

1 **The Role of Convection in Tropical Ozone Trends (1998-2018) Based on SHADOZ**
2 **Profiles** **12 June 2020**

3 **Anne M. Thompson^{1*}, Ryan M. Stauffer^{1,2}, Jacquelyn C. Witte³, Debra E. Kollonige^{2,4},**
4 **Krzysztof Wargan^{1,4}, Jerry R. Ziemke^{1,5}**

5
6 ¹NASA/Goddard Space Flight Center (GSFC), Greenbelt, MD, USA
7 anne.m.thompson@nasa.gov; ORCID: 0000-0002-7829-0920

8
9 ²Earth System Science Interdisciplinary Center, University of Maryland, College Park, MD,
10 USA
11 ryan.m.stauffer@nasa.gov; ORCID: 0000-0002-8583-7795

12
13 ³National Center for Atmospheric Research Earth Observations Laboratory, Boulder, CO, USA
14 jwitte@ucar.edu; ORCID: 0000-0002-4110-5277

15
16 ⁴Science Systems and Applications, Inc., Lanham, MD, USA
17 debra.e.kollonige@nasa.gov; ORCID: 0000-0002-6597-328X; krzysztof.wargan-1@nasa.gov;
18 ORCID: 0000-0002-3795-2983

19
20 ⁵Morgan State University, Baltimore, Maryland, USA
21 jerald.r.ziemke@nasa.gov; ORCID: 0000-0002-5575-3654

22
23 *Corresponding author: Anne M. Thompson (anne.m.thompson@nasa.gov)

24 **Key Points:**

- 25 • Trends (1998-2018) in free tropospheric (FT) and lowermost stratospheric (LMS) ozone
26 and a convective proxy at SHADOZ sites were computed
- 27 • One station displayed an annual FT ozone trend (~5%/dec) and LMS loss (-3%/dec).
28 Ozone changed only in certain months at four other sites
- 29 • LMS ozone increases (decreases) occur in the low (high)-ozone months; these may tend
30 to counteract one another

31
32 **Keywords:** Tropical Ozone Trends, Lower Stratosphere, Ozonesondes, Free Troposphere,
33 SHADOZ

34 **Index Terms:** 345, 365, 1620, 3309, 3314

Abstract

Quantifying variability in the lowermost stratosphere (LMS) is important because of feedbacks among changing temperature, dynamics and species like ozone. We used reprocessed Southern Hemisphere Additional Ozonesondes data from 1998-2018 in a Multiple Linear Regression (MLR) model to analyze variability and trends in free tropospheric (FT) and LMS ozone across five well-distributed tropical regions. The MLR also computed trends in a proxy for convection as determined from laminae in each ozonesonde-radiosonde pair. Only the equatorial Americas exhibits statistically significant annual trends in FT or LMS ozone. At the other sites, ozone trends occur in isolated layers during months when convection has changed, February-April or July-November. Our results imply that large FT ozone increases reported for populated tropical areas may be caused by growing pollution overlying smaller changes caused by perturbed dynamics. They also provide regional data for evaluating LMS ozone trends based on zonal averages of often sparse satellite measurements.

Plain Language Summary

Understanding variability in lowermost stratosphere (LMS) ozone is an important topic in the climate assessment community because of feedbacks among changing temperature, dynamics and species like ozone. Most LMS evaluations are based on satellite observations. Tropospheric ozone assessments rely heavily on profiles from commercial aircraft. Ozonesonde measurements constitute an independent dataset that encompasses both LMS and troposphere. We used v06 Southern Hemisphere Additional Ozonesondes data from 1998-2018 in a Multiple Linear Regression model to analyze variability and trends in free tropospheric (FT) and LMS ozone across five well-distributed tropical sites. Our findings: (1) Only one SHADOZ site, in the equatorial Americas, exhibits small positive FT and negative LMS ozone trends on an annually averaged basis. (2) At the other 4 sites, trends only occur in isolated layers during months with decreasing (February-April) or increasing (July-September) convection. (3) The latter ozone changes are always positive in the FT. Because most SHADOZ stations are very remote, the results do not suggest large-scale tropical FT O₃ increases. They do imply that in the urban tropics where rising emissions create additional ozone, the trends observed in aircraft profiles may overlie smaller FT ozone increases caused by perturbed dynamics.

67 **1 Introduction**

68 Ozonesonde data are widely used by the scientific community for satellite validation,
69 model evaluation and analysis of trends, especially in the free troposphere (FT) through
70 lowermost stratosphere (LMS), i.e., from ~5-20 km, where uncertainties in most satellite
71 measurements are relatively large (*SPARC/IozoneC/GAW*, 2019). Many studies have used data
72 from SHADOZ (Southern Hemisphere Additional Ozonesondes; *Thompson et al.*, 2003; 2012), a
73 14-station tropical network with > 8300 profiles since 1998, to investigate FT and LMS ozone
74 variability, layers in which there are critical feedbacks among temperature, dynamics and species
75 like water vapor and ozone.

76 **1.1 Variability in FT and LMS Ozone: Role of Convection**

77 Early studies of FT and LMS ozone variability with SHADOZ profiles focused on
78 convective influences (*Folkins et al.*, 2002) and biomass burning (*Oltmans et al.*, 2001;
79 *Thompson et al.*, 2003). ENSO-perturbed patterns of convection, precipitation and fire induce
80 variability in FT and LMS ozone that vary station to station (*Randel and Thompson*, 2011).
81 *Thompson et al.* (2011) reported significant connections between LMS ozone vertical structure
82 and convectively-generated waves inferred from SHADOZ profiles. Convective links to FT
83 ozone structure are clearly evident when profiles are classified by Self-Organizing Maps (SOM;
84 *Jensen et al.*, 2012; *Stauffer et al.*, 2018).

85 **1.2 Trends in FT and LMS Tropical Ozone. Scope of Present Study**

86 Studies with satellite data, including Aura OMI and MLS ozone, reflect uncertainty in
87 both FT and LMS trends over the past 15-20 years. A review of various FT satellite products
88 displays a range of spatial ozone changes with disagreements in magnitude and sign (*Gaudel et*
89 *al.*, 2018). Recent work with merged satellite datasets (*SPARC/IO3C/GAW*, 2019) in the mid- to
90 lower stratosphere, along with chemistry transport and assimilation models, indicate the
91 uncertainty of LMS ozone trends (*Ball et al.*, 2018; *Chipperfield et al.*, 2018; *Wargan et al.*,
92 2018), at least on a zonally averaged basis. We address this situation with ozone profiles over a
93 range of stations using v06 SHADOZ data (*Thompson et al.*, 2017; *Witte et al.*, 2017; 2018) that
94 are better resolved than satellite measurements below 20 km. First, we review seasonal and
95 regional variations in FT and LMS ozone, then quantify their convective activity through
96 analysis of ozone and radiosonde laminae. Second, trends in ozone profiles and convection are

97 determined with a standard Multiple Linear Regression (MLR) model. Data and analysis
98 methods appear in **Section 2** with Results and Discussion in **Section 3**. **Section 4** is a summary.

99

100 **2 Data and Methods of Analysis**

101 **2.1 Reprocessed SHADOZ Data**

102 Ozone data are taken from the SHADOZ archive (<https://tropo.gsfc.nasa.gov/shadoz/>);
103 they originate from electrochemical concentration cell ozonesondes coupled to standard
104 radiosondes. In order to focus on convective impacts in the tropics we use v06 data from eight of
105 the 14 long-term stations (**Table 1**). For more reliable statistics three of the “stations” or “sites”
106 as they are referred to (**Figure 1**), are based on combining profiles from pairs of launch locations
107 abbreviated as SC-Para; Nat-Asc; KL-Java. The v06 data, reprocessed in 2016-2018, reduced
108 inhomogeneities due to instrument or data-handling changes (*Witte et al.*, 2017; 2018) such that
109 sonde total ozone column (TOC) amounts agree with ground-based or satellite data within 2%
110 for all but one station. Data from a number of SHADOZ stations display a 3-6% dropoff in TOC
111 after 2013 (*Sterling et al.*, 2018; *Stauffer et al.*, 2020) relative to satellite and/or ground-based
112 readings. For the stations analyzed here, the dropoff is confined to readings above 50 hPa (~20
113 km) and does not affect the results.

114 **2.2 Free Tropopause and LMS Definitions**

115 Illustrations in **Section 3** span the surface to 20 km and refer to two FT segments: 5-10
116 km; 10-15 km. We use 15-20 km for the lowermost stratosphere (LMS), because this is where
117 convective impacts on waves maximize (*Thompson et al.*, 2011) and where *Randel et al.* (2007)
118 identified a distinct ozone annual cycle driven by the Brewer-Dobson circulation. The LMS
119 includes most of the tropical tropopause layer (13.5-18.5 km) and several km above the tropical
120 cold-point and thermal lapse-rate tropopauses over the selected SHADOZ sites (*Thompson et al.*,
121 2012).

122 **2.3 Multiple Linear Regression Model (MLR)**

123 In order to quantify factors leading to seasonal and interannual variability as well as
124 trends, a standard multiple linear regression model (MLR; original version *Stolarski et al.*, 1991,
125 updated in *Ziemke et al.*, 2019) is applied to monthly mean ozone profiles for the 5 sites: the 3
126 combined sites, Nairobi, Samoa. The model includes terms for annual and semi-annual cycles

127 and oscillations prevalent in tropics: QBO, SOI (Southern Oscillation Index) and DMI (Indian
128 Ocean Dipole Moment Index):

$$129 \quad O_3(t) = A(t) + B(t)t + C(t)SOI(t) + D(t)QBO(t) + E(t)DMI(t) + \varepsilon(t)$$

130 where t is month. The coefficients are as follows: A is periodic with 12, 6, 4, and 3 month
131 cycles, and B through D have a period of 12 months, where A is the mean seasonal cycle and B
132 represents the month-dependent linear trend. The model includes data from the SOI
133 (<https://www.ncdc.noaa.gov/teleconnections/enso/indicators/soi/>), the u30 QBO index
134 (<https://www.cpc.ncep.noaa.gov/data/indices/qbo.u30.index>), and DMI
135 (<https://stateoftheocean.osmc.noaa.gov/sur/ind/dmi.php>). The $\varepsilon(t)$ is the residual, i.e., the
136 difference between the best-fit model and the raw data. Monthly ozone data and model fits for
137 the mid FT (5-10 km) and LMS (**Figures S1 and S2**) are well-correlated; for the LMS the
138 correlation coefficients r are ~ 0.8 (**Figure S2**).

139 **2.4 Lamina Identification (LID) and GW Indices**

140 The Lamina Identification (LID) method was used to identify convective signatures in
141 ozone profiles for the 1998-2009 SHADOZ data (*Thompson et al.*, 2011). The LID technique,
142 applied here to the 1998-2018 record (**Table 1**), is based on the coherence of laminae in each
143 ozone and potential temperature profile pair; laminae are identified as deviations from running
144 means calculated every 0.5 km from surface to 20 km. When the potential temperature and ozone
145 laminae at a given level are strongly correlated, as often occurs in the LMS, the presence of a
146 convectively-generated gravity wave (GW) is inferred. The GW occurrence is a proxy for a
147 convective event. Convective influence is quantified by the monthly GW frequency (GWF),
148 defined as the percent ratio of profiles exhibiting the GW signal relative to the total number of
149 profiles within a given month. A GW Index (GWI), defined as the fraction of the 15-20 km
150 ozone column (in Dobson Units, DU) that exhibits a GW signature, combines convection and its
151 LMS ozone impact. Monthly mean GWI and the altitude of the 380 K potential temperature
152 surface, often used to mark the tropical tropopause, over 1998-2018 are also ingested in the MLR
153 model.

154 **2.5 Self-Organizing Maps (SOM)**

155 We have used SOM, a machine-learning technique, to classify ozone profiles in terms of
156 meteorological or chemical influences (*Stauffer et al.*, 2016). The entire set of ozone profiles for
157 each station is ingested into the SOM code to obtain initial nodes (i.e., cluster centroids) via a

158 linear interpolation between the two largest components of the ensemble. Subsequent iterations
159 assign a given profile to its “best match” until a cluster mean is obtained. We adopt key elements
160 of the procedure in *Stauffer et al. (2018)*: 1) a four-cluster 2x2 SOM is used to avoid clusters
161 with too few members for meaningful statistics (cf *Jensen et al., 2012*); 2) SOM clusters are
162 numbered 1 to 4 based on the cluster “mean” ozone profile. The result is a consistent definition
163 of Cluster 1 and Cluster 4 as “low” and “high” ozone for each site, respectively.

164

165 **3 Results and Discussion**

166 **3.1 Seasonal Cycles in Ozone and Convective Influence**

167 **Figure 2** displays the 5-site monthly ozone climatology from the surface to 20 km.
168 Regional differences in vertical structure are pronounced. Red to yellow (~90-60 ppbv) colors
169 never appear in mid FT ozone over the equatorial Americas (SC-Para, **Figure 2a**), KL-Java or
170 Samoa (**Figures 2d,e**). Conversely, FT ozone values ≤ 30 ppbv never appear over Nat-Asc or
171 Nairobi (**Figures 2b,c**). These contrasts partly reflect regional differences in ascending vs.
172 descending nodes of the Walker circulation. The mean total ozone column thickness over the
173 south tropical Atlantic Ocean is 5% greater than over the western Pacific, giving rise to the well-
174 known tropospheric zonal wave-one (*Thompson et al., 2003*). Compared to the FT, there is less
175 regional variability in LMS ozone (Fig. 8 in *Thompson et al., 2017*) and a large seasonal cycle
176 (**Figure 3c**; cf *Randel et al., 2007*).

177 FT ozone seasonality is unique at each site due to the timing of various dynamical and
178 chemical influences. Localized FT ozone maxima occur largely from imported fire pollution: SC-
179 Para in March and after August; at KL-Java in April-May (**Figures 2a,d**); features at 6-8 km
180 over Nat-Asc and Samoa August to November (**Figures 2b,e**); Nairobi (**Figure 2c**) in June and
181 after August. Month-to-month changes viewed as anomalies from annual mean ozone (**Figure**
182 **3a,b**) appear complex but they reveal 3-4 distinct transitions when filtered with a criterion of a 5
183 ppbv gradient (**Figure S3**). The transition times (white vertical lines in **Figure 2**), March-April,
184 June-August, September-November, are similar at all locations. Convective influence, given by
185 GWF (**Figure S4**), with transitions marked as for ozone, shifts during the same periods. GWF
186 reaches 50-70% February-April at all locations (**Figure S4**), during which ozone minima above 8
187 km, attributed to convective redistribution of near-surface lower ozone air (**Figure 2**), appear
188 over all stations except Nairobi.

189 3.2 FT and LMS Ozone Changes (1998-2018)

190 In **Figure 4** FT and LMS changes in ozone mixing ratio (%/decade during 1998-2018)
 191 are displayed, based on monthly mean trends computed with the MLR model. Corresponding
 192 values in three layers appear in **Table 1**. Shades of red (blue) in **Figure 4** represent ozone
 193 increases (decreases); cyan hatching denotes statistical (95%) significance. For four stations
 194 (**Figures 4a-d**) there is a similar pattern in February through April with significant ozone trends
 195 at various altitudes in the FT and/or LMS. At SC-Para (**Figure 4a**) LMS ozone losses set in after
 196 May, extending in some layers to November. Mid-late year LMS ozone losses also occur over
 197 the four other sites (**Figures 4b-d**). However, **Table 1** (bold values) shows that these LMS ozone
 198 losses are only significant in isolated months. Thus, there is no overall trend except at SC-Para
 199 where a mean LMS loss (-3%/decade) overlies a positive annual FT ozone trend of ~5%/decade.

200 The dominant impact of southern African and South American fires on Nat-Asc and
 201 Samoa FT ozone in July through November is well-documented (*Oltmans et al., 2001; Thompson*
 202 *et al., 2003*). A near-absence of trends over these sites in the second half of the year (**Figures**
 203 **4b,e**) may signify little change in fires since 1998. FT ozone increases over KL-Java (**Figure 4d**)
 204 in February-March may be related to the southeast Asia fire season and/or to growing urban
 205 emissions (*Zhang et al., 2016*).

206 The annual cycles illustrated in **Figure 3** provide context for the changes shown in
 207 **Figure 4** and **Table 1**:

- 208 • FT ozone changes (5-15 km) are never significantly negative for any month
- 209 • In the mid FT (5-10 km), ozone trends are significantly positive only in the
 210 lowest-ozone, convectively active time of year (February to May)
- 211 • In the LMS, statistically significant ozone increases occur only during the low-
 212 ozone time of year (January to May) and decreases only during the higher-ozone
 213 period (June/July through November/December)

214 *Zhang et al. (2016)* and *Gaudel et al. (2018)* presented analyses of tropospheric ozone
 215 changes at different periods within 1994-2015. In those studies both satellite-derived
 216 tropospheric ozone columns and commercial aircraft profiles include boundary-layer ozone so
 217 they exceed the FT changes calculated here. The satellite trends, e.g., in *Zhang et al. (2016;*
 218 *supplement)*, do not capture the negligibly small FT ozone changes over Nat-Asc and Nairobi.

219

220 **3.3 Convective Influences in Ozone Trends**

221 **Sections 3.1 and 3.2** described an implicit role for convection in the seasonal variability
222 of FT and LMS ozone. Here, we examine links between ozone profile variability and convection
223 using the LID and SOM methods (**Sections 2.4 and 2.5**). The classification of ozone profiles for
224 several SHADOZ sites in a 2x2 SOM (*Stauffer et al.*, 2018) established an anticorrelation
225 between FT ozone mixing ratios and convective activity, where the latter was quantified by
226 meteorological parameters at sonde launch time (Figure 7 in *Stauffer et al.*, 2018). The SOM in
227 **Figure S5** shows similar relationships. The characteristic S-shapes in the upper FT in Cluster 1
228 (**Figure S5a**) display the lowest mixing ratios whereas much of the elevated ozone in Cluster 4
229 (**Figure S5b**) derives from imported pollution at 5-10 km. The GWF Cluster 1 (**Figure S5c**),
230 representing maximum convection, is dominated by January-May profiles (not shown), that is,
231 when there are positive FT ozone changes at all sites except Samoa.

232 We consider whether changes in GWI (the parameter that combines GWF and its impact
233 on LMS ozone) and 380 K altitude trends (**Figure S6**) can explain ozone trends (**Table 1**).
234 Statistically significant negative trends in GWI during January/February and March at Nat-Asc
235 and Nairobi (**Figure 5b,c**) coincide with increasing LMS ozone (**Figure 4b,c**). This combination
236 implies less wave (convective) activity. With suppressed convection, there are positive FT ozone
237 changes in January and February at Nat-Asc and Nairobi (cf **Figures 4b,c**). Samoa (**Figure 5e**)
238 exhibits a January loss in GWI but no significant LMS or FT ozone change.

239 There are large GWI increases at Nat-Asc (**Figure 5b**) in October and November but no
240 LMS ozone changes, consistent with increasing convection in the latter part of the year. This
241 pattern could also be explained by significant positive trends in the tropopause altitude at Nat-
242 Asc as well as at Nairobi and SC-Para in July to September (**Figure S6a-c**). Increasing
243 convection at KL-Java (**Figure 5d**) is implied by June and July GWI increases coincident with a
244 July LMS ozone loss. There is an insignificant positive 380 K altitude trend (**Figure S6d**).

245

246 **4 Summary**

247 The 21-year SHADOZ record (1998-2018) of ozone profiles from five well-distributed
248 tropical regions was used to compute trends in the FT (5-15 km) and LMS (15-20 km). Only at
249 one station, SC-Para, is there an annually averaged FT ozone increase, ~5%/decade, or annual
250 LMS ozone loss, -3%/decade. Changes in both FT and LMS ozone vary considerably from site to

251 site, with four of five stations displaying significant increases during February to April. Using
252 proxies for convection, it appears that these FT ozone increases may be due to reduced vertical
253 mixing. LMS ozone losses later in the year may take place when convective influence and the
254 tropopause altitude are both increasing.

255 *Randel et al. (2007)* and *Stolarski et al. (2014)* used satellite observations and
256 meteorological analyses to describe multiple dynamical influences on LMS ozone. Our
257 simplified study interprets FT and LMS ozone changes with reference to a single proxy for
258 vertical motion that is inferred from the sounding data. Nonetheless, the relatively small,
259 geographically distinct changes provide a reference for evaluating ozone trends derived from
260 satellite products that are typically presented as zonal averages (*Ball et al., 2018*). Model
261 interpretations of our results are required to assess whether recent reports of large tropical ozone
262 increases (*Zhang et al., 2016; Gaudel et al., 2018*) might reflect growing urban emissions
263 superimposed on smaller trends due to changes in dynamics. Model diagnostics are also required
264 to evaluate the contributions of diverse dynamical processes to ozone changes in the LMS.

265

266 **Acknowledgments**

267 Support is gratefully acknowledged from the NASA Upper Air Research Program (K. W.
268 Jucks, Program Manager), S-NPP and JPSS (J. F. Gleason, Project Scientist) and the NASA
269 Post-doctoral Program to RMS. We are grateful to O. R. Cooper (CIRES/NOAA-CSL) and W.
270 Randel (NCAR) for helpful comments. SHADOZ v06 profile data are available at
271 <https://tropo.gsfc.nasa.gov/shadoz/Archive.html>.

272

273 **References**

274 Ball, W. T., Alsing, J., Mortlock, D. J., Staehelin, J., Haigh, J. D., Peter, T., et al. (2018).
275 Continuous decline in lower stratospheric ozone offsets ozone layer recovery, *Atmos. Chem.*
276 *Phys.*, 18, 1379–1394, <https://doi.org/10.5194/acp-18-1379-2018>

277 Chipperfield, M. P., Dhomse, S., Hossaini, R., Feng, W., Santee, M. L., Weber, M., et al.
278 (2018). On the cause of recent variations in lower stratospheric ozone, *Geophys. Res. Lett.*, 45,
279 <https://doi.org/10.1029/2018GL078071>.

280 Folkins, I. Braun, C., Thompson, A. M., Witte, J. C. (2002). Tropical ozone as in
281 indicator of deep convective outflow, *J. Geophys. Res.*, 107, D13, doi: 10.1029/2001JD001178.

- 282 Gaudel, A., Cooper, O. R., Ancellet, G., Barret, B., Boynard, A., Burrows, J. P., et al.
 283 (2018). Tropospheric Ozone Assessment Report: Present-day distribution and trends of
 284 tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation,
 285 *Elem. Sci. Anth.*, 6: 39, doi: <https://doi.org/10.1525/elementa.291>.
- 286 Jensen, A. A., Thompson, A. M., Schmidlin, F. J. (2012). Classification of Ascension
 287 Island and Natal ozonesondes using self-organizing maps, *J. Geophys. Res.*, 117, D04302,
 288 doi:10.1029/2011JD016573.
- 289 Oltmans, S. J., Johnson, B. J., Harris, J. M., Vömel, H., Thompson, A. M., et al. (2001).
 290 Ozone in the Pacific tropical troposphere from ozonesonde observations, *J. Geophys. Res.*, 106,
 291 32503-32526, doi: <https://doi.org/10.1029/2000JD900834>.
- 292 Randel, W. J., M. Park, and F. Wu (2007). A large annual cycle in ozone above the
 293 tropical tropopause linked to the Brewer–Dobson circulation, *J. Atmos. Sci.*, 64, 4479-4488, doi:
 294 10.1175/2007JAS2409.1.
- 295 Randel, W. J., and A. M. Thompson (2011), Interannual variability and trends in tropical
 296 ozone derived from SHADOZ ozonesondes and SAGE II satellite data, *J. Geophys. Res.*, 116,
 297 D07303, doi:10.1029/2010JD015195.
- 298 SPARC/IO3C/GAW (2019). SPARC/IO3C/GAW Report on Long-term Ozone Trends
 299 and Uncertainties in the Stratosphere, I. Petropavlovskikh, S. Godin-Beekmann, D. Hubert, R.
 300 Damadeo, B. Hassler, V. Sofieva (Eds.), SPARC Report No. 9, GAW Report No. 241, WCRP-
 301 17/2018, doi: 10.17874/f899e57a20b; www.sparc-climate.org/publications/sparc-reports.
- 302 Stauffer, R. M., Thompson, A. M., Witte, J. C. (2018). Characterizing global ozonesonde
 303 profile variability from surface to the UT/LS with a clustering technique and MERRA-2
 304 reanalysis, *J. Geophys. Res. Atmos.*, 123, 6213–6229, <https://doi.org/10.1029/2018JD028465>.
- 305 Stauffer, R. M., Thompson, A. M., Kollonige, D. E., Witte, J. C., Tarasick, D. W.,
 306 Davies, J., et al. (2020). A post-2013 dropoff in total ozone at a third of global ozonesonde
 307 stations: Electrochemical concentration cell instrument artifacts? *Geophys. Res. Lett.*, 47,
 308 e2019GL086791. <https://doi.org/10.1029/2019GL086791>.
- 309 Sterling, C. W., Johnson, B. J., Oltmans, S. J., Smit, H. G. J., Jordan, A. F., Cullis, P. D.,
 310 et al. (2018). Homogenizing and estimating the uncertainty in NOAA's long-term vertical ozone
 311 profile records measured with the electrochemical concentration cell ozonesonde, *Atmos. Meas.*
 312 *Tech.*, 11, 3661-3687, <https://doi.org/10.5194/amt-11-3661-2018>.

313 Stolarski, R. S., Bloomfield, P. R., McPeters, R. D., Herman, J. R. (1991). Total ozone
 314 trends deduced from Nimbus 7 TOMS data, *Geophys. Res. Lett.*, 18, 1015-1018,
 315 <https://doi.org/10.1029/91GL01302>.

316 Stolarski, R. S., Waugh, D. W., Wang, L., Oman, L. D., Douglass, A. R., Newman, P. A.
 317 (2014). Seasonal variation of ozone in the tropical lower stratosphere: Southern tropics are
 318 different from northern tropics, *J. Geophys. Res. Atmos.*, 119, 6196–6206,
 319 doi:10.1002/2013JD021294.

320 Thompson, A. M., Witte, J. C., Oltmans, S. J., Schmidlin, F. J., Logan, J. A., et al.
 321 (2003). Southern Hemisphere ADDitional Ozonesondes (SHADOZ) 1998-2000 tropical ozone
 322 climatology. 2. Tropospheric Variability and the zonal wave-one, *J. Geophys. Res. Atmos.*, 108,
 323 8241, doi: <https://doi.org/10.1029/2002JD002241>.

324 Thompson, A. M., Allen, A. L., Lee, S. Miller, S. K., Witte, J. C. (2011). Gravity and
 325 Rossby wave signatures in the tropical troposphere and lower stratosphere based on Southern
 326 Hemisphere Additional Ozonesondes (SHADOZ), 1998–2007, *J. Geophys. Res.*, 116, D05302,
 327 doi:10.1029/2009JD013429.

328 Thompson, A. M., Miller, S. K., Tilmes, S., Kollonige, D. W., Witte, J. C., et al. (2012).
 329 Southern Hemisphere Additional Ozonesondes (SHADOZ) tropical ozone climatology:
 330 Tropospheric and tropical tropopause layer (TTL) profiles with comparisons to OMI based ozone
 331 products. *J. Geophys. Res.*, 117, D23301, doi: 10.1029/2010JD016911.

332 Thompson, A. M., Witte, J. C., Sterling, C., Jordan, A., Johnson, B. J., Oltmans, S. J., et
 333 al. (2017). First reprocessing of Southern Hemisphere Additional Ozonesondes (SHADOZ)
 334 ozone profiles (1998–2016): 2. Comparisons with satellites and ground-based instruments, *J.*
 335 *Geophys. Res. Atmos.*, 122, 13,000–13,025, <https://doi.org/10.1002/2017JD027406>.

336 Wargan, K., Orbe, C., Pawson, S., Ziemke, J. R., Oman, L. D., Olsen, M. A., et al.
 337 (2018). Recent decline in extratropical lower stratospheric ozone attributed to circulation
 338 changes, *Geophys. Res. Lett.*, 45, 5166–5176, <https://doi.org/10.1029/2018GL077406>.

339 Witte, J. C., Thompson, A. M., Smit, H. G. J., Fujiwara, M., Posny, F., et al. (2017). First
 340 reprocessing of Southern Hemisphere ADDitional Ozonesondes (SHADOZ) profile records
 341 (1998-2015): 1. Methodology and evaluation, *J. Geophys. Res. Atmos.*, 122,
 342 doi:10.1002/2016JD026403.

343 Witte, J. C., Thompson, A. M., Smit, H. G. J., Fujiwara, M., Johnson, B. J., et al. (2018).
344 First reprocessing of Southern Hemisphere ADDitional Ozonesondes (SHADOZ) profile records
345 (1998-2016): 3. Methodology and evaluation, *J. Geophys. Res. Atmos.*, 123,
346 doi:10.1002/2017JD027791.

347 Zhang, Y. Cooper, O. R., Gaudel, A., Thompson, A. M., Nédelec, P., Ogino, S.-Y., West,
348 J. J. (2016). Equatorward redistribution of emissions dominates the 1980 to 2010 tropospheric
349 ozone change, *Nature-Geoscience*, doi: 10.1038/NGEO282.

350 Ziemke, J. R., Oman, L. D., Strode, S. A., Douglass, A. R., Olsen, M. A., et al. (2019).
351 Trends in Global Tropospheric Ozone Inferred from a Composite Record of
352 TOMS/OMI/MLS/OMPS Satellite Measurements and the MERRA-2 GMI Simulation, *Atmos.*
353 *Chem. Phys.* 19, 3257–3269, doi: <https://doi.org/10.5194/acp-19-3257-2019>.

354

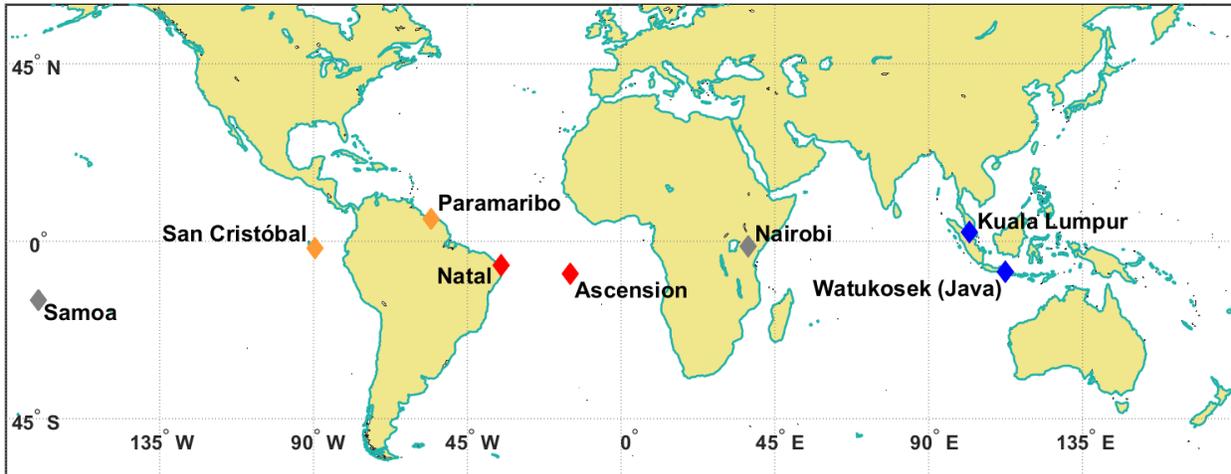
355

356 Table 1. SHADOZ site metadata including number of profiles and index terms used in MLR
 357 ozone calculations. Monthly MLR partial column ozone linear trends are shown, with significant
 358 trends in bold. Significant annual trends occur only at SC-Para (all levels) and Nat-Asc (10 to 15
 359 km). Note: As an independent check of the ozone profile trends (Figure 4), partial column ozone
 360 for each layer was calculated and subsequently input into the MLR to derive these statistics.
 361

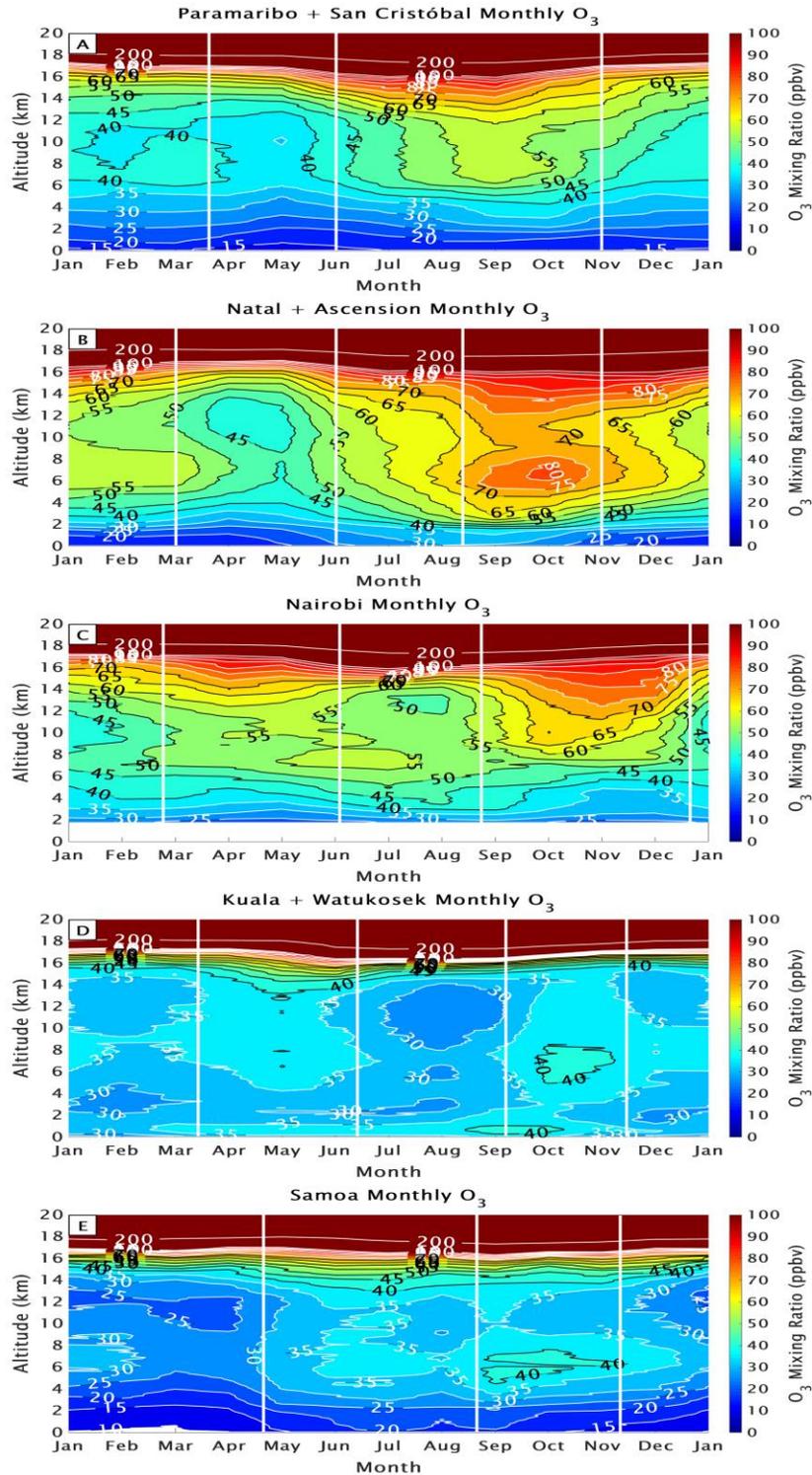
Site	Lat, Lon (°)	N	MLR Terms	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Ann
SC-Para	5.8, -55.21/ -0.92, -89.62	1190	ENSO+QBO													
5-10 km				-2.1	6.4	12.3	9.7	4.3	4.3	7.1	7.0	5.2	4.8	2.6	-2.0	5.0
10-15 km				-7.8	-4.1	12.6	17.9	1.1	-4.3	4.3	12.4	11.2	11.0	11.5	2.9	5.7
15-20 km				1.2	3.3	2.5	0.1	-3.3	-6.6	-8.1	-7.7	-5.9	-4.8	-4.6	-2.6	-3.0
Nat-Asc	-5.42, -35.38/ -7.58, 14.24	1363	ENSO+QBO													
5-10 km				-1.1	0.1	1.0	1.1	2.3	4.0	3.5	0.9	-0.7	-0.3	0.1	-0.7	0.8
10-15 km				7.9	7.7	3.3	1.5	2.8	3.4	4.2	5.5	4.3	1.5	0.3	3.3	3.8
15-20 km				8.4	11.1	6.3	2.3	1.2	-2.1	-5.6	-5.9	-3.2	-1.6	-1.5	1.7	0.9
Nairobi	-1.27, 36.8	905	ENSO+QBO													
5-10 km				4.3	12.6	13.7	4.8	-3.3	-3.8	-0.4	1.5	1.2	1.1	0.7	0.4	2.7
10-15 km				-1.1	4.4	6.9	4.4	1.3	2.2	3.2	-1.9	-6.1	-5.2	-2.6	-1.9	0.3
15-20 km				4.5	10.0	11.8	6.5	-0.8	-5.2	-5.9	-4.3	-1.1	1.6	1.9	1.7	1.7
KL-Java	2.73, 101.27/ -7.5, 112.6	770	ENSO+QBO +DMI													
5-10 km				-3.0	9.5	14.0	4.4	-1.1	1.8	3.2	-1.5	-1.0	3.7	2.1	-4.7	2.3
10-15 km				-6.2	3.9	12.2	11.7	6.9	2.4	-0.5	-1.3	0.5	0.9	-3.2	-8.2	1.6
15-20 km				-2.1	1.2	1.0	1.2	2.2	-0.6	-5.6	-7.4	-4.7	-2.7	-4.0	-4.9	-2.2
Samoa	-14.23, -170.56	752	ENSO+QBO													
5-10 km				3.7	6.4	6.4	-1.5	-5.6	-1.1	4.1	0.9	-4.7	-4.3	0.4	3.0	0.6
10-15 km				12.4	19.6	16.2	11.3	3.1	-3.5	-5.3	0.1	4.4	-0.5	-5.9	-1.4	4.2
15-20 km				0.3	6.8	3.8	-4.2	-5.3	-1.7	-1.3	-2.3	-0.7	0.8	-1.8	-4.0	-0.8

362

363

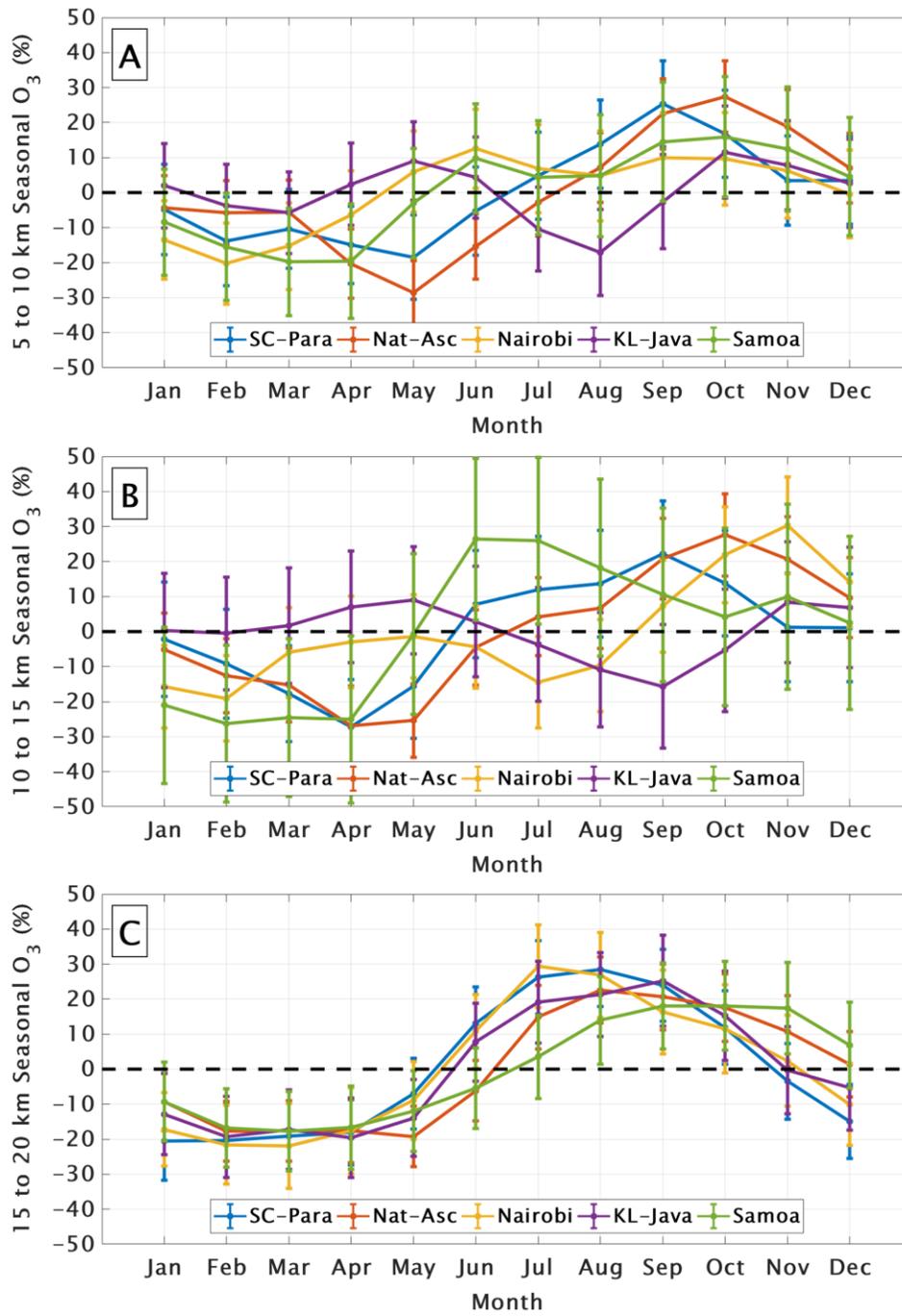


364
365 Figure 1. Map of SHADOZ stations used in this study. Stations whose combined records are
366 examined are colored orange (San Cristóbal and Paramaribo), red (Natal and Ascension), and
367 blue (Watukosek and Kuala Lumpur). Samoa and Nairobi records are studied individually and
368 colored grey. Sample numbers appear in Table 1.
369



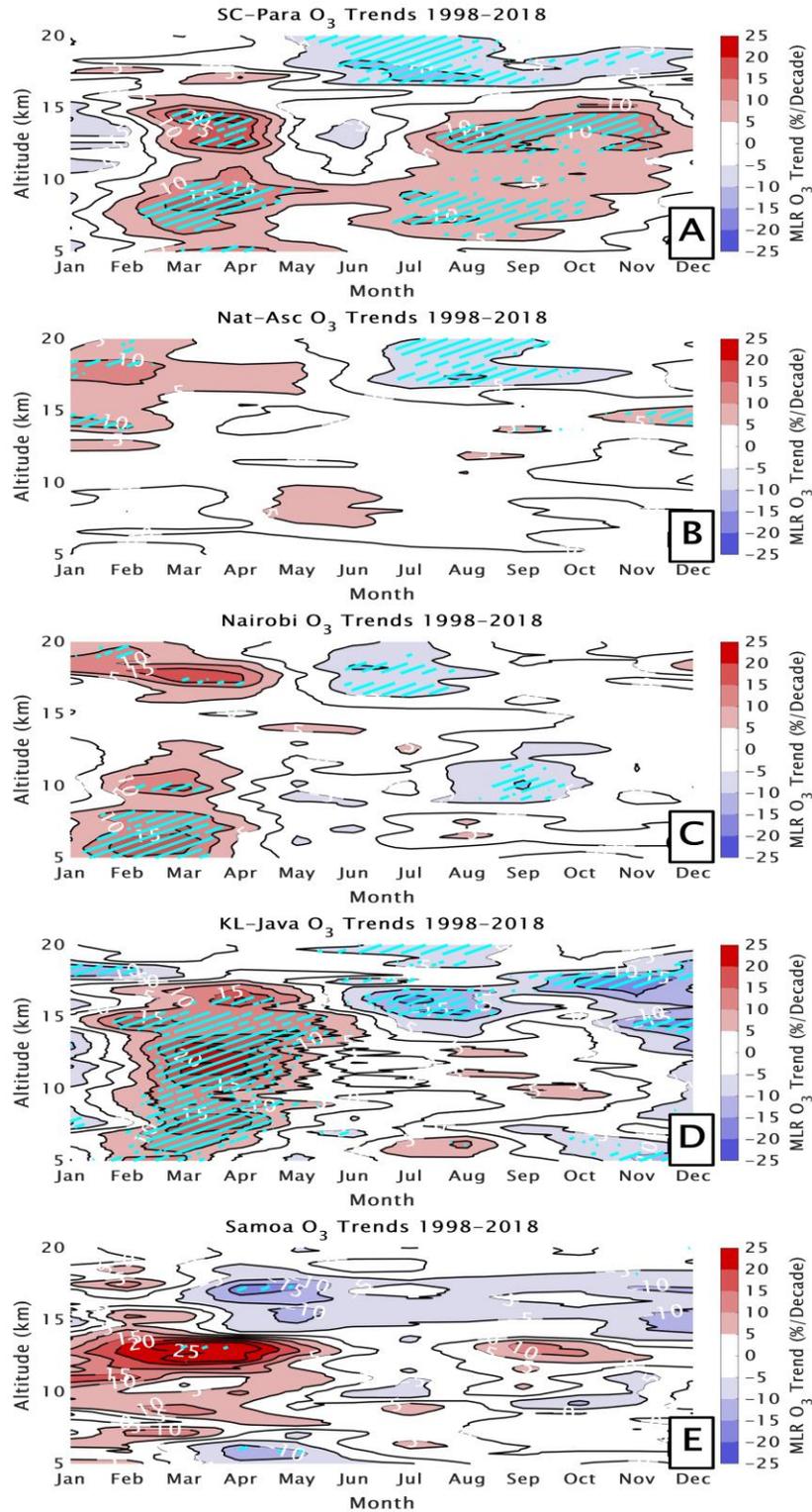
370
 371
 372
 373
 374
 375

Figure 2. Monthly averaged ozone mixing ratios from the surface to 20 km altitude for the five sites: two individual and three combinations. White dashed lines indicate transition periods marked by > 5 ppbv changes to the climatological FT and LMS ozone distributions (Figure S3).



376
 377
 378
 379
 380

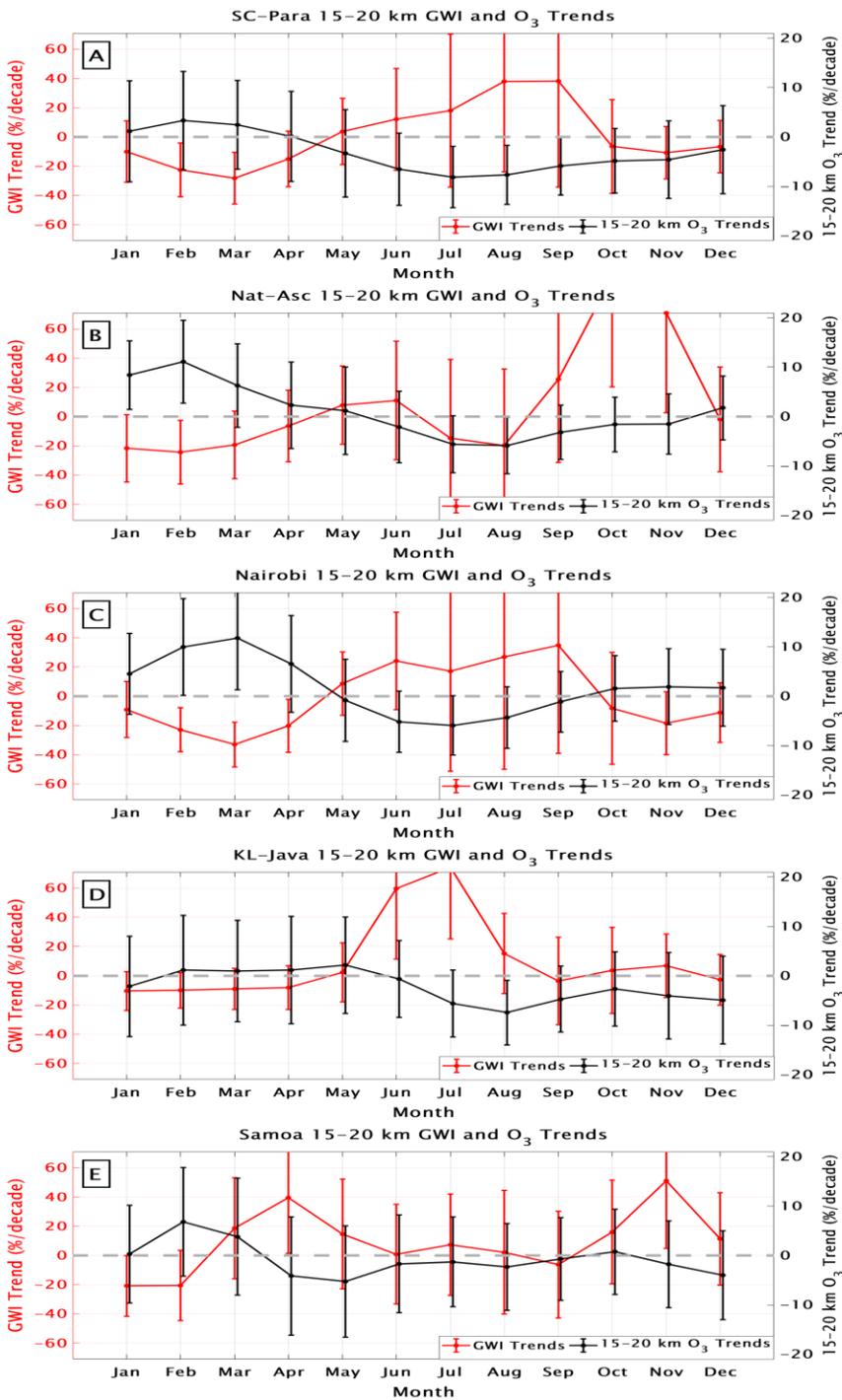
Figure 3. Seasonal variability in ozone in the FT (a and b), and LMS (c) expressed as percent anomaly, based on ozone mixing ratio deviation from the annual mean at the two individual and three combination sites.



381
382
383
384
385
386

Figure 4. Monthly MLR ozone linear trends from 5 to 20 km in percent per decade for the two individual and three combination sites. Positive trends are shown in red and negative trends are shown in blue. Trends that are significant with 95% confidence are shown with cyan hatching.

387



388

389

390 Figure 5. Monthly MLR gravity wave index (GWI) linear trends (red) and 15 to 20 km (LMS)
 391 partial column ozone linear trends (black) for the two individual and three combination sites. The
 392 dots represent the values and the error bars indicate the 95% confidence intervals. Values for the
 393 black lines can be found in Table 1.

Figure 1.

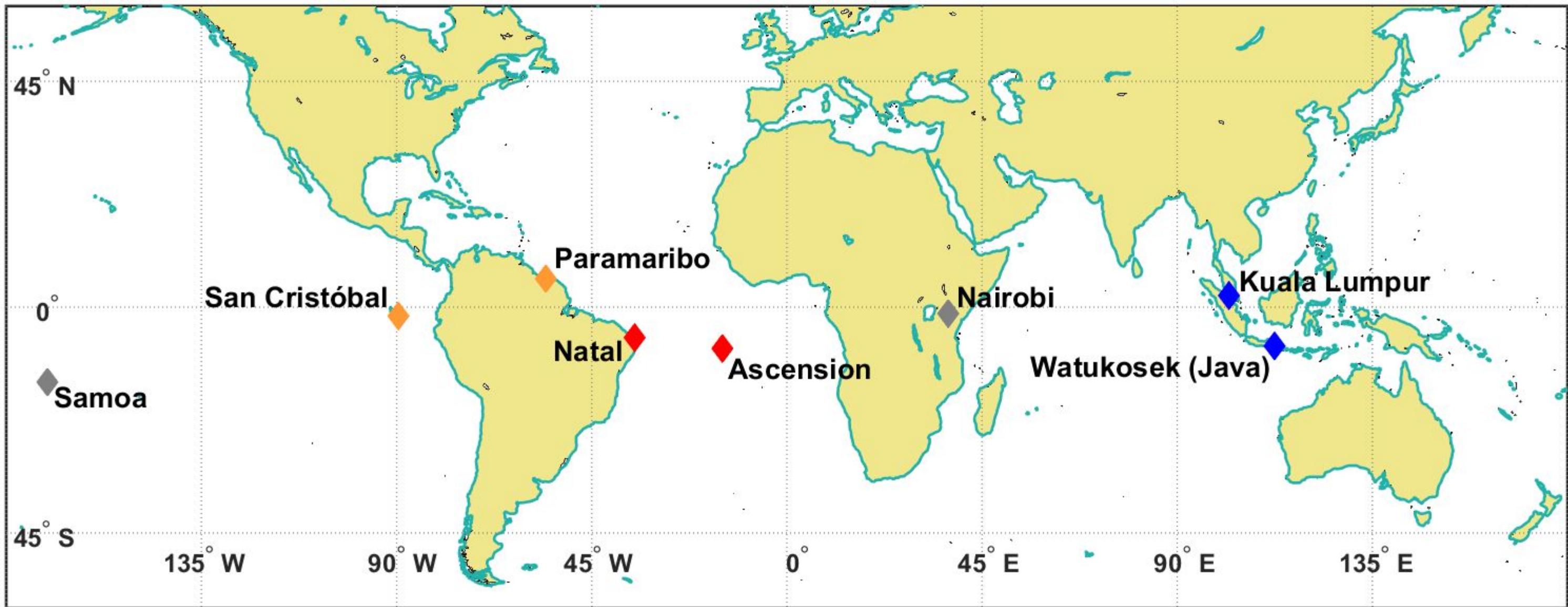
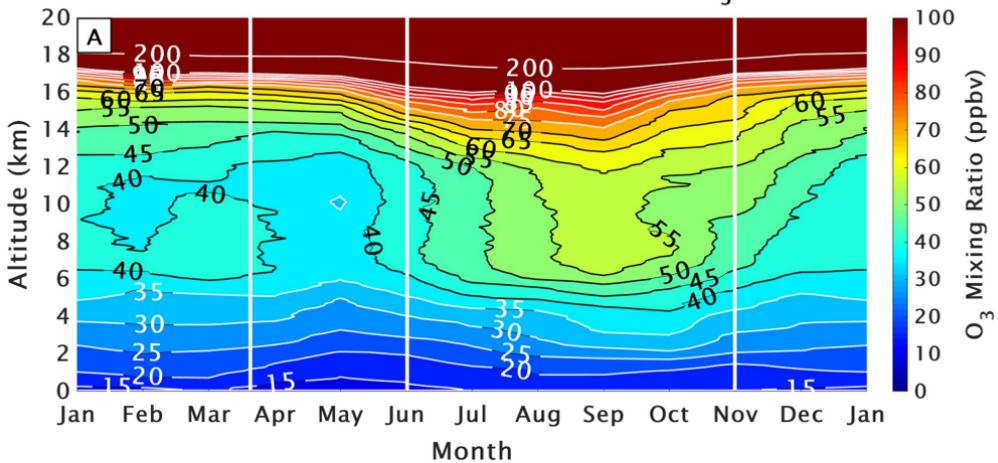
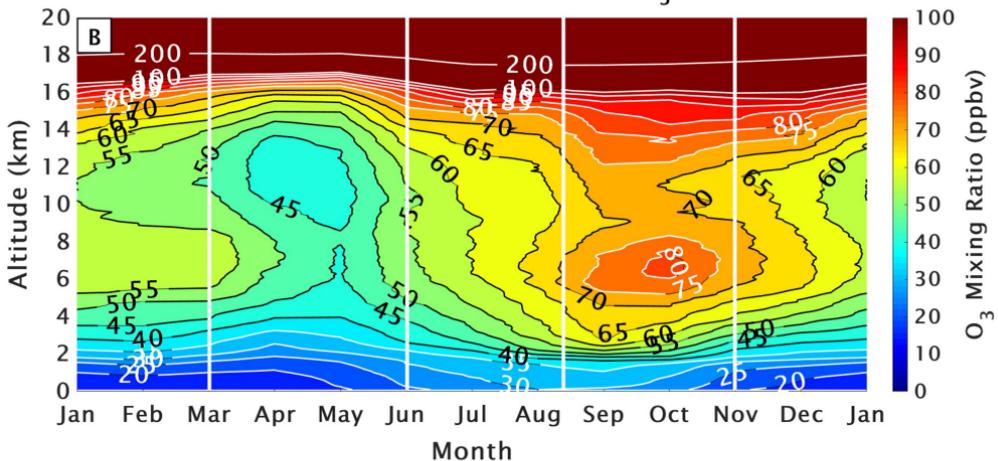


Figure 2.

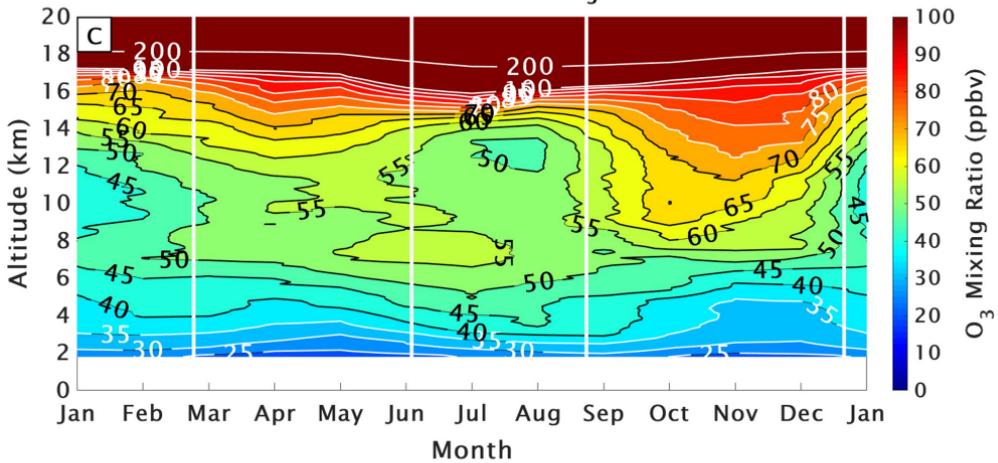
Paramaribo + San Cristóbal Monthly O₃



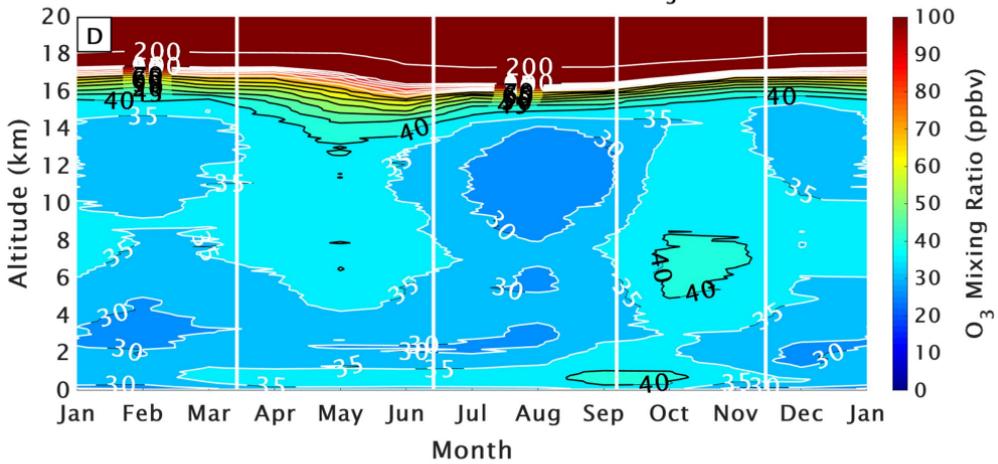
Natal + Ascension Monthly O₃



Nairobi Monthly O₃



Kuala + Watukosek Monthly O₃



Samoa Monthly O₃

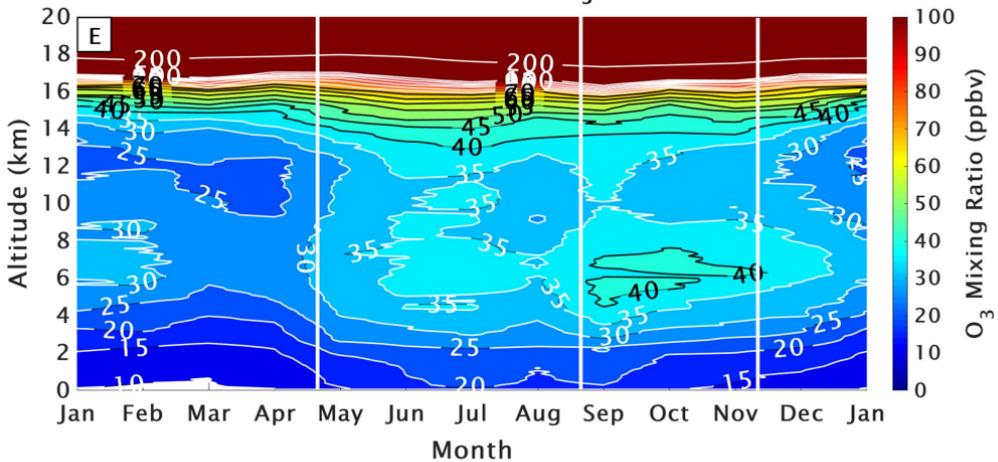


Figure 3.

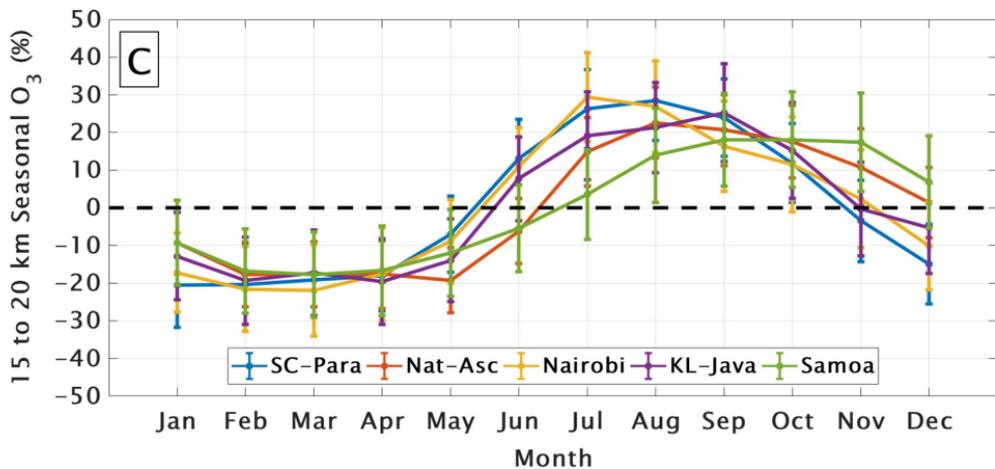
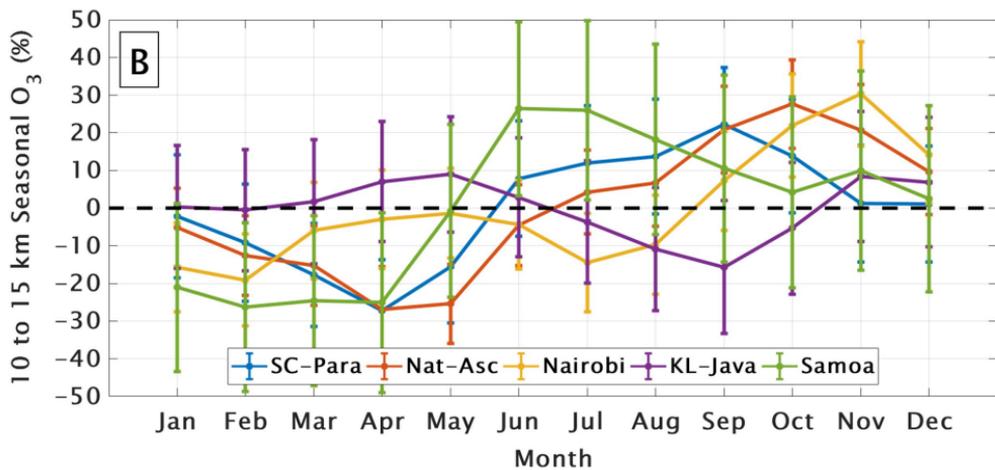
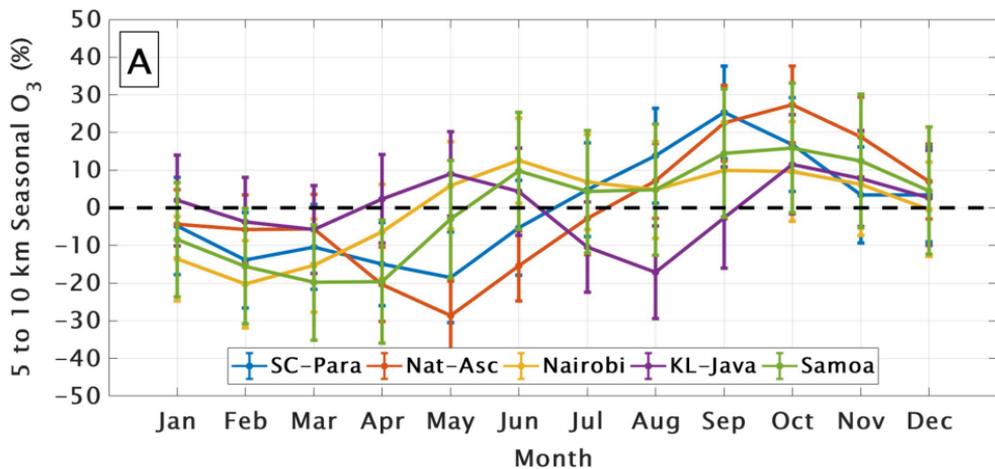
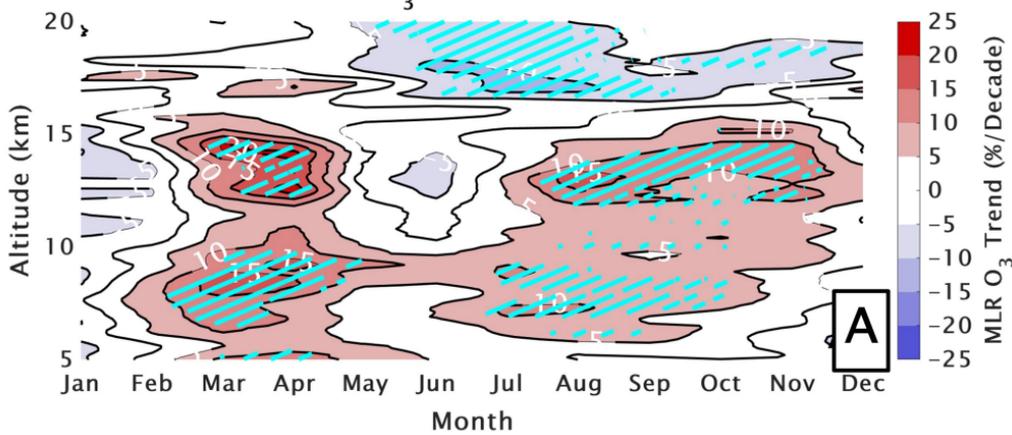
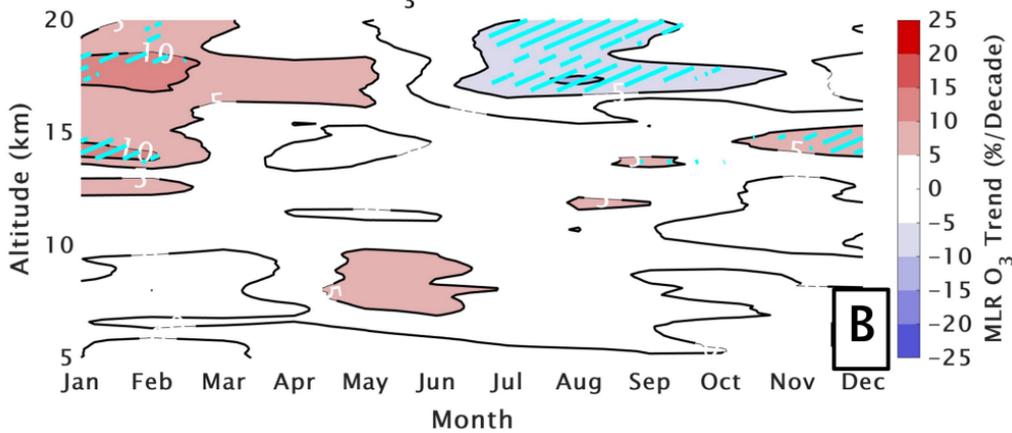


Figure 4.

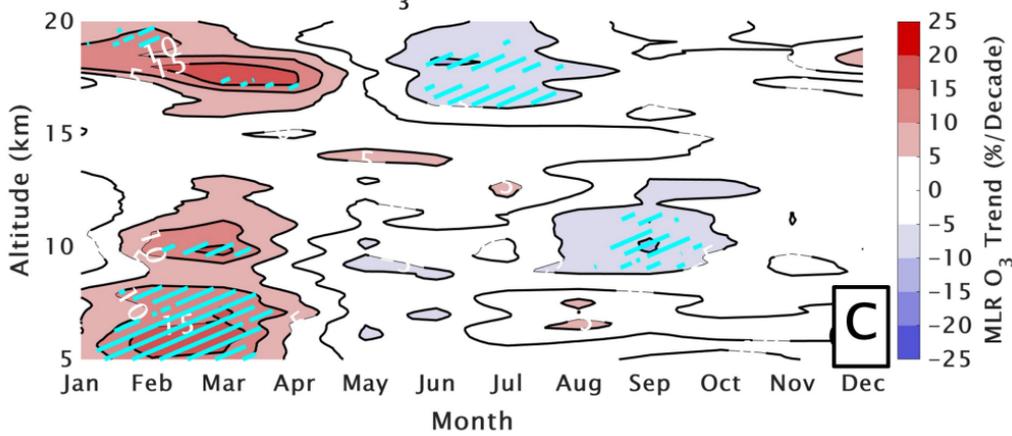
SC-Para O₃ Trends 1998–2018



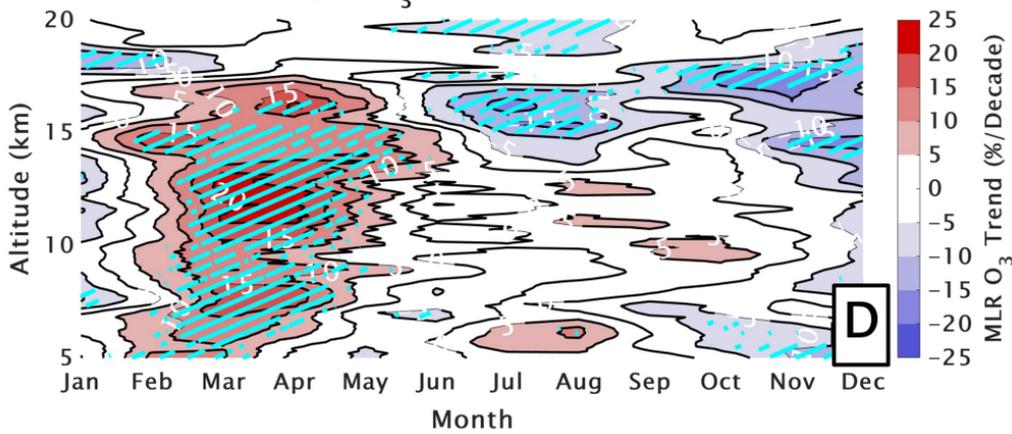
Nat-Asc O₃ Trends 1998–2018



Nairobi O₃ Trends 1998–2018



KL-Java O₃ Trends 1998–2018



Samoa O₃ Trends 1998–2018

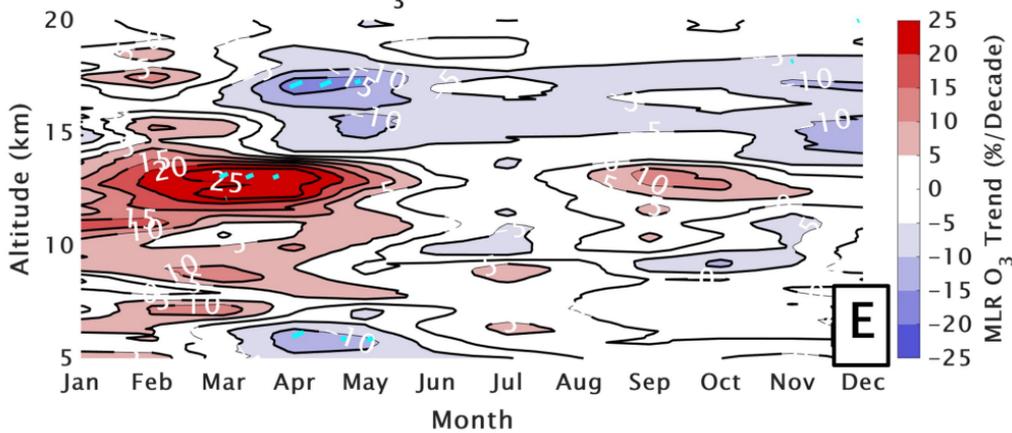
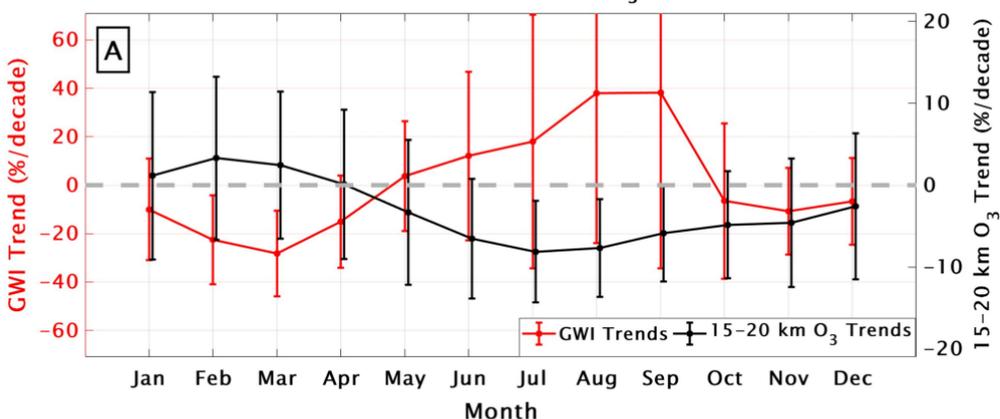
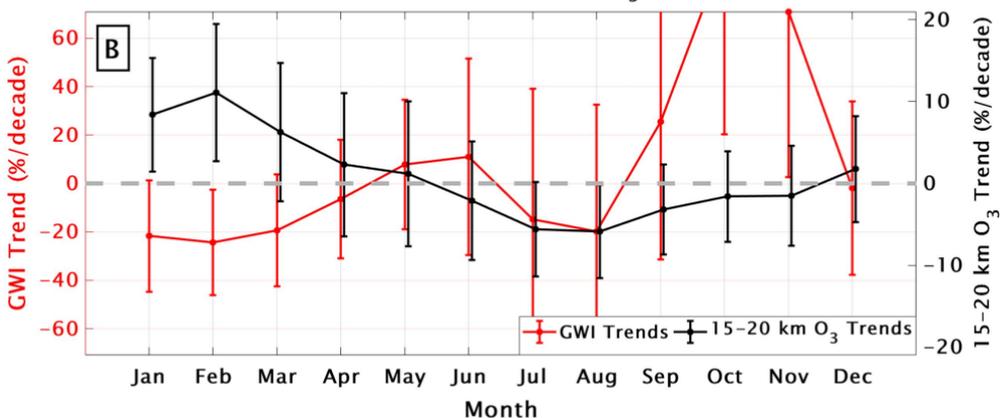


Figure 5.

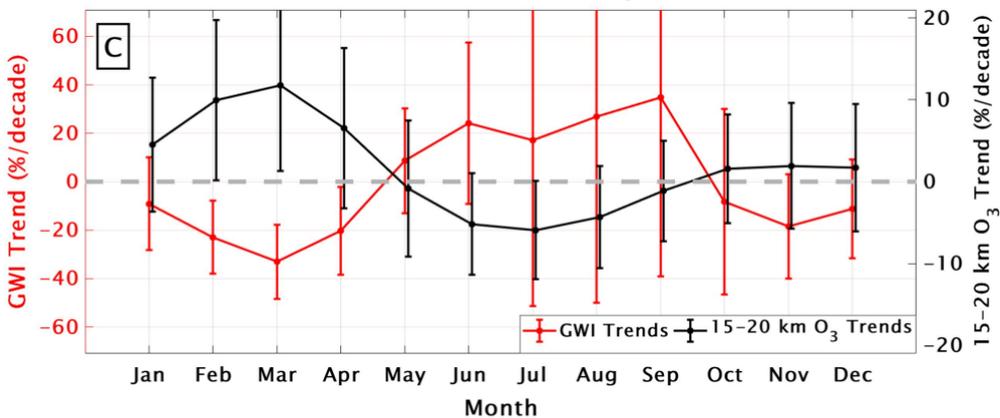
SC-Para 15-20 km GWI and O₃ Trends



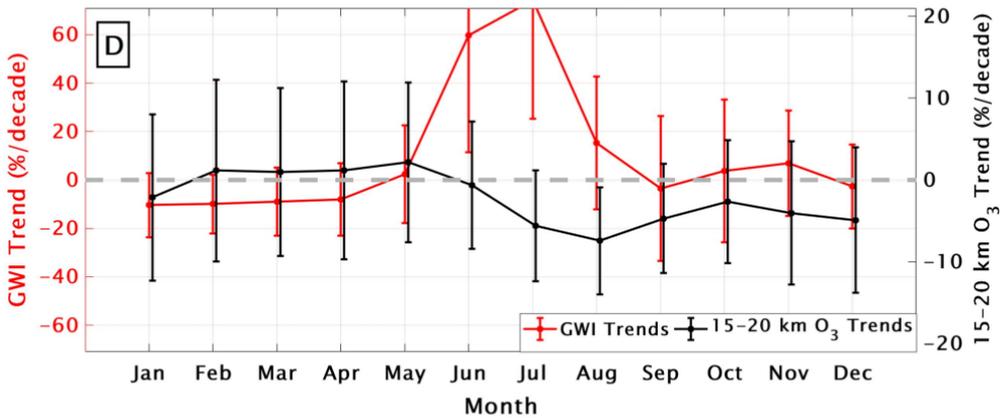
Nat-Asc 15-20 km GWI and O₃ Trends



Nairobi 15-20 km GWI and O₃ Trends



KL-Java 15-20 km GWI and O₃ Trends



Samoa 15-20 km GWI and O₃ Trends

