

## Arctic Tropospheric Ozone Trends

K. S. Law<sup>1</sup>, J. Liengard Hjorth<sup>2</sup>, J. B. Pernov<sup>2,3</sup>, C. H. Whaley<sup>4</sup>, H. Skov<sup>2</sup>, M. Collaud Coen<sup>5</sup>, J. Langner<sup>6</sup>, S. R. Arnold<sup>7</sup>, D. Tarasick<sup>8</sup>, J. Christensen<sup>2</sup>, M. Deushi<sup>9</sup>, P. Effertz<sup>10,14</sup>, G. Faluvegi<sup>11,12</sup>, M. Gauss<sup>13</sup>, U. Im<sup>2</sup>, N. Oshima<sup>9</sup>, I. Petropavlovskikh<sup>10,14</sup>, D. Plummer<sup>4</sup>, K. Tsigaridis<sup>11,12</sup>, S. Tsyro<sup>13</sup>, S. Solberg<sup>15</sup> and S.T. Turnock<sup>16,7</sup>

<sup>1</sup>Sorbonne Université, LATMOS-IPSL, UVSQ, CNRS, Paris, France

<sup>2</sup>Department of Environmental Science, Interdisciplinary Centre for Climate Change, Aarhus University, Frederiksborgvej 4000, Roskilde, Denmark

<sup>3</sup>Extreme Environments Research Laboratory, École Polytechnique Fédérale de Lausanne, 1951 Sion, Switzerland

<sup>4</sup>Canadian Centre for Climate modeling and analysis, Environment and Climate Change Canada, Victoria, BC, Canada

<sup>5</sup>Federal Office of Meteorology and Climatology, MeteoSwiss, Payerne, Switzerland

<sup>6</sup>Swedish Meteorological and Hydrological Institute, Norrköping, Sweden

<sup>7</sup>Institute for Climate and Atmospheric Science, School of Earth and Environment, University of Leeds, Leeds, United Kingdom

<sup>8</sup>Air Quality Research Division, Environment and Climate Change Canada, Toronto, ON, Canada

<sup>9</sup>Meteorological Research Institute, Japan Meteorological Agency, Tsukuba, Japan

<sup>10</sup>Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, CO, USA.

<sup>11</sup>NASA Goddard Institute for Space Studies, New York, NY, USA

<sup>12</sup>Center for Climate Systems Research, Columbia University; New York, USA

<sup>13</sup>Norwegian Meteorological Institute, Oslo, Norway

<sup>14</sup>National Oceanic and Atmospheric Administration (NOAA) ESRL Global Monitoring Laboratory, Boulder, CO, USA.

<sup>15</sup>Norwegian Institute for Air Research (NILU), Kjeller, Norway.

<sup>16</sup>Met Office Hadley Centre, Exeter, UK

Corresponding author: [Kathy.Law@latmos.ipsl.fr](mailto:Kathy.Law@latmos.ipsl.fr)

### Key Points:

- Coherent ozone trend analysis methodology applied to multi-decade, pan-Arctic surface and ozonesonde datasets and multi-model medians.
- Increasing winter Arctic tropospheric ozone overestimated by models in the free troposphere, Alaskan spring surface increases not captured.
- Spring (summer) decreases (increases) in observed ozone throughout the troposphere, not always simulated by models.

## 37 **Abstract**

38 Trends in tropospheric ozone, an important air pollutant and short-lived climate forcer (SLCF),  
39 are estimated using available surface and ozonesonde profile data for 1993-2019. Using a  
40 coherent methodology, observed trends are compared to modeled trends (1995-2015) from the  
41 Arctic Monitoring Assessment Programme SLCF 2021 assessment. Statistically significant  
42 increases in observed surface ozone at Arctic coastal sites, notably during winter, and concurrent  
43 decreasing trends in surface carbon monoxide, are generally captured by multi-model median  
44 (MMM) trends. Wintertime increases are also estimated in the free troposphere at most Arctic  
45 sites, but tend to be overestimated by the MMMs. Springtime surface ozone increases in northern  
46 coastal Alaska are not simulated while negative springtime trends in northern Scandinavia are  
47 not always reproduced. Possible reasons for observed changes and model behavior are discussed,  
48 including decreasing precursor emissions, changing ozone sinks, and variability in large-scale  
49 meteorology.

## 50 **Plain Language Summary**

51 The Arctic is warming much faster than the rest of the globe due to increases in carbon dioxide,  
52 and other trace constituents like ozone, also an air pollutant. However, improved understanding  
53 is needed about long-term changes or trends in Arctic tropospheric ozone. A coherent  
54 methodology is applied to determine trends in surface and regular profile measurements over the  
55 last 20-30 years, and results from six chemistry-climate models. Statistically significant increases  
56 in observed ozone are found at the surface and in the free troposphere during winter in the high  
57 Arctic. Paradoxically, decreases in nitrogen oxide emissions at mid-latitudes appear to be leading  
58 to increases in ozone during winter, but associated increases in Arctic tropospheric ozone tend to  
59 be overestimated in the models. Increases are also found at the surface in northern Alaska during  
60 spring but not reproduced by the models. The causes are unknown but could be related to  
61 changes in local sources or sinks of Arctic ozone or in large-scale weather patterns. Declining  
62 mid-latitude emissions may also explain negative surface ozone trends over northern  
63 Scandinavia in spring that are not always captured by the models. Further work is needed to  
64 understand changes in Arctic tropospheric ozone.

## 65 **1 Introduction**

66 Tropospheric ozone (O<sub>3</sub>) is a short-lived climate forcer (SLCF) contributing to global and Arctic  
67 warming (AMAP, 2015; Sand et al, 2016; von Salzen et al. 2022), and a critical secondary air  
68 pollutant, detrimental to human health (Anenberg et al., 2010) and ecosystems (Arnold et al.,  
69 2018). The Arctic tropospheric O<sub>3</sub> budget is complex, as recently discussed in a companion  
70 paper, Whaley et al. (2023). It originates from photochemical production of anthropogenic or

71 natural emissions of O<sub>3</sub> precursors, including nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO) and  
72 methane (CH<sub>4</sub>), in the Arctic, or following air mass transport from mid-latitudes, as well as  
73 transport of O<sub>3</sub> from the stratosphere (Law et al., 2014; Schmale et al., 2018). Sinks include  
74 photochemical destruction, including reactions involving halogens leading to so-called ozone  
75 depletion events (ODEs) (Barrie, et al., 1988; Simpson et al., 2007), and surface dry deposition  
76 (Clifton et al., 2020). Growth in anthropogenic emissions since pre-industrial times has led to  
77 increases in tropospheric O<sub>3</sub> throughout the Northern Hemisphere (NH) (Tarasick et al., 2019;  
78 Turnock et al., 2020; Cooper et al., 2020) contributing to observed global and Arctic warming  
79 over the past century (e.g. Griffiths et al., 2021). Since the mid-1990s, a mix of relatively weak  
80 positive and negative trends (+1 to -1 parts per billion by volume (ppbv) per decade) have been  
81 reported in the NH at the surface and in the free troposphere (FT), with largest increases over  
82 south and eastern Asia, associated with increasing anthropogenic emissions (Cooper et al., 2020;  
83 Wang et al., 2022a).

84 To date, only a few studies have focused on assessing tropospheric O<sub>3</sub> trends in the Arctic. While  
85 positive O<sub>3</sub> trends were diagnosed at several surface sites, results are not always statistically  
86 significant, and both positive and negative trends were reported at some Canadian sites (Tarasick  
87 et al., 2016; Sharma et al., 2019; Cooper et al., 2020). In the Arctic FT, studies found significant  
88 positive trends (Christiansen et al., 2017; Wang et al., 2022a), no trends (Tarasick et al., 2016),  
89 or mixed trends in different seasons (Bahramvash Shams et al., 2019). Differences in the periods  
90 analyzed, sign or magnitude of trends emphasizes the need to further examine trends using the  
91 same methodology. Coherent estimation of observed trends, and evaluation of modeled trends, is  
92 needed to better understand O<sub>3</sub> changes and impacts on Arctic climate that are sensitive to the  
93 altitude where O<sub>3</sub> perturbations occur (Rap et al., 2015). This study assesses annual and monthly  
94 trends, together with possible evolution in seasonal cycles, of Arctic tropospheric O<sub>3</sub> over the last  
95 20-30 years. Observed changes are compared to results from atmospheric chemistry-climate  
96 models run as part of the recent Arctic Monitoring and Assessment Programme (AMAP) SLCF  
97 assessment (AMAP, 2021; Whaley et al., 2022; von Salzen et al., 2022). Results are discussed in  
98 light of possible changes in sources and sinks of Arctic tropospheric O<sub>3</sub>.

99 **2 Methods**100 **2.1 Measurements**

101 The location of surface and ozonesonde sites used in this study are displayed in Fig. 1, together  
 102 with the Arctic Circle at 66.6°N, used to define the Arctic. Annual surface trends are shown in  
 103 the table grouped into 1) high Arctic coastal sites (Alert, Utqiagvik/Barrow, Villum), Zeppelin  
 104 (situated at 474m on Svalbard) and Summit (high altitude (FT) site on Greenland (3211m), and  
 105 2) European continental sites within (Pallas, Esrange), and just south (Tustervatn) of the Arctic  
 106 Circle.



Site	Annual trend (%)	Significance level	Period
<b>High Arctic:</b>			
Alert	<b>0.29</b>	95%	1999-2019
	<b>0.24</b>	95%	1993-2019
Utqiagvik	0.53	<90%	1999-2019
	0.26	<90%	1993-2019
Villum	<b>1.98</b>	95%	1999-2019
	0.68	90%	1996-2019
Zeppelin	-0.19	<90%	1999-2019
	<b>0.18</b>	90%	1993-2019
Summit	-0.28	<90%	2001-2019
<b>European continental Arctic and near-Arctic:</b>			
Esrange	0.08	<90%	1999-2019
	0.00	<90%	1993-2019
Pallas	<b>-0.30</b>	90%	1998-2019
	<b>-0.40</b>	90%	1995-2019
Tustervatn	<b>-0.52</b>	99%	1999-2019
	-0.18	>90%	1994-2019

107 **Figure 1.** Left: Location of surface (bold) and ozonesonde (italic) sites and showing the Arctic  
 108 Circle (66.55°N). Right: annual O<sub>3</sub> trends at surface sites in % per year (left column), the  
 109 significance level (middle column), calculated over periods shown in the right column.  
 110 Statistically significant trends (above 90% confidence level) are in bold. Geographical  
 111 coordinates for all sites are provided in Whaley et al. (2023). See text for details.  
 112

113 Surface observations are from EBAS Level 2 data, station owners for Villum before 2001,  
 114 Canada's Open Government Portal for Alert, and National Oceanic and Atmospheric

115 Administration (NOAA) for Summit, and Barrow Atmospheric Observatory, Utqiagvik  
116 (Utqiagvik from now on). Ozonesonde data are from the World Ozone and Ultraviolet Radiation  
117 Data Centre (WOUDC) and Network for the Detection of Atmospheric Composition Change  
118 (NDACC). See also the Supplementary Information (Text S1, Figs. S1 and S2, including data  
119 coverage).

## 120 **2.2 Trend analysis**

121 Observed monthly and annual trends in surface O<sub>3</sub> concentrations at different sites are  
122 determined using the non-parametric Mann-Kendall test at the 90<sup>th</sup> and 95<sup>th</sup> confidence level  
123 (CL) and Sen's slope methodology (Theil, 1950; Sen, 1968) (see Text S2). Daily median data are  
124 sorted into different months and pre-whitened, due to the presence of autocorrelation, via the  
125 3PW algorithm from Collaud Coen et al. (2020). Trends using ozonesonde profiles are calculated  
126 based on weekly medians for selected pressure levels. For the calculation of relative trends, data  
127 are normalized by division with median values and multiplied by 100.

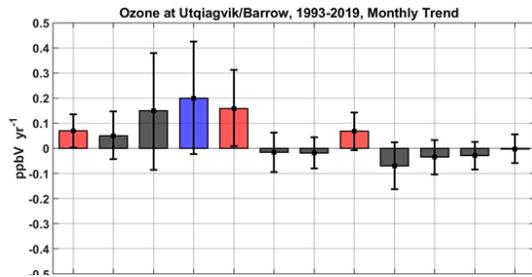
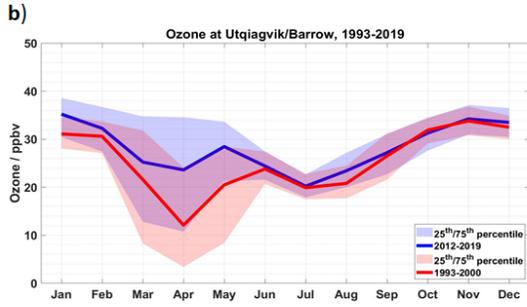
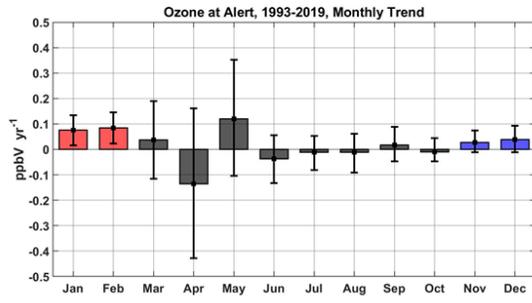
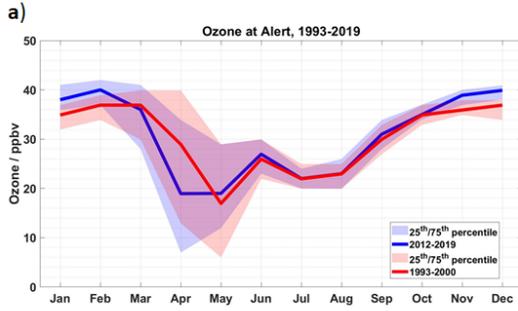
## 128 **2.3 Modeled trends**

129 Modeled trends at the surface and different altitudes are calculated for 1995-2015 using results  
130 from four global chemistry-climate models (CMAM, GISS-E2.1, MRI-ESM2, UKESM1) and  
131 two chemistry-transport models (DEHM, EMEP MSC-W) run using the same ECLIPSEv6b  
132 anthropogenic emissions, and nudged with meteorological reanalyses as part of AMAP (2021).  
133 Details can be found in Whaley et al. (2022), Text S3 and Table S1. Simulated monthly mean O<sub>3</sub>  
134 volume mixing ratios from the model grid box containing the measurement location are used to  
135 compute multi-model medians (MMMs). For ozonesonde comparisons, modeled vertical profiles  
136 are interpolated onto the same vertical bins as the measurements before trends are computed.

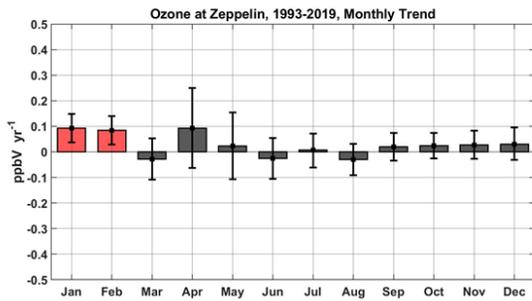
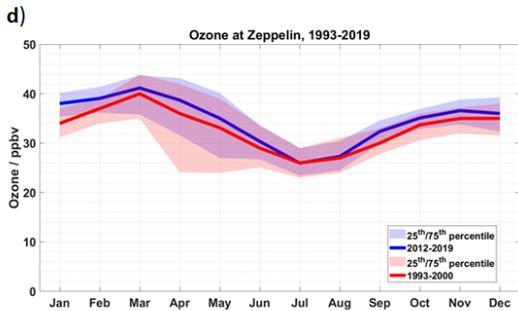
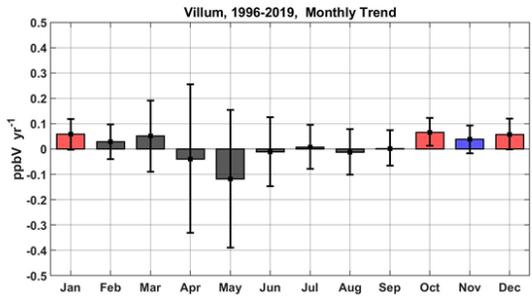
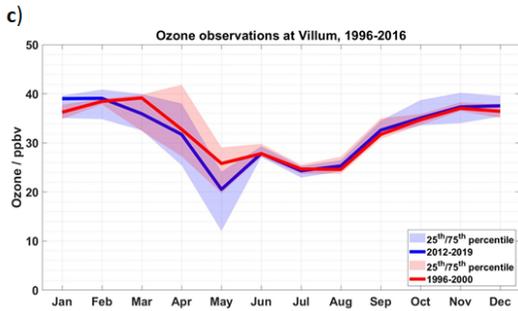
# 137 **3 Surface ozone trends in the Arctic**

## 138 **3.1 Observed ozone trends**

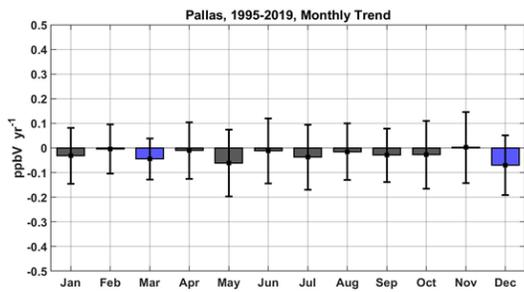
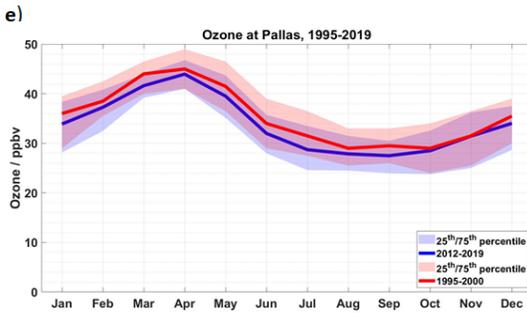
139 Annual trends are calculated for 1993-2019, or for the longest period with sufficient data, for all  
140 the sites (see Fig. 1, Table S2).



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144 **Figure 2.** Observed surface  $O_3$  trends and seasonal cycles. Left: seasonal cycles of monthly  
145 median  $O_3$  (ppbv) at a) Alert, b) Utqiagvik, c) Villum, d) Zeppelin, and e) Pallas for 1993-2000  
146 (blue lines) vs 2012-2019 (red lines). Shaded areas show upper and lower quartiles of hourly  
147 values. Right: monthly trends for 1993-2019. Boxes represent the slope of the trend in ppbv per  
148 year with red boxes significant at 95<sup>th</sup>% CL, blue boxes at 90<sup>th</sup>% CL, and black boxes not  
149 statistically significant. Error bars show 95<sup>th</sup>% CLs. Results are shown for shorter periods  
150 depending on data availability.

151 Average  $O_3$  seasonal cycles are also calculated for earlier (1993-2000) and later (2012-2019)  
152 periods, to examine possible changes, together with monthly trends (Fig. 2) at selected sites (see  
153 Fig. S3 for other sites). Monthly trends are also analyzed for different 21-year periods (1993-  
154 2012, 1999-2019) (Fig. S4).

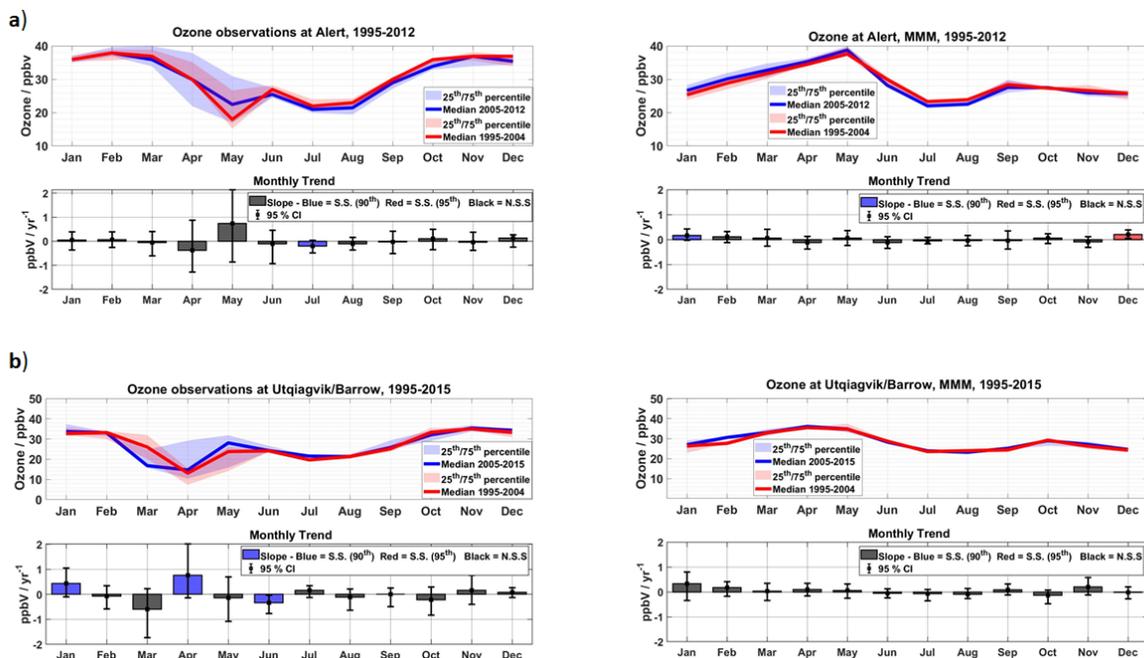
155 First considering high Arctic sites at coastal locations that exhibit a winter maximum with low  
156 spring concentrations attributed to ODEs, as discussed in Whaley et al. (2023). Alert has  
157 statistically significant (“ss”) positive  $O_3$  annual trends, as does Villum for the shorter time  
158 period 1999-2019, while annual trends at Utqiagvik are not significant (see Fig. 1). Ss trends are  
159 also calculated in particular seasons, as shown in Fig. 2. Notably, ss positive trends are found  
160 during late autumn and/or winter at Alert, Villum and Utqiagvik. Positive trends are also  
161 calculated for spring at Utqiagvik (April-May). Winter trends at Alert and spring trends at  
162 Utqiagvik are more pronounced when using the later record (1999-2019) (see Fig. S4). To  
163 further characterize these changes, probability distributions in observed  $O_3$  concentrations are  
164 calculated for months with ss trends (see Fig. S5). Positive ss trends during winter and spring at  
165 Utqiagvik are the result of a decrease (increase) in the frequency of low (high) concentrations  
166 (Jan.-May), whereas wintertime  $O_3$  concentrations shifted recently towards higher values at Alert  
167 (Nov.-Feb.) and Villum (Oct.-Jan.). Zeppelin shows a different seasonal behavior compared to  
168 Arctic sea-level coastal sites with a spring maximum, more similar to remote mid-latitude sites.  
169 Here, ss positive annual trends are estimated for 1993-2019 (Fig. 1), and in winter (Fig. 2),  
170 driven by increases in the earlier part of the record (1993-2013) (Fig. S4).

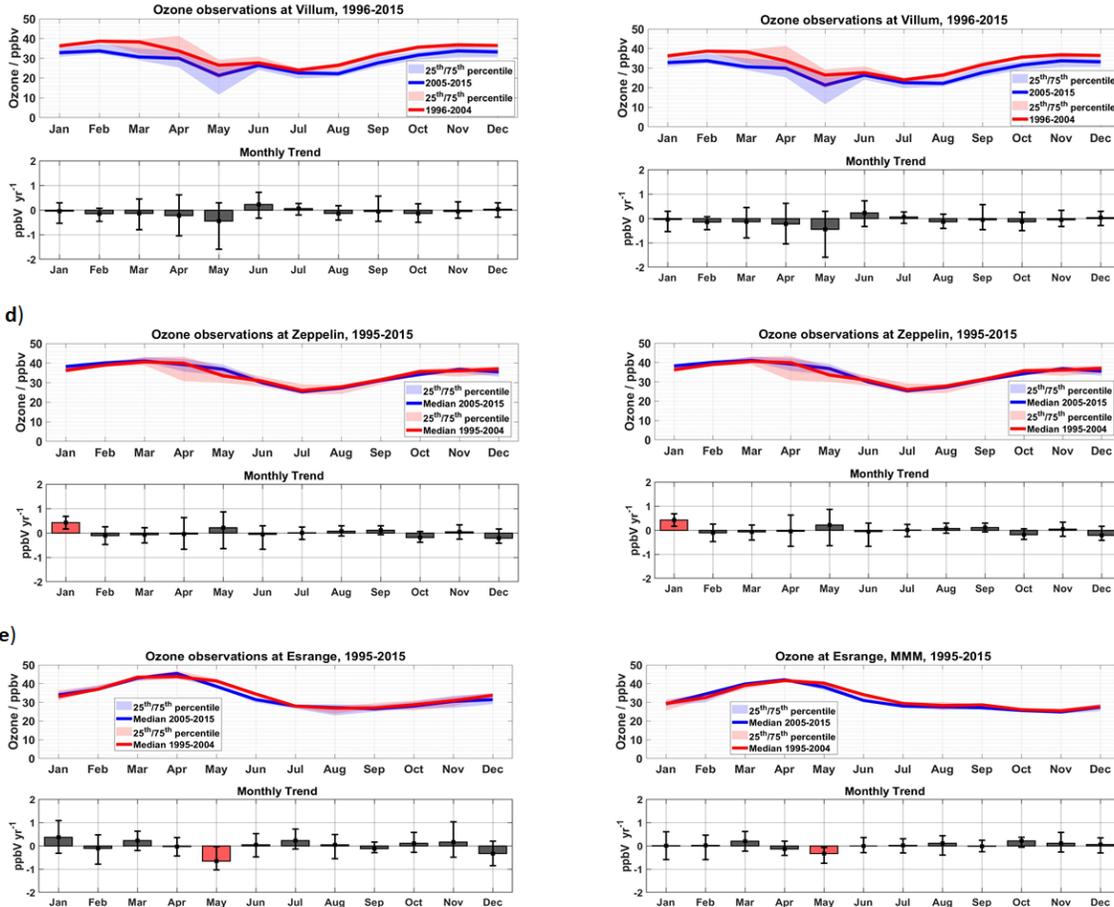
171 Continental northern Scandinavian sites exhibit a different behavior with Pallas and Tustervatn  
172 showing ss negative annual trends but no ss annual (or monthly) trends at Esrange over any of  
173 the periods considered. The shape of the seasonal cycle for the earlier versus the later period is

174 similar at these sites, which also have a spring maximum like Zeppelin. O<sub>3</sub> appears to be  
 175 decreasing throughout the year when comparing earlier and later periods although ss negative  
 176 trends are only evident at Pallas (March, December), and at Tustervatn in spring and early  
 177 summer (Fig. S4, 1999-2019 trends). Summit is more representative of the FT and samples air  
 178 masses transported from North America and Asia, or of stratospheric origin (Dibb, 2007;  
 179 Schmeisser et al., 2018). The annual trend, calculated over the shorter 2001-2019 record, is not  
 180 ss at the 90<sup>th</sup> % CL, but ss negative monthly trends are estimated for January, March-May and  
 181 September.

### 182 3.2 Comparison of observed and modeled surface trends

183 Figure 3 compares observed monthly and MMM trends for 1995-2015, or the closest possible  
 184 time interval in case of years with missing observations. Results for other sites are shown in Fig.  
 185 S6. Observed ss trends are more frequently diagnosed over 1993-2019 (Fig. 2) than over the  
 186 shorter period ending in 2015 (Fig. 3). While the MMMs simulate O<sub>3</sub> seasonal cycles reasonably  
 187 well, low O<sub>3</sub> concentrations are missed in spring, and wintertime O<sub>3</sub> is underestimated (Whaley  
 188 et al., 2023). The MMMs simulate ss positive and negative trends at Zeppelin (Jan.) and Esrange  
 189 (May), respectively, but not ss positive trends at Utqiagvik (April). Ss trends are simulated, but  
 190 not observed, at Alert (January, December) and Tustervatn (March).





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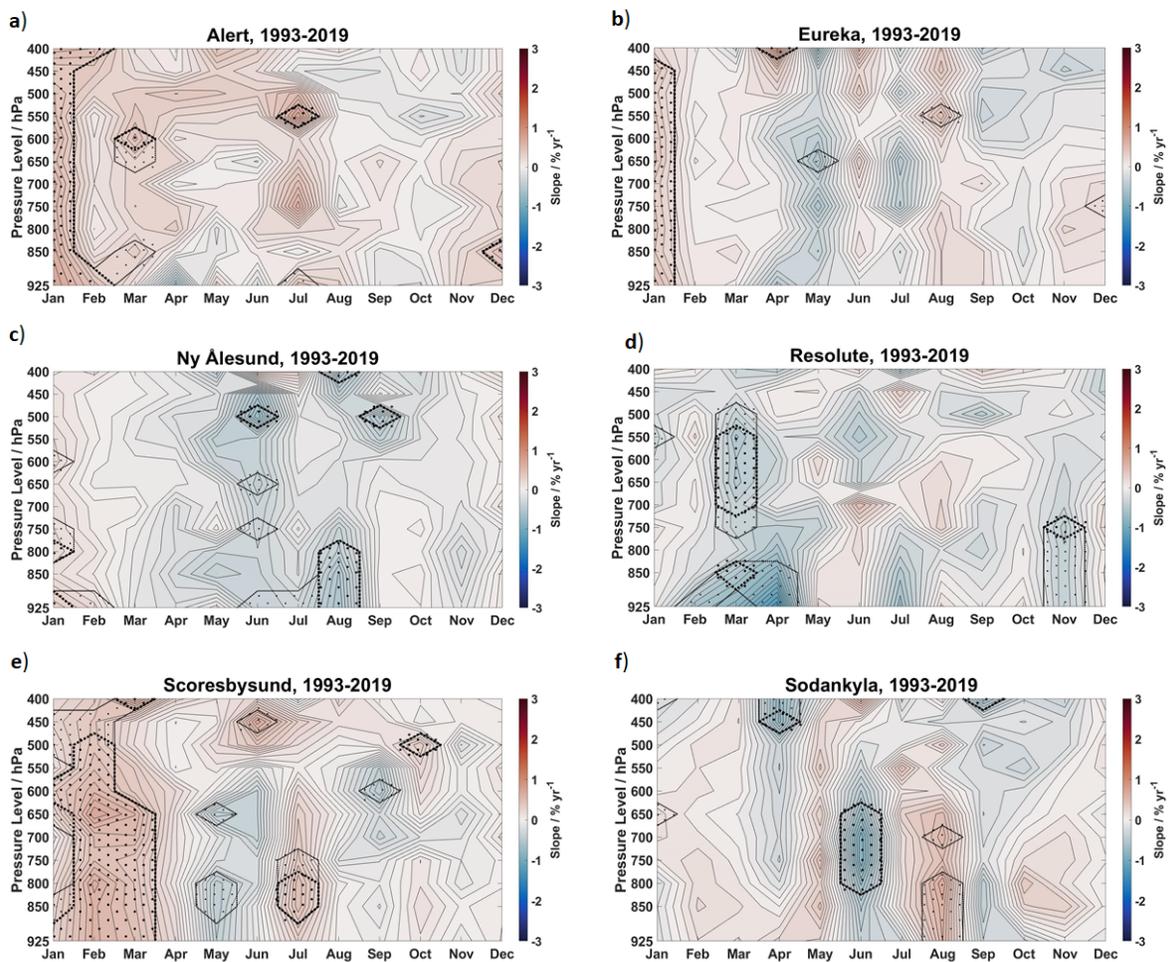
194 **Figure 3:** Comparison of observed (left) and MMM (right) surface  $O_3$  trends and seasonal cycles  
 195 at a) Alert, b) Utqiagvik, c) Villum, d) Zeppelin, and e) Esrange. Upper panels: seasonal cycles  
 196 for 1995-2004 (red lines) vs 2005-2015 (blue lines). Shaded areas show upper and lower  
 197 quartiles of monthly values (observations only). Lower panels: monthly median trends in ppbv  
 198 per year for 1995-2015, or shorter periods depending on data availability. Box coloring and  
 199 error bars are the same as Fig. 2.

## 200 4 Arctic ozone trends in the free troposphere

### 201 4.1 Observed vertical trends

202 This analysis focuses on  $O_3$  changes in the lower and mid-troposphere. Figure 4 shows observed  
 203 relative trends at six Arctic ozonesonde sites from 925-400 hPa for 1993-2019. Absolute trends  
 204 above and below 400 hPa, and relative trends from 925-100 hPa, are also calculated (Figs. S7a,

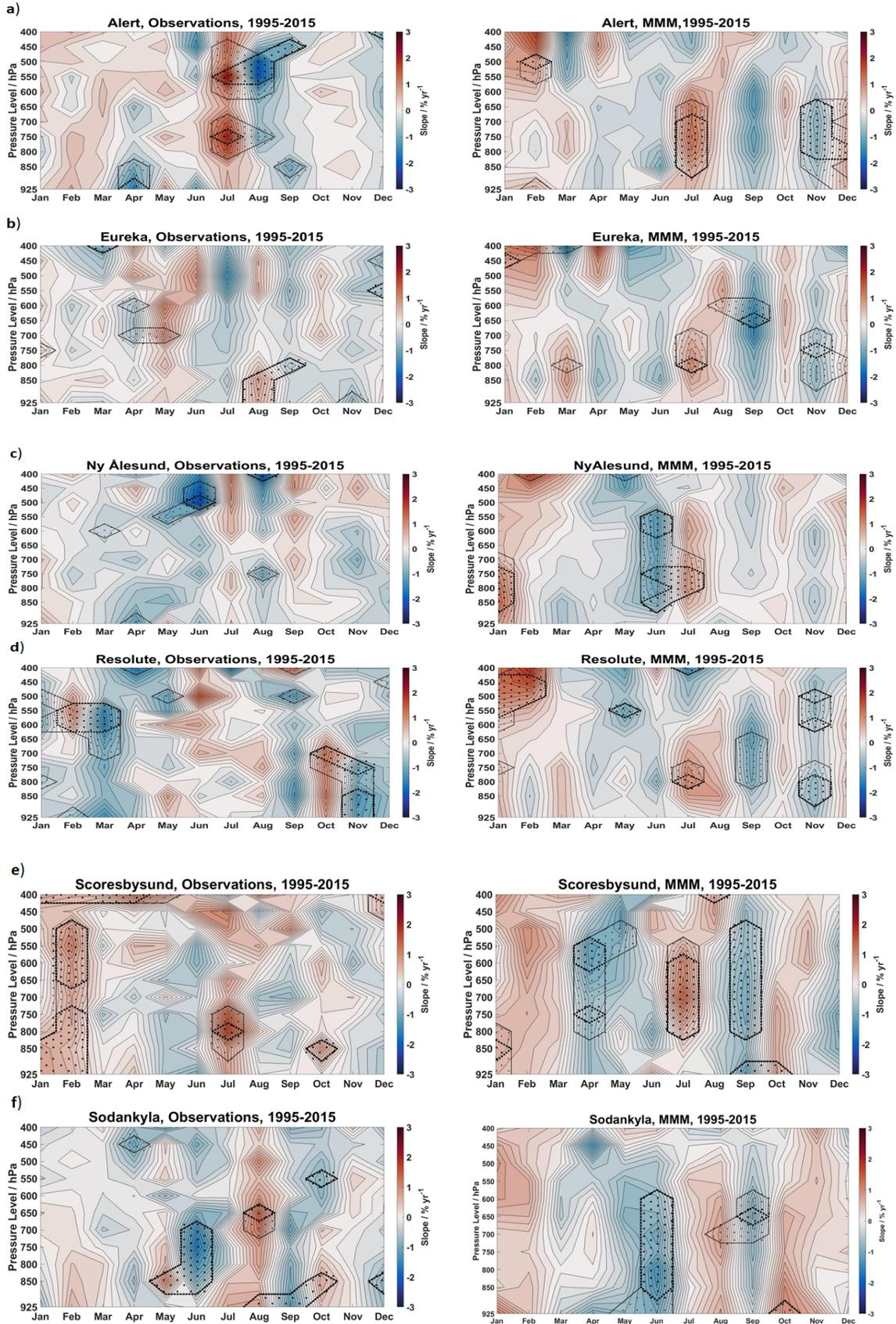
205 S7b). Overall, while there are few ss trends, there seems to be a “dipole effect” with positive  
 206 trends in winter and summer, and negative trends in spring and autumn. Positive ss winter  
 207 (Jan/Dec) trends are found up to 400 hPa at most sites (except Resolute), and also at  
 208 Scoresbysund in early spring. Positive wintertime trends are more evident in the earlier period in  
 209 the upper troposphere (UT) and lower stratosphere (LS) (Fig. S8). Eureka, Resolute, and  
 210 Sodankyla have periods with negative trends especially during spring and early summer in the  
 211 lower troposphere. Resolute decreases extend up to 500 hPa in March-April. Relative ss trends  
 212 vary from -1.5% to +0.5-1.0 % per year (Figs. 4, S7b) while stronger negative trends are  
 213 diagnosed in later years (1999-2019) compared to 1993-2013 at all sites (Fig. S8).



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216 **Figure 4:** Vertical trends in observed monthly  $O_3$  for 1993-2019, relative to monthly median  
 217 concentrations, in % per year, from 925-400 hPa at a) Alert, b) Eureka, c) Ny Alesund, d)  
 218 Resolute, e) Scoresbysund, and f) Sodankyla. Stippled lines/areas show statistical significance at  
 219 the 90<sup>th</sup> % CL (smaller marker size) and 95<sup>th</sup> % CL (larger marker size).



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223 **Figure 5:** Comparison of observed (left) and MMM (right) vertical trends in monthly  $O_3$ , relative  
224 to monthly medians, in % per year, from 925-400 hPa over 1995-2015 at a) Alert, b) Eureka, c)  
225 Ny Alesund, d) Resolute, e) Scoresbysund, and f) Sodankyla. Shading/symbols are as in Fig. 4.

## 226 **4.2 Comparison of observed and modeled vertical trends**

227 Figure 5 shows observed ozonesonde and MMM trends for 1995-2015 up to 400 hPa (see Fig.  
228 S9 for results up to 100 hPa). Only results from 5 models are used, since EMEP MSC-W only  
229 provided surface  $O_3$ . The MMMs appear to capture the observed “dipole effect” seen in the  
230 observed trends. Models also capture observed increases in the winter but trends are  
231 overestimated at most sites, especially Ny Alesund and Sodankyla. Negative winter trends at  
232 Resolute are not simulated. This may be linked to positive modeled trends above 500 hPa at all  
233 sites (Fig. S9). Summertime positive ss MMM trends are larger than observed trends at some  
234 sites, e.g. Resolute and Ny Alesund.

## 235 **5 Discussion and conclusions**

236 Increasing annual surface  $O_3$  trends at Arctic coastal sites, and at Zeppelin, are in qualitative  
237 agreement with Cooper et al. (2020), but in contrast to negative or non-significant surface trends  
238 at Canadian ozonesonde sites (Tarasick et al., 2016). A notable finding is that ss positive trends  
239 occur mainly in the winter months. While such increases were reported previously at Utqiagvik  
240 (Gaudel et al. 2018; Christiansen et al., 2022) and Alert (Sharma et al., 2019), we confirm this  
241 tendency over the wider Arctic. Emission reductions of  $NO_x$  in Europe and North America, and  
242 more recently over eastern Asia, have led to increasing wintertime  $O_3$  at mid-latitudes due to less  
243 NO titration of  $O_3$  (Jhun et al., 2015, Wang et al., 2022b, Bowman et al., 2022). This can explain  
244 observed increases in wintertime surface Arctic  $O_3$ , influenced primarily by transport of air  
245 masses from Europe (Hirdman et al., 2010). Evidence for declining  $O_3$  precursor trends is  
246 supported by decreases in observed CO in the Arctic during autumn and winter (Fig. S10). At the  
247 same time,  $CH_4$  continues to increase globally contributing to rising  $O_3$  in the NH (Zeng et al.,  
248 2022) (see also Text S4 on Arctic  $O_3$  precursor trends).

249 Another intriguing finding is springtime ss surface  $O_3$  increases at Utqiagvik (especially over  
250 1999-2019, Fig S4), but no ss changes at Alert and Villum. Changes in  $O_3$  concentrations at this

251 time of year may be driven by changes in ODE frequency linked to climate change or weather  
252 patterns (Oltmans et al, 2012). ODEs lead to zero or very low springtime O<sub>3</sub> due to bromine  
253 released from frost flowers or blowing snow (on sea-ice) (Simpson et al., 2007) or iodine  
254 compounds with a possible oceanic source (Benevant et al., 2022). Increases in springtime  
255 tropospheric bromine oxide have been observed from satellites, especially along the north coast  
256 of Greenland and central Arctic Ocean, correlating weakly with an increasing frequency in first  
257 year sea-ice (Bougoudis et al., 2020). Indeed, the frequency of low springtime O<sub>3</sub> concentrations  
258 has been increasing at Canadian high Arctic sites (see Fig. S11) but no ss springtime monthly  
259 trends are determined at Alert or Villum in our analysis. Springtime increases at Utqiagvik could  
260 be due to stronger transport from mid-latitudes to this site during periods with a more northerly  
261 extension of the Pacific storm track, hampering conditions for ODEs (Koo et al., 2012). They  
262 could also be due to an increasing influence from local emissions, such as shipping or Alaskan  
263 petroleum extraction, when photochemistry becomes active in spring (Gunsch et al., 2017).

264 Decreases in springtime/early summer O<sub>3</sub> in northern Scandinavia, especially over the later  
265 1999-2019 period, are consistent with negative trends reported at Tustervatn (Cooper et al.,  
266 2020), and sites in northern Sweden during summer (Andersson et al., 2017). These decreases  
267 are associated with lower maximum O<sub>3</sub> concentrations linked to reductions in European  
268 precursor emissions leading to less photochemical O<sub>3</sub> production (Cooper et al., 2020) although  
269 no ss trends in observed Arctic CO are found at this time of year (Fig. S10). Springtime ss  
270 negative trends at Summit may also be due to emission reductions over North America. Our  
271 results do not suggest a shift in the O<sub>3</sub> seasonal cycle toward higher concentrations in the spring  
272 (i.e. moving back toward pre-industrial O<sub>3</sub> seasonality) as reported at NH mid-latitudes  
273 (Bowman et al., 2022). Another explanation for decreasing springtime O<sub>3</sub> at the surface could be  
274 that reductions in snow cover (Mudryk et al., 2020) are leading to more O<sub>3</sub> dry deposition to  
275 Scandinavian forests.

276 The observed and modeled surface trend comparison covers 1995-2015, thereby missing the later  
277 time period when stronger observed O<sub>3</sub> trends are found, especially ss positive trends in winter.  
278 MMMs capture wintertime O<sub>3</sub> increases at Zeppelin, but overestimate at Alert and miss increase  
279 at Utqiagvik. However, Whaley et al. (2023) noted that these models underestimate wintertime  
280 Arctic O<sub>3</sub> due to deficiencies modeling shallow boundary layers, O<sub>3</sub> deposition or NO<sub>x</sub> lifetimes.

281 Nevertheless, decreasing winter trends in surface CO are captured at Alert and Utqiagvik (Fig.  
282 S10). Ss positive spring O<sub>3</sub> trend at Utqiagvik is not evident in MMM trends over 1995-2015.  
283 However, the models do not capture springtime O<sub>3</sub> seasonality due to incorrect simulation of  
284 transport patterns (Oltmans et al., 2012) or missing surface halogen chemistry (Whaley et al.,  
285 2023). Negative ss springtime (May) trends are not always reproduced, possibly reflecting issues  
286 in the emission trends or modeled dry deposition.

287  
288 FT O<sub>3</sub> trends are ss positive in winter at all Arctic sites, except Resolute, in common with several  
289 coastal Arctic surface sites. These results are in-line with increases reported at NH mid-latitudes  
290 (Cooper et al., 2020), and at Canadian ozonesonde sites (up to 400 hPa), except Resolute (Wang  
291 et al., 2022a). MMM trends are similar to observed trends over 1995-2015, including where they  
292 are ss. Patterns in observed trends are quite well captured, notably positive ss trends in winter  
293 and summer, although they tend to be overestimated. Observed negative trends in spring,  
294 extending from near the surface into the FT, are generally reproduced, and are likely to be due to  
295 decreasing NO<sub>x</sub> emissions leading to lower FT O<sub>3</sub> where photochemical production is NO<sub>x</sub>-  
296 limited. Overestimation of winter trends contrasts to previous studies where models  
297 underestimated NH trends (Wang et al., 2022a; Christiansen et al., 2022). This may be due to  
298 differences in model transport or O<sub>3</sub> precursor emission trends, including NO<sub>x</sub> reductions (see  
299 also Text S4). AMAP models overestimate mid-latitude FT O<sub>3</sub> (Whaley et al., 2023), possibly  
300 suggesting a larger sensitivity to precursor emission changes.

301  
302 Observed trends in the UT (LS) appear to have switched from positive to negative since 1993 in  
303 winter/spring, which may explain stronger positive FT trends in the earlier part of the record  
304 (1993-2013). More frequent positive phases of the Arctic Oscillation in recent years may be  
305 contributing with a weaker Brewer-Dobson circulation leading to less transport of stratospheric  
306 O<sub>3</sub> into the Arctic UT-LS, a higher tropopause height, and thus lower O<sub>3</sub> concentrations in this  
307 region (Zhang et al., 2017). However, Liu et al. (2020) did not detect any trend in the  
308 stratospheric O<sub>3</sub> flux into the Arctic UT. On the other hand, Wang et al. (2022a) attributed FT  
309 increases in NH mid-high latitude O<sub>3</sub> to increases in aircraft NO<sub>x</sub> emissions.

310 Overall, this study finds significant robust trends in Arctic tropospheric O<sub>3</sub>. Observed trends are  
311 generally quite well captured by multi-model median results, although for example, they

312 overestimate wintertime free tropospheric increases, and miss Alaskan surface increases in  
313 spring. Further investigation into the causes of observed trends, and model performance, are  
314 needed taking into account uncertainties in the observations and models (Young et al., 2018;  
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## 323 **Data Availability Statement:**

324 Surface O<sub>3</sub> monitoring datasets are provided by EMEP (European Monitoring and Evaluation  
325 Program), and Global Atmosphere Watch (GAW) World Data Centre for Reactive Gases. EMEP  
326 and GAW O<sub>3</sub> data are available via the EBAS data portal (from end of 1989 to present). CO data  
327 at Utqiagvik/Barrow and Zeppelin are also available via the EBAS data portal:  
328 <http://ebas.nilu.no>. Select the station name, and the component (CO, O<sub>3</sub>) to access the data files.  
329 Canadian surface O<sub>3</sub> data can be downloaded from: [https://data-](https://donnees.ec.gc.ca/data/air/monitor/networks-and-studies/alert-nunavut-ground-level-ozone-study/)  
330 [donnees.ec.gc.ca/data/air/monitor/networks-and-studies/alert-nunavut-ground-level-ozone-](https://donnees.ec.gc.ca/data/air/monitor/networks-and-studies/alert-nunavut-ground-level-ozone-study/)  
331 [study/](https://donnees.ec.gc.ca/data/air/monitor/networks-and-studies/alert-nunavut-ground-level-ozone-study/). Canadian surface CO is available at: [https://data-](https://donnees.ec.gc.ca/data/air/monitor/national-air-pollution-surveillance-naps-program/?lang=en)  
332 [donnees.ec.gc.ca/data/air/monitor/national-air-pollution-surveillance-naps-program/?lang=en](https://donnees.ec.gc.ca/data/air/monitor/national-air-pollution-surveillance-naps-program/?lang=en).  
333 Click on folders Data, Year, ContinuousData, then HourlyData. Surface O<sub>3</sub> records for  
334 Utqiagvik/Barrow (BRW) and Summit (SUM) are provided by PE and IE via NOAA GML. Data  
335 is available at <https://gml.noaa.gov/aftp/data/ozwv/SurfaceOzone/>. Click on the directories for  
336 BRM or SUM to obtain the data. Surface O<sub>3</sub> measurements at Summit are made possible via the  
337 U.S. National Science Foundation Office of Polar Programs and their contract with Battelle  
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339 Sodankylä ozonesonde data are obtained as part of the Network for the Detection of Atmospheric

340 Composition Change (NDACC). Data is available via  
341 <https://ndacc.larc.nasa.gov/index.php/stations>. Click on the relevant site location to access the  
342 data files. Ozonesonde data for Alert, Resolute and Eureka have been reprocessed according to  
343 Tarasick et al. (2016), available at <https://hegiftom.meteo.be/datasets/ozonesondes>.

344 All model output files in NetCDF format from the simulations used in this study can be found  
345 here: <https://open.canada.ca/data/en/dataset/c9a333ea-b81c-4df3-9880-ea7c3daeb76f>. Model  
346 codes for GISS-E2.1 are available at: <https://www.giss.nasa.gov/tools/modelE/>.

347 Open-source codes for the Mann-Kendall test associated with Sen’s slope are distributed under  
348 the BSD 3-Clause License in dedicated GitHub repositories hosted within the “mannkendall”  
349 organization (<https://github.com/mannkendall>), a Matlab (Collaud Coen and Vogt, 2020,  
350 <https://doi.org/10.5281/zenodo.4134618>, <https://github.com/mannkendall/Matlab>), Python (Vogt,  
351 2020, <https://doi.org/10.5281/zenodo.4134435>, <https://github.com/mannkendall/Python>), and R  
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353 <https://github.com/mannkendall/R>). Last access for all codes 27 January 2023.

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362 [assessment-2021-impacts-of-short-lived-climate-forcers-on-arctic-climate-air-quality-and-human-](https://www.amap.no/documents/doc/amap-assessment-2021-impacts-of-short-lived-climate-forcers-on-arctic-climate-air-quality-and-human-health/3614)  
363 [health/3614](https://www.amap.no/documents/doc/amap-assessment-2021-impacts-of-short-lived-climate-forcers-on-arctic-climate-air-quality-and-human-health/3614)

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