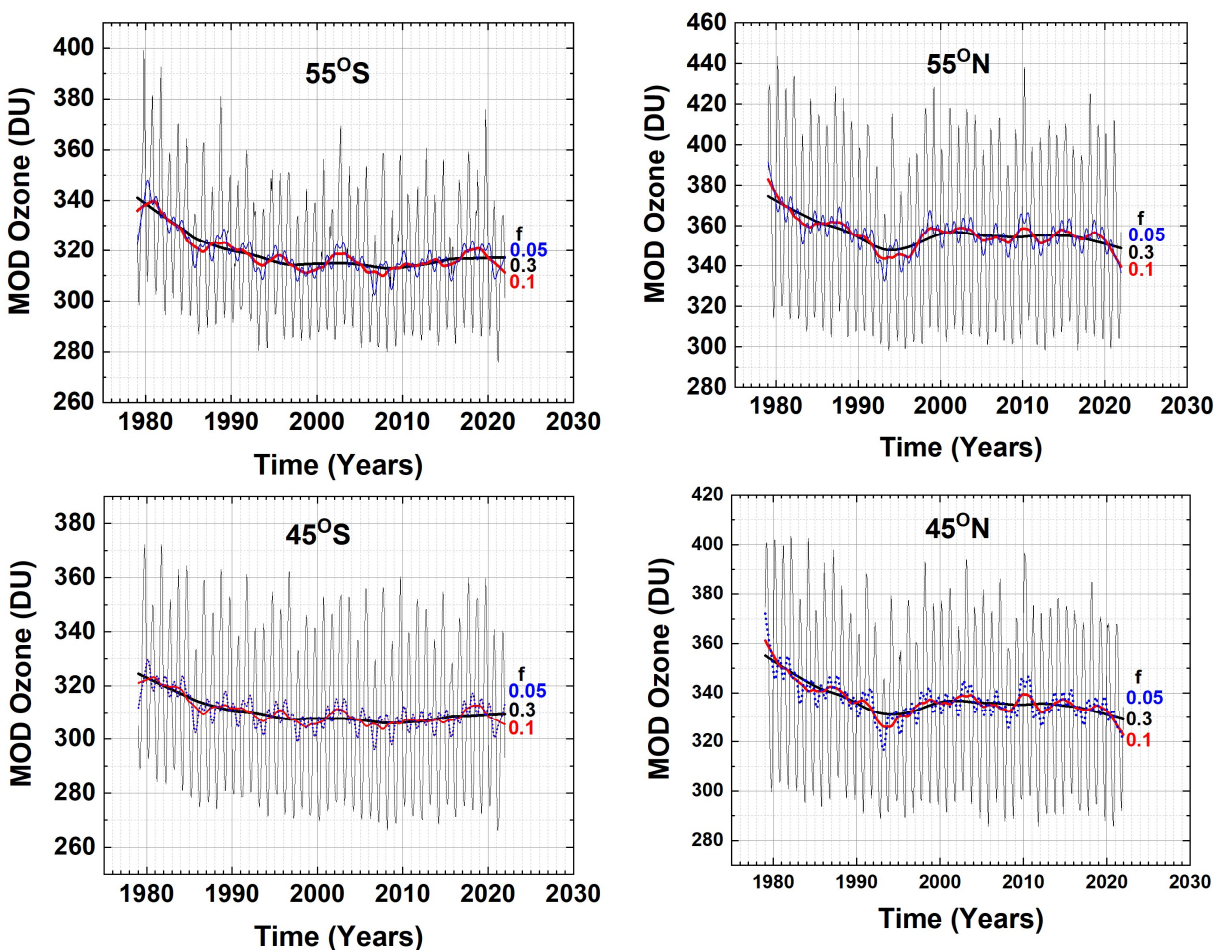


Fig. 5 Ω_{MOD} in four latitude bands and Lowess(0.3) fitting functions ($f = 0.3$, black lines). Examples of different $f = 0.1$ (Red) and 0.05 (blue dots) are shown at 45°S and 45°N . Note the slight downturn since 2010 in the Lowess(0.3) at 45°N and 55°N .

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

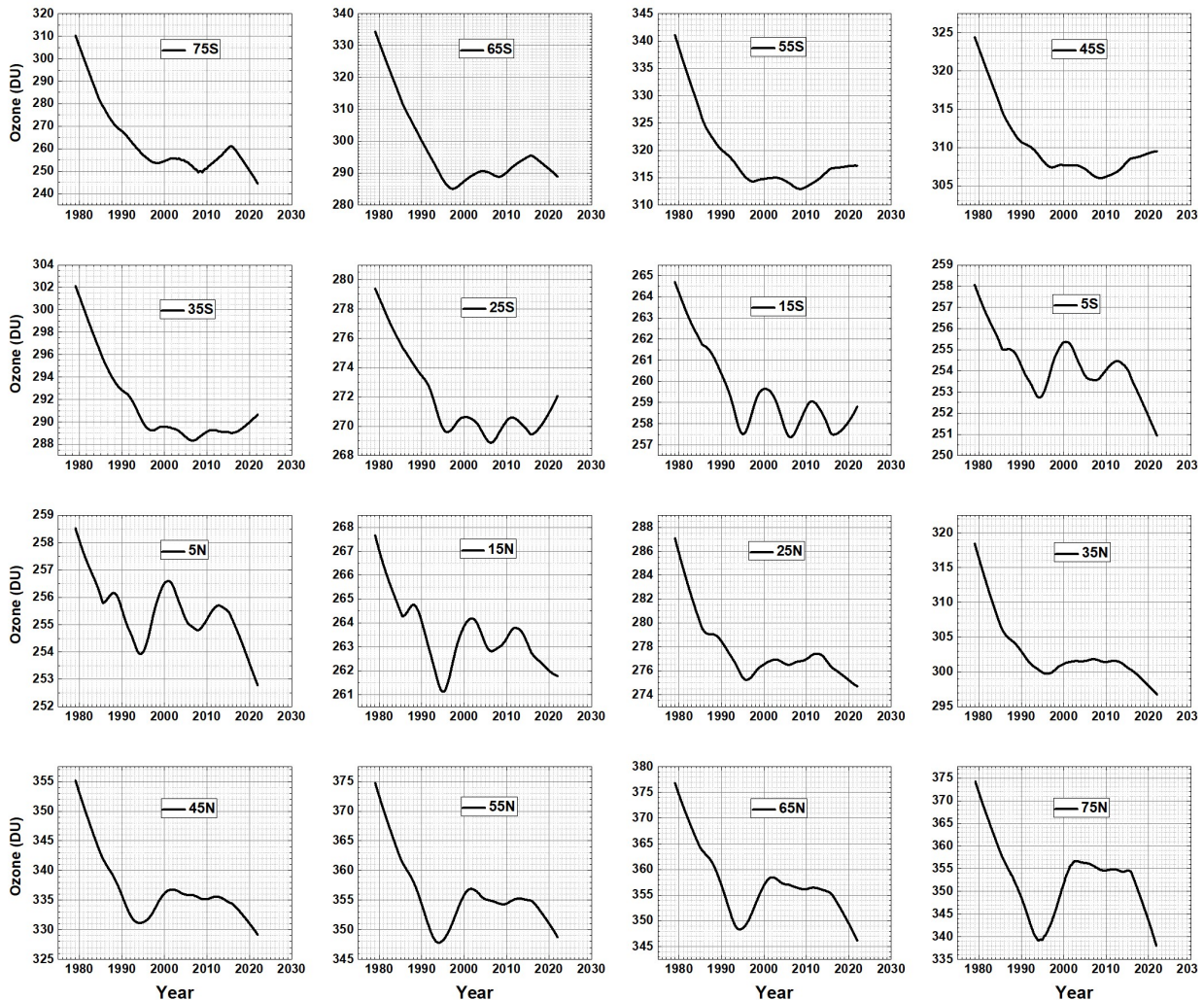


Fig. 6 Lowess(0.3) fits to the Ω_{MOD} data for 16 latitude bands **used** to determine $T_A(\theta)$. **Note that the ozone scale varies for each latitude.**

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

231 1994-1996 for low latitudes between $\pm 25^\circ$ that corresponds to the Brewer-Dobson ozone upwelling
 232 region (Brewer et al., 1926; Dobson, 1949; Butchart, 2014) where most of the ozone is created by
 233 sunlight and then transported poleward. At poleward latitudes, the turnaround dates are quite
 234 different, with a delayed date, 1997, at high SH latitudes ($35^\circ\text{S} - 65^\circ\text{S}$), 1998 at 75°S compared to 1994
 235 at high NH latitudes (45°N to 75°N).

236 The T_A delay to 1997 for latitudes $35^\circ\text{S} - 65^\circ\text{S}$ follows the delayed recovery of ozone depletion within
 237 the Spring Antarctic Ozone Hole (Solomon, 1990; Stone et al., 2021, their Fig. 3; Bodeker and Kremser,
 238 2021, their Figs. 6 and 9) and backfilling (air exchange with lower latitude ozone-rich air) during the
 239 summer months after the polar vortex winds break down in October – November.

240

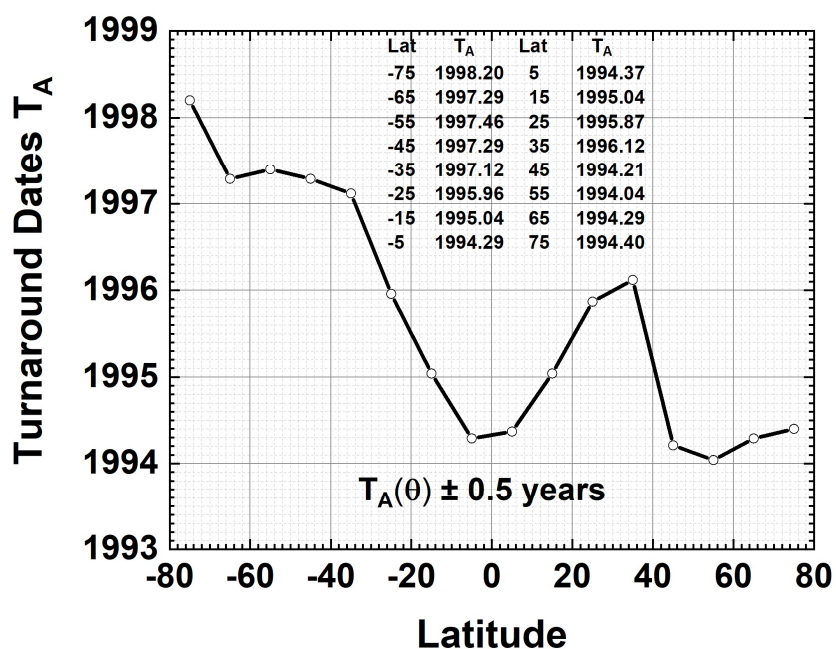


Fig 7 Turnaround dates $T_A(\theta)$ as a function of latitude from Fig. 6 with an estimated accuracy of ± 0.5 years based on the analysis in Fig. 5.

241

242 The general $T_A(\theta)$ pattern shown in Fig. 7 should appear in model calculations as a signature of the
 243 combined effects of photochemistry, dynamics, and volcanic eruptions on the cessation of decreasing
 244 ozone in the mid-1990s.

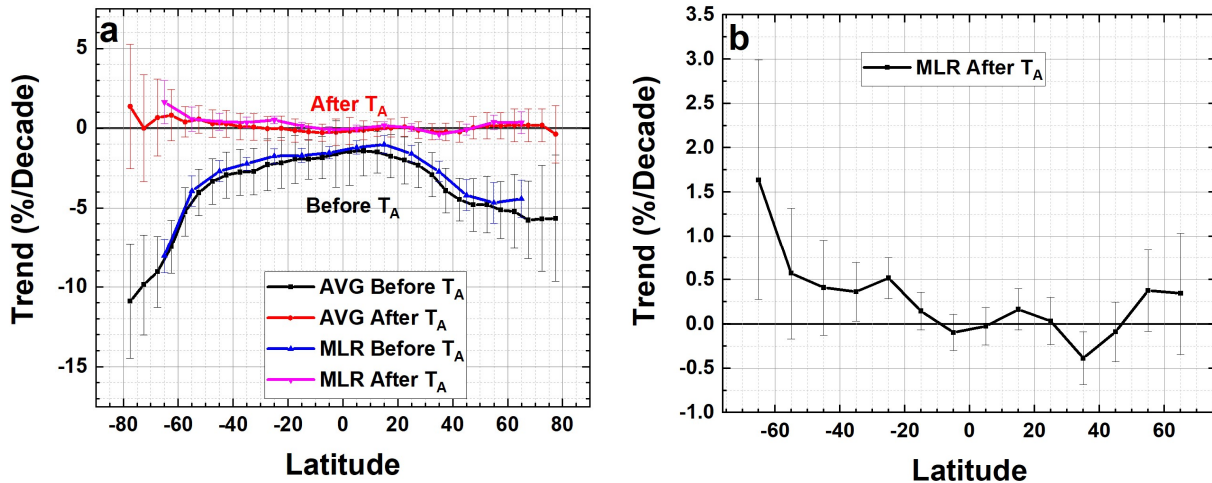


Fig. 8a Ozone trends $P_D(\theta)$ (percent per decade) using the MLR and Annual Average methods before and after $T_A(\theta)$. 8b A magnified version of the MLR estimated trends after T_A with 2σ uncertainties.

245

246 Trends (linear slopes) $P_D(\theta)$ in percent per decade are estimated (Eqn. 3) for the separate periods before
 247 and after $T_A(\theta)$ in each latitude band (Fig. 8) and for the entire period (Fig. 3). The linear slopes obtained
 248 by the two methods, MLR and annual average closely agree (Figs. 3 and 8) with the annual average
 249 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 |

250

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

254 earlier, the trend estimates are not very sensitive to the exact T_A , but the shape of $T_A(\theta)$ should be a
 255 model validation marker contained in model calculations for all effects, not just ODSs.

256 The delayed (1997) Southern Hemisphere mid and high latitude values of T_A are caused by coupling to
 257 the increasing Antarctic spring ozone loss after 1979 until a recovery starting in about 1998-2000
 258 (Solomon et al., 2016). The mid and high latitude, from 35°S to 65°S , delay is caused by the summer
 259 mixing of ozone poor air from the Antarctic region with SH midlatitude ozone-rich air once the polar
 260 vortex winds break down in November-December.

261
 262 The asymmetry between the Arctic and Antarctic is caused by the lower winter Antarctic temperatures
 263 (-80°C) leading to the formation of low altitude clouds containing ice crystals along with the isolating
 264 Antarctic polar vortex winds (Solomon et al., 2007; 2016). In the spring sunlight the ice and water
 265 droplets (Tritscher, et al., 2021) release ODS and depletes ozone to a monthly average of about 155 DU.
 266 During the summer, air exchange with ozone rich air from lower latitudes comes into the polar latitudes and fill
 267 in the ozone layer above Antarctica (monthly average about 300 DU. Smaller but significant ozone losses
 268 occurred in the Arctic region caused by occasional low temperatures and ODSs. The Arctic does not
 269 routinely have the low temperatures needed for winter ice clouds nor does it have the persistent
 270 isolating polar vortex winds because of wave action forced by the land topography The latitude band at
 271 75°N (Fig.1) has the highest amount of monthly average winter ozone 450 ± 25 DU that decreases to
 272 290 ± 20 DU monthly average during the summer that are comparable to mid-latitude values. The result
 273 is earlier values of T_A in the NH compared to the SH. The NH T_A is earlier than the 1997 minimum in
 274 stratospheric halogens (Weber et al., 2022; Newman et al., 2007). Note that T_A is not the time of the
 275 start of recovery, but rather the time for the end of rapid ozone decrease.

276
 277 Before the SH T_A , total column ozone decreased at a rate of $P_D = -10.9\pm 3.6\%$ at 77.5°S and $-8.0\pm 1.1\%$ per
 278 decade at 65°S , during the period from 1979 to 1997 with smaller decreases from 55°S to 25°S (Fig. 8a).
 279 After the turnaround period T_A , ozone at 65°S increased at $P_D = 1.6\pm 1.4\%$ /decade based on the MLR
 280 method. After T_A , most other latitudes (Fig. 8b) show stationary ozone amounts within 2σ . In the NH the
 281 decreases were smaller than in the SH before T_A because of the absence of an Arctic ozone hole region.
 282 At 77.5°N was $P_D = -5.6\pm 4\%$ /decade and at 65°N $P_D = -4.4\pm 0.35\%$ /decade.

283 An analysis of ozone trends prior to the start of reliable satellite data in late 1978 showed that the
 284 annual rate of ozone loss (%/Year) increased after 1978 (Stahelin et al., 2001). Based on the first
 285 derivatives of the data in Fig. 6, the maximum annual rate of ozone reduction occurred in 1979 and 1980
 286 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
 287 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
 288 where the loss rate is almost constant between 8% /Year and 10% /Year compared to the larger SH loss
 289 rates caused by the presence of the springtime Antarctic ozone hole.

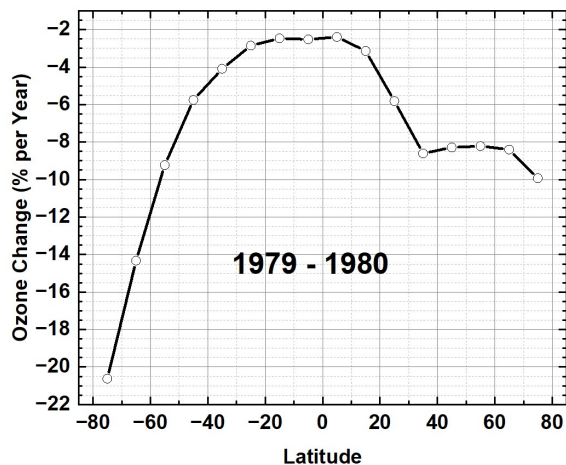


Fig. 9 The percent change in ozone per year in 1979 or 1980

290

291 The Lowess(0.3) plots in Fig. 6 suggest that Ω_{MOD} has been declining since approximately 2010 from 5°S
 292 to 65°N but still increasing from 45°S to 65°S (Fig. 6). However, computing the trends (Fig. 10) from
 293 $\Omega_{MOD}(t, \theta)$ using either the MLR (Eq. 1) or annual average methods suggest that the declines in ozone
 294 from 25°S to 65°N are not yet significant at the 2σ level over the period 2010 – 2021.

295

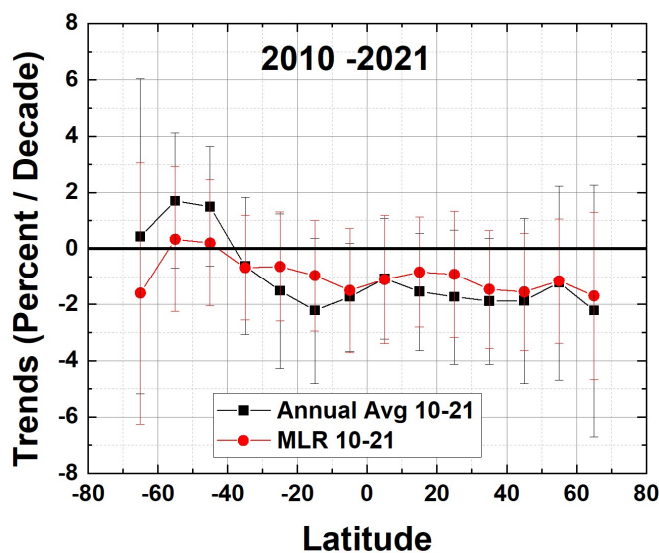
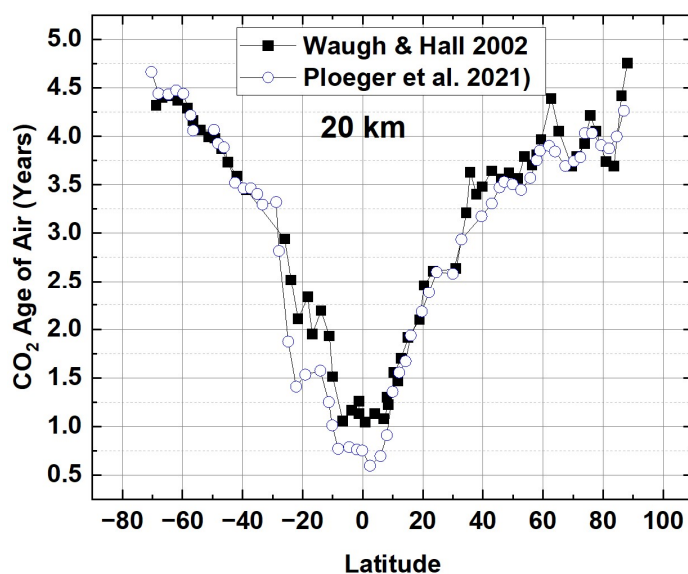


Fig. 10 Ozone trends $P_D(\theta)$ (Percent per Decade) for the period 2010 – 2021 for the Annual Average and MLR methods applied to $\Omega_{MOD}(t, \theta)$.

296 Comparing deseasonalized $\Omega_{MOD}(t, \theta)$ with deseasonalized Microwave Limb Sounder MLS (see Appendix
 297 Figs. A1, A2, and A3) Stratospheric Ozone from 2005 to 2021 shows small average (Lowess(0.3))
 298 differences that are within ± 1 DU except for 2021 when the differences at both 65°S and 65°N are about
 299 -2.5DU. This suggests that the calibrations of the later SBUV-2 and OMPS-NP instruments are stable. For

300 2016 to 2018, Ω_{MOD} is obtained from NOAA-19 SBUV plus OMPS-NP and from just OMPS-NP since 2018.
 301 Figure A3 suggests that there was a decrease in tropospheric ozone in 2020 that may correspond to
 302 reduced economic activity during the COVID-19 pandemic.

303 Age of air AoA is a measure of how long a parcel of air resides in the stratosphere after it leaves the
 304 troposphere (Linz et al., 2016; Ploeger et al, 2021). A comparison of T_A with AoA estimates from the
 305 relatively inert tracer gas CO_2 (Fig. 11) for the altitude range near the ozone maximum (approximately
 306 20 km) vs latitude (based on Waugh and Hall, 2002, their Fig. 6a and Ploeger et al, 2021 their Fig. 10a)
 307 shows near symmetry between the hemispheres with the shortest AoA in the equatorial region. The
 308 turnaround dates T_A in Fig. 6 are also symmetric in the equatorial zone corresponding the upwelling
 309 Brewer Dobson circulation and the smaller AoA. This suggests that the combined effects of chemistry
 310 and dynamics on ozone amounts are similar between $\pm 25^\circ$. The precursors to ODS are also lifted into
 311 the equatorial stratosphere and transported towards the polar regions (Newman et al., 2004; 2007)
 312 where they can be photo-dissociated into ODS. Ozone at higher latitudes, NH and SH, with longer AoA,
 313 will be dependent on transported ozone and ODS and their photochemistry, and especially the different
 314 dynamics and chemistry in the Arctic and Antarctic regions.



315 Fig. 11 Age of air derived from CO_2 data (Waugh and Hall, 2002; Ploeger et al., 2021)

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322 **3.0 Summary**

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

351

352

353 **Appendix**

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

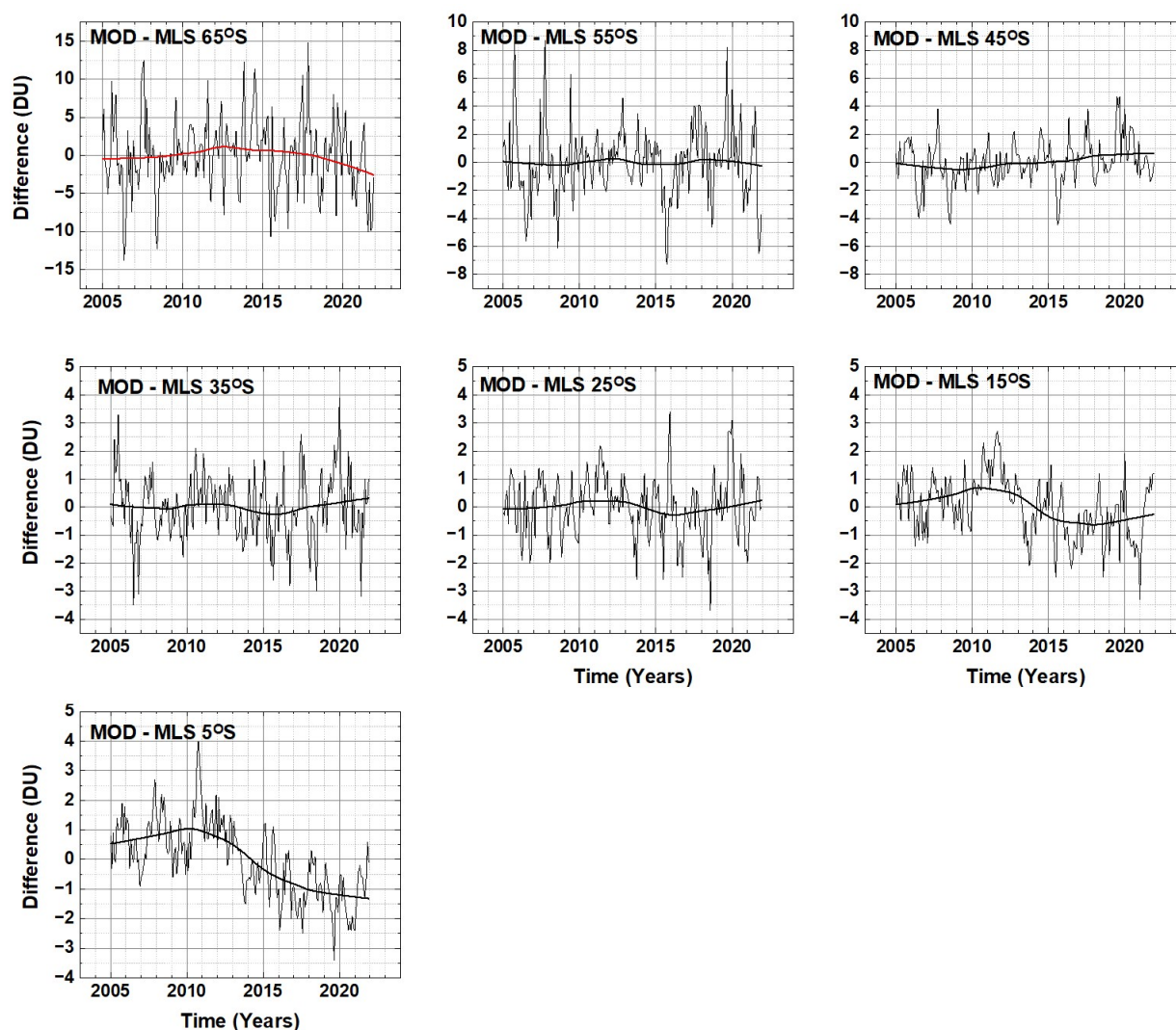


Fig. A1 A comparison of deseasonalized Ω_{MOD} with deseasonalized MLS stratospheric column ozone for 65°S to 5°S.

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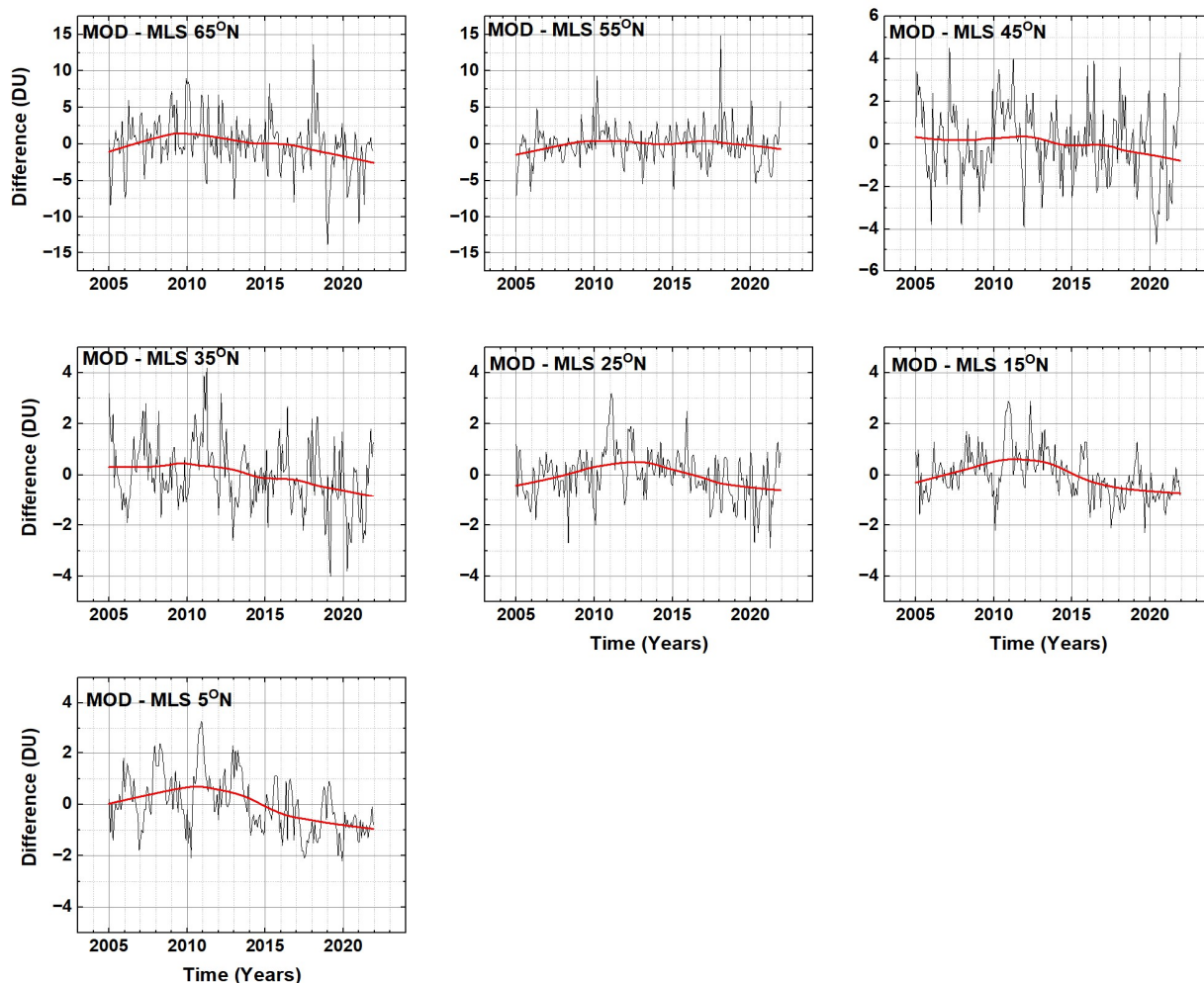


Fig. A2 A comparison of deseasonalized MOD total ozone with deseasonalized MLS stratospheric column ozone for 5°N to 65°N. Variations of ± 3 DU are within the MOD merged record uncertainties.

365

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

368

369 Since both MOD and MLS time series were deseasonalized, the mean values would be zero unless there
 370 were changes in tropospheric ozone or instrument calibration drift. The differences are summarized in
 371 Fig. A3 along with the $2\sigma'$, (σ' = standard deviation from the mean) error bars estimated from the
 372 average of each deseasonalized time series. In 2020 there appears to be a systematic change in $\langle \text{MOD} -$
 373 $\text{MLS} \rangle$ that may be a reduction in tropospheric ozone amount of about 3 DU caused by the economic
 374 slowdown associated with COVID-19 (Ziemke et al, 2022). The systematic change mostly recovered in
 375 2021 (Fig. A3) except for -1DU near the equator (5°S to 15°N).

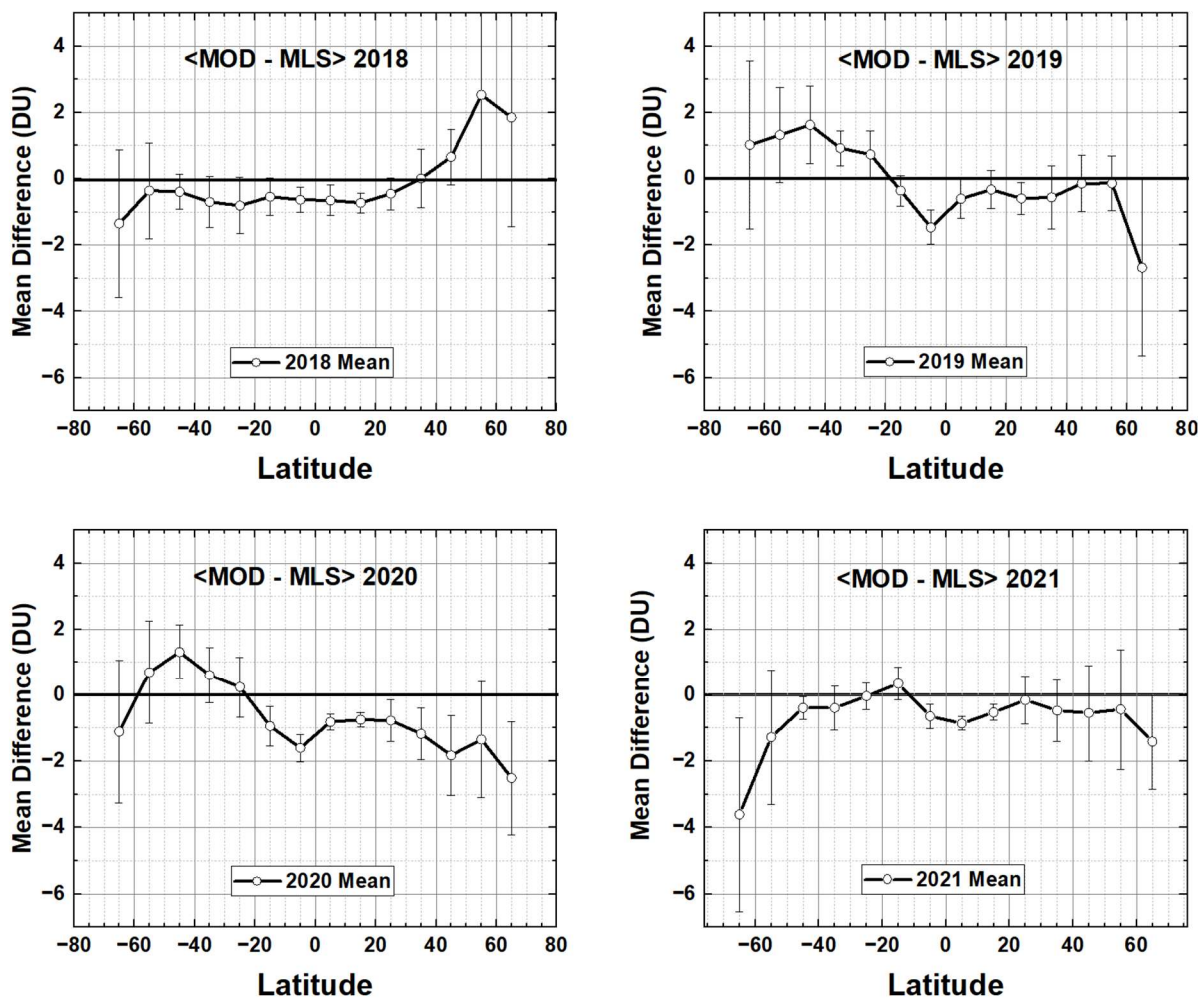


Fig. A3 Annual average <MOD – MLS> for the years 2018 to 2021. Error bars are $2\sigma'$, where σ' = standard error of the mean estimated from the average of the deseasonalized time series for each year shown in Figs. A1 and A2.

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505

506 **Author contribution:**

507 Jay Herman is responsible for writing the text, the annual integral trend calculations, and all the
508 figures. Jerald Ziemke supplied the MLR trend calculations and the comparison with MLS. Richard
509 McPeters supplied the MOD ozone as a continuous function of time from 1979 to 2021 for each
510 latitude band.

511 **Data Availability**

512 The original data used are publicly available in an ASCII format.

513 https://acd-ext.gsfc.nasa.gov/Data_services/merged/

514 and processed data in Excel format

515 https://avdc.gsfc.nasa.gov/pub/DSCOVER/JayHerman/MOD_Ozone_Trends/

516

517

518 **Competing interests:**

519 The authors declare that they have no conflict of interest.

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523

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527 added some important corrections.

528