## The unusual 2020 Arctic ozone hole as seen in the CAMS reanalysis

Antje Inness<sup>1</sup>, Simon Chabrillat<sup>2</sup>, Johannes Flemming<sup>1</sup>, vincent huijnen<sup>3</sup>, Bavo Langenrock<sup>4</sup>, Julien Nicolas<sup>1</sup>, Inna Polichtchouk<sup>5</sup>, and Miha Razinger<sup>1</sup>

<sup>1</sup>ECMWF <sup>2</sup>BIRA-IASB <sup>3</sup>KNMI <sup>4</sup>BIRA <sup>5</sup>European Center for Medium-Range Weather Forecasts

November 23, 2022

#### Abstract

A reanalysis dataset produced by the Copernicus Atmosphere Monitoring service (CAMS reanalysis, 2003 - present day) augmented by ERA5 data for the years before 2003 is used to describe the evolution of the 2020 Arctic ozone season and to compare it with years back to 1979. Ozone columns over large parts of the Arctic reached record low values in March and April 2020 because of an exceptionally cold and persistent Arctic polar vortex. Minimum ozone columns were below 250 DU for most of March and the first half of April, with the lowest values of 211 DU in the CAMS reanalysis found on 18 March. Such low values are extremely unusual for the Arctic. The previous years with similarly strong Arctic ozone depletion were 2011 and 1997 with minimum values of 232 DU and 217 DU, respectively. The performance of the CAMS ozone analysis is assessed by comparison with ozone sonde data. We find a clear sign of chemical ozone destruction with ozone severely depleted in a layer between 80-50 hPa in late March and early April when partial pressure values below 2 mPa were observed. Profiles from the limb sounders ACE-FTS and MLS show clear signs of chlorine activation and the presence of polar stratospheric clouds. Monthly mean ozone columns in March 2020 were up to 180 DU or 40% lower than the CAMS climatology (2003-2019) while values for 2011 and 1997 were lower by 31% and 35% respectively.



#### [JGR-Atmosphere]

Supporting Information for

#### The unusual 2020 Arctic ozone hole as seen in the CAMS reanalysis

A. Inness<sup>1</sup>, S. Chabrillat<sup>2</sup>, J. Flemming<sup>1</sup>, V. Huijnen<sup>3</sup>, B. Langenrock<sup>2</sup>, J. Nicolas<sup>1</sup>, I. Polichtchouk<sup>1</sup> and M. Razinger<sup>1</sup>

<sup>1</sup>ECMWF, Shinfield Park, Reading, RG2 9AX, UK.

<sup>2</sup>Royal Belgian Institute for Space Aeronomy (BIRA-IASB), Ringlaan 3, 1180 Brussels, Belgium

<sup>3</sup>Royal Netherlands Meteorological Institute, De Bilt, the Netherlands.

#### Contents of this file

Text S1 Figures S1 to S3

#### Introduction

Here we present a targeted validation of the Arctic stratospheric ozone fields from the CAMS reanalysis for the period January 2003 to April 2020 to illustrate that the data are of sufficient quality to allow us to study the evolution of the 2020 Arctic stratospheric ozone field.

## S1 Additional validation of Arctic stratospheric ozone from the CAMS reanalysis.

Figure S1 shows a comparison between the CAMS reanalysis and Fourier-Transform Infrared Spectroscopy (FTIR) observations from the Network for the Detection of Atmospheric Composition Change (NDACC) network. A detailed description of the instruments and the applied methodologies for all NDACC FTIR observations can be found in Langerock et al. (2015). FTIR measurement techniques are sensitive to the stratosphere and have a typical uncertainty of 5% for the total column. Figure S1 illustrates that the CAMS reanalysis TCO3 values show a slight underestimation against NDACC FTIR data, which is below 10% for most stations and falls within the measurement uncertainty of 5% for a large part of the timeseries.

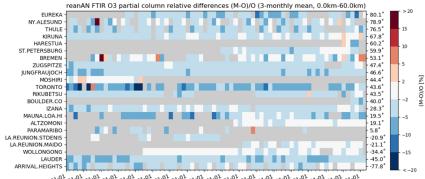


Figure S1. Timeseries of relative TCO3 bias of CAMS reanalysis against FTIR data [(CAMS-obs)/obs] in % for the period January 2003 to April 2020. The stations on the y-axis are sorted by latitude from north (top) to south (bottom). Arctic stations (i.e. north of 600N) are Eureka, Ny-Ålesund, Thule, Kiruna and Harestua.

A comparison of Arctic stratospheric ozone from the CAMS reanalysis with observations from limb scanning satellite instruments MLS (Livesey et al., 2018), Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS; Sheese et al., 2017) and MIPAS (Laeng et al., 2015) is shown in Figure S2. According to Hubert et al. (2016), MLS is among the most stable and accurate instruments for ozone above the UTLS and has a large coverage between 82° South and 82° North; MIPAS has also a good coverage but has some stability issues and tends to globally overestimate ozone concentrations. Both instruments were assimilated in the CAMS reanalysis and therefore should not be considered as fully independent observations. Ozone profile retrievals from ACE-FTS, which has a very sparse coverage, are an independent dataset not assimilated by CAMS. Availability of data from MLS starts in August 2004, ACE-FTS starts in February 2004, MIPAS ends in April 2012. In the middle upper stratosphere (10-30hPa, Figure S2a), the bias is generally smaller than  $\pm 5\%$  for all instruments, except in 2003-2004 when no profile data were available

for assimilation in the CAMS reanalysis leading to larger biases (Inness et al., 2019). As expected, the bias against MLS is very small (<1.5%), with a slight increase since 2018. This increase coincides with the change from using reprocessed MLS V4 data to using NRT MLS V4 data in the CAMS reanalysis in March 2018. A small negative bias against ACE-FTS and MIPAS is almost systematically present. In the middle lower stratosphere (30-70hPa, Figure S2b), the spread of the biases against the different instruments is slightly larger. Here, the bias against MLS is still low (<1.5%), the bias against MIPAS is negative (0-9%) and the bias against ACE-FTS is also negative (0-4%).

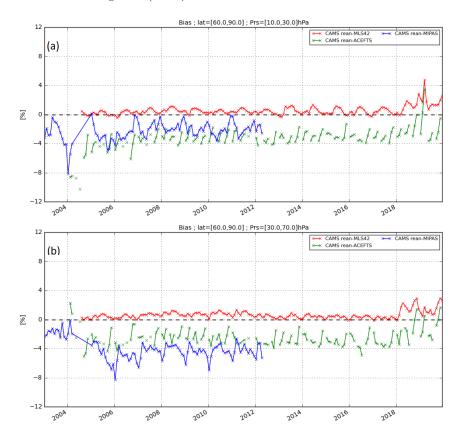


Figure S2. Time series comparing the normalized mean ozone bias [(CAMS-obs)/obs in %] of the CAMS reanalysis with observations from MIPAS (blue), ACE-FTS v3.6 (green) and MLS v4.2 (red) for the period 2003 to 2019 in the (a) middle stratosphere (10-30 hPa averages) and (b) lower stratosphere (30-70 hPa averages) for the Arctic (latitude band 90-600N).

Figure S3 shows relative stratospheric ozone biases from the CAMS reanalysis against ozone sondes averaged over the Arctic and at Ny-Ålesund. The ozone sonde data used for the validation were acquired from a variety of data centres: World Ozone and Ultraviolet Radiation Data Centre (WOUDC), Southern Hemisphere ADditional OZonesondes (SHADOZ), NDACC, and campaigns for the Determination of Stratospheric Polar Ozone Losses (MATCH). The precision of electrochemical concentration cell (ECC) ozone sondes is on the order of  $\pm 5\%$  in the range between 200 and 10 hPa, between -14% and +6% above 10 hPa, and between -7% and +17% below 200 hPa (Komhyr et al., 1995). Larger errors are found in the presence of steep gradients and where the ozone amount is low. The same order of precision was found by Steinbrecht et al. (1998) for Brewer–Mast sondes.

Over much of the stratosphere (10-100 hPa) the biases of the CAMS reanalysis in the Arctic are small and mainly negative (below 5-10%). Larger relative biases are found above 10 hPa and below 100 hPa. An

exception is the year 2004 when no profile data were assimilated in the CAMS reanalysis resulting in a reduced quality of the ozone analysis and larger biases with respect to the independent observations (Inness et al., 2019). The timeseries averaged over the Arctic stations shows increased positive biases after 2015. Such an increase in bias was not seen against the FTIR and satellite data in Figures S1 and S2 and is likely to be the result of changes in the ozone sondes. Stauffer et al. (2020) documented a drop-off in ozone values observed by ozone sondes at several stations after 2014-2016. The reason for this drop-off has to date not been identified. The sondes at Ny-Alesund seem to suffer less from this problem in the stratosphere than other sondes that go into the Arctic mean and we will therefore use the Ny-Alesund sondes further for validation of CAMS reanalysis profiles in Section 3 of the main paper.

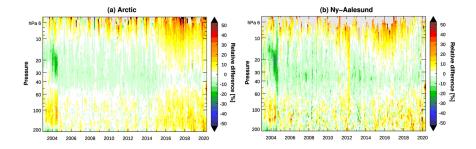


Figure S3. Timeseries of relative biases of stratospheric ozone (200 - 5 hPa) from the CAMS reanalysis against ozone sondes [(CAMS-obs)/obs in %] (a) for all available stations in the Arctic (60-900N) and (b) at Ny-Ålesund (78055'N, 11055'E) for the period January 2003 to April 2020.

The comparisons against FTIR data, limb sounding satellite data and ozone sondes shown in this section illustrate that the CAMS reanalysis generally has a small negative bias (5-10%) for stratospheric ozone in the Arctic, often within the measurement uncertainties of the instruments.

1 2	The unusual 2020 Arctic ozone hole as seen in the CAMS reanalysis
3 4	A. Inness <sup>1</sup> , S. Chabrillat <sup>2</sup> , J. Flemming <sup>1</sup> , V. Huijnen <sup>3</sup> , B. Langenrock <sup>2</sup> , J. Nicolas <sup>1</sup> , I. Polichtchouk <sup>1</sup> and M. Razinger <sup>1</sup>
5	<sup>1</sup> ECMWF, Shinfield Park, Reading, RG2 9AX, UK.
6 7	<sup>2</sup> Royal Belgian Institute for Space Aeronomy (BIRA-IASB), Ringlaan 3, 1180 Brussels, Belgium
8	<sup>3</sup> Royal Netherlands Meteorological Institute, De Bilt, the Netherlands.
9	
10	Corresponding author: Antje Inness (a.inness@ecmwf.int)
11	
12	Key Points:
13	• Arctic ozone columns in spring 2020 were the lowest since 1979
14 15 16 17 18 19	<ul> <li>The Artic polar vortex in spring 2020 was unusually strong, cold and long lasting with reduced wave driving from the troposphere</li> <li>Profiles from limb sounding satellites show clear signs of chlorine activation and the presence of polar stratospheric clouds over the Arctic in March 2020</li> <li>The CAMS reanalysis captures stratospheric ozone well and shows good agreement with ozone sondes reproducing the low Arctic ozone values in late March and early April 2020</li> </ul>
20	
21	

#### Abstract 22

- 23 A reanalysis dataset produced by the Copernicus Atmosphere Monitoring service (CAMS
- reanalysis, 2003 present day) augmented by ERA5 data for the years before 2003 is used to 24
- describe the evolution of the 2020 Arctic ozone season and to compare it with years back to 25
- 1979. Ozone columns over large parts of the Arctic reached record low values in March and 26
- 27 April 2020 because of an exceptionally cold and persistent Arctic polar vortex. Minimum ozone
- columns were below 250 DU for most of March and the first half of April, with the lowest values 28
- of 211 DU in the CAMS reanalysis found on 18 March. Such low values are extremely unusual 29 30
- for the Arctic. The previous years with similarly strong Arctic ozone depletion were 2011 and 1997 with minimum values of 232 DU and 217 DU, respectively. The performance of the CAMS 31
- ozone analysis is assessed by comparison with ozone sonde data. We find a clear sign of 32
- 33 chemical ozone destruction with ozone severely depleted in a layer between 80-50 hPa in late
- March and early April when partial pressure values below 2 mPa were observed. Profiles from 34
- the limb sounders ACE-FTS and MLS show clear signs of chlorine activation and the presence 35
- of polar stratospheric clouds. Monthly mean ozone columns in March 2020 were up to 180 DU 36
- or 40% lower than the CAMS climatology (2003-2019) while values for 2011 and 1997 were 37
- lower by 31% and 35% respectively. 38

#### **Plain Language Summary** 39

- 40 The stratosphere is the layer of the atmosphere between about 15-50 km where most of the ozone
- resides. Usually, Arctic ozone reaches maximum values in the stratosphere during boreal spring. 41
- 42 However, in spring 2020 the Arctic stratosphere was exceptionally cold, and ozone transport
- from the mid-latitudes was inhibited. Because of the low temperatures, polar stratospheric clouds 43
- could form over the Artic region and lead to stratospheric ozone destruction. This led to record 44
- low ozone columns in the Arctic at the end of March and the beginning of April 2020, with 45
- minimum values of 211 DU. This is lower than in 1997 and 2011, the previous two years that 46
- had exceptionally low ozone columns over the Arctic during spring. In spring 2020, the Arctic 47
- ozone layer showed clear signs of chemical ozone depletion with ozone almost completely 48
- destroyed in a layer around 18 km in a way that is usually only seen over the Antarctic during the 49
- 50 austral spring, when the ozone hole forms. We can therefore talk about an Arctic ozone hole in
- spring 2020. 51

#### **1** Introduction 52

While we are used to ozone holes developing over the Antarctic every year during 53 Austral spring (World Meteorological Organization (WMO), 2018), the conditions that are 54 needed for such strong ozone depletion are normally not found in the Northern Hemisphere 55 (NH). There, the winter-time polar vortex is usually weaker and more perturbed than in the 56 Southern Hemisphere (SH), and stratospheric temperatures are not as low as over the Antarctic 57 during austral winter and spring (Waugh et al., 2017). This is the result of greater planetary wave 58 activity in the NH (Shepherd, 2008) because of the larger topographic and land-sea contrast than 59 in the SH. These waves can propagate upwards in the winter hemisphere when stratospheric 60 winds are westerly and dissipate in the upper stratosphere ('wave breaking') decelerating the 61 Arctic polar vortex and leading to a warmer and more perturbed Arctic stratosphere (Garcia and 62 Boville, 1994). Because the Arctic polar vortex is less stable than the Antarctic one, more mixing 63

occurs between the Arctic and ozone-richer midlatitudes than in the SH (Manney et al., 1994). 64

The wave drag also drives a poleward transport in the stratosphere from the ozone source regions in the tropics, with connected downward transport in the extratropics and upward transport in the tropics, as part of the large-scale overturning Brewer Dobson circulation (Brewer, 1949; Dobson, 1956). Because the wave activity is stronger in the NH during winter/spring than in the SH, the Brewer-Dobson circulation is not symmetric in both

- <sup>70</sup> hemispheres (Shepherd, 2008) but leads to a stronger build-up of ozone in the lower stratosphere
- at high northern latitudes during boreal spring than in the SH during austral spring. The
- downward transport at high latitudes also leads to adiabatic warming and overall higher
   temperatures in the lower stratosphere of the NH than the SH. Consequently, fewer polar
- temperatures in the lower stratosphere of the NH than the SH. Consequently, fewer polar
   stratospheric clouds (PSCs) form in the Arctic stratosphere during winter/spring resulting
- 75 generally in less severe catalytic ozone depletion than in the SH during austral winter/spring.

76 Typical total column ozone (TCO3) values at the beginning of winter are around 450 DU in the NH but only around 330 DU in the SH (WMO, 2018). Consequently, even if severe 77 chemical ozone depletion happened in the Arctic the resulting ozone columns would be larger 78 than the very low values (below 100 DU) that were found in some years over the Antarctic (see 79 NASA's Ozone Watch https://ozonewatch.gsfc.nasa.gov/meteorology/annual\_data.html). 80 Manney et al. (2011) argue that ozone values below 250 - 275 DU in the Arctic during winter are 81 exceptional and can be referred to as a NH ozone hole even though 220 DU is the usual SH 82 83 ozone hole threshold.

Ozone columns over large parts of the Arctic reached record low values below 250 DU in 84 85 March and April 2020 with ozone severely depleted in a layer between 80-50 hPa between mid-March and mid-April 2020. The last time similarly low ozone columns were observed over the 86 Arctic was during boreal spring 2011 (Manney et al, 2011) and 1997 (Newman et al., 1997). 87 Lawrence et al. (2020) showed that the record breaking strong polar vortex in winter/spring 2020 88 developed as a combination of weak wave driving from the troposphere and a wave-reflecting 89 configuration in the upper stratosphere. The 2020 Arctic polar vortex was cold enough to allow 90 91 PSCs to form, leading to large stratospheric ozone losses.

92 The Copernicus Atmosphere Monitoring Service (CAMS, atmosphere.copernicus.eu), 93 operated by the European Centre for Medium-Range Weather Forecasts (ECMWF) on behalf of the European Commission, provides daily analyses and 5-day forecasts of atmospheric 94 composition, including analyses of stratospheric ozone in near-real time (NRT). CAMS followed 95 the evolution of the 2020 Arctic ozone depletion in NRT making use of the full 3-dimensional 96 ozone analysis fields to assess the evolution of the TCO3 field as well as the vertical structure of 97 the ozone field. In addition to providing daily NRT analyses and forecasts, CAMS also produces 98 99 a reanalysis of atmospheric composition (Inness et al., 2019), which covers the years from 2003 onwards and again includes stratospheric ozone. This reanalysis uses a single version of the 100 CAMS model and data assimilation system, taking care to minimize changes in the versions of 101 the used emissions or assimilated satellite retrievals. It is now running close to NRT and allows 102 us to intercompare recent years and to derive anomalies against climatologies calculated over the 103 whole period of the CAMS reanalysis (2003-2019) while avoiding spurious effects from changes 104 105 in the model and the assimilation system.

In this paper, we use ozone data from the CAMS reanalysis to document the evolution of the 2020 Arctic ozone field, as well as stratospheric temperatures and zonal winds, and compare stratospheric ozone during the Arctic winter and spring 2019/2020 with other years covered by the CAMS reanalysis. We use data from the ERA5 reanalysis (Hersbach et al., 2020) to extend the TCO3 timeseries back to 1979 and assess the performance of the CAMS reanalysis with ozonesondes and independent satellite retrievals.

The paper is structured in the following way. Section 2 describes the CAMS model and data assimilation system and gives some information about the quality of the CAMS ozone analysis fields. Section 3 looks at the Arctic stratospheric ozone field during the winter 2019/2020 and compares it with the seasons 2010/2011 and 1996/1997, and Section 4 finishes with the conclusions.

## 117 2 Data sets

118 2.1 The CAMS model and data assimilation system

The chemical mechanism of ECMWF's Integrated Forecast System (IFS) is a modified 119 and extended version of the Carbon Bond 2005 chemistry scheme (CB05, Yarwood et al. 2005) 120 chemical mechanism for the troposphere, as also implemented in the chemical transport model 121 (CTM) TM5 (Huijnen et al., 2010). CB05 is a tropospheric chemistry scheme with 55 species 122 and 126 reactions. Stratospheric ozone chemistry in IFS(CB05) is parameterized by a linear 123 ozone scheme (Cariolle and Déqué, 1986; Cariolle and Teyssèdre, 2007). This combination 124 allows a realistic representation of tropospheric and stratospheric ozone concentrations (Inness et 125 al., 2019; Huijnen et al., 2020; Wagner et al., 2020). The chemistry module of the IFS together 126 with its emissions are documented in more detail in Flemming et al. (2015) and Flemming et al. 127 (2017) and the updates used in the CAMS reanalysis in Inness et al. (2019). 128

The IFS uses an incremental four-dimensional variational (4D-Var) data assimilation 129 system (Courtier et al. 1994) and ozone is one of the atmospheric composition fields in the 130 CAMS reanalysis that is included in the control vector and minimized together with the 131 meteorological control variables. More details about the data assimilation system and 132 133 background errors can be found in Inness et al. (2015) and Inness et al. (2019). Ozone retrievals from a range of satellites are assimilated in the CAMS reanalysis. These data include TCO3 134 retrievals from the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY 135 (SCIAMACHY) instrument, the Ozone Monitoring Instrument (OMI) and the Global Ozone 136 Monitoring Experiment-2 (GOME-2), ozone profile data from the Michelson Interferometer for 137 Passive Atmospheric Sounding (MIPAS) and Microwave Limb Sounder (MLS) and partial 138 139 ozone columns from Solar Backscatter ULTra-Violet (SBUV/2). More information about the assimilated ozone retrievals can be found in Inness et al. (2019). Particularly important for a 140 realistic vertical distribution of the ozone analysis in the CAMS system is the assimilation of 141 profile retrievals from MIPAS and MLS (Inness et al., 2013; Flemming et al., 2011; Lefever et 142 al., 2015; Huijnen et al., 2020) that give height resolved ozone information throughout the 143 stratosphere, including data during the polar night. 144

Some validation of the CAMS reanalysis is given in Inness et al. (2019). Wagner et al.
(2020) present a comprehensive validation of the CAMS reanalysis against independent

observations, including a detailed validation of stratospheric ozone, and Huijnen et al. (2020)

provide an evaluation of tropospheric ozone for the 2003-2016 time period. There are also

149 extensive validation reports of the CAMS reanalysis available from

150 https://atmosphere.copernicus.eu/eqa-reports-global-services. These studies show that

stratospheric ozone in the CAMS reanalysis agrees to within  $\pm 5-10$  % with ozonesondes and

satellite observations and does not show any discernible trends in the bias. Additional targeted

validation of the Arctic stratospheric ozone fields from the CAMS reanalysis for the period

154 January 2003 to April 2020 is shown in the supplement and confirms that the CAMS reanalysis

generally has a small negative bias (5-10%) for stratospheric ozone in the Arctic, often within the

156 measurement uncertainties of the instruments. It gives us confidence that the CAMS reanalysis is 157 a useful dataset to study stratospheric ozone and will allow us to describe the evolution of Arctic

- 157 a useful dataset to study stratospheric ozone and will allow us to desc 158 stratospheric ozone field during the winter and spring of 2019/2020.
- stratospherie ozone neta daring the whiter and spring of
- 159 2.2 ERA5 total column ozone record

The CAMS reanalysis only goes back to 2003 because most of the atmospheric 160 161 composition satellite retrievals became available with the launch of the Envisat and Aura satellites in 2002 and 2004, respectively. To extend the timeseries of Arctic TCO3 back in time 162 163 we use ozone columns from the ERA5 reanalysis (Hersbach et al., 2020) for the years 1979 to 2002. This reanalysis is also produced with the ECMWF IFS system and while it does not 164 include a comprehensive tropospheric chemistry it uses the same stratospheric chemistry 165 parameterization as the CAMS reanalysis. Therefore, stratospheric ozone or TCO3 can well be 166 compared with the CAMS reanalysis data. In ERA5, ozone retrievals from GOME, GOME-2, 167 MIPAS, MLS, OMI, SCIAMACHY, SBUV and the Total Ozone Mapping Spectrometer 168 (TOMS) were assimilated as well as ozone sensitive radiances from several infrared sounders: 169 the High-resolution Infrared Radiation Sounder (HIRS), Atmospheric Infrared Sounder (AIRS), 170 Infrared Atmospheric Sounding Interferometer (IASI) and Cross-track Infrared Sounder (CrIS), 171 see Hersbach et al. (2020) for more details. We restrict our use of ERA5 ozone data to TCO3 172 173 fields. These are usually well constrained by the assimilated satellite data, while vertical profiles may suffer from the lack of adequate ozone profile data during some of the years (Hersbach et 174 al., 2020; Dragani, 2011). Hersbach et al. (2020) mention, for instance, that in the northern 175 winter of 1996/1997, the ERA5 ozone values in the upper stratosphere at high northern latitudes 176 are many times larger than normal, but that this problem does not significantly affect total 177 column ozone. 178

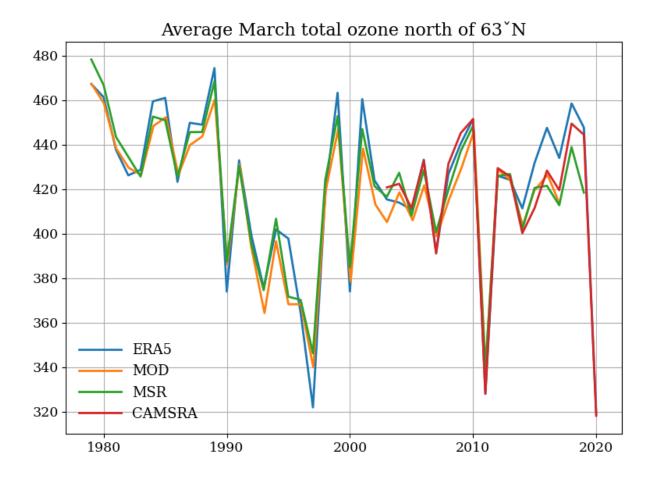
Figure 1 shows the monthly mean TCO3 values for March averaged over the area north 179 of 63°N from the CAMS reanalysis, ERA5, the Multi Sensor Reanalysis (MSR) of total ozone 180 (van der A, 2015) and NOAA's Merged Ozone data set (MOD; Stolarski and Frith, 2006). It 181 shows a good agreement between ERA5, MSR and MOD data going back to 1979 with all data 182 sets reproducing the interannual variability of TCO3 and agreeing to within 5%. ERA5 and 183 CAMS reanalysis TCO3 values agree well for the years where the datasets overlap, but there is 184 an increased discrepancy between ERA5 and the other datasets between 2014 and 2017. Such a 185 positive anomaly was also noted by Hersbach et al. (2020) against a range of other reanalysis 186 datasets and has now been traced back in part to a change from using reprocessed MLS V3 data 187 to NRT MLS data in ERA5 at the beginning of 2015. However, we only use ERA5 data for years 188 before 2003 and the good agreement seen between the CAMS reanalysis and ERA5 for the 189 period 2003-2013 and between MSR, MOD and ERA5 for the earlier years gives us confidence 190 to extend our total column dataset back in time to 1979 using ERA5 data. 191

Figure 1 illustrates that there is a large interannual variability in Arctic ozone in March, much of which is a result of changes in transport (both in the large scale downward transport as well as meridional mixing with midlatitudes), and a downward trend to lower ozone columns in the Arctic during the 1990s (see also WMO, 2018). The figure also shows three years with 196 exceptionally low Arctic ozone columns in March: 2020, 2011 and 1997. These three years will

be compared in more detail in Section 3.3. Low ozone columns are generally associated with a

strong polar vortex, while high values are found when the polar vortex is weak or broken up

- 199 (Newman et al., 1997). The lowest values in the timeseries are found in March 2020 with
- average TCO3 values north of 63°N in the CAMS reanalysis of 318 DU.



201

Figure 1. Timeseries of monthly mean March TCO3 in Dobson Units (DU) averaged over the area north of 63°N from NASA's Merged Ozone data set (MOD, orange), ERA5 (blue), Multi Sensor Reanalysis (MSR, green) and CAMS reanalysis (CAMSRA, red).

205

## 206 **3 Arctic stratospheric ozone during the winter 2019/2020**

3.1 Climatological means of TCO3, temperatures and zonal winds at 50 hPa

Figure 2 shows the climatological monthly mean fields from the CAMS reanalysis

209 (averaged over the period 2003-2019) for TCO3, temperature (T) at 50 hPa and zonal wind (U)

at 50 hPa for the months December to April. The climatological TCO3 values (Figure 2a)

increase from December to March with values above 450 DU in places in February and March.

212 This spring-time maximum is the result of descent in the Arctic stratosphere during the winter

- 213 months and minimum photochemical loss during the polar night. The lowest temperatures at 50
- hPa (Figure 2b) are usually found in December with values between -75°C and -80°C, illustrating
- that the climatological mean temperatures at 50 hPa in the Arctic during winter and spring are
- usually above the threshold for PSC formation (-78°C). The lowest temperatures are found north
- of Scandinavia with values increasing from December through to April as sunlight returns to the polar regions. The low temperatures are an indication of the climatological position of the polar
- vortex and coincide with the lowest TCO3 values. The highest temperatures are found over
- 220 Kamchatka and the Bering Sea. This coincides with the location of the highest ozone columns.
- 221 During winter, the zonal winds are westerly in the stratosphere because of the large-scale
- 222 meridional temperature gradient between the cold pole (no solar heating during the polar night)
- and the warmer mid latitudes. Figure 2c shows the belt of westerlies surrounding the North Pole
- that make up the polar vortex at this altitude, with the strongest zonal winds found in January and
- February. In an average year the polar vortex in the lower stratosphere usually forms in
- November, peaks in January and dissipates in early April (Coy et al., 1997). Figure 2c confirms
- this, showing the weakening of the zonal winds at 50 hPa from February to April, as sunlight
- returns to high northern latitudes and the temperature contrast between the pole and the
- extratropics becomes weaker. The fact that the multi-year means of TCO3, T and u are not
- symmetrical illustrates the impact that topography has on the circulation in the Arctic
- stratosphere.

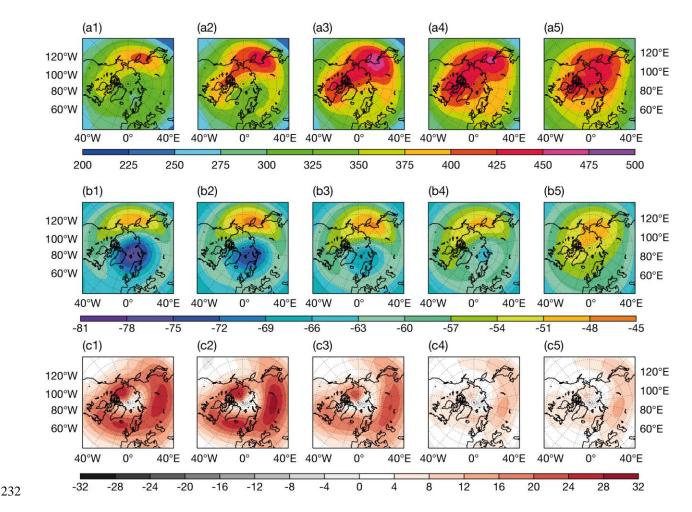


Figure 2. Climatological monthly means of (a) TCO3 (in DU), (b) temperature at 50 hPa (in °C)
and (c) zonal wind (m/s) from the CAMS reanalysis averaged over the years 2003-2019 for
December (column 1), January (column 2), February (column 3), March (column 4) and April
(column 5).

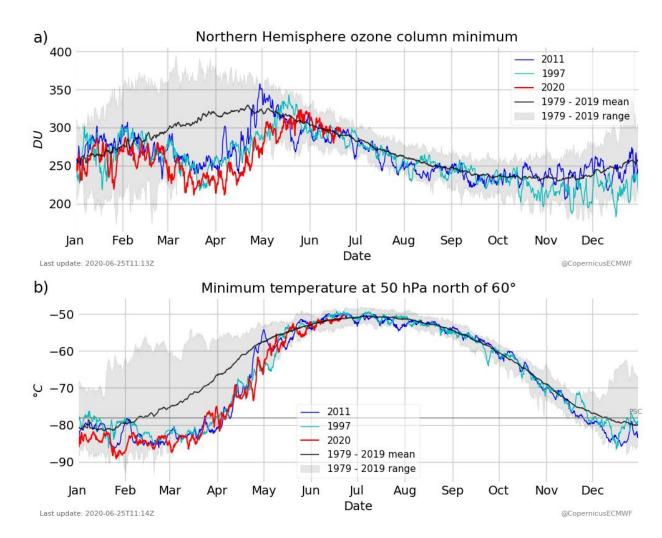
238

### 3.2 Anomalies of TCO3, temperatures and zonal winds at 50 hPa in 2020

239 Figure 3 shows timeseries of Arctic minimum TCO3 and minimum temperatures at 50 hPa (both calculated for the area north of 60°N) from the CAMS reanalysis (2003-2020) and 240 from ERA5 for the earlier years (1979-2002). The grey shading shows the range of the minimum 241 values per day for the whole period 1979-2019 and the solid black line the mean values. The 242 figure illustrates that minimum TCO3 values in the Arctic in late winter/spring show a large 243 spread from year to year and lie between about 200 and 400 DU. Tegtmeier et al. (2008) showed 244 245 that interannual variability in chemistry and the dynamical ozone supply due to transport processes (i.e. meridional mixing across vortex edge, mean transport by residual circulation and 246 diabatic descent in polar vortex) contribute equally to the variability of Arctic wintertime TCO3. 247 The average minimum TCO3 values increase from January to April and lie above 250 DU the 248 whole time. In 2020 (red line in Figure 3a) the situation is completely different. While minimum 249

TCO3 values in January and February 2020 lie within the range of minimum values observed in other years, in March and April 2020 they are the lowest observed ozone columns in the whole record with the absolute minimum value of 211 on 18 March 2020. The previous two years with exceptionally low ozone values (2011 and 1997) are also shown in Figure 3 and will be discussed in more detail in Section 3.3 below.

The timeseries of minimum temperatures at 50 hPa (Figure 3b) shows that the lowest 255 temperatures are found during the winter months December and January and that on average 256 temperatures increase from January onwards as sunlight returns to the polar regions. We also see 257 a large spread in minimum temperature values with some years having values no lower than -258 60°C while others have minimum temperatures below the PSC formation threshold of -78°C. 259 This illustrates the large interannual variability in the Arctic stratosphere. The mean temperatures 260 are just below the threshold for PSC formation for about two months from mid-December to 261 early February but above it for the rest of winter and spring. Minimum temperatures in 2020 262 were some of the lowest in our record and below -78°C from the beginning of January until the 263 last week of March. This was low enough for PSCs to form and suggests that the low Arctic 264 ozone columns seen in 2020 are at least in part the result of in-situ chemical. In particular, the 265 low temperatures in March, when more of the vortex is illuminated by the sun, create a strong 266 potential for catalytic ozone destruction. This will be discussed further in section 3.3. below. 267



269

Figure 3. (a) Timeseries of minimum TCO3 (in DU) and (b) minimum temperature (in <sup>o</sup>C) at 50 270 hPa north of 60°N from CAMS reanalysis (covering the years 2003-2020) and ERA5 (1979-271 2002). The shaded grey area shows the range of values per day over the entire period 1979-2019, 272 the black curve the mean values averaged over the period 1979-2019, the red curve the values for

273

274 2020, the blue curve the values for 2011 and the cyan curve the values for 1997.

Figure 4 shows the monthly mean anomalies of TCO3, T and u at 50 hPa for December 275 2019 to April 2020 against the climatologies from the CAMS reanalysis (Figure 2). 276 Anomalously low TCO3 values (Figure 4a) were found over the Arctic during January to April 277 2020 with the largest negative anomaly of more than -180 DU seen in March 2020, about 40% 278 below the climatological values. The negative ozone anomaly lasted well into April when TCO3 279 values were still more than 140 DU (or 35%) below the climatology. These ozone anomalies 280 were associated with the anomously strong and long-lasting polar vortex illustrated by the 281 stronger than average zonal winds at 50 hPa (Figure 4c) between January and April 2020, with 282 the largest positive anomaly in March 2020. The polar vortex lasted well into April before it split 283 284 into two around 19 April 2020. It was very stable and remained centered near the North Pole throughout, making it more akin to the polar vortex seen over Antarctic during winter/spring, and 285

- less like the more perturbed type usually found over the Arctic. The polar vortex was
- considerably colder than usual throughout the winter/spring of 2020 (Figure 4b). The largest
- temperature anomalies at 50 hPa were found in February and March 2020, when temperatures
- were more than 18°C and 21°C, respectively, below the climatological values. The low
- temperatures continued into April 2020 giving the potential for catalytic ozone depletion at a
- time when sunlight had returned to the Arctic region.

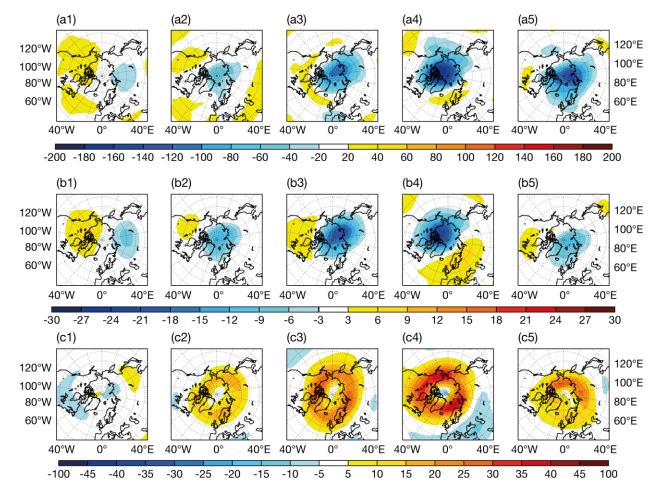


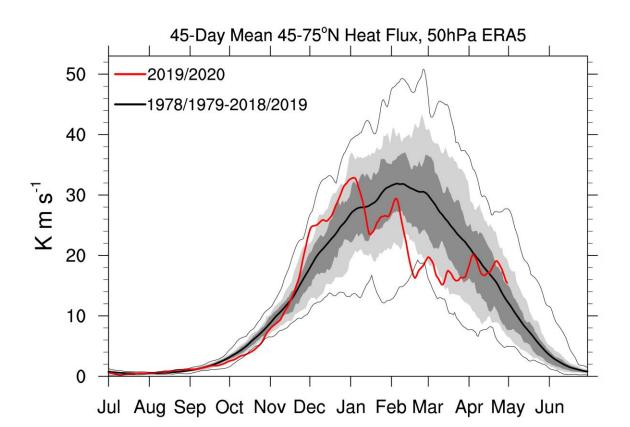
Figure 4. Monthly mean anomalies of (a) TCO3 (in DU), (b) temperature at 50 hPa (in °C) and (c) zonal wind (m/s) from the CAMS reanalysis against the climatology calculated over the years 2003-2019 (see Figure 2) for December 2019 (column 1), January 2020 (column 2), February 2020 (column 3), March 2020 (column 4) and April 2020 (column 5).

Newman et al. (2001) showed that the temperatures of the Arctic lower stratosphere in 297 early March are driven by the strength and duration of planetary waves propagating into the 298 stratosphere in mid to late winter. This can be illustrated by looking at the poleward eddy heat 299 flux in mid to late winter, a key indicator of the upward propagation of planetary waves, which is 300 highly correlated with the mean polar stratospheric temperature during late winter. Low 301 302 stratospheric temperatures are the result of weak wave forcing, and higher than average temperatures the result of strong wave forcing. Figure 5 shows the poleward 45-day running 303 mean eddy heat flux at 50 hPa averaged between 45°N and 75°N from ERA5 for the 304

winter/spring 2019/2020 compared to the years from 1979 onwards. It illustrates that spring 2020
 was characterized by below average planetary wave driving between January and the end of
 March and that values between mid-February to mid-March were below the 10th percentile of

the 1979-2019 period. This low wave driving led to the large cold temperature anomalies and

309 exceptionally strong polar vortex seen in Figure 4.



310

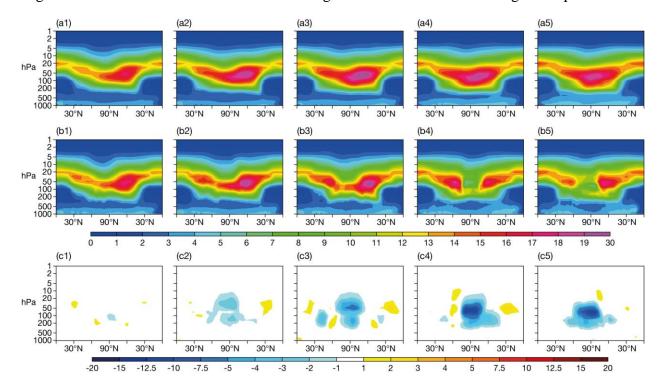
Figure 5. Eddy heat flux in Km/s at 50 hPa averaged between 45°N and 75°N for the 45-day period prior to the date indicated, calculated from ERA5 data. The light shading is the 10th to 90th percentile and the darker shading the 75-25th percentile. The thin black lines are maximum and minimum values and the red line the values for winter/spring 2019/2020.

The 3-dimensional distribution of ozone in the stratosphere as well as the corresponding 315 vertical ozone anomalies are shown in Figure 6. Figure 6a shows cross sections of climatological 316 monthly mean ozone partial pressure values (2003-2019) from the surface to the upper 317 stratosphere along one meridian (0°E/180°E) over the North Pole. They illustrate that the ozone 318 layer is at higher altitudes in the tropics where maximum ozone partial pressure values are found 319 around 20 hPa and at lower altitudes over the North Pole. Figure 6a also illustrates increased 320 ozone maximum values in the lower stratosphere in the Arctic during February and March 321 associated with the large scale diabatic descent. The cross sections for the winter/spring months 322 of 2020 (Figure 6b) are very different from the climatologies. In 2020, the ozone layer is 323 324 considerably thinner than in the climatologies. Ozone values in the layer decrease and anomalies

- against the climatology (Figure 6c) increase from January to March, and values over the North
- Pole remain low in April 2020.

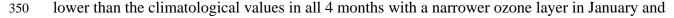
327 While some of the lower ozone values were the result of reduced meridional mixing and

- reduce diabatic downwelling during spring 2020 as a result of reduced wave activity as seen in
- low eddy heat fluxes at 50 hPa (Figure 5), there is the clear signature of chemical ozone
   depletion (see also Figure 12 below) leading to the extremely low ozone values over the North
- Pole in March and April 2020. In March 2020 ozone values in the ozone layer over the North
- pole are reduced to around 4 mPa in the monthly mean, more than 10 mPa below the
- climatological values. These low values last into April 2020. The anomaly plots for 2020 (Figure
- 6c) illustrate the vertical extent of the ozone anomalies during spring 2020, with the largest
- negative anomalies found over the Arctic during March and anomalies lasting into April.



**Figure 6.** (a) Cross sections of monthly mean climatologically averaged ozone partial pressure (in mPa) from the CAMS reanalysis (2003-2019) along one meridian (0°E/180°E) from the equator north via the North Pole and back to the equator, (b) corresponding cross sections for 2019/2020 and (c) cross sections of anomalies for 2019/2020 for December (column 1), January (column 2), February (column 3), March (column 4) and April (column 5).

342 Figure 7 compares ozone and temperature profiles for January to April 2020 from ozone sondes at Ny-Ålesund with profiles from the CAMS reanalysis and against climatological ozone 343 profiles. Ozone sondes are not assimilated in the CAMS reanalysis and therefore are completely 344 independent validation data. The climatological monthly ozone profiles at Ny-Ålesund show the 345 increase of ozone values in the lower stratosphere due to the large scale diabatic descent between 346 January and April with a broader ozone layer and increased partial pressure values between 50 347 and 200 hPa. The climatological monthly means calculated from the CAMS reanalysis agree 348 very well with sonde averages. Figure 7a shows that in 2020 the monthly mean ozone values are 349



- February 2020, which must be the result of reduced large-scale transport and associated downwelling in 2020. This is followed by a clear indication of ozone depletion in March 2020
- and April 2020, when the 2020 monthly mean values are below 5 mPa between 50 and 70 hPa.
- 354 Such ozone profiles are very unusual for the Arctic in boreal spring and much more akin to
- ozone profiles found over the Antarctic during the SH ozone hole season in October. The
- temperature profiles (Figure 7b) show that 2020 is considerably colder than the CAMS
- climatology between 30-200 hPa throughout the period, with monthly mean temperatures below
- -78°C in January, February and March. Figure 8 shows four individual ozone profiles at Ny-
- Ålesund from the CAMS reanalysis and sondes and illustrates that in the last week of March
- 2020 ozone was almost completely depleted in a layer between 50 and 80 hPa, with partial
- <sup>361</sup> pressures below 2 mPa. This layer of low ozone values remained visible in ozone sonde profiles
- at Ny-Ålesund until 15 April 2020 (see also Figure 11 below), while the subsequent ascent on 22
- April 2020 had values between 10-18 mPa all the way between 20-200 hPa, indicating the end of
- the 2020 Arctic 'ozone hole' at Ny-Ålesund.

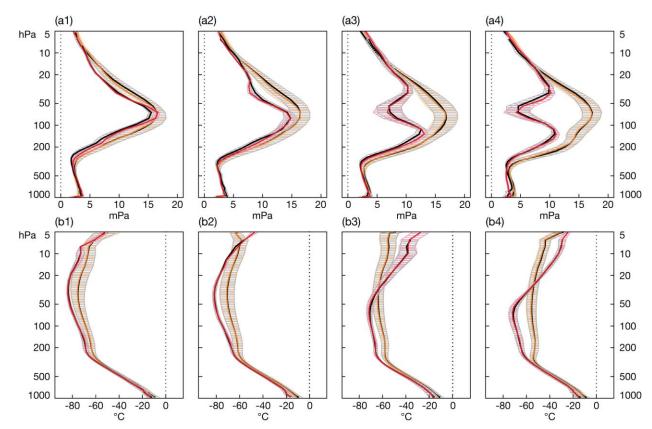
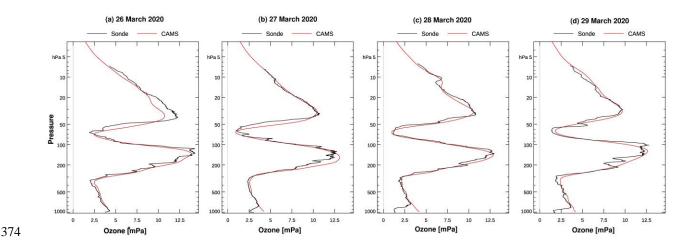
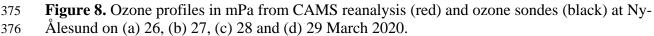


Figure 7. Comparisons of (a) ozone (in mPa) and (b) temperature (in °C) profiles from the
CAMS reanalysis and ozonesondes at Ny-Ålesund for January (column 1), February (column 2),
March (column 3) and April (column 4). The red profiles show monthly mean CAMS reanalysis
values for 2020, the orange profiles the climatological mean from the CAMS reanalysis (20032019). Black lines show the corresponding means from the ozone sondes. The hatched area
depicts +/- one standard deviation. The number of ozone profiles that went into the averages

- 372 were: January 2020: 6, February 2020: 8, March 2020: 16, April 2020: 13, January climatology:
- 187, February climatology: 178, March climatology: 177 and April climatology: 98.





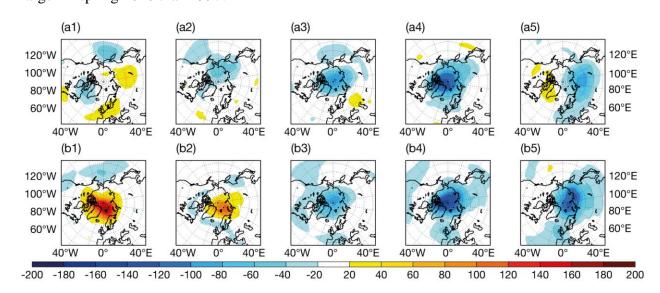
377 3.3. Comparison of Arctic ozone in spring 2020 with 2011 and 1997

The timeseries of average TCO3 north or 63°N in Figure 1 shows how exceptionally low 378 ozone columns over the Arctic were during March 2020 with averaged Arctic values down to 379 318 DU. Only two other years in the timeseries have had similarly low Arctic mean ozone values 380 in March since 1979: 2011 with 327 DU and 1997 with 321 DU. In all three years, it was the 381 unusual meteorological situation with a long lasting and cold polar vortex centered over the 382 North Pole (Manney et al., 2011; Coy et al., 1997) that led to exceptional ozone losses. All three 383 years were characterized by weak planetary wave driving from the troposphere (Newman et al., 384 2001; Hurwitz et al., 2011; Shaw and Perlwitz, 2014) and seen in Figure 5 for 2020. The 385 timeseries in Figure 3a shows the daily TCO3 minimum values for 2020, 2011 and 1997 and 386 illustrates that from late January until the end of April the minimum values in 2020 were lower 387 than in 2011 and 1997. The lowest TCO3 minimum value in 2020 was 211 DU (on 18 March), 388 compared to 232 DU in 2011 (on 26 February) and 217 DU in 1997 (on 21 March). The 389 timeseries of the minimum temperatures at 50 hPa north of 60°N (Figure 3b) shows that in 1997 390 temperatures were around average until early February and then fell in February. Temperatures 391 in the first half of January 2011 were also around or above average, but in 2020 temperatures in 392 the lower stratosphere were below average from the beginning of January and remained below 393 the PSC formation threshold of -78°C until the last week of March. 394

Figure 9 shows the monthly TCO3 anomalies for the winters 1996/97 and 2010/2011 395 against the CAMS climatology (using the same reference period 2003-2019). The corresponding 396 anomalies for 2019/2020 can be found in Figure 4. All three winters had a long-lasting, cold 397 polar vortex centered over the North Pole. However, in 1996/97 the polar vortex did not form 398 until late December and was weaker than normal with above average temperatures until the end 399 of January (Coy et al., 1997). The vortex was then strong and centered around the North Pole 400 until it broke up in late April. As a result, we see positive TCO3 anomalies in December 1996 401 and January 1997 (Figure 9b), while in January 2020 and 2011 (Figure 9a) negative TCO3 402

anomalies can already be seen over the North Pole. In February, March and April large negative
TCO3 anomalies are found in all three winters with the largest overall anomalies found in 2020
when values were about 27% (February), 40% (March) and 35% (April) below the CAMS
climatology. This compares with anomalies for February, March and April in 2011 of -25%, 31% and -22%, respectively, and -22%, -35% and -30% in 1997.

The situation in 2020 looks even more exceptional, especially compared to 1997, when one considers that Arctic ozone columns were at their lowest during the 1990s owing to the higher concentrations of ozone depleting substances (WMO, 2018) and have been slowly recovering (Steinbrecht et al., 2017). The lower TCO3 background values in 1997 can be seen in the negative anomalies in midlatitudes in 1996/1997 (Figure 9). Despite the lower TCO3 background values in 1997, the negative ozone anomalies against the CAMS climatology were larger in spring 2020 than 1997.

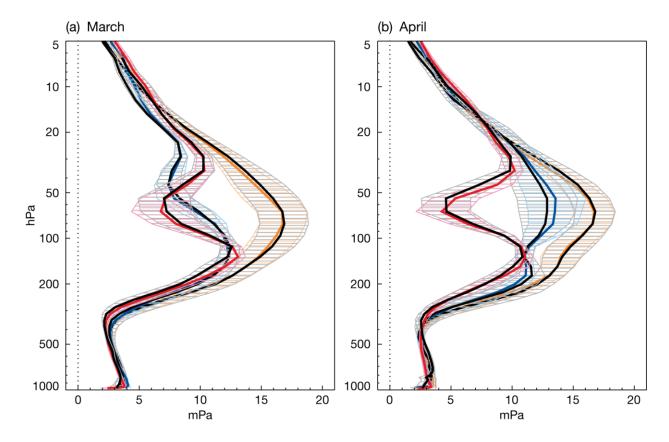


415

Figure 9. Monthly mean anomalies of TCO3 from (a) the CAMS reanalysis for 2010/2011 and
(b) ERA 5 1996/1997 against climatologies calculated over the years 2003-2019 (see Figure 2)
for December (column 1), January (column 2), February (column 3), March (column 4) and
April (column 5).

In Figure 10 we compare ozone profiles at Ny-Ålesund from sondes and the CAMS 420 reanalysis for March and April 2020 and 2011 with the CAMS climatology. The figure shows 421 422 that in 2020 the exceptionally low ozone values in the lower stratosphere lasted into April, while 423 in 2011 values had already increased again in April. Figure 11 shows timeseries of ozone profiles at Ny-Ålesund from sondes and the CAMS reanalysis for 2020 and 2011. Figure 11 shows 424 425 clearly very low ozone partial pressure values in 2020 between mid-March to mid-April in the altitude range of 50-80 hPa. In 2011 low ozone partial pressure values were also seen throughout 426 427 March, located at slightly higher altitude, but minimum values were not as low as in 2020. The period of very low ozone values ended at the end of March 2011 and by 4 April 2011 ozone 428 profiles at Ny-Ålesund had values over 10 mPa everywhere between 30-200 hPa. However, this 429 recovery did not happen until much later in 2020. An ascent at Ny-Ålesund on 15 April 2020 still 430 431 showed ozone partial pressures down to 2 mPa between 60-70 hPa and only the subsequent

432 ascent on 22 April 2020 did not show the low values any more.



433

**Figure 10**. Monthly mean ozone profiles in mPa at Ny-Ålesund for (a) March and (b) April. The red profiles show the 2020 monthly means, the blue profiles the 2011 monthly means and the orange profiles the climatological means from the CAMS reanalysis (2003-2019). Black lines show the corresponding means from the ozone sondes. The hatched areas depict +/- one standard deviation. The number of ozone profiles that went into the averages are: March 2020: 16, March 2011: 17, March climatology: 177, April 2020: 13, April 2011: 10 and April climatology: 98.

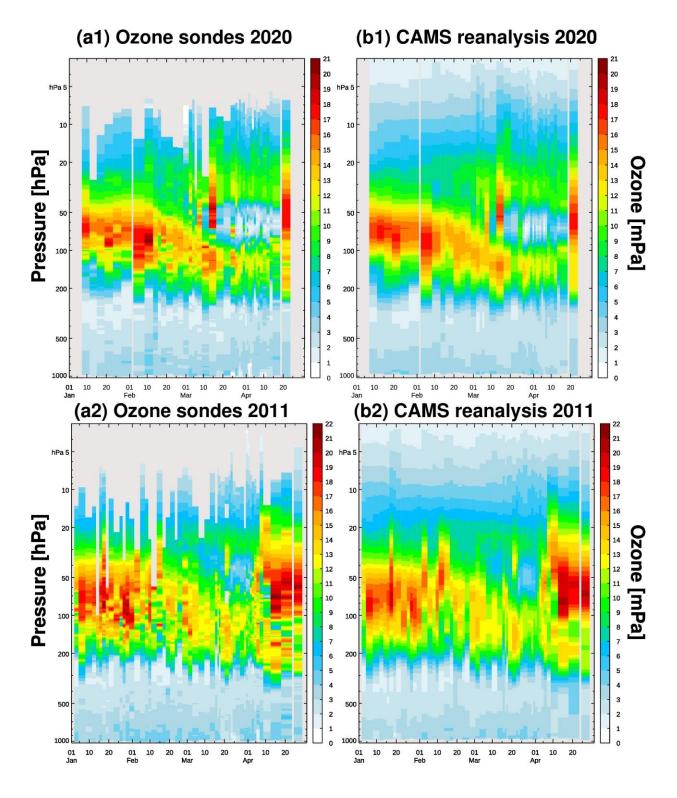
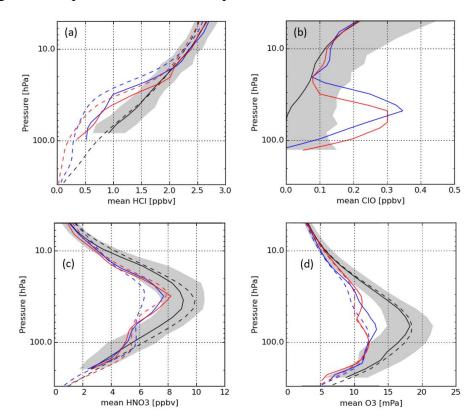


Figure 11. Timeseries from January to April of ozone profiles (in mPa) at Ny-Ålesund from ozone sondes (a) and the CAMS reanalysis (b) for 2020 (row 1) and 2011 (row 2).

Evidence that the exceptionally low ozone values seen in 2020 (and 2011) were the result 443 of chemical ozone depletion comes from the Atmospheric Chemistry Experiment - Fourier 444 Transform Spectrometer (ACE-FTS, Sheese et al., 2017) and MLS (Livesey et al., 2018) profiles 445 (Figure 12). Monthly mean ASC-FTS and MLS profiles averaged over the latitude band 60-90°N 446 show reduced concentrations of the chlorine reservoir species hydrochloric acid (HCL) and 447 exceptionally high abundances of chlorine monoxide (CLO) during March 2020 and 2011 448 relative to the ACE-FTS or MLS climatologies. There are also signs of denitrification in both 449 years with reduced concentrations of nitric acid (HNO3). This is evidence that PSCs formed 450 during spring 2020 (and 2011), chlorine activation happened and catalytic ozone depletion led to 451 the very low ozone values seen during March 2020 (and 2011). Overall MLS and ACE-FTS 452 453 agree very well, despite the much sparser sampling of ACE-FTS. The only exception is HNO3 where ACE-FTS indicates more HNO3 depletion in 2011 than in 2020 while MLS shows nearly 454 the same abundances. This apparent disagreement is due to the more limited sampling in ACE-455 FTS, i.e. during the first half of March 2011 ACE-FTS happened to sample latitudes with very 456 strong HNO3 depletion while MLS sampled more locations. 457



458

**Figure 12.** (a) HCL, (b) CLO, (c) HNO3, (d) ozone mean vertical profiles for March from MLS v4.2 retrievals (dashed lines) and ACE-FTS v3.6 retrievals (solid lines) and poleward of 60°N. Shown are 2005-2019 climatologies excluding 2011 (black lines), March 2011 (blue line) and March 2020 (red lines). The grey envelopes represent two standard deviations of all profiles in each MLS climatology. All retrievals are expressed in volume mixing ratios [ppbv] except for ozone which is in partial pressure [mPa].

465

## 466 4 Conclusions

Data from a reanalysis of atmospheric composition produced by the Copernicus 467 Atmosphere Monitoring Service, the so-called CAMS reanalysis, augmented by ERA5 data for 468 years prior to 2003 were used to assess stratospheric Arctic ozone during the winter and spring of 469 2019/2020 and to compare it with other years since 1979. During winter and spring 2020, the 470 Arctic polar vortex was exceptionally strong, long-lived, cold and remained centered around the 471 North Pole until the second half of April. This was the result of weak planetary-scale wave 472 forcing during mid to late winter. For several months, minimum temperatures in the Arctic lower 473 stratosphere were low enough to allow the formation of PSCs, and ozone columns over large 474 parts of the Arctic reached record low values in March and April 2020. Minimum total column 475 ozone values were below 250 DU for most of March and the first half of April with the lowest 476 values of 211 DU found on 18 March 2020 in the CAMS reanalysis. Such low ozone columns 477 are extremely unusual for the NH winter/spring. They were even lower than during the 478 previously recorded cases of large Arctic ozone depletion in 2011 and 1997 when the 479 CAMS/ERA5 dataset had minimum values of 232 DU and 217 DU, respectively. Monthly mean 480 Arctic TCO3 values in March 2020 were up to 180 DU or 40% lower than the CAMS 481 climatology (calculated over the period 2003-2019) while values for 2011 and 1997 were up to 482 31% and 35% lower, respectively. 483

While some of the lower ozone values were the result of reduced diabatic downwelling 484 and reduced meridional transport during spring 2020 because of reduced tropospheric planetary 485 wave activity, there is evidence that chemical ozone destruction occurred and led to extremely 486 low values in the ozone layer over the North Pole in March and April 2020. Profiles from MLS 487 and ACE-FTS show large increases in ClO and reductions in the concentrations of HNO3 and 488 HCl in March 2020, consistent with the presence of PSCs. Ozone profiles from sondes and the 489 CAMS reanalysis at Ny-Ålesund show that ozone was severely depleted in a layer between 80-490 50 hPa where ozone partial pressure values below 2 mPa were observed at the end of March 491 492 2020. The layer of severely depleted ozone concentrations lasted until the middle of April 2020 as seen in CAMS ozone profiles and ozone sondes, longer than in 2011 when low ozone values 493 ceased at the end of March. 494

The CAMS reanalysis was able to capture the exceptional Arctic ozone hole of 2020 well 495 with good agreement between CAMS ozone profiles and independent ozone sonde data. Studies 496 are on-going to determine why tropospheric planetary wave forcing was so weak in winter/spring 497 2020 and if there was any feedback from the Arctic ozone depletion onto the dynamics that 498 helped to prolong the strong vortex. An interesting question would also be to investigate whether 499 500 and to what extent ash and SO<sub>2</sub> that were injected into the Arctic stratosphere after the Raikoke eruption in June 2019 helped to enhance PSC formation and the subsequent ozone depletion in 501 the Arctic during spring 2020. It remains to be seen if this kind of Arctic ozone hole will 502 continue to be an exceptional event or occur more often in a changing climate. 503

## 504 Acknowledgments, Samples, and Data

505 The Copernicus Atmosphere Monitoring Service is operated by the European Centre for

506 Medium-Range Weather Forecasts on behalf of the European Commission as part of the

507 Copernicus programme (http://copernicus.eu). CAMS reanalysis data are freely available from

the CAMS Atmosphere Data Store (ADS, https://ads.atmosphere.copernicus.eu/cdsapp#!/home)

and ERA5 data are available from the Climate Data Store (CDS,

- 510 https://cds.climate.copernicus.eu/#!/home). Thanks to Luke Jones for providing the tools for the
- validation against ozone sondes and to Anabel Bowen for improving some of the figures.
- 512 Thanks to the data providers of the data assimilated in the CAMS reanalysis and the data used
- for the validation studies in this paper. The FTIR data used in this publication were obtained as
- 514 part of the Network for the Detection of Atmospheric Composition Change (NDACC) and are
- 515 publicly available (see http://www.ndacc.org). The MSR data was downloaded from the Temis
- 516 website (http://www.temis.nl). ACE-FTS is an instrument of the Canadian-led Atmospheric
- 517 Chemistry Experiment (also known as SCISAT) that is mainly supported by the CSA and
- 518 NSERC. We acknowledge the MLS mission scientists and associated NASA personnel for the
- 519 production of the data used in this study. The ozone sondes at Ny-Ålesund were launched by the
- 520 Alfred-Wegener-Institut Helmholtz-Zentrum für Polar- und Meeresforschung (AWI).
- 521

## 522 **References**

- Brewer, A. W. (1949), Evidence for a world circulation provided by measurements of helium
  and water vapour distribution in the stratosphere, *Q. J. R. Meteorol. Soc.*, 75, 351,
  doi:10.1002/qj.49707532603
- Cariolle, D. and Déqué, M. (1986), Southern hemisphere medium-scale waves and total ozone
   disturbances in a spectral general circulation model, *J. Geophys. Res.*, *91*, 10825–10846
- Cariolle, D. and Teyssèdre, H. (2007), A revised linear ozone photochemistry parameterization
   for use in transport and general circulation models: multi-annual simulations, *Atmos. Chem. Phys.*, 7, 2183–2196, https://doi.org/10.5194/acp-7-2183-2007
- Courtier, P., Thépaut, J.-N., and Hollingsworth, A. (1994), A strategy for operational
   implementation of 4D-Var, using an incremental approach, *Q. J. Roy. Meteor. Soc.*, 120,
   1367–1388
- Coy L., P. A. Newman, and E. R. Nash (1997), Meteorology of the polar vortex: March 1997,
   *Geophys. Res. Lett.*, Vol. 24, No. 22, 2693-2696.
- Dee, D. P. and Uppala, S. (2009): Variational bias correction of satellite radiance data in the
   ERA-Interim reanalysis, *Q. J. Roy. Meteor. Soc.*, 135, 1830–1841
- Dobson, G. M. G. (1956), Origin and distribution of polyatomic molecules in the atmosphere,
   *Proc. R. Soc. Lond. A.*, 236, 187–193
- 540 Dragani, R. (2011): On the quality of the ERA-Interim ozone reanalyses:comparisons with
  541 satellite data, Q. J. Roy. Meteor. Soc., 137,1312–1326,
  542 oi:http://dx.doi.org/10.1002/qj.82110.1002/qj.821,
- Flemming, J., Inness, A., Jones, L., Eskes, H. J., Huijnen, V., Schultz, M. G., Stein, O., Cariolle,
  D., Kinnison, D., and Brasseur, G. (2011): Forecasts and assimilation experiments of the
  Antarctic ozone hole 2008, *Atmos. Chem. Phys.*, 11, 1961–1977,
  https://doi.org/10.5194/acp-11-1961-2011
- Flemming, J., Huijnen, V., Arteta, J., Bechtold, P., Beljaars, A., Blechschmidt, A.-M.,
  Diamantakis, M., Engelen, R. J., Gaudel, A., Inness, A., Jones, L., Josse, B., Katragkou,
  E., Marecal, V., Peuch, V.-H., Richter, A., Schultz, M. G., Stein, O., and Tsikerdekis, A.

(2015), Tropospheric chemistry in the Integrated Forecasting System of ECMWF, 550 Geosci. Model Dev., 8, 975-1003, https://doi.org/10.5194/gmd-8-975-2015 551 Flemming, J., Benedetti, A., Inness, A., Engelen, R. J., Jones, L., Huijnen, V., Remy, S., 552 Parrington, M., Suttie, M., Bozzo, A., Peuch, V.-H., Akritidis, D., and Katragkou, E. 553 (2017): The CAMS interim Reanalysis of Carbon Monoxide, Ozone and Aerosol for 554 2003-2015, Atmos. Chem. Phys., 17, 1945-1983, https://doi.org/10.5194/acp-17-1945-555 2017 556 557 Garcia, R. R., and Boville, B. A. (1994): "Downward Control" of the Mean Meridional Circulation and Temperature Distribution of the Polar Winter Stratosphere, J. Atmos. 558 Sci., 51, 2238–2245, https://doi.org/10.1175/1520-559 0469(1994)051<2238:COTMMC>2.0.CO;2 560 Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horanyi, A., Munoz-Sabater, J., Nicolas, J., 561 562 Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdallaa, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, 563 P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, 564 A., Haimberger, L., Healy, S., Hogan, R.J., Holm, E., Janiskova, M., Keeley, S., 565 Laloyaux, P., Lopez, P., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, 566 S., Thepaut, J.N. (2020), The ERA5 Global Reanalysis, Quarterly Journal of the Royal 567 Meteorological Society, https://rmets.onlinelibrary.wiley.com/doi/abs/10.1002/qj.3803 568 Hubert, D., Lambert, J.-C., Verhoelst, T., Granville, J., Keppens, A., Baray, J.-L., Bourassa, A. 569 570 E., Cortesi, U., Degenstein, D. A., Froidevaux, L., Godin-Beekmann, S., Hoppel, K. W., Johnson, B. J., Kyrölä, E., Leblanc, T., Lichtenberg, G., Marchand, M., McElroy, C. T., 571 Murtagh, D., Nakane, H., Portafaix, T., Querel, R., Russell III, J. M., Salvador, J., Smit, 572 H. G. J., Stebel, K., Steinbrecht, W., Strawbridge, K. B., Stübi, R., Swart, D. P. J., Taha, 573 G., Tarasick, D. W., Thompson, A. M., Urban, J., van Gijsel, J. A. E., Van Malderen, R., 574 von der Gathen, P., Walker, K. A., Wolfram, E., and Zawodny, J. M. (2016), Ground-575 based assessment of the bias and long-term stability of 14 limb and occultation ozone 576 profile data records, Atmos. Meas. Tech., 9, 2497-2534, https://doi.org/10.5194/amt-9-577 2497-2016 578 Huijnen, V., Williams, J., van Weele, M., van Noije, T., Krol, M., Dentener, F., Segers, A., 579 Houweling, S., Peters, W., de Laat, J., Boersma, F., Bergamaschi, P., van Velthoven, P., 580 Le Sager, P., Eskes, H., Alkemade, F., Scheele, R., Nédélec, P., and Pätz, H.-W. (2010), 581 The global chemistry transport model TM5: description and evaluation of the 582 tropospheric chemistry version 3.0, Geosci. Model Dev., 3, 445-473, 583 https://doi.org/10.5194/gmd-3-445-2010 584 Huijnen, V., Miyazaki, K., Flemming, J., Inness, A., Sekiya, T., and Schultz, M. G.: An 585 intercomparison of tropospheric ozone reanalysis products from CAMS, CAMS interim, 586 TCR-1, and TCR-2, Geosci. Model Dev., 13, 1513-1544, https://doi.org/10.5194/gmd-587 13-1513-2020, 2020. 588 Hurwitz, M. M., P. A. Newman, and C. I. Garfinkel (2011): The Arctic vortex in March 2011: 589 Adynamical perspective. Atmos. Chem. Phys., 11, 11 447-11 453. 590 Inness, A., Baier, F., Benedetti, A., Bouarar, I., Chabrillat, S., Clark, H., Clerbaux, C., Coheur, 591 P., Engelen, R. J., Errera, Q., Flemming, J., George, M., Granier, C., Hadji-Lazaro, J., 592

593 594 595 596 597	<ul> <li>Huijnen, V., Hurtmans, D., Jones, L., Kaiser, J. W., Kapsomenakis, J., Lefever, K.,</li> <li>Leitão, J., Razinger, M., Richter, A., Schultz, M. G., Simmons, A. J., Suttie, M., Stein,</li> <li>O., Thépaut, JN., Thouret, V., Vrekoussis, M., Zerefos, C., and the MACC team (2013):</li> <li>The MACC reanalysis: an 8 yr data set of atmospheric composition, <i>Atmos. Chem. Phys.</i>,</li> <li>13, 4073–4109, https://doi.org/10.5194/acp-13-4073-2013</li> </ul>
598 599 600 601 602 603 604	<ul> <li>Inness, A., Blechschmidt, AM., Bouarar, I., Chabrillat, S., Crepulja, M., Engelen, R. J., Eskes, H., Flemming, J., Gaudel, A., Hendrick, F., Huijnen, V., Jones, L., Kapsomenakis, J., Katragkou, E., Keppens, A., Langerock, B., de Mazière, M., Melas, D., Parrington, M., Peuch, V. H., Razinger, M., Richter, A., Schultz, M. G., Suttie, M., Thouret, V., Vrekoussis, M., Wagner, A., and Zerefos, C. (2015): Data assimilation of satellite-retrieved ozone, carbon monoxide and nitrogen dioxide with ECMWF's Composition-IFS, <i>Atmos. Chem. Phys.</i>, 15, 5275–5303, https://doi.org/10.5194/acp-15-5275-2015</li> </ul>
605	<ul> <li>Inness, A., Ades, M., Agustí-Panareda, A., Barré, J., Benedictow, A., Blechschmidt, AM.,</li></ul>
606	Dominguez, J. J., Engelen, R., Eskes, H., Flemming, J., Huijnen, V., Jones, L., Kipling,
607	Z., Massart, S., Parrington, M., Peuch, VH., Razinger, M., Remy, S., Schulz, M., and
608	Suttie, M. (2019), The CAMS reanalysis of atmospheric composition, <i>Atmos. Chem.</i>
609	<i>Phys.</i> , 19, 3515–3556, https://doi.org/10.5194/acp-19-3515-2019
610 611 612	<ul> <li>Komhyr, W. D., Barnes, R. A., Borthers, G. B., Lathrop, J. A., Kerr, J. B., and Opperman, D. P. (1995): Electrochemical concentration cell ozonesonde performance evaluation during STOIC 1989, <i>J. Geophys. Res.</i>, 100, 9231–9244</li> </ul>
<ul> <li>613</li> <li>614</li> <li>615</li> <li>616</li> <li>617</li> <li>618</li> </ul>	<ul> <li>Laeng,A., von Clarmann, T., Stiller, G., Grabowski, U., Kiefer, M., Hubert, D., Verhoelst, T., Keppens, A., Lambert, J. C., Dinelli, B. M., Dudhia, A., Raspollini, P., Sofieva, V., Froidevaux, L., Walker, K. A., Zehner, C. (2018), The ozone climate change initiative: Comparison of four Level-2 processors for the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), Remote Sensing of Environment, ISSN: 0034-4257, Vol: 162, Page: 316-343, https://doi.org/10.1016/j.rse.2014.12.013</li> </ul>
619	Langerock, B., De Mazière, M., Hendrick, F., Vigouroux, C., Desmet, F., Dils, B., and
620	Niemeijer, S. (2015), Description of algorithms for co-locating and comparing gridded
621	model data with remote-sensing observations, <i>Geosci. Model Dev.</i> , 8, 911–921,
622	https://doi.org/10.5194/gmd-8-911-2015, 2015.
623	Lawrence, A. D., Perlwitz, J., Butler, A. H., Manney, G. L., Newman, P. A., Lee, S. H. and
624	Nash, E. R. (2020), The Remarkably Strong Arctic Stratospheric Polar Vortex of Wnter
625	2020: Links to Record-Breaking Arctic Oscillation and Ozone Loss, Submitted to J.
626	Geophys. Res., https://doi.org/10.1002/essoar.10503356.1
627	Lefever, K., van der A, R., Baier, F., Christophe, Y., Errera, Q., Eskes, H., Flemming, J., Inness,
628	A., Jones, L., Lambert, JC., Langerock, B., Schultz, M. G., Stein, O., Wagner, A., and
629	Chabrillat, S. (2015), Copernicus stratospheric ozone service, 2009–2012: validation,
630	system intercomparison and roles of input data sets, <i>Atmos. Chem. Phys.</i> , 15, 2269–2293,
631	https://doi.org/10.5194/acp-15-2269-2015
632	Livesey, N J., Read, William G., Wagner, P. A., Froidevaux, L., Lambert, A., Manney, G. L.,
633	Millán, L. F., Valle, H. C. Pumphrey, M. L., Santee, M. J., Schwartz, S. W., Fuller, R. A.,
634	Jarnot, R. F., Knosp, B. W., Martinez, E., Lay, R. R. (2018): Version 4.2x Level 2 data

- quality and description document, Tech. rep., Jet Propulsion Laboratory, available from
   http://mls.jpl.nasa.gov/.
- Manney, G., Zurek, R. W., O'Neill, A. and Swinbank, R. (1994): On the Motion of Air through
  the Stratospheric Polar Vortex, J. Atmos. Sci., 51, 2973–2994,
  https://doi.org/10.1175/1520-0469(1994)051<2973:OTMOAT>2.0.CO;2.
- Manney, G., Santee, M., Rex, M. et al. (2011), Unprecedented Arctic ozone loss in 2011. *Nature*,
   478, 469–475. https://doi.org/10.1038/nature10556
- McPeters, R. D., Bhartia, P. K., Haffner, D., Labow, G. J., and Flynn, L. (2013): The version 8.6
  SBUV ozone data record: An overview, *J. Geophys. Res.-Atmos.*, 118, 8032–8039, https://doi.org/10.1002/jgrd.50597
- Newman, P. A., E. R. Nash, and J. E. Rosenfield (2001), What controls the temperature in the
  Arctic stratosphere in the spring? *J. Geophys. Res.*, 106 (D17), 19 999–20 010.
- Newman, P. A., J. F. Gleason, and R. S. Stolarski (1997), Anomalously low ozone over the
   Arctic, *Geophys. Res. Lett.*, Vol. 24, No. 22, 2689-2692.
- Sheese, P. E., Walker, K. A., Boone, C. D., Bernath, P. F., Froidevaux, L., Funke, B., Raspollini,
  P., von Clarmann, T. (2015), ACE-FTS ozone, water vapour, nitrous oxide, nitric acid,
  and carbon monoxide profile comparisons with MIPAS and MLS, Journal of Quantitative
  Spectroscopy and Radiative Transfer, Volume 186, January 2017, Pages 63-80,
  https://doi.org/10.1016/j.jqsrt.2016.06.026
- Shepherd, T. G. (2008), Dynamics, stratospheric ozone, and climate change, *Atmosphere-Ocean*,
   46:1, 117-138, DOI: 10.3137/ao.460106
- Stauffer, R. M., Thompson, A. M., Kollonige, D. E., Witte, J. C., Tarasick, D. W., Davies, J., et
   al. (2020). A post-2013 dropoff in total ozone at a third of global ozonesonde stations:
   Electrochemical concentration cell instrument artifacts? *Geophysical Research Letters*,
   47, https://doi.org/10.1029/2019GL086791
- Steinbrecht, W., Froidevaux, L., Fuller, R., Wang, R., Anderson, J., Roth, C., Bourassa, A., 660 Degenstein, D., Damadeo, R., Zawodny, J., Frith, S., McPeters, R., Bhartia, P., Wild, J., 661 Long, C., Davis, S., Rosenlof, K., Sofieva, V., Walker, K., Rahpoe, N., Rozanov, A., 662 Weber, M., Laeng, A., von Clarmann, T., Stiller, G., Kramarova, N., Godin-Beekmann, 663 S., Leblanc, T., Querel, R., Swart, D., Boyd, I., Hocke, K., Kämpfer, N., Maillard Barras, 664 E., Moreira, L., Nedoluha, G., Vigouroux, C., Blumenstock, T., Schneider, M., García, 665 O., Jones, N., Mahieu, E., Smale, D., Kotkamp, M., Robinson, J., Petropavlovskikh, I., 666 Harris, N., Hassler, B., Hubert, D., and Tummon, F. (2017), An update on ozone profile 667 trends for the period 2000 to 2016, Atmos. Chem. Phys., 17, 10675-10690, 668 https://doi.org/10.5194/acp-17-10675-2017 669
- Steinbrecht, W., Shwartz, R., and Claude, H. (1998), New pump correction for the Brewer-Mast
   ozonesonde: Determination from experiment and instrument intercomparisons, *J. Atmos. Ocean. Tech.*, 15, 144–156

# Stolarski, R. S. and Frith, S. M. (2006), Search for evidence of trend slow-down in the long-term TOMS/SBUV total ozone data record: the importance of instrument drift uncertainty, Atmos. Chem. Phys., 6, 4057–4065, https://doi.org/10.5194/acp-6-4057-2006.

676	Tegtmeier, S., Rex, M., Wohltmann, I. & Krüger, K. (2008), Relative importance of dynamical
677	and chemical contributions to Arctic wintertime ozone. <i>Geophys. Res. Lett.</i> , Vol. 35,
678	L17801 http://dx.doi.org/10.1029/2008GL034250
679 680 681	van der A, R. J., Allaart, M. A. F., and Eskes, H. J.: Extended and refined multi sensor reanalysis of total ozone for the period 1970–2012 (2015), <i>Atmos. Meas. Tech.</i> , 8, 3021–3035, https://doi.org/10.5194/amt-8-3021-2015.
682	Wagner et al. (2020): Comprehensive Evaluation of the Copernicus Atmosphere Monitoring
683	Service (CAMS) reanalysis against independent observations. Submitted to Elementa:
684	Science of the Anthropocene, www.elementascience.org
685	Waugh, D.W, A Sobel, L.M. Polvani (2017), What is the Polar Vortex and how does it influence
686	weather? <i>Bulletin of the American Meteorological Society</i> , 98, 37-44.
687	doi.org/10.1175/BAMS-D-15-00212.1
688	WMO (World Meteorological Organization), Scientific Assessment of Ozone Depletion: 2018,
689	Global Ozone Research and Monitoring Project – Report No. 58, 588 pp., Geneva,
690	Switzerland, 2018.
691 692 693 694	Yarwood, G., Rao, S., Yocke, M., and Whitten, G. (2005), Updates to the carbon bond chemical mechanism: CB05, Final report to the US EPA, <i>EPA Report Number</i> : RT-0400675, available at: http://www.camx.com