

# The use of QBO, ENSO and NAO perturbations in the evaluation of GOME-2/MetopA total ozone measurements

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**Abstract.** In this work we present evidence that quasi cyclical perturbations in total ozone (Quasi Biennial Oscillation (QBO), El Nino Southern Oscillation (ENSO) and North Atlantic Oscillation (NAO)) can be used as independent proxies in evaluating Global Ozone Monitoring Experiment-2 aboard MetopA (GOME-2A) satellite total ozone data, using ground-based measurements, other satellite data and chemical transport model calculations. The analysis is performed in the frame of the validation strategy on longer time scales within the European Organisation for the Exploitation of Meteorological Satellites (EUMETSAT), Satellite Application Facility on Atmospheric Composition Monitoring (AC SAF) project, and covers the period 2007-2016. Comparison of GOME-2A total ozone with ground observations shows mean differences of about  $-0.7 \pm 1.4\%$  in the tropics (0-30 deg.), about  $+0.1 \pm 2.1\%$  in mid-latitudes (30-60 deg.), and about  $+2.5 \pm 3.2\%$  and  $0.0 \pm 4.3\%$  over the northern and southern high latitudes (60-80 deg.), respectively. In general, we find that GOME-2A total ozone data depict the QBO/ENSO/NAO natural fluctuations in concurrence with co-located Solar Backscatter Ultraviolet Radiometer (SBUV), GOME-type Total Ozone Essential Climate Variable (GTO-ECV; composed of total ozone observations from GOME (Global Ozone Monitoring Experiment), SCIAMACHY (SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY), GOME-2A, and OMI (Ozone Monitoring Instrument) combined into one homogeneous time series) and ground-based (GB) observations. Total ozone from GOME-2A is well correlated with the QBO (highest correlation in the tropics of +0.8) in agreement with SBUV, GTO-ECV and GB data which also give the highest correlation in the tropics. The differences between deseasonalised GOME-2A and GB total ozone in the tropics are within  $\pm 1\%$ . These differences were tested further as to their correlations with the QBO. The differences had practically no QBO signal, providing an independent test of the stability of the long-term variability

40 of the satellite data. Correlations between GOME-2A total ozone and the Southern Oscillation Index (SOI) were  
41 studied over the tropical Pacific Ocean after removing seasonal, QBO and solar cycle related variability.  
42 Correlations between ozone and SOI are on the order of +0.5, consistent with SBUV and GB observations.  
43 Differences between GOME-2A and GB measurements at the station of Samoa (American Samoa; 14.25° S, 170.6°  
44 W) are within  $\pm 1.9\%$ . We also studied the impact of NAO on total ozone in the northern mid-latitudes in winter. We  
45 find very good agreement between GOME-2A and GB observations over Canada and Europe as to their NAO-  
46 related variability, with mean differences reaching the  $\pm 1\%$  levels. The agreement and small differences which were  
47 found between the independently produced total ozone data sets as to the influence of QBO, ENSO and NAO show  
48 the importance of these climatological proxies as additional tool for monitoring the long-term stability of satellite-  
49 ground truth biases.

## 50 **1 Introduction**

51 Ozone is an important gas of the Earth's atmosphere. In the stratosphere, ozone is considered *good ozone* because it  
52 absorbs ultraviolet-B radiation from the Sun, thus protecting the biosphere from a large part of the Sun's harmful  
53 radiation (e.g. Eleftheratos et al., 2012; Hegglin et al., 2015). In the lower atmosphere and near the surface, natural  
54 ozone has an equally important beneficial role because it initiates the chemical removal of air pollutants from the  
55 atmosphere such as carbon monoxide, nitrogen oxides and methane. Above natural levels however, ozone is  
56 considered *bad ozone* because it can harm humans, plants and animals. In addition, ozone is a greenhouse gas,  
57 warming the Earth's surface. In both the stratosphere and the troposphere, ozone absorbs infrared radiation emitted  
58 from Earth's surface, trapping heat in the atmosphere. As a result, increases or decreases in stratospheric or  
59 tropospheric ozone induce a climate forcing (Hegglin et al., 2015).

60 Ozone in the atmosphere can be measured by ground-based instruments, balloons, aircraft and satellites and can be  
61 calculated by chemical transport model (CTM) simulations. Measurements by satellites from space provide ozone  
62 profiles and column amounts over nearly the entire globe on a daily basis (e.g. WMO, 2014). The three Global  
63 Ozone Monitoring Experiment-2 (GOME-2) instruments carried on Metop platforms A, B and C serve this purpose.  
64 The first was launched on 19 October 2006, the second on 19 September 2012 and the last on 7 November 2018.  
65 The three GOME-2 instruments will provide unique long-term data sets of more than 15 years (2007-2024) related  
66 to atmospheric composition and surface ultraviolet radiation using consistent retrieval techniques (Hassinen et al.,  
67 2016). The GOME-2 off-line data is set to make a significant contribution towards climate and atmospheric research  
68 while providing near real-time data for use in weather forecasting and air quality forecasting applications (Hassinen  
69 et al., 2016).

70 Validation of satellite ozone measurements is performed with ground-based (GB) measurements as well as other  
71 satellite instruments (Hassinen et al., 2016). Validation of GOME-2A total ozone for the period 2007-2011 was  
72 performed by Loyola et al. (2011) and Koukouli et al. (2012). It was found that GOME-2 total ozone data agree at  
73 the  $\pm 1\%$  level with GB measurements and other satellite data sets (Hassinen et al., 2016). The consistency between  
74 GOME-2A and GOME-2B total ozone columns, including a validation with GB measurements, was presented by

75 Hao et al. (2014). An updated time series of the differences between GOME-2A and GOME-2B with GB  
76 observations can be found in Hassinen et al. (2016). The long-term stability of the two satellite instruments was also  
77 noted in that study. Both satellites are consistent over the Northern Hemisphere with negligible latitudinal  
78 dependence, while over the Southern Hemisphere there is a systematic difference of 1% between the two satellite  
79 instruments (Hassinen et al., 2016).

80 Chiou et al. (2014) compared zonal mean total column ozone inferred from three independent multi-year data  
81 records, namely, SBUV (v8.6) total ozone (McPeters et al., 2013), GOME-type Total Ozone Essential Climate  
82 Variable (GTO-ECV) (Coldewey-Egbers et al., 2015; Garane et al., 2018), and GB total ozone for the period 1996-  
83 2011. Their analyses were conducted for the latitudinal zones of 0-30° S, 0-30° N, 50-30° S, and 30-60° N. It was  
84 found that, on average, the differences in monthly zonal mean total ozone vary between -0.3 and 0.8% and are well  
85 within 1%. In that study it was concluded that despite the differences in the satellite sensors and retrievals methods,  
86 the SBUV v8.6 and GTO-ECV data records show very good agreement both in the monthly zonal mean total ozone  
87 and the monthly zonal mean anomalies between 60°S and 60°N. The GB zonal means showed larger scatter in the  
88 monthly mean data compared to satellite-based records, but the scatter was significantly reduced when seasonal  
89 zonal averages were analysed. The differences between SBUV and GB total ozone data presented in Chiou et al.  
90 (2014) are well in agreement with Labow et al. (2013), who systematically compared SBUV (v8.6) total ozone data  
91 with that measured by Brewer and Dobson instruments at various stations as a function of time, satellite solar zenith  
92 angle, and latitude. The comparisons showed good agreement (within  $\pm 1\%$ ) over the past 40 years with very small  
93 bias approaching zero over the last decade. Comparisons with ozone sonde data showed good agreement in the  
94 integrated column up to 25 hPa with differences not exceeding 5% (Labow et al., 2013).

95 The observed small biases (at the percentage level) between satellite and GB observations of total ozone, as have  
96 been documented in the above studies, ensure the provision of accurate satellite ozone measurements. The high  
97 accuracy and stability of the satellite instruments is essential for monitoring the expected recovery of the ozone layer  
98 resulting from measures adopted by the 1987 Montreal protocol and its amendments (e.g., Zerefos et al., 2009;  
99 Loyola et al., 2011; Solomon et al., 2016; de Laat et al., 2017; Kuttippurath and Nair, 2017; Pazmiño et al., 2018;  
100 Stone et al., 2018; Strahan and Douglass, 2018). It is known that total ozone varies strongly with latitude and  
101 longitude as a result of chemical and transport processes in the atmosphere. Total ozone also varies with season.  
102 Seasonal variations are larger over middle and high latitudes and smaller in the tropics (e.g. WMO, 2014). On longer  
103 time scales total ozone variability is related to large scale natural oscillations such as the Quasi-Biennial Oscillation  
104 (QBO) (e.g. Zerefos et al., 1983; Baldwin et al., 2001), the El Nino Southern Oscillation (ENSO) (e.g. Zerefos et al.,  
105 1992; Oman et al., 2013; Coldewey-Egbers et al., 2014), the North Atlantic Oscillation (NAO) (e.g. Ossó et al.,  
106 2011; Chehade et al., 2014) and the 11-year solar cycle (e.g. Zerefos et al., 2001; Tourpali et al., 2007; Brönniman et  
107 al., 2013). Moreover, volcanic eruptions may also alter the thickness of the ozone layer (Zerefos et al., 1994;  
108 Frossard et al., 2013; Rieder et al., 2013; WMO, 2014). These natural perturbations affect the background  
109 atmosphere and consequently the distribution of the ozone layer. In this context, the study of the effect of known  
110 natural fluctuations in total ozone could serve as additional tool for evaluating the long-term variability of satellite  
111 total ozone data records.

112 The objective of the present work is to examine the ability of the GOME-2A total ozone data to capture the  
113 variability related to dynamical proxies of global and regional importance such as the QBO, ENSO and NAO, in  
114 comparison to GB measurements, other satellite data and model calculations. The variability of total ozone from  
115 GOME-2A is compared with the variability of total ozone from the other examined data sets during these naturally-  
116 occurring fluctuations in order to evaluate the ability of GOME-2A to depict natural perturbations. The analysis is  
117 performed in the frame of the validation strategy of GOME-2A data on longer time scales within the project of  
118 EUMETSAT, AC SAF. The evaluation of GOME-2A data performed here includes the study of monthly means of  
119 total ozone, the annual cycle of total ozone, the amplitude of the annual cycle [i.e.,  $(\max-\min)/2$ ], the relation with  
120 the QBO (correlation with zonal wind at the equator at 30 hPa), the relation with ENSO (correlation with SOI) and  
121 the relation with the NAO (correlation with the NAO index in winter (DJF mean)).

122 The annual cycle describes regular oscillations in total ozone that occur from month to month within a year. In  
123 general, month-to-month variations of total ozone are larger in middle and high latitudes than in the tropics. The  
124 QBO dominates the variability of the equatorial stratosphere (~16-50 km) and is easily seen as downward  
125 propagating easterly and westerly wind regimes, with a variable period averaging approximately 28 months.  
126 Circulation changes induced by the QBO affect temperature and chemistry (Baldwin et al., 2001). ENSO and NAO  
127 are naturally-occurring patterns or modes of atmospheric and oceanic variability, which orchestrate large variations  
128 in climate over large regions with profound impacts on ecosystems (Hurrell and Deser, 2009). We present the level  
129 of agreement between satellite-derived GOME-2A and GB total ozone in depicting natural oscillations like QBO,  
130 ENSO and NAO, highlighting the importance of these climatological proxies to be used as additional tools for  
131 monitoring the long-term stability of satellite-ground truth biases.

## 132 **2 Data sources**

133 The analysis uses GOME-2 satellite total ozone columns for the period 2007-2016. This data forms part of the  
134 operational EUMETSAT AC SAF GOME-2/MetopA GDP4.8 data product provided by the German Aerospace  
135 Center (DLR). The GOME-2 total ozone data have been monthly averaged on a  $1^\circ \times 1^\circ$  latitude longitude grid. The  
136 overview of the GOME-2A satellite instrument and of the GOME-2 atmospheric data provided by AC SAF can be  
137 found in Hassinen et al. (2016).

138 To examine the natural variability of ozone on longer time scales, we have additionally analysed the GOME/ERS-2,  
139 SCIAMACHY/Envisat, GOME-2A, and OMI/Aura merged prototype level 3 harmonized data record (GTO-ECV,  
140  $1^\circ \times 1^\circ$ ) for the period 1995-2016 (Coldewey-Egbers et al., 2015; Garane et al., 2018). This GTO-ECV ozone data  
141 product was generated and provided by DLR as part of the European Space Agency Ozone Climate Change  
142 Initiative (ESA O3 CCI) project. The ESA O3 CCI merged level-3 record, which is based on  
143 GOME/SCIAMACHY/GOME-2A/OMI level-2 data, was obtained using the GODFIT v3.0 retrieval algorithm.  
144 More on ESA O3 CCI datasets can be found in the studies by Van Roozendaal et al. (2012), Lerot et al. (2014),  
145 Koukouli et al. (2015) and Garane et al. (2018).

146 Both datasets are compared with a combined TOMS/OMI/OMPS satellite total ozone data set constructed using data  
147 from the Total Ozone Mapping Spectrometer (TOMS) on Nimbus 7 (1979-1993), TOMS on Meteor 3 (1991-1994),  
148 TOMS on Earth Probe (1996-2005), the Ozone Monitoring Instrument (OMI) onboard the NASA Earth Observing  
149 System (EOS) Aura satellite (2005-present) and data from the next generation Ozone Mapping Profiler Suite  
150 (OMPS) nadir profiler instrument, launched in October 2011 on the Suomi National Polar-orbiting Partnership  
151 (NPP) satellite (McPeters et al., 2015). The total ozone data are available at  $1^\circ \times 1.25^\circ$  (TOMS) or  $1^\circ \times 1^\circ$   
152 (OMI/OMPS) resolution from <https://acd-ext.gsfc.nasa.gov/anonftp/toms/> (last access: 15 June 2018). From these  
153 data we constructed monthly mean total ozone data on a  $5^\circ \times 5^\circ$  grid. To account for known biases between the  
154 instruments (e.g., Labow et al., 2013) we use the Solar Backscatter Ultraviolet (SBUV) version 8.6 Merged Ozone  
155 Data Set (MOD) monthly zonal mean total ozone ([https://acd-ext.gsfc.nasa.gov/Data\\_services/merged/index.html](https://acd-ext.gsfc.nasa.gov/Data_services/merged/index.html),  
156 also see next paragraph; last access: 15 June 2018) as a reference. We adjust each instrument such that the zonal  
157 mean in each  $5^\circ$  band averaged over the instrument lifetime matches the corresponding SBUV MOD zonal mean  
158 average. Thus the inherent longitudinal variability is retained from the TOMS/OMI/OMPS measurements but any  
159 latitude-dependent bias between the instruments is removed. With the exception of Meteor 3 TOMS in the northern  
160 hemisphere, all offsets were within 2% at low and mid-latitudes. Such a data set should not be used for long-term  
161 trends but is sufficient for analyzing periodic variability such as QBO, ENSO and NAO. We used data for the period  
162 1995-2016. We note here that another long-term data set which has been analysed for QBO, ENSO, NAO and other  
163 perturbations comes from the Multi-Sensor Reanalysis (Knibbe et al., 2014), but is not examined here.

164 In addition, we compare with satellite SBUV station overpass data from 1995 to 2016. The satellite data are based  
165 on measurements from three SBUV-type instruments from April 1970 to the present (continuous data coverage from  
166 November 1978). Even though the time series includes different versions of the SBUV instrument, the basic  
167 measurement technique remains the same over the advancement of the instrument from the Backscatter Ultraviolet  
168 (BUV) to SBUV/2 (Bhartia et al., 2013). Satellite overpass data over various ground-based stations are provided per  
169 day from <https://acd-ext.gsfc.nasa.gov/anonftp/toms/sbuV/MERGED/> (last access: 15 June 2018). These overpass  
170 data are analogous to the SBUV MOD monthly zonal mean data previously mentioned. Both are constructed by first  
171 filtering lesser quality measurements and then averaging data from individual satellites when more than one  
172 instrument is operating. Monthly averages have been calculated by averaging the daily merged ozone overpass data  
173 for stations listed in Supplement Table S1. Details about the data are provided by McPeters et al. (2013) and Frith et  
174 al. (2014).

175 We also compare with GB observations of total ozone from a number of stations contributing to the World Ozone  
176 and Ultraviolet Radiation Data Centre (WOUDC). The WOUDC data centre is one of six World Data Centres which  
177 are part of the Global Atmosphere Watch programme of the World Meteorological Organization (WMO). The  
178 WOUDC data centre is operated by the Meteorological Service of Canada, a branch of Environment Canada. In  
179 total, we analysed total ozone daily summaries from 193 ground-based stations operating either Brewer, Dobson,  
180 filter, SAOZ or microtops instruments. The GB total ozone measurements are available from the website  
181 [https://woudc.org/archive/Summaries/TotalOzone/Daily\\_Summary/](https://woudc.org/archive/Summaries/TotalOzone/Daily_Summary/) (last access: 15 June 2018). The various stations  
182 used in this study are listed in Table S1.

183 We have also analysed simulations of total ozone from the global 3-D chemical transport model (CTM) Oslo CTM3  
184 (Søvde et al., 2012). The Oslo CTM3 has traditionally been driven by 3-hourly meteorological forecast data from  
185 the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecast System (IFS) model,  
186 whereas in this study we apply the OpenIFS model (<https://software.ecmwf.int/wiki/display/OIFS/>) (last access: 15  
187 June 2018), cycle 38r1, which is an improvement from Søvde et al. (2012). Details on the model are given in Søvde  
188 et al. (2012). The Oslo CTM3 comprises both detailed tropospheric and stratospheric chemistry. Photochemistry is  
189 calculated using fast-JX version 6.7c (Prather, 2012), and chemical kinetics from JPL 2010 (Sander et al., 2011).  
190 Total ozone columns compare well with measurements and other model studies (Søvde et al., 2012 and references  
191 therein). The horizontal resolution of the model is  $2.25^\circ \times 2.25^\circ$ . We used the global monthly mean total ozone  
192 columns for the period 1995-2016.

193 To examine the QBO component on total ozone we made use of the monthly mean zonal winds at Singapore at 30  
194 hPa. The zonal wind data at 30 hPa were provided by the Freie Universität Berlin (FU-Berlin) at <http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/qbo.dat> (last access: 15 June 2018) (Naujokat, 1986). The impact of ENSO in  
195 the tropics was investigated by using the Southern Oscillation Index (SOI) from the Bureau of Meteorology of the  
196 Australian Government (<http://www.bom.gov.au/climate/current/soi2.shtml>) (last access: 15 June 2018). The  
197 correlation between total ozone and the NAO index was mainly computed for the winter-mean (DJF) when the NAO  
198 amplitude is large (e.g. Hurrell and Deser, 2009), but it is also addressed in other seasons. Emphasis is given over  
199 Canada, Europe and the North Atlantic Ocean in winter. The principal component (PC)-based NAO index (DJF)  
200 provided by the Climate Analysis Section, NCAR, Boulder, USA (available at:  
201 <https://climatedataguide.ucar.edu/climate-data/hurrell-north-atlantic-oscillation-nao-index-pc-based>) (last access: 15  
202 June 2018) was used. Total ozone variability is also related to dynamical variability, for example variability in  
203 tropopause height (e.g. Dameris et al., 1995; Hoinka et al., 1996; Steinbrecht et al., 1998). The impact of tropopause  
204 height variations on total ozone variability was examined by analyzing the tropopause pressure from the  
205 independently produced NCEP/NCAR (National Centers for Environmental Prediction/National Center for  
206 Atmospheric Research) reanalysis 1 data set computed on a  $2.5^\circ$  grid. The NCEP/NCAR reanalysis data were  
207 provided from the web site at <https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.tropopause.html> (last  
208 access: 15 June 2018) (Kalnay et al., 1996).  
209

## 210 **3 Results and discussion**

### 211 **3.1 Monthly zonal means and annual cycle**

212 Figure 1 compares monthly mean total ozone from GOME-2A and SBUV (v8.6) satellite overpass data for stations  
213 shown in Table S1 (Supplement). The GOME-2A data were taken at a spatial resolution of  $1^\circ \times 1^\circ$  around each of the  
214 ground-based monitoring stations listed in Supplement Table S1 and then averaged over the tropics, middle and high  
215 latitudes of both Hemispheres in  $30^\circ$  latitudinal zones to provide the large scale monthly zonal means for the  
216 GOME-2A data. Accordingly, SBUV satellite overpass data were averaged over each geographical zone to provide  
217 the large scale zonal means for the SBUV observations. Mean differences and standard deviations between GOME-

218 2A and SBUV total ozone were found to be  $+0.1 \pm 0.7\%$  in the tropics (0-30 deg.), about  $+0.8 \pm 1.6\%$  in mid-  
219 latitudes (30-60 deg.), about  $+1.3 \pm 2.2\%$  over the northern high latitudes (60-80 deg. N) and about  $-0.5 \pm 2.9\%$  over  
220 the southern high latitudes (60-80 deg. S). The differences were estimated as  $[\text{GOME-2A} - \text{SBUV}] / \text{SBUV} (\%)$   
221 from January 2007 to December 2016. Small differences were also found between GOME-2A and GB  
222 measurements (Figure 2 and Table 1), where here GB stations data have been averaged over each geographical zone  
223 to provide the large scale zonal means for the GB measurements. Mean differences and standard deviations between  
224 GOME-2A and GB total ozone were found to be  $-0.7 \pm 1.4\%$  in the tropics (0-30 deg.),  $+0.1 \pm 2.1\%$  in mid-latitudes  
225 (30-60 deg.),  $+2.5 \pm 3.2\%$  over the northern high latitudes (60-80 deg. N) and  $0.0 \pm 4.3\%$  over the southern high  
226 latitudes (60-80 deg. S). Recall that all estimates refer to the period between January 2007 and December 2016.

227 In summary, the largest differences between GOME-2A, SBUV (v8.6) and GB measurements are found over the  
228 northern high latitudes (60°-80° N) and the highest variability (standard deviation of the mean difference) is  
229 observed over the latitude belt (60°-80° S). In addition, these differences (especially at the high latitudes) can be  
230 affected by the fact that the same days have not always been used for the construction of the monthly mean values  
231 for the different datasets. In the tropics and mid-latitudes the respective differences are within  $\pm 1\%$  or less, in line  
232 with Chiou et al. (2014). Validation results were also presented by Loyola et al. (2011), Koukouli et al. (2012),  
233 Coldewey-Egbers et al. (2015), Koukouli et al. (2015), updates of which are included in Hassinen et al. (2016). Our  
234 results based on data updated to 2017 largely confirm those studies, pointing to the good performance of GOME-2A  
235 when extending the period of record.

236 Next, we have studied the correlation between total ozone from GOME-2A and SBUV satellite data using linear  
237 regression analysis for the period 2007–2016. The statistical significance of the correlation coefficients,  $R$ , was  
238 calculated using the  $t$ -test formula for  $R$  with  $N-2$  degrees of freedom, as used in Zerefos et al. (2018). The  
239 regression model showed statistically significant correlations between the different datasets as follows:  $R = +0.99$  in  
240 the tropics, mid-latitudes and the northern high latitudes and  $R = +0.97$  in the southern high latitudes. All correlation  
241 coefficients are highly statically significant (99.9% confidence level). In the long-term, statistically significant  
242 correlation coefficients ( $R \geq +0.94$ ) are also found between GOME-2A satellite and GB measurements (Figure 2)  
243 despite the different type of instruments used to measure total ozone from the ground. The regression parameters for  
244 the correlation coefficients shown in Figures 1 and 2 are provided in Table 2.

245 A large part of the strong correlations shown in Figures 1 and 2 is attributable to the seasonal variability of total  
246 ozone which is presented in Figure 3 for GOME-2A, SBUV and GB data. More specifically, Figure 3 shows the  
247 seasonal variations of total ozone from station data, averaged per 10 degree latitude zones north and south. At high  
248 latitudes our analysis stops at 80 degrees. There is a very good agreement between the annual cycles of total ozone  
249 from the three datasets denoting the consistency of the satellite retrievals with GB observations. Similar annual  
250 cycles are also found with the GTO-ECV ozone data (not shown). Similar consistency is also revealed for the  
251 amplitudes of the annual cycles, computed as  $[(\text{maximum value} - \text{minimum value})/2]$  in Dobson Units (DU). Figure  
252 4 shows global maps of the amplitude of annual cycle of total ozone for the period 2007-2016 from GOME-2A  
253 (upper left panel), GTO-ECV (upper right) and the TOMS/OMI/OMPS (lower left) satellite data. All maps are

254 plotted against the sine of latitude north and south in order to show areas according to their actual size. As can be  
255 seen from Figure 4, the amplitude of annual cycle is less than 20 DU in the tropics, increasing as we move towards  
256 middle and high latitudes up to 75 DU. Interestingly, there is a region with small amplitude of annual cycle in the  
257 southern mid-latitudes with values of about 10-15 DU, seen in Figure 4 as a blue curved line crossing the longitudes  
258 around 60 degrees south, which points to small seasonal variations of total ozone in these parts. The seasonal  
259 increase in Antarctic ozone is delayed by 2-3 months compared to the north polar region. Only with the breakdown  
260 of the polar vortex in late spring, i.e. at a time when the poleward transport over lower latitudes has already ceased,  
261 does a strong ozone influx occur in the Antarctic. With this delay the amplitude of the seasonal variation stays much  
262 smaller poleward of 55-60° in the south than in the north (Dütsch, 1974). These features are consistent between all  
263 examined satellite data sets and are reproduced to a large extent by the Oslo CTM3 model as well, except in the  
264 southern mid-latitudes where the model seems to underestimate the observed annual cycle (Figure 4 lower right).

265 In summary, we find a similar pattern and amplitude of annual cycle between total ozone from GOME-2A and the  
266 other examined total ozone data sets. The mean differences in the annual cycles of GOME-2A and SBUV satellite  
267 data are small in the tropics (0-30 deg.:  $0.3 \pm 2.4$  DU), and increase as we move to mid-latitudes (30-60 deg.:  $2.4 \pm$   
268  $4.4$  DU) and higher latitudes (60-80 deg.:  $1.7 \pm 4.8$  DU). These numbers are consistent with the ones found between  
269 GOME-2A and GB measurements (tropics:  $1.1 \pm 2.3$  DU; mid-latitudes:  $1.2 \pm 5.1$  DU; high latitudes:  $5.1 \pm 7.1$  DU).  
270 In all latitude zones the correlation coefficients between the annual cycles of GOME-2A – SBUV and GOME-2A –  
271 GB data pairs were found to be greater than 0.9.

272 Before examining correlations with the large scale natural fluctuations QBO, ENSO and NAO, the mean annual  
273 cycle has been removed from the ozone data sets as described in the next section.

### 274 **3.2 Correlation with QBO**

275 We then studied how changes in dynamics affect the ozone columns in the atmosphere. The time series obtained  
276 have been deseasonalised by subtracting the long-term monthly mean from each individual monthly mean value.  
277 Ozone column variations for different latitude zones in the Northern and Southern Hemispheres have been  
278 compared. Figure 5 compares total ozone deseasonalised anomalies (in % of the mean) from GOME-2A and SBUV  
279 satellite retrievals in the tropics (10° N–10° S), sub-tropics (10°–30°) and mid-latitudes (30°–60°). The right panel of  
280 Figure 5 shows the respective anomalies from GTO-ECV data. Mean differences between GOME-2A and SBUV  
281 deseasonalised monthly zonal means between 60° N and 60° S are less than  $\pm 0.5\%$ .

282 The line with dots superimposed on the ozone anomalies in Figure 5 shows the equatorial zonal winds at 30 hPa  
283 which were used as a proxy index to study the impact of QBO on total ozone. The general features include a QBO  
284 signal in total ozone at latitudes between 10° N and 10° S which almost matches with the phase of QBO in the zonal  
285 winds. At higher northern and southern latitudes there is a phase shift in the QBO impact on total ozone. The impact  
286 of QBO is most pronounced in the tropics and less pronounced in the sub-tropics and mid-latitudes. Strong positive  
287 correlations with the QBO are found in the tropics (correlation between GOME-2A and QBO of about +0.77, t-test  
288 = 12.91) and weaker (usually of opposite sign) less significant correlations are found at higher latitudes (about –0.15



289 in the northern and about  $-0.45$  in the southern extra tropics). Similar correlation patterns with the QBO are found  
290 for the GTO-ECV, SBUV and GB data. These correlations suggest that the variability that can be attributed to the  
291 QBO in the tropics is about 60%, and about 2% and 20% in the northern and the southern extra tropics, respectively.

292 Table 3 summarizes the correlation and regression coefficients between total ozone and QBO at 30 hPa for the  
293 different latitude zones and the different datasets. For latitudes between  $10^\circ$  N and  $10^\circ$  S correlations between total  
294 ozone from GOME-2A, GTO-ECV, SBUV, GB data and the QBO are all positive. At latitudes between  $10^\circ$  and  $30^\circ$   
295 the correlations turn to negative, in agreement with Knibbe et al. (2014) results, who noted that moving from the  
296 tropics towards higher latitudes the regression estimates switch to negative values at approximately  $10^\circ$  N and  $10^\circ$  S.  
297 The correlations with the QBO at 30 hPa remain negative up to  $60^\circ$ , a consistent result among all our data sets,  
298 something also reported by Knibbe et al. (2014) with the MSR ozone data. The correlation and regression  
299 coefficients between GOME-2A and QBO are fairly similar to those found between SBUV and QBO, as well as  
300 among all data sets as seen in Table 3, despite the different periods of records.

301 These features are also evident in Figure 6 which compares GOME-2A (and GTO-ECV) satellite total ozone with  
302 GB observations with respect to the QBO. Mean differences and standard deviations between GOME-2A and GB  
303 and between GTO-ECV and GB deseasonalised total ozone data do not exceed one percent. Again, correlation  
304 coefficients between deseasonalised GOME-2A and deseasonalised GB data are highly significant in all latitude  
305 zones ( $30^\circ$ – $60^\circ$  N:  $+0.91$  (slope= $0.818$ , error= $0.035$ , t-value= $23.466$ , N= $119$ );  $10^\circ$ – $30^\circ$  N:  $+0.91$  (slope= $0.786$ ,  
306 error= $0.033$ , t-value= $23.529$ , N= $119$ );  $10^\circ$  N– $10^\circ$  S:  $+0.94$  (slope= $0.973$ , error= $0.034$ , t-value= $28.449$ , N= $109$ );  $10^\circ$ –  
307  $30^\circ$  S:  $+0.87$  (slope= $0.864$ , error= $0.044$ , t-value= $19.659$ , N= $119$ );  $30^\circ$ – $60^\circ$  S:  $+0.88$  (slope= $0.858$ , error= $0.043$ , t-  
308 value= $19.854$ , N= $119$ ). The same stands for the correlations between GTO-ECV and GB data pairs ( $30^\circ$ – $60^\circ$  N:  
309  $+0.94$ ;  $10^\circ$ – $30^\circ$  N:  $+0.89$ ;  $10^\circ$  N– $10^\circ$  S:  $+0.94$ ;  $10^\circ$ – $30^\circ$  S:  $+0.87$ ;  $30^\circ$ – $60^\circ$  S:  $+0.85$ ). Our results are in line with  
310 Eleftheratos et al. (2013) and Isaksen et al. (2014) who compared QBO-related ozone column variations from the  
311 chemical transport model Oslo CTM2 with SBUV satellite data for shorter time periods. In summary, it has been  
312 shown that GOME-2A depicts the significant effects of QBO on stratospheric ozone in concurrence with SBUV and  
313 GB measurements. The instrument captures correctly the variability of ozone in the tropics and the mid-latitudes,  
314 which is nearly in phase with the QBO in the tropics and out of phase in the northern and the southern mid-latitudes  
315 as have been shown by earlier studies (e.g. Zerefos, 1983; Baldwin et al., 2001).

### 316 3.3 Correlation with ENSO

317 Apart from the QBO, which affects the variability of total ozone in the tropics, an important mode of natural climate  
318 variability in the tropics is ENSO. To examine the impact of ENSO on total ozone in the tropics we first removed  
319 variability related to the QBO and the solar cycle, and then performed the correlation analysis with the SOI. The  
320 effect of the QBO was removed from the time series by using a linear regression model for the total ozone variations  
321 at each grid box, of the form:

$$322 \quad D(t) = a_0 + a_1 * QBO(t) + residuals(t); 0 < t \leq T \quad (1)$$

323 where  $D(t)$  is the monthly deseasonalised total ozone and  $t$  is the time in months with  $t=0$  corresponding to the initial  
 324 month and  $t=T$  corresponding to the last month. The term  $a_0$  is the intercept of the statistical model. To model QBO  
 325 we made use of the equatorial zonal winds at 30 hPa. The term  $a_1$  is the regression coefficient of QBO. The QBO  
 326 component was removed from the time series by using a phase lag with maximum correlation of 28 months (month  
 327 lag -14 to month lag 13). The QBO-related coefficients  $a_0$  and  $a_1$  of Eq. (1) for the deseasonalized GOME-2A,  
 328 GTO-ECV, TOMS/OMI/OMPS and Oslo CTM3 zonal mean data are presented in Table 3. Additional information  
 329 for the regression coefficients  $a_1$  of QBO is provided in the Supplement Figure S1, which shows the spatial  
 330 distribution of the regression coefficients in latitude-longitude maps.

331 The residuals from Eq. (1) were then inserted in a second regression (Eq. 2) to account for the effect of solar cycle  
 332 on total ozone, as follows:

$$333 \quad O_3(t) = \beta_0 + \beta_1 * F10.7(t) + residuals(t); 0 < t \leq T \quad (2)$$

334 where  $\beta_0$  and  $\beta_1$  are now the intercept and regression coefficients of solar cycle, respectively. To model the solar  
 335 cycle we used the 10.7 cm wavelength solar radio flux (F10.7) as a proxy, taken from the National Research Council  
 336 and Natural Resources Canada at [ftp://ftp.geolab.nrcan.gc.ca/data/solar\\_flux/monthly\\_averages/solflux\\_monthly\\_average.txt](ftp://ftp.geolab.nrcan.gc.ca/data/solar_flux/monthly_averages/solflux_monthly_average.txt) (last access 12  
 337 December 2018). We use the absolute solar fluxes, which are adjusted to account for variations in Earth-Sun  
 338 distance and uncertainty in antenna gain and waves reflected from the ground. Latitude-longitude maps of the  
 339 regression coefficients  $\beta_1$  of the solar cycle are presented in the Supplement Figure S2. We note that the global  
 340 pattern of the regression coefficients of solar cycle from GOME-2A data matches well with what has been shown by  
 341 Knibbe et al. (2014) with the reanalysis MSR data.

343 The remainders from Eq. (2) were used in a third regression (Eq. 3) to study the correlations between total ozone  
 344 and SOI at each individual grid box:

$$345 \quad O_3(t) = c_0 + c_1 * SOI(t) + residuals(t); 0 < t \leq T \quad (3)$$

346 where  $c_0$  and  $c_1$  are now the intercept and regression coefficients of ENSO, accordingly. Estimates of the regression  
 347 coefficients  $c_1$  are shown in the Supplement Figure S3.

348 Figure 7 presents the correlations between SOI and total ozone from GOME-2A (upper left panel), GTO-ECV  
 349 (upper right) and TOMS/OMI/OMPS satellite data (bottom left), as well as between SOI and the Oslo model  
 350 simulations (bottom right). All four plots refer to the period 2007-2016. As can be seen from Figure 7 (upper left),  
 351 correlations of  $>0.3$  between GOME-2A total ozone and SOI are found in the tropical Pacific Ocean at latitudes  
 352 between 25 deg. north and south. These correlations were tested as to their statistical significance in the period  
 353 2007-2016 using the  $t$ -test for  $R$  with  $N-2$  degrees of freedom (as in Zerefos et al., 2018), and were found to be  
 354 statistical significant. A similar picture of correlation coefficients is also observed by the GTO-ECV and  
 355 TOMS/OMI/OMPS data. Both data sets show similar results as to the range of correlations ( $>0.3$ ) in the tropical  
 356 Pacific for the common period of observations. Nevertheless, the spatial resolution is higher in the GOME-2A and

357 GTO-ECV (1x1 deg.) data than in the TOMS/OMI/OMPS (5x5 deg.) data, so the former data sets perform better  
358 when looking at smaller space scales. We have to note here that in both maps there are larger areas with correlation  
359 coefficients  $>0.3$  in the southern part of the tropics than in the northern part. However, this was mostly observed  
360 during the period 2007-2016. By examining the longer-term data record of the TOMS/OMI/OMPS data which  
361 extend back to 1979, we find symmetry in the pattern of correlations north and south of the equator in the tropical  
362 Pacific Ocean (Figure A1 of Appendix A), which indicates that both sides of the tropical Pacific are affected more  
363 or less in a similar way by El Niño/La Niña events. Finally, the Oslo CTM3 gives small correlations ( $<0.3$ ) in the  
364 tropical Pacific Ocean around the equator, except over the northern and southern subtropics where the model  
365 compares better with the observations.

366 The small rectangle in Figure 7 corresponds to the South Pacific region ( $10^{\circ}$ - $20^{\circ}$  S,  $180^{\circ}$ - $220^{\circ}$  E) and the blue cross  
367 to the station Samoa (American Samoa;  $14.25^{\circ}$  S,  $189.4^{\circ}$  E), at which total ozone has been studied as for the impact  
368 of ENSO after removing variability related to the annual cycle, QBO and the solar cycle. Figure 8 shows an example  
369 of the ENSO impact on total ozone in the South Pacific Ocean. The upper panel shows the time series of total ozone  
370 anomalies from GOME-2A, GTO-ECV and TOMS/OMI/OMPS satellite data together with the SOI (Figure 8a).  
371 Comparisons of GOME-2A data with GTO-ECV data, SBUV overpass data and GB measurements at the station  
372 Samoa are shown in Figure 8b. The dotted line shows the respective tropopause pressure anomalies from NCEP  
373 reanalysis. All data sets point to the strong influence of ENSO on total ozone. Most evident is the strong decrease of  
374 about 4% in 1997/98 which was caused by the strongest El Niño event in the examined period. A strong decrease is  
375 also observed in the tropopause pressures by NCEP. Notable also is the strong La Niña event in 2010 which caused  
376 total ozone to increase by about 4%. We calculate a strong correlation between total ozone from GTO-ECV and SOI  
377 of  $+0.66$  (99% confidence level), which accounts for about 40% of the variability of total ozone over the tropical  
378 Pacific Ocean when the annual cycle, QBO signal and solar cycle are removed. From the regression with SOI we  
379 estimated an ENSO-related term from which we calculated the amplitude of ENSO in total ozone as [maximum  
380 ozone - minimum ozone]/2. The amplitude of ENSO in total ozone was estimated to be 8.8 DU or 3.5% of the  
381 annual mean. This is comparable to the amplitude of annual cycle (7.7 DU or 3.0% of the mean) and larger than the  
382 amplitude of QBO (2.2 DU or 0.8% of the mean) and the amplitude of solar cycle in this region (4.1 DU or 1.6% of  
383 the mean). These results are based on the GTO-ECV total ozone data. Similar results were also found at the station  
384 Samoa from GB observations (i.e. correlation with SOI:  $+0.55$ , amplitude of ENSO: 7.7 DU or 3.0% of the mean,  
385 amplitude of annual cycle: 6.7 DU or 2.7% of the mean). Statistics of total ozone such as mean, amplitude of annual  
386 cycle, amplitude of QBO, amplitude of solar cycle and amplitude of ENSO in total ozone over the selected areas are  
387 presented in Table 4. Both satellite, GB and model data show consistent results. It also appears that the station  
388 Samoa represents well the greater area in the Southern Pacific as to the impact of ENSO.

389 Differences between GOME-2A and its data pairs in the southern Pacific Ocean are the order of  $-0.2 \pm 1.0\%$   
390 between GOME-2A and TOMS/OMI/OMPS data,  $-0.3 \pm 0.9\%$  between GOME-2A and GTO-ECV, and  $-0.9 \pm 1.8\%$   
391 between GOME-2A and Oslo CTM3. Accordingly, differences at Samoa are:  $-0.6 \pm 1.9\%$  between GOME-2A and  
392 GB data,  $0.0 \pm 1.4\%$  between GOME-2A and GTO-ECV, and  $-0.1 \pm 1.3\%$  between GOME-2A and SBUV. Despite  
393 the small differences found, we note here that GOME-2A values in the last 4 years of Figures 8 and 9 slightly

394 deviate from the other data sets, and correlate weaker with SOI than the other years in the time series. For instance,  
395 we estimate a drop in the correlation coefficient between GOME-2A and SOI at the station Samoa (+0.58 in the  
396 period 2007-2012 and +0.47 in the period 2007-2016), which nevertheless does not alter the statistical significance  
397 of the correlation.

398 From Figure 8 it also appears that there are high correlations with the tropopause height. The correlation coefficient  
399 between the NCEP tropopause pressure and GOME-2A total ozone over the South Pacific Ocean is of the order of  
400 +0.59 (Student's t-test statistics results: t-value = 7.946, p-value <0.0001, N = 119). Accordingly, the correlation  
401 with GTO-ECV ozone data is the order of +0.64 (t-value = 13.165, p-value <0.0001, N = 252) and with  
402 TOMS/OMI/OMPS the order of +0.58 (t-value = 10.913, p-value <0.0001, N = 241). The high correlation between  
403 the tropopause pressure and total ozone on interannual and longer time scales points to the very strong link between  
404 these parameters. These links were already documented in the past (e.g. Steinbrecht et al., 1998, 2001) and are  
405 verified with the GOME-2A data. At the same time a strong correlation is also evident between tropopause pressure  
406 and SOI, again on interannual and longer time scales ( $R = +0.66$ , t-value = 13.825, p-value <0.0001, N = 252). The  
407 above results point to the strong impact of ENSO on the tropical ozone column through the tropical tropopause;  
408 warm (El Niño) and cold (La Niña) events affect the variability of the tropopause which in turn affects the  
409 distribution of stratospheric ozone. In the tropics, where total ozone is mainly stratospheric, as the tropopause moves  
410 to higher altitudes (lower pressure), the stratosphere is compressed, reducing the amount of stratospheric (total)  
411 ozone. This happens during warm (El Niño) episodes. The opposite phenomenon occurs during cold (La Niña)  
412 events when the tropopause height decreases (higher pressure) and total ozone is then increased. These events can  
413 affect the long-term ozone trends in the tropics when looking at time periods when strong El Niño and La Niña  
414 events occur at the beginning and the end of the trend period respectively (Coldewey-Egbers et al., 2014).

415 Furthermore, in Figure 8 we have marked 7 stations in the greater South Asia region ( $35^{\circ}$ - $45^{\circ}$  N,  $45^{\circ}$ - $125^{\circ}$  E) where  
416 total ozone is anti-correlated with the SOI. Admittedly, these anti-correlations are weak (about -0.3) but we thought  
417 worthwhile presenting the time series in these areas as well. Figure 9 shows the variability of total ozone after  
418 removing seasonal, QBO and solar cycle related variations, over the South Asia region (upper panel) and over the 7  
419 stations averaged within this region (lower panel). As can be seen from this figure, the explained variance by ENSO  
420 is small, not exceeding 9%. All correlations from the comparisons with the SOI are summarized in Table 5. In spite  
421 the small correlations with the SOI, the consistency between GOME-2A, GTO-ECV, TOMS/OMI/OMPS and Oslo  
422 CTM3 data anomalies is very high and their differences are within  $\pm 1\%$ . Differences at the 7 stations in South Asia  
423 are as follows:  $-1.3 \pm 2.4\%$  between GOME-2A and GB data,  $-0.4 \pm 1.0\%$  between GOME-2A, and GTO-ECV and -  
424  $0.5 \pm 1.0\%$  between GOME-2A and SBUV.

425 In summary, our findings indicate that GOME-2A captures well the disturbances in total ozone during ENSO events  
426 with respect to satellite SBUV and GB observations. Our findings on the ENSO-related total ozone variations (low  
427 ozone during ENSO warm events, high ozone during ENSO cold events, and magnitude of changes) are in line with  
428 recent studies (e.g. Randel and Thompson, 2011; Oman et al., 2013, Sioris et al., 2014) included in the 2014 Ozone

429 Assessment report (Pawson et al., 2014; WMO, 2014). Our results are also in agreement with Knibbe et al. (2014)  
430 who showed negative ozone effects of El Niño between 25° S and 25° N, especially over the Pacific.

### 431 **3.4 Correlation with NAO**

432 The residuals from Eq. (3), free from seasonal, QBO, solar and ENSO related variations, were later used to study the  
433 correlation between total ozone and NAO in winter. The results are presented in Figure 10 which shows the  
434 correlation coefficients between total ozone and NAO index in winter from the GOME-2A (upper left), GTO-ECV  
435 (upper right) and TOMS/OMI/OMPS satellite data (lower left), and the Oslo CTM3 model calculations (lower  
436 right). Negative correlations between total ozone and NAO are presented with blue colours while positive  
437 correlations with red colours. From Figure 10 (upper left) it appears that total ozone is strongly correlated with NAO  
438 in many regions. Strong negative correlation coefficients are observed in the majority of the northern mid-latitudes  
439 (R about  $-0.6$ ) while positive correlations exist in the tropics and some negative correlations in the southern mid-  
440 latitudes. These characteristics are observed in both GTO-ECV and TOMS/OMI/OMPS datasets and are reproduced  
441 by the Oslo model as well, all for the common period 2007-2016. The regression coefficients on these comparisons  
442 are presented in the Supplement Figure S4.

443 We note here that the results of the correlation analysis for the period 2007-2016 were based on a relative small  
444 sample of data from 10 winters and therefore many of these correlation coefficients may not be statistically  
445 significant. The statistical significance of the correlation coefficients in every grid box was tested only with the  
446 TOMS/OMI/OMPS data (Figure A2, Appendix A), which provided us the opportunity to calculate the respective  
447 correlations using data for the whole period of record 1979-2016. It appears that when extending the data back to the  
448 1980's the negative correlations in the southern mid-latitudes in winter disappear while the positive correlations in  
449 the tropics become weaker; yet the observed anti-correlation between total ozone and NAO index in the northern  
450 mid-latitude zone holds strong. The dotted line in the plot shows areas with statistically significant correlation  
451 coefficients (99% confidence level). Indeed, in the long-term, statistically significant correlations between total  
452 ozone and the NAO index during winter are mostly found over the northern mid-latitudes and the sub-tropics. A  
453 small, statistically significant signal is also seen over Antarctica but it was not analysed further.

454 According to this finding we have restricted the analysis of NAO to the northern mid-latitudes. Rectangles (Figure  
455 10, upper left) correspond to two regions in the North Atlantic, i.e., 35°-50° N, 20°-50° W and 15°-27° N, 30°-60° W  
456 respectively, which were studied for the impact of NAO on total ozone after removing variability related to the  
457 annual cycle, QBO, solar cycle and ENSO. In addition we have studied a number of stations in Canada, USA, and  
458 Europe contributing ozone data to WOUDC, which are marked by red and green crosses in Figure 10. The red  
459 crosses refer to the monitoring stations in Canada and the US, and the green crosses to the stations in Europe. In  
460 Figure 11 we present the times series of total ozone anomalies from GOME-2A, GTO-ECV and TOMS/OMI/OMPS  
461 satellite data along with the NAO index in winter over the North Atlantic. Model calculations are shown as well.  
462 The dotted line shows the respective tropopause pressure anomalies from NCEP reanalysis. Comparisons between  
463 GOME-2A, GTO-ECV, SBUV (v8.6) overpass data and GB measurements over the various stations in Canada,  
464 USA and Europe are shown in Figure 12.

465 The observed anomalies over the North Atlantic Ocean point to the strong influence of NAO on total ozone in  
466 winter. Most evident is the strong increase in total ozone in 2010 of more than 8% particularly over 35°-50° N and  
467 20°-50° W. This increase was accompanied by a strong increase in tropopause pressures. Both changes (in total  
468 ozone and tropopause pressures) occurred under a strong negative phase of NAO, the strongest one in the past 20  
469 years. We observe strong anti-correlation among total ozone and NAO index in winter ( $R = -0.74$  over 35°-50° N,  
470 20°-50° W), which is statistically significant at the 99% confidence level. This anti-correlation suggests that about  
471 50% of the variability of total ozone in winter is explained by NAO when the annual cycle, QBO, solar cycle and  
472 ENSO signals are removed. Differences for GOME-2A and its data pairs are estimated to be  $-0.7 \pm 1.1\%$  between  
473 GOME-2A and TOMS/OMI/OMPS data,  $+0.1 \pm 1.0\%$  between GOME-2A and GTO-ECV, and  $-0.2 \pm 1.5\%$   
474 between GOME-2A and Oslo CTM3. From the regression with the NAO index we derived a NAO-related term  
475 from which we calculated the amplitude of NAO in total ozone as [maximum ozone - minimum ozone]/2. The  
476 amplitude of NAO over the North Atlantic region (35°-50° N, 20°-50° W) was estimated to be about 16.5 DU or  
477 5.2% of the annual mean. This is about half of the amplitude of the annual cycle (which is ~37 DU or 11.7% of the  
478 mean). These estimates are based on GTO-ECV data. Similar correlation and amplitude were also found with  
479 GOME-2A, the combined TOMS/OMI/OMPS satellite data and the Oslo CTM3 model simulations.

480 A similar but opposite correlation is found over the southern part of the North Atlantic (15°-27° N, 30°-60° W).  
481 Here, we estimate a significant correlation coefficient with NAO of +0.60, amplitude of NAO of about 7.2 DU  
482 (2.6% of the annual mean) and amplitude of annual cycle of about 15.8 DU (5.7% of the mean). Again, similar  
483 estimates are found with the GOME-2A and the TOMS/OMI/OMPS satellite data and reproduced by the model  
484 calculations as well. The annual mean total ozone and the amplitudes of annual cycle, QBO, solar cycle and NAO in  
485 total ozone over the studied regions in the North Atlantic are summarised in Table 6. Differences between GOME-  
486 2A and GTO-ECV data at the southern part of North Atlantic are the order of  $-0.6 \pm 0.7\%$ . Differences with the  
487 TOMS/OMI/OMPS data are estimated to be  $-0.9 \pm 0.8\%$ , and with the Oslo CTM3  $-0.1 \pm 0.7\%$ .

488 The time series of total ozone anomalies and of the NAO index for the examined stations in Canada, USA and  
489 Europe are presented in Figure 12. Table 7 presents the respective statistics. The correlation between total ozone and  
490 the NAO index in winter after removing from ozone variability related to the annual cycle, QBO, solar cycle and  
491 ENSO is  $-0.40$  (90% confidence level). Again, a particular feature was the total ozone increase in 2010 by 6% of the  
492 mean associated with the negative NAO phase. Noteworthy on this increase is the consistency with the GB  
493 measurements and the satellite SBUV overpass data, and in general the agreement found between the variability of  
494 the tropopause pressures and total ozone. Differences between GOME-2A and GB data are  $-1.0 \pm 1.8\%$ .  
495 Accordingly we estimate differences of about  $-1.1 \pm 0.5\%$  between GOME-2A and GTO-ECV data and of about -  
496  $1.3 \pm 0.6\%$  between GOME-2A and SBUV data. On the basis of GTO-ECV data we estimate that in Canada and  
497 USA, the amplitude of NAO in total ozone in winter is about 7 DU (or 2.2% of the mean), while it is estimated to be  
498 about 9 DU (or 2.7% of the mean) over Europe. These numbers are slightly smaller than the GOME-2A, GB and  
499 SBUV estimates, less than about one percent (Table 7).

500 The anti-correlation between total ozone column and the NAO index during winter also applies to southern Europe  
501 and the Mediterranean. Following the study of Ossó et al. (2011) who reported a reversal in the correlation pattern  
502 between NAO and total ozone from winter to summer in southern Europe, we have looked at the correlations during  
503 summer as well. Figure 13 presents the comparisons for 21 ground-based stations located in the region bounded by  
504 latitudes 30°-47° N and by longitudes 10°W-40°E. Figure 13a shows results for the summer and Figure 13b shows  
505 results for winter. As evident, the observed anti-correlation between GB total ozone and NAO in winter ( $R = -0.43$ ,  
506 slope =  $-0.980$ , t-value =  $-2.095$ , p-value =  $0.0499$ ,  $N = 21$ ) reverses sign and becomes positive in the summer ( $R =$   
507  $+0.60$ , slope =  $0.874$ , t-value =  $3.309$ , p-value =  $0.0037$ ,  $N = 21$ ), indicating that the NAO explains about 36% of ozone  
508 variability in the summer in this region. A similar picture is also seen from GOME-2A, GTO-ECV and SBUV data.

509 In summary, our findings based on GOME-2A, GTO-ECV and SBUV overpass data are in line with those found by  
510 Ossó et al. (2011) and Steinbrecht et al. (2011) who analysed TOMS and OMI satellite data, and GB measurements  
511 at the station Hohenpeissenberg, respectively. During winter, total ozone variability associated with the NAO is  
512 particularly important over northern Europe, the U.S. East Coast, and Canada, explaining up to 30% in total ozone  
513 variance for this region (Ossó et al., 2011). Also, both studies found unusually high total ozone columns in 2010  
514 over much of the Northern Hemisphere and related them to the negative phase of NAO or AO (the Arctic  
515 Oscillation).

#### 516 **4 Conclusions**

517 We have evaluated the ability of GOME-2/MetopA (GOME-2A) satellite total ozone retrievals to capture known  
518 natural oscillations such as the QBO, ENSO and NAO. In general, GOME-2A depicts these natural oscillations in  
519 concurrence with GTO-ECV, TOMS/OMI/OMPS, SBUV (v8.6) satellite overpass data, ground-based  
520 measurements (Brewer, Dobson, filter and SAOZ) and chemical transport model calculations (Oslo CTM3).

521 Mean differences between GOME-2A and SBUV total ozone were found to be  $+0.1 \pm 0.7\%$  in the tropics (0-30  
522 deg.), about  $+0.8 \pm 1.6\%$  in mid-latitudes (30-60 deg.), about  $+1.3 \pm 2.2\%$  over the northern high latitudes (60-80  
523 deg. N) and about  $-0.5 \pm 2.9\%$  over the southern high latitudes (60-80 deg. S). These differences were estimated as  
524  $[GOME-2A - SBUV] / SBUV$  (%) from January 2007 to December 2016. Small differences were also found  
525 between GOME-2A and GB measurements, with standard deviations of the differences being  $\pm 1.4\%$  in the tropics,  
526  $\pm 2.1\%$  in mid-latitudes, and  $\pm 3.2\%$  and  $\pm 4.3\%$  over the northern and the southern high latitudes respectively.

527 The variability of total ozone from GOME-2A has been compared with the variability of total ozone from other  
528 examined data sets as to their agreement depicting natural atmospheric phenomena such as the QBO, ENSO and  
529 NAO. First, we studied correlations between total ozone and the QBO after removing from the ozone data sets  
530 variability related to the seasonal cycle. Then, we examined correlations between total ozone and ENSO, after  
531 removing variability related to the QBO and solar cycle, and finally correlations with the NAO after removing  
532 variability related to the QBO, solar cycle and ENSO. Our main results are as follows:

533 **QBO:** Total ozone from GOME-2A is well correlated with the Quasi-Biennial Oscillation (+0.8 in the tropics) in  
534 agreement with GTO-ECV, SBUV and GB data. The amplitude of QBO on total ozone maximizes around the  
535 equator and it is estimated to about 2.6% of the mean. Going from low to mid-latitudes there is a phase shift in the  
536 QBO impact on total ozone. Correlation coefficients between GOME-2A total ozone and the QBO over 30-60 deg.  
537 north and south are -0.1 and -0.5 respectively, in agreement with the correlations between GB total ozone and the  
538 QBO (-0.2 and -0.5, accordingly). On the basis of GOME-2A, the amplitude of QBO in total ozone is estimated to  
539 be 0.6% of the mean in the northern mid-latitudes and 1.4% of the mean in the southern mid-latitudes.

540 **ENSO:** Correlation coefficients among GOME-2A total ozone and SOI in the tropical Pacific Ocean are estimated  
541 to be about +0.6, consistent with GTO-ECV, SBUV and GB observations. It was found that the El Niño Southern  
542 Oscillation (ENSO) signal is evident and consistent in all examined datasets. The amplitude of ENSO in total ozone  
543 is about 6–9 DU corresponding to about 2.5–3.5% of the annual mean. Differences between GOME-2A, GTO-ECV  
544 and GB measurements during warm (El Niño) and cold (La Niña) events are within  $\pm 1.5\%$ . Similar estimates also  
545 result from the Dobson measurements at American Samoa, indicating that Samoa station represents well the greater  
546 area in the Southern Pacific for satellite evaluations as to the impact of ENSO.

547 **NAO:** The respective results as far as the impact of the North Atlantic Oscillation over the northern mid-latitudes  
548 showed a clear NAO signal in winter in all data sets, with amplitudes of about 16-19 DU (about 5–6% of the annual  
549 mean) in the North Atlantic, 9-12 DU (3-4% of the mean) over Europe, and 7-10 DU (2-3% of the mean) over  
550 Canada and the US. Comparison with GB observations over Canada and Europe showed very good agreement  
551 between GOME-2A, GTO-ECV and GB observations as to the influence by NAO, with differences within  $\pm 1\%$ .

552 In addition to the usual validation methods, which compare monthly mean and zonal mean total ozone data and  
553 analyse the differences between satellite and GB instruments, we showed here that quasi cyclical perturbations such  
554 as the QBO, ENSO and NAO can serve as independent proxies of spatiotemporal variation to qualitatively evaluate  
555 GOME-2A satellite total ozone against ground-based and other satellite total ozone data sets. The agreement and  
556 small differences which were found between the variability of total ozone from GOME-2A and the variability of  
557 total ozone from other satellite retrievals and ground-based measurements during these naturally-occurring  
558 oscillations verify the good quality of GOME-2A satellite total ozone to be used in ozone-climate research studies.

#### 559 **Data availability**

560 Satellite SBUV (v8.6) total ozone station overpass data were downloaded from [https://acd-](https://acd-ext.gsfc.nasa.gov/Data_services/merged/index.html)  
561 [ext.gsfc.nasa.gov/Data\\_services/merged/index.html](https://acd-ext.gsfc.nasa.gov/Data_services/merged/index.html) (last access: 15 June 2018) (McPeters et al., 2013; Bhartia et al.,  
562 2013). GTO-ECV total ozone data are available at <http://www.esa-ozone-cci.org/?q=node/160> (last access: 15 June  
563 2018) (Coldewey-Egbers et al., 2015; Garane et al., 2018). Ground-based total ozone daily summaries were obtained  
564 from the World Ozone and UV Data Centre (WOUDC) at  
565 [https://woudc.org/archive/Summaries/TotalOzone/Daily\\_Summary/](https://woudc.org/archive/Summaries/TotalOzone/Daily_Summary/) (last access: 15 June 2018). The QBO  
566 component on total ozone was examined by using the monthly mean zonal winds at Singapore at 30 hPa. Zonal



567 wind data at 30 hPa were provided by the Freie Universität Berlin (FU-Berlin) at [http://www.geo.fu-](http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/qbo.dat)  
568 [berlin.de/met/ag/strat/produkte/qbo/qbo.dat](http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/qbo.dat) (last access: 15 June 2018) (Naujokat, 1986). The Southern Oscillation  
569 Index (SOI) was provided by the Bureau of Meteorology of the Australian Government at  
570 <http://www.bom.gov.au/climate/current/soi2.shtml> (Australian Government – Bureau of Meteorology, 2018). The  
571 NAO index for December, January and February was provided by the Climate Analysis Section, NCAR, Boulder,  
572 USA at <https://climatedataguide.ucar.edu/climate-data/hurrell-north-atlantic-oscillation-nao-index-pc-based> (last  
573 access: 15 June 2018) (Hurrell and Deser, 2009). The tropopause pressures from the NCEP/NCAR reanalysis 1 data  
574 set were downloaded from <https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.tropopause.html> (last  
575 access: 15 June 2018) (Kalnay et al., 1996).

## 576 **Competing interests**

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

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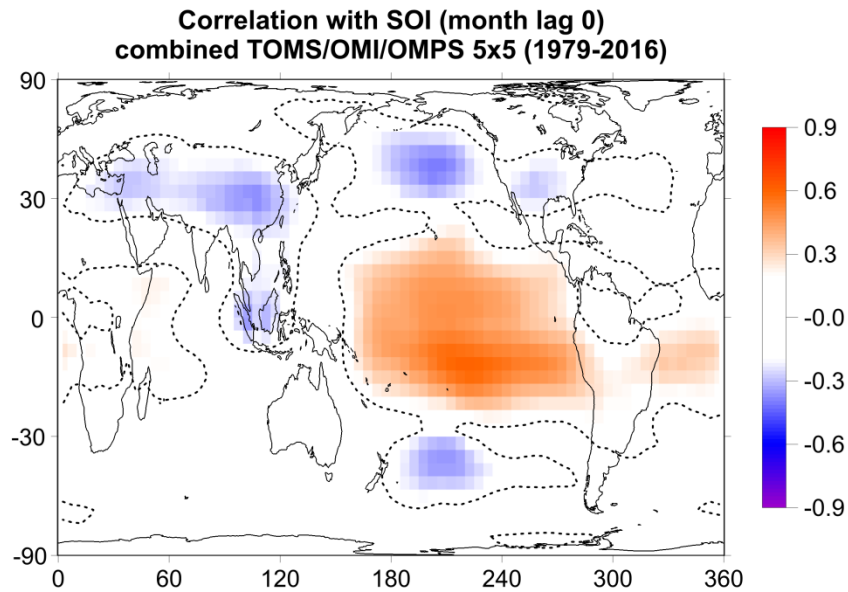
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761 Appendix A

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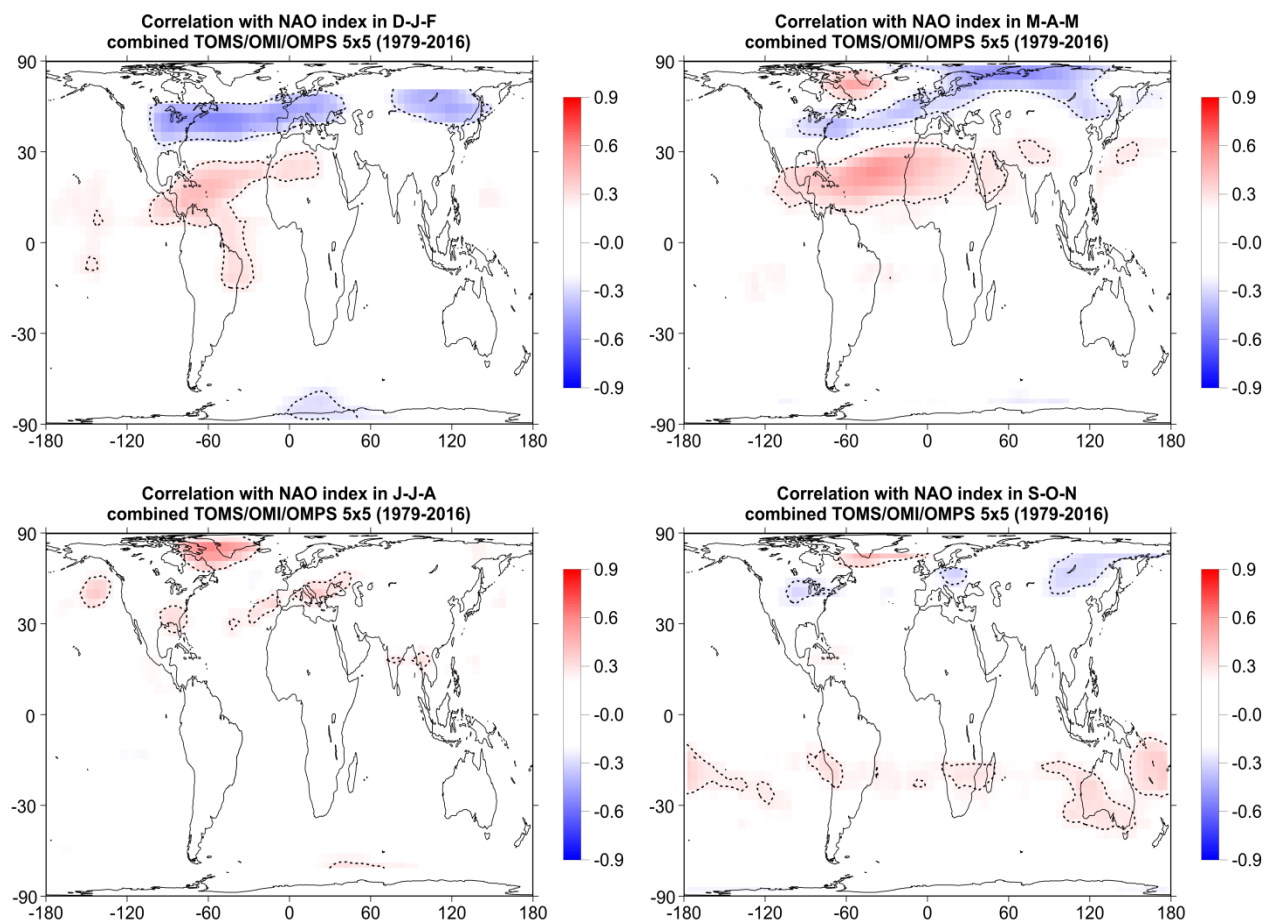
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765 **Figure A1. Map of correlation coefficients between total ozone from TOMS/OMI/OMPS satellite data and**  
766 **SOI for the whole period 1979-2016, after removing variability related to the seasonal cycle, QBO and solar**  
767 **cycle. The dotted line bounds the regions where the correlation coefficients are statistically significant at the**  
768 **99% confidence level (t-test). Only correlation coefficients above/below  $\pm 0.2$  are shown. Ozone data for the**  
769 **period 1991-1993 after the Mt Pinatubo eruption were not used in the correlation analysis to avoid any data**  
770 **contamination by the volcanic aerosols.**

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774

775 **Figure A2. Map of correlation coefficients between total ozone from TOMS/OMI/OMPS satellite data and the**  
 776 **NAO index during winter (December, January, February (D-J-F); upper left), spring (March, April, May (M-**  
 777 **A-M); upper right), summer (June, July, August (J-J-A); lower left) and autumn (September, October,**  
 778 **November (S-O-N); lower right) for the whole period 1979-2016, after removing variability related to the**  
 779 **seasonal cycle, QBO, solar cycle and ENSO. The dotted line bounds the regions where the correlation**  
 780 **coefficients are statistically significant at the 99% confidence level (t-test). Only correlation coefficients**  
 781 **above/below  $\pm 0.2$  are shown. Ozone data for the period 1991-1993 after the Mt Pinatubo eruption were not**  
 782 **used in the correlation analysis to avoid any data contamination by the volcanic aerosols.**

783

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785

786 **Table 1. Mean differences and their standard deviations in percent between total ozone from GOME-2A,**  
 787 **SBUV (v8.6) satellite overpass data and ground-based observations over different latitude zones, as shown in**  
 788 **Figures 1 and 2.**

789

	[GOME-2A – SBUV] / SBUV (%) Stations mean data	[GOME-2A – GROUND] / GROUND (%) Stations mean data
60°-80° N	+1.3 ± 2.2	+2.5 ± 3.2
30°-60° N	+0.8 ± 1.6	+0.1 ± 1.9
0°-30° N	0.0 ± 0.7	-0.5 ± 1.2
0°-30° S	+0.1 ± 0.7	-0.9 ± 1.6
30°-60° S	+0.9 ± 1.6	0.0 ± 2.4
60°-80° S	-0.5 ± 2.9	0.0 ± 4.3

790

791

792 **Table 2. Statistics of the correlations shown in Figures 1 and 2 between total ozone from a) GOME-2A data**  
 793 **and SBUV (v8.6) overpass data, and b) GOME-2A data and ground-based measurements.**

794

(a) GOME-2A and SBUV (v8.6)	Correlation	Intercept (DU)	Slope*	Error	t-value	p-value	N
60°-80° N	+0.987	4.925	0.999	0.015	65.224	<0.0001	117
30°-60° N	+0.984	5.002	0.993	0.017	59.784	<0.0001	118
0°-30° N	+0.989	28.304	0.894	0.012	72.404	<0.0001	118
0°-30° S	+0.981	21.575	0.919	0.017	53.874	<0.0001	118
30°-60° S	+0.977	-4.198	1.023	0.021	49.123	<0.0001	118
60°-80° S	+0.974	2.944	0.984	0.025	39.985	<0.0001	88

795

(b) GOME-2A and Ground-based	Correlation	Intercept (DU)	Slope*	Error	t-value	p-value	N
60°-80° N	+0.973	7.651	1.002	0.022	45.155	<0.0001	118
30°-60° N	+0.977	15.772	0.952	0.019	49.671	<0.0001	119
0°-30° N	+0.982	49.534	0.810	0.014	56.951	<0.0001	119
0°-30° S	+0.916	56.520	0.778	0.032	24.655	<0.0001	119
30°-60° S	+0.946	12.423	0.958	0.030	31.612	<0.0001	119
60°-80° S	+0.939	0.405	0.999	0.039	25.439	<0.0001	89

796 \* Error, t-value and p-value refer to slope.

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799

800 **Table 3. Statistics of correlations between deseasonalized total ozone and the QBO at 30 hPa for a) GOME-**  
 801 **2A data, b) GTO-ECV data, c) SBUV (v8.6) overpass data, and d) ground-based measurements.**

802

(a) GOME-2A and QBO	Correlation	Intercept (%)	Slope*	Error	t-value	p-value	N
30°-60° N	-0.073	-0.045	-0.008	0.010	-0.791	0.4307	119
10°-30° N	-0.099	-0.048	-0.008	0.008	-1.077	0.2835	119
10° N-10° S	+0.767	0.654	0.114	0.009	12.910	<0.0001	119
10°-30° S	-0.472	-0.273	-0.048	0.008	-5.799	<0.0001	119
30°-60° S	-0.424	-0.262	-0.046	0.009	-5.063	<0.0001	119

803

(b) GTO-ECV and QBO	Correlation	Intercept (%)	Slope*	Error	t-value	p-value	N
30°-60° N	-0.116	-0.090	-0.012	0.007	-1.869	0.0628	259
10°-30° N	-0.142	-0.100	-0.014	0.006	-2.293	0.0226	259
10° N-10° S	+0.779	0.705	0.109	0.005	19.949	<0.0001	259
10°-30° S	-0.484	-0.306	-0.046	0.005	-8.873	<0.0001	259
30°-60° S	-0.417	-0.312	-0.048	0.007	-7.345	<0.0001	259

804

(b) SBUV v(8.6) and QBO	Correlation	Intercept (%)	Slope*	Error	t-value	p-value	N
30°-60° N	-0.165	-0.112	-0.018	0.007	-2.694	0.0075	262
10°-30° N	-0.177	-0.114	-0.018	0.006	-2.901	0.0040	263
10° N-10° S	+0.748	0.648	0.104	0.006	18.223	<0.0001	263
10°-30° S	-0.488	-0.287	-0.046	0.005	-9.037	<0.0001	263
30°-60° S	-0.458	-0.328	-0.051	0.006	-8.333	<0.0001	263

805

(b) Ground-based and QBO	Correlation	Intercept (%)	Slope*	Error	t-value	p-value	N
30°-60° N	-0.158	-0.123	-0.017	0.007	-2.594	0.0100	264
10°-30° N	-0.142	-0.083	-0.016	0.007	-2.317	0.0213	264
10° N-10° S	+0.695	0.553	0.095	0.006	15.327	<0.0001	253
10°-30° S	-0.490	-0.268	-0.046	0.005	-9.091	<0.0001	264
30°-60° S	-0.431	-0.322	-0.048	0.006	-7.734	<0.0001	264

806 \* The slope is in % per unit change of the explanatory variable. Error, t-value and p-value refer to slope.

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809

810 **Table 4. Annual mean total ozone, amplitude of annual cycle, amplitude of QBO, amplitude of solar cycle and amplitude of ENSO in the period 1995-**  
 811 **2016 from GOME-2A, GTO-ECV, the combined TOMS/OMI/OMPS satellite data and Oslo CTM3 model calculations over the South Pacific region**  
 812 **(10°-20° S, 180°-220° E) and at station Samoa (14.25° S, 189.4° E) located within this region.**

813

	South Pacific Ocean				station Samoa			
	GOME-2A*	GTO-ECV	TOMS/OMI/OMPS	Oslo CTM3	GOME-2A*	GTO-ECV	GROUND	SBUV (v8.6)
Annual mean	255.3 DU	254.7 DU	253.0 DU	259.5 DU	252.7 DU	252.2 DU	249.2 DU	251.9 DU
Amplitude of annual cycle	7.4 DU (2.9%)	7.7 DU (3.0%)	7.3 DU (2.9%)	5.2 DU (2.0%)	7.1 DU (2.8%)	6.7 DU (2.7%)	6.7 DU (2.7%)	7.3 DU (2.9%)
Amplitude of QBO	2.7 DU (1.0%)	2.2 DU (0.9%)	2.4 DU (0.9%)	2.3 DU (0.9%)	3.0 DU (1.2%)	2.2 DU (0.9%)	2.7 DU (1.1%)	2.0 DU (0.8%)
Amplitude of solar cycle	2.1 DU (0.8%)	4.1 DU (1.6%)	4.6 DU (1.8%)	1.8 DU (0.7%)	2.0 DU (0.8%)	4.5 DU (1.8%)	1.6 DU (0.6%)	4.5 DU (1.8%)
Amplitude of ENSO	6.2 DU (2.4%)	8.8 DU (3.5%)	6.0 DU (2.4%)	8.8 DU (3.4%)	5.6 DU (2.2%)	7.7 DU (3.0%)	5.5 DU (2.2%)	7.5 DU (3.0%)

814 \*period 2007-2016

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817

818 **Table 5. Statistics of the comparisons between total ozone, tropopause pressures and SOI for a) South Pacific**  
 819 **(10°-20° S, 180°-220° E), b) station Samoa (14.25° S, 189.4° E), c) South Asia (35°-45° N, 45°-125° E) and d) 7**  
 820 **stations in South Asia.**

821

(a) South Pacific	Correlation with SOI	Intercept (%)	Slope*	Error	t-value	p-value	N
GOME-2A	+0.56	-0.238	0.118	0.016	7.236	<0.0001	119
GTO-ECV	+0.66	-0.069	0.145	0.010	14.014	<0.0001	252
TOMS/OMI/OMPS	+0.62	-0.139	0.134	0.011	12.285	<0.0001	241
Oslo CTM3	+0.55	-0.064	0.144	0.014	10.501	<0.0001	252
Tropopause	+0.66	-0.761	0.241	0.017	13.825	<0.0001	252

822

(b) Samoa	Correlation with SOI	Intercept (%)	Slope*	Error	t-value	p-value	N
GOME-2A	+0.47	-0.217	0.108	0.018	5.823	<0.0001	119
GTO-ECV	+0.55	-0.100	0.127	0.012	10.366	<0.0001	252
SBUV overpass	+0.59	-0.114	0.127	0.011	11.398	<0.0001	251
GB (WOUDC)	+0.42	-0.058	0.106	0.017	6.194	<0.0001	178
Tropopause	+0.65	-0.799	0.223	0.017	13.405	<0.0001	252

823

(c) South Asia	Correlation with SOI	Intercept (%)	Slope*	Error	t-value	p-value	N
GOME-2A	-0.23	0.090	-0.044	0.018	-2.525	0.0129	119
GTO-ECV	-0.30	0.073	-0.074	0.015	-5.047	<0.0001	252
TOMS/OMI/OMPS	-0.28	-0.212	-0.073	0.016	-4.553	<0.0001	241
Oslo CTM3	-0.18	0.140	-0.040	0.014	-2.877	0.0044	252
Tropopause	-0.27	-0.188	-0.129	0.029	-4.476	<0.0001	252

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(d) South Asia (7 stations mean)	Correlation with SOI	Intercept (%)	Slope*	Error	t-value	p-value	N
GOME-2A	-0.23	0.090	-0.043	0.017	-2.518	0.0132	119
GTO-ECV	-0.30	0.067	-0.072	0.014	-5.040	<0.0001	252
SBUV overpass	-0.27	0.086	-0.066	0.015	-4.464	<0.0001	251
GB (WOUDC)	-0.36	0.427	-0.103	0.017	-5.912	<0.0001	240
Tropopause	-0.28	-0.122	-0.160	0.035	-4.597	<0.0001	252

825 \* The slope is in % per unit change of the explanatory variable. Error, t-value and p-value refer to slope.

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829 **Table 6. Annual mean total ozone, amplitude of annual cycle, amplitude of QBO, amplitude of solar cycle and amplitude of NAO in the period 1995-**  
 830 **2016 from GOME-2A, GTO-ECV, the combined TOMS/OMI/OMPS satellite data and Oslo CTM3 model calculations over the North Atlantic Ocean:**  
 831 **(a) region 35°-50° N, 20°-50° W, and (b) region 15°-27° N, 30°-60° W.**

832

	North Atlantic Ocean							
	(a) 35°-50° N, 20°-50° W				(b) 15°-27° N, 30°-60° W			
	GOME-2A*	GTO-ECV	TOMS/OMI/OMPS	Oslo CTM3	GOME-2A*	GTO-ECV	TOMS/OMI/OMPS	Oslo CTM3
Annual mean	319.7 DU	315.9 DU	317.3 DU	311.2 DU	276.6 DU	276.4 DU	274.4 DU	282.6 DU
Amplitude of annual cycle	37.4 DU (11.7%)	37.0 DU (11.7%)	36.9 DU (11.6%)	32.0 DU (10.3%)	12.7 DU (4.6%)	15.8 DU (5.7%)	15.1 DU (5.5%)	15.5 DU (5.5%)
Amplitude of QBO	2.5 DU (0.8%)	2.3 DU (0.7%)	2.6 DU (0.8%)	3.2 DU (1.0%)	3.0 DU (1.1%)	2.8 DU (1.0%)	3.9 DU (1.4%)	4.3 DU (1.5%)
Amplitude of solar cycle	0.4 DU (0.1%)	0.3 DU (0.1%)	2.2 DU (0.7%)	2.3 DU (0.7%)	3.5 DU (1.3%)	2.7 DU (1.0%)	3.3 DU (1.2%)	1.0 DU (0.3%)
Amplitude of NAO (winter)	18.3 DU (5.7%)	16.5 DU (5.2%)	18.4 DU (5.8%)	18.3 DU (5.9%)	4.2 DU (1.5%)	7.2 DU (2.6%)	5.0 DU (1.8%)	8.0 DU (2.8%)

833 \*period 2007-2016

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837 **Table 7. Annual mean total ozone, amplitude of annual cycle, amplitude of QBO, amplitude of solar cycle and amplitude of NAO in the period 1995-**  
 838 **2016 from GOME-2A, GTO-ECV satellite data, ground-based observations and SBUV (v8.6) satellite overpass data over: (a) Canada and USA (11**  
 839 **stations mean), and (b) Europe (41 stations mean).**

840

	(a) Canada and USA				(b) Europe			
	30°-50° N, 60°-110° W (11 stations mean)				35°-55° N, 10° W-40° E (41 stations mean)			
	GOME-2A*	GTO-ECV	GROUND	SBUV (v8.6)	GOME-2A*	GTO-ECV	GROUND	SBUV (v8.6)
Annual mean	324.2 DU	320.6 DU	322.5 DU	320.9 DU	329.9 DU	325.7 DU	326.9 DU	326.8 DU
Amplitude of annual cycle	38.1 DU (11.7%)	34.1 DU (10.6%)	33.2 DU (10.3%)	34.0 DU (10.6%)	39.3 (11.9%)	40.5 DU (12.4%)	39.2 DU (12.0%)	40.7 DU (12.4%)
Amplitude of QBO	2.1 DU (0.6%)	2.5 DU (0.8%)	3.5 DU (1.1%)	2.6 DU (0.8%)	2.7 DU (0.8%)	1.9 DU (0.6%)	2.8 DU (0.8%)	2.2 DU (0.7%)
Amplitude of solar cycle	0.3 DU (0.1%)	0.5 DU (0.2%)	1.4 DU (0.4%)	0.5 DU (0.2%)	2.1 DU (0.6%)	0.8 DU (0.2%)	1.0 DU (0.3%)	0.3 DU (0.1%)
Amplitude of NAO (winter)	9.8 DU (3.0%)	6.9 DU (2.2%)	8.7 DU (2.7%)	9.3 DU (2.9%)	9.8 DU (3.0%)	8.9 DU (2.7%)	11.8 DU (3.6%)	9.9 DU (3.0%)

841 \*period 2007-2016

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845 **Table 8. Statistics of the comparisons between total ozone, tropopause pressures and NAO index in winter**  
 846 **(DJF mean) for a) the northern part of North Atlantic (35°-50° N, 20°-50° W), b) its southern part (15°-27° N,**  
 847 **30°-60° W), c) 11 stations in Canada and USA, and d) 41 stations in Europe.**

848

(a) Northern part of North Atlantic	Correlation with NAO in winter	Intercept (%)	Slope*	Error	t-value	p-value	N
GOME-2A	-0.85	0.035	-2.474	0.568	-4.355	0.0033	9
GTO-ECV	-0.74	0.412	-2.188	0.453	-4.827	0.0001	21
TOMS/OMI/OMPS	-0.74	0.734	-2.386	0.538	-4.436	0.0004	18
Oslo CTM3	-0.75	0.639	-2.457	0.498	-4.937	<0.0001	21
Tropopause	-0.83	0.665	-3.112	0.480	-6.478	<0.0001	21

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(b) Southern part of North Atlantic	Correlation with NAO in winter	Intercept (%)	Slope*	Error	t-value	p-value	N
GOME-2A	+0.54	-0.132	0.661	0.386	1.712	0.1306	9
GTO-ECV	+0.60	-0.202	1.097	0.333	3.291	0.0038	21
TOMS/OMI/OMPS	+0.58	-0.334	1.138	0.402	2.832	0.0120	18
Oslo CTM3	+0.65	-0.077	1.188	0.316	3.761	0.0013	21
Tropopause	+0.59	-0.702	1.547	0.482	3.207	0.0046	21

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(a) CA/USA (11 stations mean)	Correlation with NAO in winter	Intercept (%)	Slope*	Error	t-value	p-value	N
GOME-2A	-0.71	-0.042	-1.305	0.493	-2.647	0.0331	9
GTO-ECV	-0.40	0.308	-0.904	0.479	-1.886	0.0746	21
SBUV overpass	-0.50	0.318	-1.209	0.476	-2.541	0.0199	21
GB (WOUDC)	-0.46	0.268	-1.046	0.477	-2.190	0.0419	20
Tropopause	-0.41	0.268	-0.739	0.377	-1.959	0.0650	21

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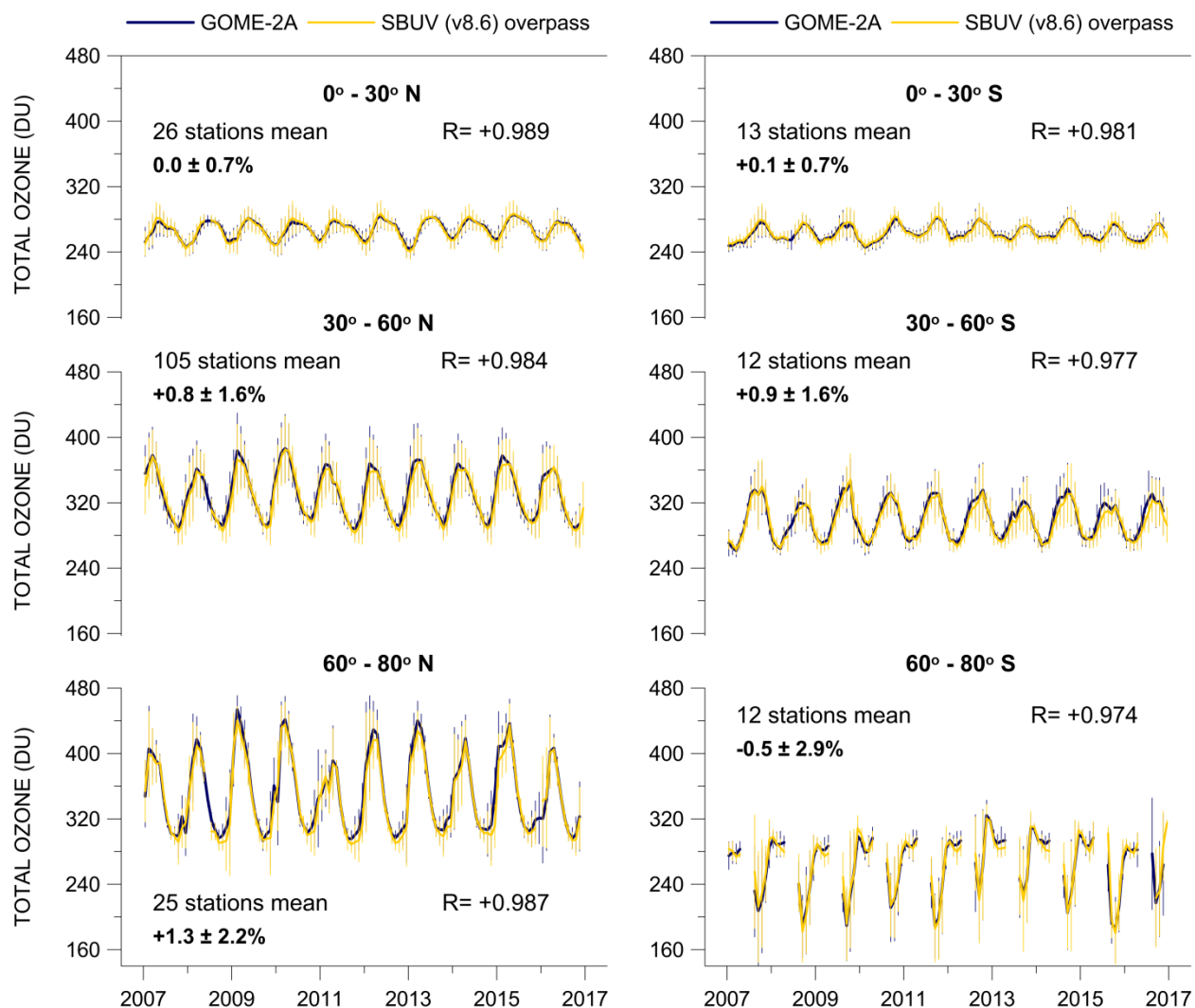
(b) Europe (41 stations mean)	Correlation with NAO in winter	Intercept (%)	Slope*	Error	t-value	p-value	N
GOME-2A	-0.46	0.089	-1.282	0.897	-1.428	0.1963	9
GTO-ECV	-0.42	0.315	-1.141	0.573	-1.992	0.0609	21
SBUV overpass	-0.47	0.389	-1.264	0.543	-2.329	0.0311	21
GB (WOUDC)	-0.48	0.625	-1.327	0.560	-2.368	0.0287	21
Tropopause	-0.40	0.048	-0.989	0.523	-1.891	0.0739	21

852 \* The slope is in % per unit change of the explanatory variable. Error, t-value and p-value refer to slope,

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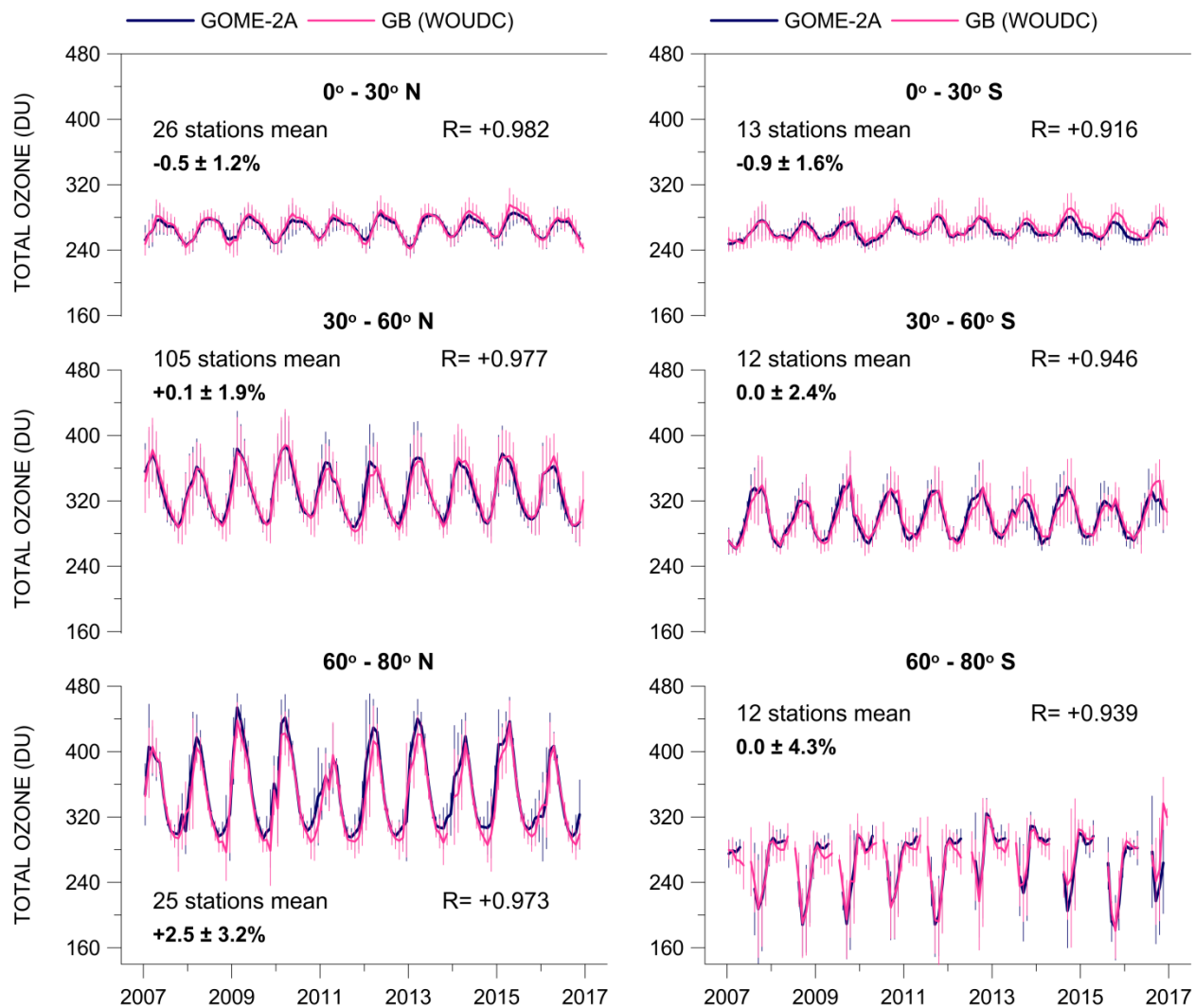
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859 **Figure 1. Monthly mean total ozone from GOME-2A as compared with monthly mean total ozone from**  
 860 **SBUV (v8.6) satellite overpass data for the period 2007-2016 over the Northern and the Southern Hemisphere**  
 861 **based on stations mean data. *R* is the correlation coefficient between the two lines. Error bars show the**  
 862 **standard deviation of each monthly mean. Mean differences ±  $\sigma$  are given as [GOME-2A – SBUV] / SBUV**  
 863 **(%).**

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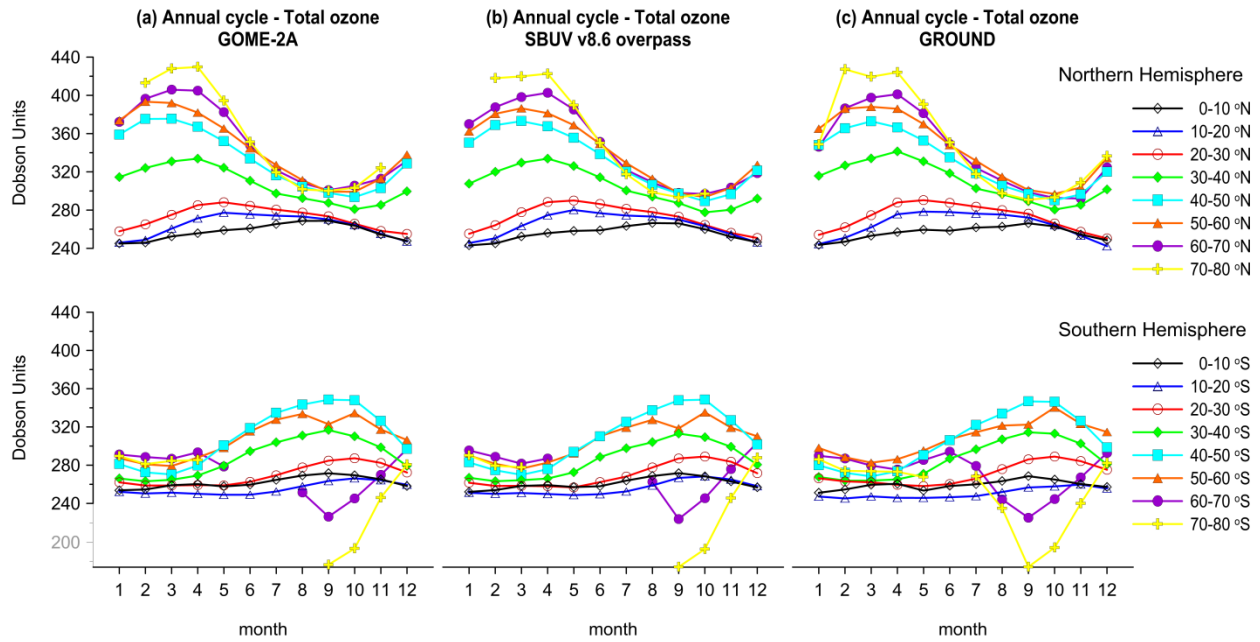
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869 **Figure 2.** Same as in Figure 1 but for GOME-2A and GB observations. *R* is the correlation coefficient  
 870 between the two lines. Error bars show the standard deviation of each monthly mean. Mean differences ±  $\sigma$   
 871 are given as [GOME-2A – GROUND] / GROUND (%).

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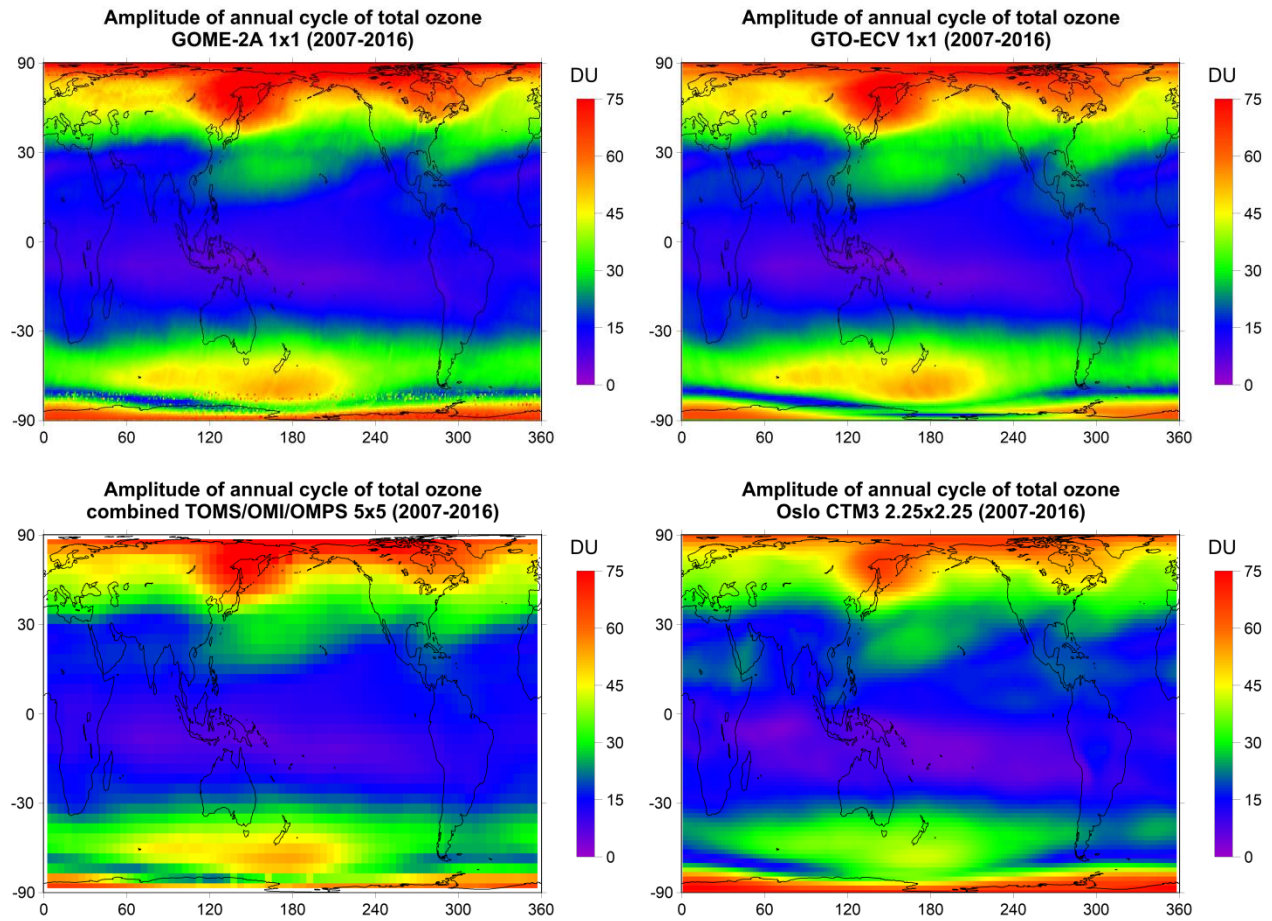
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877 **Figure 3. Comparison of the annual cycle of total ozone from GOME-2A with that from SBUV (v8.6) satellite**  
878 **overpass data and GB observations in the period 2007-2016 based on stations data averaged per 10 degree**  
879 **latitude zones. The annual cycle is distorted above 60 deg. S due to the Antarctic ozone hole.**

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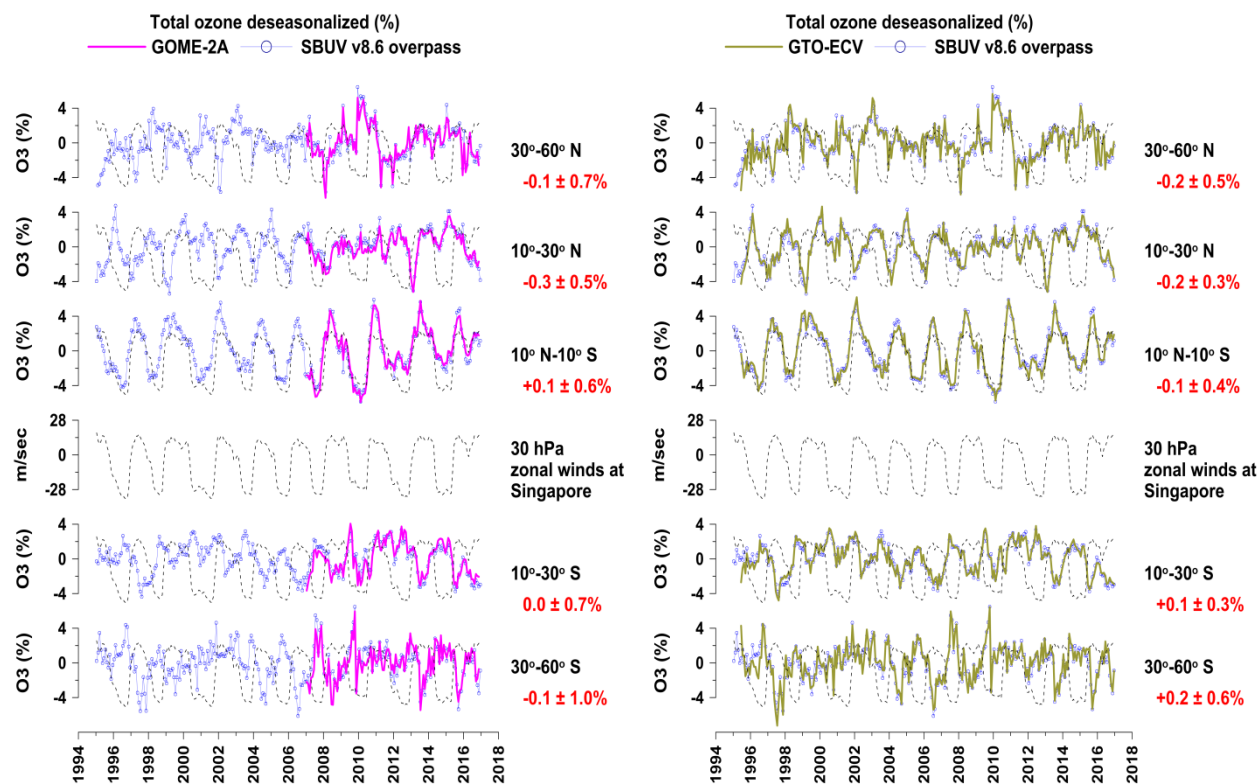


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884 **Figure 4. Comparison of the amplitude [i.e., (max-min)/2] of the annual cycle of total ozone from GOME-2A**  
885 **(upper left) with the amplitude of the annual cycle of total ozone from GTO-ECV (upper right), the combined**  
886 **TOMS/OMI/OMPS satellite data (lower left) and Oslo CTM3 model simulations (lower right).**

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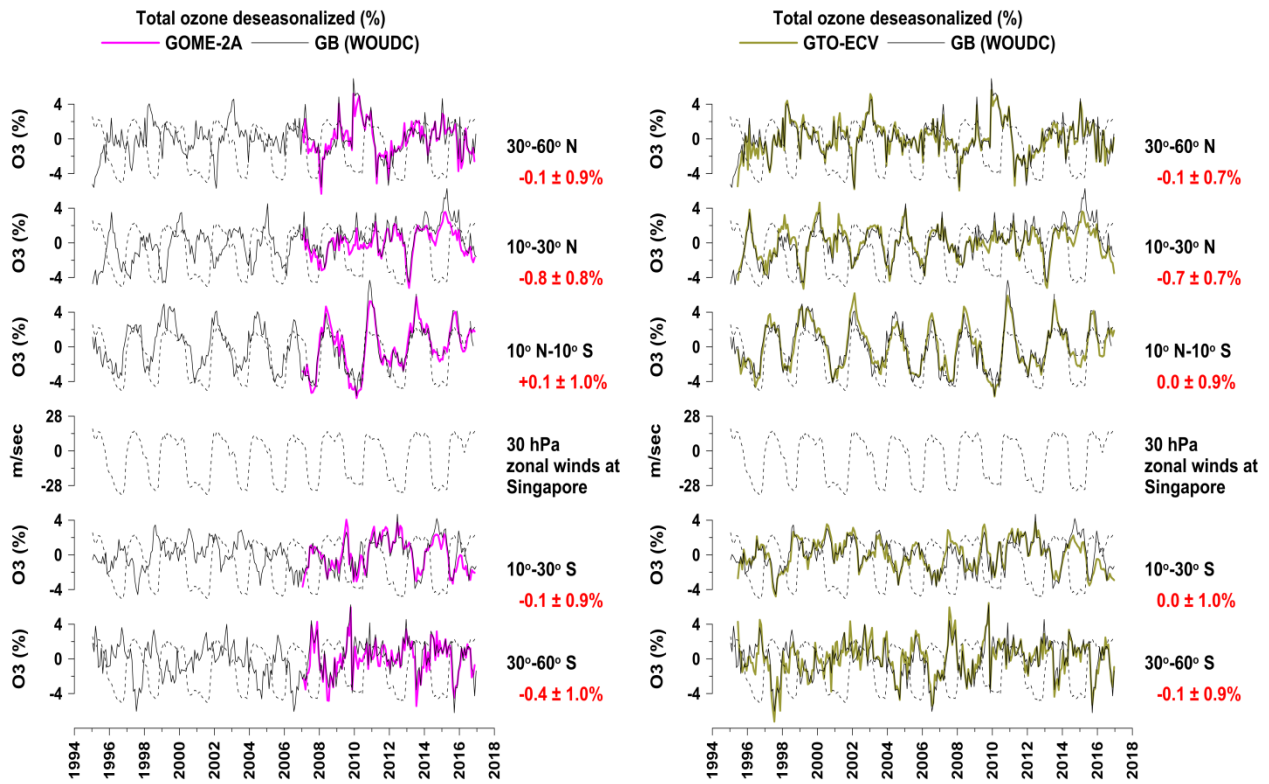
890

891 **Figure 5. (Left panel) Time series of deseasonalised total ozone from GOME-2A and SBUV (v8.6) satellite**  
 892 **overpasses over different latitude zones along with the equatorial zonal winds at the**  
 893 **QBO; (Right panel) same as in left panel but for GTO-ECV and SBUV. Values with red colour refer to**  
 894 **the mean differences  $\pm \sigma$  (in %) between GOME-2A and SBUV deseasonalised data averaged over various**  
 895 **WOUDC stations (150 stations in the northern mid-latitudes (30°-60° N), 21 stations in the northern**  
 896 **subtropics (10°-30° N), 8 stations in the tropics (10° S-10° N), 10 stations in southern subtropics (10°-30° S) and**  
 897 **12 stations in the southern mid-latitudes (30°-60° S)). The QBO proxy is superimposed on the ozone**  
 898 **anomalies.**

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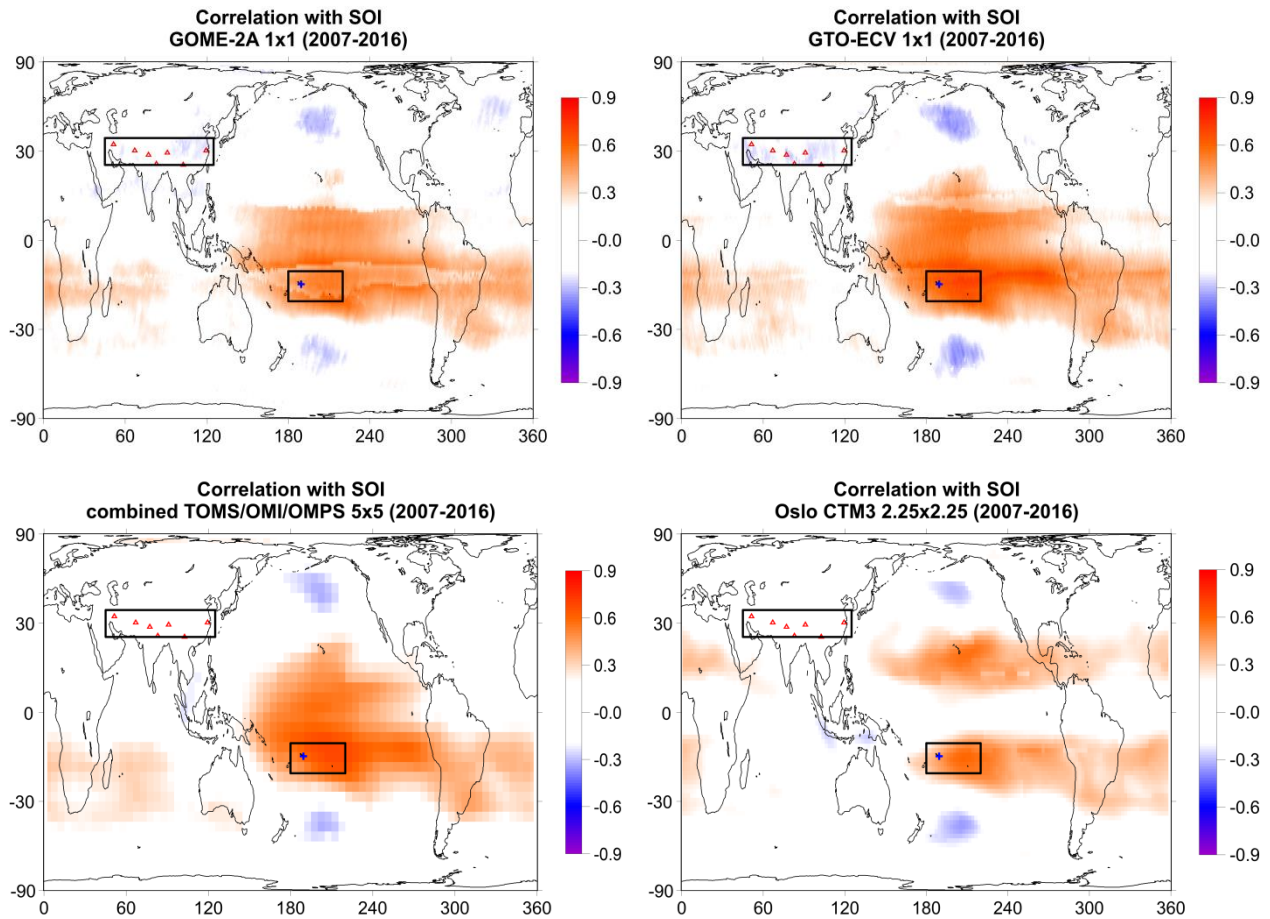
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903 **Figure 6. Same as in Figure 5 but for GOME-2A and GB observations (left panel), and for GTO-ECV and**  
904 **GB observations (right panel). The QBO proxy is superimposed on the ozone anomalies.**

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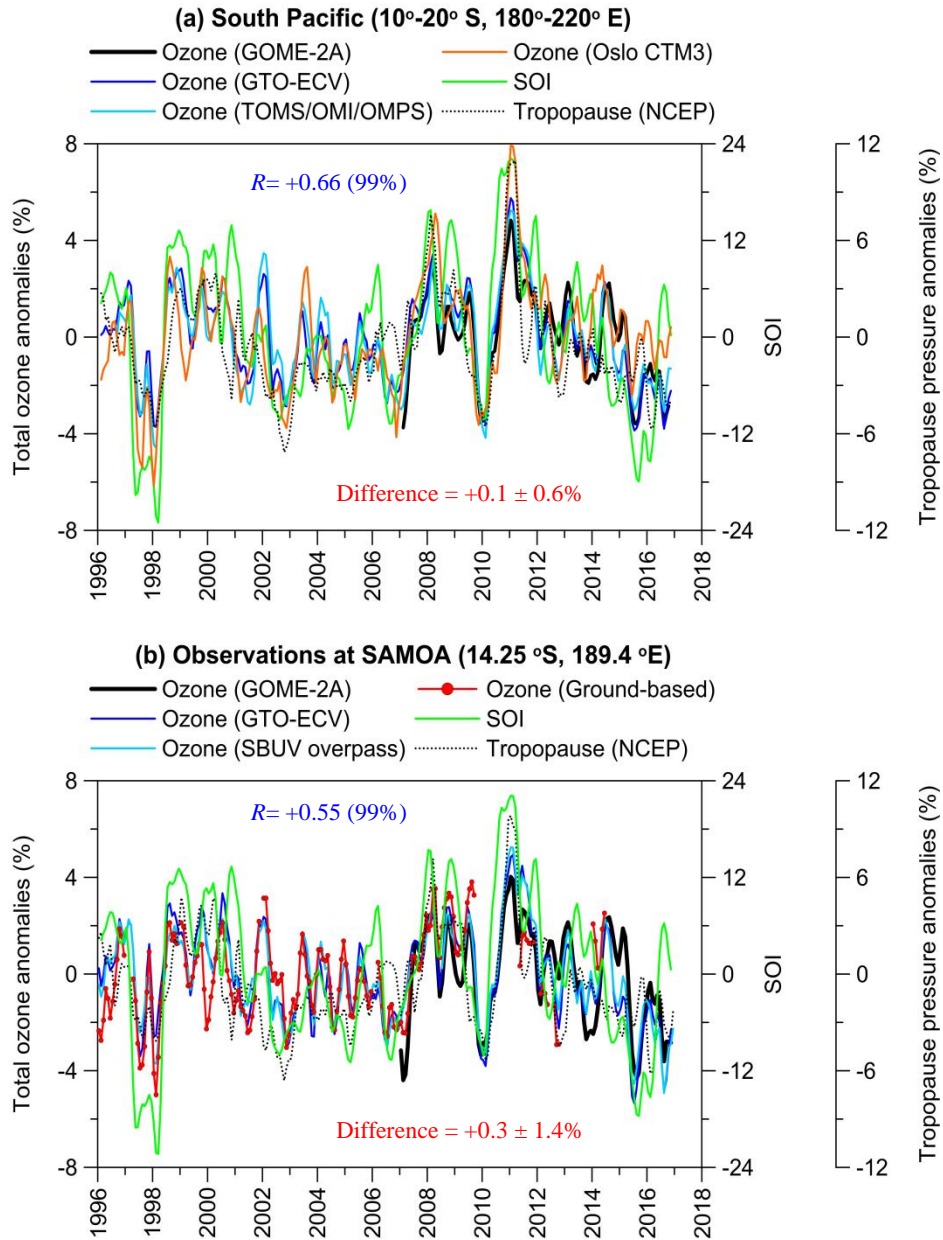


908

909 **Figure 7. Map of correlation coefficients between total ozone and SOI for GOME-2A (upper left), GTO-ECV**  
910 **(upper right), TOMS/OMI/OMPS satellite data (lower left) and Oslo CTM3 model simulations (lower right).**  
911 **Rectangles correspond to the South Pacific region (10-20 °S, 180-220 °E) and South Asia region (35-45 °N, 45-**  
912 **125 °E), blue cross to the station Samoa (14.25 °S, 189.4 °E) and red triangles to stations in South Asia, in**  
913 **which total ozone has been studied as for the impact of ENSO after removing variability related to the annual**  
914 **cycle, QBO and solar cycle. Positive correlations are shown by red colours while negative correlations by blue**  
915 **colours. Only correlation coefficients above/below  $\pm 0.2$  are shown.**

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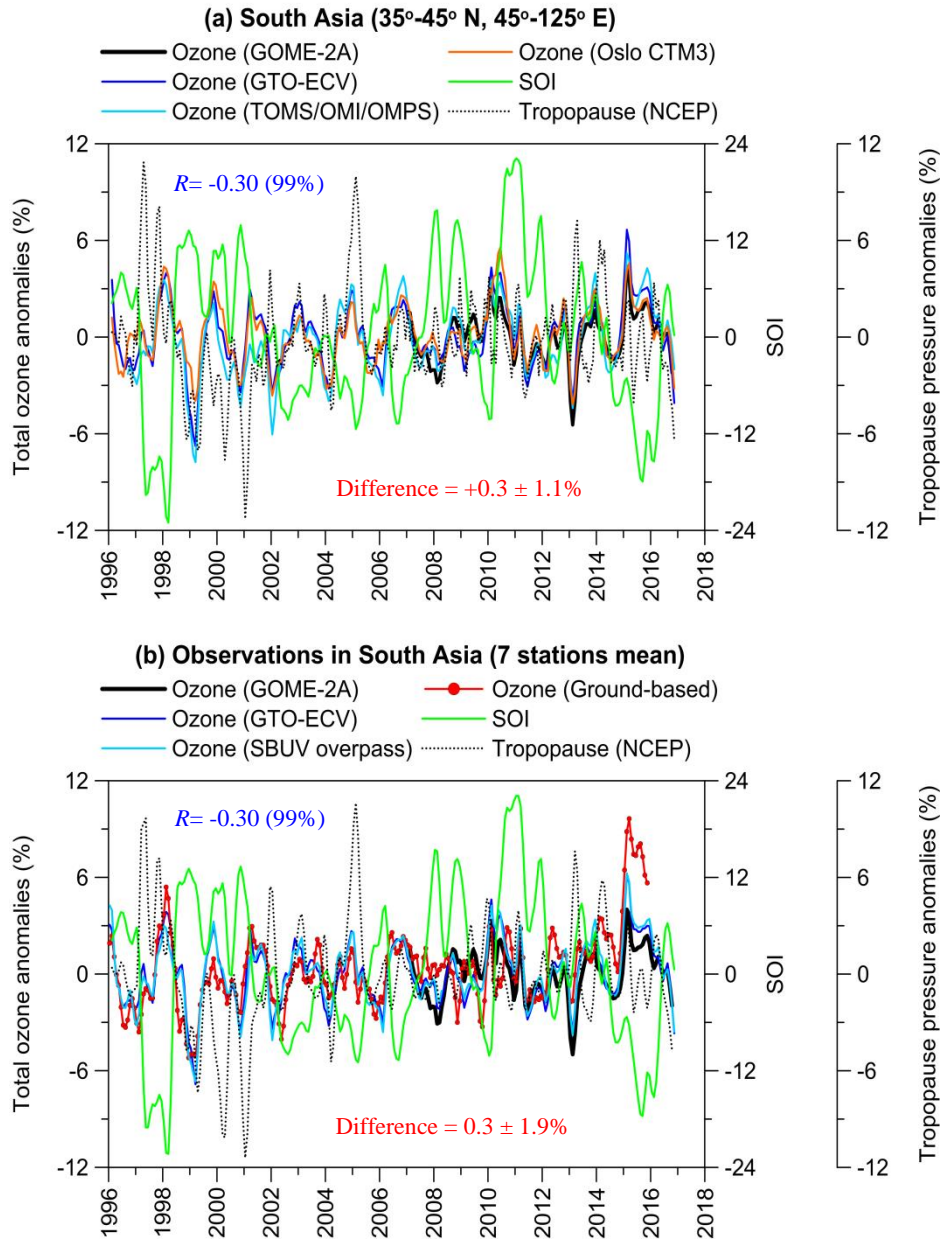


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919 **Figure 8. (a) Example of regional time series of total ozone (%) over the South Pacific region (10°-20° S, 180°-**  
 920 **220° E) along with SOI. The dotted line shows the respective tropopause pressure variability from NCEP.  $R$  is**  
 921 **the correlation coefficient between GTO-ECV total ozone and SOI (statistical significance of  $R$  is given in**  
 922 **parentheses). The difference refers to the mean difference  $\pm \sigma$  (in %) between GTO-ECV and the combined**  
 923 **TOMS/OMI/OMPS satellite data. (b) Same as in (a) but for SBUV overpass and GB data at the station**  
 924 **Samoa. The difference refers to the mean difference  $\pm \sigma$  (in %) between GTO-ECV and GB data.**

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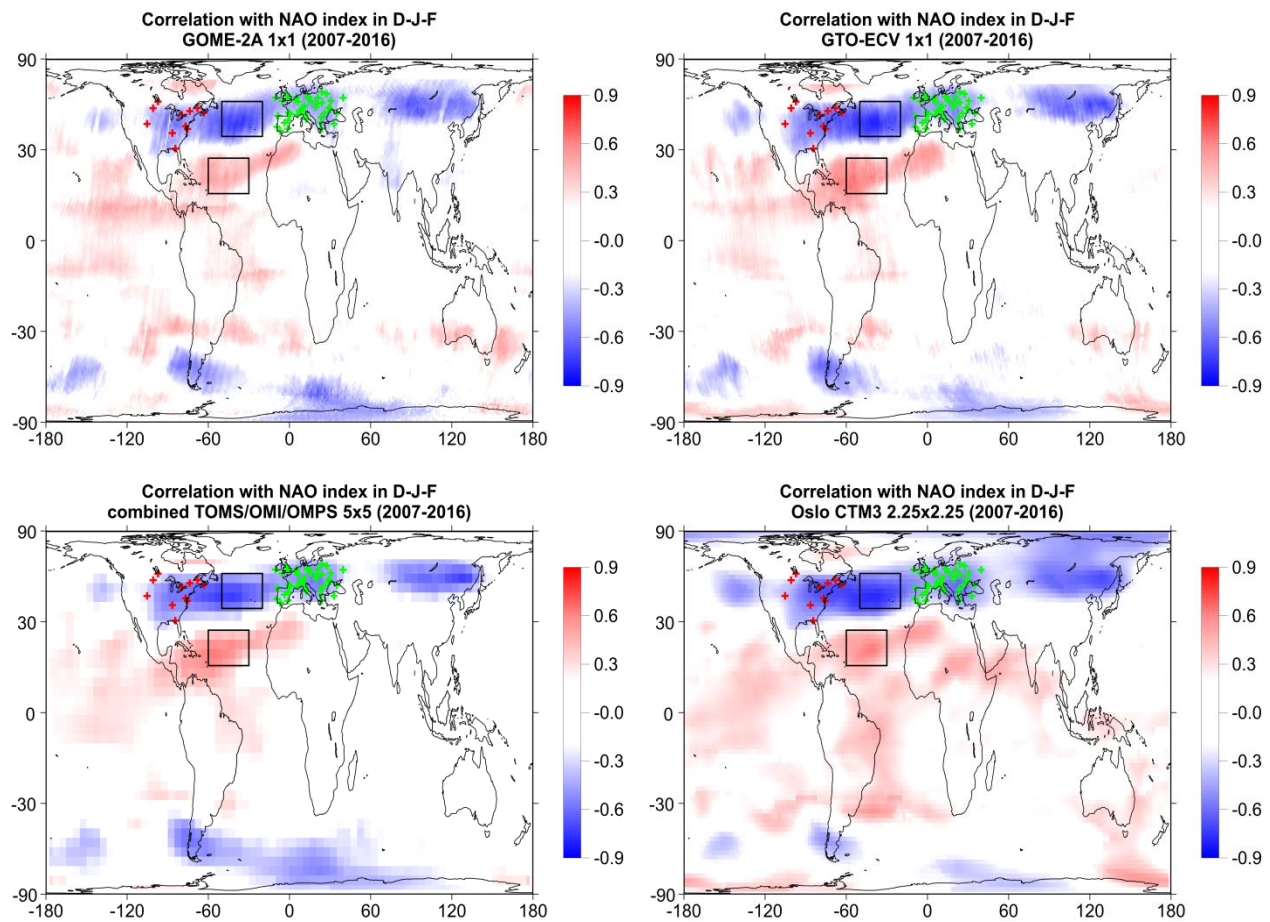
928 **Figure 9. (a) Example of regional time series of total ozone (%) over South Asia (35°-45° N, 45°-125° E) along**  
 929 **with SOI. The dotted line shows the respective tropopause pressure variability from NCEP.  $R$  is the**  
 930 **correlation coefficient between GTO-ECV total ozone and SOI (statistical significance of  $R$  is given in**  
 931 **parentheses). The difference refers to the mean difference  $\pm \sigma$  (in %) between GTO-ECV and the combined**  
 932 **TOMS/OMI/OMPS satellite data. (b) Same as in (a) but with SBUV overpass and GB data averaged at 7**  
 933 **stations in South Asia. The difference refers to the mean difference  $\pm \sigma$  (in %) between GTO-ECV and GB**  
 934 **data.**

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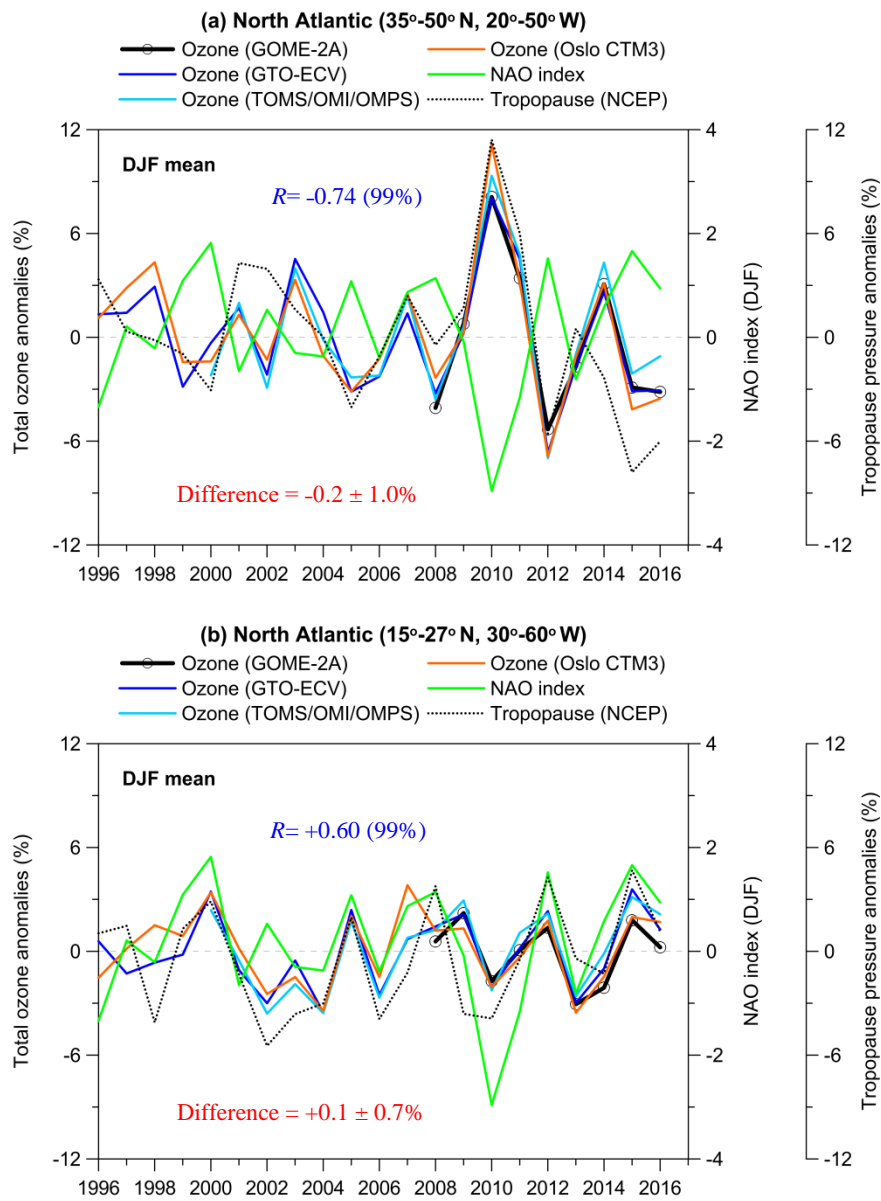


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939 **Figure 10. Map of correlation coefficients between total ozone and the NAO index during winter (December,**  
940 **January, February; D-J-F) for GOME-2A (upper left), GTO-ECV (upper right), TOMS/OMI/OMPS satellite**  
941 **data (lower left) and Oslo CTM3 model simulations (lower right). Rectangles correspond to regions in the**  
942 **North Atlantic (35°-50° N, 20°-50° W; 15°-27° N, 30°-60° W), and red and green crosses to stations in**  
943 **Canada/USA and Europe, in which total ozone has been studied as for the impact of NAO after removing**  
944 **variability related to the annual cycle , QBO, solar cycle and ENSO. Positive correlations are shown by red**  
945 **colours while negative correlations by blue colours. Only correlation coefficients above/below  $\pm 0.2$  are shown.**

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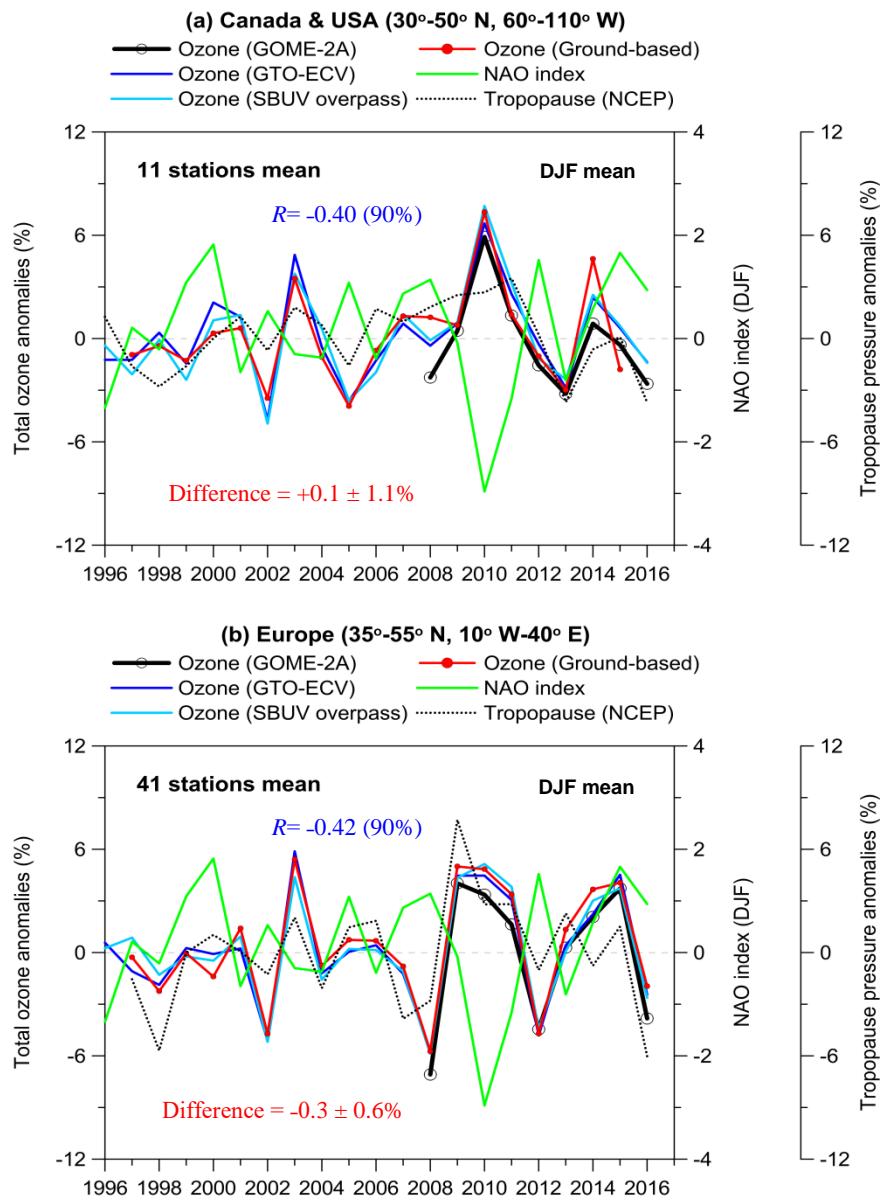


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950 **Figure 11. Example of regional time series of total ozone (%) over the North Atlantic regions (a) 35°-50° N,**  
 951 **20°-50° W and (b) 15°-27° N, 30°-60° W in winter (DJF mean) along with the NAO index. The dotted line**  
 952 **shows the respective tropopause pressure variability from NCEP reanalysis.  $R$  is the correlation coefficient**  
 953 **between GTO-ECV total ozone and the NAO index. The differences refer to the mean differences  $\pm \sigma$  (in %)**  
 954 **between GTO-ECV and the combined TOMS/OMI/OMPS satellite data.**

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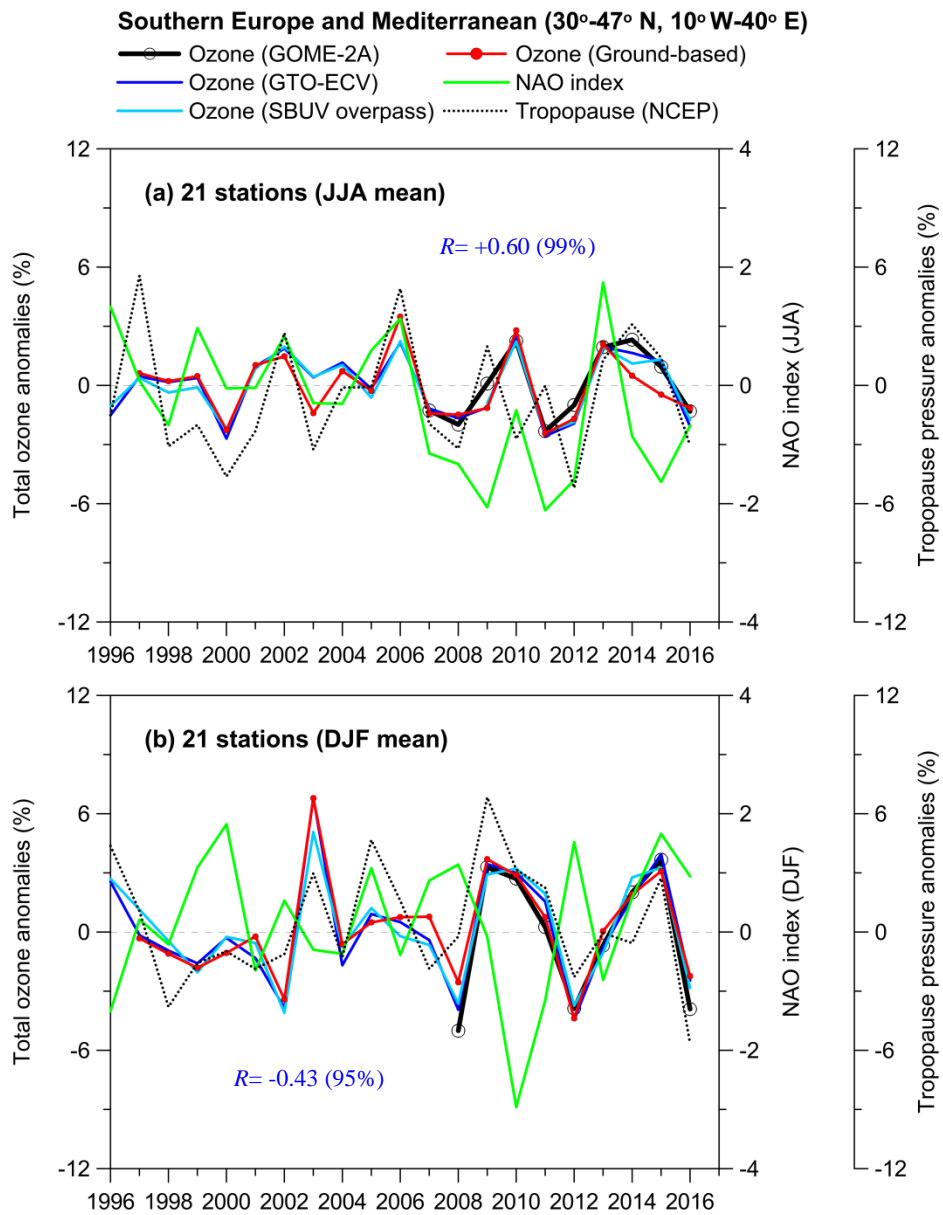
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959 **Figure 12. Comparison with GB observations over: (a) Canada and USA and (b) Europe in winter (DJF**  
 960 **mean).  $R$  is the correlation coefficient between GTO-ECV total ozone and the NAO index. The differences**  
 961 **refer to the mean differences  $\pm \sigma$  (in %) between GTO-ECV and GB data.**

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968 **Figure 13. Relation between total ozone and the NAO index in summer (JJA mean) and winter (DJF mean)**  
 969 **for 21 stations in southern Europe. The correlation coefficients refer to NAO index and GB total ozone after**  
 970 **removing variability related to the seasonal cycle, QBO, solar cycle and ENSO.**

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