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1 Ozone trends in homogenized Umkehr, Ozonesonde, and COH

² overpass records

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19 Abstract. This study presents an updated evaluation of stratospheric ozone profile trends at 20 Arosa/Davos/Hohenpeißenberg, Switzerland/Germany, Observatory de Haute Provence (OHP), France, Boulder, 21 Colorado, Mauna Loa Observatory (MLO) and Hilo, Hawaii, and Lauder, New Zealand with focus on the ozone 22 recovery period post 2000. Trends are derived using vertical ozone profiles from NOAA's Dobson Network via the 23 Umkehr method (with a recent new homogenization), ozonesondes, and the NOAA COHesive SBUV/OMPS satellite-24 based record (COH) sampled to match geographical coordinates of the ground-based stations used in this study. 25 Analyses of long-term changes in stratospheric ozone time series were performed using the updated version (0.8.0) of the Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) Independent Linear Trend (ILT) 26 27 regression model. This study finds a consistency of the trends derived from the different observational records, which 28 is a key factor to the understanding of the recovery of the ozone layer after the implementation of the Montreal Protocol 29 and its amendments that control ozone-depleting substances production and release into the atmosphere. The Northern 30 Hemispheric Umkehr records of Aros/Davos, OHP, and MLO all show positive trends in the mid to upper stratosphere 31 with trends peaking at ~+2%/decade. Although the upper stratospheric ozone trends derived from COH satellite 32 records are more positive than those detected by the Umkehr system, the agreement is within the two sigma 33 uncertainty. Umkehr trends in the upper stratosphere at Boulder and Lauder are positive but not statistically significant, 34 while COH trends are larger and statistically significant (within 2 sigma). In the lower stratosphere, trends derived 35 from Umkehr and ozonesonde records are mostly negative (except for positive ozonesonde trends at OHP), however 36 the uncertainties are quite large. Additional dynamical proxies were investigated in the LOTUS model at five ground-37 based sites. The use of additional proxies did not significantly change trends, but equivalent latitude reduced the 38 uncertainty of the Umkehr and COH trends in the upper stratosphere and at higher latitudes. In lower layers, additional 39 predictors (tropopause pressure for all stations, two extra components of Quasi-Biennial Oscillation at MLO, Arctic





- 40 Oscillation at Arosa/Davos, OHP and MLO) improve the model fit and reduce trend uncertainties as seen by Umkehr
- 41 and sonde.

42 1 Introduction

43 The WMO Ozone Assessments (WMO, 2018; WMO, 2022), indicate that for some geographical regions, the 44 stratospheric ozone layer is recovering in accordance with the reduction of ozone depleting substances (ODS) whose 45 production was restricted by the Montreal Protocol and its amendments. The US Clean Air Act requires NOAA to 46 monitor prohibited chemicals and the ozone layer to ensure the success of the Montreal Protocol. NOAA's long-term 47 network of measurements helps to interpret total column and vertically resolved ozone changes and link ozone 48 recovery to the reduction of ODS levels in the stratosphere, changes in the lower stratosphere that are associated with 49 climate changes, and to the increases in the troposphere that are influenced by the stratosphere/troposphere exchange 50 and long-range transported pollution. The ongoing recovery of the stratospheric ozone layer is of great importance to 51 human health (i.e. cancer from enhanced UV exposures, Madronich et al., 2021), the sustained production of crops, 52 and the success of fisheries (dangerous algae blooms). For more information see the Environmental Effects 53 Assessment Panel 2022 Quadrennial Assessment (EEAP, 2023). 54 Studies of ozone recovery require long-term datasets often consisting of data merged from several instruments, or 55 from a single instrument type on multiple satellite platforms, or at a ground-based (GB) station. 2011 saw the initiation 56 of the SPARC/IO3C/IGACO-O3/NDACC (SI2N) activity to evaluate ozone trends in the depletion and recovery phases using both GB and merged satellite observations. The resulting report (Harris et al., 2015) emerged near the 57 release of the 2014 WMO Ozone Assessment (WMO, 2014). The two studies resulted in broadly similar trend values 58 59 in both the depletion and recovery phases. But the WMO report determined the recovery trends to be statistically 60 significant, whereas the SI2N study did not. The discrepancy centered on differing error analysis techniques for the 61 merged datasets: SI2N using distribution of the individual variances around the mean and WMO using weighted mean 62 of the individual standard deviations. The Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) study sought to reconcile the differences 63 between the WMO Assessment and the SI2N study. Phase 1 focused on developing best practices for data merging, 64 65 trend determination and error analyses. Results focused on analysis of broad latitudinal regions, near global, Northern 66 and Southern Hemisphere, and Tropics as were used in the SI2N studies. Results are found in the 2019 report 67 (SPARC/IO3C/GAW, 2019). Phase 2 refined the trend models, and extended the study to gridded, and GB ozone data 68 sets. The development of methods used in trend detection is built on the community knowledge gained during the 69 Tiger Team project in early 1990s (Reinsel et al., 2005), collaborations through the SPARC, WMO and IO3C

- supported LOTUS activity (Hassler et al., 2014; Harris et al., 2015; Godin-Beekmann et al., 2022) and the most recent
- 71 contributions to the WMO Ozone assessment analyses published in Chapter 3, "Update on Global Ozone: Past, Present

72 and Future" (Hassler et al., 2022).

- 73 Understanding the causes of the differences between GB and satellite records can create improvements not only in the
- 74 internal consistency of data sets, but also in the uncertainties of overall ozone trends. Further, development of
- 75 techniques to directly assess uncertainties in the merged records resulting from discrepancies that cannot be completely





76 reconciled, such as small relative drifts and differences resulting from coordinate transformations and sampling 77 differences, allows for a more precise estimate of significance of the mean trends. For the GB and satellite data used 78 in the 2019 LOTUS Report, information on stability and drifts of the measurement was incomplete. The 79 homogenization of many ozonesonde records was recently addressed and data were reprocessed (Tarasick et al., 2016; 80 Van Malderen et al., 2016; Witte et al., 2017; Sterling et al., 2018; Witte et al., 2018; Ancellet et al., 2022) while some 81 instrumental artifacts still need to be addressed (Smit, 2021). 82 The first attempt to evaluate representativeness of the trends derived from GB station records in the middle and upper 83 stratosphere using SBUV data was done as a part of the LOTUS activity and was discussed in the 2019 LOTUS report. 84 Comparisons of trends derived from satellite data sub-sampled at the station location (overpass) to those derived from 85 the relevant zonal average provide a measure of potential sampling errors when comparing satellite and GB trends 86 (Zerefos et al., 2018; Godin-Beekmann et al., 2022). This paper continues that work by comparing trends derived from 87 several GB and satellite records that are matched spatially. We further investigate the impact of temporal matching on 88 trends. 89 The common statistical linear regression trend model used in the 2019 LOTUS Report and the 2022 update (Godin-90 Beekmann et al., 2022) was optimized for analyses of the zonally averaged satellite data sets. However, analyses of 91 the GB and satellite overpass ozone profile data may require reconsideration of additional proxies and optimization 92 methods to improve interpretation of the processes that impact ozone changes over limited geophysical regions and 93 reduce trend uncertainties. An assessment of model sensitivities to uncertainties in the volcanic aerosols, solar cycle, 94 QBO, El Nino Southern Oscillation (ENSO) and other mechanisms also need to be considered in the GB and satellite 95 overpass record trend analysis. The localized time series for the assessment of dynamical and chemical proxies can 96 improve attribution of ozone variability, especially in the lower stratosphere, thus reducing uncertainties in the derived 97 trends. This paper provides an assessment of uncertainties in the derived trends from the NOAA ground-based, 98 ozonesonde and SBUV/OMPS (zonally averaged and overpass) records and reports improvements in the Multiple 99 Linear Regression (MLR) trend uncertainties with addition of proxies representing interannual dynamical variability 100 or long-term changes in atmospheric circulation. 101 In the LOTUS report, the ozone trends were analyzed at low and middle latitudes, with a focus on the upper and

- 102 middle stratosphere. This paper includes middle and low latitude trends assessed in the lower stratosphere and thus
- 103 offers an opportunity to test the additional proxy of the tropopause pressure (Thompson et al., 2021).

104 **2 Data**

105 2.1 Umkehr and Ozonesonde Records at NOAA

106 The Dobson Ozone Spectrophotometer has been used to study total ozone since its development in the 1920s

- 107 (Staehelin et al., 2018). Dobson records are regularly used in satellite record validation (Bai et al., 2015; Koukouli et
- al., 2016; Boynard et al., 2018) and the development of global combined ozone data records (Fioletov et al., 2008;
- 109 Hassler et al., 2018). The NOAA Dobson ozone record was homogenized in 2017 to account for inconsistencies in
- 110 past calibration records, data processing methods and selection of representative data (Evans et al., 2017). NOAA





111 Dobson instruments at 4 stations and MeteoSwiss at Arosa/Davos also measure Umkehr profiles, which are derived 112 as partial column ozone amounts in ~5 km layers. Profiles are derived using an optimum statistical inversion of Dobson 113 measurements taken continuously at different solar zenith angles (SZAs) (Petropavlovskikh, 2005; Hassler, 2014). 114 These Umkehr data were recently homogenized to assure the removal of small but significant instrumental artifacts 115 that can impact the accurate detection of stratospheric ozone trends (Petropavlovskikh et al., 2022, Maillard Barras et 116 al., 2022). This study focuses on Umkehr records from the MeteoSwiss station of Arosa/Davos, Switzerland, and on 117 Umkehr records from the NOAA stations of Boulder, Colorado), Mauna Loa Observatory (MLO), Hawaii, Lauder, 118 New Zealand, and the Umkehr record from Observatory de Haute Provence (OHP), France). NOAA/GML for Umkehr 119 data means that the NOAA optimization process was applied to the operational records (N-values) prior to the retrieval 120 of profiles. study ozone The source data used in this are available at https://gml.noaa.gov/aftp/data/ozwv/Dobson/AC4/Umkehr/Optimized/. See Table 1 for details on the GB datasets, 121 122 locations, source of data and temporal extent of data used. Umkehr measurements are typically made twice per day 123 when there is no cloud obstruction. 124 The ozonesonde instrument has been flown at 4 NOAA stations since the 1980s. Evolving instrumentation and 125 standard operating procedures led to the development of data homogenization methods by NOAA and the international

- community (i.e., ASOPOS-1, Smit, 2014) to resolve record inconsistencies in the NOAA (Sterling et al., 2018),
- 127 Canadian (Tarasick et al., 2016) and SHADOZ (Southern Hemisphere Additional Data from Ozonesonde) networks
 128 (Witte et al., 2017; Witte et al., 2018). The effort was extended in the ASOPOS-2 (Smit et al., 2021) activity and
- 129 included a larger group of stations that are part of the NDACC (Network for Detection of Atmospheric Composition
- 130 Change) and WMO GAW (World Meteorological Organization Global Atmosphere Watch program) networks. The
- 131 error budget for each profile is calculated and included in the archived files (Sterling, 2018). Modern ozonesonde
- 132 instruments measure ozone at high vertical resolution, on the order of 100 m (Thompson et al., 2019) depending on
- the altitude.
- 134 The sondes constitute an essential component of satellite calibration and cross-calibration (Hubert et al., 2016), 135 verification and improvement of climate chemistry and chemistry-transport models (Wargan et al., 2018; Stauffer et 136 al., 2019). The Dobson total ozone, Umkehr and ozonesonde profile records provide key measurements for upper and
- 137 middle stratospheric ozone trend calculations, and are part of the NOAA benchmark network for stratospheric ozone
- 138 profile observations (SPARC/IO3C/GAW, 2019; Godin-Beekmann et al., 2022; WMO, 2022).
- 139 The ozonesonde data are used for trend analyses from OHP, Boulder, and Lauder stations where we have Umkehr
- 140 observations. Ozonesondes are launched at Hilo, Hawaii, which is nearly co-located with MLO. Ozonesonde data
- 141 for the Arosa/Davos panel are selected from Hohenpeißenberg (HOH), Germany station that is in close vicinity to
- 142 Arosa/Davos station. Sonde measurements are typically measured once or twice per week, varying somewhat with
- 143 station operational procedures.
- 144 Data for the NOAA GML ozonesonde records are publicly available from the NOAA Global Monitoring Lab (GML)
- 145 at https://gml.noaa.gov/aftp/data/ozwv/Ozonesonde/. We use the '100 Meter Average Files' in each station directory.
- 146 Other sonde datasets used in this study are also available at several other data centers including the World Ozone and
- 147 Ultraviolet Radiation Data Centre (WOUDC, www.woudc.org), Network for the Detection of Atmospheric





- 148 Composition Change (NDACC, www.ndacc.org) data centers, or at the Harmonization and Evaluation of Ground-
- 149 based Instruments for Free-Tropospheric Ozone Measurements (HEGIFTOM, <u>https://hegiftom.meteo.be/</u>) archive.
- 150 Table 1 denotes the source of each dataset used in this study.
- 151 The ozonesonde data is of significantly higher vertical resolution (even when used as 100 m averages) than the Umkehr
- 152 data layers of approximately 5000 m. In order to create a dataset with comparable resolution, we use the Umkehr
- 153 averaging kernels (AK) to smooth the sonde data. Details appear in Appendix A. We cap the sonde profile at Umkehr
- 154 layer 5 (16–32 hPa) as there is not sufficient sonde information at higher altitudes to meet the requirements of the AKs
- 155 for layers 6 and above. We further match the ozonesonde data to the dates when both Umkehr and sonde data are
- 156 available using \pm 24 hours to find a match, then generate the ozonesonde monthly mean. Appendix D explores the
- 157 impact of temporal sampling on trends. The final matched dataset, with AK averaging, is publicly available at
- 158 https://gml.noaa.gov/aftp/ozwv/Publications/2023 Umkehr Ozone Trends Paper/.
- 159

Location	WOUD C Station #	Instrument	Date Range used in trend calculations	Source
Arosa/Davos Arosa, Switzerland (46.8° N, 9.7° E) Davos, Switzerland (46.8° N, 9.8° E)	035	Umkehr	1980 – 2018 2018 – 2020	Optimization by NOAA/GML
Hohenpeißenberg (HOH), Germany (47.8° N, 11.0° E)	099	Ozonesonde	1980 - 2020	NDACC
Observatory de Haute Provence (OHP), France (43,9°N, 5,8°	040	Umkehr	1983 - 2020	NOAA/GML
E)		Ozonesonde	1991 – 2020	NDACC (same as HEGIFTOM)
Boulder, Colorado (40.0° N. 105.3° W)	067	Umkehr	1980 - 2020	NOAA/GML
		Ozonesonde	1980 - 2020	NOAA/GML - 100m average data
Mauna Loa Observatory (MLO), Hawaii (19.5°N, 155.6°W)	031	Umkehr	1982 – 2020	NOAA/GML





Hilo, Hawaii (19.7° N,155.1° W)	109	Ozonesonde	1982 – 2020	NOAA/GML - 100m average data
Lauder, New Zealand (45.0°S, 169.7°E)	256	Umkehr	1987 – 2020	NOAA/GML
		Ozonesonde	1987 – 2020	NDACC

160

161

Table 1: GB datasets, location, instrument type, temporal extent, data record source. For the trend calculations we remove 162 data during volcanic periods from 1982-1984 and 1991-1994.

163 2.2 The NOAA Cohesive (COH) Station Overpass Ozone Profile Datasets 164

NASA and NOAA have produced satellite measurements of ozone profiles through the Solar Backscatter Ultraviolet 165 166 (SBUV) on the sequence of Polar Orbiting Environmental Satellites (POES) since 1978. This measurement series is extended with the related Ozone Mapping and Profiler Suite (OMPS) nadir profiler (NP) instruments using similar 167 168 measurement techniques and retrieval algorithms. These combine to provide nearly 45 years of continuous data (1978 - present). This single instrument type dataset eliminates many homogeneity issues including varying vertical 169 resolution, or instrumentation differences. Version 8.6 SBUV data incorporates additional calibration adjustments 170 171 beyond the Version 8 release (McPeters et al., 2013). Small but evident biases remain (Kramarova et al., 2013a). 172 Several methods have been historically used to combine these datasets into a continuous series. The NASA MOD 173 version 1 dataset based on SBUV and OMPS v8.6 (Frith, 2014) combines data from all available satellites with no 174 modification or bias adjustments. NASA has developed an alternate processing for the SBUV and OMPS data (v8.7) 175 which incorporates new calibrations at the radiance level, and updated a priori with improved troposphere. 176 Additionally, the a priori is chosen to be representative of the local solar time of the measurement. MOD v2 is based 177 on the v8.7 processing (Frith et al., 2020), and further applies an adjustment to the v8.7 data to shift all measurements to a nominal measurement time of 1:30 PM local time. 178 179 The NOAA SBUV/2 and OMPS Cohesive dataset (further referred to as COH) combines data from the SBUV/2 and 180 OMPS instruments using NASA's version 8.6 for the SBUV/2 data and NOAA/NESDIS version 4r1 for the OMPS 181 Suomi National Polar-orbiting Partnership (SNPP) data. This dataset uses correlation-based adjustments providing

182 an overall bias adjustment plus an ozone-dependent factor (Wild et al., 2016) to moderate the remaining biases

183 between instruments in the series. The resulting profile product is a set of daily or monthly zonal means and is publicly

184 available at https://ftp.cpc.ncep.noaa.gov/SBUV CDR. Zones are 5° wide in latitude, identified by the central latitude

 $(2.5^{\circ}, 7.5^{\circ}, \text{etc.})$. Contributing satellites and their period of use is shown in Table 2. 185

Satellite	Dates
Nimbus 07	10/1978 - 5/1989
NOAA 11	6/1989 - 12/1993





NOAA 09	1/1994 - 6/1997
NOAA 11	7/1997 - 12/2000
NOAA 16	1/2001 - 12/2003
NOAA 17	1/2004 - 12/2005
NOAA 18	1/2006 - 12/2010
NOAA 19	1/2011 - 12/2013
SNPP	1/2014 - present

187 Table 2: Satellite mapping for COH data series

188 A previous version of this dataset using OMPS v3r2 has been used in climate reviews and trend studies (Godin-

Beekmann et al., 2022; Weber et al., 2022a, 2022b) including Chapter 3 of the WMO Ozone Assessment (Hassler et 189

190 al., 2022). Appendix B examines the differences between the data versions. The impact on trends is limited to less 191 than 1% per decade, well within the precision of the trend results.

192 We create the overpass data at a ground station by collecting all profiles from a satellite within a $\pm 2/20^{\circ}$ 193 latitude/longitude box centred on the station. The box size is chosen to ensure that one to four points are found per 194 day. Fewer points are found if the orbit passes directly over the station; more points are found if the orbits straddle 195 the station. The collected profiles are inverse-distance weighted to the station location and averaged. COH style adjustments are applied (Wild et al., 2016) creating a COH overpass time series from 1978 to present. This dataset is 196 197

available on the NOAA website at https://ftp.cpc.ncep.noaa.gov/SBUV CDR/overpass.

Figure 1 shows the ozone anomaly time series for the 40-45° N zonal average data, and for the data at 3 stations in or 198 199 near the zone. Anomalies are calculated with respect to the zonal average climatology. The series shown are for the 200 layer data with the bottom pressure of the layer displayed on the right side of the graph. This depiction retains the information about the relative differences between the stations and the zonal average. In the mid-stratosphere (25-10 201 202 hPa) the biases between the stations are most pronounced with Arosa/Davos usually showing less ozone and Boulder 203 usually showing more ozone. At the uppermost layers (1 and 4 hPa), and the lowest layer (41 hPa) the bias between 204 stations is reduced. The anomalies for Arosa/Davos and OHP, which are geographically closer than Boulder, are often 205 nearly anticorrelated with the Boulder anomalies especially in the second half of the year. Indeed at 16 hPa in 206 particular, one can see that often the Boulder anomalies are positive when the Arosa/Davos and OHP anomalies are 207 negative.







209

Figure 1. Monthly ozone anomaly relative to the zonal mean monthly averages. This process leaves intact the trend for each site and the zone, and accentuates the differences between the station values since all anomalies are referenced to the zonal product. Evident at 4 hPa is a positive trend from 2002 to 2013, then a levelling out after.

213 Figure 2 also shows the anomalies for the 40-45° N zonal average with the station anomalies, but each anomaly is 214 now created using the climatology derived from each separate dataset. This removes the bias between the stations and 215 the zones. At 1 hPa, Arosa/Davos appears to display the most variation (largest peaks and dips) in the anomalies. 216 Since the anomaly for each site is now based on the seasonality of each site's data the structure in the anomalies is 217 more uniform. For example, now at 16 hPa, the difference between Boulder and the two sites Arosa/Davos and OHP 218 in the latter half of the year is removed. In 2012, where the Boulder anomaly was positive with respect to the zonal 219 average seasonal value, and the Arosa/Davos and OHP sets were negative with respect to the zonal seasonal average, 220 all are now of the same sign with respect to their own seasonal averages. Nonetheless, there are events where one station shows opposite anomalies to the other two, for example early 2009 at 41 hPa, when the Boulder anomaly is 221 222 negative, and Arosa/Davos and OHP are positive. Thus, it is noted that when comparing daily or monthly data values 223 from GB and satellite data, the overpass data will reveal a different structure than the zonal data. The trend calculations 224 in this paper are based on the datasets of Fig. 2, where the seasonal behavior is removed using the station seasonality. 225









Figure 2. Monthly ozone anomaly relative to the monthly climatology for each station overpass dataset. This process leaves intact the trend for each site and the zone, and shows the consistency among the stations when each station climatology is removed. This dataset is used for the trends calculations. Evident at 4 hPa is a positive trend from 2002 to 2013, then a leveling out after. Trends are run on this dataset.

231 The COH overpass and zonal datasets have a similar vertical granularity as the Umkehr dataset, but use somewhat 232 different pressures for the demarcation of the top and bottom of each layer. Since no additional smoothing is required, 233 we simply use interpolation and integration to convert the COH layer profiles to the Umkehr layers. We exclude layers 234 1 to 4 since there is little sensitivity in SBUV and OMPS NP in these layers (Kramarova et al., 2013b). The overpass 235 monthly-mean dataset in this study uses all COH data matched to dates when Umkehr data also exists. This dataset is 236 publicly available at https://gml.noaa.gov/aftp/ozwv/Publications/2023 Umkehr Ozone Trends Paper/. Appendix 237 D explores the impact of temporal sampling on trends. 238 This study also uses a specialized zonal monthly-mean COH product which is the average of all daily profiles with an 239

- 239 Umkehr match at the associated GB station. Zones used for most stations are the 5° wide zone which includes the 240 geographic station latitude (Arosa/Davies: 47.5° N, OHP: 42.5° N, MLO: 17.5° N). Boulder and Lauder, however,
- are located directly on the border of two zones, so the zonal product in this study is the mathematical average of the
- 242 two adjacent zones (Boulder: 37.5° N and 42.5° N, Lauder 42.5° S and 47.5° S).





243 3 Methods

244 3.1 LOTUS Model overview - the Reference Model

- 245 The Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) activity is a project of SPARC
- 246 (Stratosphere-Troposphere Processes and their Role in Climate) and has produced a statistical Multiple Linear
- 247 Regression (MLR) model called the LOTUS model (https://usask-arg.github.io/lotus-regression/index.html).
- 248 The 2019 LOTUS report (SPARC/IO3C/GAW, 2019) and update (Godin-Beekmann et al., 2022) have quantified
- 249 stratospheric ozone trends and evaluated their uncertainties. The LOTUS model is a general-least-squares approach
- 250 MLR model. This study uses version 1 (v 0.8.0) with the independent linear trends (ILT) configuration. The
- 251 independent linear terms represent the ozone depletion period (pre-1997), the ozone recovery period (post-2000) and
- an optional gap period (1997–2000). We will call the terms "pre", "post" and "gap" for short. The version 0.8.0 adds
- an option to enforce continuity across the gap period which is used in this study. The regression uses an interactive
- 254 procedure (Cochrane and Orcutt, 1949) and the autocorrelation coefficient is adjusted with each iteration. The
- 255 covariance matrix is modified accordingly to account for measurement gaps (Savin and White, 1978).
- 256 The LOTUS model (further referred as reference model in this study) is written here:

257 $\hat{y}(t,z) = \beta_0(t,z)C_{pre}(t) + \beta_1(t,z)C_{post}(t) + \beta_2(t,z)Linear_{pre}(t) + \beta_3(t,z)Linear_{post}(t) + \sum_{i=4}^n \beta_i X_i(t,z) + \epsilon(t,z)$ (1)

where $\hat{y}(t, z)$ is the estimated ozone at time *t* and altitude *z*; β are the fitted coefficients of the model; the residual term, $\varepsilon(t, z)$ is the difference between the LOTUS model and the input data. C_{pre} and C_{post} are the constant terms as defined by:

261

$$Constant_{post} = egin{cases} 0 & ext{for } t \geq 2000\text{-Jan} \ 0 & ext{for } t < 1997\text{-Jan} \ mt & ext{for } 1997\text{-Jan} \leq t < 2000\text{-Jan} \ 1 & ext{for } t \geq 2000\text{-Jan} \ \end{array}$$

 $Constant_{pre} = \begin{cases} 1 & \text{for } t < 1997\text{-Jan} \\ 1 - mt & \text{for } 1997\text{-Jan} \le t < 2000\text{-Jan} \\ 0 & \text{for } 1997\text{-Jan} \le t < 2000\text{-Jan} \end{cases}$

262

where 0.29135 and t = month starting in January 1980 and ending in December 2020. Indeed, the constant terms are only constant in the "pre" and "post" periods. The 3-year "gap" period is represented by a line of slope m connecting

the two constant (pre and post period bias) terms.

266 The linear terms of the model are defined as:

267

268

where m=0.008487, b = -1.700240, and t = month starting in January 1980 and ending in December 2020.

270 Natural variability is a complicating factor in deriving trends associated with the changes in the ozone depleting 271 chemistry. LOTUS fits predictor variables as proxies for natural variability to the ozone data so that one can interpret

the resulting linear trend as a trend due to the changes in chemistry. The summation term is the summation of the

- 273 predictors used as a proxy for the dynamical induced ozone variability.





The natural variability proxies in the LOTUS model v 0.8.0 are Aerosol Optical Depth (AOD), El Nino/ Southern Oscillation (ENSO), and the Quasi-Biennial Oscillation (QBO) in the form of the first two principal components (also

known as an empirical orthogonal function analysis). The data sources for each are described in Table 3.

277 Large SO₂ levels after volcanic eruptions can impact the validity of sonde ozone retrievals (Yoon et al., 2022). Both

278 Umkehr and satellite ozone profiles from SBUV and OMPS are highly uncertain and/or biased because of high aerosol

279 load during volcanic eruptions (DeLuisi et al, 1989; Petropavlovskikh et al., 2005, 2022; Bhartia et al, 1993, Torres

et al., 1995, Bhartia et al, 2013). It is recommended that the data for 2 to 3 years after the El-Chichon and Pinatubo

281 large volcanic eruptions should not be used in trend analyses. Therefore, we exclude data during the volcanic periods

(1982–1983 and 1991–1993) from the analyzed time series. Moreover, this study is focused on the linear trend analyses after 2000 when there are no large stratospheric aerosol perturbations that significantly influence stratospheric ozone variability over the middle latitudes and therefore impact trend and uncertainty estimates. Since

we have eliminated the data during the volcanic period, this study does not include the AOD proxy in the calculations.
We define the 'reference' model (RM) as the proxies most commonly used for the dynamical proxies which is
equivalent to the LOTUS model v 0.8.0 minus the AOD term. The representative equation is:

288

 $\sum_{i=4}^{n} \beta_i(t,z) X_i(t) = \beta_4(t,z) QBO_A(t) + \beta_5(t,z) QBO_B(t) + \beta_6(t,z) ENSO(t) + \beta_7(t,z) Solar(t)$ (2)

The Quasi-biennial Oscillation (QBO) is derived from the Singapore radiosonde profiles (1979–2020) that detect variability in the direction of the tropical winds in the lower stratosphere. It also shows that zonal wind variation propagates downward with an average period of ~28 months [Wallace, 1973]. The principal component analysis of the 100–10 hPa zonal winds can describe the majority of the wind variability. The reference model (and LOTUS v 0.8.0) use the two leading modes of the calculated empirical orthogonal functions (EOF) for trend analyses [Wallace et al., 1993].

The El Niño/ Southern Oscillation (ENSO) is a periodic mode of climate variability of the atmosphere and sea surface 295 296 temperatures associated with the equatorial Pacific Ocean with periods ranging from 2-8 years. The Multivariate 297 ENSO index (MEI) is produced by the NOAA Physical Sciences Laboratory and is derived from the EOF analysis of 298 sea surface temperature, sea level pressure, outgoing terrestrial radiation, and surface winds in the area of the Pacific 299 basin from 30° S to 30° N and from 100° E to 70° W (Wolter and Timlin, 2011). Temperature anomalies in the 300 troposphere with corresponding stratospheric temperature anomalies during El Niño/ La Niña events modulate the 301 tropical upwelling of the Brewer-Dobson circulation (BDC) and thus the meridional transport of ozone in the 302 stratosphere. (Diallo et al., 2018).

The solar cycle is the 11-year periodic cycle of solar activity and solar irradiance that reaches the Earth's atmosphere. The change in UV radiation that is absorbed by the atmosphere, most notably in the upper stratosphere, leads to changes in atmospheric temperature and the photochemistry which produces ozone. (Lee and Smith, 2003). The 10.7 cm solar radio flux data is used as the proxy for the solar cycle in the LOTUS model.

307 Seasonal components in the form of Fourier harmonics were added into the LOTUS model with version 0.8.0. Godin-

308 Beekman et al. (2022) showed in their Fig. 7 that the model fit for the ozone profile satellite and model records is

improved by adding seasonal components to the proxies, increasing the adjusted R-squared (R2) from 0.3 or less to

310 0.3 to 0.5. The seasonality and relevant contributions of some predictor's variables are compensated in this study by





- 311 adding the seasonal components to the fitted predictors. Seasonal components are represented in the model by sine
- 312 and cosine functions with periods of 12 and 6 months that describe the variability of the proxies on these timescales.
- 313 So, for each fitted predictor in the model
- $\beta_i X_i(t,z)$ where i > 1
- 315 a seasonal variation in the form of Fourier components is added as follows:

$$\beta_m(t,z) = \beta_{m,0}(z) + \sum_{i=1}^2 \beta_{m,1,i}(z) \sin(\frac{2\pi i t}{12}) + \sum_{i=1}^2 \beta_{m,2,i}(z) \cos(\frac{2\pi i t}{12})$$

317 3.2 The Extended Model - Adding Predictors

318 Recent publications (i.e. Petropavlovskikh et al., 2019; Szelag et al., 2020; Godin-Beekmann et al, 2022; Millan et al. 319 2024) highlight the need to reduce the trend uncertainties in the lower stratosphere (LS). There is still a discrepancy between modeled and observed ozone trends in the LS but large uncertainties make comparisons difficult. In this 320 321 study, we test additional predictors in the model to account for dynamical variability of ozone in the stratosphere, thus 322 improving the model performance and reducing the uncertainty of the trends. The argument for additional predictors 323 is that the LOTUS model was developed for the regression of zonally averaged ozone data, which reduces some 324 variability that might be impacting the ground-based records on regional bases. Impact of additional proxies in trend 325 analyses were reported in other publications (Weber et al, 2022a, Bernet, 2023 and references therein), and were 326 mostly found to improve the statistical model fit at high latitudes where the impact of the descending branch of the 327 Brewer-Dobson circulation and Arctic/Antarctic oscillations has contributed to additional variability in stratospheric 328 ozone records.

329 In what we define as the 'extended' model, we add single additional predictors (one at a time) in the model as such:

 $\sum_{i=4}^{n} \beta_i(t,z) X_i(t,z) = \beta_4(t,z) QBO_A(t) + \beta_5(t,z) QBO_B(t) + \beta_6(t,z) ENSO(t) + \beta_7(t,z) Solar(t) + \beta_8(t,z) X_{predictor}(t,z)$

331 The fitted predictors contain Fourier components, like in the reference model, to allow for seasonal variation.

332 We test the following additional predictors as described below to assess the impact on trends and uncertainties:

- 333
- 334

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• Quasi-Biennial Oscillation (QBO): Two notable disruptions to the otherwise relatively periodic QBO have occurred during the study period: 2015–2016 and in 2020 (Diallo, et. al 2022). Two additional leading modes of the calculated empirical orthogonal functions (EOF) are tested to improve the trend model fit during the anomalous QBO years.

Arctic/Antarctic Oscillation (AO/AAO): the pattern of surface air pressure anomalies in the polar region and certain mid-latitude regions. The AO/AAO has strong correlations (Lawrence et al 2020) with stratospheric ozone through the strength of the polar vortex. The positive phase of the AO or AAO in the winter months is associated with low activity in the vertically propagating planetary Rossby waves, a strong polar vortex, a low vortex wavenumber, and low stratospheric temperatures. Thus, the positive (negative) phase of the AO/AAO is correlated to low (high) ozone anomalies especially in the winter months (Lawrence et al, 2020).

•





- 344 North Atlantic Oscillation (NAO): Similar to the Arctic Oscillation, this is a pattern of surface air pressure 345 anomalies between certain regions in the high altitudes of the North Atlantic Ocean. This index is calculated 346 by the pressure difference between the Azores high and the subpolar low. 347 Eddy Heat Flux (EHF): The flux of heat through a zonal plane by transport due to the Brewer-Dobson circulation, here averaged from 45-75° N/S (use EHF S for Lauder only). This represents the planetary wave 348 349 activity that drives transport of ozone. Tropopause Pressure (TP): Pressure level of boundary between the troposphere and the stratosphere. In this 350 351 study, we use the monthly mean pressure level of the tropopause from the NOAA National Centers for 352 Environmental Prediction (NCEP) reanalysis product. As the troposphere warms due to release of GHGs 353 and the stratosphere cools due to ODSs destroying stratospheric ozone, the tropopause is rising (Meng et al., 354 2021). Thompson et al. (2021) and Stauffer et al., (2023) found that the lower stratospheric ozone trends in 355 tropics become slightly positive when recomputed with respect to the tropopause height (which has its own 356 trend). This finding indicates that ozone depletion in the lower stratosphere (i.e. Ball et al., 2020) is driven 357 by climate-change-related changes in transport and mixing in the lower stratosphere. Therefore, we are 358 testing the TP proxy in the model to account for non-chemical ozone losses in order to assess chemical 359 attribution of ozone trends. 360 Equivalent Latitude (EqLat): Geographical latitude of the isoline encircling the area of equal Potential Vorticity (PV) (Lary et al, 1995). The EqLat normalizes the range of PV values that change with season and
- 361 interannual and makes it convenient for interpretation of ozone variability and trends (i.e. Wohltmann et al 362 363 2005). The dataset was generated from GMI CTM analyses (private communications with Susan Strahan, 364 June 2021) for each ground-based station overpass criteria (latitude and longitude envelope, see above) and 365 at several altitude levels coincident with Umkehr ozone profile layers. Appendix C discusses a COH dataset based on EqLat instead of geometric latitude. No advantage was found by using the EqLat coordinate system 366 367 for the COH zonal dataset.
- 368 Source datasets for all predictors in the reference and extended models are shown in Table 3.

Predictor	Description	Source				
ENSO	El Nino/Southern Oscillation	Monthly https://psl.r	Mean 10aa.gov/enso/1	Multivariate nei.old/ ¹	ENSO	Index
Solar	Solar 10.7cm flux	https://spac solaire/sola	eweather.gc.ca rflux/sx-5-en.p	ı/forecast-prevision/s hp	olar-	





QBO	Quasi-Biennial Oscillation	Principal Component Analysis of the Monthly Mean Zonal Wind https://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/qbo.dat
AOD	AOD is included in t	he LOTUS model, but not used in this study
AO	Arctic Oscillation, Monthly Mean index	http://www.cpc.ncep.noaa.gov/products/precip/CWlink/daily_ao_index/m onthly.ao.index.b50.current.ascii
AAO	Antarctic Oscillation, Monthly Mean index	https://www.cpc.ncep.noaa.gov/products/precip/CWlink/daily_ao_index/a ao/aao.shtml
NAO	The North Atlantic Oscillation, monthly mean index	https://www.cpc.ncep.noaa.gov/products/precip/CWlink/pna/norm.nao.mo nthly.b5001.current.ascii.table
EHF	Eddy Heat Flux	Cumulative Mean (from September to April) of Heat Flux at 100 hPa from MERRA2 reanalysis averaged over 45–75° N (45–75° S for Lauder), deseasonalized. It is kept constant from April to Sep. https://acd-ext.gsfc.nasa.gov/Data_services/met/ann_data.html
ТР	Tropopause Pressure	Monthly Mean NCEP-NCAR reanalysis (Kalnay et al., 1996); Tropopause pressure at the lat/lon of each station, deseasonalized. ftp://ftp.cdc.noaa.gov/Datasets/ncep.reanalysis.derived/tropopause/pres.tro pp.mon.mean.nc
EqLat	Equivalent Latitude	Monthly Mean equivalent latitude derived from MERRA2 -GMI CTM potential vorticity (PV) contours on 31 potential temperature surfaces [Susan Strahan, private communication, 8/24/2022]. The PV at each station is determined by a 1/distance weighted average of the values in a \pm 2 lat, \pm 2 lon grid, then converted to EqLat on the Umkehr layers.





369Table 3: List of predictors either previously used (bolded) in the LOTUS 0.8.0 (reference) model and additional predictors370evaluated in this study for a future use in the extended LOTUS trend regression model. Note, two components of the QBO371predictors were used in the reference model (i.e. Godin-Beekmann et al., 2022). We added two more components in the372extended model for tests described in this paper.

¹ Since the incorporation of the ENSO index into the LOTUS model, NOAA GSL has updated the index to v1.2.
 <u>https://psl.noaa.gov/enso/mei/</u>. However, for consistency with results from the Godin-Beekmann (2022) paper we use the old MEI index that is part of the LOTUS v 0.8.0 package.

376 All proxies are used as is. No de-trending (removal of the long-term trend in proxy) is applied to the proxies. Therefore,

377 we interpret any changes to the trends derived with additional proxies as approximations of trends driven by chemistry

378 and transport related to climate change. These are rough approximations as some feedbacks are known to impact

379 chemistry (e.g. changes in stratospheric temperature).

380 3.3 The Full Model - Combining Additional Predictors

381 After we have determined the impact of the additional predictors singly, we discern which predictors should be 382 combined to constitute the 'Full Model'. Prior to selecting additional predictors for the 'Full Model', we perform 383 correlation tests to identify any cross correlations between predictors. We select predictors that are not highly 384 correlated (less than +/- 0.2) to ensure that all predictors are largely independent. We use the square of the Pearson 385 correlation coefficient R2 for each pair of the predictors to test our assumptions. We find that ENSO, Solar, QBO 386 (1,2,3,4), AAO, AO, EHF (N and S), and TP (at each station) have correlations less than +/- 0.2 (with the exception 387 of R2 = 0.3 for EHF (N) and AO). Therefore, any of these predictors can be combined in the 'Full Model'. We find 388 that NAO has a correlation of .38 with AO so we do not use these two predictors in the same model. 389 We also test the independence of EqLat proxies calculated at several geographic locations (defined by the latitude and 390 longitude of each Umkehr station) and by selecting a proxy at several altitude levels centered in the middle of Umkehr 391 layers 3–9. We find that the R2 between the TP and EqLat in the lower stratosphere (Umkehr layer 3) can be large but anticorrelated -0.7 (Boulder), moderate 0.4 (MLO and Lauder), while close to zero at Arosa/Davos and low at OHP 392 393 (-0.2). In the middle and upper stratosphere, the R2 varies from -0.5 to -0.4 (MLO), 0.2 to 0.3 (Arosa/Davos and 394 OHP), 0.5 to 0.6 (Boulder), and 0.4 to 0.7 (Lauder). EqLat has mostly low correlations ($\leq \pm 0.3$) with all other proxies

except for higher correlations with QBO B in layers 5 (-0.3) and 6 (-0.4), and QBO A in layer 7 (0.3) at MLO; and with AO in layer 8 (0.3) at OHP and Arosa/Davos. Also, EqLat has no correlation with the TP proxy in layer 4 in

Boulder, in layer 9 at Lauder, and in layers 8 and 9 at OHP. Since there are occasional high correlations between

398 EqLat and TP proxies, we do not use them together in the 'Full trend Model'.

399 4 Results

400 4.1 Reference Model Trend Results

First, we discuss the reference model trends derived from the COH overpass, Umkehr and ozonesonde records at 5 geographic locations. All datasets are deseasonalized with a climatology computed from a subset of data taken from

- 403 1998–2008 prior to the trend analysis. Trend results are presented in Fig. 3 and organized in 5 panels. Each panel
- 404 shows trends at selected pressure/altitude levels detected from Umkehr (green), COH (orange) and ozonesonde (blue)





405 records at Arosa/Davos, OHP, Boulder, MLO/Hilo and Lauder ground-based stations. Ozonesonde data for the 406 Arosa/Davos panel are selected from Hohenpeißenberg, Germany station that is in close vicinity to Arosa/Davos 407 station. We show trends for layers where the measurement is of highest quality: Umkehr (layer 3 through 8), COH

408 (layers 5 through 9) and ozonesonde (layers 3 through 5) records.

- 409 The Umkehr data used in this analysis is the monthly mean of all available Umkehr data (one or two measurements
- 410 per day). The sonde and COH monthly means use only those profiles that have corresponding Umkehr measurements
- 411 on that date. We explore the impact of temporal sampling on trends in Appendix D. For COH with the Umkehr
- 412 matched data, trends are slightly larger at OHP but well within the error bars. At all other stations the COH trends are
- 413 not impacted by sampling. At OHP the ozonesonde trends matched to Umkehr are slightly larger at layer 4 only and
- 414 well within the error bars; while at Lauder in layers 4 and 5 trends are smaller, but barely within the error bars.
- 415 In the upper (above 10 hPa) stratosphere, Umkehr (green) and COH (orange) trends are positive and agree within the
- 416 error bars (+/- 2 standard errors). The exception is found at 8–2 hPa pressure level over the Lauder station, where
- 417 Umkehr trends are near zero and COH trends are $\sim +3-4$ %/decade. The error bars show +/- 2 standard errors, and the
- 418 fact that they do not overlap suggests that the differences in trends are statistically significant. This could be related
- 419 to the relatively large uncertainties in the instrumental corrections applied to homogenize the Umkehr record
- 420 (Petropavlovskikh et al, 2022). Björklund (2023) discusses relative drifts in Umkehr, ozonesonde, FTIR and MW
- 421 ozone records over Lauder. The authors are not able to identify instrumental artifacts that may have caused the 422 discrepancies in the co-located records, but point out that it is not related to the sampling biases.
- 423 In the middle stratosphere (60–10 hPa) agreement between Umkehr and COH is within uncertainty of the trend except 424 at Arosa/Davos where COH trends are statistically different from Umkehr trends at 16–8 hPa. COH trends at 32–16 425 hPa are mostly negative (-2–3 %/decade) with the exception of Lauder where trends are near zero and similar to 426 Umkehr trends. Umkehr trends between 32–16 hPa are close to zero. The ozonesonde trends (blue) agree with COH 427 (orange) and Umkehr (green) trends in layer 63–16 hPa at Arosa/Davos, Boulder and MLO. However, at OHP 428 (Lauder) the ozonesonde trends are found to be positive at +3±3 %/decade (negative at -3±1.5 %/decade) and
- 429 significantly different from the near-zero trends seen in the COH and Umkehr results.
- In the lower stratosphere (125–63 hPa), Umkehr trends vary between small positive (+1–2 %/decade at Hilo and
 Lauder) and negative (-2–3 %/decade at Arosa/Davos, OHP and Boulder); however, trend uncertainties are the largest
 (2 standard errors are 2–3 %/decade, (see Table 4 below) in comparison to the middle and upper stratospheric trends.
 Ozonesonde trends at OHP station are positive (+4 %/decade), and negative over Lauder (-2 %/decade). They also
- 434 feature large uncertainties (±5 %/decade) that are larger than the uncertainties found in Umkehr trends which could
- 435 be caused by the limited sampling (see Appendix D). Sonde trends at Hilo show negative trend values with large
- 436 uncertainties. But the data in this study at Hilo is not corrected for the ozonesonde drop off after 2014 known to occur
- 437 at this station (Stauffer, 2022), so the deviation from the Umkehr results at these levels may be misleading.
- 438 Figure 3 also shows trends derived from the zonal-mean COH data associated with each station (orange dashed line).
- 439 These are shown for comparison with the overpass COH data (solid line) to study the impact of the spatial sampling
- 440 biases on the trends. Though Figs. 1 and 2 show clear interannual differences between the records from the individual
- stations, and the associated zonal average, we find very small differences in trends (mostly in the upper stratosphere





- 442 at middle latitude stations). Therefore, the station overpass sampling provides trends that are representative of the
- 443 zonal averaged trends (Zerefos, 2018) and the discrepancies in trends between GB and satellite records do not strongly
- 444 depend on the spatial sampling differences.
- 445



446

Figure 3: The 2000–2020 ozone trends are shown at 7 altitude/pressure levels. The LOTUS model v 0.8.0 is used for trend analyses. Umkehr trends (green), COH (orange) and ozonesonde (blue) are shown for 5 ground-based stations: Arosa/Davos, OHP, Boulder, MLO and Lauder (panels left to right). Ozonesonde data for the Arosa/Davos panel are selected from Hohenpeißenberg, Germany that is in close vicinity to Arosa/Davos. Trends from the zonal-mean COH data (orange dashed line) are shown for comparison with the overpass COH data (solid line). The error bars indicate ± 2 standard errors.

453 4.2 Standard Error of Reference Model

	LOTUS Model Proxy Tests: Standard Error for Reference Model																
Height	leight Umkehr Arosa/Davos					OHP			Boulder			MLO			Lauder		
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	
1-2	9		0.92			0.91			0.62			0.43			0.63		
2-4	8	0.85	0.59		1.06	0.68		0.51	0.52		0.52	0.37		0.72	0.57		
4-8	7	0.69	0.59		0.77	0.54		0.41	0.52		0.58	0.62		0.57	0.66		
8-16	6	0.66	0.68		0.75	0.59		0.42	0.43		0.55	0.49		0.61	0.56		
16-32	5	0.66	0.75	0.76	0.89	0.68	1.26	0.54	0.51	0.77	0.82	0.55	0.75	0.73	0.54	0.73	
32-63	4	1.05		1.04	1.13		1.95	0.90		1.04	0.90		0.94	0.83		1.16	
63-127	3	1.55		1.60	0.15		2.75	1.15		1.63	0.87		1.07	1.11		1.50	

454

455 Table 4: Standard Error (SE) for the Reference model 2000-2020 trend for five ground-based station locations

(Arosa/Davos, OHP, Boulder, MLO and Lauder). Results are provided for trend analyses of the Cohesive satellite (COH),
 Dobson Umkehr (UMK) and ozonesonde (SND) records and for Umkehr. The layers are selected to represent the best
 quality of data. Values of SE shown are the actual errors in DU.

459 We will use the standard error of the trend fit to the data to evaluate the improvements in the model fit after additional

460 proxies are included. We use the standard error as a metric instead of standard deviation to reduce dependence on the





461 number of points in the trends model. The Table 4 provides the Standard Errors for the Reference Model fit and 462 represents uncertainty of the trend in DU of the mean ozone in each layer at the station. The standard errors of the 463 trend detected in three co-located ozone records at each station (or in the nearby location as in case of Arosa/Davos 464 or MLO comparisons) do not significantly differ, although in general ozonesonde errors are slightly larger than 465 Umkehr errors most likely due to the larger sampling errors in ozonesonde monthly mean record. Also, the errors in 466 trends detected in COH layers 5-8 are on average smaller than for Umkehr trends (with the exception of layer 7 at 467 Boulder, MLO and Lauder) which could be explained by an overpass method that averages several satellite profiles 468 from adjacent orbits and therefore reduces meteorological scale variability in averaged ozone data.

469 **4.3 Adjusted R2**

470 The adjusted R2 values of the 2000–2020 trends are shown in Fig. 4 and Table 5 for the data fit using the Reference

471 model. The adjusted R2 is a modified version of R2 that adjusts for the number of predictors in a regression model

472 and represents the 'goodness' of the model fit to the data. For COH adjusted R2 is shown for both the overpass and

473 the zonal datasets.

474 Though values are significantly less than the high values usually seen when comparing data that includes the prevalent

475 seasonal variation, the adjusted R2 values for the COH zonal mean record are similar in magnitude and vertical shape

476 to the results of the (60°S-60°N) broadband trend analyses published in Godin-Beekmann (2022), Fig. 7 varying

477 between 0.1 and 0.5. We designate the average values (0.3) as a threshold for satisfactory fit indicating conformance

with prior LOTUS results. We indicate in bold in Table 5 adjusted R2 values of 0.3 or greater to note achievement ofthat threshold.

The adjusted R2 for the Reference model fit is slightly better for the zonal mean COH data than for the COH overpass over the Northern middle latitude stations. This is expected as much of the variability of the time series is reduced in the zonal average as compared to the station overpass data as shown in Fig. 2, and more easily explained by the typically used predictors. Indeed, the goal of this study is to determine if the additional predictors help to explain the

484 additional variation as measured at point locations.

The model fit to the GB data is similar to the COH overpass results in the middle stratosphere (layers 5 and 6), but the model explains less ozone variability in the Umkehr records in the upper stratosphere (layers 7 and 8). In the lower stratosphere (layers 3, 4 and 5), the model fit to the ozonesonde and Umkehr records is similar with the exception of Lauder (Umkehr has larger adjusted R2 in layers 4 and 5). The adjusted R2 for COH overpass in layer 5 is similar to Umkehr and sonde with a larger difference at OHP. The adjusted R2 in the lower stratosphere is less than in the middle stratosphere, which points to other processes (e.g., transport) that drive ozone variability. In this paper we investigate

491 improvement to the trend model fit by introducing additional proxies that can improve representation of the

- 492 dynamically-driven ozone variability in the stratosphere.
- 493







494

495 Figure 4: The adjusted R2 is plotted as a function of altitude/pressure for the LOTUS model fit to the Umkehr (green),

496 ozonesonde (blue), COH overpass (orange, solid), and COH zonal-mean (orange, dashed). Results are shown in 5 panels
 497 that represent trend analyses of ozone records over Arosa/Davos (Hohenpeißenberg for sondes), OHP, Boulder, MLO (Hilo

for sondes) and Lauder ground-based stations.

	LOTUS Model Proxy Tests: Adjusted R2 for Reference Model															
Height	Height Umkehr Arosa/Davos					OHP			Boulder			MLO		Lauder		
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.31			0.27			0.29			0.11			0.29	
2-4	8	0.23	0.38		0.14	0.30		0.17	0.37		0.11	0.32		0.17	0.32	
4-8	7	0.25	0.25		0.19	0.31		0.19	0.27		0.26	0.32		0.12	0.24	
8-16	6	0.19	0.19		0.19	0.25		0.28	0.28		0.31	0.41		0.16	0.11	
16-32	5	0.21	0.24	0.13	0.14	0.33	0.24	0.28	0.34	0.16	0.31	0.41	0.25	0.37	0.34	0.26
32-63	4	0.10		0.10	0.16		0.21	0.13		0.09	0.22		0.24	0.34		0.20
63-127	3	0.05		0.05	0.15		0.08	0.09		0.02	0.09		0.14	0.11		0.10

499Table 5: Similar to Table 4, but for the adjusted R2. Values of 0.30 and above are indicated in Bold as a threshold to500indicate a satisfactory fit.

501 4.4 Reference Model P-Values:

The p-values are often used to evaluate statistical significance of predicted results and results labelled "significant" if they remain below a threshold of 0.05. However, Chang et al. (2021) argued as Wasserstein et al. (2019) does that all trends should be reported with their associated p-values and a thorough discussion of the certainty of trend detection as described by the p-values. Therefore, the p-values can be used for understanding the certainty of the trend. Under the IGAC TOAR activity, p-values are scored to define a consistent scale for comparison of the trends between different analyses (see Table 3, Chang et al., 2023).

LOTUS Model Proxy Tests: Reference Model P Value																
Height	Umkehr	A	rosa/Dav	'OS		OHP			Boulder			MLO		Lauder		
(hPa)	Layer	UMK	СОН	SND	UMK	UMK COH SND		UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND





1-2	9		0.00			0.00			0.03			0.10			0.00	
2-4	8	0.00	0.00		0.00	0.00		0.05	0.00		0.02	0.00		0.47	0.00	
4-8	7	0.00	0.00		0.02	0.00		0.12	0.00		0.00	0.00		0.43	0.00	
8-16	6	0.00	0.62		0.84	0.98		0.05	0.66		0.01	0.17		0.85	0.50	
16-32	5	0.17	0.08	0.05	0.87	0.15	0.01	0.58	0.00	0.04	0.56	0.00	0.93	0.61	0.62	0.00
32-63	4	0.55		0.57	0.62		0.06	0.21		0.11	0.61		0.11	0.10		0.03
63-127	3	0.23		1.00	0.17		0.15	0.22		0.25	0.47		0.08	0.03		0.49

508Table 6: Similar to Table 4 but for p-values. Values of less than 0.05 (high certainty of trend detection) are shown in green.509Values between .05 and 0.1 (yellow) medium certainty, between 0.1 and 0.33 (orange) low certainty of trend detection, and510above 0.33 (red) very low certainty or no evidence of trend detection.

511 Table 6 provides p-values for the Reference Model. These are further used as a baseline for comparison to model fits

512 with additional predictors. P-values of the reference model fit suggest a high certainty (p<0.05) in detected trends in

513 the COH data in layers 7, 8 and 9 at almost all stations with the exception of the higher p-value (0.1, medium certainty)

514 found at MLO in layer 9. Also, high certainty in derived trends is reached for COH records in layer 5 at Boulder and 515 MLO.

516 Umkehr trend analyses also show high confidence in trend detection at Arosa/Davos and MLO stations in layers 6, 7

517 and 8, at OHP in layers 6 and 8, and in Boulder in layers 7 and 8. For the ozonesonde data the high confidence (i.e.

low uncertainty) is found for Hohenpeißenberg, OHP, and Boulder trends detected in layer 5, and at Lauder in layers4 and 5.

520 The medium level of the certainty $(0.05 \le p \le 0.10)$ is found in trends detected in layer 5 of COH ozone time series at

521 Arosa/Davos, layer 4 of ozonesonde records at OHP, layer 9 COH and layer 3 of ozonesonde at MLO, and in layer 4

522 of Umkehr at Lauder.

Low certainty in detected trends at p-value of 0.10 (not inclusive) to 0.33 is found in Umkehr layer 3 and 5 at Arosa/Davos; in COH layer 5, Umkehr and ozonesonde layer 3 at OHP; in Umkehr layers 3, 4 and 7, and ozonesonde layers 3 and 4 at Boulder; and in ozonesonde layer 4 and COH layer 6 record at MLO.

Highest (lowest certainty) p-values (>0.33) were found in layer 6 of COH overpass records at most stations (except for MLO where p-values are medium high). We note that the COH trends are close to zero and the uncertainty envelope crosses the zero line. Therefore, these results point to the trend model's inability to detect non-zero trends and account for all ozone variability in this layer. Similarly, near-zero Umkehr trends with relatively large SE in layer 6 at OHP and Lauder, layer 5 at all (except Arosa/Davos) stations, and in layer 3 and 4 at MLO show the same level of high p-

531 values thus suggesting that additional proxies should be added in the trend model to assess the impacts of the natural

532 variability and instrumental noise on trend uncertainty.

533 It is also important to note that the reference trend model fit to ozone in Umkehr layers 7 and 8 at Lauder has high p-

values, which is related to the near-zero trends that shows large disagreement with COH trend. This difference could

be caused by remaining instrumental step changes that were not fully removed during the record homogenization

536 (Petropavlovskikh et al., 2022).





538 While near-zero trends and high p-values are found in the fit of the Hilo ozonesonde record in layer 5, the p-values in 539 layers 3 and 4 show only medium p-values for near zero trends. It is possible that infrequent launches of ozonesonde 540 observations at Hilo could create the temporal sampling bias and appear noisy. The ozonesonde record at 541 Hohenpeißenberg has sufficiently frequent sampling (3 times per week) for successful trend analyses (Chang et al., 542 2020; Chang, 2023 preprint), but the p-values remain high in layers 3-4. The p-values for Umkehr fit at Arosa/Davos 543 are in the medium to high range for layers 3,4,5, but somewhat smaller which could be due to non-zero trends in layers 544 3 and 5. The p-value difference could be also related to the different location of the ozonesonde (HOH) and Umkehr 545 (Arosa/Davos) observations, thus the records could contain different atmospheric variability that might impact the 546 model fit.

547 We will discuss changes to the p-values in the next section after we add more proxies to the trend model in an attempt

548 to improve confidence in trend detection.

549 5 Trends with the Extended Model - testing the addition of single predictors

550 The LOTUS styled Reference Model is developed and optimized for zonal average datasets. Modeling and trend analysis for GB and satellite overpass data may improve by the addition of other proxies not used in the reference 551 552 model to improve capturing processes that impact ozone changes over limited geographical regions. The Extended 553 Model tests the addition of single predictors to see if fit statistics can be improved for GB and overpass datasets. We 554 judge success of the Extended Model by examination of the reduction in the Standard Error of the trend term, and by evaluation of the impact on the adjusted R2 of the model fit. Table 7 displays the change in the Standard Error of the 555 556 post 2000 trend for each proxy tested determined as SE_{ref} - SE_{ext} as a percent of SE_{ref}. As such positive values 557 correspond to the desired reduction of SE, and are highlighted in the table in green. Low impact changes in the SE 558 are highlighted in yellow, and increases in SE (negative values) are highlighted in orange, or red. It may seem unusual 559 for the addition of proxies to increase the SE (negative values in the table) which indicates less confidence in the fit. But these SE are the uncertainty in the trend term, not in the overall model fit. The new proxies considered each have 560 561 a possible trend and associated error budget for that trend. Whether the additional proxy increases trend uncertainty 562 can depend on how well the trend of the new proxy can be characterized. The adjusted R2 is a better indicator of the overall model improvement. Table 8 displays the adjusted R2 for the Extended Model for each proxy tested. Values 563

of 0.30 and above are indicated in bold as a threshold to indicate a satisfactory fit.

	a) LOTUS Model Proxy Tests: Adding Tropopause Pressure (% difference in Std. Error of Model)															
Height	Umkehr	mkehr Arosa/Davos				OHP		Boulder				MLO		Lauder		
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.33			0.11			0.49			1.39			3.01	
2-4	8	-0.71	-0.51		-0.09	-0.44		-0.19	0.38		-0.58	-0.27		1.26	2.62	
4-8	7	-0.29	0.00		0.26	1.30		0.25	-0.19		2.61	0.33	ĺ	3.71	1.36	ĺ
8-16	6	-1.07	-0.73	ĺ	0.00	0.34	ĺ	0.72	-0.23	ĺ	0.55	0.82	ĺ	3.11	5.39	ĺ
16-32	5	-0.15	2.14	-0.93	1.13	5.34	2.28	-0.37	0.59	0.60	4.54	9.31	2.72	0.00	0.74	2.44





32-6	3	4	6.60	6.07	5.87	9.81	3.35	7.46	7.02	6.05	8.03	9.44
63-12	7	3	12.80	10.17	12.80	10.91	6.81	6.01	5.77	4.55	9.76	7.94

		b)	LOTU	S Mode	Proxy T	ests: Ad	lding Eq	uivalent	Latitude	(% diffe	erence in	Std. Eri	or of M	odel)		
Height	Umkehr	A	rosa/Dav	05		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		8.37			2.85			1.94			-7.18			2.85	
2-4	8	-0.47	0.68		0.09	1.17	ĺ	-0.39	1.53	ĺ	-3.48	-5.42	ĺ	0.98	3.14	
4-8	7	3.75	3.19		2.08	0.56	ĺ	5.41	4.08	ĺ	-2.61	-3.90	ĺ	0.53	1.21	
8-16	6	6.11	8.28		2.54	10.88	ĺ	2.39	7.75	ĺ	5.29	7.76	ĺ	3.44	7.72	
16-32	5	7.93	10.55	5.87	1.92	13.35	7.97	-1.85	0.00	2.84	0.25	0.73	0.65	0.82	3.91	-0.77
32-63	4	-1.44		-1.80	3.20		0.39	-0.22		-0.10	0.33		1.03	-0.24		0.44
63-127	3	1.29		2.02	-1.43		-3.26	-0.79		-0.79	9.57		2.32	1.36		1.88

			c) I	LOTUS	Model Pı	oxy Test	ts: Addin	g QBO	CD (% d	ifference	in Std. 1	Error of	Model)			
Height	Umkehr	A	rosa/Dav	os		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		-3.70			1.54			-1.30			-3.70			-4.91	
2-4	8	0.24	5.09		5.65	13.78		0.00	5.74		-4.26	-1.90		0.00	-2.27	
4-8	7	-2.74	-1.34		2.20	0.37		-2.21	-2.33		-2.26	-3.25		-0.18	-0.60	
8-16	6	-3.51	-4.51		-3.07	-3.57		-0.72	0.94		-0.91	0.20		-2.13	-3.95	
16-32	5	-2.59	-1.34	-0.40	-3.84	-1.78	1.14	-1.48	-0.59	-1.94	14.23	9.67	12.55	-1.09	0.74	1.67
32-63	4	-1.44		0.28	-2.22		-1.18	0.00		0.10	9.81		7.38	-0.84		-2.56
63-127	3	-3.04		-1.78	-1.09		-1.00	-0.17		-0.79	-0.58		2.41	0.00		-3.63

			d) I	LOTUS	Model Pr	oxy Test	ts: Addin	ng AO/A.	AO (% d	lifference	e in Std.	Error of	Model)			
Height	Umkehr	Α	rosa/Dav	'OS		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		1.20			-1.64			0.32			-1.85			-0.48	
2-4	8	-0.83	0.00		-3.77	-1.17		-0.78	-0.38		-2.13	-2.44		0.84	-1.92	
4-8	7	-0.72	1.68		-4.15	-2.60		3.19	4.66		1.22	-3.41		1.24	-1.21	
8-16	6	-0.15	-0.58	ĺ	-2.41	-3.91	ĺ	1.20	0.47	ĺ	1.64	-1.63	ĺ	-0.33	2.51	
16-32	5	-1.22	-0.40	-1.20	0.45	-2.08	-2.28	0.19	-0.59	-2.09	3.93	1.82	0.65	-1.64	-1.49	1.41
32-63	4	5.84		7.78	0.36		5.47	-1.23		-1.71	7.58		1.74	-0.72		2.47
63-127	3	13.12		12.86	5.45		8.00	-1.40		-3.22	4.38		1.34	-1.08		2.42

			e)	LOTUS	S Model 1	Proxy Te	ests: Add	ling NAC) (% diff	erence i	ı Std. Er	ror of M	odel)			
Height	Umkehr	Aı	osa/Dav	05		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND





1-2	9		0.54			-2.52			-0.16			-3.70			-1.27	
2-4	8	-0.24	0.00		-3.11	-1.91		-0.39	-1.91		-1.74	-3.79		-1.96	-1.75	
4-8	7	-0.58	0.67		-1.95	-2.04		0.00	3.88		2.61	-1.14		-2.83	-2.41	
8-16	6	0.15	-0.87		-1.74	-3.40		-2.15	-2.82		2.37	-0.41		-1.97	-0.54	
16-32	5	-0.46	-1.20	-1.07	0.68	-2.23	-3.98	-0.37	-1.38	-1.20	-1.35	-0.73	-2.98	-2.46	-4.28	-1.54
32-63	4	2.58		3.13	-0.62		-0.39	-0.22		0.40	1.45		-0.82	-2.64		-4.77
63-127	3	10.60		6.74	2.65		1.67	0.44		-2.73	1.73		-0.45	-2.26		-4.91

569

		f) LO	TUS Moo	iel Proxy	Tests: A	Adding E	ddy Hea	t Flux (?	% differe	nce in S	td. Error	of Mod	el)		
Height	Umkehr	A	rosa/Dav	os		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		4.89			4.61			4.38			-3.24			0.16	
2-4	8	-1.42	4.58		2.64	6.01		3.12	8.80		-1.55	-2.98		0.70	1.92	
4-8	7	-2.74	-3.36		-0.39	-3.90		-2.95	-2.33		5.04	-4.39		1.77	4.52	
8-16	6	-3.21	-3.20		-2.54	-4.76		-2.39	-3.52		-1.09	0.41		-0.16	1.08	
16-32	5	-3.35	-2.80	-3.20	-2.15	-3.71	-2.28	-2.59	-2.37	-2.09	9.33	-0.36	4.14	0.68	2.42	0.90
32-63	4	-1.91		-1.61	-2.04		-1.97	-2.79		-2.82	8.70		2.97	1.92		2.21
63-127	3	1.49		1.35	-0.88		-1.79	-2.53		-2.61	0.92		0.80	2.08		1.68

570 571

 Table 7: Change in Standard Error (SE) of the post-2000 trend estimate, in percent of SE of Reference Model for adding single predictors. Panel a: Tropopause Pressure; b: Equivalent Latitude; c: QBO terms C and D; d: AO/AAO; e: NAO; f:

				a) LO	TUS Mo	odel Prox	y Tests:	Adding	Tropopa	use Pres	sure (Ad	justed R	2 of Mo	lel)		
Height	Umkehr	A	rosa/Dav	os		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.31			0.27			0.29			0.11			0.31	
2-4	8	0.23	0.38		0.15	0.30		0.17	0.38		0.10	0.31		0.18	0.34	
4-8	7	0.24	0.24		0.19	0.32		0.19	0.27		0.28	0.32		0.14	0.24	
8-16	6	0.19	0.19		0.19	0.25		0.29	0.28		0.32	0.42		0.19	0.15	
16-32	5	0.21	0.26	0.13	0.15	0.39	0.26	0.27	0.35	0.16	0.34	0.47	0.28	0.36	0.35	0.30
32-63	4	0.21		0.22	0.29		0.32	0.19		0.18	0.29		0.29	0.42		0.31
63-127	3	0.24		0.23	0.42		0.21	0.22		0.11	0.14		0.18	0.25		0.21

				b) LO	DTUS M	odel Pro	xy Tests	: Adding	Equival	ent Latit	ude (Adj	justed R2	2 of Mod	el)		
Height	Umkehr	A	rosa/Dav	05		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.43			0.37			0.36			0.15			0.32	
2-4	8	0.23	0.39		0.14	0.31		0.17	0.39		0.10	0.30		0.18	0.34	
4-8	7	0.35	0.34		0.31	0.41		0.27	0.33		0.29	0.36		0.17	0.27	
8-16	6	0.31	0.35		0.33	0.45		0.33	0.40		0.40	0.51		0.25	0.23	
16-32	5	0.34	0.39	0.26	0.25	0.51	0.33	0.31	0.40	0.18	0.31	0.42	0.26	0.42	0.41	0.29





32-63	4	0.11	0.09	0.19	0.21	0.12	0.08	0.22	0.25	0.34	0.21
63-127	3	0.08	0.07	0.16	0.08	0.12	0.02	0.18	0.19	0.14	0.12

				c)	LOT	US Mode	el Proxy	Tests: A	dding Ql	BO CD (Adjusted	R2 of N	lodel)			
Height	Umkehr	A	rosa/Dav	'OS		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.31			0.30			0.31			0.10			0.28	
2-4	8	0.25	0.44	ĺ	0.18	0.43		0.19	0.44		0.09	0.32		0.19	0.33	
4-8	7	0.24	0.26		0.22	0.34		0.20	0.28		0.25	0.31		0.13	0.25	
8-16	6	0.19	0.18		0.18	0.24		0.30	0.32		0.32	0.43		0.17	0.10	
16-32	5	0.22	0.24	0.15	0.13	0.34	0.27	0.29	0.36	0.16	0.40	0.48	0.35	0.38	0.37	0.31
32-63	4	0.12		0.13	0.17		0.22	0.14		0.10	0.29		0.32	0.35		0.20
63-127	3	0.05		0.07	0.17		0.09	0.11		0.02	0.10		0.18	0.13		0.10

				d)	LOT	US Mode	el Proxy '	Tests: A	dding A()/AAO (Adjusted	R2 of N	lodel)			
Height	Umkehr	A	rosa/Dav	05		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.33			0.26			0.32			0.11			0.30	
2-4	8	0.23	0.39		0.13	0.30		0.18	0.38		0.11	0.31		0.18	0.31	
4-8	7	0.24	0.26		0.20	0.31		0.23	0.33		0.29	0.32		0.14	0.23	
8-16	6	0.20	0.19		0.18	0.24		0.31	0.30		0.34	0.42		0.19	0.16	
16-32	5	0.22	0.24	0.13	0.15	0.33	0.24	0.29	0.34	0.15	0.34	0.44	0.27	0.37	0.34	0.28
32-63	4	0.17		0.19	0.18		0.28	0.13		0.09	0.30		0.28	0.33		0.22
63-127	3	0.18		0.19	0.24		0.18	0.09		0.02	0.15		0.18	0.11		0.14

					e) LO	TUS Mo	del Prox	y Tests:	Adding 1	NAO (Ao	djusted I	R2 of Mo	del)			
Height	Umkehr	Α	rosa/Dav	'OS		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.33			0.26			0.31			0.11			0.30	
2-4	8	0.24	0.39		0.13	0.30		0.19	0.37		0.12	0.31		0.17	0.32	
4-8	7	0.25	0.26		0.18	0.31		0.20	0.32		0.30	0.34		0.11	0.23	
8-16	6	0.21	0.20		0.18	0.24		0.28	0.27		0.35	0.44		0.16	0.12	
16-32	5	0.22	0.23	0.14	0.16	0.33	0.23	0.28	0.34	0.17	0.31	0.42	0.24	0.36	0.33	0.28
32-63	4	0.14		0.14	0.17		0.24	0.15		0.13	0.25		0.25	0.33		0.19
63-127	3	0.15		0.13	0.20		0.14	0.12		0.03	0.12		0.16	0.10		0.09

				f)	LOTUS	Model Pi	roxy Tes	ts: Addiı	ıg Eddy	Heat Flu	ıx (Adjus	sted R2 o	of Model))		
Height	Umkehr	Arosa/Davos				OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK COH SND		UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	





1-2	9		0.39			0.34			0.35			0.11			0.29	
2-4	8	0.25	0.44	ĺ	0.17	0.38	ĺ	0.23	0.46	ĺ	0.12	0.31		0.18	0.33	
4-8	7	0.24	0.24		0.21	0.31		0.20	0.28		0.30	0.31		0.13	0.28	
8-16	6	0.19	0.19		0.18	0.24		0.28	0.28		0.31	0.42		0.16	0.13	
16-32	5	0.21	0.24	0.14	0.15	0.34	0.24	0.28	0.34	0.17	0.36	0.42	0.29	0.37	0.35	0.27
32-63	4	0.11		0.11	0.17		0.20	0.13		0.09	0.28		0.28	0.35		0.21
63-127	3	0.07		0.09	0.18		0.07	0.09		0.01	0.11		0.17	0.12		0.11

Table 8: Adjusted R2 after adding single predictors. Panel a: Tropopause Pressure; b: Equivalent Latitude; c: QBO terms
 C and D; d: AO/AAO; e: NAO; f: Eddy Heat Flux.

580 5.1 Tropopause pressure (TP)

581 Adding the TP proxy to the standard LOTUS model produces the most consistent results between different techniques 582 (COH, Umkehr and ozonesonde) and also have similar magnitude of standard error changes among different latitudes 583 (i.e. Arosa/Davos, OHP, Boulder, MLO, Lauder). The most significant impact in improving the SE is found in the 584 lower stratosphere (layers 3, 4) and in the middle stratosphere (layer 5) at the MLO tropical station. The impact of the 585 TP proxy on the COH trend uncertainty in the model stratosphere (layer 5) is somewhat larger, likely due to the 586 satellite AK extending into the lower stratosphere. Similarly, larger reduction of the standard error in the Umkehr 587 trends in the lowermost stratosphere (layer 3) in comparison to the AK-smoothed ozonesonde record could be due to 588 sampling biases in the ozonesonde record. Adding the TP proxy to the Reference Model improves the adjusted R2 in 589 layers 3-5, whereas the SE improvements are also consistent across geo-locations and measurement techniques. 590 Several improvements resulted in adjusted R2 to exceed the 0.3 threshold (Umkehr at OHP in layer 3, sonde at OHP 591 in layer 4, and sonde at Lauder in layers 3 and 4) and in many cases the adjusted R2 increased by more than 0.02.

592 **5.2 Equivalent Latitude (EqLat)**

In the mid-latitudes, the addition of EqLat as a predictor shows consistent results across measurement techniques and stations with few exceptions. The reduction in the SE of the model fit is evident in the COH data in the upper stratosphere (above 4 hPa or ~ 40 km), but is less pronounced in Umkehr profiles. The impact on MLO SE of the trend fit in the upper stratosphere is negative (in both COH and Umkehr records) which can be explained by the fact that the EqLat is much closer to geometric latitude near the equator than at the middle/high latitudes and therefore its use as a proxy would not provide any additional information in interpretation of the tropical upper stratospheric ozone variability. It could also suggest that the addition of EqLat will overfit the record.

The ozone record trend fits in the middle stratosphere (32–4 hPa or 25–40 km) benefit from adding the EqLat proxy at most locations. Improvement in the SE of the trends in the lower stratosphere (127–63 hPa or ~15–20 km) is minimal, limited to some locations and instrumental records (Arosa/Davos Umkehr and HOH sonde, MLO Umkehr and sonde, and Lauder Umkehr and ozonesonde), which could be related to the location of subtropical jet that modulates mixing of tropical and subtropical (and occasionally polar) air masses and influences the strat/trop exchange. Unexpectedly, the addition of the EqLat proxy to the MLR statistical model for trend detection in Boulder Umkehr and ozonesonde low stratospheric ozone records increases the uncertainties of the fit, while the influence of





- 607 subtropical jet on Boulder lower stratosphere is well known (Manney et al, 2018). Perhaps, the data analyses also
- 608 need to consider the tropopause variability.
- In terms of the impact on the adjusted R2, the EqLat proxy significantly improves model fit for multiple instruments,
- 610 mostly in layers 5–7, and in COH fit in layer 9. The adjusted R2 improvements also often exceeded 0.3 threshold. No
- 611 significant improvement is found in the ozonesonde model fit in layer 5 with the exception of the OHP record (0.09
- 612 increase).

613 5.3 Extra QBO terms C and D

614 QBO is an important driver of ozone variability at tropical stations. Based on the results of adding 2 extra terms of the 615 QBO to the standard model, the recommendation could be to exercise this option only for the tropical station trends. 616 At the Northern middle latitudes (i.e in Arosa/Davos, OHP and, to a lesser degree, in Boulder) an improvement to the 617 trend SE uncertainties in layer 8 is noted. There seems to be a similar pattern for the upper stratosphere in trends 618 derived with Heat Flux. Tweedy et al. (2017) show that the first two EOFs of the QBO did not describe the anomalous 619 QBO behavior, while Anstey et al. (2021) show that the addition of two more EOFs of the QBO could capture the 620 effect of the disruptions on the zonal winds. Therefore, including additional QBO EOFS could benefit attribution of 621 ozone variability in the middle stratosphere (layers 4 and 5) in the tropical latitudes (reduced errors in MLO/Hilo 622 trends) and in the upper stratosphere (layer 8 in Umkehr and COH trends) in the NH middle latitude stations 623 (Arosa/Davos, OHP, Boulder) related to the global circulation pattern that are also represented by the Heat Flux proxy. 624 A slight reduction in the errors at SH middle latitude (COH and sonde at Lauder, New Zealand) could be invoked by 625 the EqLat variability that has a small correlation with the QBO-D proxy. Reduction of SE in the trend fit of the layer 626 5 ozonesonde record at OHP (up to 2 %) is not found in the Umkehr or COH results, which suggests overfitting and 627 sampling bias (see results in Appendix D). 628 The addition of extra QBO terms improves the adjusted R2 model fit for all COH station overpass records in layer 8

- The addition of extra QBO terms improves the adjusted K2 model in for an COH station overpass records in layer s
- and occasionally improves Umkehr adjusted R2 (except at MLO). The most significant improvement is found at MLO
 in layers 3–5 in all three instrument records.

631 5.4 Arctic and Antarctic Oscillations (AO/AAO)

632 AO/AAO proxies reduce SE (green colored cells) in the lower stratosphere (layers 3 and 4) at Arosa/Davos, OHP, and MLO, although the reduction somewhat differs between the Umkehr and ozonesonde records. At the same time, at 633 634 Boulder and Lauder the SE does not show an improvement after the addition of the AO/AAO proxy (AAO is used 635 instead of AO at Lauder). In the middle stratosphere (layer 7), a reduction in SE is found over Boulder in both COH 636 and Umkehr records. The addition of AO/AAO proxies improves the SE of the trend at MLO and Lauder but only in 637 Umkehr records, while it worsens the COH SE. At Lauder, the COH SE in layer 6 shows an improvement, but not in 638 Umkehr record. Since results in the middle stratosphere (layers 5-7) are not always consistent among different techniques (reductions are not in the same layers) it could indicate statistical model overfit into the record's noise, or 639 640 vertical smoothing of the Umkehr or COH technique that combines ozone variability in the layer with a portion of ozone variability in the adjacent layers, thus partially or completely reducing the correlation with the proxy. 641





The addition of the AO predictor increases the adjusted R2 in the lower stratosphere at Arosa/Davos, OHP and MLO.
Also, a small enhancement of the adjusted R2 is seen in the middle and upper stratosphere, including in Umkehr layers
643 6 and 7 and COH layers 6, 7 and 9 over Boulder, as well as in Umkehr fit in layers 5–7 at MLO, and at Lauder (AAO)
645 for Umkehr and COH records in layer 6. These results are not very consistent across different geolocations, but seem
646 to be consistent across instrumental records at some stations (Umkehr and ozonesonde in the lower layers, and COH
647 and Umkehr in the upper layers).

648 5.5 North Atlantic Oscillation (NAO)

649 Including the NAO proxy in the trend model appears to have a similar pattern (i.e., in latitude and altitude) of changes 650 in the standard error as compared to the result of inclusion of the AO/AAO proxy. It is not a surprise, since indices of 651 the NAO and AO are highly correlated in time due to their common link to the downward propagation of stratospheric 652 anomalies. Standard errors are somewhat reduced in the lower stratospheric layers at the middle NH latitude and 653 tropical Umkehr records, but the change is less significant than in AO/AAO cases. The impacts on ozonesonde trend 654 uncertainties are very minimal and inconclusive at Boulder (layers 3 and 5), OHP (layer 4) and MLO (layer 3) records. 655 The impacts on Lauder are similar or stronger (SE is increased for both Umkehr and sonde records) to the impacts of 656 the AO/AAO. In the middle and upper stratosphere the standard errors are typically reduced. The exception is found 657 in layer 7 of the COH record at Boulder and Arosa/Davos, and in layers 6 and 7 of the Umkehr record at MLO. Similar negative results are found when AO/AAO proxies are added, which suggests that the observed time series are 658 659 overfitted and potentially some instrumental or sampling anomalies are misinterpreted with addition of these proxies.

660 **5.6 Eddy Heat Flux (EHF)**

The EHF represents a dynamical proxy for assessment of the impact of the Brewer Dobson Circulation (BDC). It is expected to have an impact on the upper stratospheric ozone by accelerating the transport in the upper branch that brings more ozone at higher latitudes (i.e. Arosa/Davos) and middle latitudes (i.e. OHP, Boulder, and Lauder). It could possibly represent changes in the lower branch of the BDC circulation and the expansion of the tropical band, thus modulating ozone in the lower stratosphere at tropics (i.e. MLO). In the Southern middle latitudes (i.e. Lauder), the correlations could be related to the shift in the subtropical wave activities to the higher latitudes in response to the ozone hole healing.

The addition of the EHF predictor leads to the reduced SE uncertainties in the upper stratosphere in COH and Umkehr trends at OHP and Boulder, and in COH only trends at Arosa/Davos. It has a much smaller reduction of SE for the Lauder trend and even an increase in uncertainties if used to fit upper stratospheric ozone time series at MLO. At the same time, the SE in the Umkehr and ozonesonde middle stratosphere (layers 4–5) is substantially reduced, including smaller improvements at Lauder. In the lower stratospheric (layer 3) ozone trend SE in Umkehr and sonde records at MLO, Lauder and Arosa/Davos are somewhat reduced when using the EHF proxy.

675 records in layers 8 and 9 in all but MLO records, which indicates impact of the BDC upper branch on the middle





- high measurement noise in the station records. There is, however, a small increase in adjusted R2 in the Umkehr
- 678 record in layer 7 at MLO (whereas COH does not show a change).
- The increase in adjusted R2 is found at MLO in Umkehr and sonde layers 4 and 5, including a small increase in layer
- 680 3, which probably is related to the EHF-driven changes in the middle stratosphere . Ozone variability in Umkehr and
- sonde records at MLO appears to contain information about the circulation changes in the shallow BDC branch.

682 6 The Full Model - adding multiple predictors

683 In this paper we seek to develop an improved model and thus trend estimates for point located measurements of ozone 684 through modifications of a model optimized for zonal data. Our criteria for model improvement are based on reduction of the SE of the trend with either improvement (at best) or moderate impact (at worst) on the model adjusted R2. From 685 the results of the previous section, we see several opportunities to improve the model and improve confidence in the 686 trend estimates. This section examines if the gains of the above are improved while adding several predictors together. 687 688 As stated above the TP as a predictor exhibits the most consistent results for all stations and measurement techniques. 689 The other predictors have successes in SE reduction, but only at some layers, and some stations. Some results are 690 instrument dependent. Based on the tests above we expect combining predictors can improve the model fit and trend SE reduction, but it is 691 692 clear that the predictor selection should vary by station and level. Appendix E details the choices made for the Full

693 Model which combines 1 to 3 additional proxies beyond the Reference Model.

694 6.1 Predictors added for the Full Model

695 Reduction of the SE of the trend while improving (or at least not impacting) model adjusted R2 is the basis of predictor

696 choice for the Full Model. To qualify a predictor should exhibit consistent results for all measurement techniques.

697 Improvement at multiple stations is preferred to single station improvements. In general, we avoid combining highly

698 correlated predictors. Table 9 shows final choices for the Full Model.

		LOTUS F	ull Model predictor	r selection	
	Arosa/Davos	OHP	Boulder	MLO	Lauder
Layer					
9	EqLat	EqLat	EqLat	Reference only	EqLat
8	EqLat	EqLat	EqLat	Reference only	EqLat
7	EqLat	EqLat	EqLat	Reference only	EqLat
6	EqLat	EqLat	EqLat	EqLat	EqLat
5	EqLat	EqLat	EqLat	EqLat, QBO CD, AO	EqLat
4	TP, AO	TP, AO	TP	TP, QBO CD, AO	TP
3	TP, AO	TP, AO	TP	TP, QBO CD, AO	TP





701 Table 9: Added predictors for the Full model are tuned for each layer and each station. For layers 7 to 9 the SE and 702 adjusted R2 parameters at MLO are not improved by additional predictors, and the original LOTUS based Reference 703 Model is used. Appendix E explains the logic of the predictor selection.

704 6.2 Impact of the Full Model on trends

- 705 Figure 5a shows the trends for the stations (with COH overpass) for the Reference and Full Models. An impact of the
- 706 Full Model on ozone trends derived in the upper stratosphere (above 16 hPa) is neutral. Addition of proxies to the
- 707 LOTUS model does not change trends which remain the same magnitude as those derived using the Reference Model,
- 708 i.e. positive and statistically significant at the SH and MH middle latitudes and over tropics. The largest difference
- 709 (outside of the SE uncertainty) between upper stratospheric Umkehr and COH trends is found over Boulder, MLO
- 710 and Lauder.



711

712 Figure 5a: Post 2000 trends for the Full and Reference Model. In this figure the COH data shown in orange is the overpass data. Solid lines depict Reference Model values (unchanged from Fig. 3). Dashed lines depict Full Model values for all 3

713 714 instrument types.

> LOTUS Post-2000 Trends Proxy Comparison Arosa/Davos (46.8°N) OHP (43.9°N) Boulder (40.0°N) MLO (19.5°N) Lauder (45.0°S) 50 45 Height [hPa] 10 20 100 15 Ozone trend fit [%/decade] Umkehr Post Trends (Ref) Cohesive Post Trends (Ref) Sonde Post Trends (Ref) Umkehr Post Trends (Full) ____ Cohesive Zonal Post Trends (Ref) ____ Sonde Post Trends (Full)





Figure 5b: Post 2000 trends for the Full and Reference Model. In this case the orange lines are with the zonal data instead of the COH overpass data. Dashed lines depict Full Model values for all 3 instrument types. The Umkehr and sonde trends are unchanged from Fig. 5a.

- 719 In the middle stratosphere, additional proxies do not change trend values across locations and instrumental records
- 720 (outside of the SE). At OHP, Boulder and Lauder Umkehr trends in layer 6 (8–16 hPa) are barely positive while COH
- 721 trends are negative. At Arosa/Davos and MLO, COH trends in layer 6 are barely negative and Umkehr trends are
- significantly positive. Most COH trends in layer 5 (16-32 hPa) are statistically negative (except at Lauder), while
- 723 Umkehr trends are near zero.
- 724 In the lower stratosphere, Umkehr and sonde trends Arosa/Davos and MLO change after the Full model is used.
- 725 However, Umkehr and sonde trend changes at MLO are within the SE and therefore can be deemed not significant.
- 726 Ozonesonde trends at Arosa/Davos in layer 3 (125-63 hPa) change from zero to positive. Umkehr trends at
- 727 Arosa/Davos in layer 3 change from negative to near zero. Large differences between ozonesonde and Umkehr trends
- 728 at Lauder and OHP remain unchanged after the Full model is applied although respective SE envelopes overlap.
- 729 Figure 5b also shows the trends for the Reference and Full Models, but the COH data shown is the associated zonal
- 730 data relevant to each station. Incorporation of the additional proxies does not change the trend values for the zonal
- 731 COH data. Impact on error estimates for the trends are discussed next.

732 6.2 Impact of the Full Model on the Trend SE

733 Table 10 summarizes the reduction in the SE for the Full model. Selection of the EqLat predictor for the Full model 734 in the layers 5-9 and for all stations (except MLO/Hilo, to be discussed later) shows the improvement in the SE (as 735 discussed in the previous section). Also, the TP predictor is selected for inclusion to the Full model for trend analyses 736 at Boulder and Lauder stations in layers 3 and 4. The combination of several predictors are used for individual stations 737 based on the additional reduction in the SE. For the Arosa/Davos and OHP stations we select a combination of the TP 738 and AO to reduce the SE almost twice as much in some layers. Inclusion of AO proxy is in support of the interpretation 739 of seasonal and interannual ozone variability recorded over stations in Europe that are north of 40 degrees latitudes 740 and are exposed to the seasonal events of ozone depleted air masses transported from the Polar region during the spring season (Steinbrecht et al., 2011; Manney et al., 2011; Knudsen and Grooss, 2000; Fioletov and Shepherd, 2003; 741 742 Zhang et al., 2017; Weber et al., 2022a). The strong impact of AO/AAO on the lower stratosphere ozone variability 743 are not detected in Boulder or Lauder and we choose not to include it in the Full model for trend analyses at these 744 stations.

				LOTU	S Model	Proxy T	ſests: (%	6 Differe	ence in S	td. Erro	r of Mo	del)				
Height	Umkehr	A	rosa/Dav	/05		OHP			Boulder	r		MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		8.35			2.74			1.94			0.00			2.85	
2-4	8	-0.47	0.68		0.09	1.03		-0.39	1.53		0.00	0.00		0.98	3.14	
4-8	7	3.75	3.04		2.08	1.86	ĺ	5.41	4.08	Ì	0.00	0.00	ĺ	0.53	1.21	
8-16	6	6.11	8.36		2.54	10.88	ĺ	2.39	7.75	Ì	0.55	0.82	ĺ	3.44	7.72	
16-32	5	7.93	10.72	5.87	1.92	13.33	7.97	-1.85	0.00	2.84	19.39	13.32	15.27	0.82	3.91	-0.77





32-63	4	8.71	9.96	6.13	9.92	3.35	7.46	20.51	9.64	8.03	9.44
63-127	3	20.30	18.49	13.48	14.01	6.81	6.01	6.00	4.73	9.76	7.94

745	Table 10: Change in post 2	2000 trend SE in t	the Full	Model as a %	differe	ence of the Ref	erence Model.	Color codi	ing is the
746	same as introduced in Table	e 7.							0

747 The MLO/Hilo location is close to the Tropical belt and therefore has different processes impacting stratospheric 748 ozone variability as discussed in the previous section. We find that EqLat proxy can be added to the Full model in 749 layer 6 and 5 (similar to other stations); however, above layer 6, EqLat or TP is not useful for interpretation of tropical ozone variability and therefore we believe the trend model in these layers should remain as it currently is used in 750 751 Godin-Beekmann et al. (2022) analyses. The EqLat and TP are mildly correlated (-0.4) in the stratosphere, and 752 therefore we decided against combining both of these proxies in the Full model. However, we also found that adding 753 AO and QBO C/D proxies in layers 3, 4 and 5 improved the model fit and reduced the SE. These combined additional 754 proxies are not correlated and reduce SE more than when using them separately. 755 The Full Model showed impacts on the SE in the upper stratosphere (above 8 hPa). The trend errors were reduced

756 with the exception of Umkehr trends at 4–2 hPa over Boulder and Arosa/Davos where errors did not change. No

changes in SE are found at MLO with additional proxies, thus the Full Model is kept the same as the Reference Model

758 for this station in the upper stratosphere.

Similarly, in the middle stratosphere SE were mostly reduced after the Full Model was applied (except for slightly
 larger SE in trends derived from ozonesonde at OHP and from Umkehr at Boulder).

761 After applying the Full Model in the lower stratosphere, we still found high uncertainty due to higher ozone variability

762 (natural variability), but SE were reduced. Arosa/Davos and MLO Umkehr and sonde trends changed after Full Model

763 was used. Change in ozonesonde trends at HOH in layer 3 (125-63 hPa) goes from zero to positive and trend detection

764 becomes highly confident (p-value <0.05). Umkehr trends at Arosa/Davos in layer 3 changed from negative to near

765 zero but results have low certainty (p-value >0.1). Larger trend differences remain between ozonesonde and Umkehr

- 766 at Lauder and OHP after the Full Model is applied.
- 767

	LOTUS Model Proxy Tests: (% difference of SE of Trend): overpass and zonal COH														
									_						
Height	Umkehr	Arosa/I	Davos	ОН	Р	Bould	ler	ML	0	Lauc	ler				
(hPa)	Layer	Overpass	Zonal												
1-2	9	7.61	2.89	2.20	1.30	1.61	1.34	NA	NA	3.17	1.97				
2-4	8	0.00	0.90	1.47	1.26	0.00	0.63	NA	NA	1.75	2.76				
4-8	7	3.39	0.47	1.85	2.55	5.77	1.53	NA	NA	0.00	1.11				
8-16	6	7.35	2.75	10.17	8.98	9.30	4.30	0.00	1.79	8.93	5.34				
16-32	5	10.67	1.74	11.76	5.54	0.00	2.36	12.73	4.81	3.70	-1.11				

768 769

Table 11: Change in Standard Error of Trend, as percent of Reference Model SE, for the COH overpass data and zonal data at the 5 ground stations. MLO Full Model in layers 9-7 is the same as the Reference Model (change is marked as NA).







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Figure 6: Change in Standard Error of Trend, as percent of Reference Model SE, for the COH overpass data (blue) and
 COH zonal data (red) at the 5 ground stations.

It is instructive to ponder if the addition of proxies that yield improvements via reduction of the standard error in the localized GB or overpass measurements also have the potential to improve uncertainties in the zonal data. To explore this Table 11 and Fig. 6 show the percent change in SE of the trend when adding the proxies for the Full model. Values are shown for both the COH overpass and the COH zonal data. In general, except when the improvement in the SE for the overpass COH is small (3% or less), addition of proxies has much less impact on the zonal results than on overpass results. This suggests that indeed the Reference LOTUS model is well tuned for zonal datasets, but can be improved with select addition of proxies for overpass or localized GB data.

781 6.3 Impact of the Full Model on adjusted R2

Table 12 shows the adjusted R2 for the Full Model. In the upper stratosphere, the Full Model increases the adjusted

- 783 R2 above 8 hPa (except in Umkehr at 4–2 hPa). Over MLO there is no change because the Full Model is kept the
- same as the Reference Model for layers 7, 8 and 9.

					LOTU	S Model	Proxy	Fests: (A	djusted	of Mod	el)					
Height	Umkehr	Aı	·osa/Dav	/05		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.42			0.37			0.36			0.11			0.32	
2-4	8	0.23	0.39		0.14	0.31		0.17	0.39		0.11	0.32		0.18	0.34	
4-8	7	0.35	0.35		0.31	0.41		0.27	0.33		0.26	0.32		0.17	0.27	
8-16	6	0.31	0.35		0.33	0.45		0.33	0.40		0.40	0.51		0.25	0.23	
16-32	5	0.34	0.38	0.26	0.25	0.51	0.33	0.31	0.40	0.18	0.44	0.53	0.39	0.42	0.41	0.29
32-63	4	0.23		0.25	0.29		0.34	0.19		0.18	0.42		0.38	0.42		0.31
63-127	3	0.31		0.31	0.44		0.26	0.22		0.11	0.19		0.24	0.25		0.21

785 Table 12: Adjusted R2 of the Full Model. Values of 0.30 and above are indicated in Bold as a threshold to indicate a 786 satisfactory fit. Compare to Table 4 containing values for the Reference Model.



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- In the middle stratosphere (32-8 hPa) adjusted R2 increases are found in all records (although smaller increases are found in ozonesonde and Umkehr records at OHP, Boulder and Lauder at 32-64 hPa). At Arosa/Davos, Boulder and Lauder the adjusted R2 in the COH and Umkehr trend models increase and continue to be very close in value. The COH adjusted R2 is larger at OHP and MLO than in Umkehr and sonde records thus suggesting that overpass conditions might have smoothed some natural variability observed in the GB records. In general, the adjusted R2 is the largest at the 32-64 hPa level. This suggests that the Full Model shows an improvement for regional trend analyses
- 793 in the middle stratosphere.
- 794 Although Umkehr and sonde trend changes at MLO in the low stratosphere are within the SE and therefore can be
- 795 deemed not significant, the adjusted R2 is increased which suggests a better model fit in the Full Model. The adjusted
- R2 increases in both Umkehr and ozonesonde data, while the largest increases are found in the Arosa/Davos, OHP 796 797 and MLO records.
- 798 In the lower stratosphere, the adjusted R2 remains low in both Umkehr and sonde records at Boulder (only TP is added 799 for the Full model). While the p-values at 63-32 hPa are significantly reduced (see discussion in the next section), 800 they still remain relatively high. These results suggest that additional research is needed to identify the best set of proxies for Boulder records in the lower stratosphere. At Lauder, the ozonesonde record shows smaller adjusted R2 801 802 as compared to Umkehr partially due to low sampling biases.
- It is valuable to further explore the impact of the Full Model on the adjusted R2 for the zonal and overpass COH data. 803
- 804 Fig. 7a shows the adjusted R2 for the Reference and Full Models at each of the 5 stations using the COH overpass 805 data. In all cases the Full Model improves the adjusted R2 except for MLO layers 7, 8 and 9 where the Full and 806 Reference Model are identical. The most significant improvements are seen by Umkehr at layers 3 to 7, COH overpass 807 at Layers 5, 6 and 7, and sonde layers 3-5. Figure 7b shows similar results using COH zonal data instead of overpass. 808 There is practically no further improvement in the adjusted R2 for the zonally averaged COH results (except for a 809
- small increase for MLO layer 5). Comparison of results reveals that for OHP the implementation of the Full model
- for the COH overpass data (Fig. 7a, dashed line) improves the adjusted R2 to values nearing that of the Reference 810 811 Model zonal data in layer 7 and below (Fig. 7b, solid line). For MLO and Lauder the use of the Full Model on the
- COH overpass data improves the adjusted R2 over the Reference Model beyond the improvement seen in the COH 812
- 813 zonal results for layers 5 and 6. At Arosa/Davos and Boulder the implementation of the Full Model does not fully
- 814 reach the magnitude of the COH zonal adjusted R2.







Figure 7a: Adjusted R2 for the Full Model (dashed lines) and Reference Model (solid lines) at 5 stations. The COH data in
this figure is the overpass data at each station.



818

Figure 7b: Adjusted R2 for the Full Model (dashed lines) and Reference Model (solid lines) at 5 stations. The COH data in
 this figure is the zonal data for each station. The Umkehr and sonde lines are identical to those in Fig. 7a.

821 6.3 Examination of the p-values of the Full Model

822 In the upper stratosphere (above 8 hPa), the confidence in Umkehr trends remained high (see Table 13) for most

823 stations except at Boulder (medium to low) and Lauder (very low, although some improvement was found). COH

trends confidence was very slightly degraded over Boulder at 1–2 hPa, but mostly has not changed.

825 In the middle stratosphere (between 32 and 8 hPa), p-values were significantly reduced in COH records. At 8–16 hPa

remained high, but at 16-32 hPa the confidence improved (continued) to high over Arosa/Davos and OHP (Boulder

827 and MLO). In case of Umkehr analyses in layer 8-16 hPa at Arosa/Davos, Boulder and MLO the confidence remained

828 high. However, at 16-32 hPa the Umkehr trend detection confidence was degraded over Arosa/Davos and Lauder.

829 For the ozonesonde record, the p-values remained low (<0.05) except at MLO where some improvement was found

830 after the Full Model was used, but the p-value remained high. It suggests that some instrumental records have either





- 831 high atmospheric or instrumental noise and therefore perhaps high certainty in trend detection cannot be achieved with
- 832 linear trend models. For near zero trends with high variability, the p-values are not a good criterion for trend
- 833 detectability.
- 834 In the lower stratosphere (between 125 and 32 hPa), analyses of p-values for the Full Model fit show significant
- 835 improvement for Umkehr trends at MLO between 63-32 hPa (while the p-value was increased at other stations at this
- 836 level). In addition, improvement in p-values was found for ozonesonde trends at all stations. Specifically, very low p-
- values for the Full model were reached at Arosa/Davos (125-63 hPa), OHP (125-63 and 63-32 hPa), MLO (125-63
- 838 and 63–32 hPa), and Lauder (63–31 hPa).

					LOT	US Mod	el Proxy	Tests: (P Value	of Mod	el)					
Height	Umkehr	Aı	osa/Dav	/05		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.00			0.00			0.06			0.10			0.00	
2-4	8	0.00	0.00		0.00	0.00		0.06	0.00		0.02	0.00		0.41	0.00	
4-8	7	0.00	0.01		0.02	0.00		0.13	0.00		0.00	0.00		0.34	0.00	
8-16	6	0.01	0.17		0.81	0.47		0.02	0.83		0.00	0.21		0.92	0.71	
16-32	5	0.74	0.00	0.00	0.86	0.03	0.02	0.50	0.00	0.05	0.51	0.00	0.57	0.74	0.73	0.00
32-63	4	0.56		0.34	0.67		0.01	0.27		0.15	0.09		0.02	0.08		0.01
63-127	3	0.66		0.04	0.32		0.01	0.31		0.35	0.63		0.02	0.02		0.30

Table 13: P Value of the Full Model. High certainty of trend detection is seen for values below .05 (green). Values between .05 and 0.1 (yellow) medium certainty, between 0.1 and 0.33 (orange) low certainty of trend detection, and above 0.33 (red)

841 very low certainty or no evidence of trend detection.

842 7 Summary of the Full Model findings.

843 We find that upper stratospheric trends in COH overpass and Umkehr records detect ozone recovery with high

confidence (p<0.05) above 8 hPa (with the exception of near-zero positive Umkehr trends over Lauder and Boulder).

- 845 We note the largest difference between Umkehr and COH trends (outside of the SE uncertainty) at Boulder, Mauna
- Loa and Lauder.

847 Confidence for the middle stratosphere (between 32 and 8 hPa) trends vary between high, medium and low. Although

848 most of the trends are narrowly different from zero (especially when error bars are considered), there are some

849 differences in results across instrumental groups: trends in COH and sonde (except at OHP) between 32 and 16 hPa

tend to be small negative, while Umkehr trends are slightly positive. Some trends are statistically different from zero.

851 However, instrument-specific error bars often overlap and thus making differences in trends not significant.

852 Confidence in lower stratosphere trends is highly variable and even lower than in the middle stratosphere due to higher

ozone variability unaccounted for by Solar, QBO and ENSO proxies used in the Reference Model. However, high

- confidence (p<0.05) is still found in ozonesonde trends at Arosa/Davos, OHP, MLO and Lauder (although not at all
- 855 layers). Umkehr trends in the lower stratosphere show lower confidence than ozonesonde trends (except at Lauder
- 856 and Arosa/Davos in the lowermost altitudes). The low confidence levels could be related to the near-zero trends
- 857 derived from Umkehr data, whereas ozonesonde trends are often different from zero lines. Also, we apply AK-



858



859	ozonesonde trends (see Appendix A) and did not find any significant impacts. Most notably, ozonesonde and Umkehr
860	trends significantly disagree in the lower stratosphere at OHP and Lauder and therefore require further investigation.
861	The instrumental drifts and differences in Lauder trends are also discussed in Bjorkland et al. (2023 preprint) and are
862	consistent with our findings.
863	8 Conclusions
864	This paper is a follow up to Godin-Beekmann et al. (2022) with a focus on the GB record trend assessment. Therefore,
865	our trend analyses focus on the questions:
866	1) Do proxies for evaluating trends of GB stations need to be different from those of the optimized set for zonal
867	data?
868	2) Are station records representative of the small geophysical region or semi-global changes?
869	3) Do uncertainties of the zonal averaged trends improve with additional proxies?
870	
871	The Full Model developed in this paper for station and overpass data adds proxies to the LOTUS models of Godin-
872	Beckmann (2022). Our trend analysis of stratospheric ozone records from the Umkehr, ozonesonde and COH station
873	overpass data at 5 geographical regions using the Full Model (LOTUS v 0.8.0) show similar trends to those published

smoothing to the sondes to account for the wide AKs in the Umkehr retrieval. We tested the impacts of the AK on

overpass data at 5 geographical regions using the Full Model (LOTUS v 0.8.0) show similar trends to those published in Godin-Beekmann et al. (2022) paper. We analyze trends for instrumental records converted to 7 Umkehr layers that represent ozone changes in the upper, middle and lower stratosphere over NH and SH middle latitudes and over high tropics of the NH. We also analyze GB station records at Arosa/Davos, Hohenpeißenberg and OHP separately in contrast to the "European regional" trend analyses presented in Godin-Beekmann et al. (2022) and included COH overpass records for comparisons with the GB records. Our analyses include evaluation of the adjusted R2 (aka

- goodness of the model fit), standard error and p-values.
- 880 We also investigate differences between satellite trends as detected in the records sampled for individual geographical
- 881 locations (spatial and temporal overpass criteria) versus zonal average datasets. We find that COH overpass ozone 882 records capture ozone variability of the ground-based station records (Umkehr and sonde) better than COH zonal data.
- 883 We do not find that the COH zonal record is improved by using EqLat instead of geometric latitude to construct the
- dataset (see Appendix C), but EqLat can be an important additional proxy at some levels for GB data. To determine
- the improvement to the model fit we use the Standard Error and adjusted R2 for the Full and Reference model fit.
- 886 Using the Reference model for the zonal mean COH data we find slightly better adjusted R2 than for the COH overpass
- 887 data fit over the Northern middle latitude stations. This is expected as much of the variability of the overpass time
- series is reduced in the zonal average data. Therefore, we also explore the impact of additional predictors in the trend
- 889 model fit applied to the more variable GB and satellite COH overpass data to determine if that will reduce the SE and
- 890 improve the adjusted R2. We also apply the Full model to the zonally averaged data to assess the benefits of additional
- 891 proxies to further reduce trend uncertainties.
- 892 We find that adding predictors (with few exceptions) does not change the trends but often reduces SEs and increases
- 893 the adjusted R2 (with the exception of the upper stratospheric ozone trends at MLO). We also find that the p-values





894 are useful for interpretation of improvements of the model fit in the data, although improvements in the SE do not 895 always result in improved confidence in derived trends, especially when the trends are close to zero. In these cases we 896 conclude that either longer records are needed to discern trend information outside of the atmospheric noise or further 897 research into the inconsistencies between instrumental records and homogenization procedures is required. We also 898 find the small changes in trends in the lower stratosphere and improvements in the model fit after additional proxies 899 are used. However, the sampling tests indicate that trends can depend on the temporal selection of the records when 900 AK are used to smooth ozonesonde high resolution profiles (see discussion in Appendix D). 901 This paper concludes that additional proxies bring improvements to trend detectability in case of GB and gridded 902 satellite data analyses and better agreement is achieved between satellite overpass and GB trends. We also find that 903 zonally averaged and gridded satellite records produce comparable trends over the studied middle latitudes and 904 subtropical regions. Therefore, the GB trends are representative of the stratospheric ozone changes over the semi-905 global area. Finally, zonally averaged data do not benefit from addition of proxies beyond what LOTUS model uses

for global trend detection whereas the uncertainties in GB and gridded trends are significantly reduced and sometimes
 (Boulder, MLO, Lauder) become comparable to the uncertainties of the zonally averaged trends in the upper and

- middle stratosphere. Based on analyses presented in this paper we strongly recommend using additional proxies for
- trend analyses of GB and gridded satellite stratospheric ozone records. Additional proxies should be selected based
- 910 on the latitude and altitude of the observational ozone record to adequately represent stratospheric transport and mixing
- 911 processes impacting interannual and seasonal ozone variability.

912 Appendices

913 Appendix A: AK Smoothing for ozonesondes

914 Ozonesonde profiles have high vertical resolution (purple line in Fig. A1) in comparison to the Umkehr (green solid 915 line) or COH (orange dashed line) ozone profiles. Each Umkehr layer is referenced to the atmospheric pressure at the bottom of the layer, which is constructed using half of the pressure in the layer below. Averaging Kernels (AK) as 916 917 shown in Fig. A1, panel b, define the granularity of the Umkehr vertical grid. In order to compare trends from three 918 instrumental records in the same vertical system, we convert the ozonesonde and COH profiles to the Umkehr layers 919 and DU. The COH overpass data is in units of DU, but on different layers than the defined Umkehr layers, so only vertical grid modification is required. The sonde profiles (purple thin line) are in units of partial pressure and are first 920 921 converted to DU, then converted to the Umkehr grid (blue solid line in panel a). Conversion to the Umkehr grid can 922 be done either by interpolation, or by AK smoothing. The equation describing the process of applying AK smoothing 923 is

$$Ozone_{smoothed}(i) = \sum_{j} \left\{ AK_{i,j} * Ozone_{true}(j) - Ozone_{apriori}(j)
ight\} + Ozone_{apriori}(i)$$

924

where AK is the Averaging Kernel for layer i, Ozone*smoothed* is the smoothed ozone result, Ozone*true* is the ozonesonde profile, and Ozone*apriori* is the Umkehr a priori (climatological) profile. The AK for each Umkehr layer





927 is used as a weighting function applied to the ozonesonde profile (Ozone*true*) prior to the integration which simulates

928 the Umkehr optimal estimation method used for estimating the ozone content in the targeted layer (Rodgers, 2000).







Figure A1: a) An example of ozone observations over the Boulder, CO station. The purple line is 100-m averaged ozone partial pressure (hPa) vertical profile measured by sonde on 13 November, 2018. The green line with solid circles is the ozone profile derived from Dobson Umkehr observations on the same day. The blue line with blue dots is the ozonesonde profile converted to the Umkehr layers and smoothed with the Umkehr AK. The orange dashed line with open squares is the COH ozone profile observed over Boulder on the same day and interpolated to the Umkehr layer vertical grid. b) The Umkehr AK for the ozone profile derived from observations in Boulder on 13 November, 2018. Each line represents the smoothing function for one of 10 Umkehr layers (see color legend).

938 Although the ozonesonde measurement typically reaches altitudes between 32 and 10 hPa, the balloon often bursts 939 before reaching the top of layer 6 (16 hPa), therefore only partially covering the ozone content in that layer. We also 940 note that Umkehr AKs are relatively wide and therefore will incorporate (weight in) ozone variability from the layer 941 above and layer below of the targeted Umkehr layer. (See layer 6, green line in Fig. A1, panel b.) Therefore, there 942 are two sources of error in ozonesonde comparisons with Umkehr ozone in layer 6: a) burst level for ozonesonde does 943 not reach the top of the layer 6, thus the integrated ozone is smaller than expected. b) the Umkehr AK for layer 6 is 944 relatively wide and therefore the Umkehr layer partially contains information from above the burst altitude of the 945 ozonesonde, thus making smoothed ozonesonde concentration lower than expected. In order to avoid these errors, we 946 only show ozonesonde results up to layer 5. 947 Similarly, we explored smoothing COH profiles with Umkehr AKs. Figure A2 demonstrates the time series of the 948 COH ozone over the Mauna Loa station. The trend model was fitted to the COH record with and without AK applied. 949 The reference trend model included proxies and trends. To focus on ozone variability that contributes to the trends we

- 950 subtracted the modeled ozone variability from the COH data and then added the trend component back. The COH
- 951 record residuals in Fig. A2 are shown in Umkehr layers where COH is either smoothed with AK (red lines) or not
- 952 (green lines). We notice that the AK-smoothing of the COH profile in layer 9 does not have a lot of independent





953 information from layer 8. In this example it clearly shows that the trends in layer 8 are embedded in the COH layer 9 954 ozone time series, which was confirmed when we compared trends derived from the AK-smoothed COH in layers 8 955 and 9. In case of the integrated COH ozone record, the trends in layers 8 and 9 differed. In order to avoid biasing the 956 COH trends at layer 9 we decided to not apply Umkehr AKs for COH smoothing and only use COH profiles 957 interpolated into the Umkehr layers. This result makes sense since COH overpass data are derived from UV 958 backscatter radiances also using an Optimal Estimation technique. COH overpass data has a comparable vertical 959 resolution to Umkehr, simply with different layer definitions. Interpolation makes the most sense for rendering COH 950 deta in the Umkehr sention.

960 data in the Umkehr vertical coordinate system.

COH Tir	me Se	eries a	t Mauna	Loa.	Proxv	Tested:	AKs
001111			c maama	Lou,	110/19	icocca.	,



961

Figure A2: Modified residuals (seasonal cycle, Solar, QBO, and ENSO are removed, but trend is retained) of COH overpass
 data at Mauna Loa (20N, 156W). Red: AK smoothed to Umkehr layers; Green: Interpolated to Umkehr layers. Vertical
 lines show the dates of satellite records in COH. The largest impact of the AK is seen between 1997 and 2001 where two
 curves separate in layers 7, 8 and 9, and also after 2001 in layer 9.

966 Figure A3 demonstrates time series of monthly mean ground-based records the lower stratosphere at 5 stations. The

967 Umkehr data (blue) are compared with the ozonesonde anomalies either interpolated to the Umkehr layer 3 (green),

968 or ozonesonde profiles matched with Umkehr profiles in time and smoothed using the Umkehr averaging kernels

969 (crimson). All three datasets have been deseasonalized using their respective climatological (using 1998-2008

- 970 climatology) average monthly mean ozone. The application of the Averaging Kernels has the effect of smoothing the
- 971 temporal variability.







972

Figure A3: Time series of monthly averaged and de-seasonalized (in %) ozone anomalies of Umkehr (green) and
ozonesondes records are compared at 5 ground-based stations. Ozonesonde data are either calculated using only profiles
that are interpolated in Umkehr layer 3 (blue) or matched with Umkehr profile in time and smoothed with the Umkehr
averaging kernels (crimson).

977 Appendix B: COH using OMPS v3r2 vs OMPS v4r1

978 OMPS SNPP v4r1 uses updated SDRs as input which incorporate unified and consistent calibration algorithms

979 removing artificial jumps caused by operational changes, instrument anomalies, or contamination for anomaly views

980 of the environment or spacecraft. Also included are new interpolated band-passes, and updated soft calibration based

981 on the new input SDR's.

982 Differences between the v3r2 and v4r1 versions of the resulting COH dataset are typically less than 1 percent (Fig.

983 A4 and A5). Small seasonal variation is apparent at all levels. Larger differences are visible in 2020 when the soft

984 calibration for v3r2 is extended beyond its period of relevance. Figure A6 shows the drift between the two versions.

985 Drift between the datasets is less than +- 1% at all levels. This is a reasonable estimate of the resulting expected trend

986 difference in using the newest COH version as compared to the v3r2 results used in Godin-Beekmann (2022).







987 988

Figure A4: Differences in the COH monthly average zonal product as generated from SNPP v4r1 and v3r2 processing.
Also shown is the annual cycle in this difference as depicted by the average over all years for each month. Exhibited at 4045N is a less than 2% difference with an annual cycle. A somewhat different pattern is seen in 2020 where the soft calibration for v3r2 is extended beyond its period of relevance.









997 the drift between the two versions.







998



1000 Appendix C: Impact of using equivalent latitude in generation of the COH product

The COH overpass data used in this paper collects all profiles during the day within a latitude and longitude box of +/- 2 degrees by +/- 20 degrees, then generates a 1/distance averaged value for the station. The box is based on geometric latitude and longitude. With 15 orbits per day, the chosen box size guarantees 2 to 4 possible profiles within the box depending on whether the orbit overpasses or straddles the site as shown in Fig. A7. Also shown is a scenario when the equivalent latitude (EqLat) near the site is particularly non-zonal. In such cases the profiles selected using a geometric coordinate box will select SBUV profiles from an Eq Lat that is different from that of the measurement station.







1010Figure A7: Shows orbits of SNPP and positions of OMPS NP ozone profiles on January 20, 2012 and January 5, 2012. The1011second row displays a possible EqLat contour overlaid.

1012 It is informative to create an overpass product using boxes based on EqLat and determine the impact on the data. 1013 Since EqLat is layer dependent, the included profiles must be selected independently for each layer. Figure A8 shows

1014 COH overpass data for Boulder using geometric coordinates, EqLat based coordinates, and the associated Umkehr

1015 data. Color coding shows the EqLat at Boulder for each measurement day with dark blue and yellow indicating days

- 1016 with extreme variation from 40N.
- 1017

1009







1020Figure A8: COH overpass data generated with geometric coordinates, EqLat based coordinates, and the associated Umkehr1021dataset at Boulder for 2012. Data points are color coded for the EqLat at the measurement site. Boulder is at 40 N.

Variation in EqLat is most apparent in Winter months and transitional Fall and Spring, less so in Summer. Yet the value of the COH ozone is not dramatically altered in the time series. Figure A9 shows correlation plots of the COH overpass to Umkehr for the data at layer 7 (4-8 hPa). The pattern of the scatter and the value of the correlation coefficient are not substantially altered for overpass determination using geometric latitude (left) and EqLat (right). Figure A10 shows the vertical distribution of the Correlation coefficient and the RMS Difference for the two COH datasets vs Umkehr. These two metrics are minimally impacted for this sample year in the layers where COH is valid.



1029





1030Figure A9: Correlation between Umkehr and COH overpass using Geometric Latitude (left) and EqLat (right) to select1031included profiles for layer 7 (4-8 hPa).

1032



1033

1034Figure A10: Profiles of Correlation coefficients and RMS differences between COH overpass data at Boulder for 2012 using1035Geometric Latitude (blue) and EqLat (red) to select data points included in the average.

1036

1037 The use of geometric latitude appears to be sufficient in the choice of included data points in the overpass COH product 1038 at the layers used in this paper. Likely this is a ramification of the smooth horizontal resolution of the satellite product.

1039 Appendix D: Temporal Sampling and Impact on Trends

1040 This paper compares trends for three instrument types each with differing measurement frequency. From each set of 1041 constructed. measurements а monthly average is See the data files at 1042 https://gml.noaa.gov/aftp/ozwv/Publications/2023 Umkehr Ozone Trends Paper/ for the data and the number of 1043 data points in each monthly average with the sampling variations. Umkehr measures once or twice per day depending 1044 on cloud interference with the measurement. At Arosa/Davos and Lauder, Umkehr measurements are sparser than 1045 the other GB stations, often less than 10 per month. At Boulder beginning in 1983 measurements number 20 or more 1046 per month. At OHP the Umkehr record begins in 1983 with a strong 20 or more measurements per month. From 1047 1999 to 2016, however, measurements per month are often less than 15 per month. The most Umkehr measurements 1048 at MLO are the most abundant, especially after 1985 measuring multiple times in a day, resulting in 50-70 data points 1049 contributing to the monthly average. The COH overpass dataset is typically available once per day at each station with 1050 occasional misses, contributing usually 27-30 data points per month. Since Umkehr can measure multiple times per 1051 day, the COH data matched to Umkehr can contain more profiles in the monthly average than the original full COH 1052 data, since the COH overpass data will appear twice in the monthly average, once per each Umkehr measurement.





This occurs often at MLO. Ozonesonde launches are typically one to three times per week depending on the station. At Arosa/Davos, sonde measurements are typically about 15 per month. Sonde measurements at the other stations usually have approximately 5 measurements per month, with some periods of up to 10 per month. As with COH overpass measurements, the sonde dataset matched to Umkehr can have more contributions to the monthly average resulting from dates with more than one Umkehr measurement, resulting in multiple sonde matches.

1058 The trend results in this paper use all available Umkehr data to generate the monthly means. The COH and sonde data 1059 are matched to Umkehr to use the Umkehr temporal sampling for COH, and to be able to use the Umkehr averaging 1060 kernels for sonde. It is important to determine how the temporal sampling within the monthly mean data may impact 1061 trend results. To aid this understanding, we create three subsets of Umkehr data each with different temporal sampling 1062 and create the corresponding monthly mean: 1) all observations in Umkehr record; 2) Umkehr matched to the COH dataset; and 3) Umkehr matched to the sonde dataset. In this way we use the same data, but only vary the temporal 1063 sampling. Since the COH is measured every day, except in the rare case that the satellite data is missing due to 1064 1065 instrument issues, sampling 1 and 2 should provide nearly identical results. We expect a strong change in the monthly 1066 mean and resulting trends for Umkehr record when it is matched with infrequent sampling of ozonesonde profiles 1067 (especially in Boulder, Hilo and Lauder).

1068 Figure A11 summarizes the results. Each line in Fig. A11 is trend derived from Umkehr data, but with sampling of all 1069 data, data matched to COH dates, and data matched to sonde dates. In general, the differences are within the envelope of trend uncertainty (+/- 2 std errors). As expected, the trends and standard errors for all (green) and COH-matched 1070 1071 subsampled (orange) Umkehr records are nearly the same. The largest differences in all Umkehr and COH matched 1072 Umkehr lines are apparent at OHP. We have determined that this arises from occasional months when there is a short 1073 satellite outage coupled with sparse Umkehr observations at the station. However, trends derived from sonde-matched 1074 Umkehr data (blue) show deviations from other observations. This is especially clear at Arosa/Davos in the upper 1075 stratosphere (~2-3 % above 10 hPa). But since this is above the measurement capability of the ozonesonde, this will 1076 not impact the ozonesonde trend results at Arosa/Davos. At Lauder the most significant differences are seen in layer 1077 3 (2.5%), but unfortunately not in the direction to explain sonde differences in the Lauder trend curves as compared to 1078 Umkehr. Smaller differences are seen at other layers (very small, less than 1 %, differences in layers 6 and 4). At

1079 OHP small differences of less than 1 % are seen between 50 and 10 hPa, well within error estimates.









1081Figure A11: Trend results for the Reference Model using Umkehr data mimicking the temporal sampling of COH and1082sonde. Green is all available Umkehr data; orange is Umkehr data matched to COH measurements dates; blue is Umkehr1083data matched to sonde measurement dates.

Figure A12 further explores sampling differences by examination of trends of COH data using the full COH dataset, and data sampled to the Umkehr dates in generation of the monthly mean datasets. As with Fig A11, the trend lines are nearly identical at all stations except OHP. At OHP in the early 2000's there are significantly fewer COH points matched to Umkehr because of the drop in Umkehr measurements. This likely impacts the post-2000 trend estimate. The differences remain below 2%, and are within the error estimate of the trends. In summary, the sampling biases between COH overpass and Umkehr data cannot explain the difference in the derived trends (see Fig. 3, most notable

1090 in layers 7 and 8 at Boulder and Lauder).

1091









Figure A13 explores the impact on trends from sampling differences of the sonde data. Shown are trends with all sonde data, and trends with Umkehr matched data. In this figure only, the sonde data is not AK smoothed since the Umkehr AK are only available on dates when there is an Umkehr measurement. So shown here are trends from sonde data integrated to the Umkehr levels. As with Fig. A11 the only visible impact is seen at OHP and Lauder, though both are within error estimates. At Lauder the trends remain negative for both samplings, but sonde sampled to Umkehr moves closer to the zero line. At OHP the sonde trends are positive, but sonde sampled to Umkehr moves slightly closer to zero. The sampling impact on trends for both OHP and Lauder are likely due to the reduced number

1102 of Umkehr data at these sites.



1103

1104Figure A13: Trend results for Reference model exploring sampline of the sonde data. Solid blue is all sonde data; dashed1105is Umkehr matched sonde data.

1106 Figure A14 explores the impact of sampling on the adjusted R2 using the COH overpass data. Shown are the adjusted

1107 R2 for all available COH overpass data, and the same using only COH overpass with matches to the Umkehr data.

1108 For Arosa/Davos, OHP and Lauder the differences are small. For Boulder and MLO at some layers (Boulder, layers

1109 6,7; MLO layers 6,9), the impact is more apparent with the Full COH exhibiting higher adjusted R2 at these stations.







1110

1111Figure A14: Adjusted R2 for the Reference Model exploring variations in sampling of the COH data. Solid orange is COH1112data matching Umkehr sampling; dotted orange is all available COH data.

1113 Appendix E: Decision process for the Full Model

1114 The LOTUS styled Reference Model is developed and optimized for zonal average datasets. The Extended Model 1115 tests the addition of single predictors to see if fit statistics can be improved for GB and overpass datasets. For 1116 Tropospheric Pressure (TP), improvements are consistent among layers and among instrument types. The addition of 1117 EqLat also yields consistent results for instrument types and at most stations, though not Mauna Loa. Addition of other predictors gives mixed results depending on level and station. The potential for improving confidence in trend 1118 1119 results exists by combining predictors using different choices depending on layer and station. We choose additional 1120 predictor combinations with consideration of three criteria: 1) combined predictors should not have a high correlation 1121 with each other (usually .2 or less); 2) predictors should reduce the SE of the trend consistently for all instrument 1122 types; 3) addition of the predictor should not greatly reduce the adjusted R2 of the model fit, but preferentially increase 1123 it. As seen in Tables 7e and 7f the NAO and the EHF predictors do not make a significant improvement when added 1124 to the Reference Model, so we do not include either in the Full Model.

1125 Mixed Model:

1126 We have noted a high correlation between the TP and EqLat predictors at all levels especially for Boulder, Mauna 1127 Loa and Lauder with correlation adjusted R2 of .4 to .7, and somewhat less correlated at Arosa/Davos and OHP with 1128 adjusted R2 of .2 and .3. Subsequently, we choose to not use these two predictors together (at the same station/layer 1129 combination). The addition of TP at all stations for layers 3 and 4 uniformly decreases the standard errors at all 1130 stations for both Umkehr and sonde. The addition of EqLat (with the exception of Umkehr at Boulder, level 5) almost uniformly decreases the standard errors at all stations for layers 5 and 6. There is additional reduction in the SE for 1131 1132 layers 7 to 9 for all stations except at Mauna Loa. Thus, we choose TP and EqLat as additional predictors at these 1133 layers. QBO C and D, have significant impact in decreasing the SE in layers 4 and 5 for both Umkehr and sonde, and 1134 layer 3 for sonde with only a small degradation for Umkehr. QBO-CD shows an improvement in layer 8 at OHP, both





1135 COH and Umkehr, and Arosa/Davos and Boulder for COH only. We have tested adding both QBO and EqLat for 1136 layer 8 at these 3 stations. For Umkehr measurements, there is no improvement beyond EqLat only with QBO-CD 1137 also included. For COH there is additional improvement, but not to the extent of QBO-CD alone. Since the 1138 improvement is limited to one layer, and for only COH, we choose to only add the additional QBO-CD for the tropical 1139 MLO. Table A1 shows the resulting combination of additional predictors for this Mixed Model.

1140

	LOTUS Mixed Model								
Layer	Arosa/Davos	ОНР	Boulder	MLO	Lauder				
9	EqLat	EqLat	EqLat	Ref	EqLat				
8	EqLat	EqLat	EqLat	Ref	EqLat				
7	EqLat	EqLat	EqLat	Ref	EqLat				
6	EqLat	EqLat	EqLat	EqLat	EqLat				
5	EqLat	EqLat	EqLat	EqLat, QBO CD	EqLat				
4	ТР	ТР	ТР	TP, QBO CD	TP				
3	ТР	TP	ТР	TP, QBO CD	TP				

1141

1142 Table A1: Details of additional predictor combinations for each level and station in the Mixed Model

1143 The resulting change in SE from the Reference Model is shown in Table A2. For most stations/layers this is simply a 1144 composite of the values from the single EqLat or TP Extended Model results. There remain a few 1145 instrument/station/layers where the SE is slightly increased - OHP sonde Layer 5, Arosa/Davos Umkehr layer 8 and 1146 Boulder Umkehr layer 8, but these are negligible. At Boulder Layer 5 Umkehr the increase in SE is somewhat more 1147 at 1.85% difference, but this is still small enough to not be of great concern. For Mauna Loa at layers 3,4 and 5 the 1148 model is rerun adding two predictors together and the results are new. Indeed in these cases the SE is improved 1149 beyond the single predictor results of either QBO alone, or TP or EqLat alone with the exception of Sonde layer 5 1150 where the change in SE is just slightly degraded from QBO alone (13.42% vs 13.69% reduction in SE).

			I	LOTUS	Model	Proxy T	ests: (%	b Differ	ence in	Std. Eri	ror of M	lodel)				
Height	Umkehr	Arosa/Davos		OHP			Boulder			MLO			Lauder			
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		8.35			2.74			1.94			0.00			2.85	
2-4	8	-0.47	0.68		0.09	1.03		-0.39	1.53		0.00	0.00		0.98	3.14	
4-8	7	3.75	3.04		2.08	1.86		5.41	4.08		0.00	0.00		0.53	1.21	
8-16	6	6.11	8.36		2.54	10.88		2.39	7.75		0.55	0.82		3.44	7.72	
16-32	5	7.93	10.72	5.87	1.92	13.33	-0.56	-1.85	0.00	0.90	15.21	11.13	13.42	0.82	3.91	0.00
32-63	4	6.60		6.07	5.87		7.04	3.35		2.97	13.15		9.70	8.03		4.13





63-127	3	12.80		10.17	12.80		6.91	6.81		2.46	3.81		4.19	9.76		4.41
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1151 Table A2: Change in the SE of the trend using the Mixed Model.

- 1152 Table A3 shows the associated adjusted R2 for the proposed Mixed Model. Similarly to the change in SE the adjusted
- R2 is a composite of the individual EqLat or TP results from the extended model with the exception of the results for layers 3,4, and 5 at Mauna Loa where both predictors are included concurrently. At these layers the adjusted R2 in
- some cases matches the higher Adj R2 values of the two predictors, and in others improves with the combination of
- some cases matches the ingret Auj K2 values of the two predictors, and in others improves with the combination of
- 1156 QBO and TP or EqLat.
- 1157

				I	OTUS	Model I	Proxy T	ests: (A	ljusted	R2 of M	lodel)					
Height	Umkehr	Ar	osa/Dav	/05		OHP			Boulder			MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		0.42			0.37			0.36			0.11			0.32	
2-4	8	0.23	0.39		0.14	0.31		0.17	0.39		0.11	0.32		0.18	0.34	
4-8	7	0.35	0.35		0.31	0.41		0.27	0.33		0.26	0.32		0.17	0.27	
8-16	6	0.31	0.35		0.33	0.45		0.33	0.40		0.40	0.51		0.25	0.23	
16-32	5	0.34	0.38	0.26	0.25	0.51	0.21	0.31	0.40	0.19	0.40	0.35	0.37	0.42	0.41	0.24
32-63	4	0.21		0.22	0.29		0.32	0.19		0.13	0.34		0.35	0.42		0.25
63-127	3	0.24		0.23	0.42		0.26	0.22		0.13	0.14		0.21	0.25		0.19

1158 Table A3: Adjusted R2 for the Mixed Model

1159 Augmented Mixed Model

- 1160 It is hard to ignore the substantial reduction of SE when adding the AO/AAO predictor especially for layers 3,4 and 5
- at Mauna Loa, and for layers 3 and 4 at Arosa/Davos. The results for OHP layers 3 and 4 are still compelling, though

somewhat less so. So we explore the addition of AO/AAO at these three stations only, for the layers specified. Table

- 1163 A4 summarizes the predictor choices for this Augmented Mixed Model.
- 1164
- 1165

		LOTUS Augn	nented Mixed M	lodel	
	Arosa/Davos	OHP	Boulder	MLO	Lauder
Layer					
9	EqLat	EqLat	EqLat	Ref	EqLat
8	EqLat	EqLat	EqLat	Ref	EqLat
7	EqLat	EqLat	EqLat	Ref	EqLat
6	EqLat	EqLat	EqLat	EqLat	EqLat
5	EqLat	EqLat	EqLat	EqLat, QBO, AO/AAO	EqLat
4	TP, AO/AAO	TP, AO/AAO	ТР	TP, QBO CD, AO/AAO	TP
3	TP, AO/AAO	TP, AO/AAO	ТР	TP, QBO CD, AO/AAO	TP





1166 1167	Table A4: from Table	Table A4: Details of additional predictor choices for each level and station in the Augmented Mixed Mo from Table A1 by adding AO/AAO at some levels for Arosa/Davos, OHP and Mauna Loa.									
				LOTUS	8 Model Proxy Tests: (%	Difference in Std. Erro	or of Model)				
	TT 1 1 (T T			00	OTTR	D 11					

Height	Umkehr	Aı	rosa/Dav	vos		OHP			Boulder	r		MLO			Lauder	
(hPa)	Layer	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND	UMK	СОН	SND
1-2	9		8.35			2.74			1.94			0.00			2.85	
2-4	8	-0.47	0.68		0.09	1.03		-0.39	1.53		0.00	0.00		0.98	3.14	
4-8	7	3.75	3.04		2.08	1.86		5.41	4.08		0.00	0.00		0.53	1.21	
8-16	6	6.11	8.36		2.54	10.88		2.39	7.75		0.55	0.82		3.44	7.72	
16-32	5	7.93	10.72	5.87	1.92	13.33	-0.56	-1.85	0.00	0.90	19.39	13.32	15.70	0.82	3.91	0.00
32-63	4	8.71		9.96	6.13		7.04	3.35		2.97	20.51		10.45	8.03		4.13
63-127	3	20.30		18.49	13.48		5.46	6.81		2.46	6.00		4.85	9.76		4.41

Table A4: Details of additional predictor choices for each level and station in the Augmented Mixed Model . This differs

1168 1169

Table A5: Change in the SE of the trend using the Mixed Model.

1170

1171 Table A5 displays the change in the SE from the Reference Model now for the Augmented Mixed Model. Adding 1172 AO/AAO at Arosa/Davos (layers 3 and 4) and Mauna Loa (layers 3 to 5) greatly reduces the SE beyond that of the 1173 Mixed Model results in Table A2. For OHP (layers 3 and 4) the impact is less dramatic for Umkehr. For sonde 1174 measurements at layer 4 the AO/AAO addition has no impact beyond the Mixed Model; for layer 3 the addition of 1175 AO/AAO results in less reduction of the SE.

1176

				L	OTUS	Model F	roxy T	ests: (A	djusted	R2 of N	lodel)					
Height	Umkehr	Arosa/Davos			ОНР			Boulder			MLO			Lauder		
(hPa)	Layer	UMK	сон	SND	UMK	сон	SND	UMK	сон	SND	UMK	сон	SND	UMK	СОН	SND
1-2	9		0.42			0.37			0.36			0.11			0.32	
2-4	8	0.23	0.39		0.14	0.31		0.17	0.39		0.11	0.32		0.18	0.34	
4-8	7	0.35	0.35		0.31	0.41		0.27	0.33		0.26	0.32		0.17	0.27	
8-16	6	0.31	0.35		0.33	0.45		0.33	0.40		0.40	0.51		0.25	0.23	
16-32	5	0.34	0.38	0.26	0.25	0.51	0.21	0.31	0.40	0.19	0.44	0.53	0.40	0.42	0.41	0.24
32-63	4	0.23		0.25	0.29		0.34	0.19		0.13	0.42		0.39	0.42		0.25
63-127	3	0.31		0.31	0.44		0.26	0.22		0.13	0.19		0.26	0.25		0.19

1177 Table A6: Adjusted R2 for the Augmented Mixed Model

1178 Table A6 displays the Adj R2 for the Augmented Mixed Model. Adding AO/AAO improves the Adj R2 results for

1179 Arosa/Davos and MLO and has little to no impact at OHP. Based on the criteria outlined at the beginning of this

1180 appendix, we assign the Augmented Mixed Model as the 'Full Model' in the body of this paper.

1181

1182 Code/Data availability: All dataset used in this study are publicly available at the website





1183 https://gml.noaa.gov/aftp/ozwv/Publications/2023_Umkehr_Ozone_Trends_Paper/.

1184 **Competing interests:** The authors declare that they have no conflict of interest.

1185 Author contributions: IP and JW conceptualized the paper, and IP led the paper preparation. PE, KA, and JW 1186 performed the data analysis. KM is responsible for the production of the spatial and temporally matched ground-based 1187 and satellite ozone profile data. JW is responsible for producing COH zonally averaged and station overpass ozone 1188 profile records. LF is responsible for the retrieval and calibration of the OMPS data. GM, PE, KM and KA are 1189 responsible for NOAA Umkehr measurements. EMB is responsible for measurements in Arosa/Davos. RQ is 1190 responsible for Umkehr and ozonesonde observations in Lauder, New Zealand. BJ and PC are responsible for 1191 ozonesonde observations in Boulder and Hilo. GA is responsible for the ozonesonde observations in OHP. RVM is 1192 responsible for HEGIFTOM ozonesonde records and data analyses. RD, SGB, DZ provided context of the LOTUS 1193 model use and interpretation of trend analyses. All authors contributed to the writing of the paper.

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