

1 **Investigating zonal asymmetries in stratospheric ozone**  
2 **trends from satellite limb observations and a chemical**  
3 **transport model**

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10 **Key Points:**

- 11 • A longitudinal asymmetry in stratospheric ozone trends at northern high latitudes  
12 is found in satellite observations in the past two decades
- 13 • The asymmetry is particularly large in springtime and the TOMCAT chemistry  
14 transport model well reproduces the pattern
- 15 • Changes in polar wave activity and in the position and strength of the polar vor-  
16 tex are found to be relevant to explain this pattern

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**Abstract**

This study investigates the origin of the zonal asymmetry in stratospheric ozone trends at northern high latitudes, identified in satellite limb observations over the past two decades. We use a merged dataset consisting of ozone profiles retrieved at the University of Bremen from SCIAMACHY and OMPS-LP measurements to derive ozone trends. We also use TOMCAT chemical transport model (CTM) simulations, forced by ERA5 reanalyses, to investigate the factors which determine the asymmetry observed in the long-term changes. By studying seasonally and longitudinally resolved observation-based ozone trends, we find, especially during spring, a well-pronounced asymmetry at polar latitudes, with values up to +6 % per decade over Greenland and -5 % per decade over western Russia. The control CTM simulation agrees well with these observed trends, whereas sensitivity simulations indicate that chemical mechanisms, involved in the production and removal of ozone, or their changes, are unlikely to explain the observed behaviour. The decomposition of TOMCAT ozone time series and of ERA5 geopotential height into the first two wavenumber components shows a clear correlation between the two variables in the middle stratosphere and demonstrates a weakening and a shift in the wavenumber-1 planetary wave activity over the past two decades. Finally, the analysis of the polar vortex position and strength points to a decadal oscillation with a reversal pattern at the beginning of the century, also found in the ozone trend asymmetry. This further stresses the link between changes in the polar vortex position and the identified ozone trend pattern.

**Plain Language Summary**

Monitoring long-term ozone changes in the stratosphere is important to assess the evolution of the ozone layer in response to the Montreal Protocol and climate changes. In this study, we investigate the origin of a zonal asymmetry in stratospheric ozone trends over the past two decades, which was identified at northern polar latitudes by analyzing satellite observations. To this aim, we use a merged dataset consisting of ozone profiles retrieved at the University of Bremen from SCIAMACHY and OMPS-LP measurements to derive ozone trends. We also use TOMCAT chemical transport model (CTM) simulations to investigate the factors which determine the asymmetry observed in the long-term ozone changes. The asymmetry is found to be largest in springtime, and the CTM simulation agrees well with the observation-based trends. Sensitivity simulations indicate that chemical mechanisms, involved in the production and removal of ozone, are unlikely to explain the observed pattern. On the contrary, changes in atmospheric dynamics are found to be relevant. In particular, the analysis of the polar vortex position and strength points to a decadal oscillation with a reversal pattern at the beginning of the century, which is also found in the ozone trend asymmetry.

**1 Introduction**

The variations of the ozone concentration, as a function of time, altitude and latitude are explained by several dynamical, chemical and photochemical processes (e.g., Seinfeld & Pandis, 2016; WMO, 2022). In the lower stratosphere, where the chemical lifetime is relatively long (i.e. many years), except during polar spring, ozone is transported from the tropics to high latitudes, and it is affected by changes in atmospheric dynamics. In the upper stratosphere ozone has a relatively short photochemical lifetime, implying that changes in the transport of long-lived chemical species and in temperature play important roles in determining ozone concentrations at those levels.

The stratospheric circulation comprises an upper branch of the Brewer–Dobson circulation (BDC), involving upwelling in the tropics, meridional poleward transport, and then descent in the polar regions, and a lower branch, having a more rapid meridional poleward transport on isentropic surfaces (Butchart, 2014). This circulation is driven

67 by the wave breaking in the stratosphere and therefore is subject to strong inter-annual  
68 variability. The wave breaking happens in the so called “surf zone” at the edge of the  
69 polar vortex, so that its position and vertical structure has an indirect impact on the BDC,  
70 as well as BDC impacts the vortex position and strength (McIntyre & Palmer, 1984).  
71 An acceleration of the stratospheric mean mass transport has been predicted by several  
72 model studies (e.g., Garcia & Randel, 2008), but strong inter-annual variations prevent  
73 a robust detection of this trend from observations. In addition there may be decadal-  
74 scale oscillations. A large inter-annual variability also characterizes the polar vortex, with  
75 climate models not agreeing on whether it will weaken or strengthen during the 21st cen-  
76 tury (Karpechko et al., 2022). Several studies have addressed decadal changes of the po-  
77 lar vortex position and strength (e.g., Zhang et al., 2016; Seviour, 2017), pointing out  
78 a vortex weakening and shift of its mean position towards Eurasia, particularly at the  
79 end of the last century. In contrast, Hu et al. (2018) presented a strengthening of the  
80 stratospheric polar vortex over the last two decades, that could be related to a weaken-  
81 ing of the propagation of wavenumber-one wave flux, which was connected by the au-  
82 thors to sea-surface temperature warming over the north Pacific sector.

83 Among various anthropogenic influences on the stratospheric ozone, two most rel-  
84 evant are the release of halogen-containing ozone-depleting substances (ODSs) and of  
85 greenhouse gases (GHGs). With the adoption of the Montreal Protocol and its amend-  
86 ments the industrial production of ODSs, e.g. chlorofluorocarbon compounds (CFCs),  
87 was regulated. This reduced their emissions during the 1990s and is expected to lead to  
88 a recovery of the ozone layer globally (e.g., WMO, 2018, 2022). On the other hand, the  
89 increasing concentration of GHGs such as  $CO_2$  and  $CH_4$  in the troposphere, is causing  
90 a cooling of the stratosphere, through radiative transfer feedback. This cooling affects  
91 the ozone chemistry in the upper stratosphere, as the rate coefficients of reactions involved  
92 in catalytic cycles removing ozone have a direct dependence on temperature (Waugh et  
93 al., 2009). At the same time, the termolecular reaction  $O_2 + O + M \rightarrow O_3 + M$  has a  
94 rate inversely proportional to temperature so that the cooling also accelerates the ozone  
95 production (Groves et al., 1978).

96 The coupling between the described chemical and dynamical processes controlling  
97 stratospheric ozone is expected to have a complex spatial structure, varying in altitude,  
98 latitude, longitude and time. Therefore, to study long-term variations of the ozone field,  
99 there is a need for consistent long-term time series with a good temporal and spatial cov-  
100 erage over the whole globe.

101 In order to study long-term changes in ozone vertical profiles and test our under-  
102 standing of the impact of natural phenomena and anthropogenic activities on atmospheric  
103 ozone, single instrument time series are generally inadequate. Several studies have used  
104 satellite merged datasets to investigate stratospheric ozone trends, but the majority of  
105 them focused only on zonal mean changes (e.g., WMO, 2022). By exploiting the dense  
106 spatial sampling provided by limb observations, recently, Arosio et al. (2019) and Sofieva  
107 et al. (2021) looked at longitudinally resolved trends and highlighted the presence of zonal  
108 asymmetries, especially at northern high latitudes. In particular, poleward of 60 °N, they  
109 identified a bi-polar structure having positive values over the Atlantic/Greenland sec-  
110 tor and close to zero or negative changes over Siberia.

111 As discussed in the following paragraphs, some studies also showed zonal asymme-  
112 tries in the BDC and its impact on the distribution of trace gases and ozone trends in  
113 winter-time at northern high latitudes, by using model simulations and satellite datasets.  
114 Most studies focused on total ozone column measurements.

115 Longitudinally varying changes in total ozone were already pointed out in the study  
116 by Hood and Zaff (1995), who investigated total ozone at northern mid-latitudes dur-  
117 ing winter in the 1980s, using TOMS measurements. The authors identified the typical  
118 asymmetric ozone distribution related to quasi-stationary planetary waves, i.e. a pro-

119 nounced maximum over eastern Russia related to the Aleutian low and a secondary max-  
 120 imum over eastern Canada associated with the Icelandic low. In addition, a distinct lon-  
 121 gitudinal dependence of the mid-latitude ozone trends over this period was identified:  
 122 the largest negative trends (-40 DU per decade) occurred over Russia and western Pa-  
 123 cific, whereas positive trends were found over the northern Atlantic sector. Another study  
 124 using TOMS data was performed by Peters and Entzian (1999) who investigated decadal  
 125 total ozone changes in the months December-February over the period 1979-1992 in the  
 126 northern hemisphere. They found a strong anti-correlation between the long-term to-  
 127 tal ozone changes and the 300-hPa geopotential height (GPH) changes. This means that  
 128 decadal changes in the UTLS dynamics led to longitude-dependent changes in the to-  
 129 tal ozone.

130 Asymmetries in the ozone climatology were investigated by Bari et al. (2013), us-  
 131 ing models, reanalysis and satellite data, focusing on the northern mid-latitudes in win-  
 132 ter. The authors stressed the importance of a 3-D approach in studying the BDC. They  
 133 found that the distribution of winds and trace gases is related to the zonal wavenumber-  
 134 1 pattern in geopotential hight (GPH) observed in the northern hemispheric stratosphere  
 135 during winter at high and mid-latitudes. They showed that air masses are driven south-  
 136 wards and upwards to the upper stratosphere over the Pacific ocean, whereas over Eu-  
 137 rope and Asia the flow is northward and downward.

138 More recently, Kozubek et al. (2015) investigated the meridional component of strato-  
 139 spheric winds as a function of altitude at northern mid-latitudes to study its longitudi-  
 140 nal dependency. A well-defined two-core structure was identified at 10 hPa in the north-  
 141 ern hemisphere, with opposite wind directions, related to the Aleutian pressure high at  
 142 10 hPa. They also computed meridional wind changes over two periods: 1970-1995 and  
 143 1996-2012. They found that meridional wind trends are negative in the first period and  
 144 positive in the second period, i.e. the two-core structure became stronger in the last 2  
 145 decades. As a follow up, Kozubek et al. (2017) investigated the long-term variations of  
 146 stratospheric winds over the whole globe at 10 hPa using four reanalysis datasets. The  
 147 trends were reported for winter months before and after the ozone trend turnaround point  
 148 at the end of the 1990s. They found hints of an acceleration of the BDC and change in  
 149 the ozone trend asymmetries before and after 1997.

150 Within this framework, and in light of the findings in Arosio et al. (2019) and Sofieva  
 151 et al. (2021), the present paper aims to analyze vertically and longitudinally resolved ozone  
 152 trends from satellite observations and to exploit simulations from the TOMCAT chem-  
 153 istry transport model (CTM) to identify the mechanisms driving the observed zonal asym-  
 154 metry in the ozone linear trends in the period 2004 to 2021. Sect. 2 introduces the satel-  
 155 lite dataset used in this study and the TOMCAT CTM. Sect. 3 shows a comparison of  
 156 the measured and simulated ozone anomalies and of the respective zonally and longitu-  
 157 dinally resolved trends, where the asymmetry at northern high latitudes is evident. Sect. 4  
 158 presents the results of TOMCAT runs, which were designed to assess the impact of chem-  
 159 ical processes on the observed longitudinally asymmetric pattern in ozone trends. In Sect. 5  
 160 we explore in more detail the seasonally-resolved long-term changes in ozone and tem-  
 161 perature, which leads to Sect. 6 where geopotential height and ozone fields are decom-  
 162 posed into wavenumber-1 and -2 to assess similarities in their behaviour. Finally, in Sect. 7  
 163 we present some potential vorticity trends to further investigate changes in the polar vor-  
 164 tex over the past two decades, followed by concluding remarks.

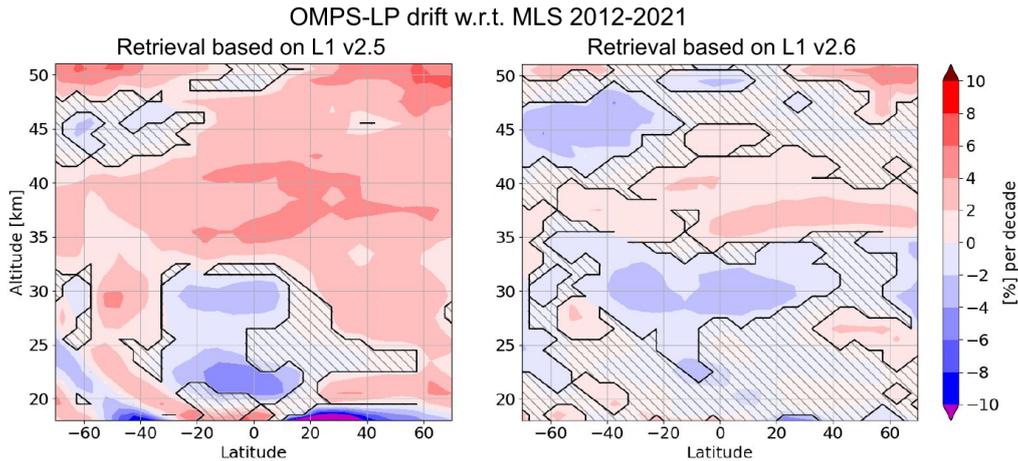
## 165 **2 Datasets**

### 166 **2.1 Satellite observations**

167 The merged satellite dataset consisting of observations from the SCanning Imag-  
 168 ing Absorption spectroMeter for Atmospheric CHartography (SCIAMACHY) and the

169 Ozone Mapper and Profiler Suite - Limb Profiler (OMPS-LP) has been produced at the  
 170 University of Bremen and is described in Arosio et al. (2019). Here, it will be referred  
 171 to as SCIA+OMPS. This dataset is longitudinally resolved with a grid size of  $5^\circ$  in lat-  
 172 itude and  $20^\circ$  in longitude and has a vertical resolution of 3.3 km. The time series has  
 173 been recently updated after the re-processing of the OMPS-LP dataset by using improved  
 174 Level 1 gridded (L1G) data. In the new L1G data version (v2.6), the NASA team im-  
 175 plemented some calibration corrections, a wavelength registration adjustment and an im-  
 176 proved pointing correction. The main aim of the re-processing is the removal of the posi-  
 177 tive drift identified in the previous OMPS-LP ozone product with respect to indepen-  
 178 dent time series (Kramarova et al., 2018), e.g., from the Microwave Limb Sounder (MLS),  
 179 which has proven its long-term stability in previous studies, e.g., Hubert et al. (2016).  
 180 We define the drift as the linear trend of the relative difference between OMPS-LP and MLS.

181 The drift w.r.t. MLS time series is shown in Fig. 1. The left panel refers to the OMPS-  
 182 LP time series retrieved using L1G v2.5 data, whereas the right panel refers to the up-  
 183 dated time series, based on L1G v2.6 data. The comparison between the left and the right  
 184 panel shows that the strong positive drift w.r.t. MLS has been significantly reduced, par-  
 185 ticularly above 35 km. The striped areas indicate values which are lower than the respec-  
 186 tive  $2\sigma$  uncertainty, i.e. they are not statistically significant at 95 % confidence level. Drift  
 187 values are still significant at some altitude-latitudes but generally with values half as large  
 188 as for the previous data version. This result provides improved confidence in the scien-  
 189 tific value of the ozone trends derived from the SCIA+OMPS merged time series.



**Figure 1.** Drift of the OMPS-LP ozone product retrieved at the University of Bremen w.r.t. MLS during the period 2012-2021 in % per decade. Left panel: using L1G v2.5 data. Right panel: using L1G v2.6 data. Striped areas are non-significant at  $2\sigma$ .

## 190 2.2 TOMCAT chemical transport model

191 TOMCAT/SLIMCAT is a three-dimensional off-line chemical transport model (CTM)  
 192 (M. Chipperfield, 2006). The model is forced by winds and temperatures from meteo-  
 193 rological analyses, which, in this study, are taken from the European Centre for Medium-  
 194 Range Weather Forecasts (ECMWF) reanalysis v5 (ERA5). Once the atmospheric trans-  
 195 port and temperatures are prescribed, the model calculates the abundances of chemi-  
 196 cal species in the troposphere and stratosphere. A full-chemistry reference run was used  
 197 as baseline for this study and other dedicated runs were produced. The resolution of the  
 198 model was kept  $2.8^\circ \times 2.8^\circ$  latitude and longitude, and about 1.5 km altitude in the strato-

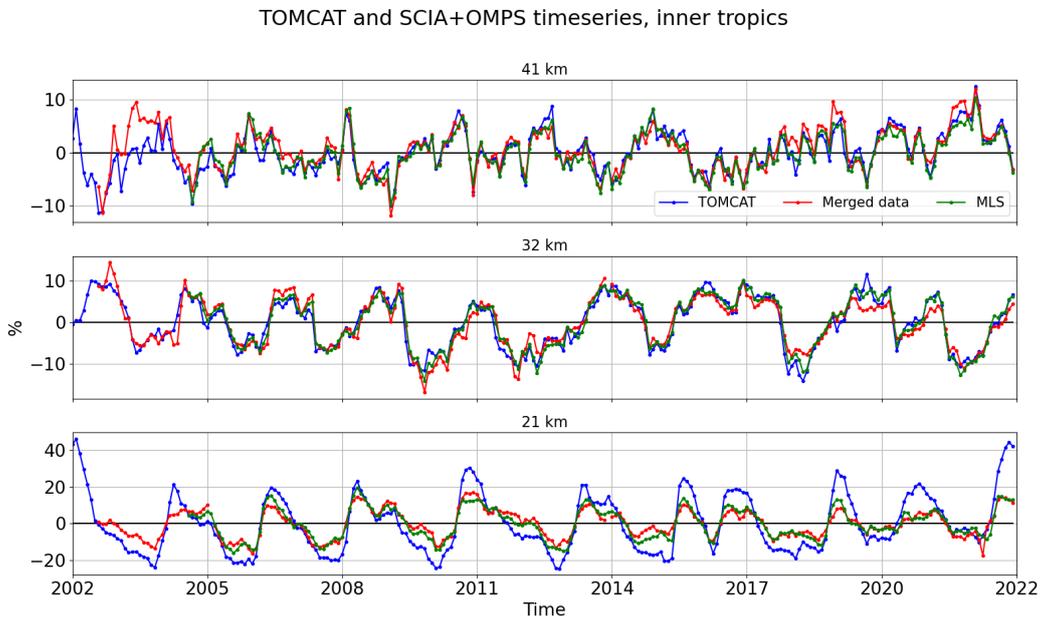
199 sphere, and was interpolated to match the merged satellite dataset resolution. Monthly  
 200 averaged values are considered.

201 The TOMCAT CTM simulations were used in this study for three important rea-  
 202 sons:

- 203 1. The CTM provides a continuous time series without spatial or temporal gaps, so  
 204 that it is possible, for example, to explore polar winter conditions, which are not  
 205 sampled by limb scattering sounders;
- 206 2. The possibility to study trends going back in time until 1979, when satellite limb  
 207 observations were sparse;
- 208 3. The possibility to investigate the mechanisms that determine the trend asymme-  
 209 tries by running dedicated simulations using different settings.

### 210 3 Comparison with TOMCAT: time series and trends

211 As a preliminary consistency check, we looked into the absolute bias between SCIA+OMPS  
 212 and TOMCAT time series, and noticed that the CTM underestimates ozone content in  
 213 the upper stratosphere and overestimates it in the lower stratosphere, which is a known  
 214 feature (Dhomse et al., 2021): further investigations on this issue are outside the scope  
 215 of this paper. For this reason and because we are interested in ozone trends, deseason-  
 216 alized (relative) anomalies of the time series were calculated and found to be in good agree-  
 217 ment with SCIA+OMPS, as shown in Fig. 2. In the lower tropical stratosphere, the am-  
 218 plitude of the oscillations, probably due to the Quasi Biennial Oscillation (QBO), is more  
 219 pronounced in TOMCAT than in SCIA+OMPS. MLS time series is also included as a  
 reference in this plot.

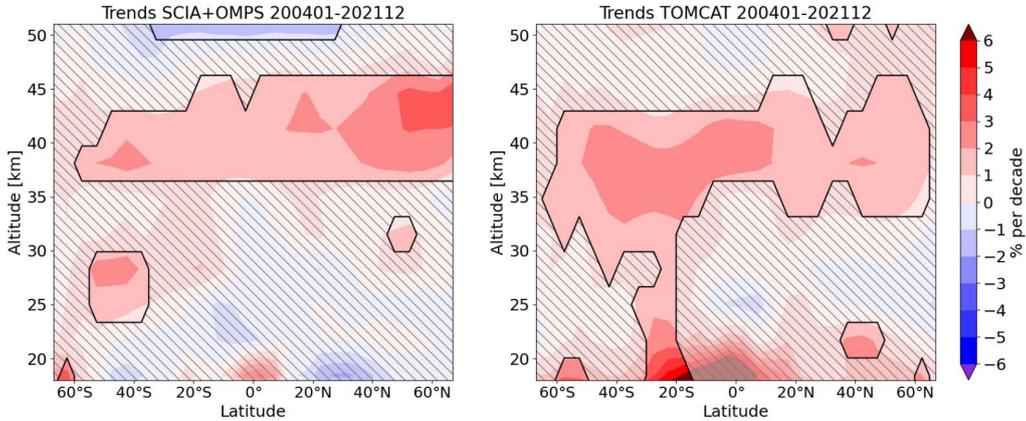


**Figure 2.** Deseasonalized anomalies from the reference TOMCAT simulation, the merged SCIA+OMPS dataset and MLS time series, in the inner tropics ( $[5^{\circ} \text{ S}, 5^{\circ} \text{ N}]$ ) at three altitudes (41, 32 and 21 km).

221 We applied to both TOMCAT and SCIA+OMPS time series a multivariate linear  
 222 regression model, based on the Long-term Ozone Trends and Uncertainties in the Strato-  
 223 sphere (LOTUS) model and including several proxies. In particular, we included in the  
 224 regression model traditionally employed proxies (e.g., Petropavlovskikh et al., 2019), such  
 225 as the first two principal components of the QBO, the Multivariate El Nino Southern  
 226 Oscillation (ENSO) index (MEI) and the Mg II index for solar activity, but also dynam-  
 227 ical proxies such as the yearly integrated eddy heat fluxes and the Atlantic/Antarctic  
 228 oscillation (AO/AAO). For a more detailed description of the used proxies, we refer to  
 229 Weber et al. (2022).

230 Due to the discrepancies in the SCIAMACHY time series w.r.t. other satellite prod-  
 231 ucts found in the first year of its lifetime (Sofieva et al., 2017), and because of the Hunga  
 232 Tonga volcanic eruption, which occurred in January 2022, with its large stratospheric  
 233 perturbation (Lu et al., 2023), we focused on the period 2004-2021 to study ozone trends.

234 The resulting zonal mean ozone trends are reported in Fig.3. Striped areas also in  
 235 the following plots indicate values which are smaller than the  $2\sigma$  uncertainty (non-significant).



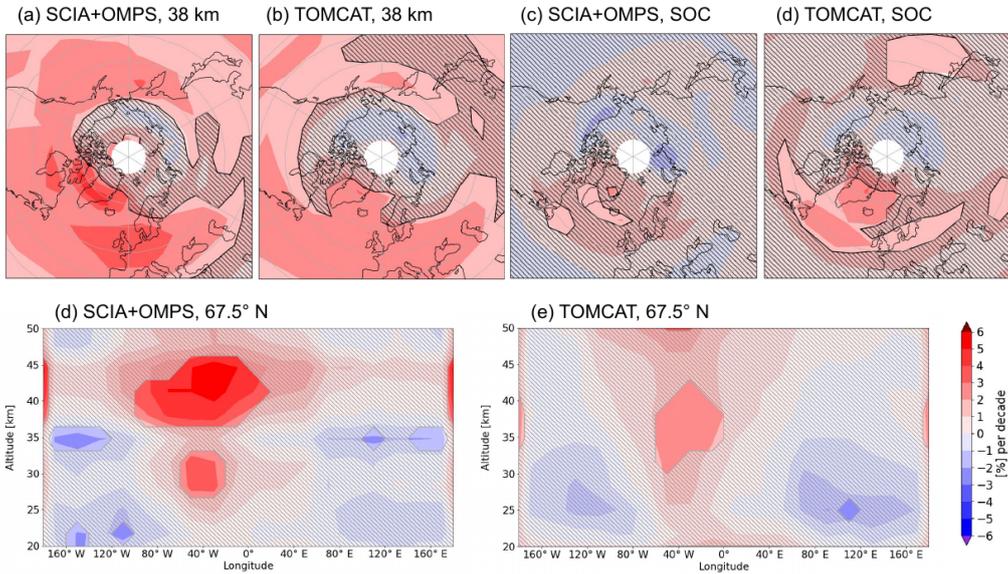
**Figure 3.** Zonal mean ozone trends from SCIA+OMPS on the left and from TOMCAT on the right, computed over the 2004-2021 period. Striped areas are non-significant at  $2\sigma$ .

236 Generally a good agreement between model and observations is found, with the ex-  
 237 pected positive trends in the middle and upper stratosphere, related to the ongoing ozone  
 238 recovery. The most significant discrepancy is located below 25 km where TOMCAT shows  
 239 overall positive trends, whereas SCIA+OMPS shows negative values, though non-significant,  
 240 except for the inner tropics at 19 km. The detection of negative trends in the lower trop-  
 241 ical and extra-tropical stratosphere has been extensively debated (Ball et al., 2018; M. P. Chip-  
 242 perfield et al., 2018). A possible reason for the discrepancy between TOMCAT and SCIA+OMPS  
 243 in the lower stratosphere is related to ERA5 forcing, as pointed out by Li et al. (2022,  
 244 2023).

245 In comparison with the long-term 2000-2020 ozone trends shown in Godin-Beekmann  
 246 et al. (2022), trend values in the lower stratosphere are not significant and closer to zero.  
 247 The negative values identified in SCIA+OMPS above 47 km are not shown by other merged  
 248 datasets.

249 Longitudinally resolved trends from TOMCAT and SCIA+OMPS are compared  
 250 in Fig. 4, at a specific altitude (38 km), in terms of stratospheric ozone column (SOC)  
 251 and for a longitude-altitude cross section at 67.5° N. In the top row, we notice a pro-  
 252 nounced longitudinal variability and some common patterns in CTM and in SCIA+OMPS

253 trends, especially the zonal asymmetry above 45° N. This asymmetry is more evident  
 254 in TOMCAT, with negative (though non-significant) values over the Siberian and East-  
 255 Canadian sectors, and positive values over the Atlantic sector. Looking at the plot show-  
 256 ing the trends in SOC and the longitude-altitude cross section at 67.5° N, we can see that  
 257 the asymmetry is vertically consistent. The positive SOC trend in the Atlantic sector  
 258 are significant at  $2\sigma$ , especially for TOMCAT. A similar structure was identified also in  
 259 MLS time series and in the MEGRIDOP dataset (Sofieva et al., 2021). Above 35 km,  
 260 TOMCAT shows smaller positive values over the Atlantic sector as compared to the satel-  
 261 lite observations. To further test the robustness of this pattern and its independence from  
 262 ERA5 forcing, in the Supplements, in Fig. S2 we compared trends from the MERRA 2  
 263 - Global Modeling Initiative (M2-GMI) CTM time series, which is forced with MERRA-  
 264 2 meteorology, with TOMCAT, and found a bias in term of absolute trend values but  
 265 a good agreement in terms of asymmetric pattern.



**Figure 4.** Top row: longitudinally resolved trends for SCIA+OMPS (panel a) and TOMCAT (panel b) datasets at 38 km, in terms of stratospheric columns for both datasets in panels (c) and (d) respectively. Bottom row: trends for a longitude-altitude cross section at 67.5° N. Striped areas denote non-significant values at  $2\sigma$  level.

266 A TOMCAT simulation with a higher spatial resolution ( $1.4^\circ \times 1.4^\circ \times 0.75$  km  
 267 grid) was run to investigate whether the discrepancies between the CTM and SCIA+OMPS  
 268 could be reduced. This simulation was sampled at the locations of the satellite obser-  
 269 vations to make the CTM time series more consistent with the merged dataset, in terms  
 270 of temporal and spatial sampling. The resulting dataset was re-gridded and the two parts  
 271 of the time series, covering SCIAMACHY and OMPS-LP periods respectively, were de-  
 272 biased to remove the discrepancy related to the different local time of the satellite ob-  
 273 servations. The comparison of the resulting higher resolution data did not show any sig-  
 274 nificant differences w.r.t. the standard run, neither in the trends nor in the time series  
 275 (here not shown). As a result, we use the standard run as reference in this study.

## 4 Investigation of the potential influence of chemical processes on the trend asymmetry

We performed two additional sensitivity (SEN) simulations to investigate the potential influence of chemical processes on the origin of the ozone trend asymmetry. The first simulation 'SEN-fDyn' was forced using constant ERA5 data, corresponding to the year from July 1999 to June 2000, which were repeated each year over the 2004-2021 period. The choice of the 1999/2000 year is arbitrary, as far as a winter with an average-strong polar vortex is considered; we tested the use of the July 2002 to June 2003 period for the repeating forcing without finding any significant difference. In the second simulation 'SEN-noPSC', polar stratospheric clouds (PSCs)-related heterogeneous chemistry was inhibited, by not allowing temperature to drop below 200 K in the model chemistry scheme to prevent PSC formation. The results are shown in Fig. 5. The longitude-altitude cross section of the ozone trends at 67.5° N over 2004-2021 from the reference full-chemistry TOMCAT (control) run is shown, together with the trends for the 'SEN-noPSC' and 'SEN-fDyn' simulation.

Fig. 5 shows that the zonally asymmetrical trend pattern from the SEN-noPSC simulation is almost identical to the one from the control simulation. As expected, the trends over the Atlantic sector are smaller due to reduced ozone losses in the absence of PSCs. This indicates that heterogeneous chemistry does not play a relevant role in producing trends variable with longitude. To further test this hypothesis and the robustness of the zonal asymmetry we computed the ozone trends for the 2004-2019 period, i.e. excluding the cold 2019/2020 Arctic winter. As discussed in the Supplements, Fig. S1, we did not find relevant differences, highlighting the robustness of the pattern.

Trend values in the 'repeating forcing' scenario show zonal symmetry and are overall smaller with respect to the reference run. In this case no long-term temperature trend is present in the forcing, which plays an important role for the ozone trend in the upper stratosphere. The fact that no zonal asymmetry is observed for this run indicates that gas-phase chemistry alone cannot directly explain either the asymmetry in trends. However, an indirect impact of atmospheric dynamics on gas-phase chemistry cannot be excluded (Galytska et al., 2019).

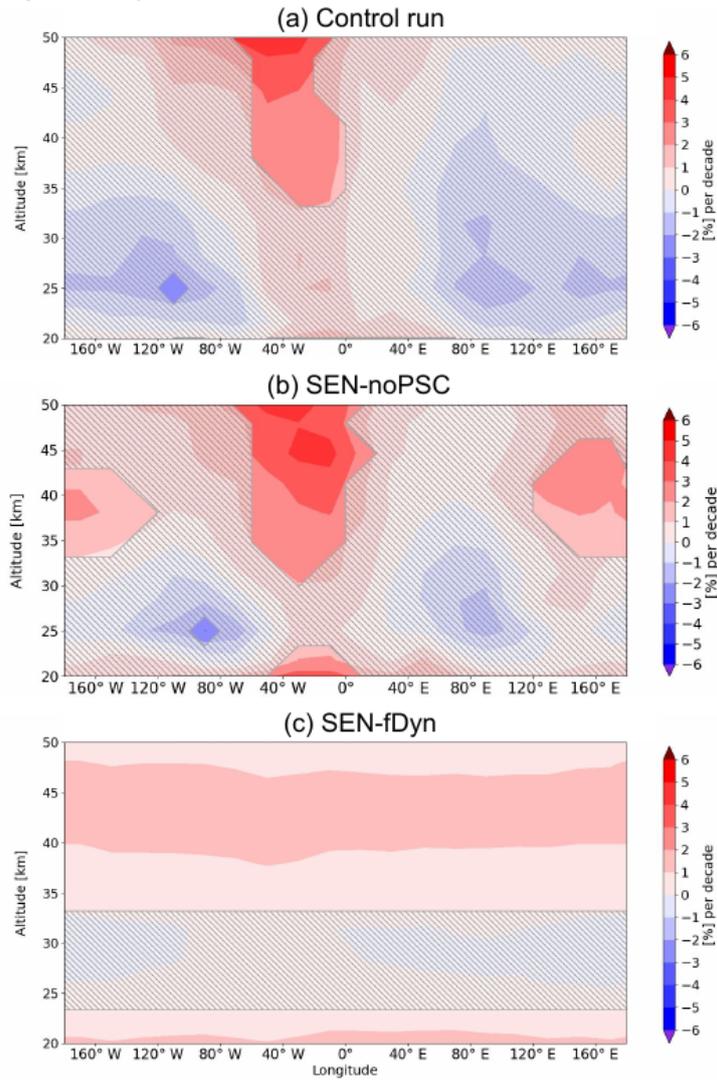
In addition, we compared the trend results computed for the TOMCAT reference run and for ERA5 ozone data. As shown in Fig. 6, the zonal trends in ERA5 are significantly different from Fig. 3, pointing out that ozone reanalysis data should not be used to compute long-term ozone changes, unless a careful de-biasing of the time series is performed (e.g., Bernet et al., 2020). However, longitudinally resolved trends shown in Fig. 6 at 32 km have a remarkable similarity with the pattern found in TOMCAT. This provides more evidence that atmospheric dynamics is mainly driving the observed asymmetric pattern, as TOMCAT is forced with ERA5 meteorology.

## 5 Seasonal ozone trends

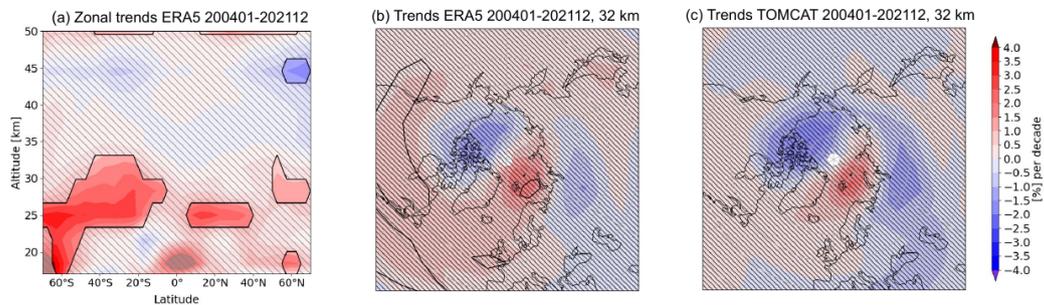
To further investigate the longitudinal asymmetry at northern high latitudes, seasonal trends were analyzed. Two approaches to obtain seasonal time series for the SCIA+OMPS dataset are described in Appendix Appendix A. In the following, we show trend values obtained by merging the two seasonally averaged single-instrument time series.

In Fig. 7, seasonal ozone trends are shown for SCIA+OMPS (top row) and for the reference TOMCAT run (middle row) at 32 km altitude for spring (MA), summer (JJA) and autumn (SO). Only two months are used in spring and autumn to get a better coverage of the polar regions. The TOMCAT time series was masked to mirror the availability of satellite data. ERA5 temperature trends are displayed in the bottom row of Fig. 7 for the same three seasons.

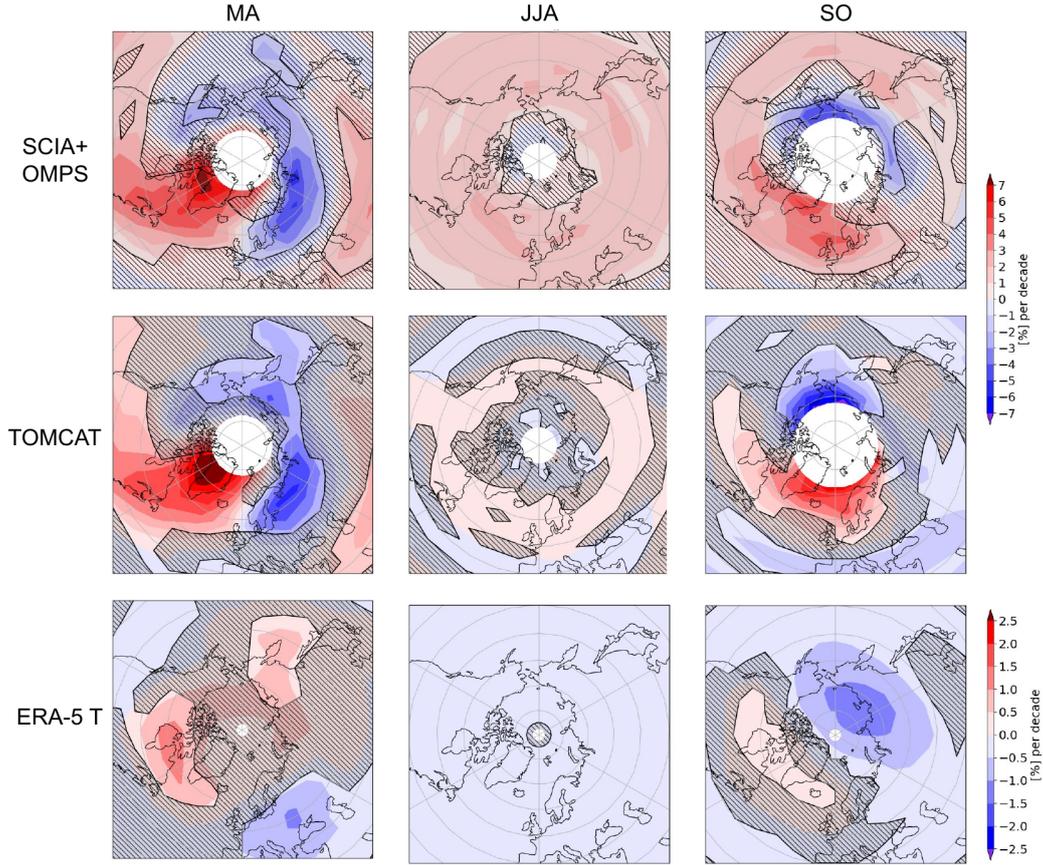
Longitudinally resolved O3 trends TOMCAT, 200401 - 202112



**Figure 5.** Longitudinally-resolved ozone trend cross section at  $70^\circ$  N, over 2004-2021 for three TOMCAT scenarios: (a) reference control run, (b) PSC-inhibited scenario and (c) repeating forcing.



**Figure 6.** Panel (a) shows zonal ozone trends for ERA5 time series over 2004-2021. Panels (b) and (c) show the longitudinally resolved trends at 32 km for ERA5 and TOMCAT, respectively.



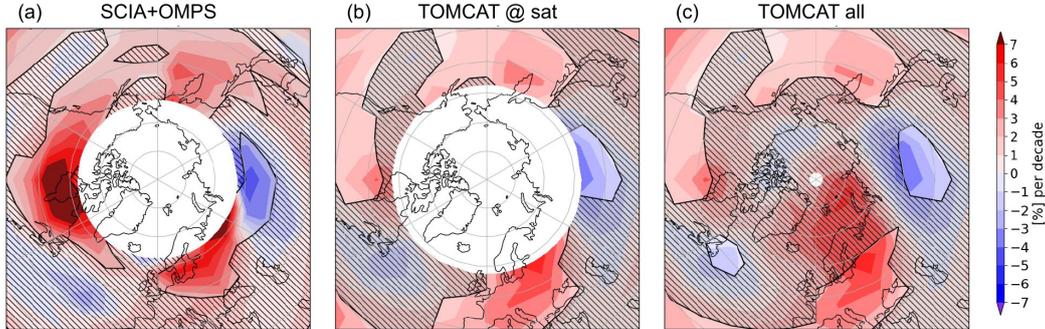
**Figure 7.** Seasonally resolved trends at 32 km from SCIA+OMPS dataset (top row), TOMCAT reference simulation (middle row) and ERA5 temperature (bottom row). The left column shows trends for spring (MA), the middle column for summer (JJA) and the right one for autumn (SO).

325 During summer (JJA, middle column) the trend fields are fairly homogeneous over  
 326 longitude, displaying significant positive values of about 1 % per decade for SCIA+OMPS  
 327 and close to zero for TOMCAT. In contrast, during spring (left column) and autumn (right  
 328 column) the asymmetry is well pronounced. In particular, we notice a strong zonal asym-  
 329 metry in the spring-time trends in SCIA+OMPS that is very well captured by TOM-  
 330 CAT, with the positive maximum located over the North Atlantic sector. The negative  
 331 values between Scandinavia and Siberia are also statistically significant (at  $2\sigma$  level) for  
 332 both observations and model. A similar bi-polar pattern is also found in SO, but more  
 333 confined to polar latitudes and shifted in longitude. The good agreement of TOMCAT  
 334 with observations also holds in this case.

335 Regarding temperature, in summer we find a close-to-zero negative trend, whereas  
 336 in spring and autumn the pattern is also zonally asymmetric, however no strong corre-  
 337 lation with the patterns observed in the ozone trends was found. In conclusion, we find  
 338 no strong evidence to relate the catalytic destruction of ozone in the polar vortex to the  
 339 longitudinal asymmetry pattern observed in the Arctic.

340 A comparison between TOMCAT and SCIA+OMPS during winter months is more  
 341 difficult, as limb scattering observations do not sample polar night conditions, as shown

342 in Fig. 8, panel (a), where trends in DJF are shown at 32 km. SCIA+OMPS shows large  
 343 positive values with maxima over Canada and Scandinavia. The CTM, sampled in the  
 344 same manner as the SCIA+OMPS monthly time series, shows a comparable pattern (panel  
 345 (b)), with less pronounced positive values. The trends calculated using the full TOM-  
 346 CAT profiles (non-sampled, averaged over all the model time steps) give a better pic-  
 347 ture of the two positive (over Canada and Scandinavia) and two negative cores (over Siberia  
 348 and South of Greenland), although mostly not statistically significant.



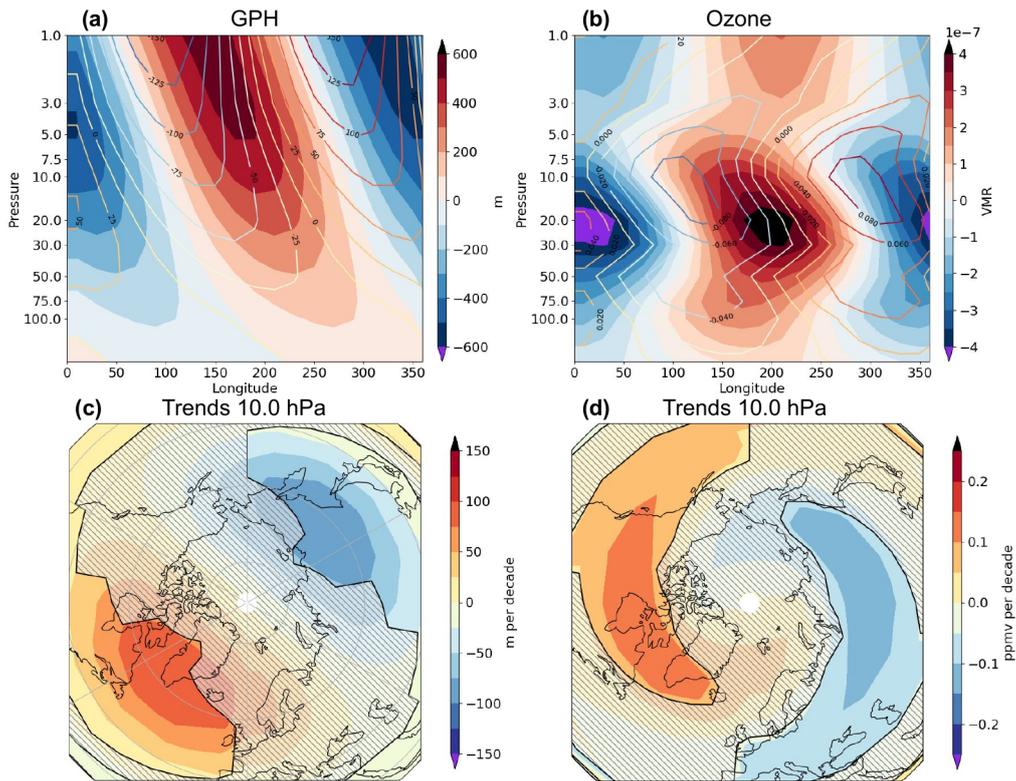
**Figure 8.** Seasonally resolved trends in winter at 32 km from SCIA+OMPS (panel a) and TOMCAT: in panel (b) the CTM simulation was sampled as the satellite data whereas in panel (c) the non-sampled TOMCAT times series was used to compute the trends.

## 349 6 Changes in GPH and atmospheric dynamics

350 To investigate changes in the wave activity at northern high latitudes during the  
 351 last two decades as a cause of the asymmetry in trends, we analyzed the time series of  
 352 geopotential height (GPH) from ERA5. In particular, we considered the longitudinally  
 353 resolved vertical structure of the GPH field and decomposed it in wavenumber one (wave-  
 354 1) and wavenumber two (wave-2) components, using a fast Fourier transform. We focused  
 355 on the January-March period, where the largest asymmetric pattern in trends was found.  
 356 This analysis is based on the theory of linear interference of waves (Smith & Kushner,  
 357 2012), according to which a negative correlation exists between changes in the climato-  
 358 logical stationary wavefield and the stratospheric jet strength.

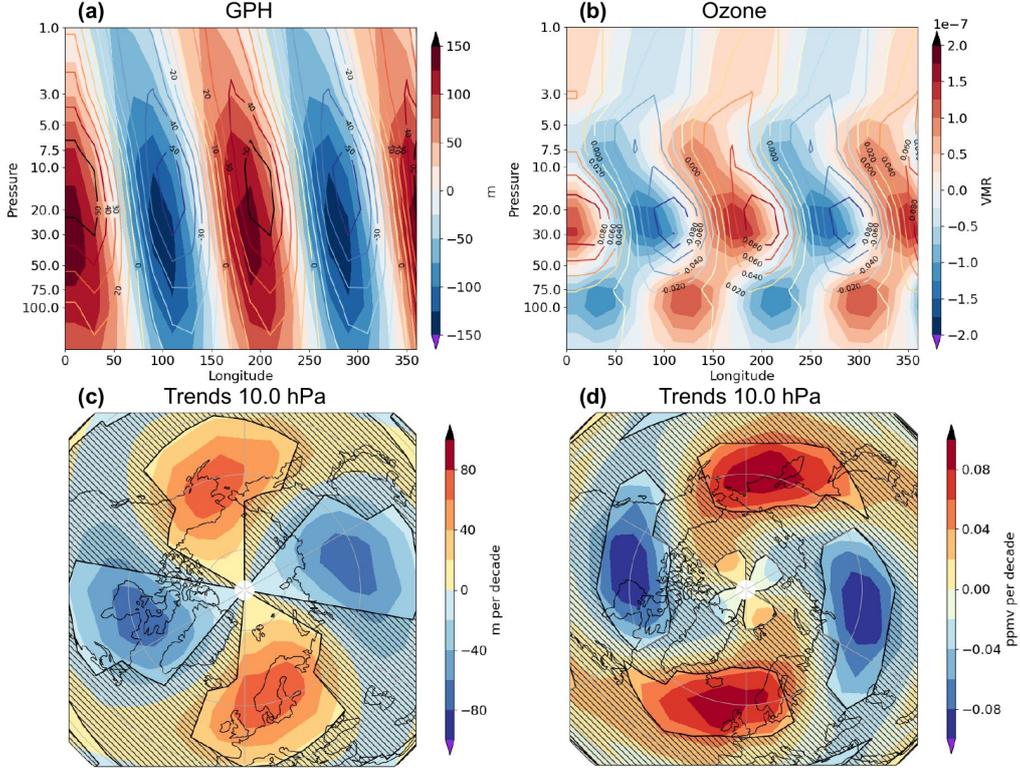
359 First we obtained the 2004-2021 climatology of the wave-1 component, after av-  
 360 eraging the GPH over the  $[45^\circ \text{ N}, 70^\circ \text{ N}]$  latitude band. Then, we computed the linear  
 361 trends of the wave-1 component over the same time period. Fig. 9a shows the wave-1  
 362 climatology in colors and the respective linear trends in m per decade in contours. The  
 363 position of the positive wave-1 GPH anomalies is approximately collocated with the re-  
 364 gion showing a negative trend, and vice-versa, i.e. they are approximately in quadrature.  
 365 In particular, a  $100/120^\circ$  eastward shift between the climatology and the wave-1 trend  
 366 maxima is visible, pointing out an eastward shift in the wave-1 forcing and a weaken-  
 367 ing of the wavenumber-1 planetary wave, according to the linear wave theory (Matsuno,  
 368 1970), over the last two decades.

369 We then performed a similar analysis for the ozone field, choosing the TOMCAT  
 370 time series with a complete coverage of the polar regions. We find a similar baroclinic  
 371 pattern in the climatology of the wave-1 component of TOMCAT ozone particularly in  
 372 the middle stratosphere, as shown in Fig. 9, panel (b). Above 5.0 hPa and below 50.0 hPa  
 373 the correlation between the two panels breaks down. The trends of the ozone anomaly  
 374 wave-1 component are superimposed in panel (b) in ppmv per decade and they are, sim-



**Figure 9.** Top row, JFM climatology of the wave-1 component of GPH (left) and TOMCAT ozone (right) averaged over  $[45^\circ \text{ N}, 70^\circ \text{ N}]$ . Superimposed the trends of the same quantities are shown as contours, with values in m per decade (left) and ppmv per decade (right). Bottom row, the wave-1 trend values at 10.0 hPa are shown (striped regions indicate no statistical significance at  $2\sigma$ ).

375 ilarly to GPH, out of phase w.r.t. their climatological values. Panels (c) and (d) show  
 376 the GPH and ozone trends at 10.0 hPa, respectively, with the striped areas indicating  
 377 values smaller than their  $1\sigma$  uncertainty. The similarity is evident, although the shape  
 378 of the two cores is more elongated for ozone trends.



**Figure 10.** Same as Fig. 9, but for the wave-2 components of GPH and ozone.

379 As shown in Fig. 10, a similar analysis was performed for the wave-2 components  
 380 of ERA5 GPH and TOMCAT ozone anomalies. The climatologies of wave-2 GPH and  
 381 ozone anomalies in the middle stratosphere show again a baroclinic structure, with val-  
 382 ues that are approximately in phase with their respective trends (about  $30^\circ$  east-shift  
 383 between the two maxima or minima). This indicates that the wavenumber-2 wave forc-  
 384 ing in the stratosphere has intensified in the last 2 decades. Panels (c) and (d) show the  
 385 wave-2 GPH and ozone trends at 10.0 hPa, respectively. The similarity is in this case  
 386 striking.

387 To quantify the correlation between climatology and trends, we calculated the pressure-  
 388 weighted pattern correlation as in Fletcher and Kushner (2011) for the GPH pattern and  
 389 found a correlation of -0.75 for wave-1, i.e. out of phase, and of 0.77 for wave-2, i.e. close  
 390 to in-phase. This analysis of the wave-1 and -2 components points out the strong cor-  
 391 relation between changes in ozone and in GPH, which are themselves related to changes  
 392 in wave activity. In our case, the pattern in GPH wave-1 and -2 components is consis-  
 393 tent with a long-term shift and a strengthening of the polar vortex: the weakening and  
 394 shift of the wavenumber-1 planetary wave activity is leading to a strengthening of the  
 395 polar vortex, partially offset by the strengthening of the wavenumber-2 wave activity.  
 396 This seems to be the main driver of the asymmetry in the long-term ozone changes.

397 The identified GPH patterns are consistent with previous literature findings, e.g.,  
 398 Hu et al. (2018), which were related by the authors to a weakening of the Aleutian low  
 399 and to warmer sea-surface temperature over the central North Pacific. However other  
 400 authors, e.g., Zhang et al. (2016); Seviour (2017), pointed out a weakening of the polar  
 401 vortex over the 1980-2010 period. In the next Sect. we directly investigate changes in  
 402 the polar vortex over the last 4 decades to reconcile these findings.

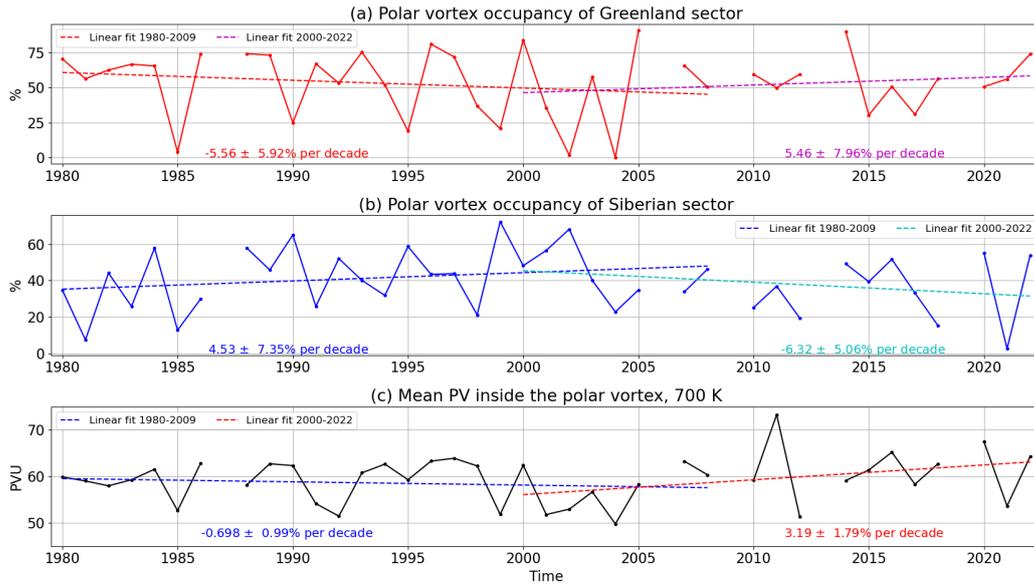
## 403 7 Potential vorticity trends and polar vortex changes

404 In this section we present the changes in the polar vortex over the last four decades,  
 405 following studies such as Zhang et al. (2016). We defined the polar vortex boundary us-  
 406 ing the methodology described in Nash et al. (1996); in particular, we used ERA5 mod-  
 407 ified potential vorticity at 700 K and wind on potential vorticity isolines. 700 K is con-  
 408 sidered to be representative of the middle stratosphere, around 30 km. The determina-  
 409 tion of the polar vortex boundary is based on the peak of the potential vorticity gradi-  
 410 ent in the equivalent latitude space (Butchart & Remsberg, 1986), collocated with a hor-  
 411 izontal wind peak. After determining the polar vortex boundary for the Februaries since  
 412 1980, we investigated the change in its position and strength.

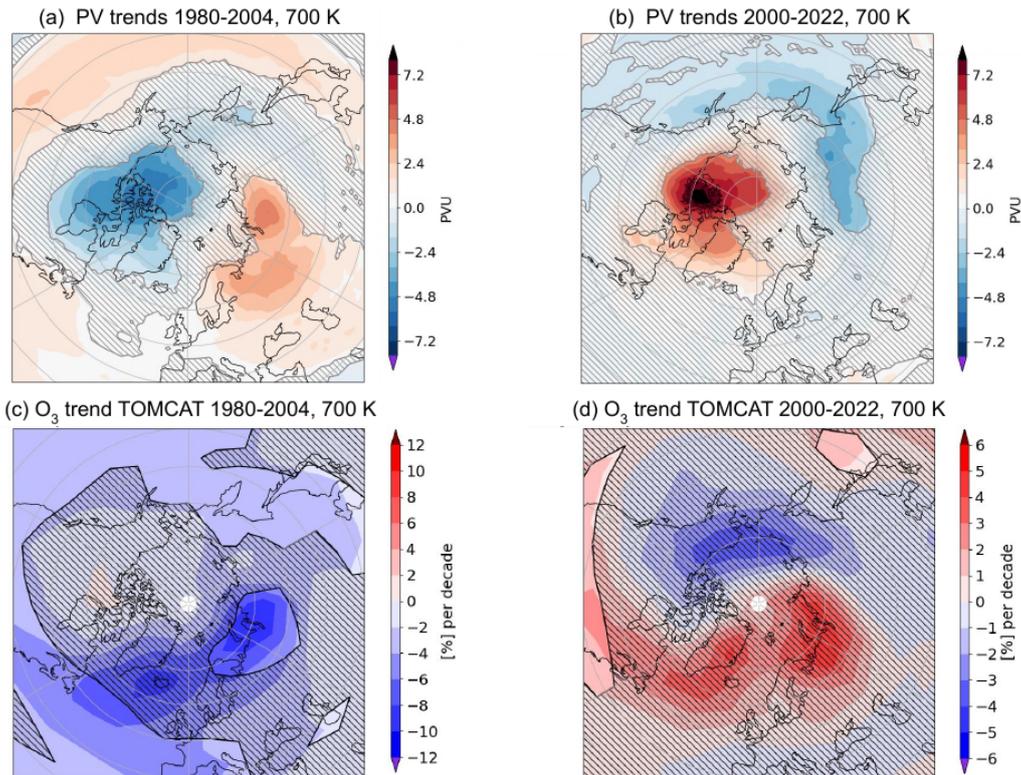
413 We defined two relevant sectors where the ozone asymmetric pattern is relevant:  
 414 the first around Greenland and the second over Siberia, as shown in the Supplements  
 415 in Fig. S3. We computed the polar vortex relative occupancy of these two sectors in each  
 416 February to assess decadal oscillations in the position of the polar vortex. As shown in  
 417 Fig. 11, a change in the linear trends of the sector occupancy occurred at the beginning  
 418 of the century: as reported in Zhang et al. (2016) over the period 1980-2009 the polar  
 419 vortex underwent a shift to the Eurasian sector, however, from the beginning of the cen-  
 420 tury, an opposite shift seems to have occurred. This tendency is not, however, as sound  
 421 as the shift in the previous period, as several years (1987, 2006, 2009, 2013 and 2019)  
 422 needs to be screened out, because of a weak polar vortex or major sudden stratospheric  
 423 warming (SSW) events in February. Due to the high interannual variability, trends are  
 424 mostly not significant, even at  $1\sigma$  level as reported in the panels, and should be consid-  
 425 ered as decadal oscillations rather than a long-term change of the polar vortex.

426 In panel (c) of Fig. 11 the mean potential vorticity inside the polar vortex is shown:  
 427 in the first two decades a negative linear trend is found, i.e. a weakening of the polar vor-  
 428 tex as reported by Zhang et al. (2016) and Seviour (2017). In the last two decades, in  
 429 contrast, the trend becomes positive, indicating a strengthening of the polar vortex as  
 430 reported by Hu et al. (2018). The strengthening of the polar vortex is consistent with  
 431 a positive shift in the Arctic oscillation (Weber et al., 2022).

432 Finally, we investigate the trends of the modified potential vorticity in the middle  
 433 stratosphere (700 K) over the two periods 1980-2004 and 2000-2022. In Fig. 12 top pan-  
 434 els, we clearly see a reversal of the pattern over the polar regions, with panel (a) show-  
 435 ing similar results to the findings of Zhang et al. (2016), pointing out a shift of the po-  
 436 lar vortex to Eurasia, whereas panel (b) indicates a shift of its mean position again to-  
 437 wards North America over the last 20 years. Looking at the ozone trends on the 700 K  
 438 isentropic surface from the TOMCAT time series in the respective periods, shown in  
 439 the bottom row of Fig. 12, we also notice a reversal of the pattern: the negative values  
 440 were largest over the Atlantic/Scandinavian sector during the first period, whereas dur-  
 441 ing the last two decades the positive trends are largest in the same region.



**Figure 11.** Panels (a) and (b) show the relative occupancy of the Greenland sector and of the Siberian sector, respectively, by the polar vortex. Panel (c) shows the mean modified potential vorticity within the polar vortex at 700 K isentropic surface. Respective trends with  $1\sigma$  uncertainties are reported in the panels.



**Figure 12.** Trends of the ERA5 modified potential vorticity (panels (a) and (b)) and of the TOMCAT ozone (panels (c) and (d)) at the 700 K isentropic surface are shown for the periods 1980-2005 and 2000-2022.

## 8 Conclusions

In this study we have presented a comparison between satellite limb observations and simulations from the TOMCAT CTM to investigate the zonal asymmetry in ozone trends identified at northern high latitudes. The OMPS-LP product has been recently updated at the University of Bremen by using the improved L1G data provided by the NASA team, leading to a better long-term stability of the ozone time series w.r.t. the previous version. A preliminary comparison between SCIA+OMPS and TOMCAT time series and zonal trends demonstrated the overall good agreement between the two, when considering deseasonalized anomalies. We then presented the longitudinal asymmetry in trends observed at northern high latitudes over the period 2004-2021, which is well captured not only by the CTM but also by the ERA5 time series, hinting at the dynamical origin of this feature.

By using dedicated TOMCAT runs, we further showed that the identified patterns are dynamically driven, as neither gas-phase chemistry nor heterogeneous chemistry was found to have a relevant direct role in the discussed asymmetry. By investigating the trends at a seasonal level, we found that the asymmetry shows the largest amplitude in late winter/early spring. In this season, we found positive values up to 6-7 % per decade over Greenland and negative values of 3-4 % per decade over Eurasia. This seasonal trend pattern observed in SCIA+OMPS is very well reproduced by TOMCAT.

We decomposed ERA5 geopotential height (GPH) and TOMCAT ozone fields in wave-1 and -2 components for months JFM, finding a strong similarity in the changes of the two quantities in the middle stratosphere. According to the linear wave interference, the findings are consistent with a long-term shift and a strengthening of the polar vortex, i.e. weakening of the wavenumber-1 planetary wave. In this way, it was possible to link the zonal asymmetric pattern in ozone trends to changes in the wave activity in the stratosphere.

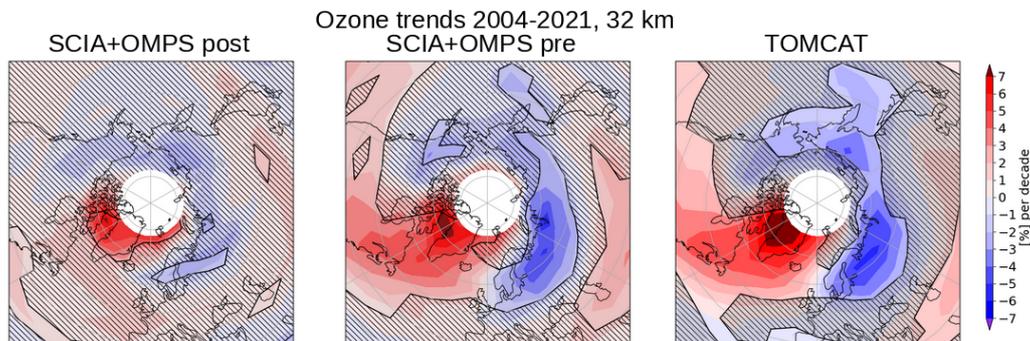
The analysis of the polar vortex position and of the trends in potential vorticity in the middle stratosphere in Sect. 7 qualitatively confirms the proposed relationships between the shift in the mean polar vortex position and the ozone trend asymmetry. The overall pattern underwent decadal changes over the last 40 years, with the last 2 decades seeing a probable strengthening of the vortex and a shift towards North America. This final section of the manuscript is related to the study of the long-term variations of the polar vortex due to climate change and requires further investigations to understand its causes.

In summary, this study has pointed out the role of decadal variations in atmospheric dynamics in explaining ozone trends at northern high latitudes. The observed asymmetry of ozone trends during the past decades is a consequence of decadal climate variability originating in the troposphere. This asymmetric pattern shall be taken into account when calculating ozone trends in the polar region in particular when using ground-based observations, e.g., ozonesondes and Fourier transform infrared spectrometers.

## Appendix A Methods to merge SCIAMACHY and OMPS-LP datasets

For the study of seasonal trends, two approaches have been employed. In the first case, we compute the seasonal averages of the merged monthly SCIA+OMPS dataset. In the second case, the merging is applied to seasonal averages of both dataset anomalies. A filtering is necessary to remove latitude bins for which not all months in the defined season are available or when the latitude coverage of the two instruments differs (at high latitudes). It was found that the second method provides better agreement with CTM simulations compared to the first approach.

490 This is illustrated using, as an example, the March-April (MA) trends at 32 km  
 491 displayed in Fig.A1. The “SCIA+OMPS post” indicates the computation of seasonal av-  
 492 erages using the merged monthly dataset (first method), whereas the case “SCIA+OMPS  
 493 pre” in the middle panel indicates the merging performed on seasonal averages (second  
 494 method). The comparison with TOMCAT significantly improves in the second case.



**Figure A1.** Comparison of seasonal ozone trends in MA at 32 km from SCIA+OMPS (left-most two panels) and TOMCAT (right panel). In the left panel, the merging of the two satellite datasets is performed in terms of monthly time series (“post”); in the central panel, the merging is in terms of averaged seasonal values (“pre”).

## 495 Open Research Section

496 The Merged SCIA+OMPS dataset produced at the University of Bremen and used  
 497 for this study is available at the following link: <https://doi.org/10.5281/zenodo.10033299>.  
 498 TOMCAT simulations and the extracted PV and GPH values used for this study can  
 499 be found, respectively, at: <https://zenodo.org/doi/10.5281/zenodo.10054832> and  
 500 <https://doi.org/10.5281/zenodo.10054575>

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