

Arctic Tropospheric Ozone Trends

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RESEARCH LETTER

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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, and spring surface changes not captured
- Spring (summer) decreases (increases) in observed ozone throughout the troposphere, not always simulated by models

Supporting Information:

Supporting Information may be found in the online version of this article.

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Arctic Tropospheric Ozone Trends

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Abstract Observed trends in tropospheric ozone, an important air pollutant and short-lived climate forcer (SLCF), are estimated using available surface and ozonesonde profile data for 1993–2019, using a coherent methodology, and compared to modeled trends (1995–2015) from the Arctic Monitoring Assessment Program SLCF 2021 assessment. Increases in observed surface ozone at Arctic coastal sites, notably during winter, and concurrent decreasing trends in surface carbon monoxide, are generally captured by multi-model median trends. Wintertime increases are also estimated in the free troposphere at most Arctic sites, with decreases during spring months. Winter trends tend to be overestimated by the multi-model medians. Springtime surface ozone increases in northern coastal Alaska are not simulated while negative springtime trends in northern Scandinavia are not always reproduced. Possible reasons for observed changes and model performance are discussed including decreasing precursor emissions, changing ozone dry deposition, and variability in large-scale meteorology.

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

1. Introduction

Tropospheric ozone (O_3) is a short-lived climate forcer (SLCF) contributing to global and Arctic warming (AMAP, 2015; Sand et al., 2016; von Salzen et al., 2022), and a critical secondary air pollutant, detrimental to human health (Anenberg et al., 2010) and ecosystems (Arnold et al., 2018). The Arctic tropospheric O_3 budget is complex, as recently discussed in a companion paper, Whaley et al. (2023). It originates from photochemical

LAW ET AL. 1 of 12



Formal analysis: Jens L. Hjorth, Jakob B. Pernov, Cynthia H. Whaley, Henrik Skov, Martine Collaud Coen, David Tarasick, Peter Effertz, Irina Petropavlovskikh, Sverre Solberg Investigation: Kathy S. Law, Jens L. Hjorth, Jakob B. Pernov, Cynthia H. Whaley, Henrik Skov, Martine Collaud Coen, Joakim Langner, Stephen R.

Software: Jens L. Hjorth, Jakob B. Pernov, Martine Collaud Coen Visualization: Jens L. Hjorth, Jakob B. Pernov, Cynthia H. Whaley, Henrik Skov, Martine Collaud Coen, David Tarasick Writing – original draft: Kathy S. Law, Jens L. Hjorth, Jakob B. Pernov, Cynthia H. Whaley, Henrik Skov, Martine Collaud Coen, Joakim Langner, Stephen R. Arnold

Writing – review & editing: Kathy S. Law, Jens L. Hjorth, Jakob B. Pernov, Cynthia H. Whaley, Henrik Skov, Martine Collaud Coen, Joakim Langner, Stephen R. Arnold, David Tarasick, Jesper Christensen, Peter Effertz, Michael Gauss, Ulas Im, Naga Oshima, Irina Petropaylovskikh, David Plummer, Svetlana Tsyro, Stephen Turnock production of anthropogenic or natural emissions of O_3 precursors, including nitrogen oxides (NO_x), carbon monoxide (CO) and methane (CH_4), in the Arctic, or following air mass transport from mid-latitudes, as well as transport of O_3 from the stratosphere (Law et al., 2014; Schmale et al., 2018). Sinks include photochemical destruction, including reactions involving halogens leading to so-called ozone depletion events (ODE_3) (Barrie, et al., 1988; Simpson et al., 2007), and surface dry deposition (Clifton et al., 2020). Growth in anthropogenic emissions since pre-industrial times has led to increases in tropospheric O_3 throughout the Northern Hemisphere (OOE_3) (Squdel et al., 2018; Tarasick et al., 2019; Turnock et al., 2020) contributing to observed global and Arctic warming over the past century (e.g., Griffiths et al., 2021; Szopa et al., 2021). Since the mid-1990s, a mix of relatively weak positive and negative trends (+1 to -1 parts per billion by volume (ppbv) per decade) have been reported in the OEE_3 0 NH at the surface and in the free troposphere (OEE_3 1), with largest increases over south and eastern Asia, associated with increasing anthropogenic emissions (Cooper et al., 2020; H. Wang et al., 2022).

To date, only a few studies have focused on assessing tropospheric O_3 trends in the Arctic. While positive O_3 trends were diagnosed at several surface sites, results do not always have high certainty, and both positive and negative trends were reported at some Canadian sites (Cooper et al., 2020; Sharma et al., 2019; Tarasick et al., 2016). In the Arctic FT, studies found significant positive trends (B. Christiansen et al., 2017; H. Wang et al., 2022), no trends (Tarasick et al., 2016), or mixed trends in different seasons (Bahramvash Shams et al., 2019). Differences in the periods analyzed, sign or magnitude of trends, based on different methodologies, data averaging, etc. emphasizes the need to further examine trends using the same methodology. Coherent estimation of observed trends, and evaluation of modeled trends, is needed to better understand O_3 changes and impacts on Arctic climate that are sensitive to the altitude where O_3 perturbations occur (Rap et al., 2015). This study assesses annual/decadal and monthly trends, together with possible evolution in seasonal cycles, of Arctic tropospheric O_3 over the last 20–30 years. Observed changes are also compared to results from atmospheric chemistry-climate models run as part of the recent Arctic Monitoring and Assessment Programme (AMAP) SLCF assessment (AMAP, 2021; Whaley et al., 2022; von Salzen et al., 2022), taking into account reported model deficiencies (Whaley et al., 2023). Results are discussed in light of possible changes in sources and sinks of Arctic tropospheric O_3 .

2. Methods

2.1. Measurements

The location of surface and ozonesonde sites used in this study are displayed in Figure 1, together with the Arctic Circle at 66.6°N, used to define the Arctic. Decadal surface trends are shown in the table grouped into (a) high Arctic coastal sites (Alert, Utqiagvik/Barrow, Villum), Zeppelin (situated at 474 m on Svalbard) and Summit (high altitude (FT) site on Greenland (3,211 m)) and (b) European continental sites within (Pallas, Esrange), and just south (Tustervatn) of, the Arctic Circle.

Surface observations are from EBAS Level 2 data, station owners for Villum before 2001, Canada's Open Government Portal for Alert, and National Oceanic and Atmospheric Administration (NOAA) for Summit, and Barrow Atmospheric Observatory, Utqiagʻvik (Utqiagʻvik from now on). Ozonesonde data are from the World Ozone and Ultraviolet Radiation Data Center (WOUDC) and Network for the Detection of Atmospheric Composition Change (NDACC). See also Text S1, Figures S1 and S2 in Supporting Information S1, including data coverage.

2.2. Trend Analysis

Observed monthly and annual/decadal trends in surface O_3 concentrations at different sites are determined using a non-parametric Mann-Kendall method based on the 90th and 95th confidence limits (CLs) and Sen's slope methodology (Sen, 1968; Theil, 1950) and p-values (probability that trends occurred by chance). Daily median data are sorted into different months and pre-whitened, due to the presence of autocorrelation, via the 3PW algorithm from Collaud Coen et al. (2020). Trends using ozonesonde profiles are calculated based on weekly medians for selected pressure levels. See Text S2 in Supporting Information S1 for details and justification for use of our methods. We focus on discussing trends with high (95% CL, $p \le 0.05$) and medium certainty (90% CL, 0.05). CLs are shown in the Figures and <math>p-values are given in Figure 1 and Tables S2–S5 in Supporting Information S1.

LAW ET AL. 2 of 12

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Site	Trend (ppbv/10 years)	p-value	Period
High Arctic	:	•	
Alert	0.94	0.044	1999-2019
	0.79	0.038	1993-2019
Utqiagvik	1.51	0.129	1999-2019
	0.75	0.258	1993-2019
Villum	3.24	0.034	1999-2019
	1.13	0.162	1996-2019
Zeppelin	-0.36	0.497	1999-2019
	0.62	0.089	1993-2019
Summit	-1.28	0.142	2001-2019
European c	ontinental Arctic o	and near-	Arctic:
Esrange	0.24	0.697	1999-2019
	0.27	0.610	1993-2019
Pallas	-0.98	0.327	1999-2019
	-1.33	0.067	1995-2019
Tustervatn	-1.79	0.003	1999-2019
	-0.63	0.102	1994-2019

Figure 1. Left: Location of surface (bold) and ozonesonde (italic) sites and showing the Arctic Circle (66.55°N). Right: O_3 trends at surface sites in ppbv per decade and p-values. Trends (>90% confidence limit, $p \le 0.1$) are in bold. Geographical coordinates for all sites are provided in Whaley et al. (2023). See text for details.

2.3. Modeled Trends

Modeled trends at the surface and different altitudes are calculated for 1995–2015 using results from four global chemistry-climate models (CMAM, GISS-E2.1, MRI-ESM2, UKESM1) and two chemistry-transport models (DEHM, EMEP MSC-W) run using the same ECLIPSEv6b anthropogenic emissions, and nudged with meteorological reanalyses as part of AMAP (2021). Details can be found in Whaley et al. (2022), Text S3 and Table S1 in Supporting Information S1. Simulated monthly mean O_3 volume mixing ratios from the model grid box containing the measurement location are used to compute multi-model medians (MMMs). For ozonesonde comparisons, modeled vertical profiles are interpolated onto the same vertical bins as the measurements before trends are computed.

3. Surface Ozone Trends in the Arctic

3.1. Observed Ozone Trends

Annual and decadal trends are calculated for 1993–2019, or for the longest period with sufficient data, for all the sites (see Figure 1, Table S2 in Supporting Information S1).

Average O_3 seasonal cycles are also calculated for earlier (1993–2000) and later (2012–2019) periods, to examine possible changes, together with monthly trends (Figure 2, Table S3 in Supporting Information S1 for p-values) at selected sites (see Figure S3 in Supporting Information S1 for other sites). Monthly trends are also analyzed for different 21-year periods (1993–2013, 1999–2019) (Figure S4 in Supporting Information S1).

First considering high Arctic sites at coastal locations that exhibit a winter maximum with low spring concentrations attributed to ODEs, as discussed in Whaley et al. (2023). Alert has positive O_3 annual trends (p=0.044), as does Villum (p=0.034) for the shorter time period 1999–2019, while annual trends at Utqiagʻvik are not apparent (see Figure 1/Table S2 in Supporting Information S1). Trends are also calculated in particular seasons, as shown in Figure 2. Notably, positive trends are found during late autumn and/or winter at Alert, Villum and Utqiagʻvik (p's ≤ 0.022). Positive trends are also calculated in spring (April-May) and August at Utqiagʻvik. Winter trends at Alert and spring trends at Utqiagʻvik are more pronounced over the later record (1999–2019) (see Figure S4

LAW ET AL. 3 of 12

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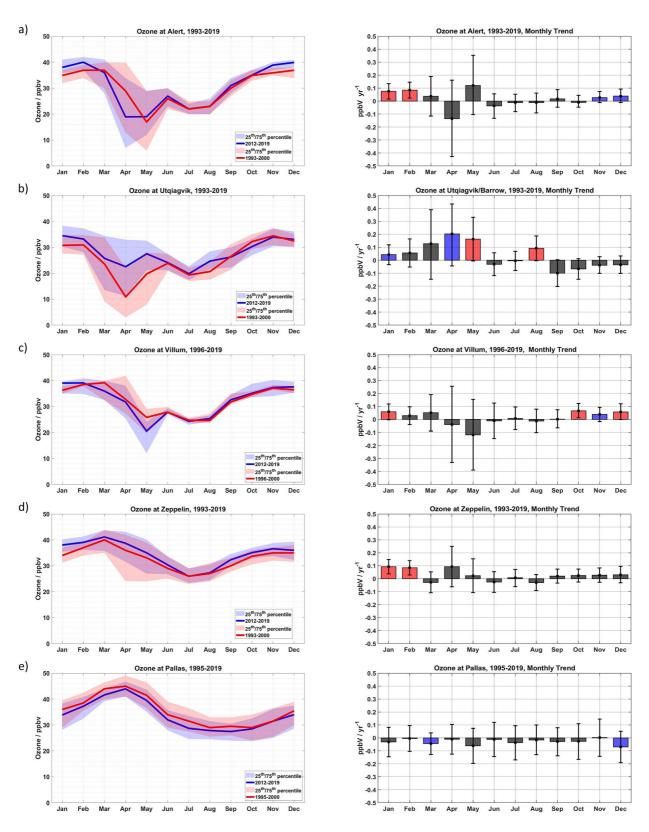


Figure 2. Observed surface O_3 trends and seasonal cycles. Left: seasonal cycles of monthly median O_3 (ppbv) at (a) Alert, (b) Utqiagʻvik, (c) Villum, (d) Zeppelin, and (e) Pallas for 1993–2000 (red lines) versus 2012–2019 (blue lines). Shaded areas show upper and lower quartiles of hourly values. Right: monthly trends for 1993–2019. Boxes represent the slope of the trend in ppbv per year with red boxes having 95th% confidence limit (CL), blue boxes 90th% CL, and black boxes are trends with low certainty. Error bars show 95th% CLs. Results are shown for shorter periods depending on data availability.

LAW ET AL. 4 of 12

in Supporting Information S1). To further characterize these changes, probability distributions in observed O_3 concentrations are calculated for periods with at least 90% CL monthly trends (see Figure S5 in Supporting Information S1). Positive trends during winter and spring at Utqiagʻvik are the result of a decrease (increase) in the frequency of low (high) concentrations (January–May), whereas wintertime O_3 concentrations shifted recently towards higher values at Alert (November–February) and Villum (October–January). Zeppelin shows a different seasonal behavior compared to Arctic sea-level coastal sites with a spring maximum, more similar to remote mid-latitude sites. Here, positive annual trends are estimated for 1993–2019 (Figure 1, p = 0.089), and in Jan./Feb (Figure 2, $p \le 0.001$), driven by increases in the earlier part of the record (1993–2013) (Figure S4 in Supporting Information S1).

Continental northern Scandinavian sites exhibit a different behavior with Tustervatn (p = 0.003), and Pallas, with lower certainty (p = 0.067), showing negative annual trends but no clear annual (or monthly) trends at Esrange (p > 0.151) over any of the periods considered. The shape of the seasonal cycle for the earlier versus the later period is similar at these sites, which also have a spring maximum like Zeppelin. O_3 appears to be decreasing throughout the year when comparing earlier and later periods although negative trends are only evident at Pallas (March, December), and at Tustervatn in spring and early summer for 1999–2019 trends (Figure S4 in Supporting Information S1). Summit is more representative of the FT and samples air masses transported from North America and Asia, or of stratospheric origin (Dibb, 2007; Schmeisser et al., 2018). No clear annual trend, calculated over the shorter 2001–2019 record, is seen, but negative monthly trends are estimated for January, March-May and September ($p \le 0.060$).

3.2. Comparison of Observed and Modeled Surface Trends

Figure 3 compares observed monthly and MMM trends for 1995–2015, or the closest possible time interval in case of years with missing observations. Results for other sites are shown in Figure S6 of Supporting Information S1 and p-values in Table S4 of Supporting Information S1. Observed trends are more frequently diagnosed over 1993–2019 (Figure 2) than over the shorter period ending in 2015 (Figure 3). While the MMMs simulate O_3 seasonal cycles reasonably well, low O_3 concentrations are missed in spring, and wintertime O_3 is underestimated (Whaley et al., 2023). The MMMs simulate positive trends at Zeppelin (Jan., p = 0.048) and negative trends at Esrange (May, p = 0.017), respectively, but not observed positive trends at Utqiagvik (April, p = 0.035). Trends are simulated, but not observed, at Alert (January, December, p = 0.058, 0.014), Zeppelin (April, p = 0.032), Villum (Sept., p = 0.035), and Tustervatn (March, p = 0.057).

4. Arctic Ozone Trends in the Free Troposphere

4.1. Observed Vertical Trends

This analysis focuses on O₃ changes in the lower and mid-troposphere. Figure 4 shows observed relative trends at six Arctic ozonesonde sites from 925 to 400 hPa for 1993–2019 (see *p*-values in Table S5 of Supporting Information S1). Absolute trends above and below 400 hPa, and relative trends from 925 to 100 hPa, providing information on changes in the upper troposphere (UT) and lower stratosphere (LS), are also calculated (Figures S7a and S7b in Supporting Information S1). Overall, while there are few high confidence trends, there seems to be a "dipole effect" with positive trends in winter and summer, and negative trends in spring and autumn. Positive winter (notably Jan.) trends are found up to 400 hPa at most sites (except Resolute and Sodankyla), and at Scoresbysund in early spring. Positive wintertime trends are more evident in the earlier period in the UTLS (Figure S8 in Supporting Information S1). Eureka, Resolute, and Sodankyla have periods with negative trends especially during spring and early summer in the lower troposphere (LT). Resolute decreases extend up to 500 hPa in March-April. Relative trends vary from -1.5% to +0.5-1.0% per year (Figure 4 and Figure S7b in Supporting Information S1) while stronger negative trends are diagnosed in later years (1999–2019) compared to 1993–2013 at all sites (Figure S8 in Supporting Information S1).

4.2. Comparison of Observed and Modeled Vertical Trends

Figure 5 shows observed ozonesonde and MMM trends for 1995–2015 up to 400 hPa (see Figure S9 in Supporting Information S1 for results up to 100 hPa). Only results from five models are used, since EMEP MSC-W only

LAW ET AL. 5 of 12

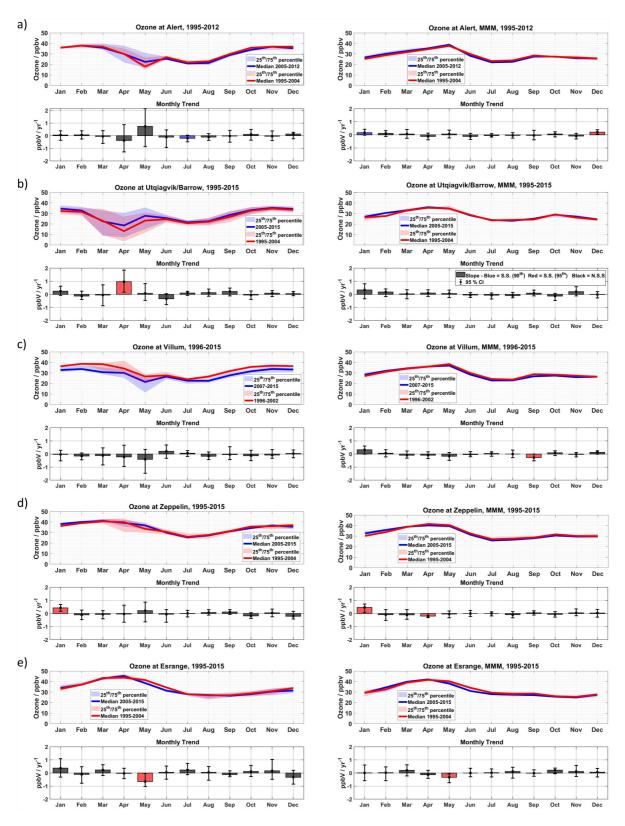


Figure 3. Comparison of observed (left) and multi-model median (right) surface O₃ trends and seasonal cycles at (a) Alert, (b) Utqiagvik, (c) Villum, (d) Zeppelin, and (e) Esrange. Upper panels: seasonal cycles for 1995–2004 (red lines) versus 2005–2015 (blue lines). Shaded areas show upper and lower quartiles of monthly values (observations only). Lower panels: monthly median trends in ppbv per year for 1995–2015, or shorter periods depending on data availability. Box coloring and error bars same as Figure 2.

LAW ET AL. 6 of 12

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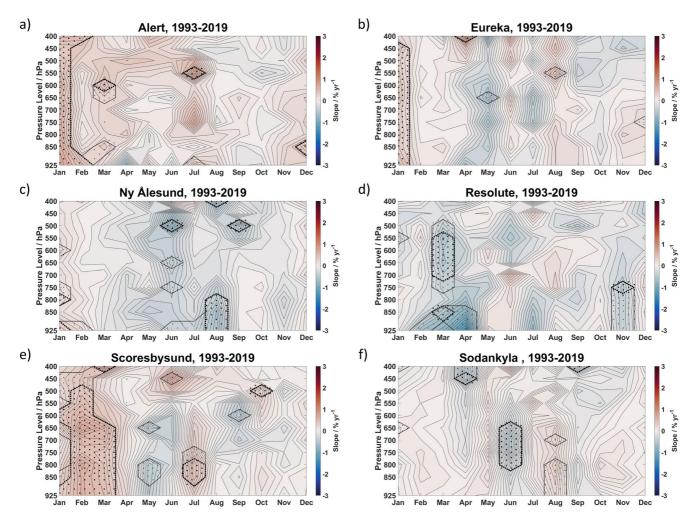


Figure 4. Vertical trends in observed monthly O_3 for 1993–2019, relative to monthly median concentrations, in % per year, from 925 to 400 hPa at (a) Alert, (b) Eureka, (c) Ny Ålesund, (d) Resolute, (e) Scoresbysund, and (f) Sodankyla. Stippled lines/areas show trends having 90th% confidence limit (CL) (smaller marker size) and 95th % CL (larger marker size).

provided surface O₃. The MMMs appear to capture the observed "dipole effect" seen in the observed trends. Models also capture observed increases in the winter but trends are overestimated at most sites, especially at Ny Ålesund. Negative winter trends at Resolute are not simulated. This may be linked to positive modeled winter trends above 500 hPa at all sites (see also Figure S9 in Supporting Information S1). Summertime positive MMM trends are larger than observed trends at some sites, for example, Resolute and Ny Ålesund.

5. Discussion and Conclusions

Increasing annual surface O_3 trends at Arctic coastal sites, and at Zeppelin, are in qualitative agreement with Cooper et al. (2020), but in contrast to negative or non-significant surface trends at Canadian sites (Tarasick et al., 2016). A notable finding is that positive trends occur mainly in the winter months. While such increases were reported previously at Utqiagʻvik (A. Christiansen et al., 2022; Cooper et al., 2020) and Alert (Sharma et al., 2019), we confirm this tendency over the wider Arctic. Emission reductions of NO_x in Europe and North America, and more recently over eastern Asia, have led to increasing wintertime O_3 at mid-latitudes due to less nitrogen oxide titration of O_3 (Bowman et al., 2022; Jhun et al., 2015; T. Wang et al., 2022). This can explain observed increases in wintertime surface Arctic O_3 , influenced primarily by transport of air masses from Europe (Hirdman et al., 2010). Evidence for declining O_3 precursor trends is supported by decreases in observed CO in the Arctic during autumn and winter (Figure S10 in Supporting Information S1). At the same time, CH_4 continues to increase globally contributing to rising O_3 in the NH (Zeng et al., 2022) (see also Text S4 in Supporting Information S1 on Arctic O_3 precursor trends).

LAW ET AL. 7 of 12

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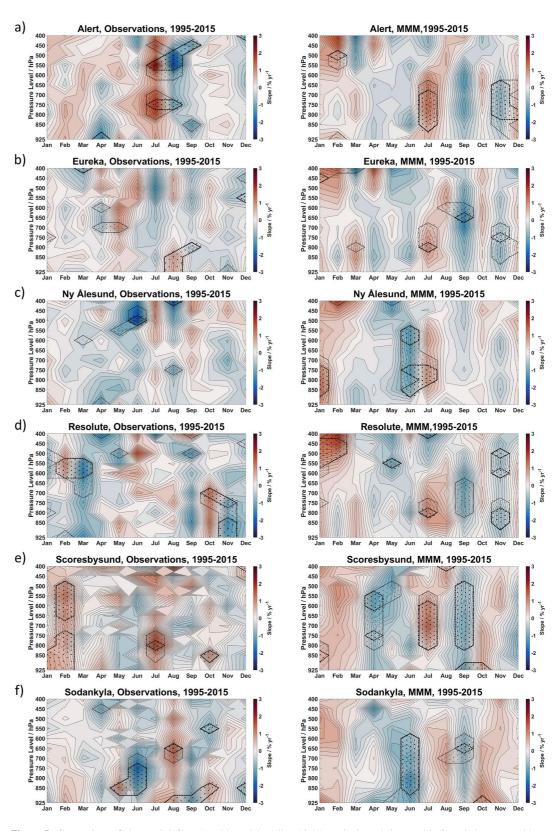


Figure 5. Comparison of observed (left) and multi-model median (right) vertical trends in monthly O_3 , relative to monthly medians, in % per year, from 925 to 400 hPa over 1995–2015 at (a) Alert, (b) Eureka, (c) Ny Ålesund, (d) Resolute, (e) Scoresbysund, and (f) Sodankyla. Shading/symbols are as in Figure 4.

LAW ET AL. 8 of 12

Another intriguing finding is springtime surface O₃ increases at Utqiagvik (especially over 1999–2019, Figure S4 in Supporting Information S1), but no discernible trends at Alert and Villum. Changes in O₃ concentrations at this time of year may be driven by changes in ODE frequency linked to climate change or weather patterns (Oltmans et al., 2012). ODEs lead to zero or very low springtime O₃ due to bromine released from frost flowers or blowing snow (on sea-ice) (Simpson et al., 2007; Yang et al., 2008, 2010) or iodine compounds with a possible oceanic source (Benavent et al., 2022). Increasing prevalence of first year sea-ice leading to increasing sea-spray aerosols from blowing snow (Confer et al., 2023) may explain increases in springtime tropospheric bromine oxide, observed from satellites, along the north coast of Greenland and central Arctic Ocean (Bougoudis et al., 2020). Indeed, the frequency of low springtime O₃ concentrations has been increasing at Canadian high Arctic sites (see Figure S11 in Supporting Information S1) but no clear springtime monthly trends are determined at Alert or Villum in our analysis. Springtime increases at Utqiagvik could be due to stronger transport from mid-latitudes to this site during periods with a more northerly extension of the Pacific storm track, hampering conditions for ODEs (Koo et al., 2012). They could also be due to an increasing influence from local emissions, such as shipping or Alaskan petroleum extraction, when photochemistry becomes active in spring (Gunsch et al., 2017).

Decreases in springtime/early summer O_3 in northern Scandinavia, especially over the later 1999–2019 period, are consistent with negative trends reported at Tustervatn since 1995 (Cooper et al., 2020), and in northern Sweden during summer (Andersson et al., 2017). These decreases are associated with lower maximum O_3 concentrations linked to reductions in European precursor emissions leading to less photochemical O_3 production (Cooper et al., 2020) although no clear trends in observed springtime CO are found at Zeppelin (Figure S10 in Supporting Information S1). Springtime negative trends at Summit may also be due to emission reductions over North America. Our results do not suggest a shift in the O_3 seasonal cycle toward higher concentrations in the spring (i.e., moving back toward pre-industrial O_3 seasonality) as reported at NH mid-latitudes (Bowman et al., 2022). Another explanation for decreasing springtime O_3 at the surface could be that reductions in snow cover due to climate warming (Mudryk et al., 2020) are leading to more O_3 dry deposition to Scandinavian forests.

The observed and modeled surface trend comparison covers 1995–2015, thereby missing the later time period when stronger observed O_3 trends are found, especially positive trends in winter. MMMs capture wintertime O_3 increases at Zeppelin, but overestimate at Alert although they simulate decreasing surface winter CO at Alert and Utqiagvik (Figure S10 in Supporting Information S1). This suggests that while anthropogenic emission changes may be captured, other model deficiencies may be contributing, such as modeling shallow boundary layers, O_3 deposition or NO_x lifetimes, as noted by Whaley et al. (2023). The MMMs miss springtime increases at Utqiagvik. This could be due to incorrect simulation of transport patterns (Oltmans et al., 2012) or missing surface halogen chemistry leading to incorrect modeled seasonality (Whaley et al., 2023). Negative springtime (May) Scandinavian trends are not always reproduced, possibly reflecting issues in the emission trends or modeled dry deposition.

Positive FT O₃ trends in winter at most Arctic sites are found, in common with several coastal Arctic surface sites, and in-line with increases reported at NH mid-latitudes (Gaudel et al., 2018), and at Canadian ozonesonde sites, except Resolute (H. Wang et al., 2022). Patterns in observed trends are quite well captured by the MMMs over 1995–2015, notably positive trends in winter and summer, although they tend to be overestimated. Positive summer trends may be linked to increased photochemical production from increased lightning and boreal fires due to climate warming (Veraverbeke et al., 2017). Observed negative trends in spring, extending from near the surface into the FT, are generally reproduced, and are likely to be due to decreasing NO_x emissions leading to lower FT O₃ where photochemical production is NO_x-limited. Negative LT trends could also be due to increasing ODEs extending over 100 kms and up to 1.5 km (Yang et al., 2020; Zilker et al., 2023). Overestimation of winter trends contrasts to previous studies where models underestimated NH trends (A. Christiansen et al., 2022; H. Wang et al., 2022). This may be due to differences in model transport or O₃ precursor emission trends, including NO_x reductions (see also Text S4 in Supporting Information S1). AMAP models overestimate mid-latitude FT O₃ (Whaley et al., 2023), possibly suggesting a larger sensitivity to precursor emission changes.

Observed trends in the UT (LS) appear to have switched from positive to negative since 1993 in winter/spring, which may explain stronger positive FT trends in the earlier part of the record (1993–2013). More frequent positive phases of the Arctic Oscillation in recent years may be contributing with a weaker Brewer-Dobson circulation leading to less transport of stratospheric O_3 into the Arctic UTLS, a higher tropopause height, and thus lower O_3 concentrations in this region (Zhang et al., 2017). However, Liu et al. (2020) did not detect any trend in the

LAW ET AL. 9 of 12

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stratospheric O₃ flux into the Arctic UT. On the other hand, H. Wang et al. (2022) attributed FT increases in NH mid-high latitude O₃ to increases in aircraft NO_x emissions.

Overall, this study identifies trends with high-medium certainty in observed Arctic tropospheric O₃ that are generally quite well captured by MMM results, Further investigation into the causes of observed trends, and model performance, are needed taking into account uncertainties in the observations and models (Fiore et al., 2022; Young et al., 2018), including known model issues (Whaley et al., 2023).

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

Surface O₃ monitoring datasets are provided by EMEP (European Monitoring and Evaluation Program), and Global Atmosphere Watch (GAW) World Data Centre for Reactive Gases. EMEP and GAW O₂ data are available via the EBAS data portal (from end of 1989 to present). CO data at Utqiagvik/Barrow and Zeppelin are also available via the EBAS data portal: http://ebas.nilu.no. Select the station name, and the component (CO, O₃) to access the data files. Canadian surface O₃ data can be downloaded from: https://data-donnees.ec.gc.ca/data/ air/monitor/networks-and-studies/alert-nunavut-ground-level-ozone-study/. Canadian surface CO is available https://data-donnees.ec.gc.ca/data/air/monitor/national-air-pollution-surveillance-naps-program/?lang=en. Click on folders Data, Year, Continuous Data, then Hourly Data. Surface O₃ records for Utqiagvik/Barrow (BRW) and Summit (SUM) are provided by P. Effertz and I. Petropavlovskikh via NOAA GML. Data is available at https://gml.noaa.gov/aftp/data/ozwv/SurfaceOzone/, Click on the directories for BRM or SUM to obtain the data. Surface O₃ measurements at Summit are made possible via the U.S. National Science Foundation Office of Polar Programs and their contract with Battelle Arctic Research Operations (contract #49100420C0001). Ny Ålesund, Scoresbysund and Sodankylä ozonesonde data are obtained as part of the NDACC. Data is available via https:// ndacc.larc.nasa.gov/index.php/stations. Click on the relevant site location to access the data files. Ozonesonde data for Alert, Resolute and Eureka have been reprocessed according to Tarasick et al. (2016), available at https:// hegiftom.meteo.be/datasets/ozonesondes.

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

Open-source codes for the Mann-Kendall test associated with Sen's slope are distributed under the BSD 3-Clause Licence in dedicated GitHub repositories hosted within the "mannkendall" directory (https://github.com/ mannkendall): Matlab (Collaud Coen & Vogt, 2020, https://doi.org/10.5281/zenodo.4134618, https://github. com/mannkendall/Matlab), Python (Vogt, 2020, https://doi.org/10.5281/zenodo.4134435, https://github.com/ mannkendall/Python), and R (Bigi & Vogt, 2020, https://doi.org/10.5281/zenodo.4134632, https://github.com/ mannkendall/R). Last access for all codes 27 January 2023.

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