

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  $\Omega_{\text{MOD}}(t, \theta)$  from the NASA Merged Ozone Data set  
19 (MOD) were examined to show that the latitude-dependent,  $\theta$ , ozone depletion turnaround dates  $T_A(\theta)$   
20 range from 1994 to 1998.  $T_A(\theta)$  is defined as the approximate date when the zonally average ozone  
21 ceased decreasing.  $\Omega_{\text{MOD}}$  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 new calculated systematic latitude-dependent hemispherically asymmetric**  
24  **$T_A(\theta)$  shape that currently do not appear in the suite of chemistry-climate models that are part of**  
25 **Chemistry-Climate Model Validation Activity CCMVal, the Chemistry-Climate Model Validation Activity,**  
26 **which combine the effects of photochemistry, volcanic eruptions, and dynamics in their estimate of**  
27 **ozone recovery.** Trends of zonally averaged total column ozone in percent per decade were computed  
28 before and after  $T_A(\theta)$  using two different trend estimate methods that closely agree, Fourier Series  
29 Multivariate Linear Regression and linear regression on annual averages. During the period 1979 to  $T_A(\theta)$   
30 the most dramatic rates of SH ozone loss were  $P_D = -10.9 \pm 3$  % per decade at  $77.5^\circ\text{S}$  and  $-8.5 \pm 0.9$ % per  
31 decade at  $65^\circ\text{S}$ , which is about double the NH rate of loss of  $P_D = -5.6 \pm 4$  %/decade at  $77.5^\circ\text{N}$  and  
32  $4.4 \pm 1$ %/decade at  $65^\circ\text{N}$  for the period 1979 to  $T_A(\theta)$ . After  $T_A(\theta)$ , there has been an increase at  $65^\circ\text{S}$  of  
33  $P_D = 1.6 \pm 1.4$ % per decade with smaller increases from  $55^\circ\text{S}$  to  $25^\circ\text{S}$  and a small decrease at  $35^\circ\text{N}$  of -  
34  $0.4 \pm 0.3$  %/decade. Except for the Antarctic region, there only has been a small recovery in the Southern  
35 Hemisphere toward 1979 ozone values and almost none in the Northern Hemisphere.

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## 39 1.0 Introduction

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

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

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

79 1926) that transports ozone from an upwelling in the equatorial region into the stratosphere and to  
80 downwelling into mid- and high-latitudes.

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

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

### 107 **1.1 The Merged Ozone Data Set MOD**

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

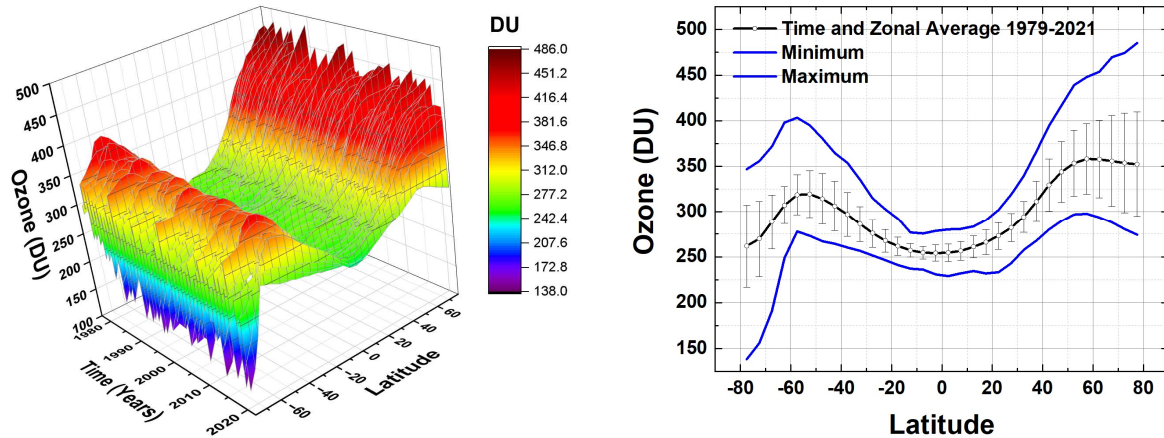


Fig. 1 Left: The zonally and monthly averaged  $\Omega_{\text{MOD}}$  data set 1979 – 2021 and  $-77.5^{\circ}$  to  $77.5^{\circ}$ . Right: Time and zonal averaged ozone and its maxima and minima 1979 – 2021. Error bars are 1 standard deviation  $\pm 1\sigma$ .

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

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

## 143 2.0 Trend Estimates from the MOD Ozone Data

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

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

153

$$\Omega_{MOD}(t, \theta_i) = A(\theta_i, t) + B(\theta_i, t) \cdot t + C(\theta_i, t) \cdot QBO_1(t) + D(\theta_i, t) \cdot QBO_2(t) + E(\theta_i, t) \cdot ENSO(t) + F(\theta_i, t) \cdot Solar(t) + R(t, \theta_i) \quad (1)$$

154

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

167

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

168

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

172

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

176

177

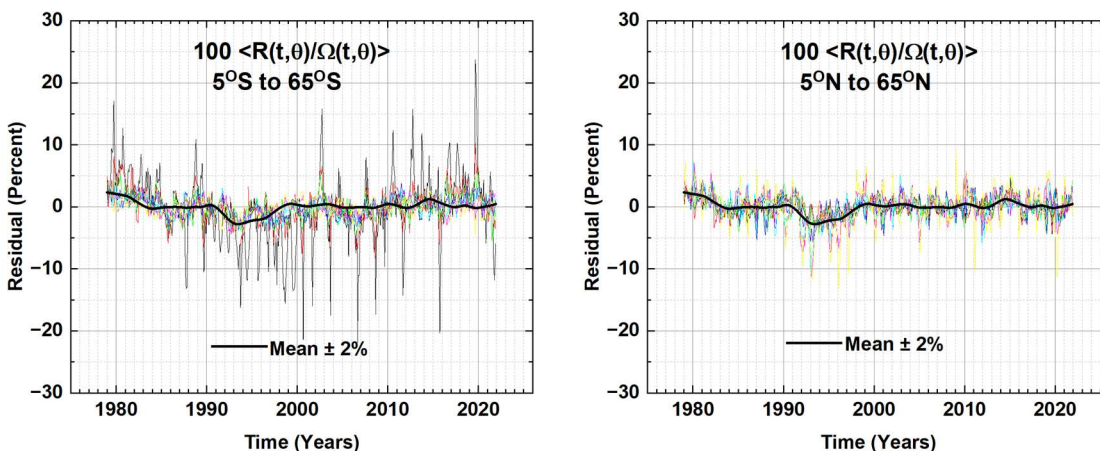


Fig. 2 The latitude average residual term from Eq. 1 in percent  $100 \langle R(t, \theta_i) / \Omega_{MOD}(t, \theta_i) \rangle$ . The black line is the Lowess(0.1) fit (Cleveland, 1979) to the  $R(t, \theta_i)$  with an average error estimate of  $\pm 2\%$ . The light-colored lines are each latitude's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

178  
 179 The latitude average residual  $R(t)$  in percent of the MOD ozone amount ( $100 \langle R(t, \theta_i) / \Omega_{MOD}(t, \theta_i) \rangle$ ) is shown  
 180 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 The SH  $R(t, \theta)$  is more variable than the NH with the largest variations arising in the  $55^\circ S$  and  $65^\circ S$  latitude  
 183 bands. On average Eq. 1 fits the original data  $\Omega_{MOD}(t, \theta_i)$  to within  $\pm 2\%$ .

184  
 185 The linear deseasonalized trend results  $B(\theta_i)$  are obtained for 14 latitude bands  $\theta_i$  (centered on  $65^\circ S$  to  
 186  $65^\circ N$ ). The latitudinal trends  $P_D(\theta_i)$  are expressed in %/Decade given by Eq. 3, where the denominator  $D$   
 187 is either the time average  $\langle \Omega \rangle$  of the area weighted global ozone average (Fig 1) or the time average  $D(\theta_i)$   
 188  $= \langle \Omega_{MOD}(t, \theta_i) \rangle$  for each latitude band over the considered period. The whole year period considered is  
 189 1979 – 2021.

$$190 \quad P_D(\theta_i) = 1000 B(\theta_i) / D(\theta_i) \quad (\% / \text{Decade}) \quad (3)$$

191  
 192 In the second method, the trend is estimated using annual integrals (annual averages) that remove the  
 193 seasonality and other short-term oscillations but ignore longer term oscillations such as the 28-to-29-  
 194 month QBO cycle and the average 11.3-year solar cycle. A comparison of the two trend estimating  
 195 methods is shown in Fig. 3 for the entire 1979 to 2021 period showing that they agree quite closely, but  
 196 that the annual average method has slightly larger two standard deviations  $2\sigma$  than the MLR method.

197  
 198 The MLR method (Eqns. 1 and 2) are not applied poleward of the Arctic and Antarctic circles where  
 199 latitude dependent extended winter night periods occur. Additional latitude dependent terms of varying  
 200 periods would be needed for latitudes greater than  $70^\circ$ . The annual average method does not have  
 201 these complications.

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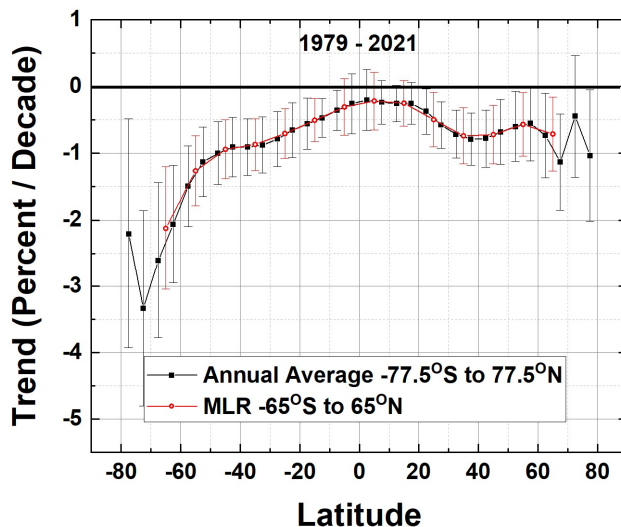


Fig. 3 The ozone trend  $P_D(\theta)$  for the entire period 1979 – 2021 for two methods, MLR and Annual Average. The latitude grids for the two methods are offset to show the agreement in the trends and  $2\sigma$  error bars.

203

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

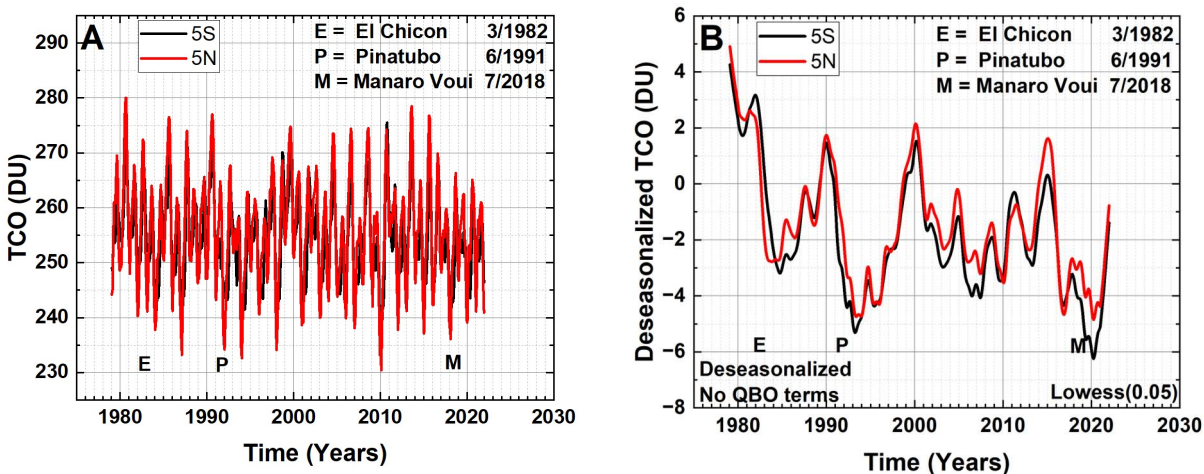


Fig. 4. A.  $\Omega_{\text{MOD}}$  time series for  $\theta = 5^\circ\text{N}$  and  $5^\circ\text{S}$ . B. The deseasonalized TOC time series for  $\theta = 5^\circ\text{N}$  and  $5^\circ\text{S}$  without removing QBO effects (Eq. 1). The approximate dates are shown of volcanic eruptions that injected large amounts of  $\text{SO}_2$  into the stratosphere leading to minima approximately 1 year later.

210



211 Figure 4A shows the  $\Omega_{\text{MOD}}$  time series for  $5^{\circ}\text{S}$  and  $5^{\circ}\text{N}$ , and Fig. 4B the deseasonalized and smoothed  
 212 (Lowess(0.05))  $\Omega_{\text{MOD}}$  time series. After deseasonalizing, but not removing QBO effects (Eq. 1), both the  
 213 2.3-year QBO oscillation and the reduced ozone effects from volcanic eruptions, are shown in Fig. 4B.  
 214 Some volcanos (e.g., from El Chicon March 1982, Mt. Pinatubo June 1991, and Manaro Voui July 2018)  
 215 inject significant amounts of  $\text{SO}_2$  into the lower stratosphere leading to the formation of aerosols that  
 216 reduce UV light and the production of ozone, especially in the equatorial region.

217 Figure 5 shows the Lowess(0.3) fits (black curves) to the  $\Omega_{\text{MOD}}$  data for four sample latitude bands  $55^{\circ}\text{S}$ ,  
 218  $45^{\circ}\text{S}$ ,  $55^{\circ}\text{N}$ , and  $45^{\circ}\text{N}$  that tracks the longer-term changes in the  $\Omega_{\text{MOD}}$  time series. Also shown are  
 219 examples of  $f = 0.1$  (red) and  $f = 0.05$  (blue dots). The Lowess(0.05) fit (blue dots) shows considerable  
 220 structure with a minimum in 1993 that is likely related to the Mt. Pinatubo eruption and a modest El  
 221 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  
 222 determined when  $f=0.3$  because of short term oscillations. The Lowess(0.3) degree of smoothing  
 223 removes most of the short-term effects on ozone such as QBO and those from volcanic eruptions from  
 224 El Chichon (1982) and Mt. Pinatubo (1991), both well before the earliest estimated  $T_A$  in 1994.

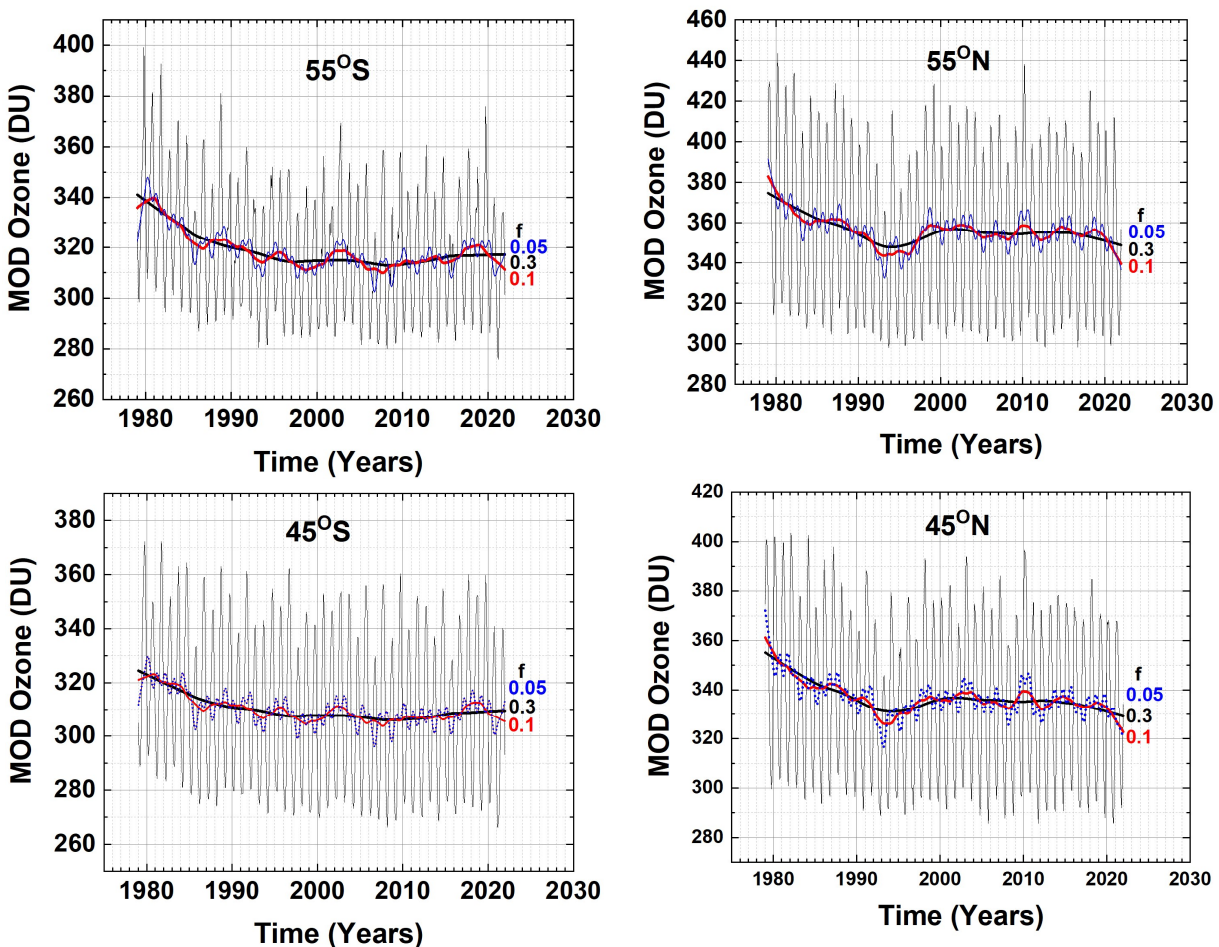


Fig. 5  $\Omega_{\text{MOD}}$  in four latitude bands and Lowess(0.3) fitting functions ( $f = 0.3$ , black lines). Examples of different  $f = 0.1$  (Red) and 0.05 (blue dots) are shown at  $45^{\circ}\text{S}$  and  $45^{\circ}\text{N}$ . Note the slight downturn since 2010 in the Lowess(0.3) at  $45^{\circ}\text{N}$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

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

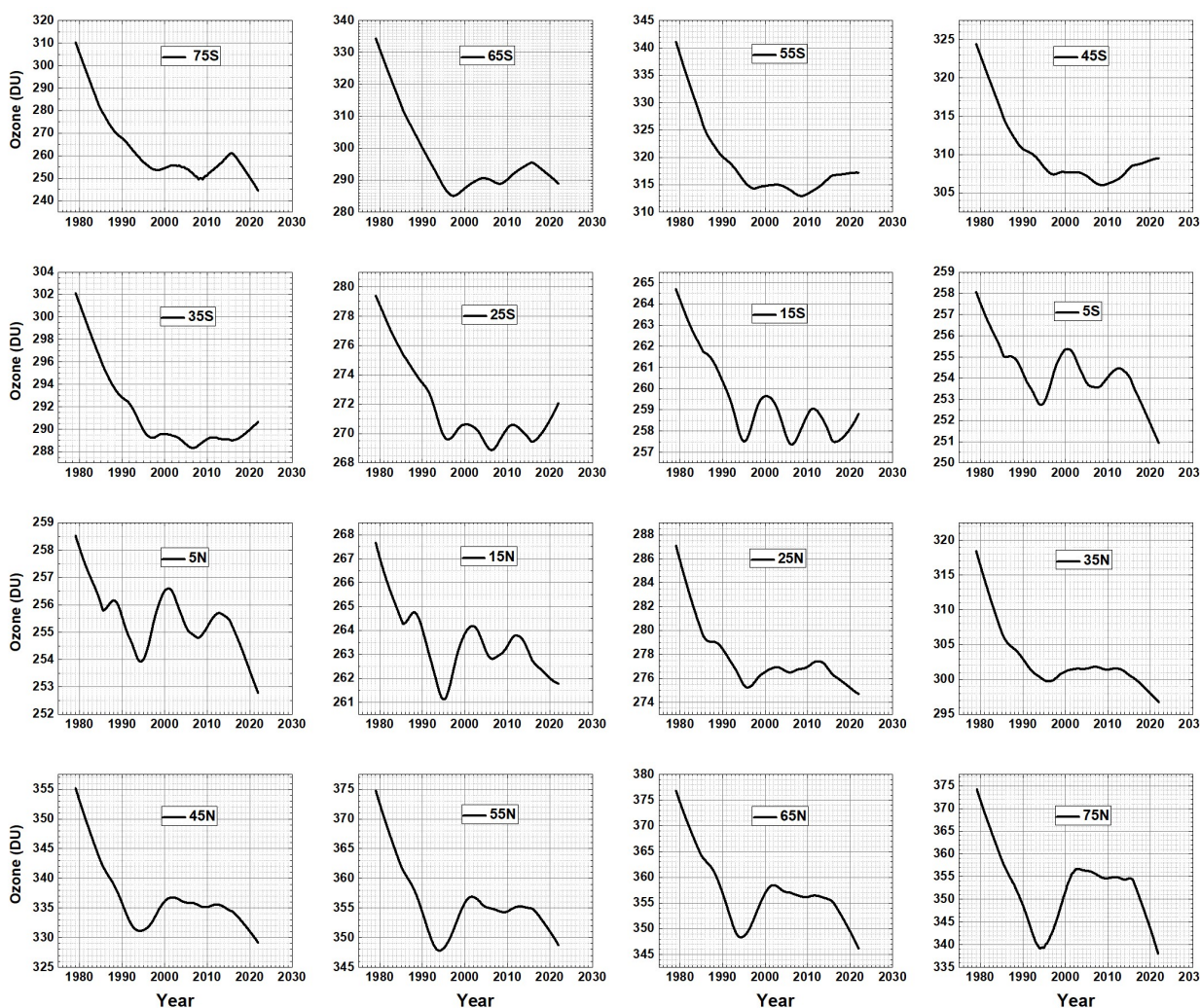


Fig. 6 Lowess(0.3) fits to the  $\Omega_{\text{MOD}}$  data for 16 latitude bands used to determine  $T_A(\theta)$ . Note that the ozone scale varies for each latitude.

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

237 1994-1996 for low latitudes between  $\pm 25^\circ$  that corresponds to the Brewer-Dobson ozone upwelling  
 238 region (Brewer et al., 1926; Dobson, 1949; Butchart, 2014) where most of the ozone is created by  
 239 sunlight and then transported poleward. At poleward latitudes, the turnaround dates are quite  
 240 different, with a delayed date, 1997, at high SH latitudes ( $35^\circ\text{S} - 65^\circ\text{S}$ ), 1998 at  $75^\circ\text{S}$  compared to 1994  
 241 at high NH latitudes ( $45^\circ\text{N}$  to  $75^\circ\text{N}$ ).

242 The  $T_A$  delay to 1997 for latitudes  $35^\circ\text{S} - 65^\circ\text{S}$  follows the delayed recovery of ozone depletion within  
 243 the Spring Antarctic Ozone Hole (Solomon, 1990; Stone et al., 2021, their Fig. 3; Bodeker and Kremser,  
 244 2021, their Figs. 6 and 9) and backfilling (air exchange with lower latitude ozone-rich air) during the  
 245 summer months after the polar vortex winds break down in October – November.

246

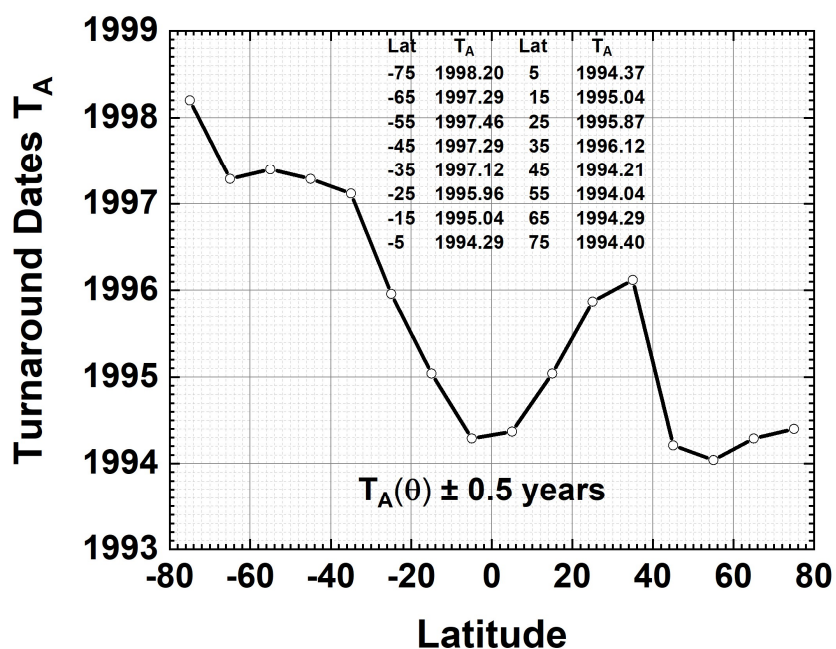


Fig 7 Turnaround dates  $T_A(\theta)$  as a function of latitude from Fig.6 with an estimated accuracy of  $\pm 0.5$  years based on the analysis in Fig. 5.

247

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

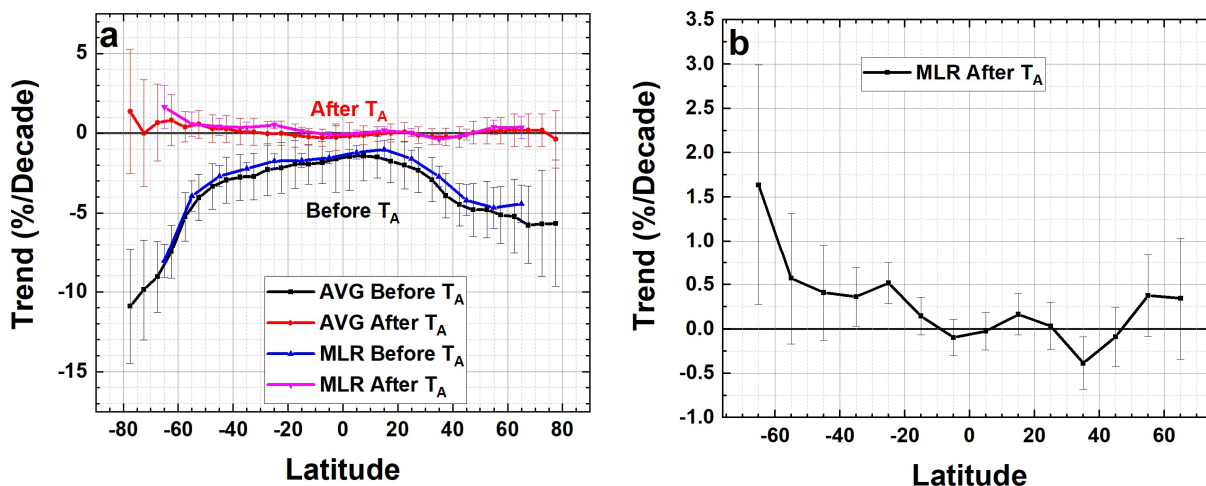


Fig. 8a Ozone trends  $P_D(\theta)$  (percent per decade) using the MLR and Annual Average methods before and after  $T_A(\theta)$ . 8b A magnified version of the MLR estimated trends after  $T_A$  with  $2\sigma$  uncertainties.

251

252 Trends (linear slopes)  $P_D(\theta)$  in percent per decade are estimated (Eqn. 3) for the separate periods before  
 253 and after  $T_A(\theta)$  in each latitude band (Fig. 8) and for the entire period (Fig. 3). The linear slopes obtained  
 254 by the two methods, MLR and annual average closely agree (Figs. 3 and 8) with the annual average  
 255 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	$-8.04 \pm 1.1$	$1.64 \pm 1.4$
-55	$-3.93 \pm 1.0$	$0.57 \pm 0.7$
-45	$-2.69 \pm 0.7$	$0.41 \pm 0.5$
-35	$-2.22 \pm 0.4$	$0.36 \pm 0.3$
-25	$-1.75 \pm 0.5$	$0.52 \pm 0.2$
-15	$-1.71 \pm 0.4$	$0.15 \pm 0.2$
-5	$-1.54 \pm 0.4$	$-0.10 \pm 0.2$
5	$-1.21 \pm 0.4$	$-0.03 \pm 0.2$
15	$-1.01 \pm 0.6$	$0.16 \pm 0.2$
25	$-1.61 \pm 0.5$	$0.03 \pm 0.3$
35	$-2.71 \pm 0.6$	$-0.39 \pm 0.3$
45	$-4.20 \pm 1.0$	$-0.09 \pm 0.3$
55	$-4.67 \pm 1.3$	$0.38 \pm 0.5$
65	$-4.43 \pm 1.2$	$0.35 \pm 0.7$

256

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

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

262 The delayed (1997) Southern Hemisphere mid and high latitude values of  $T_A$  are caused by coupling to  
263 the increasing Antarctic spring ozone loss after 1979 until a recovery starting in about 1998-2000  
264 (Solomon et al., 2016). The mid and high latitude, from  $35^\circ\text{S}$  to  $65^\circ\text{S}$ , delay is caused by the summer  
265 mixing of ozone poor air from the Antarctic region with SH midlatitude ozone-rich air once the polar  
266 vortex winds break down in November-December.

267  
268 The asymmetry between the Arctic and Antarctic is caused by the lower winter Antarctic temperatures  
269 ( $-80^\circ\text{C}$ ) leading to the formation of low altitude clouds containing ice crystals along with the isolating  
270 Antarctic polar vortex winds (Solomon et al., 2007; 2016). In the spring sunlight the ice and water  
271 droplets (Tritscher, et al., 2021) release ODS and depletes ozone to a monthly average of about 155 DU.  
272 During the summer, air exchange with ozone rich air from lower latitudes comes into the polar latitudes and fill  
273 in the ozone layer above Antarctica (monthly average about 300 DU. Smaller but significant ozone losses  
274 occurred in the Arctic region caused by occasional low temperatures and ODSs. The Arctic does not  
275 routinely have the low temperatures needed for winter ice clouds nor does it have the persistent  
276 isolating polar vortex winds because of wave action forced by the land topography The latitude band at  
277  $75^\circ\text{N}$  (Fig.1) has the highest amount of monthly average winter ozone  $450\pm 25$  DU that decreases to  
278  $290\pm 20$  DU monthly average during the summer that are comparable to mid-latitude values. The result  
279 is earlier values of  $T_A$  in the NH compared to the SH. The NH  $T_A$  is earlier than the 1997 minimum in  
280 stratospheric halogens (Weber et al., 2022; Newman et al., 2007). Note that  $T_A$  is not the time of the  
281 start of recovery, but rather the time for the end of rapid ozone decrease.

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

289 An analysis of ozone trends prior to the start of reliable satellite data in late 1978 showed that the  
290 annual rate of ozone loss (%/Year) increased after 1978 (Stahelin et al., 2001). Based on the first  
291 derivatives of the data in Fig. 6, the maximum annual rate of ozone reduction occurred in 1979 and 1980  
292 in the NH and SH (Fig. 9) except for  $65^\circ\text{N}$  in 1992 where the rate of loss is  $-8.75\%$ /Year. The loss rates  
293 range from  $-20.6\%$ /Year at  $75^\circ\text{S}$  to  $2.39\%$ /Year at  $5^\circ\text{N}$ . A smaller loss rate occurred for  $35^\circ\text{N}$  to  $75^\circ\text{N}$   
294 where the loss rate is almost constant between  $8\%$ /Year and  $10\%$ /Year compared to the larger SH loss  
295 rates caused by the presence of the springtime Antarctic ozone hole.

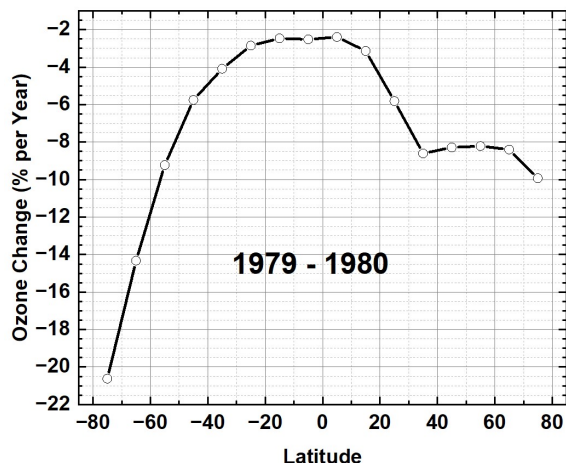


Fig. 9 The percent change in ozone per year in 1979 or 1980

296

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

301

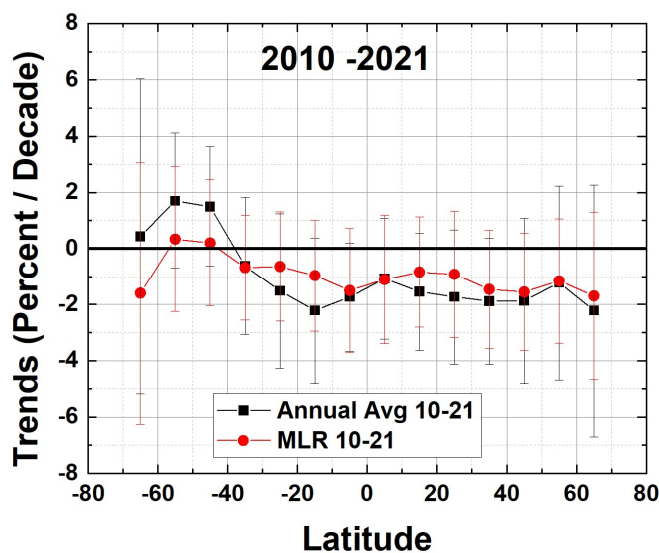


Fig. 10 Ozone trends  $P_D(\theta)$  (Percent per Decade) for the period 2010 – 2021 for the Annual Average and MLR methods applied to  $\Omega_{\text{MOD}}(t, \theta)$ .

302 Comparing deseasonalized  $\Omega_{\text{MOD}}(t, \theta)$  with deseasonalized Microwave Limb Sounder MLS (see Appendix  
 303 Figs. A1, A2, and A3) Stratospheric Ozone from 2005 to 2021 shows small average (Lowess(0.3))  
 304 differences that are within  $\pm 1\text{DU}$  except for 2021 when the differences at both 65°S and 65°N are about  
 305  $-2.5\text{DU}$ . This suggests that the calibrations of the later SBUV-2 and OMPS-NP instruments are stable. For

306 2016 to 2018,  $\Omega_{\text{MOD}}$  is obtained from NOAA-19 SBUV plus OMPS-NP and from just OMPS-NP since 2018.  
 307 Figure A3 suggests that there was a decrease in tropospheric ozone in 2020 that may correspond to  
 308 reduced economic activity during the COVID-19 pandemic.

309 Age of air AoA is a measure of how long a parcel of air resides in the stratosphere after it leaves the  
 310 troposphere (Linz et al., 2016; Ploeger et al, 2021). A comparison of  $T_A$  with AoA estimates from the  
 311 relatively inert tracer gas  $\text{CO}_2$  (Fig. 11) for the altitude range near the ozone maximum (approximately  
 312 20 km) vs latitude (based on Waugh and Hall, 2002, their Fig. 6a and Ploeger et al, 2021 their Fig. 10a)  
 313 shows near symmetry between the hemispheres with the shortest AoA in the equatorial region. The  
 314 turnaround dates  $T_A$  in Fig. 6 are also symmetric in the equatorial zone corresponding the upwelling  
 315 Brewer Dobson circulation and the smaller AoA. This suggests that the combined effects of chemistry  
 316 and dynamics on ozone amounts are similar between  $\pm 25^\circ$ . The precursors to ODS are also lifted into  
 317 the equatorial stratosphere and transported towards the polar regions (Newman et al., 2004; 2007)  
 318 where they can be photo-dissociated into ODS. Ozone at higher latitudes, NH and SH, with longer AoA,  
 319 will be dependent on transported ozone and ODS and their photochemistry, and especially the different  
 320 dynamics and chemistry in the Arctic and Antarctic regions.

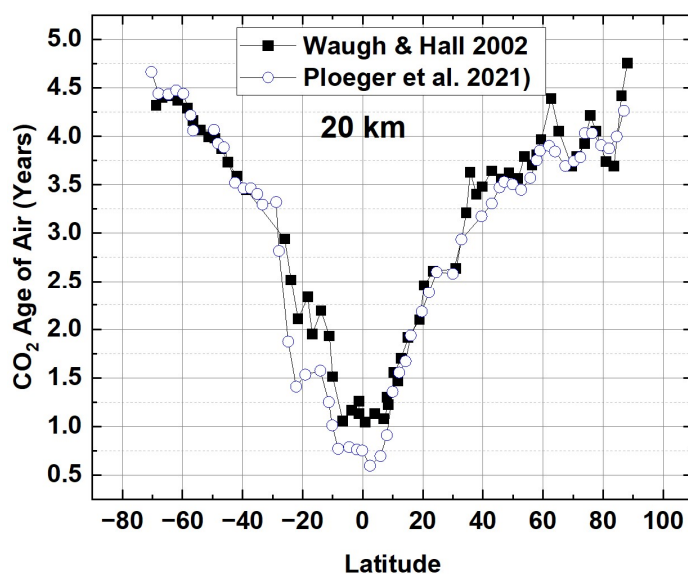


Fig. 11 Age of air derived from  $\text{CO}_2$  data (Waugh and Hall, 2002; Ploeger et al., 2021)

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328 **3.0 Summary**

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

359

360



361 **Appendix**

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

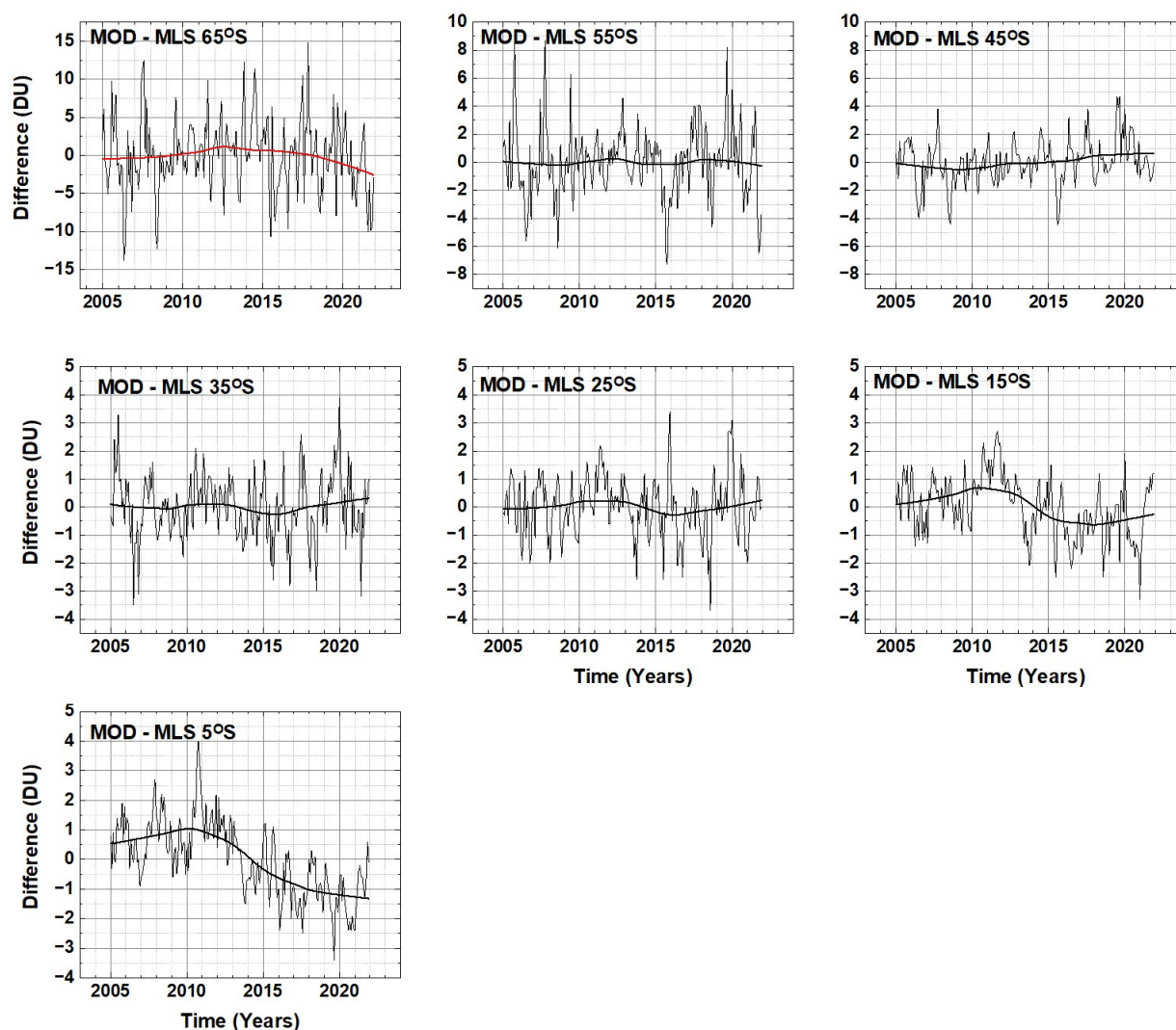


Fig. A1 A comparison of deseasonalized  $\Omega_{\text{MOD}}$  with deseasonalized MLS stratospheric column ozone for 65°S to 5°S.

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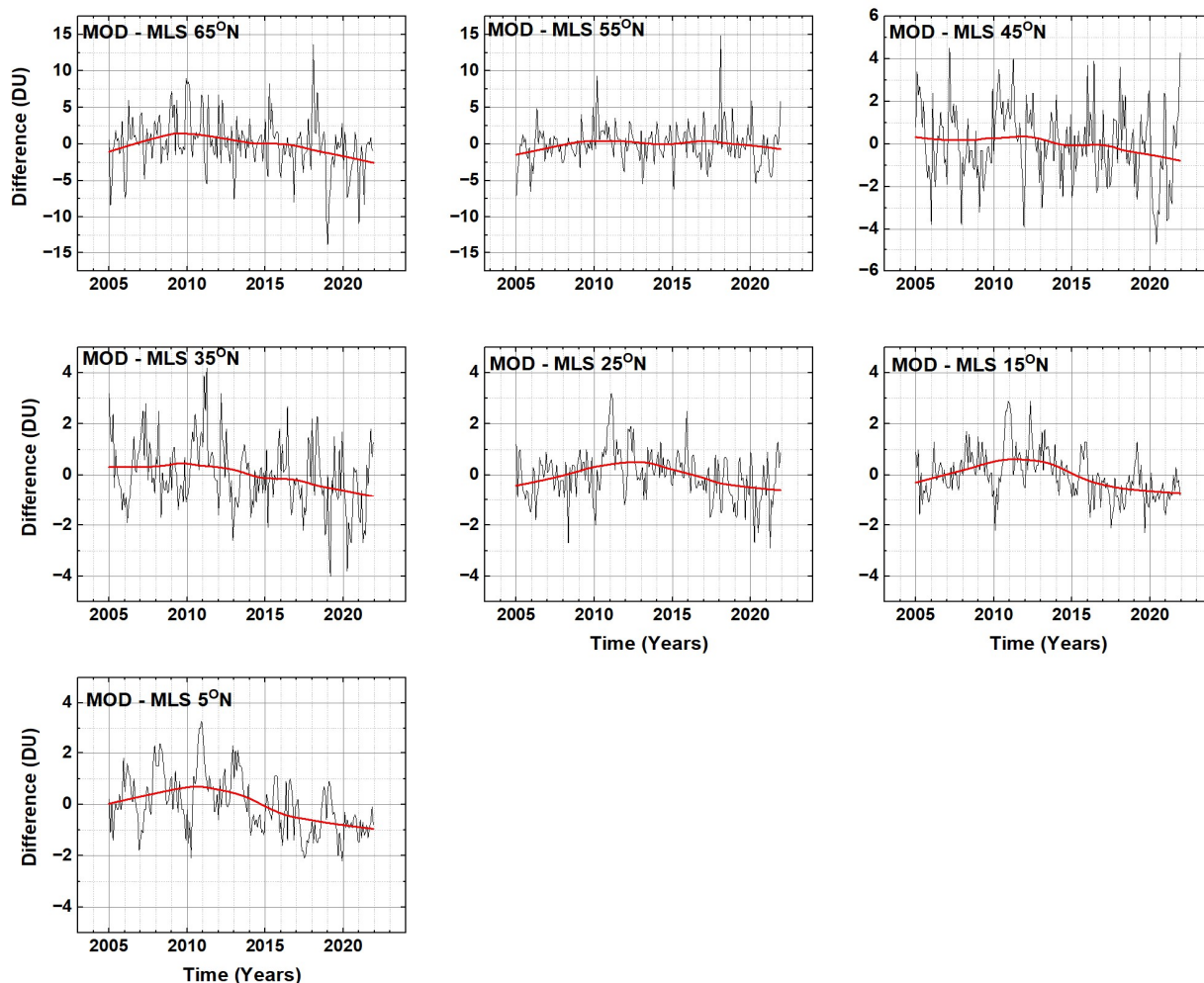


Fig. A2 A comparison of deseasonalized MOD total ozone with deseasonalized MLS stratospheric column ozone for 5°N to 65°N. Variations of  $\pm 3$ DU are within the MOD merged record uncertainties.

373

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

376

377 Since both MOD and MLS time series were deseasonalized, the mean values would be zero unless there  
 378 were changes in tropospheric ozone or instrument calibration drift. The differences are summarized in  
 379 Fig. A3 along with the  $2\sigma'$ , ( $\sigma'$  = standard deviation from the mean) error bars estimated from the  
 380 average of each deseasonalized time series. In 2020 there appears to be a systematic change in  $\langle \text{MOD} -$   
 381  $\text{MLS} \rangle$  that may be a reduction in tropospheric ozone amount of about 3 DU caused by the economic  
 382 slowdown associated with COVID-19 (Ziemke et al, 2022). The systematic change mostly recovered in  
 383 2021 (Fig. A3) except for -1DU near the equator (5°S to 15°N).

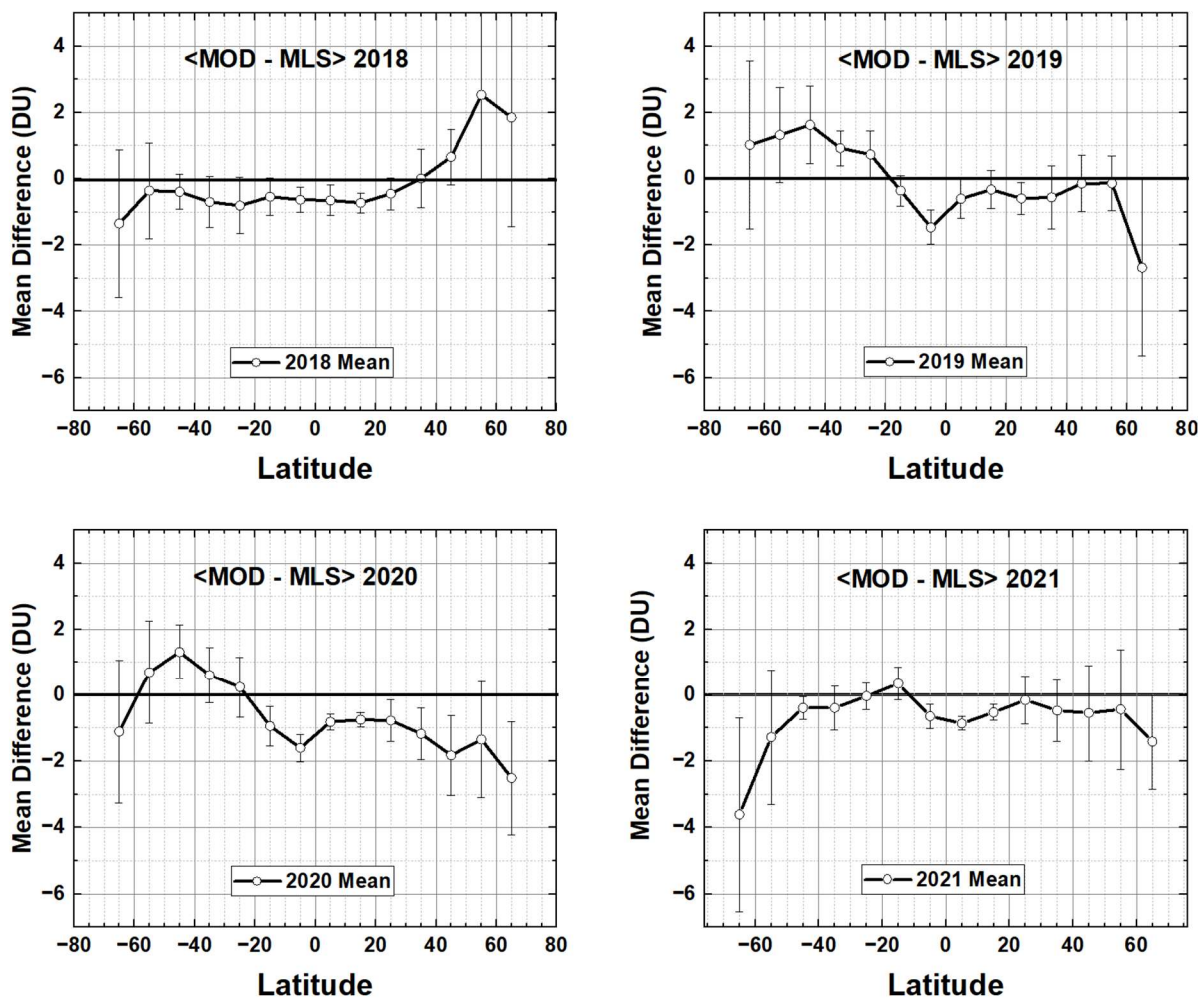


Fig. A3 Annual average <MOD – MLS> for the years 2018 to 2021. Error bars are  $2\sigma'$ , where  $\sigma'$  = standard error of the mean estimated from the average of the deseasonalized time series for each year shown in Figs. A1 and A2.

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536 **Author contribution:**

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

#### 541 **Data Availability**

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

543 [https://acd-ext.gsfc.nasa.gov/Data\\_services/merged/](https://acd-ext.gsfc.nasa.gov/Data_services/merged/)

544 and processed data in Excel format

545 [https://avdc.gsfc.nasa.gov/pub/DSCOVER/JayHerman/MOD\\_Ozone\\_Trends/](https://avdc.gsfc.nasa.gov/pub/DSCOVER/JayHerman/MOD_Ozone_Trends/)

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#### 548 **Competing interests:**

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

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