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Investigation of the summer 2018 European ozone air pollution episodes using novel satellite data and modelling

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Abstract:

In the summer of 2018, Europe experienced an intense heat wave which coincided with several persistent large-scale ozone (O_3) pollution episodes. Novel satellite data of lower tropospheric column O_3 from the Global Ozone Monitoring Experiment-2 (GOME-2) and Infrared Atmospheric Sounding Interferometer (IASI) on the MetOp satellite showed substantial enhancements in 2018 relative to other years since 2012. Surface observations also showed ozone enhancements across large regions of continental Europe in summer 2018 compared to 2017. Enhancements to surface temperature and the O_3 precursor gases carbon monoxide and methanol in 2018 were co-retrieved from MetOp observations by the same scheme. This analysis was supported by the TOMCAT chemistry transport model (CTM) to investigate processes driving the observed O_3 enhancements. Through several targeted sensitivity experiments we show that meteorological processes, and emissions to a secondary order, were important for controlling the elevated O_3 concentrations at the surface. However, mid-tropospheric (~500 hPa) O_3 enhancements were dominated by meteorological processes. We find that contributions from stratospheric O_3 intrusions ranged between 15 - 40%. Analysis of back trajectories indicates that the import of O_3-enriched air masses into Europe originated over the North Atlantic substantially increasing O_3 in the 500 hPa layer during summer 2018.

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1. Introduction

Over the past two decades there have been several intense summer-time heatwaves over Europe (e.g. 2003 over continental Europe (Scott et al., 2004), 2006 over north-western Europe (Rebetez et al., 2008) and 2010 across eastern Europe and Russia (Matsueda et al., 2011)). With current and future climate change, increasing average global surface temperature is expected to trigger more frequent and intense heatwaves (Lhotka et al., 2017; Guerreiro et al., 2018). The summer-time 2018 heatwave across predominantly north-western and central Europe and Scandinavia generated temperature anomalies of approximately 2.0-4.0 K (Li et al., 2020; Drouard et al., 2020). Dynamically, it was caused by a combination of intense anticyclonic blocking systems, Rossby wave dynamics and the positive phase of the summer-time North Atlantic Oscillation (NAO+) (Li et al., 2020; Liu et al., 2020; Drouard et al., 2020). Environmentally, the summer 2018 heatwave caused severe drought conditions with decreased precipitation and soil moisture content (Bastos et al., 2020; Dirmeyer et al., 2020), while negatively impacting natural vegetation (e.g. decreased gross primary productivity (Smith et al., 2020; Bastos et al., 2020)). From a human health perspective, the 2018 heatwave caused 863 temperature related excess deaths in the UK (PHE, 2019).

As well as dynamical and vegetation responses, enhancements in atmospheric pollutants from heatwaves can lead to a degradation in air quality (AQ). Firstly, anticyclonic conditions (i.e. atmospheric blocking) have been shown to cause the accumulation of primary air pollutants such as carbon monoxide (CO; Thomas and Devasthale, 2014), nitrogen dioxide (NO_2; Pope et al., 2014) and particulate matter (i.e. $PM_{2.5}$; Graham et al., 2020) to hazardous levels. Secondly, higher temperatures during blocking events, which can trap and accumulate existing pollutants (e.g. Pope et al., 2016), can lead to the secondary formation of tropospheric ozone (O_3). Elevated surface O_3 is associated with adverse health impacts (Doherty et al., 2017; Jerrett et al., 2009) with ailments such as asthma, reduced lung function and disease (WHO, 2021). It also has adverse impacts on the natural biosphere (Sitch et al., 2007) and agriculture (Hollaway et al., 2012; van Dingenen et al., 2009).

In this study, we use surface and satellite observations of O_3, in combination with the well-evaluated TOMCAT global chemical transport model (CTM), to investigate the impact of the summer 2018 heatwave on European AQ and determine the key processes driving observed surface/tropospheric O_3 enhancements. We describe the observations and model we have used in Section 2. Section 3 and Section 4 discusses our results and discussion/conclusions, respectively.

2. Observations and Model

2.1. Satellite and Surface Observations

We use satellite observations of lower tropospheric O_3 (i.e. sub-column O_3 (SCO_3) between the surface and 450 hPa) from the Global Ozone Monitoring Experiment (GOME-2) and the Infrared Atmospheric Sounding Interferometer (IASI) instruments on-board ESA's MetOp-A satellite, which was launched in 2006 into a sun-synchronous polar orbit with equator crossing times of 9:30 (day) and 21:30 (night). GOME-2 is a nadir-viewing spectrometer with spectral coverage in the ultraviolet-visible (UV-Vis) of 240–790 nm (Riese et al., 2012) and a ground footprint of 40 km × 80 km in the first part of the mission and 40 km x 40 km from 2013 (once Metop-B was commissioned). IASI is a Michelson interferometer which observes the infrared spectral range 645 to 2760 cm^{-1} with spectral sampling of 0.25 cm^{-1} (Illingworth et al., 2011). It measures simultaneously in four fields of view

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(circular at nadir with a diameter of 12 km) which are scanned across track to sample a 2200 km-wide swath (Clerbaux et al., 2009).

For GOME-2, the Rutherford Appleton Laboratory (RAL) scheme uses an optimal estimation algorithm (Rodgers, 2000) to retrieve ozone height-resolved ozone distributions spanning the stratosphere and troposphere (Miles et al., 2015). The scheme applied to GOME-2 has been developed from that used first for GOME-1 on-board ERS-2 (Munro et al., 1998; Forster et al., 2007). This is a multi-step scheme in which profile information is first retrieved in the stratosphere by exploiting wavelength-dependent absorption in the O_3 Hartley band (270-307nm) and is then extended into the troposphere by exploiting temperature-dependent spectral structure in the O_3 Huggins bands (325-335nm). For IASI, O_3 profiles are retrieved using an extended version of RAL's Infrared-Microwave-Sounding (IMS) scheme, which is described in Pope et al., (2021) and Palmer et al., (2022). The IMS core scheme was originally developed to retrieve temperature, water vapour and stratospheric O_3 profiles along with surface spectral emissivity and cloud jointly from co-located measurements by IASI, the Microwave Humidity Sounder (MHS) and the Advanced Microwave Sounding Unit (AMSU-A) on MetOp (RAL Space, 2015). GOME-2 O_3 data were filtered for a geometric cloud fraction less than 0.2, a solar zenith angle less than 80°, a cost function less than 200.0 and a convergence flag equal to 1.0. IASI data were filtered for a geometric cloud fraction less than 0.2 and a cost function less than 1000.0.

We also use surface O_3 observations from the European Monitoring and Evaluation Programme (EMEP) network for May-August 2017 and 2018. In total, we used 83 spatial collocated EMEP sites in both years years across Europe. Here, data at individual sites were selected where the corresponding data flag was set to 0.0.

2.2. Modelling & Sensitivity Experiments

In this study the TOMCAT CTM (Chipperfield, 2006) is forced by European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim reanalysis meteorology (Dee et al., 2011) and run at a horizontal resolution of 2.8° × 2.8° with 31 vertical levels from the surface to 10 hPa. The model includes detailed tropospheric chemistry, including 229 gas-phase reactions and 82 advected tracers (Monks et al., 2017), and heterogeneous chemistry driven by size-resolved aerosol from the GLOMAP module (Mann et al., 2010). Simulations used here include year-specific anthropogenic emissions from MACCity (Granier et al., 2011) and fire emissions from the Global Fire Assimilation System (GFAS, Kaiser et al., 2012). Year-specific off-line biogenic volatile organic compounds (VOCs) emissions for acetone, methanol, isoprene and monoterpenes were simulated by the Joint UK Land Environment Simulator (JULES – Pacifico et al., 2011; Best et al., 2011; Clark et al., 2011). All other biogenic VOC emissions are climatological values and provided by the Chemistry-Climate Model Initiative (CCMI) (Morgenstern et al., 2017). The global budgets of the JULES VOC emissions are low in comparison to the climatological CCMI emissions, so were scaled up on a regional basis, while retaining the 2017-2018 step change related to the 2018 summer heat wave. The full details of JULES VOC emissions scaling can be found in **Supplementary Material (SM) 3**. The model was run for 2017 and 2018 with output at 6-hourly intervals (i.e. 00, 06, 12 and 18 UTC). Here, each year was run with its respective meteorology and emissions and given the labels Met17_Emis17 (representing 2017) and Met18_Emis18 (representing 2018).

To explore the importance of emission and meteorological processes behind the elevated European summer 2018 tropospheric O_3 levels, a 1-year model sensitivity experiment was performed using

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2018 meteorology but 2017 emissions (i.e. Met18_Emis17). Therefore, the difference between Met18_Emis17 and Met17_Emis17 highlights the impact of fixed emissions (i.e. 2017 emissions for both years), while the Met18_Emis18 minus Met18_Emis17 highlights the impact of fixed meteorology (i.e. 2018 meteorology for both years). These are compared with the control differences for 2018-2017 (Met18_Emis18- Met17_Emis17). From here on in, we refer to the control differences, fixed emission differences and the fixed meteorology differences as CTL_DIFF, FIXED_EMIS_DIFF and FIXED_MET_DIFF, respectively. TOMCAT also includes a stratospheric O_3 tracer (i.e. tags O_3 in the model which originated in the stratosphere). This was used to investigate the impact of stratospheric O_3 intrusion into the troposphere.

TOMCAT has been used in a number of previous studies to investigate air quality and tropospheric composition (e.g. Richards et al., 2013; Emmons et al., 2015; Pope et al., 2016; Pope 2020) whose results give confidence in the model's ability to simulate European tropospheric O_3 in this study. Overall, when compared with observations, TOMCAT has good spatial agreement with both GOME-2 and IASI and can reasonably reproduce the 2018 SCO_3 enhancement in 2018 verses 2017 (**SM 4**). The model also has good agreement, both in magnitude and seasonality, with the EMEP observed surface concentrations (**SM 4**).

2.3 ROTRAJ Back-trajectories

We use the Reading Offline Trajectory Model (ROTRAJ) to generate air mass back-trajectories (Methven et al., 2003) to assess the import of tropospheric O_3 into Europe. ROTRAJ is a Lagrangian atmospheric transport model driven by meteorology from the same ECMWF ERA-Interim reanalyses as used by TOMCAT. Velocity fields at the Lagrangian particle positions are determined by cubic Lagrange interpolation in the vertical, bilinear interpolation in the horizontal and linear interpolation in time. Kinematic back-trajectories were released at 6-hourly intervals (i.e. at 00, 06, 12 and 18 UTC) from Paris and Berlin, both central locations over Europe in the region of summer-time 2018 O_3 enhancements, between the 1st May and 31st August for both 2017 and 2018. The trajectories were released at the surface and at approximately 500 hPa and integrated for 10 days with 6-hourly output (i.e. 41 trajectory points including the starting location) to investigate the origin of air masses arriving in these altitude regions of enhanced summer-time O_3 in 2018. In total, ROTRAJ was therefore run 8 times (2 years × 2 altitudes × 2 locations).

To quantify the import of tropospheric O_3 into Europe, for each trajectory, all the trajectory points were co-located with corresponding TOMCAT O_3 mixing ratio values (i.e. the horizontal and vertical grid box the trajectory point sits within and corresponding time stamp) and then the average O_3-weighted back-trajectory (O_3-WBT) determined (i.e. back-trajectories with larger O_3WBT values come from air masses with larger O_3 content).

3. Results

3.1 Surface Temperature

Several studies (e.g. Li et al., 2020; Liu et al., 2020; Drouard et al., 2020) have documented the intense heat wave across Europe in the summer of 2018. This is further shown in **Figure 1** which compares surface temperature, co-retrieved with ozone and other variables from MetOp-A by the IMS scheme, between 2017 and 2018. In May, higher temperatures occur across Scandinavia (5.0-10.0 K), eastern Europe (3.0-7.0 K) and the UK (1.0-3.0 K), but temperatures are lower (-3.0 to -1.0 K) across Iberia. In June, a similar spatial distribution occurs but the magnitude of the differences is

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smaller. In July the largest temperature increases range from 6.0-8.0 K in Scandinavia to 2.0-6.0 K in the UK/France. Iberia continued to experience temperatures lower by -2.0 to 0.0 K. In August, there are near-zero differences over the UK, Iberia and most of Scandinavia but with increases of 1.0-3.0 K over eastern Europe and Finland.

3.2 Satellite Ozone

We investigate the longer term variability in tropospheric O_3 (i.e. SCO_3) to determine if 2017 is a suitable comparator for the 2018 summer O_3 enhancements as it is for temperature. **Figure 2** shows the 2012-18 SCO_3 average between May and August for a domain over the Atlantic and Europe and the difference for the same season between specific years and the 2012-18 average. In 2012 and 2013, there are significant positive differences from the average between 1.0 DU and 5.0 DU over much of the domain. Over continental Europe, the differences are smaller. Here, the significance of differences between the year-specific and long-term averages are determined using the Wilcoxon Rank test (Pirovano et al., 2012) at the 95% confidence level. In 2014 and 2015, there are negative differences across Europe (-4.0 DU to -1.0 DU). In 2016, similar negative differences are primarily across the north and south-east of the domain. In 2017, there are near-zero differences across the Atlantic, UK and western Europe. Over eastern Europe and Mediterranean, there are significant negative differences of between -2.0 DU and -1.0 DU. In 2018, across continental Europe there are significant positive differences between 2.0 DU and 4.0 DU. As the 2017 differences are relatively small in magnitude with a low proportion of significant pixels (i.e. Sig Pixels % = 32.7 is the lowest across the 7 years), it is representative of average conditions for comparison with 2018. For 2018, the summer SCO_3 enhancements across continental Europe are the largest for the years shown with a coherent cluster of significant differences. This illustrates that the summer 2018 SCO_3 enhancements are a substantial deviation from the average conditions (which we represent as 2017 hereon) and that this is an intense O_3 event.

Investigation of SCO_3 retrieved from both GOME-2 (**Figure 3**) and the IMS scheme (**Figure 4**) show consistent enhancements in summer 2018. In 2017, between May and August, GOME-2 typically observed SCO_3 values between 20.0-30.0 DU across continental Europe. Peak SCO_3 values occurred over the Mediterranean (30.0-38.0 DU); relatively high ozone is a typical feature of the Mediterranean in summer (Richards et al., 2013). In 2018, the seasonality is consistent with 2017, but the continental European SCO_3 values ranged between 25.0 DU and 35.0 DU. For the 2018-2017 difference, SCO_3 enhancements occur across continental Europe in all four months, but peaked in May and July between 3.0 DU and 8.0 DU, while typically 1.0-5.0 DU in June and August. The spatial distribution of IMS-retrieved SCO_3 is similar to that of GOME-2 in 2017 and 2018, although the absolute values tend to be systematically lower by 3.0-4.0 DU. However, despite this systematic offset, the 2018-2017 differences are reasonably consistent with GOME-2. Across continental Europe, IMS SCO_3 shows 2018 enhancements in all months investigated, but peaks in May and July, like GOME-2, between 3.0 DU and 6.0 DU. The differences range from 1.0 DU to 3.0 DU in June and are approximately 1.0 DU in August (though a peak enhancement of 3.0-5.0 DU occurs over the Mediterranean). Spatial correlations between the GOME-2 and IASI difference (i.e. 2018-2017) maps for the months investigated ranged between 0.21 and 0.47 (see **SM 4**).

The GOME-2 and IASI instruments observe UV-Vis and IR wavelengths, with peak vertical sensitivities to tropospheric O_3 in the lower and mid/upper troposphere, respectively. Consistency in the 2018

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enhancements in SCO_3 indicates that these extend over the bulk of the troposphere and increases confidence in the differences for both sensors.

Investigation of several satellite-retrieved O_3 precursor gases (see **SM 1**) showed enhancements in total column methanol ($TCCH_3OH$, **Figure S1**), especially linked to May and July temperature enhancements (**Figure 1**), minor increases in tropospheric column NO_2 ($TCNO_2$, **Figure S2**) in May and July over central Europe and widespread enhancements (weakest in July and strongest in August) in total column carbon monoxide (TCCO, **Figure S3**). Investigation of the GOME-2 and IASI total column O_3 (TCO_3) differences between 2017 and 2018 (**Figures S4 & S5**) showed these to be in close agreement. Some spatial structure is similar to that of the SCO_3 difference patterns (**Figures 3 and 4**), with correlations of approximately 0.5 between TCO_3 and SCO_3 for each instrument (see **SM 2**). Given the complex relationship between tropospheric O_3, precursor gases, atmospheric chemistry (e.g. NO_x or VOC-limited regimes), surface deposition and meteorological conditions (e.g. atmospheric temperatures and transport), a detailed chemistry transport model is required to assess the key processes leading to the observed SCO_3 enhancements over Europe.

3.3 Surface Ozone

Increased temperatures during heat waves have been shown to enhance surface O_3 concentrations (e.g. Jacob and Daniel, 2009; Vieno et al., 2010; Pyrgou et al., 2018). In the summer (May-June-July-August, MJJA) of 2018, EMEP recorded larger O_3 mixing ratios across most of Europe in comparisons to 2017 (**Figure 5a & b**). Over central Europe, surface O_3 mixing ratios ranged from approximately 45.0 ppbv to over 60.0 ppbv, while in 2017 it was 35.0 ppbv to 50.0 ppbv. Over the UK and north-western Europe, surface O_3 mixing ratios ranged from 20.0 ppbv to 30.0 ppbv and then 25.0 ppbv to 35.0 ppbv in MJJA 2017 and 2018, respectively. In Scandinavia and eastern Europe, surface O_3 mixing ratios ranged from 20.0 ppbv to 35.0 ppbv in MJJA 2017, while increasing to 25.0 ppbv to approximately 40.0 ppbv in MJJA 2018. **Figure5c** highlights these widespread enhancements where domain-average surface O_3 mixing ratios are larger by typically 5.0-10.0 ppbv in May and from mid-June to mid-August in 2018. **Figure 5d** shows that the domain median surface O_3 concentration across MJJA was larger by 2.0-3.0 ppbv, but the 2018 extremes were greater with 75^{th} and 95^{th} percentiles of 42.0 ppbv and 53.0 ppbv in 2017 and 47.0 ppbv and 59.0 ppbv in 2018. Therefore, surface observations of O_3 recorded widespread enhancements in MJJA 2018 compared to 2017 with peak site differences >10.0 ppbv. This is generally consistent with the 2018 layer-averaged enhancements in the satellite-retrieved SCO_3 for regions where both datasets have spatial coverage.

3.4. Model Simulations

We use the TOMCAT model to investigate different factors potentially driving the observed enhancements in tropospheric O_3. In comparisons with the observations (see **SM 4**) the model reproduces the sign and spatial distribution of observed 2018-2017 differences reasonably well. Although it has a tendency to underestimate the absolute magnitude, we are confident in the model's ability to simulate the tropospheric O_3 enhancements relative to 2017.

At the surface (**Figure 6**), TOMCAT CTL_DIFF (i.e. Met18_Emis18 - Met17_Emis17) suggests that O_3 is enhanced in May over Scandinavia (2.0- >5.0 ppbv), north-western Europe (0.0-2.0 ppbv), the Arctic Ocean (>5.0 ppbv) and off the coast of Iberia (3.0-5.0 ppbv). However, negative values exist over eastern Europe (-3.0 ppbv to -1.0 ppb) and the Atlantic west of Ireland (-3.0 ppbv to -1.0 ppb). In June, the negative differences persist in eastern Europe (-3.0 ppbv to -1.0 ppb), but positive

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differences are located over northern Scandinavia (1.0-2.0 ppbv) and the North Atlantic (2.0-4.0 ppbv). For July, CTL_DIFF shows the largest enhancements over continental Europe (i.e. Po Valley, France, Benelux region and Iberia) and the UK (>5.0 ppbv). Negative differences of between -3.0 ppbv and -1.0 ppbv remain over eastern Europe. In August, the only clear differences are over Iberia and the western Mediterranean, ranging between 3.0 ppbv and >5.0 ppbv. Overall, TOMCAT simulates sub-regional surface O_3 enhancements over Europe, which are generally consistent with EMEP observations apart from several sites over eastern Europe.

At 500 hPa, TOMCAT CTL_DIFF shows larger-scale O_3 enhancements in 2018 compared to 2017 (>5.0 ppbv) throughout May to August. In May and August, there are, however, a few negative differences (-5.0 ppbv to -3.0 ppbv) over far eastern Europe. In June and July, the full domain is more or less dominated by O_3 enhancements in 2018. In Figures **4** and **5** (and **SM 4**), GOME-2 and IASI (and TOMCAT with the instrument averaging kernels (AKs) applied to account for the vertical sensitivity of the retrievals, see **SM 4** for more information) show SCO_3 enhancements during these months of 2018. Given the vertical extents and peak heights of their retrieval sensitivities and consistency in spatial patterns (**Figs SM-8** and **10**) it is evident that the O_3 enhancements detected by GOME-2 and IASI extend over the free troposphere. The model shows large-scale O_3 enhancements in the free troposphere and similar patterns to GOME-2 and IASI when averaging kernels applied. So the model corroborates this finding from the satellite retrievals. Signals from EMEP and TOMCAT at the surface, on the other hand, are more mixed across the domain.

The right-hand column of **Figure 6** shows the relative difference in the stratospheric O_3 contribution to the 500 hPa O_3 layer (i.e. Strat % @ 500 hPa), from CTL_DIFF, between 2017 and 2018. Here, the percentage of stratospheric O_3 contributing to the O_3 concentration at the 500 hPa is calculated for 2017 and 2018 and then the 2018-2017 difference determined. The largest enhancement to the 500 hPa layer was in July where the stratospheric O_3 contribution increased by 3.0% to >5.0% across Europe. In June and August, the spatial patterns are similar with stratospheric O_3 contribution enhancements of 3.0-5.0% across southern Europe, Scandinavia and the North Atlantic (above the UK). In the North Atlantic, UK and northern Europe, there are near-zero changes in June and August. In May, there are enhancements >5.0% across the northern region of the domain and northern Africa, while smaller enhancements (1.0%-3.0%) over the UK and near-zero changes over eastern Europe. This is partially supported by analysis of TCO_3 (see **SM 2**) where there are reasonable spatial correlations (~0.5 to 0.6) between the SCO_3 2017-2018 summer differences and the equivalent for TCO_3. Therefore, these results indicate a potentially enhanced contribution of stratospheric O_3 into the mid-troposphere during the summer of 2018 across Europe.

To quantify the separate importance of precursor emissions and meteorology in governing the summer 2018 O_3 enhancements we compare the sensitivity experiments with the control runs. **Figure 7** (left column) shows the results for the fixed emissions differences (i.e. FIXED_EMIS_DIFF) between years (i.e. Met18_Emis17 – Met17_Emis17). At the surface, the FIXED_EMIS_DIFF show similar spatial patterns to that of CTL_DIFF (**Figure 6** – left column). The domain spatial difference correlations between these simulations is greater than 0.96 for all months considered. However, FIXED_EMIS_DIFF (**Figure 7** - left column) tends to be lower than CTL_DIFF (**Figure 6** – left column) by approximately 0.0-2.9 ppbv (i.e. positive red regions are weaker and negative blue regions stronger in intensity). Therefore, the Met18_Emis17 run struggles to reproduce the absolute surface O_3 enhancements in the Met18_Emis18 run. When the fixed meteorology differences

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(FIXED_MET_DIFF, i.e. Met18_Emis18 - Met18_Emis17, **Figure 8 -** left column) are compared with CTL_DIFF, the surface 2018-2017 differences are substantially different.

Surface FIXED_MET_DIFF ranges between 0.0 ppbv and 2.0 ppbv across the domain in May and June and is more confined to continental Europe in July and August. This shows that TOMCAT simulates lower 2018 summer-time O_3 when 2017 emissions are used and indicates that emissions do have some role in controlling O_3 levels at the surface. However, as the spatial difference pattern for FIXED_MET_DIFF (**Figure** 8 – left column) is different to that of CTL_DIFF (**Figure 6** – left column), spatial correlations between them range from -0.53 to 0.54 over the four months, it suggests that meteorology is important in governing the spatial distribution of CTL_DIFF. This is supported by the fact that FIXED_MET_DIFF - CTL_DIFF (**Figure 8** left column – **Figure 6** left column) yields absolute domain variations between 0.0 ppbv and 12.2 ppbv. Therefore, the two sensitivity experiments suggest meteorology and emissions both play important roles in controlling surface O_3 during the summer of 2018, but meteorology predominantly governs the spatial pattern and absolute magnitude of the O_3 enhancements.

At 500 hPa, comparison of FIXED_EMIS_DIFF and CTL_DIFF show very consistent spatial patterns across the four months with correlations all above 0.98. In terms of the absolute differences between FIXED_EMIS_DIFF and CTL_DIFF (i.e. **Figure 7** centre column – **Figure 6** centre column) it peaks at approximately 2.8 ppbv. For FIXED_MET_DIFF, the spatial correlation with CTL_DIFF, as for the surface, is variable with values between -0.38 and 0.43. The absolute differences between FIXED_MET_DIFF and CTL_DIFF (i.e. **Figure 8** centre column – **Figure 6** centre column) ranges from 0.0 ppbv to 14.8 ppbv. Therefore, emissions have a secondary role in controlling the O_3 while meteorology is by far the dominant factor. For Strat % @ 500 hPa, the spatial correlations between CTL_DIFF and FIXED_EMIS_DIFF are above 0.95 for all months and the absolute differences between them (i.e. **Figure 7** right column - **Figure 6** right column) are near-zero. Comparison of FIXED_MET_DIFF and TC_CTL shows spatial difference correlations ranging between -0.33 and 0.71 and absolute differences (i.e. **Figure 8** right column - **Figure 6** right column) peaking at 12.9%. Therefore, as expected, meteorological processes are dominating the influence of the stratospheric O_3 contribution (i.e. through stratosphere-troposphere exchanges) to the 500 hPa layer during the summer 2018 O_3 enhancements over Europe.

To investigate the importance of stratospheric-troposphere exchange to the middle troposphere enhancement (i.e. as shown in the TOMCAT 500 hPa layer and the satellite SCO_3 data), **Figures 9** and **10** show TOMCAT control run zonal 2018-2017 difference cross-sections (for the domain longitudes) of O_3 profiles and the stratospheric O_3 contribution to each pressure layer. In May and June, in the lower troposphere (approximately surface to 800 hPa), there are negative (-3.0% to 0.0%) and positive (0.0% to 3.0%) differences between 30-50°N and 50-70°N, respectively. During June, there are positive differences (0.0% to 5.0%) across most latitudes and in August, the opposite occurs to that of May/June. In the mid-troposphere (800-300 hPa), positive differences occur in most months (0.0-5.0% in May, 0.0-7.0% in June, >10% in July and 5.0-10.0% in August), though in May and August negative differences (-5.0% to 0.0%) exist around 40°N and 55°N. This is consistent with the 500 hPa O_3 differences in **Figure 6** (centre panels). In the upper troposphere – lower stratosphere (UTLS, approximately 300-100 hPa) there are limbs of positive O_3 differences (i.e. >10%, 5.0-10.0 ppbv) propagating into the mid-troposphere (30-40°N in May, 30-50°N in June, 40-50°N in July and 30-40°N & 60-70°N in August), suggestive of stratospheric intrusion into the mid-troposphere. Using the stratospheric O_3 tracer in TOMCAT, **Figure 10** shows the enhanced proportion of O_3 originating from

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the stratosphere in the summer of 2018. Interestingly, for all months (apart from May between 30-45°N), there are enhanced contributions of stratospheric O_3 (15.0% to >50.0%) in the lower-mid troposphere (i.e. below 500 hPa). In absolute terms, this is only a minor contribution typically below 800 of <1.0 ppbv. Between 800-400 hPa, this increases to 1.0-5.0 ppbv (remains relatively consistent in percentage terms) in most months and latitude bands. In the UTLS, it increases to 5.0-10.0% enhancements in stratospheric O_3 contributions, which is consistent with its proximity to the stratosphere. In comparison between **Figures 9** and **10,** where there are enhancements in the stratospheric O_3 contribution but negative differences in O_3 (e.g. in June in the lower troposphere between 50°N and 55°N) which is suggestive of different processes influencing the O_3 concentrations (e.g. descent of relatively small stratospheric O_3 contributions but advection of tropospheric O_3 away from the region). Overall though, in the mid-troposphere, where there are larger enhancements in O_3, there are similar responses in the stratospheric O_3 contribution. For June, the mid-troposphere O_3 enhancement is approximately 5.0-7.0 ppbv with a signal of 1.0-2.0 ppbv in the stratospheric tracer. Therefore, in the more extreme cases, the stratospheric O_3 contribution is approximately 15.0-40.0% to the mid-tropospheric O_3 enhancements in summer 2018 over Europe.

The two remaining factors, linked to meteorological processes (as suggested above), which may affect the O_3 enhancements in 2018 are increased summer temperatures (e.g. through enhanced kinetic rates), and the import of tropospheric O_3 from upwind (e.g. North America from the prevailing winds). **Figure 11** shows the 2017-2018 zonal temperature differences (i.e. same as **Figure 9** but for temperature) with the correlation between the 2017 and 2018 temperature and O_3 differences overplotted. Qualitatively, the zonal differences in O_3 and temperature have some similarities. There are positive differences (temperature differences of 0.0-1.0%) between 50-60°N and the surface and 400 hPa in May and June. Then in July, collocated positive differences (peaking at 2.0% or 3.0 K) exist between 50-70°N from the surface to 300 hPa. In August, there is no clear relationship between temperature and O_3 enhancements. In all months (to a lesser extent in August), in the UTLS, there are spatial agreements with positive differences between approximately 30-45°N and negative differences between 50/55-70°N. In terms of correlations (i.e. temporal correlation in each grid box using the TOMCAT 6-hourly time series), the spatial agreement is relatively weak. In all months, most of domain has relatively small values ranging between -0.5 to 0.5. There are only a few locations with strong correlations (i.e. > 0.5), which are in the UTLS or in the lower-mid troposphere between 50-70°N (June & August) and 45-55°N in July near the surface. Overall, the relationship between increased temperatures and enhanced kinetic rates yielding more ozone formation is non-linear, so it is unsurprising that the direct comparisons of temperature and ozone 2018-2017 differences above shows no clear pattern. Therefore, future work could include a further sensitivity experiment running TOMCAT for 2018, but with 2017 temperatures used in the chemistry routines to quantify the role of temperature in the summer 2018 O_3 enhancements.

To investigate the potential advection of tropospheric O_3-rich air masses into Europe we have used ROTRAJ back-trajectories to determine the O_3WBTs (i.e. an indicator of air mass O_3 content). As shown in **SM 5**, there is large variability in the O_3WBT values and spatial distribution (i.e. **Figures SM 12** and **13**), so they have to be gridded onto the TOMCAT horizontal resolution (see **Figures SM14** and **15**). **Figure 12** shows the differences (2018-2017) between the gridded O_3WBTs where the back-trajectories have been released at the surface from Paris (**Figure 12a**), at the surface from Berlin (**Figure 12b**), at approximately 500 hPa from Paris (**Figure 12c**) and at approximately 500 hPa from Berlin (**Figure 12d**). We selected Paris and Berlin as they are situated in central Europe where the

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summer 2018 O_3 enhancements have been observed while the surface and 500 hPa are the altitudes of primary focus in the modelling work.

At the surface, Paris and Berlin show consistent patterns. Over the North Atlantic (i.e. origin of the prevailing winds into Europe), there are typically negative O_3WBT values between -5.0 ppbv and -1.0 ppbv suggesting that advection of O_3 into Europe during the summer (i.e. May-August) was predominantly larger in 2017 and did not strongly contribute to the 2018 observed surface O_3 enhancements. Advection of O_3-rich air in 2018 did originate from Scandinavia into continental Europe, though the number of trajectories is relative low (see **Figure S12**). As both locations show similar relationships, it provides confidence in this methodology. At 500 hPa, the 50-60°N spatial pattern is less defined with values typically between -5.0 and 5.0 ppbv for both locations. However, in the southern North Atlantic (30-50°N) there are positive differences of approximately 3.0-10.0 ppbv for both release locations. Note that as free-tropospheric winds tend to have larger horizontal velocities, the back-trajectories generally start from further away closer to North America. Again, given the broad similarity in differences between both release locations, it provides confidence in this approach. Overall, our results indicate a larger transport of O_3 to the surface of continental Europe in 2017, while at approximately 500 hPa the import of O_3 into Europe is larger in 2018. Here, the positive differences originate from the southern North Atlantic (i.e. a larger range of locations, absolute values and homogeneous signal than the mixed differences between 50-60°N).

One potentially important factor is dry deposition of O_3 to the land surface. Due to the heatwave, stress on the biosphere and the associated die back of vegetation could potentially reduce the efficiency of O_3 deposition decreasing the O_3 sink (i.e. O_3 is more likely to deposit onto land covered by vegetation than bare soil). Investigation of the normalised difference vegetation index (NDVI), from the IMS scheme, between the summers of 2017 and 2018 did not highlight any spatially coherent changes (not shown here). As a result, there is no obvious large-scale spatial vegetation die back in 2018 due to the heatwave and thus the impact this would have on ozone deposition in TOMCAT. Therefore, we ran two further experiments where the bare soil fraction for each grid box over Europe was increased and decreased by 25% in summer 2018. This was to investigate the sensitivity of surface ozone deposition to changes in the land surface. For the increase in bare soil fraction there was a moderate systematic increase in European summer ozone by 0.0-1.5 ppbv (i.e. less ozone deposition). When the bare soil fraction was decreased by 25%, this yielded a small decrease in surface ozone by approximately 0.5 ppbv. Overall, a sizable level of vegetation die back would be required for decreased ozone dry deposition to substantially contribute to the summer 2018 surface ozone enhancements.

4. Discussion and Conclusions

The summer of 2018 produced an intense heatwave across most of Europe with a substantial impact on tropospheric temperatures, droughts, stress on vegetation and human mortality. Observations of surface temperature, precursor gases and total column O_3 (TCO_3) experienced enhancements in 2018 relative to 2017. In this paper we have demonstrated a strong enhancement in surface and tropospheric O_3 during the heatwave between May and August 2018. The EMEP surface data suggest an average European enhancement, relative to 2017, peaking at approximately 10.0 ppbv in July and August. Investigation of lower tropospheric O_3 (i.e. surface-450 hPa sub-column O_3 – SCO_3) from the GOME-2 and IASI instruments also showed enhancements, peaking at 5.0-10.0 DU, relative to 2017. Analysis of the long-term GOME-2 SCO_3 record indicates 2017 to be a suitably

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neutral/average reference year and the enhancement in 2018 to be anomalously large. Our comparisons were therefore made between the summers of 2017 and 2018.

Consistency between the UV (GOME-2) and IR (IASI) sounders was important to our analysis because their vertical sensitivities peak in the lower and mid-upper troposphere, respectively. The similar patterns of SCO_3 enhancement detected by the two sounders therefore indicate that these extend over the bulk of the troposphere, supportive of surface/lower tropospheric ozone enhancements. This consistency also provides confidence that the complementary vertical sensitivities of GOME-2 and IASI ozone retrievals could be exploited in further investigation of tropospheric ozone (e.g. long-term trends from multiple platforms/retrieval schemes has shown large-scale inconsistencies in other studies e.g. Gaudel et al., (2018)) in the future.

Tropospheric O_3 behaviour is complex and the summer 2018 enhancements over Europe could potentially have been caused by various factors: atmospheric chemistry, meteorology (e.g. temperature, advection of O_3-rich air masses), anthropogenic and natural precursor emissions, dry deposition and stratospheric intrusion. To investigate the interactions between these processes, potentially leading to the summer 2018 O_3 enhancements, we used the well-evaluated TOMCAT 3D CTM. Evaluation of the model in this study showed that it could accurately capture the spatial pattern, temporal evolution and sign (i.e. positive 2018-2017 O_3 differences) of the O_3 enhancements and that, although it underestimated the observed enhancements, TOMCAT is an adequate tool to investigate them.

The results of several model simulations showed that the surface ozone enhancements (mainly in north-western Europe) in the summer of 2018 were predominantly driven by meteorological processes with emissions acting as a secondary factor. As the ROTRAJ back-trajectories suggest that advection of summer-time O_3 was larger in 2017, the 2018 European O_3 enhancements at surface level were likely from in-situ processes. The TOMCAT stratospheric O_3 tracer indicated a negligible contribution of stratospheric O_3 to these surface enhancements. At 500 hPa, the enhancement in tropospheric O_3 is much larger spatially across Europe and dominated by meteorological processes. Intrusion of stratospheric O_3 into the mid-troposphere has a moderate influence on the observed/modelled O_3 enhancements with contributions of up to 15.0-40.0%. Correlations between TOMCAT temperature and O_3 enhancements show broad agreement at some latitudes (e.g. 50-70°N in the lower-mid troposphere). However this relationship is non-linear and difficult to quantify without further simulations/model tracers, which was beyond the scope of this study. ROTRAJ back-trajectories suggest that in 2018, relative to 2017, there is the advection of more O_3-rich airmasses into the European mid-troposphere contributing to the summer 2018 O_3 enhancements at this altitude. Therefore, in the summer of 2018 over Europe, in-situ meteorological processes appear to be predominantly driving surface O_3 enhancements over Europe, while advection of tropospheric O_3-rich air and stratospheric intrusion are driving the corresponding tropospheric O_3.

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Conflicting Interests

The authors declare that they have no conflicts of interest.

Date Availability

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The TOMCAT simulations are publically available at http://homepages.see.leeds.ac.uk/~earrjpo/european_summer_2018_o3/tomcat, while the RAL Space satellite can be found at http://homepages.see.leeds.ac.uk/~earrjpo/european_summer_2018_o3/satellite. The EMEP surface O_3 data was obtained from http://ebas-data.nilu.no/default.aspx. The GOME-2 tropospheric column NO_2 data was downloaded from EUMETSAT at https://acsaf.org/nrt_access.php. The TOMCAT and RAL Space satellite data will be uploaded to the Zenodo open access portal (https://zenodo.org/) if this manuscript is accepted for publication in ACP after the peer-review process.

Author Contributions

RJP, MPC and BJK conceptualised and planned the research study. RJP performed the TOMCAT model simulations with support from MPC and WF. The JULES BVOC emissions were provided by ECP and GDH. RJP analysed the satellite data provided by RAL Space (BJK, RS, BGL and LJV) with support from BJK, RS and BGL. RJP undertook the EMEP analysis. RJP ran ROTRAJ with technical support from SRA and AMG. RJP prepared the manuscript with contributions from all co-authors.

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Figures:

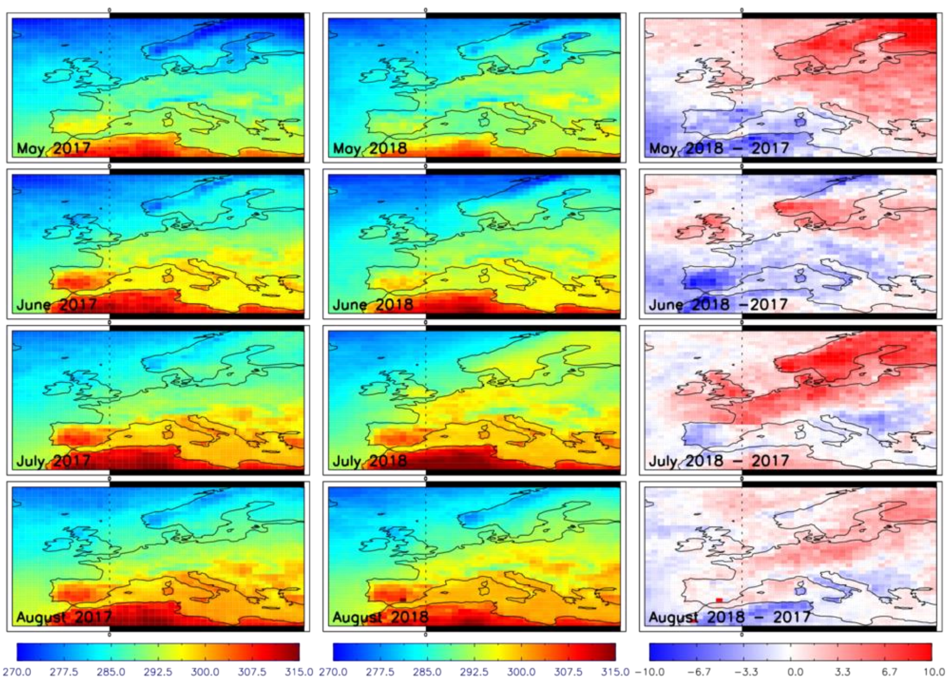

Figure 1: *Surface temperature (K) over Europe for May to August in 2017 (LHS), 2018 (middle) and 2018-2017 difference (RHS) retrieved from MetOp-A IASI, MHS and AMSU by the IMS scheme.*

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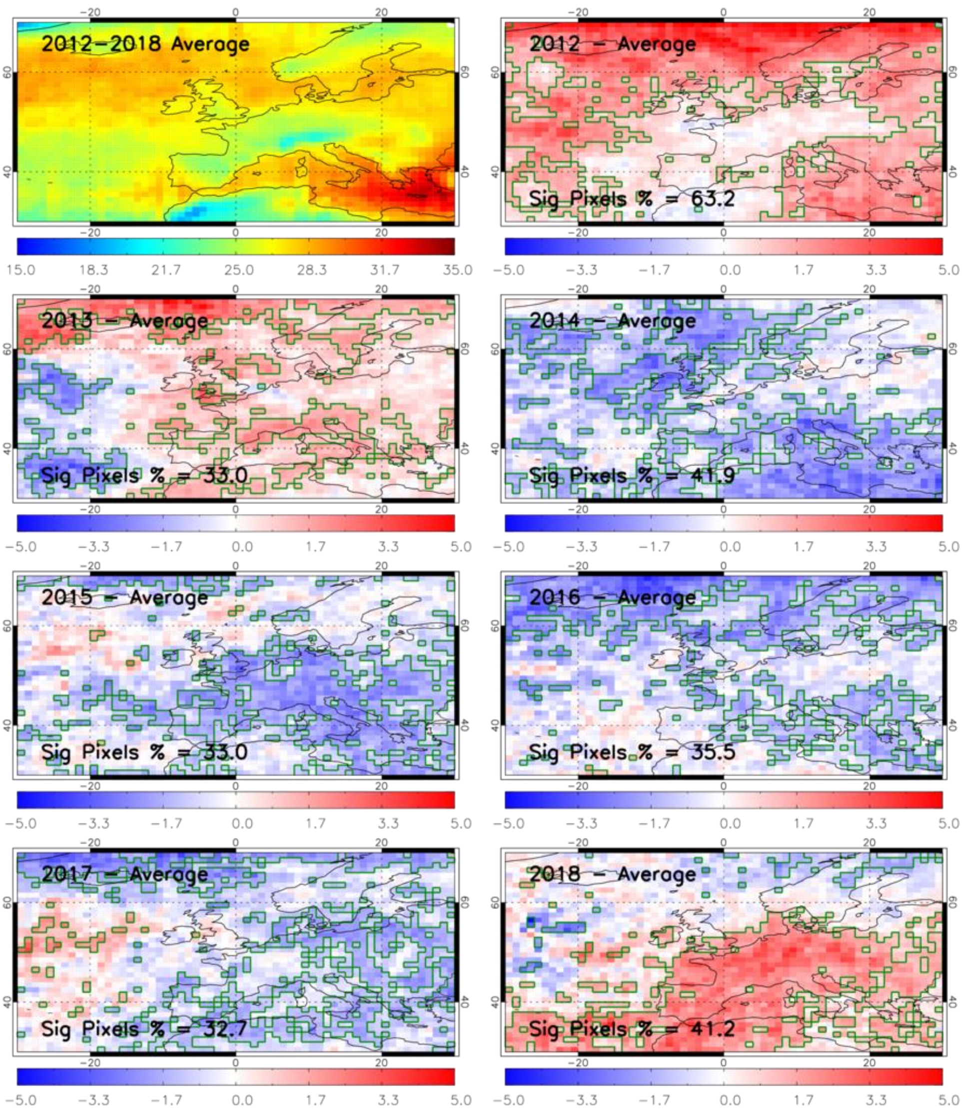

Figure 2: *Sub-column ozone (SCO_3, surface-450 hPa), in Dobson units (DU), retrieved from GOME-2 on Metop-A averaged across May to August between 2012 and 2018 (top left panel) and the corresponding difference from the 2012-18 mean for each year, respectively. The green-polygon-outlined regions show where the year-specific seasonal average is significantly different (95% confidence level based on the Wilcoxon Rank Test (WRT)) from the long-term (2012-2018) seasonal average. The "Sig Pixel %" label indicates the number of pixels in the domain with significant differences.*

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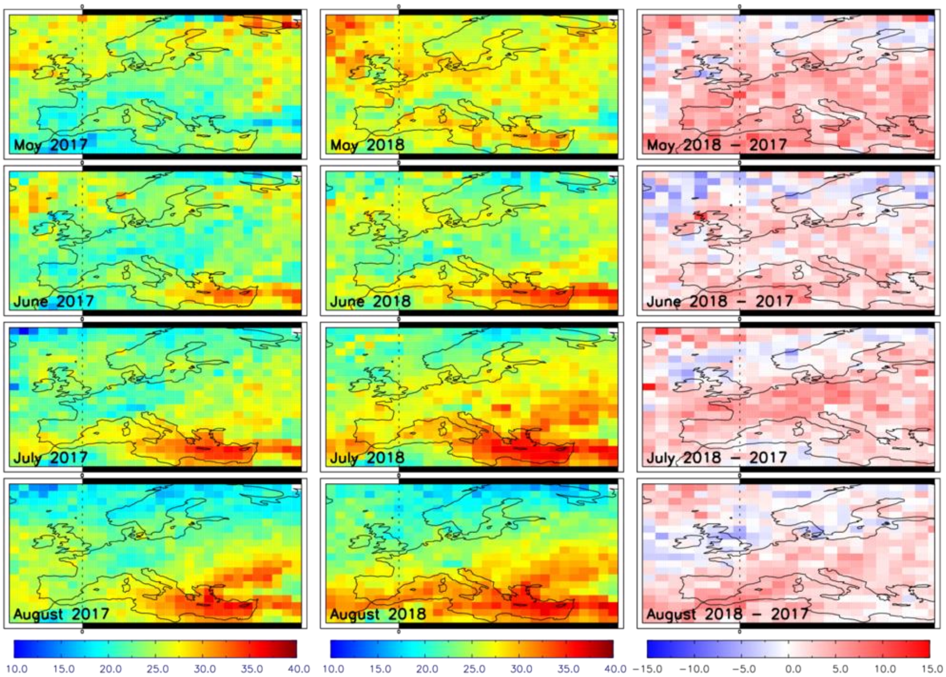

Figure 3: *SCO_3 (DU) from GOME-2 over Europe for May to August in (left column) 2017, (middle column) 2018 and (right column) 2018-2017 difference.*

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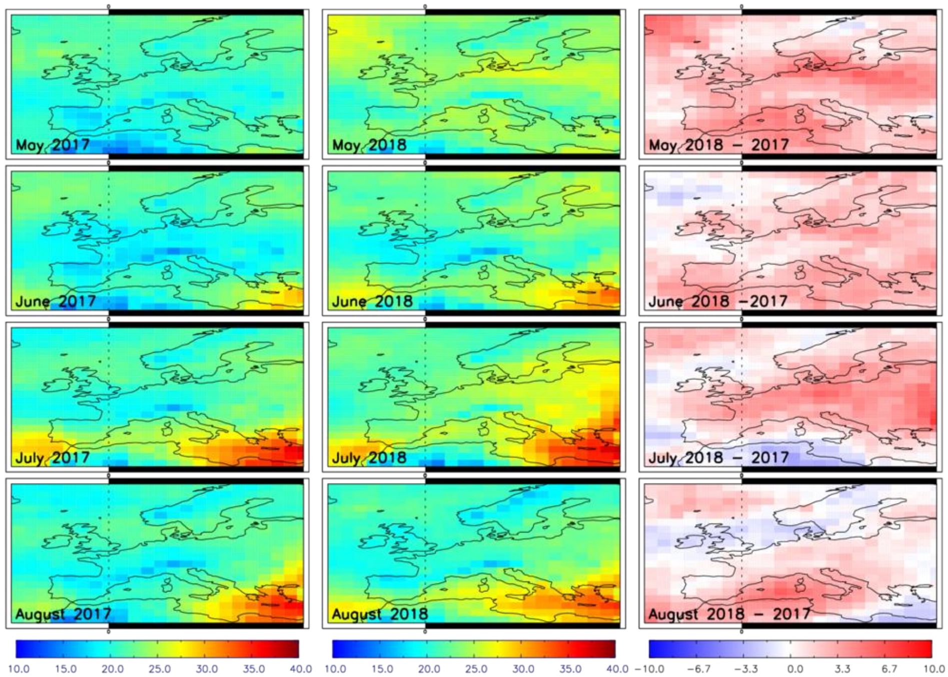

Figure 4: *SCO_3 (DU) for May to August in 2017 (LHS), 2018 (middle) and 2018-2017 difference (RHS) over Europe retrieved from MetOp-A IASI, MHS and AMSU by the IMS scheme.*

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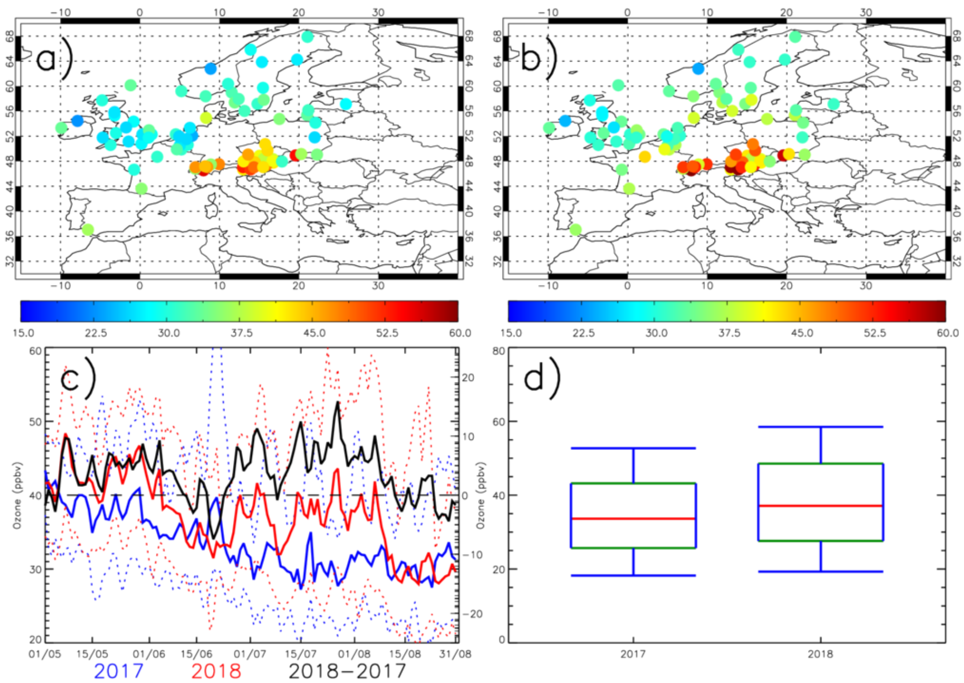

Figure 5: *European surface ozone (ppbv) for a) May-June-July-August (MJJA) 2017, b) MJJA 2018), c) regional time series for MJJA 2017 (blue), MJJA 2018 (red) and the 2018-2017 difference (black) and d) box-whisker plots for MJJA 2017 and 2018. In panel d) the median, 25th & 75th percentiles and 10th& 90th percentiles are shown by the red, green and blue lines, respectively.*

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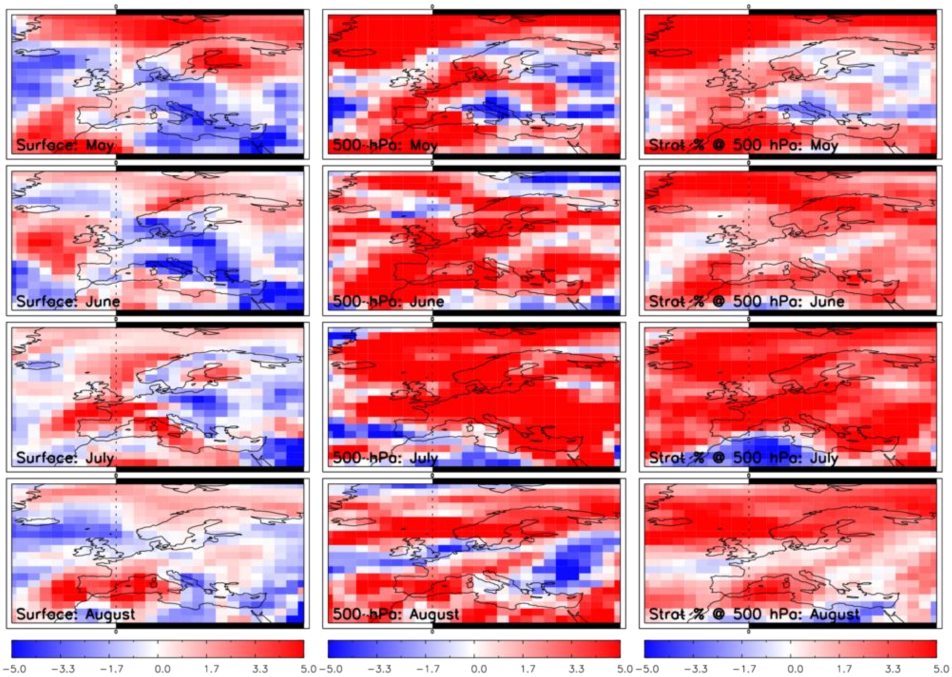

Figure 6: *TOMCAT ozone (ppbv) 2018-2017 differences for May to August for the surface (LHS), 500 hPa (middle) and the stratospheric contribution (%) to the 500 hPa layer (RHS).*

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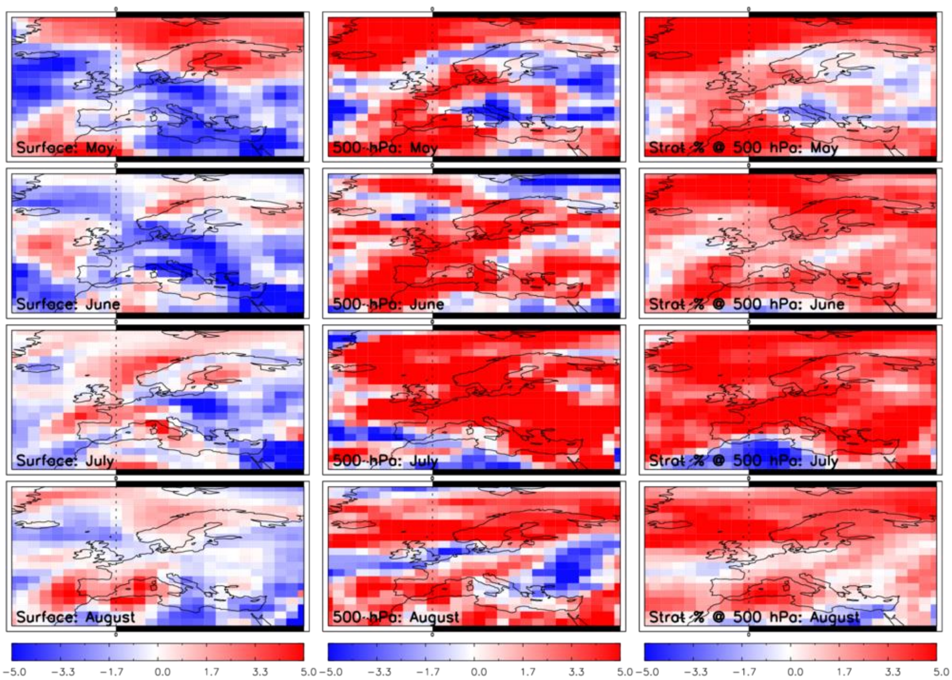

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669 **Figure 7:** *TOMCAT ozone (ppbv) 2018-2017 differences for May to August for the fixed emissions*
670 *simulation (Fixed_EMIS) for the surface (LHS), 500 hPa (middle) and the stratospheric contribution*
671 *(%) to the 500 hPa layer (RHS).*

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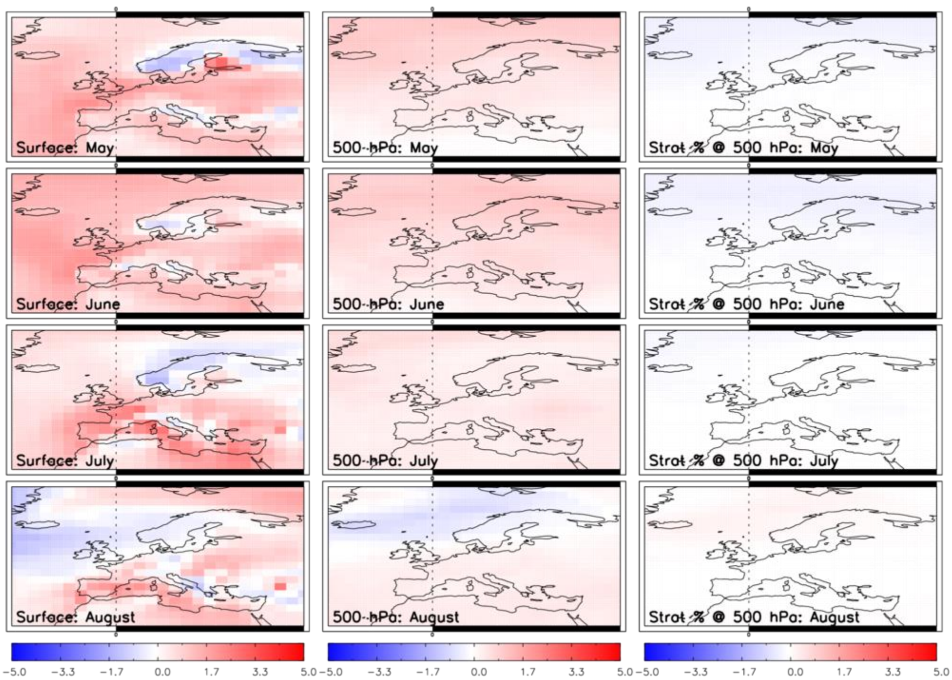

Figure 8: *TOMCAT ozone (ppbv) 2018-2017 differences for May to August for the fixed meteorology simulation (Fixed_MET) for the surface (LHS), 500 hPa (middle) and the stratospheric contribution (%) to the 500 hPa layer (RHS).*

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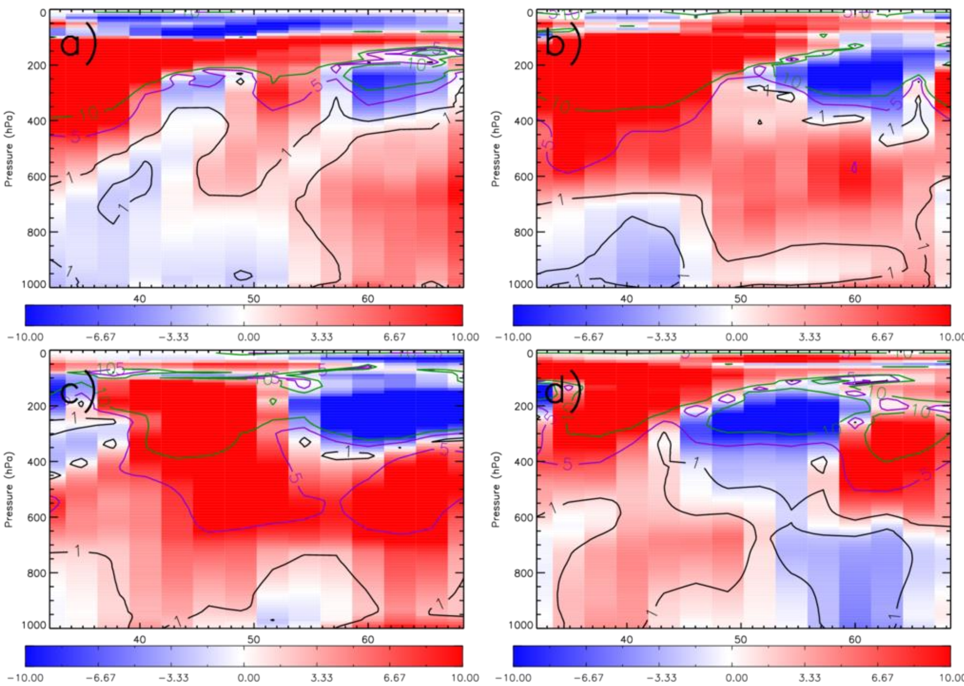

Figure 9: *TOMCAT ozone, zonally averaged between 20°W and 40°E, 2018-2017 percentage differences (absolute difference (ppbv) shown as solid lines) from the control simulation. Panels a)-d) represent the monthly averages for May, June, July and August.*

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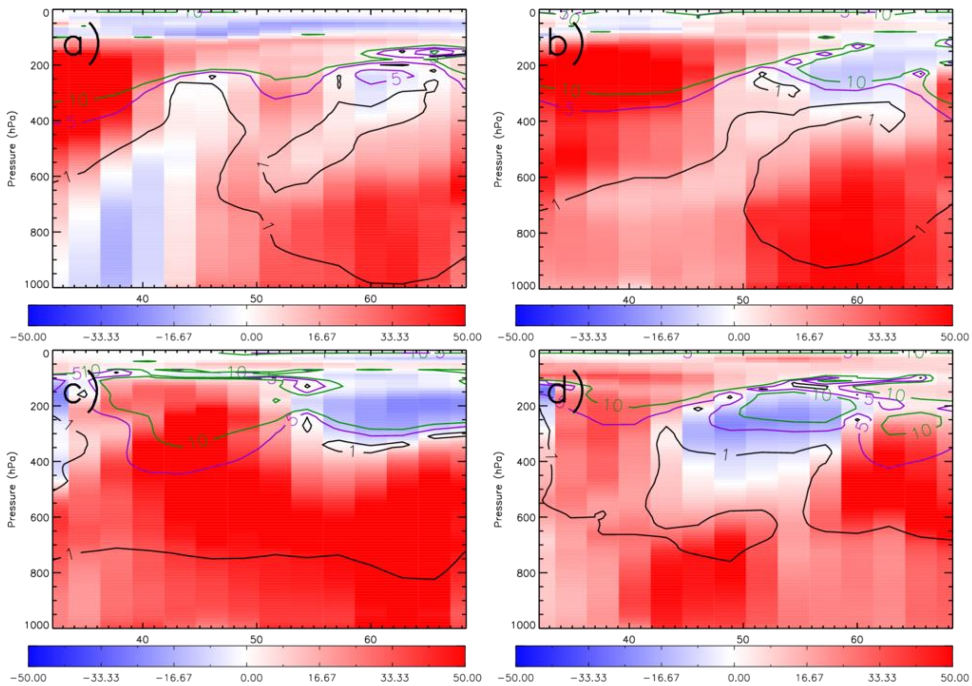

Figure 10: *TOMCAT stratospheric ozone tracer, zonally averaged between 20°W and 40°E, 2018-2017 percentage differences (absolute difference (ppbv) shown as solid lines) from the control simulation. Panels a)-d) represent the monthly averages for May, June, July and August.*

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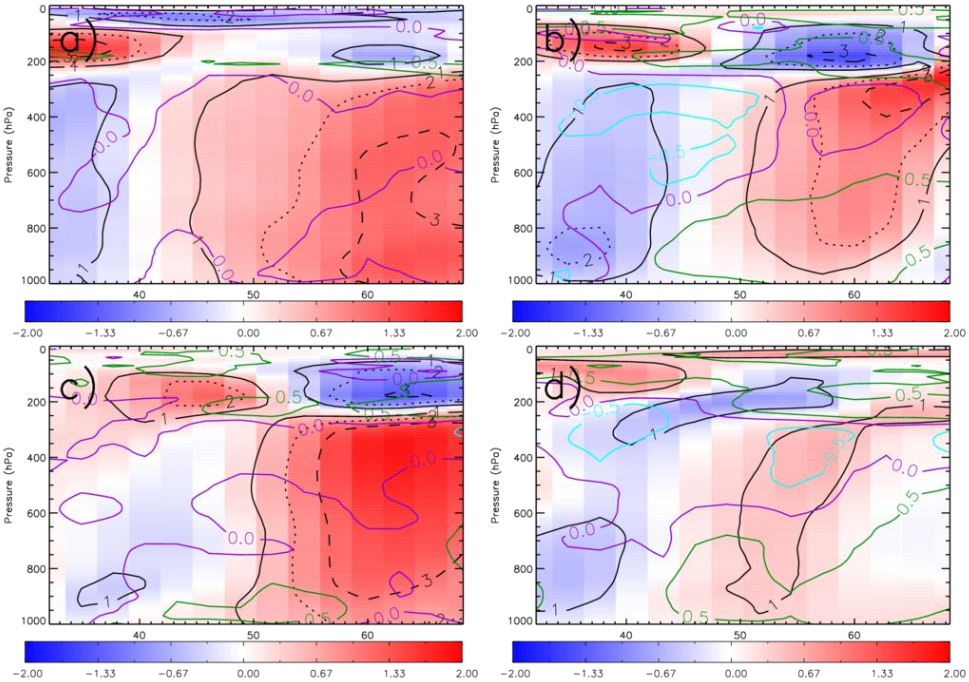

Figure 11: *TOMCAT temperature, zonally averaged between 20°W and 40°E, 2018-2017 percentage differences (absolute difference (K) shown by black solid, dotted and dashed lines) from the control simulation. Overplotted are contours of the temporal correlation (i.e. within each grid box) between the temperature and ozone 2018-2017 differences. Panels a)-d) represent the monthly averages for May, June, July and August.*

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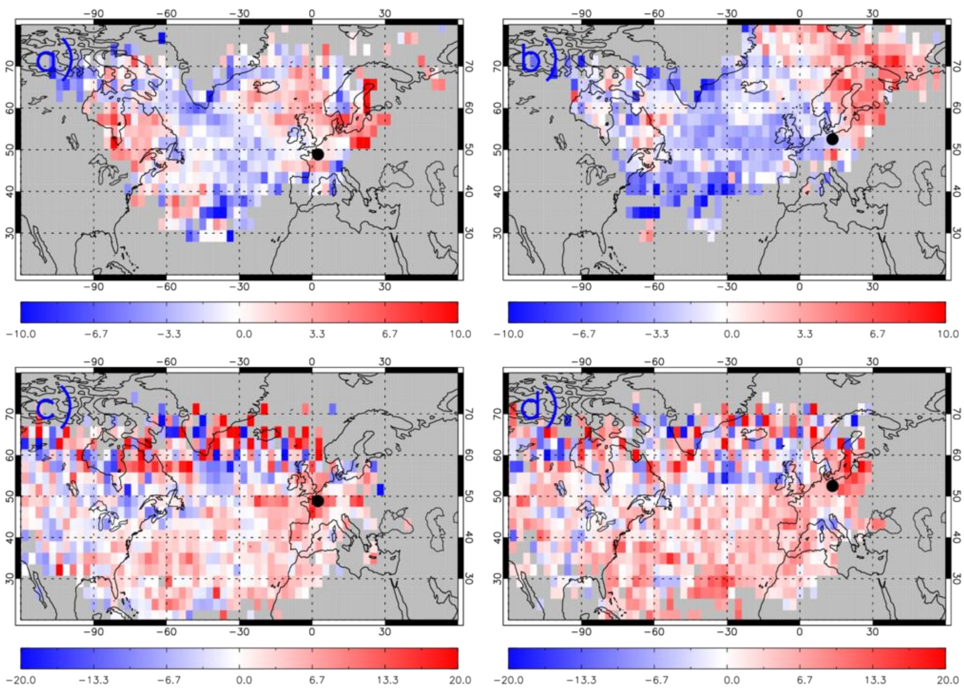

Figure 12: *The difference between May-August 2018 and May-August 2017 (i.e. 2018-2017) ROTRAJ back-trajectories (10 days), weighted by the average TOMCAT O_3 (ppbv) concentration along each trajectory path, gridded onto the TOMCAT horizontal resolution for a) Paris at the surface, b) Berlin at the surface, c) Paris at approximately 500 hPa and d) Berlin at approximately 500 hPa. The black circles represent the location of Paris or Berlin, where the trajectories where released from.*