

Trends in stratospheric ozone derived from merged SAGE II and Odin-OSIRIS satellite observations

A.E. Bourassa¹, D.A. Degenstein¹, W.J. Randel², J. M. Zawodny³, E. Kyrölä⁴, C.A. McLinden⁵, C.E. Sioris⁶, C.Z. Roth¹

1. Institute of Space and Atmospheric Studies, University of Saskatchewan, Saskatoon, Canada
2. National Center for Atmospheric Research, Boulder, CO, USA
3. NASA Langley Research Center, Hampton, VA, USA
4. Finnish Meteorological Institute, Earth Observation Unit, Helsinki, Finland
5. Environment Canada, Downsview, Ontario, Canada
6. Department of Earth and Space Science and Engineering, York University, Toronto, Canada

Abstract

Stratospheric ozone profile measurements from the Stratospheric Aerosol and Gas Experiment (SAGE) II satellite instrument (1984–2005) are combined with those from the Optical Spectrograph and InfraRed Imager System (OSIRIS) instrument on the Odin satellite (2001–Present) to quantify interannual variability and decadal trends in stratospheric ozone between 60°S and 60°N. These data are merged into a multi-instrument, long-term stratospheric ozone record (1984–present) by analyzing the measurements during the overlap period of 2002–2005 when both satellite instruments were operational. The variability in the deseasonalized time series is fit using multiple linear regression with predictor basis functions including the quasi-biennial oscillation, El Niño–Southern Oscillation index, solar activity proxy, and the pressure at the tropical tropopause, in addition to two linear trends (one before and one after 1997), from which the decadal trends in ozone are derived. From 1984–1997, there are statistically significant negative trends of 5–10% per decade throughout the stratosphere between approximately 30–50 km. From 1997–present, a statistically significant recovery of 3–8% per decade has taken place throughout most of the stratosphere with the notable exception between 40°S–40°N below approximately 22 km where the negative trend continues. The recovery is not significant between 25–35 km altitude when accounting for a conservative estimate of instrument drift.

Introduction

The variability of stratospheric ozone and the trends observed in past decades continue to be of importance for understanding the future evolution of ozone and its interaction with changing climate. Analysis of various data sets toward this end has been a focus of research for nearly three decades. An improved understanding of chemical depletion processes has led to predictions of complete future recovery of the ozone layer by 2050 at the earliest (WMO, 2011). Several recent studies have highlighted the need for high quality, altitude resolved profile measurements to quantify the different aspects of atmospheric variability and trends throughout the global stratosphere. Satellite measurements are elemental to these analyses (for example see Weatherhead et al., 2000, Randel and Wu, 2007; Randel and Thompson, 2011; Kyrölä et al., 2012). One of the key data sets in many past studies is the ozone profile measurements made by the Stratospheric Aerosol and Gas (SAGE) II satellite instrument, which obtained high quality vertical profiles of the global stratosphere by solar occultation from 1984–2005. In this paper we use the ozone profile measurements made by the Optical Spectrograph and InfraRed Imaging System (OSIRIS) on the Odin satellite, which began in 2002, to extend the SAGE II record to the present day. The OSIRIS ozone profile data set, obtained through measurements of limb scattered sunlight spectra, have similar quality and vertical resolution as the SAGE II measurements, and extend from the upper tropopause to the upper stratosphere. Also, OSIRIS provides retrievals of ozone density on altitude levels, similar to SAGE II, so that conversions from mixing ratio on pressure surfaces is not needed. The four year overlap between the two missions allows for robust comparison and analysis to merge the two measurement sets. The goal of this paper is twofold: to demonstrate the feasibility of merging the SAGE II and OSIRIS ozone measurements into a single time series through comparative analysis of the two time series, and secondly, to use the merged time series to quantify interannual variability and trends in stratospheric ozone from 1984–2013 over the latitude range 60°S to 60°N. The OSIRIS instrument is currently still fully operational and this merged time series will continue to provide long-term monitoring of these trends into the future. This work follows directly from the recent results of Sioris et al., 2013, who analyzed the SAGE II and OSIRIS time series in the tropical lower stratosphere only.

42 **Data Set Descriptions**

43

44 The SAGE II instrument, operational from 1984 to 2005, measured solar transmittance in the ultraviolet,
45 visible and near infrared wavelength range in order to infer profiles of ozone number density and other
46 stratospheric trace gases and aerosol (McCormick et al., 1989). The profiles extend from below 15 km to
47 above 50 km with 1 km resolution, are reported on a regular 0.5 km altitude grid, and have relatively sparse
48 sampling in a latitudinal distribution that varies slowly over the course of weeks to months to cover the globe.
49 The ozone product version used here is version 7.0 (Damadeo et al., 2013), which, compared to previous
50 standard product version 6.2, features decreased retrieval smoothing, updated absorption cross sections
51 (Bogumil et al., 2003), and atmospheric temperature and pressure from Modern-Era Retrospective Analysis
52 for Research and Applications (MERRA, Rienecker et al., 2011). The outlier removal, including filtering for
53 high values of volcanic aerosol, has been updated from Wang et al., 2002. See Damadeo et al, 2013 for
54 details on the SAGE II version 7.0 data product. As noted by Kyrölä et al., 2013, among others, the
55 requirements for stability of the measurements for this type of trend analysis are stringent. High quality for
56 the SAGE II occultation measurements is achieved via a self-calibration through the observation of the direct
57 sun at the beginning or end of the occultation on a sunset or sunrise measurement, respectively.

58

59 The OSIRIS instrument began standard stratospheric limb scattered sunlight measurements in early 2002,
60 shortly after launch on the Odin satellite in 2001 (Llewellyn et al, 2004; Murtagh et al., 2002; McLinden et
61 al., 2012) and remains fully operational at the present time. The spectra, which cover the 280-800 nm
62 wavelength range, are used in combination with a spherical radiative transfer model that accounts for multiple
63 scattering (Bourassa et al., 2008) and a non-linear relaxation inversion to retrieve profiles of ozone number
64 density from 10–60 km altitude with approximately 2 km vertical resolution and successive samples about
65 every 500 km along the polar orbit track (Degenstein et al., 2009). Although the limb scatter technique does
66 not measure the sun directly, the retrieval method provides a similar self-calibration of the measurement
67 through normalization of the spectral radiance by the measurement at the same wavelength at a higher tangent
68 altitude above the retrieval range. This effectively decreases the dependence on the effective scene albedo
69 (von Savigny et al., 2003) and helps to increase the long term stability of the retrieved product in a similar
70 fashion to the self-calibration of the occultation measurement. The polar orbit, which is nominally 1800h
71 local time at the ascending node, provides global coverage during spring and fall, and coverage of the tropics
72 throughout the year. No measurements are obtained during polar winter. In this work, for which we have
73 used the version 5.07 data set, only OSIRIS measurements on the descending orbit track, i.e. near local dawn,
74 are used as the slight procession of the orbit to later local times has resulted in the loss of coverage in the
75 tropics on the ascending node after 2004.

76

77 **Analysis and Results I: Merged Ozone Anomaly Time Series**

78

79 Previous work comparing OSIRIS and SAGE II ozone observations has shown generally excellent agreement
80 between the two data sets during the four year overlap period. Adams et al., 2013, performed a detailed
81 comparison of coincident measurements and found the absolute value of the resulting mean relative
82 difference profile is <5% for 13–55 km and <3% for 24–54 km. Generally the OSIRIS data were found to
83 have a slightly higher bias around 22 km, particularly at higher latitudes.

84 The long term stability of the 11-year OSIRIS data set was comprehensively characterized and assessed by
85 Adams et al., 2014, who used the Microwave Limb Sounder (MLS) and Global Ozone Monitoring by
86 Occultation of Stars (GOMOS) satellite data records in addition to and ozonesonde measurements. The
87 results of this work show that the mean percent differences between coincident measurements are within 5%
88 at all altitudes above 18.5 km for MLS, above 21.5 km for GOMOS, and above 17.5 km for ozonesondes. The
89 stability analysis found that global average drifts relative to the validation data sets are < 3% per decade in
90 comparison with MLS for 19.5–36.5 km, GOMOS for 18.5–54.5 km, and ozonesondes for 12.5–22.5 km.
91 Adams et al., 2014, concluded that the 11-year OSIRIS data set is suitable for trend analysis.

92 Even in light of these encouraging results, the relatively long 4-year overlap between the SAGE II and
93 OSIRIS measurements is an essential factor in the ability to reliably merge these data sets into a single time
94 series for the assessment of decadal trends, particularly since post-2000 SAGE II was capturing only one
95 occultation per orbit. Similarly, as both instruments retrieve ozone number density on an altitude grid, there is
96 no need for unit and/or vertical co-ordinate conversions that can bias ozone trends if the simultaneous trends
97 in temperature are not properly accounted for (McLinden and Fioletov, 2011). For the purposes of this paper,
98 monthly mean ozone number density time series were calculated for both instruments in 10 degree latitude
99 bins from 60°S to 60°N and in 1 km altitude bins. Extending the analysis to higher latitudes is limited by the
100 lack of OSIRIS sampling in polar winter. The raw agreement in the monthly averaged ozone number density
101 can be seen from the sample time series shown for four different latitude-altitude bins in Figure 1. The top
102 panel, Fig. 1(a), at 18.5 km in the tropics shows the strong seasonal cycle that is observed by both instruments
103 just above the tropical tropopause. The second panel, Fig. 1(b), at 22.5 km shows the dominant Quasi-
104 Biennial Oscillation (QBO) signal in the tropical lower stratosphere. For slightly higher altitudes at 27.5 km,
105 the QBO signal becomes weaker as shown in the third panel, Fig. 1(c). The lowest panel, Fig. 1(d),
106 corresponding to 44.5 km altitude and 15°S to 25°S, is more typical of the upper stratosphere where weak
107 signal variation results in poor correlation despite approximate agreement between the two instruments.
108 Overall the time series show similar sized, in-phase, cyclic variations. Somewhat larger differences can be
109 found, particularly at altitudes above 40 km, when comparing to SAGE II sunrise or sunset measurements
110 separately, or with smaller latitudinal or temporal bins.

111 For most of the stratosphere there is a high correlation (greater than 0.9) between the two time series during
112 the overlap period as shown in Figure 2. Regions in the lower stratosphere where the correlation decreases
113 (circled and marked 'a' in the plot) correspond to regions where the overall cyclic variability in the time
114 series is much smaller. This can be seen by comparing the magnitude of the cyclic variability of the time
115 series in Fig. 1(c) to those at lower altitudes in Fig 1(a) and (b). These same regions also correspond to low
116 values of normalized standard deviation in the middle panel of Fig. 2. The decreased variability results in
117 lower values of correlation despite good agreement in monthly mean values, which agree to within typically
118 5% throughout the stratosphere as shown in the bottom panel of Fig. 2. This is a similar level of agreement
119 found by detailed coincidence comparisons (Adams et al., 2013). OSIRIS has a slightly higher bias for
120 latitudes south of 50°S, which extends throughout stratospheric altitudes and lasts throughout the austral
121 summer. OSIRIS also shows a slightly larger negative bias below 20 km between 40°S and 40°N. The
122 circled region marked 'b' in the correlation plot, as well as the rest of the upper stratosphere where the
123 correlation is not as high, are regions where there is very little seasonal or QBO signal in the time series,

124 again leading to reduced correlation even though the monthly mean values are in good agreement (see Fig.
125 1(d)).

126 A merged time series of the deseasonalized ozone anomaly was created by combining the two data sets. To
127 perform the deseasonalization, climatological means are found for each month by averaging for each
128 instrument independently; this accounts for any small difference in the observed seasonal cycles, as done
129 previously in other studies (Jones et al., 2009; Randel and Thompson, 2011). These means are determined
130 using the entire time span of each data set, i.e. 1984 to 2005 for SAGE II, and 2002 to 2013 for OSIRIS.
131 Sample deseasonalized anomaly time series for each instrument independently are shown in Figure 3 for the
132 same locations as those shown in the panels in Figure 1. Note that this technique results in zero average value
133 for the time series of each instrument. The final merged time series was determined by shifting the OSIRIS
134 time series at each latitude and altitude by the difference between OSIRIS and SAGE II during the overlap
135 period, and then averaging the two time series in any bin that contains a point from both OSIRIS and SAGE
136 II. The values that are used to shift the OSIRIS anomalies to match SAGE II during the overlap period are
137 quite small, typically less than 3%.

138 The resulting merged and deseasonalized time series is shown as a function of altitude and time for several
139 latitude bins in Figure 4. Missing data due to lack of sufficient sampling or screening of the data for
140 contamination from the large volcanic eruptions is shown in white. Particularly in the tropics there is the
141 clear signature of the downward propagating signal of the QBO. Note the approximate change in phase of
142 this signal above ~27 km, which is related to the change from dynamical to chemical control of stratospheric
143 ozone around this altitude level as reported by several other studies in the past (for example Randel and
144 Thompson, 2011; Chipperfield et al., 1994). In this figure, the beginning and end of the instrument overlap
145 period is marked with vertical black lines; however, the consistency of the observed structures across these
146 time periods fosters confidence in the quality of the merged data set. Note that even by eye, a broad
147 minimum in ozone can be seen in each latitude bin between 1994–2005, particularly in the upper stratosphere.

148 **Analysis and Results II: Time Series Analysis and Decadal Trends**

149
150 We have used a standard linear multi-variate regression analysis to quantify the variability and trends in the
151 merged and deseasonalized SAGE II – OSIRIS time series. We have included standard predictor variable
152 basis functions representing the solar cycle (F10.7 cm index from
153 ftp://ftp.ngdc.noaa.gov/stp/solar_data/solar_radio/flux/penticton_adjusted/daily), the El Niño–Southern
154 Oscillation (ENSO) from the Multivariate ENSO Index (MEI) obtained from the
155 NOAA Climate Diagnostics Center (Wolter, 2013), deseasonalized tropical tropopause pressure from the
156 National Centers for Environmental Prediction (NCEP), and the QBO ([http://www.geo.fu-](http://www.geo.fu-berlin.de/en/met/ag/strat/produkte/qbo/index.html)
157 [berlin.de/en/met/ag/strat/produkte/qbo/index.html](http://www.geo.fu-berlin.de/en/met/ag/strat/produkte/qbo/index.html) (Naujokat, 1986)). Typically QBO variability is fit based
158 on two orthogonal QBO time series that can be used in linear combination to represent the QBO at all
159 pressure levels (Wallace et al., 1993; Randel and Wu, 1996). We have used a principal component analysis to
160 generate the first three principal components of the QBO. As shown in the results of the regression below,
161 the third component is a smaller amplitude, high frequency term that accounts for a small fraction of the
162 observed ozone variability, approximately on the same order as the solar cycle proxy. Each of these predictor
163 variable basis functions is shown in Figure 5. The ENSO index is used with a 2-month lag as done previously

164 (see for example, Randel and Thompson, 2011).

165

166 For simplicity of interpretation, we have also used a piece-wise linear term to account for long term changes
167 in the stratosphere, particularly the changes in equivalent effective stratospheric chlorine (EESC). The time
168 of the inflection point of this piece-wise term, corresponding to the “turn-around” time, or the beginning of
169 ozone recovery, has been fixed at 1997. This is discussed in some detail in several studies including Kyrölä
170 et al., 2013, Laine et al., 2013, Newchurch et al., 2003, and Jones et al., 2009 with overall agreement on the
171 choice of 1997. This has also been the decision of the SI²N working group for the trend analysis for 2014
172 WMO ozone assessment (Harris et al., 2014, in preparation). The estimate of the uncertainty in the results of
173 the regression analysis is determined using a bootstrap resampling technique, with one year time granularity.
174 This estimate includes the effects of serial autocorrelation (Efron and Tibshirani, 1993; Randel and
175 Thompson, 2011).

176

177 All altitude/latitude bins are analyzed independently with no smoothing between the bins. Results of the
178 regression analysis for chosen altitude and latitude bins are shown in detail in Figures 6-8. In each column of
179 these figures, the uppermost panel shows the ozone anomaly in blue and resulting fit in red. The middle panel
180 shows the pre- and post-1997 linear terms in percent per decade, and the six predictor basis functions
181 multiplied by their normalized best-fit parameters, which are shown numerically along with each curve. The
182 lower panel contains the residual ozone anomaly. The left panel of Fig. 6 corresponds to the tropics at
183 18.5 km altitude, cf. Figures 1(a) and 3(a). At this location negative trends in ozone are apparent both before
184 and after 1997 with a relatively strong correlation to tropopause pressure and ENSO. The right panel
185 corresponds to the tropics at 22.5 km altitude, cf. Figures 1(b) and 3(b), with smaller but still negative linear
186 trends both before and after 1997. Here the ozone anomaly contains a very strong QBO signature from the
187 first two principal components. In Fig. 7, the left panel corresponds to the tropics at 27.5 km altitude, cf.
188 Figures 1(c) and 3(c). At this location a small positive linear trend is observed after 1997 and the QBO
189 indices are the dominant terms in the fit. The right panel corresponds to 25°S–15°S at 44.5 km altitude, cf.
190 Figures 1(d) and 3(d), where significant recovery of 5.1% per decade is observed after 1997 following a
191 decrease of -5.9% per decade prior to 1997. In Fig. 8, the left panel corresponds to 45°S–35°S at 42.5 km
192 altitude where the strongest changes between pre- and post-1997 linear trends occur. This same latitude and
193 altitude shows the strongest changes in the northern hemisphere as well. The right panel corresponds to 5°S–
194 5°N at 32.5 km altitude. In this region, the linear trends are reversed in direction compared to the remainder
195 of the stratosphere, but are statistically insignificant. In many of these cases, the fit residual variability is
196 large and does not appear completely random; this is consistent with previous studies and remains not well
197 understood (Randel and Thompson, 2011). Also, residuals appear larger for the SAGE II part of the record in
198 some places (example Fig. 8, left panel), probably because of the relatively sparser SAGE II sampling.

199

200 The relative contribution and spatial structure of each of the fitted predictor basis functions is shown in Fig. 9
201 as relative ozone change per one standard deviation of each parameter for QBO principal components,
202 tropical tropopause pressure, ENSO index and solar proxy. Regions that are not statistically significant at the
203 95% percent level are indicated with overlaid grey stippling. The QBO is a significant term throughout the
204 stratosphere with well-known out-of-phase patterns between the tropics and middle latitudes (Randel and Wu,
205 2007). As noted above, the third principal component is small but significant in places. The solar cycle

206 proxy is also small but significant in the middle and upper stratosphere in the Southern Hemisphere. The
207 overall pattern matches that reported by Kyrölä et al., 2013, from analysis of a merged SAGE II – GOMOS
208 time series, except at 50-60°N where the GOMOS analysis shows a larger influence of the solar cycle. Both
209 the ENSO index and the tropical tropopause pressure have relatively strong and significant projections in the
210 tropical lower stratosphere in the opposite sense (ENSO index is negative, and tropopause pressure is
211 positive). This is likely linked to the fact that both ozone and temperature changes are caused by enhanced
212 tropical upwelling in ENSO warm events (Randel et al., 2009; Calvo et al., 2010). The ENSO projection also
213 shows out-of-phase patterns in the NH midlatitude lower stratosphere, corresponding to ozone enhancements
214 during ENSO warm events (consistent with observations of column ozone over midlatitudes, e.g. Bronnimann
215 et al, 2004).

216

217 The results from the piece-wise linear fit for the pre- and post-1997 time periods are shown in Fig. 10. From
218 1984–1997, there are statistically significant negative trends of 5–10% per decade throughout the
219 stratosphere, except over the tropics between 22–34 km, where there is a small insignificant positive trend.
220 Significant recovery of 3–8% per decade from 1997 to the present is observed throughout the majority of the
221 stratosphere. Recovery is also not observed in the tropics between 25-35 km where there is a small
222 insignificant negative trend. The largest positive trends are observed in the upper stratosphere above 35 km.
223 Our results show a distinct hemispherical asymmetry in the recovery with the strongest positive trends at
224 southern latitudes between 20-45°S.

225

226

227 **Summary and Discussion**

228

229 These results derived from the merged SAGE II – OSIRIS data agree in general with the results from the
230 SAGE II – GOMOS time series analysis performed by Kyrölä et al., 2013. The altitude and latitude structure
231 of the results are very similar. The negative trends also match very well in terms of magnitude. However, the
232 asymmetry in the recovery in our result is not also in the GOMOS result and the magnitude of the recovery is
233 not as high as we find with the OSIRIS data set. The GOMOS analysis shows significant recovery only for
234 altitudes 38-45 km at a rate of 1-2% per decade and does not find the decreasing trend post-1997 in the
235 tropical lower stratosphere. There also seems to be a systematic decrease in the GOMOS post-1997 trends for
236 both northern and southern latitudes above 40 degrees that does not arise in the OSIRIS analysis performed
237 here. These differences may be explained in part by the small drifts, essentially < 3% per decade, detected by
238 Adams et al., 2013, in the comparisons between OSIRIS GOMOS, and MLS. Although the drift patterns are
239 not the same between OSIRIS and the other two instruments there are some noteworthy similarities including
240 positive drifts with respect to the other instruments in the tropics throughout stratospheric altitudes, and
241 negative drifts in the southern extratropics. The OSIRIS-GOMOS analysis shows a region of higher positive
242 drift south of 40S and above 40 km altitude, and stronger positive drifts (> 5% per decade) in the extratropics
243 below 20 km altitude. Because the drift analysis remains uncertain and more comprehensive comparisons
244 with other instruments and ground based measurements need to be done, we have chosen not to directly
245 modify the resulting trends, but have attempted to account for any potential drift by adding 3% per decade in
246 quadrature to the bootstrap uncertainties. This is shown in Fig. 11 where the grey stippling to indicate the
247 significance of the trend has been re-calculated in this fashion. In general, the recovery above 35 km remains

248 significant; however all trends in the altitude range 25-35 km are insignificant with conservative drift
249 estimates taken into account. The decreasing trend in the lower stratosphere also remains significant.

250
251 Other recent studies of long term satellite measurements have reported stratospheric ozone trends by similar
252 analyses, though not by merging with the SAGE II measurements. Gebhardt et al., 2014, derived trends using
253 SCIAMACHY measurements alone from 2002 to 2012. The broad pattern of recovery in the upper
254 stratosphere is consistent with our results, including the hemispheric asymmetry that shows stronger recovery
255 in the southern hemisphere. However, the SCIAMACHY results show strong, significant, negative trends of
256 up to -20% per decade in the tropics between 30-35 km, and up to -10% per decade in the northern
257 hemisphere middle latitudes between 25-35 km that are not in agreement with our results. Additionally, the
258 SCIAMACHY analysis does not show the decreasing trend in the lower stratosphere. Eckert et al., 2014, also
259 performed similar analyses using the MIPAS measurements alone from 2002 to 2012. These results show
260 recovery in the mid-latitude upper stratosphere and a small region of significant negative trend in the tropical
261 stratosphere near 30 km, though with smaller magnitude than that found with SCIAMACHY, and no
262 significant trend below 20 km.

263
264 This study has demonstrated the feasibility of merging the SAGE II and OSIRIS ozone profile records into a
265 single time series from 1984-present for the purpose of assessing variability and trends in stratospheric ozone.
266 The data sets each have high quality and high vertical resolution with small drifts in comparison to other
267 instruments. There is excellent agreement between the two data sets during the four year overlap period,
268 2002-2005, which enhances confidence in the ability to reliably merge the two records. Analysis of the time
269 series of monthly, zonally averaged values shows that both instruments correlate well throughout the
270 stratosphere from 60°S to 60°N and agree to within typically 5%. An instrument independent
271 deseasonalization of each time series is used to merge the data sets into a single time series of interannual
272 ozone anomalies spanning 1984-2013. A multivariate linear regression is used to assess the remaining
273 variability in terms of predictor basis functions including the QBO, ENSO index, tropical tropopause
274 pressure, solar proxy and a piece-wise linear term with fixed inflection at 1997. Our analyses capture the
275 spatial patterns and magnitudes for the QBO, ENSO and solar variations in ozone reported in previous studies
276 (references). The trend results show that from 1984–1997 there are statistically significant negative trends of
277 5–10% per decade throughout the stratosphere above 30 km, except over the tropics where a small and
278 insignificant positive trend is found between 30-35 km. In contrast, from 1997–present a statistically
279 significant ozone increase (recovery) of 3–8% per decade has taken place throughout most of the stratosphere,
280 with the notable exception between 40°S–40°N below approximately 22 km where the negative trend
281 continues, except below approximately 22 km altitude between 40°S and 40°N where decreasing trends
282 continue, consistent with the tropical lower stratospheric trend reported by Sioris et al., 2013. The bootstrap
283 uncertainty estimates are more conservative than those used by Sioris et al. and in this analysis the decreasing
284 trend very near to the equator is not significant. Sioris et al. also used a single linear trend throughout the
285 entire time period and splitting the analysis into pre- and post-1997 time periods may also affect the
286 significance of this decreasing trend. Thus the long-term negative trends in the tropical lower stratosphere
287 appear distinctive from the reversible changes, i.e. ozone recovery, observed in the middle and upper
288 stratosphere.

References

- Adams, C., Bourassa, A. E., Bathgate, A. F., McLinden, C. A., Lloyd, N. D., Roth, C. Z., Llewellyn, E. J., Zawodny, J. M., Flittner, D. E., Manney, G. L., Daffer, W. H., and Degenstein, D. A.: Characterization of Odin-OSIRIS ozone profiles with the SAGE II dataset, *Atmos. Meas. Tech.*, 6, 1447-1459, doi:10.5194/amt-6-1447-2013, 2013.
- Adams, C., Bourassa, A. E., Sofieva, V., Froidevaux, L., McLinden, C. A., Hubert, D., Lambert, J.-C., Sioris, C. E., and Degenstein, D. A.: Assessment of Odin-OSIRIS ozone measurements from 2001 to the present using MLS, GOMOS, and ozonesondes, *Atmos. Meas. Tech.*, 7, 49-64, doi:10.5194/amt-7-49-2014, 2014.
- Bogumil, K., Orphal, J., Homann, T., Voigt, S., Spietz, P., Fleischmann, O., Vogel, A., Hartmann, M., Kromminga, H., Bovensmann, H., Frerick, J., and Burrows, J.: Measurements of molecular absorption spectra with the SCIAMACHY pre-flight model: instrument characterization and reference data for atmospheric remote-sensing in the 230–2380 nm region, *J. Photochem. Photobiol. A-Chemistry*, 157, 167–184, doi:10.1016/S1010-6030(03)00062-5, 2003.
- Bourassa, A.E., D.A. Degenstein and E.J. Llewellyn, SASKTRAN: A Spherical Geometry Radiative Transfer Code for Efficient Estimation of Limb Scattered Sunlight, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 109, 52-73, doi:10.1016/j.jqsrt.2007.07.007, 2007.
- Brönnimann, S., J. Luterbacher, J. Staehelin, T. M. Svendby, G. Hansen, and T. Svenoe, Extreme climate of the global troposphere and stratosphere in 1940-42 related to El Nino, *Nature*, 431(7011), 971-974, 2004.
- Calvo, N., R. R. Garcia, W. J. Randel, and D. Marsh, Dynamical mechanism for the increase in tropical upwelling in the lowermost tropical stratosphere during warm ENSO events, *J. Atmos. Sci.*, 67, 2331–2340, doi:10.1175/2010JAS3433.1, 2010.
- Chipperfield, M. P., L. J. Gray, J. S. Kinnersley, and J. Zawodny, A two-dimensional model study of the QBO signal in SAGE II NO₂ and O₃, *Geophys. Res. Lett.*, 21, 589–592, doi:10.1029/94GL00211, 1994.
- Damadeo, R. P., Zawodny, J. M., Thomason, L. W., and Iyer, N.: SAGE version 7.0 algorithm: application to SAGE II, *Atmos. Meas. Tech.*, 6, 3539-3561, doi:10.5194/amt-6-3539-2013, 2013.
- Degenstein, D. A., Bourassa, A. E., Roth, C. Z., and Llewellyn, E. J.: Limb scatter ozone retrieval from 10 to 60 km using a multiplicative algebraic reconstruction technique, *Atmos. Chem. Phys.*, 9, 6521-6529, doi:10.5194/acp-9-6521-2009, 2009.
- Efron, B., and R. J. Tibshirani, *An Introduction to the Bootstrap*, 436 pp., Chapman and Hall, New York, 1993.
- Eckert, E., von Clarmann, T., Kiefer, M., Stiller, G. P., Lossow, S., Glatthor, N., Degenstein, D. A., Froidevaux, L., Godin-Beekmann, S., Leblanc, T., McDermid, S., Pastel, M., Steinbrecht, W., Swart, D. P. J.,

Walker, K. A., and Bernath, P. F.: Drift-corrected trends and periodic variations in MIPAS IMK/IAA ozone measurements, *Atmos. Chem. Phys.*, 14, 2571-2589, doi:10.5194/acp-14-2571-2014, 2014.

Gebhardt, C., Rozanov, A., Hommel, R., Weber, M., Bovensmann, H., Burrows, J. P., Degenstein, D., Froidevaux, L., and Thompson, A. M.: Stratospheric ozone trends and variability as seen by SCIAMACHY from 2002 to 2012, *Atmos. Chem. Phys.*, 14, 831-846, doi:10.5194/acp-14-831-2014, 2014.

Harris, N. R. P., Hassler, B., Tummon, F., Bodeker, G. E., Petropavlovskikh, I., Fioletov, V. E., Steinbrecht, W., Davis, S. M., Wang, H. J., Froidevaux, L., Frith, S. M., Wild, J., Kyrölä, E., Sioris, C., and Zawodny, J.: SI2N Overview paper: Analysis and Interpretation of Changes in the Vertical Distribution of Ozone, in preparation, 2014.

Jones, A., Urban, J., Murtagh, D. P., Eriksson, P., Brohede, S., Haley, C., Degenstein, D., Bourassa, A., von Savigny, C., Sonkaew, T., Rozanov, A., Bovensmann, H., and Burrows, J.: Evolution of stratospheric ozone and water vapour time series studied with satellite measurements, *Atmos. Chem. Phys.*, 9, 6055–6075, doi:10.5194/acp-9-6055-2009, 2009.

Kyrölä, E., M. Laine, V. Sofieva, J. Tamminen, S-M. Päivärinta, S. Tukiainen, J. Zawodny, and L. Thomason, Combined SAGE II-GOMOS ozone profile data set 1984–2011 and trend analysis of the vertical distribution of ozone, *Atmos. Chem. Phys.* 13, 10645–10658, 2013.

Laine, M., Latva-Pukkila, N., and Kyrölä, E.: Analyzing time varying trends in stratospheric ozone time series using state space approach, *Atmos. Chem. Phys. Discuss.*, 13, 20503–20530, doi:10.5194/acpd-13-20503-2013, 2013.

Llewellyn, E.J., N.D. Lloyd, D.A. Degenstein, R.L. Gattinger, S.V. Petelina, A.E. Bourassa, J.T. Wiensz, et al., The OSIRIS instrument on the Odin spacecraft, *Canadian Journal of Physics*, 82, 411-422, doi:10.1139/P04-005, 2004.

McCormick, M. P., J. M. Zawodny, R. E. Viega, J. C. Larson, and P. H. Wang, An overview of SAGE I and II ozone measurements, *Planet. Space Sci.*, 37, 1567–1586, doi:10.1016/0032-0633(89)90146-3, 1989.

McLinden, C. A. and V. Fioletov, Quantifying trends in stratospheric ozone: Complications due to stratospheric cooling, *Geophys. Res. Lett.*, 38, L03808, doi:10.1029/2010GL046012, 2011.

McLinden, C. A., A. E. Bourassa, S. Brohede, M. Cooper, D. A. Degenstein, W. J. F. Evans, R. L. Gattinger, C. S. Haley, E. J. Llewellyn, N. D. Lloyd, P. Loewen, R. V. Martin, J. C. McConnell, I. C. McDade, D. Murtagh, L. Rieger, C. von Savigny, P. Sheese, C. E. Sioris, B. Solheim, and K. Strong, OSIRIS: A decade of scattered light, *Bull. Am. Met. Soc.*, 93, 1845-1863, doi: 10.1175/BAMS-D-11-00135.1, 2012.

Murtagh, Donal, Urban Frisk, Frank Merino, Martin Ridal, Andreas Jonsson, Jacek Stegman, Georg Witt et al., An overview of the Odin atmospheric mission, *Can. J. Phys.*, 80, 4:309-319, 2002.

Naujokat, B.: An update of the observed quasi-biennial oscillation of the stratospheric winds over the tropics, *J. Atmos. Sci.*, 43, 1873–1877, 1986.

Newchurch, M. J., Yang, E.-S., Cunnold, D. M., Reinsel, G. C., Zawodny, J. M., and Russell, J. M.: Evidence for slowdown in stratospheric ozone loss: First stage of ozone recovery, *J. Geophys. Res.-Atmos.*, 108, 4507, doi:10.1029/2003JD003471, 2003.

Randel, W. J., and F. Wu, Isolation of the ozone QBO in SAGE II data by singular value decomposition, *J. Atmos. Sci.*, 53, 2546–2559, doi:10.1175/1520-0469, 1996.

Randel, W. J. and Wu, F.: A stratospheric ozone profile data set for 1979-2005: Variability, trends, and comparisons with column ozone data, *J. Geophys. Res.*, 112, D06313, doi:10.1029/2006JD007339, 2007.

Randel, W. J., R. R. Garcia, N. Calvo, and D. Marsh, ENSO influence on zonal mean temperature and ozone in the tropical lower stratosphere, *Geophys. Res. Lett.*, 36, L15822, doi:10.1029/2009GL039343, 2009.

Randel, W. J., and A. M. Thompson, Interannual variability and trends in tropical ozone derived from SAGE II satellite data and SHADOZ ozonesondes, *J. Geophys. Res.*, 116, D07303, doi:10.1029/2010JD015195, 2011.

Rienecker, M. M., Suarez, M. J., Gelaro, R., Todling, R., Bacmeister, J., Liu, E., Bosilovich, M. G., Schubert, S. D., Takacs, L., Kim, G.-K., Bloom, S., Chen, J., Collins, D., Conaty, A., da Silva, A., Gu, W., Joiner, J., Koster, R. D., Lucchesi, R., Molod, A., Owens, T., Pawson, S., Pegion, P., Redder, C. R., Reichle, R., Robertson, F. R., Ruddick, A. G., Sienkiewicz, M., and Woollen, J.: MERRA: NASA's Modern-Era Retrospective Analysis for Research and Applications, *J. Climate*, 24, 3624–3648, doi:10.1175/JCLI-D-11-00015.1, 2011.

Sioris, C. E., McLinden, C. A., Fioletov, V. E., Adams, C., Zawodny, J. M., Bourassa, A. E., and Degenstein, D. A.: Trend and variability in ozone in the tropical lower stratosphere over 2.5 solar cycles observed by SAGE II and OSIRIS, *Atmos. Chem. Phys. Discuss.*, 13, 16661-16697, doi:10.5194/acpd-13-16661-2013, 2013.

Von Savigny, C., C. Sioris, C. E. Sioris, I. C. McDade, E. J. Llewellyn, D. Degenstein, W. F. J. Evans et al., Stratospheric ozone profiles retrieved from limb scattered sunlight radiance spectra measured by the OSIRIS instrument on the Odin satellite, *Geophys. Res. Lett.*, 30, 14:1755, 2003.

Wallace, J. M., R. L. Panetta, and J. Estberg, Representation of the equatorial quasi-biennial oscillation in EOF phase space, *J. Atmos. Sci.*, 50, 1751–1762, doi:10.1175/1520-0469, 1993.

Weatherhead, E. C., Reinsel, G. C., Tiao, G. C., Jackman, C. H., Bishop, L., Hollandsworth Firth, S. M., Deluisi, J., Keller, T., Oltmans, S. J., Fleming, E. L., Wuebbles, D. J., Kerr, J. B., Miller, A. J., Herman, J., and McPeters, R.: Detecting the recovery of total column ozone, *J. Geophys. Res.*, 105, 22201–22210, 2000.

Wang, H. J.: Assessment of SAGE version 6.1 ozone data quality, *J. Geophys. Res.*, 107, 1–18, doi:10.1029/2002JD002418, 2002.

Wolter, K.: Multivariate ENSO Index (MEI), available at: ww.cdc.noaa.gov/people/klaus.wolter/MEI/, 2013.

WMO: Scientific Assessment of Ozone Depletion: 2010, Global Ozone Research and Monitoring Project – Report No. 52, World Meteorological Organization, Geneva, Switzerland, 2011.

Acknowledgements

This work was supported by the Natural Sciences and Engineering Research Council (Canada) and the Canadian Space Agency (CSA). Odin is a Swedish-led satellite project funded jointly by Sweden (SNSB), Canada (CSA), France (CNES) and Finland (Tekes).

Figure Captions

Figure 1: Time series of monthly average ozone number density in latitude and altitude bins. The grey shaded area denotes the overlap time between the start of OSIRIS measurements (shown in red) and end of the SAGE II measurements (shown in blue). Note the changing scale on the vertical axes.

Figure 2: Top: correlation between SAGE II and OSIRIS monthly mean during the overlap period 2002–2005. Middle: Standard deviation of the time series, including measurements from both instruments during the overlap period, normalized to the monthly mean. Bottom: Percent difference between the SAGE II and OSIRIS monthly mean over the same period. Encircled regions marked ‘a’ and ‘b’ denote decreased correlation due to lower amplitude seasonal and QBO variability.

Figure 3: Deseasonalized ozone anomaly at the same locations as Figure 1 calculated for each instrument independently. In the top panel, the strong seasonal cycle observed just above the tropical tropopause has been effectively removed. The remaining variability is mostly due to the QBO and tropopause pressure. At high altitudes in the tropical stratosphere shown in the second and third panels the QBO signal dominates the anomaly. For the upper stratosphere, shown in the bottom panel the QBO signal has greatly decreased.

Figure 4: Merged relative ozone anomaly for selected latitude bins. The overlap period is indicated with the black bars. Missing profiles, shown in white, occur when neither instrument samples a given month. Other periods of missing data confined to the lower stratosphere are due to screening of the SAGE II data for high aerosol extinction during large volcanic eruptions. The magnitude of the relative ozone anomaly is strongest over the tropics, where the downward propagating QBO signature is clear. In each latitude bin, a broad minimum in ozone can be seen between 1994–2005, particularly in the upper stratosphere.

Figure 5: The six non-linear basis functions used to fit the ozone anomaly. From top down: The first three principal components of the multi-level QBO; the F10.7 solar proxy; the El Niño Southern Oscillation index with a two month lag; de-seasonalized tropopause pressure.

Figure 6: In each column, the uppermost panel shows the ozone anomaly in blue and resulting fit in red for a given latitude and altitude. The middle panel shows the pre- and post-1997 percent per decade linear terms, and the six non-linear basis functions multiplied by their normalized best-fit parameters (shown on the right of each curve). The basis functions are shown on the same scale as the ozone anomaly. These terms sum to form the resulting fit. The lower panel contains the residual ozone anomaly. Left: 5°S–5°N at 18.5 km altitude (cf. Figures 1a and 3a). At this location negative trends in ozone are apparent both before and after 1997 with a relatively strong correlation to tropopause pressure and ENSO. Right: 5°S–5°N at 22.5 km altitude (cf. Figures 1b and 3b) with smaller but still negative both before and after 1997.

Figure 7: Same as Fig. 6. Left: 5°S–5°N at 27.5 km altitude (cf. Figures 2c and 4c). A small positive linear trend is observed after 1997. The QBO indices are the dominant terms. Right: 25°S–15°S at 44.5 km altitude (cf. Figures 1d and 3d). Significant recovery of 5.1% per decade is observed after 1997 following a decrease of -5.9% per decade prior to 1997.

Figure 8: Same as Fig. 6. Left: 45°S–35°S at 42.5 km altitude. The strongest change between pre- and post-1997 linear trends occurs in this region and similarly for the northern hemisphere at these latitudes. Right: 5°S–5°N at 32.5 km altitude. In this region, the linear trends are reversed in direction compared to the remainder of the stratosphere, but are statistically insignificant.

Figure 9: Altitude-latitude cross sections of the normalized fit parameter values for the six predictor basis functions. The units are relative ozone change per one standard deviation of the predictor time series. Regions that are not statistically significant at the 95% level are shown with overlaid grey stippling. In

general throughout the stratosphere the greatest contribution to the relative ozone anomaly comes from the first two principal components of the QBO index.

Figure 10: Altitude-latitude cross section of linear ozone trends in percent per decade before 1997 (top) and after 1997 (bottom) derived from the merged SAGE II and OSIRIS ozone anomaly time series. Statistical significance at the 95% level is denoted by areas without grey stippling. Significant recovery is observed throughout the majority of the stratosphere, except below approximately 22 km altitude between 40°S and 40°N where decreasing trends continue and in the tropics between 25-35 km where there is no significant trend.

Figure 11: The same linear trends in ozone in the post 1997 period as those shown in Fig. 10, but with the significance interval modified to account for potential drift in the OSIRIS measurements.