Influence of synoptic and local meteorological conditions on surface ozone concentrations over Europe

Noelia Otero Felipe

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Abstract

Climate change is expected to alter global, regional and local meteorological conditions and as a result, the changes in the climate system will play an essential role on future air quality. Tropospheric ozone is considered one of the most harmful pollutants and it is strongly dependent on weather conditions. Therefore, understanding the impacts of near-term climate change on ozone concentrations is crucial for developing effective air quality policies.

This dissertation focuses on the analysis of the influence of synoptic and local meteorological conditions on ground-level ozone over Europe and it provides a comprehensive spatial characterization of the most important meteorological key-driving factors of surface ozone concentrations over the whole domain. For this purpose two approaches are proposed: i) a weather types classification and ii) regression methods.

Firstly, large-scale atmospheric circulation is examined through a weather types classification, implemented grid cell-by-grid cell over Europe. The ability of a suite of global climate models to reproduce realistic synoptic patterns in the present climate is evaluated against two reanalysis products. Additionally, the association between weather types and anomalies of maximum and minimum temperatures is investigated. In general, the models are able to capture realistic synoptic patterns when compared to the reanalyses. However, some limitations to reproduce the frequencies of certain weather types, such as low flow conditions over South Europe in summer and autumn are found. The projected changes in the frequency of weather types under future climate scenarios reveal an increase of anticyclonic days and warmer conditions affecting the British Isles in summer, and more westerlies and consequently mild winter conditions over Central Europe. As a result of a projected increase of low flow conditions over the Mediterranean basin, stagnant situations would become more frequent, favouring episodes of air pollution. Further analysis indicate that changes in the frequency of weather types represent a minor contribution of the total change of projected European temperatures. Thus, the temperature changes could be attributed to the so-called within-type variations (changes of the weather types themselves). In the context of climate change, that implies that global warming would also affect the characteristics of some weather types over time (i.e., within-type variations) that are associated with warmer temperatures under future conditions.

Secondly, the classification of weather types provides an easy physically interpretable framework for assessing the impacts of synoptic conditions on ozone concentrations. A synoptic-regression approach is developed to investigate the effect of both, synoptic and local meteorological conditions on surface ozone over the European domain. It is shown that local meteorological conditions are generally dominant factors influencing surface ozone variability, rather than the synoptic conditions. The results reveal distinctive regional and seasonal patterns of the most influential ozone drivers. In particular, local meteorological conditions have a strong influence over Central and East Europe, where maximum temperature becomes the most important driver of surface ozone in summer and relative humidity along with surface solar radiation in spring.

Finally, a multi-model assessment examines the capability of a set of state-of-the-art air quality models to reproduce the observed relationship between meteorological variables and surface ozone. The results show distinctive seasonal and regional performances in the statistical models developed for each dataset (i.e. observations and model outputs). Overall, the air quality models are in better agreement with observations over the regions referred to as internal regions: England, France, Mid-Europe, North Italy and East Europe. On the contrary, they present more limitations over the rest of the regions, referred to as the external regions: Inflow, Scandinavia, Iberian Peninsula, Mediterranean and the Balkans. There is a larger meteorological contribution in the internal regions, especially in summer where the local meteorology plays an important role in photochemical processes. A minor meteorological effect is found in the external regions, probably due to a major influence of the dynamical processes that are not captured by the statistical models. Most of the air quality models tend to overestimate the sensitivity to maximum temperature and solar radiation and none of them are able to capture the strength of the observed relationship between ozone and relative humidity appropriately. Here, dry deposition schemes may be a key for the underestimation of such relationship. Further analysis of the slopes of the ozone-temperature relationship indicates that the air quality models capture the observed relationship between ozone and temperature in most of the internal regions in summer, while in spring they overestimate it in most of the European regions.

Zusammenfassung

Da zu erwarten ist, dass der Klimawandel die globalen, regionalen und kommunalen meteorologischen Zustände verändern wird, werden die Veränderungen des Klimasystems eine wesentliche Rolle in Bezug auf die zukünftig Luftqualität spielen. Troposphärisches Ozon gilt als einer der schädlichsten Schadstoffe und ist stark abhängig von den Wetterbedingungen. Die zeitnahen Auswirkungen des Klimawandels zu verstehen ist daher dringend erforderlich, um eine effektive Luftqualitätspolitik zu entwickeln.

Diese Doktorarbeit legt den Schwerpunkt auf die Analyse des Einflusses von synoptischen und kommunalen meteorologischen Zuständen auf bodennahes Ozon in Europa und sie liefert eine umfassende räumliche Charakterisierung der wichtigsten Schlüsselfaktoren der Oberflächen-Ozon-Konzentration auf dem gesamten Gebiet. Zu diesem Zweck werden zwei Ansätze vorgeschlagen: i) eine objektive Wetterlagenklassifikation und ii) Regressionsmethoden.

Zunächst wird die großflächige atmosphärische Zirkulation durch eine objektive Wetterlagenklassifikation untersucht – umgesetzt in Form von Gitterzelle zu Gitterzelle in Europa. Es wird ein Vergleich zwischen der Fähigkeit mehrerer globaler Klimamodelle realistisch aussehende synoptische Muster im gegenwärtigen Klima zu reproduzieren einerseits und neuen Darlegungen andererseits aufgestellt und anschließend ausgewertet. Darüber hinaus wird der Zusammenhang zwischen Wetterarten und Anomalien von Maximal- und Mindesttemperaturen untersucht. Im Vergleich mit den neuen Darlegungen können die Modelle im Allgemeinen realistische synoptische Muster erfassen. Allerdings gibt es einige Einschränkungen in der Reproduktion der Frequenzen bestimmter Wetterarten, wie z. B. niedrige Strömungsbedingungen über Südeuropa im Sommer und Herbst. Die prognostizierten Veränderungen bezüglich der Häufigkeit der Wetterarten unter zukünftigen Klimaszenarien zeigen einen Anstieg antizyklonischer Tage und wärmeren Bedingungen, die die britischen Inseln im Sommer beeinflussen, sowie mehrere Westwindzonen, welche folglich milde Winterbedingungen über Mitteleuropa hervorbringen. Infolge einer prognostizierten Zunahme der niedrigen Strömungsbedingungen über dem Mittelmeerraum würden stagnierende Situationen häufiger vorkommen, was die Folgen der Luftverschmutzung

begünstigt. Eine Analyse des Abbaus zur Beurteilung der Auswirkungen der Frequenzänderungen auf die prognostizierten Temperaturen deutet darauf hin, dass Veränderungen in der Häufigkeit der Wetterarten einen geringen Beitrag zur Gesamtveränderung der europäischen Temperaturen darstellen. So könnten die Temperaturveränderungen den sogenannten In-Typ-Variationen (selbst Änderungen der Wetterarten) zugeschrieben werden. Im Kontext des Klimawandels bedeutet dies, dass die globale Erwärmung auch die Eigenschaften einiger Wetterarten im Laufe der Zeit beeinflussen würde (d.h. In-Typ-Variationen), die mit wärmeren Temperaturen unter zukünftigen Bedingungen verbunden sind.

Zweitens bietet die Einordnung von Wetterarten einen einfachen physikalisch interpretierbaren Rahmen, um die Auswirkungen von synoptischen Bedingungen auf die Ozonkonzentration zu bewerten. Ein Ansatz der synoptischen Regression wird entwickelt, um die Wirkung von sowohl synoptischen als auch kommunalen meteorologischen Bedingungen auf Oberflächen-Ozon auf europäischem Gebiet zu untersuchen. Es wird gezeigt, dass kommunale meteorologische Bedingungen in der Regel dominierende Faktoren sind, die die Oberflächen-Ozon-Variabilität beeinflussen, und nicht synoptische Bedingungen. Die Ergebnisse zeigen regionale und saisonale Muster der einflussreichsten Ozon Treiber. Die Ozon-Persistenz (vom Vortag) ist auch als Prädiktor enthalten und scheint eine wesentliche Rolle über Südeuropa zu spielen, wohingegen die kommunalen/regionalen meteorologischen Bedingungen einen starken Einfluss auf Mittel- und Osteuropa haben. Besonders die Maximaltemperatur und relative Luftfeuchtigkeit sind der wichtigste Treiber für Oberflächen-Ozon im Sommer zusammen mit Oberflächen-Sonnenstrahlung im Frühling.

Der letzte Teil der Doktorarbeit untersucht eine Multimodell-Bewertung der Fähigkeit einer Reihe von hochmodernen Modellen zur Luftqualität, um die beobachtete Beziehung zwischen meteorologischen Variablen und Oberflächen-Ozon zu reproduzieren. Die Ergebnisse zeigen deutliche saisonale und regionale Leistungen der statistischen Modellen, die für jeden Datensatz (d. H. Beobachtungen und Modellausgaben) entwickelt wurden. Insgesamt stehen die Luftqualitätsmodelle in größerer Übereinstimmung zu den Beobachtungen über die Regionen, welche als folgende interne Regionen bezeichnet werden: England, Frankreich, Mitteleuropa, Norditalien und Osteuropa. Dem gegenübergestellt sind Regionen, welche mehr Einschränkungen gegenüber den übrigen Regionen haben. Solche werden als äußere Regionen bezeichnet: Inflow, Skandinavien, die Iberische Halbinsel, das Mittelmeer und die Balkanstaaten. Es gibt einen größeren meteorologischen Beitrag in den internen Regionen, vor allem im Sommer, wo die lokale Meteorologie eine wichtige Rolle bei photochemischen Prozessen spielt. Eine kleinere meteorologische Wirkung findet sich in den äußeren Regionen, vermutlich aufgrund eines großen Einflusses der dynamischen Prozesse, die nicht durch die statistischen Modelle erfasst werden. Die meisten Luftqualitätsmodelle neigen dazu, die Empfindlichkeit gegen Maximaltemperatur und Sonneneinstrahlung zu überschätzen, und keines von ihnen kann die Stärke der beobachteten Wechselwirkung zwischen Ozon und relativer Feuchtigkeit passend erfassen. Hier könnten trockene Ablagerungsschemata ein Lösungsansatz für die Unterschätzung einer solchen Beziehung bieten. Eine weitere Analyse des Anstiegs der Beziehung zwischen Ozon und Temperatur deutet darauf hin, dass die Luftqualitätsmodelle die beobachtete Beziehung zwischen Ozon und Temperatur in den meisten internen Regionen im Sommer einfangen, während sie diese im Frühjahr sie in den meisten europäischen Regionen überschätzen.

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List of Acronyms

Roman Symbols

ACCMIP	Atmospheric Chemistry and Climate Model Intercomparison Project
AMJ	April-May-July
AQ	Air quality
CCMs	Chemistry Climate models
CH_4	methane
*CMIP*5	Coupled Model Intercomparison Project Phase 5
CO	Carbon monoxide
CO_2	Carbon dioxide
CTMs	Chemistry Transport Models
CWT	Circulation Weather Types
ECMWF	European Center for Medium-range Weather Forecasting
EDT	Eurodelta-Trends
EEA	European Environment Agency
EU	European Union
GCMs	Global Climate Models
GLMs	Generalized Linear Models
HO_x	Hydroxyl radicals
IPCC	Intergovernmental Panel on Climate Change
JAS	July-August-September

JC	Jenkinson and Collison
JJA	June-July-August
LWTs	Lamb Weather Types
MAM	March-April-May
$MDA8O_3$	Maximum daily 8-hour average ozone
MLR	Multiple Linear Regression
MME	Multimodel ensemble
NO_2	Nitrogen Dioxide
NO_x	Oxides of Nitrogen
$O(^1)D$	excited oxygen atom
O_3	Ozone
OH	Hydroxyl Radicals
PAN	Peroxyacyl nitrates
PM	Particulate Matter
PM_{10}	Particulate Matter of size 10 micron
QR	Quantile Regression
$RCP8.5$	Representative Concentration Pathways (8.5)
STE	Strato-Tropospheric Exchange
$TF-HTAP$	The Task Force on Hemispheric Transport of Air Pollution
VOCs	Volatile Organic Compounds
WHO	World Health Organisation
WT	Weather types

Chapter 1

Introduction

Air pollution is considered a critical environmental issue resulting from a combination of elevated emissions and unfavourable weather conditions that allow the accumulation of pollutants in the near-surface atmosphere (Jacob and Winner, 2009). Since the early 1970s, air pollution has been one of the main political concerns of the European Union (EU). During the 1990s a series of directives on air quality management have been adopted to set air quality limit and target values (EEA, 2010). Despite the improvements and the continued efforts for achieving international air quality standards, air pollution is considered the single largest environmental health risk in Europe (EEA, 2016). Poor air quality has also negative impacts in ecosystems, the built environment and climate. All of these effects of air pollution have considerable market costs, such as reduced labour productivity, additional health care, crop and forest yield losses, and non-market costs, such as premature mortality or degradation of air and water quality (EEA, 2016). Moreover, climate change can affect air quality in several ways, including changes in ventilation rates, chemical production and loss rates, natural emissions, and background concentrations (Jacob and Winner, 2009). Thus, air pollution and climate change represent a global concern that must be considered joinly to identify the co-benefits of reducing emissions in order to mitigate the impacts of climate change.

At the present, particulate matter (PM) and tropospheric or ground-level ozone (O_3) are two of the most problematic pollutants in Europe (EEA, 2016). Epidemiological studies have shown that both PM and tropospheric O_3 have significant impacts on human health, including premature mortality (Bell et al., 2006; Silva et al., 2013). PM is a complex mixture of extremely small particles and liquid droplets with a broad compositional range, and may have primary and/or secondary sources (HTAP, 2010). PM poses a great risk as it penetrates into sensitive regions of the respiratory system and can lead to serious health problems and premature mortality (WHO, 2013). Tropospheric ozone has also shown considerable negative health effects that may lead to premature mortality (Brauer et al., 2012; Silva et al., 2013) and is one of the most harmful pollutants in terms of damage to ecosystems (WHO, 2005). In the stratosphere ozone absorbs ultraviolet (UV) radiation and it is essential for protecting us from its harmful effect, while in the troposphere it is a damaging pollutant. The concentrations of ozone in the atmosphere depend on the amount of its precursor sources but also are strongly influenced by transport and meteorological conditions (Monks et al., 2015).

This dissertation specifically focuses on tropospheric ozone, which has been recognized as the third most important pollutant in terms of health damage and the first most harmful pollutant to ecosystems in Europe (EEA, 2016). Thus, this chapter provides a summary of tropospheric ozone, outlining its formation and the main impacts. In particular, the strong influence of climate on ozone pollution and the implications under future climate conditions are discussed.

1.1 Ozone

The presence of ozone in the troposphere is partly due to stratospheric-tropospheric exchange (STE) or it can be arise from photochemical reactions within the troposphere. Initially, the stratosphere was thought to be the primary source of tropospheric ozone: about 90% of atmospheric ozone is present in the stratosphere, while the remaining ozone, about 10%, is present in the troposphere. However, early studies pointed out that the global tropospheric ozone budget is largely controlled by photochemical production and loss within the troposphere (Chameides and Walker, 1973; Crutzen, 1974). In particular, at northern mid-latitudes tropospheric ozone has increased since 1950, and it has been attributed to increasing anthropogenic precursor emissions (Parrish et al., 2012). Despite current chemical transport and climate global models varying quantitatively in the magnitude, several modelling studies (Lamarque et al., 2005; Young et al., 2013) suggest that 30% of the present-day tropospheric ozone burden is attributable to human activity.

In the troposphere, ozone is produced by complex non-linear chemical reactions involving carbon monoxide (CO), methane (CH_4), nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of sunlight (Seinfeld and Pandis, 2006). Its precursors originate from both natural (e.g. wildfires, biogenic hydrocarbon emissions, lightning NOx, volcanic activity) and anthropogenic sources (e.g. fossil fuel and biofuel combustion, crop burning). The non-linear ozone chemistry implies that decreases in precursor emissions do not necessarily cause decreases in ozone levels. In particular, understanding the sensitivity of ozone production to two main precursors, i.e. NO_x and VOCs, is a major chanllenge especially in urban areas, where ozone production can be either NO_x-sensitive or VOC-sensitive. In the NO_x-sensitive regime, ozone increases with increasing NO_x while increasing VOC levels has little effect on ozone. In the VOC-sensitive regime, ozone increases with increasing VOC and decreases with increasing NO_x (Sillman, 1999). The complex relationship between ozone, NO_x and VOC is exacerbated by the influence of meteorological conditions. Thus, considering the impacts of meteorological conditions on ozone is also essential to develop an effective policy response under a changing climate.

Two loss processes close the ozone budget: dry deposition to the Earth's surface, which accounts for about 25% of the total O_3 removal from the troposphere (Lelieveld and Dentener, 2000), and chemical reactions (Crutzen, 1974). Chemical production and loss rates are several times larger than the influx from the stratosphere and the surface deposition flux (Stevenson et al., 2006). The atmospheric lifetime of ozone usually depends on season and altitude and it ranges from several days in the boundary layer to weeks in the free atmosphere (Jacob and Winner, 2009; Young et al., 2013). In the lower troposphere its lifetime is shorter in summer (around 5 days) due to higher water vapour concentrations (Monks et al., 2015). The relatively long lifetime of ozone allows its transport over intercontinental scales. Thus, tropospheric ozone is not only a pollutant on a regional scale, but also it is considered a global pollutant that can influence air quality in remote areas (HTAP, 2010). As a hemispheric pollutant, reductions in local or national emissions of ozone precursors do not always lead to corresponding local decrease of pollution levels.

The concentrations of tropospheric ozone vary seasonally and the strongest seasonal variations occur at northern mid-latitudes where the ozone burden is at a minimum in October and November and reaching a maximum in spring/summer (depending on the location) (Cooper et al., 2014; Monks, 2000). The seasonal variation could be partly explained by STE, which leads to a peak flux in May and a minimum in November in mid-latitudes (Hsu and Prather, 2009). Nevertheless, the response to STE is not uniform with altitude or latitude (Monks et al., 2015). Moreover, populated continental areas at northern latitudes show a summertime peak of ozone levels, which can be attributed to regional photochemical production. In remote areas the maximum of ozone is generally found in spring, mainly due to STE and the photochemical production (Monks, 2000). Furthermore, some studies have suggested a shift in the seasonal cycle at northern mid-latitudes over the last few decades as a consequence of changing anthropogenic emissions, natural variability or a changing climate. Parrish et al. (2013) found a more pronounced and earlier springtime maximum over northern mid-latitudes and they argued that it could be explained by changes in atmospheric patterns along with spatial and temporal changes in emissions. Cooper et al. (2014) also showed changes in the seasonal cycle, but in this case they only found a seasonal change in some sites. Therefore, the shift in the seasonality of tropospheric ozone at northern mid-latitudes appears not to be universal.

1.2 Impacts of ozone

Tropospheric ozone has multiple and adverse impacts, not only on human health, but also on ecosystems and climate. Being a strong oxidant and phytotoxic agent, tropospheric ozone can cause respiratory problems (Bell et al., 2006), and it damages agricultural crops and forest vegetation (Ashmore, 2005). Tropospheric ozone is considered an important greenhouse gas (IPCC, 2013). In this section the main impacts of tropospheric ozone are discussed. For the rest of thesis, tropospheric ozone (surface or ground-level ozone) will be referred to as ozone.

1.2.1 Human health and ecosystems

Tropospheric ozone is a crucial public health issue. There is evidence in support of an association between ozone levels with adverse cardiovascular and respiratory diseases (Huang et al., 2005). Exposures to ozone have been associated to short-term premature mortality (Bell et al., 2004), to increase the likelihood of wheeze, chest tightness and asthma (Mortimer et al., 2002). Several studies assessed the numbers of hospital admissions for respiratory and chronic obstructive pulmonary diseases to ambient ozone levels (Anderson et al., 1996; Burnett et al., 1997). Lung function problems have also been reported by several studies (Anderson et al., 2001; Peters et al., 1997). Long-term ambient ozone exposures may also contribute to risk of respiratory and circulatory mortality (Jerrett et al., 2009; Turner et al., 2016).

The European Union's Air Quality Directive (EU, 2008) sets four standards to reduce air pollution by ozone and its impacts on human health, (i) an information threshold defined as 1-hour average ozone concentration of $180\,\mu g/m^3$, (ii) an alert threshold defined as 1-hour average ozone concentration of $240\,\mu g/m^3$, (iii) a long-term objective defined as the maximum daily 8-hour mean concentration of ozone should not exceed $120\,\mu g/m^3$ and (iv) a target value defined as the long-term objective that should not be exceeded on more than 25 days per year, averaged over 3 years. The recommended target value by the WHO air quality guidelines is $100\,\mu g/m^3$ (daily maximum 8-hour mean)(WHO, 2005).

The target value under EU law has to be attained as far as possible by the attainment date and compliance is checked, but not legally binding. According to the recent report of the European Environment Agency (EEA, 2016) in 2014, 16 countries of the European Members States (EU-28) registered concentrations above the ozone target value more than 25 times (Fig. 1.1). Therefore, despite the improvements in the air quality legislation to control emission of ozone's precursors, a large part of the European population is still exposed to high levels of ozone that exceed the European Union (EU) standards and the World Health Organisation air quality guidelines (WHO AQG) for health protection. Moreover, some studies have shown the adverse effects to surface ozone exposure even in low concentrations (Bell et al., 2006).

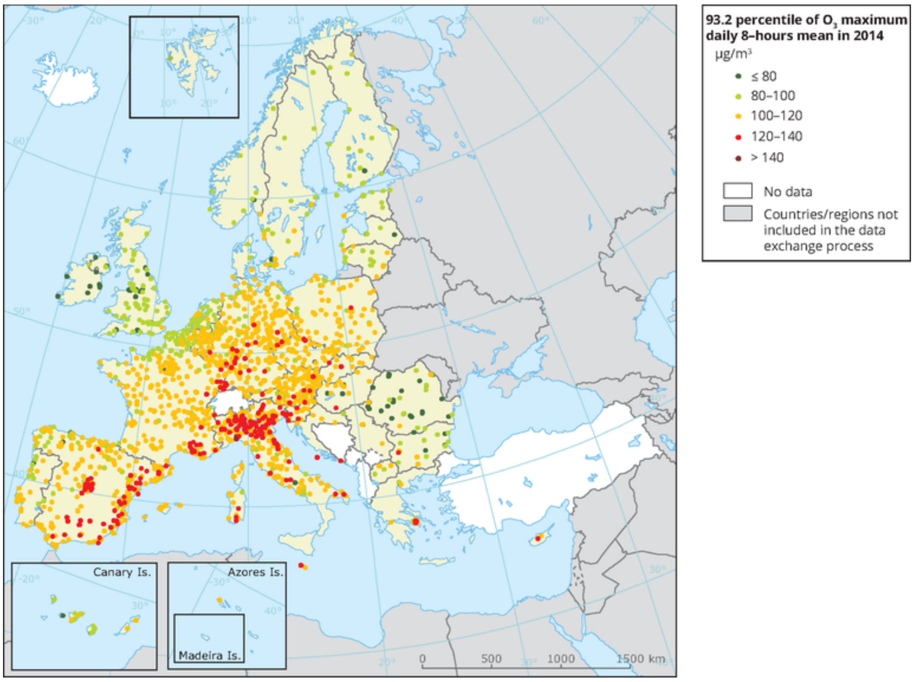

Figure 1.1 Observed concentrations of O3 in 2014. Only stations with more than 75% of valid data have been included in the map. Source:EEA, 2016.

In addition, ozone is a powerful and aggressive oxidant that has adverse effects on ecosystems. The impacts of ozone on vegetation can be in response to short-term episodes or cumulative during the growing season (LRTAP, 2010). Some of the ozone effects on plants include reduced growth, less seed production, lower functional leaf area and earlier leaf senescence (Monks et al., 2015). Previous studies suggest that ozone can exacerbate the effects of extreme weather events reducing the sensitivity of plans to drought (Wagg et al., 2012). At ground level, ozone damages agricultural crops, forests and plants by reducing their growth rates leading to substantial costs of crop yield loss (EEA, 2015). Field experiments have shown that exposure of crops to ozone to result in yield reduction and deterioration of crop quality (Fuhrer, 2009). Several studies pointed out that many species of plants are sensitive to high ozone levels, including agricultural crops such as wheat, tomato, soybean and rice, and salad crops (Mills et al., 2007). Crop sensitivities to ozone exposure are also influenced by meteorological factors, such as humidity, temperature, soil moisture and radiation. Several indices have been proposed to assess the impacts of ozone on vegetation (LRTAP, 2010; Mills et al., 2011), either based on the concentration-based critical level (ozone accumulated over a threshold of X ppb, AOT_{X}) and the uptake-based critical level (accumulated ozone dose above a threshold of Y or phytotoxic ozone dose, POD_{Y})(Mills et al., 2011).

1.2.2 Climate impacts of ozone

Tropospheric ozone interacts with both solar (shortwave) and terrestrial (longwave) radiation. Changes in the atmospheric distribution of ozone contribute to the radiative forcing of climate change (e.g. Fink et al., 2007; Lacis et al., 2015). It belongs to the category of the so-called short-lived climate pollutants (Shindell et al., 2012) due to its relatively short atmospheric lifetime compared to long-lasting greenhouse gases, such as CO_2 (Radjavi and Rosenthal, 2007; Stevenson et al., 2013). In the last IPCC report it was shown that changes in tropospheric ozone between 1750 and 2010 had led to a global mean radiative forcing of +40 Wm^{-2} (Myhre et al., 2013). Unlike the well-mixed greenhouse gases, ozone is distributed inhomogeneously and the radiative forcing from ozone, and especially its variation with time, results from a complex interplay between emissions, chemistry and transport. Ozone precursors can also affect the abundance of atmospheric OH and consequently alter the lifetimes of other greenhouse gases, such as CH_4 (Monks et al., 2015).

Several modelling studies have suggested that increased tropospheric ozone levels related to industrialization in developing economies have contributed to the accelerated warming. Shindell et al. (2006) suggested that changes in tropospheric ozone may have contributed to the spatial pattern warming over the 20th-century, mostly at high latitudes in winter and spring, and over polluted areas areas in summer. They pointed out that the increasing tropospheric ozone levels in low latitudes may have an impact on the warming detected in the tropics.

1.3 Climate influence on ozone

Tropospheric climate-chemistry interactions involve a large number of chemical processes and compounds that have shown an inhomogeneous distribution and trends, which add more complexity to a better understanding the influence of climate change on future air quality (Isaksen et al., 2009). There are a variety of atmospheric pathways in which climate change can influence regional ozone pollution levels, including changes in the ozone precursors, changes in dynamical and photochemical processes and changes in the tropospheric background, through processes such as stratosphere-troposphere exchange (Colette et al., 2015).

Future climate can alter the contribution of long-range transport and local meteorological conditions affecting ozone concentrations (Dawson et al., 2007; Doherty et al., 2013). Regional ozone pollution is expected to increase under warmer temperatures and weaker circulation (Denman et al., 2007). Climate change projections have shown to lead to an increase of up to 8 μg/m^3 in ozone concentrations over Europe by the end of the century (2071-2100) (EEA, 2016). In particular, summertime average ozone increases by 6 μg/m^3 over most of the European countries, and a similar increase for ozone peaks is found over polluted areas by the end of the century (Fig. 1.2, EEA 2016). In this section, the main impacts of long-range transport and local meteorological conditions on ozone will be discussed.

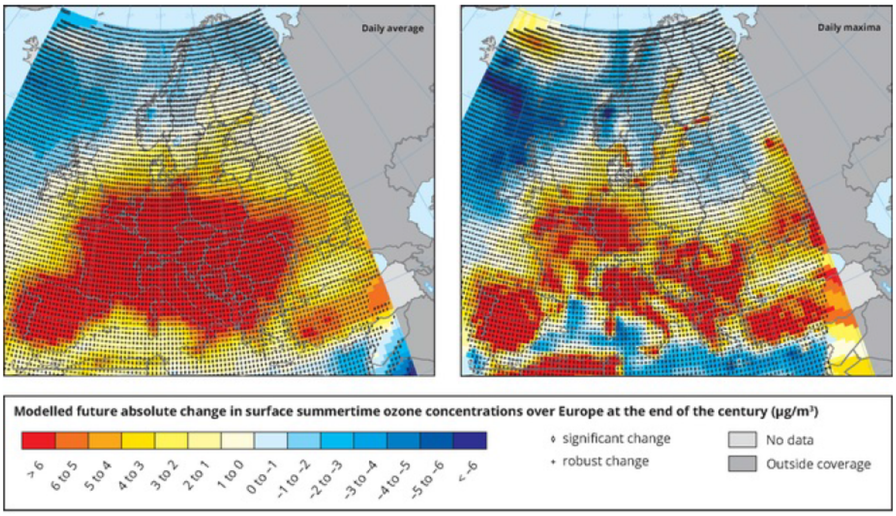

Figure 1.2 Absolute difference between future (2071-2100) and present (1960-2010) summertime average daily and maxima ozone levels in a 3 model ensemble. Significant changes are represented by a diamond sign. Source:EEA, 2016.

The effect of climate change on ozone has been termed as "climate penalty" (Wu et al., 2008). The magnitude of the climate penalty has been addressed in two main approaches. Some authors investigated the effect of climate on surface ozone through statistical analyses. For instance, Bloomer et al. (2009) defined the ozone climate penalty factor as the slope of the ozone-temperature relationship. They found that the climate penalty decreased with the reduction of the emissions of the ozone precursors. Similarly, Rasmussen et al. (2013) used the direct increase in ozone concentrations due to increasing temperatures (ppbK^{-1}) to assess the ozone climate penalty in U.S. Varotsos et al. (2013) developed a statistical model to examine the potential impact of increasing temperatures on ozone exceedances over Europe. They found that statistically significant increases of ozone exceedances could be explained by the increases in the upper temperature percentiles. Their results showed the highest ozone exceedance increases over South Europe, a moderate increase in the central regions and the lowest increase over Northwest and Northeast Europe. Temperature can influence surface ozone directly, through the temperature dependence of chemical reactions (Sillman and Samson, 1995) or indirectly, through the temperature dependence of dry deposition and biogenic emissions of ozone precursors (e.g Andersson and Engardt, 1996;

Solberg et al., 2008). This relationship will be discussed in more detail in the subsection 1.3.2.

Chemistry-transport and chemistry-climate models (CTMs and CCMs, respectively) are common tools for assessing future air quality. With a global chemical transport model, Wu et al. (2008) investigated the additional decreases in NO_x emissions to counter potential ozone increase due to climate change over U.S. They also assessed the reduced benefits of ozone precursors emissions controls under a warmer climate. Colette et al. (2015) presented an exhaustive analysis of the robustness of the climate penalty in Europe across time periods and scenarios, based on previous studies of regional and global chemical-transport models. They showed a penalty for summer surface ozone concentrations of at least 5 ppbv by the end of the century. While at global scale most of the studies showed a decrease in global background ozone due to an enhanced loss rates as a result of a projected increased absolute humidity (Doherty et al., 2013; Racherla and Adams, 2006), at regional scale a warmer climate can lead to increasing ozone concentrations under urban and pollutant regions (Nolte et al., 2008; Steiner et al., 2006). In that case, different model studies have quantified the effects of climate change on surface ozone concentrations over Europe (e.g. Andersson and Engardt, 1996; Colette et al., 2013; Meleux et al., 2007).

1.3.1 Long-range transport

Large-scale atmospheric circulation is an important factor that influences the distribution of ozone pollution and its precursors, especially because it modulates local meteorological controls on photochemical production and the build-up of regional ozone levels (Hegarty et al., 2007). Therefore, pollutants can be exported from one emission source region to another receptor region far downwind on the regional, intercontinental and even hemispheric scale (HTAP, 2010). The transport of atmospheric pollution is a serious problem since it can offset the impact of regional mitigation strategies. Climate change can modify the transport pathways since it will alter synoptic and convective transport, affecting the export and import of pollution (Doherty et al., 2013).

Global models are often used in different ways to investigate the impacts of long-range transport on air pollution and their implications under future climate conditions. In addition, global models provide boundary conditions to regional models generally with finer-grid resolution over the study region (Jacob and Winner, 2009). Previous modelling studies have investigated the impacts of climate change on long-range transport of ozone pollution. For instance, Leibensperger et al. (2008) showed that the frequency of summertime mid-latitudes cyclones is a strong predictor of stagnation and ozone pollution episodes in the eastern of US. Similarly, other studies reported an increase of ozone pollution episodes due to decreased frequency of mid-latitude cyclones (Mickley et al., 2004; Wu et al., 2008). Doherty et al. (2013) examined the influence of climate change on surface ozone and precursors by using three couple climate-chemistry models. They quantified the effects on intercontinental transport from major emissions regions to downwind receptor locations. In this case, they found stronger climate sensitivity to ozone chemistry than changes through the transport. They suggested that changes in transport might be more dominant when considering peaks of ozone.

Atmospheric patterns

Classification of atmospheric circulation patterns are useful tools to better understand the influence of atmospheric circulation on air quality, particularly on surface ozone levels. Moreover, large-scale synoptic conditions govern ozone advection and can also promote favourable conditions for ozone pollution episodes, such as high temperatures, low winds or no precipitation (i.e. stagnant conditions) (Horton et al., 2014). Changes in the frequency of synoptic patterns have been also associated with episodes of weather extremes that can occur under specific synoptic conditions (IPCC, AR5). For instance, the extreme temperatures and the lack of precipitation during the summer of 2003 in Europe have been related to the persistent anticyclonic conditions over central Europe (Fink et al., 2004). This particular episode led to exceptionally long-lasting and spatially extensive periods of high levels of ozone pollution over Europe (Fiala et al., 2003). Dole et al. (2011) suggested that the persistent blocking of westerly flow was essential during the 2010 heat weave in Russia that killed tens of thousands of people. Extreme events like the above described are likely more frequent under future climate change (Russo et al., 2015; Vautard et al., 2013).

A large number of observational studies have investigated the links between synoptic patterns with surface ozone levels. Comrie (1992), through a manual classification of synoptic types over Pennsylvania (U.S), found that high ozone concentrations occur with slow-moving anticyclones in summer. Hegarty et al. (2007) identified the most common circulation patterns and they found strong links between stagnant warm conditions and high ozone levels across northeastern U.S. In Europe, Davies et al. (1992) described marked associations between regional scale weather types and surface ozone concentrations, specifically between westerly cyclonic and high pressure types with ozone pollution levels, in winter and summer (respectively). Other representative studies found a consistent relationship between surface ozone levels and certain synoptic patterns in different regions across Europe (e.g. Demuzere et al., 2009, 2011; Saavedra et al., 2012; Tang et al., 2009). Most of these previous analyses have addressed the connection of ozone pollution levels with circulation patterns for specific regions, presenting a restrictive synoptic classification for the area of study (e.g. Central Europe, Scandinavia, Iberian Peninsula). This dissertation aims to contribute with a novel extended version of a weather types classification over the whole European domain (see Chapter 2) to investigate the impacts of atmospheric circulation on ozone pollution.

1.3.2 Meteorological conditions

Meteorology plays an essential role in ozone formation and transport. Variations in the local meteorological conditions can contribute to ozone increases. The most favourable conditions generally occur under slow moving high-pressure system that usually bring warm temperatures, clear skies and sunshine, light winds, a well-defined boundary layer and low humidity. Thus, changes in the climate system are expected to affect future air quality. Particularly, ozone concentrations would be affected by changes in meteorological conditions. Understanding the relationships between meteorological conditions and surface ozone is essential for defining control strategies and also for developing robust projections to evaluate future air quality. Different approaches have assessed the influence of meteorological conditions on tropospheric ozone (e.g. Barrero et al., 2005; Camalier et al., 2007; Davis et al., 2011; Dawson et al., 2007; Demuzere et al., 2009; Dueñas et al., 2011; Porter et al., 2015; Solberg et al., 2008). Most of the observational studies agree on the strong influence of specific meteorological factors on surface ozone concentrations.

As mentioned, temperature is one of the most important meteorological factors associated with high levels of surface ozone due to its direct influence in chemical reactions rates and its strong correlation with stagnant and sunny atmospheric conditions (Jacob et al., 1993). A wide number of statistical studies have shown the strong positive correlation between temperature and ozone (e.g. Camalier et al., 2007; Chaloulakou et al., 2003; Comrie, 1997; Dueñas et al., 2011; Lemaire et al., 2016; Ordóñez et al., 2007). Many chemical reaction rates increase with temperature (e.g., methane and non-methane hydrocarbon) leading to an increase of ozone production. In particular, high temperatures lead to high concentrations of ozone precursors from the dissociation of peroxycetylnitrate (PAN) and its homologs that act as reservoir species for NO_2 (Sillman and Samson, 1995). The influence of temperature on ozone levels can occur via increasing VOCs emissions from vegetation, which act as a significant source of precursors for surface ozone formation under high NO_x conditions (Rasmussen et al., 2013). Some authors pointed out the importance of the impacts of high temperatures on biogenic emissions and dry deposition, indicating the reduced uptake from vegetation due to dry conditions would lead to increase ozone levels. This has been suggested to play a significant role during the exteme temperature episodes (Hodnebrog et al., 2012; Solberg et al., 2008; Vautard et al., 2013). Similarly, Andersson and Engardt (1996) also found that changes in ozone dry deposition under future climate conditions would have a major impact (rather than isoprene emissions) on surface ozone concentrations over southern Europe. Moreover, temperature has an indirect effect on NO_x-VOC chemistry that further complicate the relation between ozone and temperature (Sillman, 1999). For instance, it has been shown variations in the ozone-temperature relationship between regions with different NO_x/VOC ratios (Steiner et al., 2006). Different modelling studies have assessed the effects of temperature-dependent chemistry on ozone production with temperature, showing that both temperature-dependent chemistry and isoprene emissions are important for the ozone increase with temperature (e.g. Doherty et al., 2013). Recently, Coates et al. (2016) used an idealised box model to determine how ozone levels change with temperature under different NO_x conditions. Their analyses suggest that reducing NO_x emission would be beneficial to offset the additional ozone production due to increasing temperatures.

The expected warming conditions will increase water vapour concentrations that will have an impact on the reaction rates of chemical processes (Stevenson et al., 2006). Such an increase under a warming climate would lead to increased ozone destruction and shorter ozone lifetime and lower concentrations over less polluted and remote sites (Johnson et al., 1999). However, there are competing effects on ozone because the hydroxyl radical (OH) plays an important role in a variety of atmospheric reactions (e.g. production of ozone from NO_x and VOCs). Increased ozone concentrations might be possible due to subsequent reactions (von Schneidemesser et al., 2015). Dawson et al. (2007) performed a sensitivity study to assess the effect of absolute humidity, among others meteorological parameters. They found that changes in water vapour concentrations (absolute humidity) have small effect on air-quality standard of ozone exceedances, but notable effects on daily maximum 8-hour averages were found. Overall, they reported a weak relationship between ozone and water vapour concentrations, which become more complicated under polluted conditions (Dawson et al., 2007).

Relative humidity is also an important variable and usually negative correlated with ozone concentrations (Jacob and Winner, 2009). Different statistical studies have shown the association between low concentrations of ozone with high levels of relative humidity (e.g. Barrero et al., 2005; Demuzere et al., 2009; Dueñas et al., 2011; Munir et al., 2011). Previous hypothesis suggested that the negative correlation between relative humidity and ozone might be due to the photolysis of ozone and subsequent loss of $O(^1D)$ to H_2O (Jacob and Winner, 2009). The effect of high levels of relative humidity has been suggested to be an indicator of precipitation and cloudiness events that favour with low concentration of pollutant (Elminir, 2005). Some authors pointed out that temperature could also explain the relathionship ozone-relative humidity, since it influences relative humidity (negatively correlated) and ozone (positively correlated) at the same time. For instance, Camalier et al. (2007) found that relative humidity was one of the most important variables (along with temperature) to explain ozone variability over eastern U.S., with a large effect in the southern urban areas and less pronounced in the northern urban regions. In this case, they argued that these regional differences could be due to the larger variations of the temperature in the northern areas than in the southern regions. Thus, they suggested that it could reflect a combined effect (with other meteorological variables, such as temperature) in polluted areas rather than a cause-and-effect relationship. Recently, Kavassalis and Murphy (2017) argued that the stomatal regulation of dry deposition (the uptake of ozone by trees) might explain the relationship ozone and relative humidity: with high levels of relative humidity, trees open their stomata and take up ozone, removing ozone from the air.

Increasing short-wave radiation provides the energy to initiate ozone formation. High levels of ozone are generally observed under abundant solar radiation (e.g. hot and sunny summertime weather conditions). Some observational studies also suggested that the strong correlations between ozone and solar radiation could partly reflect the link with high temperatures and clear sky (Ordóñez et al., 2005). High wind speed is associated with low ozone concentrations due to enhanced advection and deposition, although this relationship might involve more complex processes in some places (Tecer et al., 2003). Through a sensitivity study, Dawson et al. (2007) found that changes in wind speed appeared to play a secondary role on daily maximum 8-hour average ozone concentrations, but they found stronger effects on air-quality standard exceedances. Precipitation changes are expected to affect the rates of wet deposition of ozone precursors. Changes in cloud cover affect the photochemistry of ozone production and loss, with higher levels of ozone under reduced cloudiness conditions (Meleux et al., 2007). Additionally, changes in mixing height could affect reaction rates and the dilution of pollutants (Dawson et al., 2007).

1.4 Assessing climate impacts

As stated, future climate change may impact ozone in many different ways (e.g. modifying ozone precursors' concentrations, altering chemical production and loss rates, influencing local meteorology, among others). Climate-air quality interactions are particularly important and complex at the regional scale. The nonlinear behaviour of such interactions and the importance of regional variations of emissions requires the use of three-dimensional models, which are also essential for studying future projections of anthropogenic ozone precursors emissions. Future projections indicate that the European region is one of the most sensitive to climate change (Giorgi, 2006) and modelling studies have shown that projected European summer climate changes might have impacts on air quality (Giorgi and Meleux, 2007).

Climate models and chemistry-climate or air quality (AQ) models are used to investigate the impacts of climate on air pollution, which are able to operate at the global and regional scale (Giorgi and Meleux, 2007). Coupled climate-chemistry models at a global scale are complex and usually require major computational demands than the models operating at regional scale. The model choice will depend particularly on the spatial scale and the purpose of the study. Representing the interactions between chemistry and climate at all scales is a challenge. For instance, in the global models some mesoscale processes may be not well represented by a coarser resolution, while the limited area in the regional models may lead more uncertainities due to long-range transport processes that cannot be properly captured. Since model simulations are the primary tools available for making projections of future climate over the coming century and beyond, it is crucial to evaluate their performance individually and collectively (Flato et al., 2013). For that, models used for future projections must be evaluated against present-day observations. Moreover, the output of global climate models (GCMs) is an essential source of information and they have been extensively used to understand changes in the climate system. GCMs also provide the basis for different dynamical and statistical downscaling methods that assume a strong influence of large-scale weather on local-scale weather, but without reverse effects from local scales upon global scales (Maraun et al., 2010).

Climate impacts on future ozone pollution can be assessed by dynamically downscaling Global Climate Model (GCM) results to regional scales using regional climate and AQ models. In particular, a wide number of global and regional climate models (GCMs, RCMs respectively) in combination with AQ models have been employed for examining the implications of a changing climate on future ozone pollution over Europe (e.g. Andersson and Engardt, 1996; Engardt et al., 2009; Langner et al., 2005; Meleux et al., 2007). Climate and AQ models can be run in two main approaches: off-line and on-line. In the so-called off-line case, the climate model is run first and independently, and the resulting meteorological simulations are used to drive the air-quality model (with no feedback between tracer concentrations and climate fields). In the on-line approach, climate and AQ models are run simultaneously and then, exchanging information with each other (Giorgi and Meleux, 2007). Model simulations require a substantial computational cost, since many complex processes are involved. Particularly, in the case of the on-line models integrations of all meteorological and chemical composition fields are performed every time step, which requires very expensive computational systems (Baklanov et al., 2014). Despite computational improvements and the growing number of studies, understanding which are the most important interactions between meteorology and chemistry and how they should be implemented to improve model simulations still remains an issue (Kavassalis and Murphy, 2017).

An alternative approach has been frequently used based on the empirical relationships between surface ozone and meteorological factors. Statistical methods are well known to provide an alternative technique to assess the effect of climate on surface ozone. The main advantage of the statistical approach is that these methods are computationally efficient and they also can be easily applied to different model outputs (Wilby et al., 2002). Statistical approaches establish quantitative relations between large-scale atmospheric variables and regional or local climate conditions and thus, they would be beneficial for understanding the trends of future air quality under climate warming (Wise, 2009). Nevertheless, they assume stationarity between predictor and predictand relationships, which may be a weakness for assessing future changes (Wilby et al., 2004). Therefore, both dynamical and statistical approaches should be considered complementary.

1.5 Research questions

The overall scientific scope of this dissertation focuses on a better understanding of the role of large-scale circulation and local meteorological conditions on surface ozone concentrations over Europe. The results presented in this study will be particularly of interest for the development of mitigation strategies for future air quality over the continent.

In order to accomplish this goal, firstly large-scale atmospheric circulation is assessed through a novel approach of a traditional classification of weather types. In particular, an extended version of the Jenkinson and Collison scheme (automated version of the original Lamb weather types, LWTs) is developed. With this new implementation, the capability of a set of global models for reproducing realistic synoptic patterns is assessed, not only under present, but also under future climate conditions. Furthermore, the impacts of changes in the frequency of weather types on temperature anomalies are analysed. On the basis of this classification procedure, the influence of large-scale atmospheric conditions along with local meteorological conditions on surface ozone is investigated. For that, three statistical approaches were developed to assess the impacts on climatic factors on ozone levels under different assumptions considering: (i) the distribution of ozone as a whole, (ii) the peaks of ozone levels, and (iii) specific threshold of ozone pollution according the current legislations for health protection (EU, 2008; WHO, 2005). The final part of this research focuses on assessing the capability of AQ models to capture the observed relationship between meteorological parameters and surface ozone.

Dealing with these issues brings up the following research questions investigated in this dissertation:

1. *Do state-of-the-art climate models realistically reproduce the occurence of weather types over Europe? Based on their representation, what are the expected changes in the frequency of weather types under future climate projections? How do synoptic patterns relate with temperature anomalies in the present and future climate conditions?*

2. *How do regression-based models represent the influence of synoptic and local meteorological conditions on surface ozone over Europe? What are the main drivers of the variability of surface ozone over Europe and how do they influece the observed variability?*

3. *Are AQ models able to capture the basic relationship between meteorological conditions and ozone? Do AQ capture the drivers of ozone derived from observations? In particular, do they represent realistically the climate penalty?*

1.6 Thesis outline

This thesis consists out of 6 chapters, with a preceding introductory chapter (Chapter 1), following by a general description of the methodology used (Chapter 2). The thesis is structured into three main chapters (Chapters 3-5), each dealing with one topic and research questions formulated in the previous section. These chapters are written and prepared as a separate scientific article, and each one consists on a largely independent study with an introduction, data and methodology section, as well as the corresponding results, but also with some reference to the previous chapter. Chapters 3 and 4 are already published, while Chapter 5 has been submitted to a journal and is still under review. To conclude, a general summary and outlook are presented in Chapter 6.

Chapter 3 assesses the large-scale atmospheric circulation in a multi-model ensemble of coupled global climate models participants of fifth phase of the Coupled Model Intercomparison Project (CMIP5) experiment and their performance is evaluated against reanalysis data. Changes in the frequency of weather types under future climate are examined by using model's simulation from one Representative Concentration Pathway emission scenario using future (RCP8.5). Furthermore, the relationship between weather types and anomalies of maximum and minimum temperature is investigated.

In Chapter 4 a synoptic-regression approach is proposed to examine the influence of synoptic and local meteorological conditions on surface ozone concentrations over Europe. One of the main goals of this study is to determine the influence of the main key-driving factors of ozone. In this study, three different statistical methods are proposed to examine the influence of several synoptic and meteorological factors on ozone concentrations in three distinctive cases of the ozone distribution: the mean, the tail and the exceedances of the distribution. Moreover, regional patterns of ozone's drivers are identified.

Chapter 5 extends the analysis described in chapter 4, but with the primary objective of evaluating the capability of a set of AQ models to represent the observed relationship between surface ozone and meteorology. Through a statistical approach, this study focuses on investigating the role of meteorological drivers in models in order to identify potential sources of error when comparing to observations.

The thesis is concluded in Chapter 6 by summarising the main results from the previous chapters and by answering the reseach questions mentioned above. Finally, this chapter presents an outlook on issues that remained open and may be investigated in future studies.

Chapter 2

Methods

2.1 Methods

This research examines the impacts of the climatic factor on tropospheric ozone through the use of two methods described in this section: a circulation classification approach and regression methods integrating data from different sources.

2.1.1 Atmospheric classification

Synoptic climatology is the scientific field employed to relate larger-scale atmospheric conditions to a broad range of local-scale environmental elements (Barry and Perry, 1973; Yarnal et al., 2001) and it has been shown to be an efficient tool to explain these relationships. Synoptic classifications, commonly referred as circulation weather types (CWT), are used to categorize the continuum of atmospheric circulation into a number of discrete types of weather providing information about the atmospheric conditions for a given region (Beck and Philipp, 2010). The variability of atmospheric circulation is characterized by CWT in terms of frequency changes of several patterns on different time and spatial scale (Huth, 2000). In this context, a pattern can be defined through a field such as sea level pressure, geopotential or any variable that describes the atmospheric circulation for each time instant of the analysis on a grid (Huth et al., 2010). Overall, there is not one generally best individual CWT and the choice of a classification methodology will depend on the particular study and purpose (Huth et al., 2008). A wide range of classification schemes have been developed under The European project “Harmonization and Applications of Weather Types Classification for European Regions-COST733” which offers a general numerical method for assessing, comparing and classifying atmospheric situations over Europe (Huth et al., 2008, 2010; Philipp et al., 2010).

At present, circulation patterns are used for many purposes: human mortality (Kassomenos et al., 2001); understanding the links with surface climate variables, such as precipitation (Goodess and Jones, 2002; Lorenzo et al., 2008; Trigo and Dacamara, 2000) or temperature (Chen, 2000); investigating the links with extreme events, such as storms (Donat et al., 2010), or droughts (Paredes et al., 2006); analysis with environmental variables, for instance wildfire occurrence (Kassomenos, 2010) or air quality (Comrie, 1992; Dayan et al., 2012; Demuzere et al., 2009; Kallos and Pielke, 1993; Leśniok et al., 2010; Russo et al., 2014). Specifically in the context of air quality, a wide number of studies have focused on examining the relationship between air pollution and synoptic patterns in different regions around the world, for instance, in Australia (Hart et al., 2006), Canada (Cheng et al., 2007), Greece (Flocas et al., 2009; Kallos and Pielke, 1993), United States (Comrie, 1992; Davis and Gay, 1993; Yarnal, 1993), China (Chen et al., 2008), United Kingdom (Davies et al., 1992; O'Hare and Wilby, 1995), Portugal (Russo et al., 2014), Iberian Peninsula (Saavedra et al., 2012; Santurún et al., 2014), Netherlands (Demuzere and van Lipzig, 2010), Scandinavia (Tang et al., 2009), Israel (Dayan and Levi, 2002). They have shown that the circulation patterns approaches can be successfully used for air quality applications.

The usefulness of CWT resides in the substantial information on the atmospheric state and its use can provide some physical evidence in the driving of large-scale variable conditions that form the basis of the environmental variables of study (Demuzere and van Lipzig, 2010). Previous works have been investigating trends in surface climatic variables to determine whether certain atmospheric patterns are related to specific surface weather conditions and/or whether this relationship remains stable over long period of times (Canynová and Huth, 2016). Moreover, an enhanced lifetime of circulation types would indicate the persistence of atmospheric circulation, which would have implications on regional weather conditions. For instance, some studies pointed out that an increasing persistence of atmospheric circulation contributes to extreme temperature events. Domonkos et al. (2003) investigated the long-term fluctuations in the frequencies of winter and summer extreme events (cold and summer respectively) and they found strong connections between the frequency of extreme events and the residence time of circulation patterns. Kyselý (2007) found that the persistence of circulation patterns was linked to temperatures anomalies and then, circulation patterns were conductive to heat and cold weaves.

The use of CWT in this research has been mainly motivated for several reasons. Firstly, given that atmospheric circulation contains general information about local meteorological conditions, it is an effective tool to obtain information about how large-scale processes relate to other aspects of weather, such as temperature. Secondly, synoptic weather patterns may directly influence surface ozone concentrations through the transport or accumulation of ozone and its precursors. Finally, specific weather types can provide the most favourable meteorological conditions for ozone pollution, which is another indirect influence of synoptic conditions on high ozone levels. For example, during summertime anticyclonic situations are usually associated to warmer temperatures, abundant radiation and reduced cloud cover that can promote pollution episodes.

One of the most well known schemes for classifying atmospheric circulation was developed by Lamb in 1972 for the British Isles and it was extended from 1861 to 1997 (Hulme and Barrow, 1997). The original LWTs method classified manually the atmospheric patterns (mostly using sea level pressure) according to the wind direction and circulation type. Jenkinson and Collison (1977) presented an automated version of the subjective LWTs classification. A comparison of both methods (Jones et al., 1993) showed the agreement between them and the frequencies of the objective types were highly correlated to the traditional Lamb types. Jones et al. (2013) also provided an extensive review about the objective LWTs over the British Isles bases on extended reanalysis products.

The original Jenkinson and Collison (hereinafter, JC) classification uses a coarsely gridded pressure data on a 16 grid-points with a 10º resolution in zonal and a 5º resolution in meridional directions, for a central point located at 55ºN latitude and 5ºW longitude (Fig. 1). Thus, the circulation pattern for a given day is described using the locations of the centers of high and low pressure that determine the direction of the geostrophic airflow. Each day is then assigned both, a vorticity type and a wind flow direction. Based on the original catalogue and procedures (Jones et al., 1993), a set of indices associated with the direction and vorticity of geostrophic flow are calculated. These indices are: southerly flow (SF), westerly flow (WF), total flow (F), southerly shear vorticity (ZS), westerly shear vorticity (ZW) and total shear vorticity (Z). They are calculated according to the expressions detailed in the paper I (and references therein), as well as the rules for classifying the days into the types. Specifically, this classification allows 27 types of weather: 8 directional, 1 cyclonal (C), 1 anticyclonal (A), 8 hybrid cyclonal-directional, 8 hybrid anticyclonal-directional and 1 undefined.

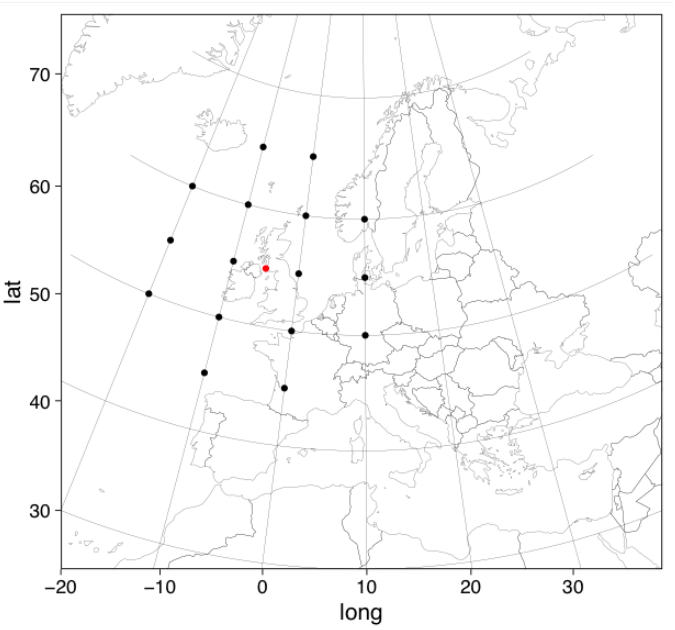

Figure 2.1 Configuration of the JC scheme originally applied over British Islands for a central point (red dot) located at 5ºW, 55ºN surrounded by the 16-points (black dots).

The JC scheme has been used in many studies with a different resolution, 5º x 5º (Lorenzo and Taboada, 2005; Trigo and Dacamara, 2000) and other configuration of points (Dessouky and Jenkinson, 1977; Grimalt et al., 2012; Spellman, 2000). It has been widely applied for many purposes involving different environmental variables, such as precipitation (Cortesi et al., 2013; Russo et al., 2015; Vicente-Serrano et al., 2011), snow depth (Vicente-Serrano and López-Moreno, 2006), temperature (Post et al., 2002)). Moreover, this classification has been used to investigate extreme events, such as storms (Donat et al., 2010), droughts (García-Herrera et al., 2007) and air pollution (Comrie, 1992; Davies et al., 1992; Demuzere et al., 2009; Pope et al., 2014; Santurún et al., 2014; Tang et al., 2009). Most of these studies were restricted to particular locations over Europe (e.g. Iberian Peninsula, British Isles, Netherlands, Germany, or Scandinavia). Therefore, motivated by this successful application in many different regions, this research consider it worthwhile to develop an extended version of the JC classification at every grid-point over the map in order to investigate directly and indirectly the influence of atmospheric circulation on surface ozone. Although the JC classification was implemented around the world, due to the scope of this dissertation, for the present study only the European domain is considered. Thereby, in order to investigated the links between synoptic weather types and European ozone pollution levels, the impacts of weather types on maximum and minimum temperatures (one of the main influential meteorological drivers of surface ozone) are further examined (paper I, Chapter 3). Finally, the second independent study uses the airflow indices derived from the classification procedure to investigate their direct influence on surface ozone concentrations (paper II, Chapter 4).

The extended classification of the JC classification developed during this dissertation will be released as a R-package to the CRAN package repository.

2.1.2 Statistical-regression methods

Statistical methods have been widely used within the context of air quality. In particular, many studies have been assessed the impact of meteorology on tropospheric ozone. Statistical techniques can be applied from a predictive point of view, to obtain ozone forecast (e.g. Barrero et al., 2005; Chaloulakou et al., 1999; Comrie, 1997), to better understand the underlying mechanisms (e.g. Ordóñez et al., 2005; Varotsos et al., 2013), but also to investigate and estimate ozone trends (e.g. Bloomer et al., 2009; Bloomfield et al., 1996; Gardner and Dorling, 2000). A representative study from Thompson et al. (2001) established a review of the main statistical approaches applied for tropospheric ozone evaluation, which were categorised into regression-based modelling, extreme values and space-time methods. Overall, they concluded that there is not one simple method most appropriate for all purposes and all meteorological scenarios, and thus, the choice of the methodology will depend on the aim of the analysis and the meteorological conditions of ozone formations for a given location. Schlink et al. (2003) comprehensively evaluated 15 different statistical techniques for ozone forecasting applied to ten data sets representing different meteorological and emission conditions throughout Europe. They suggested that those techniques that can handle nonlinearities might give better results for ozone predictions.

In addition, combined regression analyses and circulation-based methods haven been previously applied in the context of air quality, since they can reflect both local meteorological and atmospheric conditions (Cheng et al., 2007; Demuzere and van Lipzig, 2010; Demuzere et al., 2009; Tang et al., 2011). As described in Chapter 4 (paper II), a synoptic-regression approach has been used to further investigate to role of synoptic conditions along with local meteorological conditions on European surface ozone concentrations. While most of the former studies employed observational data set over specific locations, this dissertation offers an extended synoptic-regression approach over Europe, where statistical models are individually developed at each grid-point over the wide domain.

A brief description of the statistical models used in Chapters 4 and 5 is presented below.

Multiple linear regression

In the context of ozone pollution, one of the most widely used methods is linear regression that assumes normality of the data distribution and linearity of the associations between variables. Linear regressions are effective tools for identifying connections between specific meteorological conditions and the mean pollutant response. A large number of studies have applied multiple linear regression (MLR) approaches for modelling tropospheric ozone using not only meteorological variables as predictor variables, but also introducing other pollutants (Barrero et al., 2005; Chaloulakou et al., 1999, 2003; Comrie, 1997; Demuzere and van Lipzig, 2010; Demuzere et al., 2009; Dueñas et al., 2011; Sousa et al., 2006, among others). Model comparisons between linear and nonlinear regression methods suggested that nonlinear approaches might be superior might be superior from ozone forecast (e.g Chaloulakou et al., 2003; Comrie, 1997; Sousa et al., 2006)). However, MLR approaches have been successfully applied due to their simplicity and particularly because MLR models are more readily interpretable in terms of the underlying physical mechanisms between the variables involved (when compared against non-linear methods) (Demuzere et al., 2009; Gardner and Dorling, 2000). In this case, the use of MLR is an appropriate and preferable method to investigate the ozone mean pollutant response and to identify the most important drivers of ozone pollution levels as a whole.

MLR establishes a quantitative relation between the predictant and the predictor or group of predictors that can be useful for a future prediction of such predictant (Abdul-Wahab et al., 2005). The general form of the linear regression model can be expressed as:

$$\hat{O}3 = \beta_0 + \sum_{i=1}^{n} \beta_i X_i + \varepsilon \tag{2.1}$$

where β_0 is the intercept and β_i are the coefficients to be determined by the linear regression, which are independent variables (more detail about the variables used, see Chapter 4 and 5), ε refers to the residuals terms and $\hat{O}3$ are the modeled values of ozone concentrations.

Quantile regression

The link between meteorology and ozone has been shown to be considerably dependent on both space and time, which adds some non-linearities into their relationship and makes more complicated ozone modeling, due to its changing behavior (Baur et al., 2004). This implies that the contribution of the explanatory variables to ozone concentrations have significant changes at different ozone concentrations (Baur et al., 2004; Munir et al., 2012). In some specific cases, for example to examine the ozone response across the distribution, specifically at the extremes (e.g. above the 95th percentile), the use of a more flexible technique is required. Quantile regression (QR) allows the examination of the entire distribution of ozone, rather than measuring the central tendency of its distribution (Koenker and Basset, 1978). Therefore, the choice of a QR approach is mainly motivated by its flexibility to analyse the main drivers of peak ozone levels (i.e. 95th percentile).

Quantile regression (QR) (Koenker and Basset, 1978) expands the flexibility of both parametric and non-parametric regression methods, and it allows the covariates to have different impacts at different points of the distribution and the robustness to departures from normality and skewed tails (Mata and Machado, 1996). In the context of high ozone levels, QR is particularly important since it is a more flexible method for ozone modeling with heterogeneous conditional distribution. Previous studies have shown the potential of this technique in environmental research (Munir et al., 2012; Sousa et al., 2009). Baur et al. (2004) modelled the impact of meteorology and the ozone persistence (ozone concentrations from the previous day) on ozone concentrations over Athens. They found that upper air temperature had a major effect on high levels than on low levels of ozone. Similarly, Sousa et al. (2008) used a QR approach for modelling ozone over Northern Portugal and they found that wind direction was influential in the medium quantiles, while relative humidity was more important in the higher quantiles. A more recent study from (Porter et al., 2015) found key differences in covariate sensitivities of ozone across US and quantiles. Overall, they showed that the key drivers of high-quantile ozone levels were temperature and relative humidity in summer, and incoming radiation flux in winter. These studies suggested that QR methods might provide important insights on the different determinants of ozone concentrations.

QR can be seen as an extension of the least squares estimation of conditional mean models and it specifies the conditional quantile function:

$$\hat{O}3 = \beta_0^{(\tau)} + \sum_{i=1}^{n} \beta_i^{(\tau)} X_i + \varepsilon^{(\tau)} \tag{2.2}$$

In this case the constants $\beta_0^{(\tau)}$ and $\beta_i^{(\tau)}$ are estimated for the different percentiles using each time the entire dataset (e.g. 0.95 percentile). Therefore, the intercept and the coefficients will be generated independently for each selected quantile (Koenker, 2005).

Generalized linear model

As mentioned, the complex relationship between the meteorological factor and ozone implies that its behaviour varies considerably with the ozone distribution. This means that at high ozone levels the influence of meteorology might play a different role than in a normal distribution of surface ozone concentrations. Within the framework of regression-based methods, generalized linear models (GLMs) (Nelder and Wedderburn, 1972) are an extension of the classical linear regression models that somewhat relaxes the strict linearity of assumptions of linear models. The use of GLMs allow the expected value of the response to depend on a smooth monotonic function of the linear predictor and the response can follow any distribution from an exponential family (e.g., binomial, gamma etc.) (Wood, 2006). In this study, GLMs are selected to estimate the probability of ozone exceedances based on the current target values: $120\,\mu g/m^3$ and $100\,\mu g/m^3$ (EU, 2008; WHO, 2005, respectively).

GLMs have been applied for analysing daily rainfall sequences and daily maximum wind speed (Chandler and Wheater, 2002; Yan et al., 2002) and they showed their potential for representing complex relationship among variables in the climate system. GLMs are effectively probability models that can simulate realistic sequences (or occurrences) of environmental variables. Some authors successfully applied GLMs for analysing ozone concentrations and the role of meteorological variables (Camalier et al., 2007; Davis et al., 2011). Furthermore, in the context of nonlinear methods, many studies have proposed the use of generalized additive models (GAMs), which are GLMs with a linear predictor involving a sum of smooth functions of covariates (Hastie and J.Tibshirani, 1990), to investigate the nonlinear associations between ozone and meteorological variables (e.g. Carslaw et al., 2007; Davis et al., 1998; Gardner and Dorling, 2000; Munir et al., 2011). Generalized linear models (Nelder and Wedderburn, 1972) allow for response distributions other than normal, and for a degree of non-linearity in the model structure (Wood, 2006). The general equation can be expressed in a similar way as in (1), but in this case now $\hat{O}3(\tau)$represents the link function that relates the mean of the response to the linear predictors in the model. In our case, the response variable takes two possible outcomes (0 or 1) and it is modelled using a binomial distribution.

The following three chapters describe in detail the application of the methods presented in this chapter as well as the data used in each study.

Chapter 3

Paper I: Assessment of an extended version of the Jenkinson–Collison classification on CMIP5 models over Europe

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Chapter 4

Paper II: Synoptic and meteorological drivers of extreme ozone concentrations over Europe

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Synoptic and meteorological drivers of extreme ozone concentrations over Europe

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Synoptic and meteorological drivers of extreme ozone concentrations over Europe

N Otero[1], J Sillmann[2], J L Schnell[3], H W Rust[4] and T Butler[1]

[1] Institute for Advanced Sustainability Studies e.V, Potsdam, Germany
[2] CICERO Center for International Climate and Environmental Research-Oslo, Norway
[3] Department of Earth System Science, University of California, Irvine, CA, USA
[4] Freie Universität Berlin, Institut für Meteorologie, Berlin, Germany

E-mail: Noelia.OteroFelipe@iass-potsdam.de

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Abstract

The present work assesses the relationship between local and synoptic meteorological conditions and surface ozone concentration over Europe in spring and summer months, during the period 1998–2012 using a new interpolated data set of observed surface ozone concentrations over the European domain. Along with local meteorological conditions, the influence of large-scale atmospheric circulation on surface ozone is addressed through a set of airflow indices computed with a novel implementation of a grid-by-grid weather type classification across Europe. Drivers of surface ozone over the full distribution of maximum daily 8 h average values are investigated, along with drivers of the extreme high percentiles and exceedances or air quality guideline thresholds. Three different regression techniques are applied: multiple linear regression to assess the drivers of maximum daily ozone, logistic regression to assess the probability of threshold exceedances and quantile regression to estimate the meteorological influence on extreme values, as represented by the 95th percentile. The relative importance of the input parameters (predictors) is assessed by a backward stepwise regression procedure that allows the identification of the most important predictors in each model. Spatial patterns of model performance exhibit distinct variations between regions. The inclusion of the ozone persistence is particularly relevant over southern Europe. In general, the best model performance is found over central Europe, where the maximum temperature plays an important role as a driver of maximum daily ozone as well as its extreme values, especially during warmer months.

1. Introduction

Tropospheric ozone has adverse impacts on human health (Fang *et al* 2013), forests and agricultural crops (Booker *et al* 2009), and contributes to climate change (Jacob and Winner 2009). Given the harmful effects of high ozone concentrations, especially in terms of human health, ozone remains an important air quality issue. Therefore, the World Health Organization (WHO) Air Quality Guidelines (AQG) have set 100 μg m^{-3} (as a maximum daily value of the 8 h running mean) as a target value for ozone for the protection of human health, while the European Union suggests 120 μg m^{-3} (WHO 2014c).

Surface ozone concentrations are strongly dependent on meteorological variables, such as solar radiation fluxes, temperature, cloudiness, or wind speed/direction (Dueñas *et al* 2002, Gardner and Dorling 2000). Atmospheric circulation controls the short and long-term transport (Demuzere *et al* 2009) of ozone, and it can also affect the interaction among ozone precursors, facilitating its formation and destruction (e.g., Davies *et al* 1992a, 1992b, Comrie and Yarnal 1992, Saavedra *et al* 2012). In addition, the transport of emitted ozone precursors from urban and industrialised areas may even cause photochemical production of ozone in regions far from the source of the emissions (Holloway *et al* 2003). The relationship between surface ozone and meteorological variables is complex and nonlinear (Comrie 1997), but is usually strongest in summertime due to high temperatures, peak solar radiation and stagnant conditions (Jacob and Winner 2009, Andersson and Engardt 2010).

The motivation for this study is to investigate the spatial response of surface ozone to meteorology and prevailing atmospheric conditions to better understand the drivers of surface ozone and its variability. One of the main objectives of this work is to examine the relevance of different meteorological variables of surface ozone over Europe, in order to better understand how ozone air quality could be expected to change under future climatic conditions. Our approach is novel as it is not restricted to small regions or single countries but the entire European domain as we combine a recent gridded data set of interpolated surface ozone concentrations with a novel implementation of a circulation classification method applied to a gridded meteorological reanalysis data set for Europe. We aim to identify the most important drivers of maximum daily ozone levels as well as characterize the drivers of extreme ozone levels, in spring (March, April, May) and summer (June, July, August) months during the period 1998–2012. For these purposes, statistical models are built for each grid cell in the European domain using three different regression methods: multiple linear regression to assess the drivers of the mean as well as quantile and logistic regression for high percentiles and threshold exceedances respectively.

2. Data and methods

We use a recent interpolated data set of observed maximum daily 8 h average surface ozone (MDA8) concentrations provided by Schnell *et al* (2015), who have developed an objective mapping algorithm to calculate hourly surface ozone averaged over 1° by 1° grid cells, over the period 1998–2012. This interpolation of surface ozone concentrations provides a 1° × 1° product with a similar resolution to current global CTMs and allows for the examination of the influence of atmospheric circulation and meteorological conditions from different data sets in a similar resolution.

The ECMWF ERA-Interim reanalysis dataset (1° × 1°) (Dee *et al* 2011), for the same period of time, 1998–2012, is used. Daily mean values are calculated as the mean of the four available analysis fields at 00, 06, 12, and 18UTC for the following variables: mean sea level pressure, zonal (u) and meridional (v) wind components at 10 meters, temperature at 2 m, total cloud cover, geopotential and relative humidity, both at 1000 hPa. Maximum of temperature is obtained as the maximum of these four values per day. Moreover, daily means are also computed from the 3-hourly forecast fields: surface solar radiation downwards and surface thermal radiation downwards. This data defines the local meteorological conditions at each grid cell. Additionally, we define synoptic scale potential meteorological drivers in the following.

2.1. Synoptic meteorological conditions

This study uses an objective scheme developed by Jenkinson and Collinson (1977) of the Lamb weather types catalogue (Lamb 1972) to classify daily atmospheric circulation. The original scheme, developed for the British Isles, has been widely used for other regions in mid-latitudes, mostly in the north of the European continent (e.g., Spellman 2000, Trigo and Dacamara 2000, Linderson 2001, Goodess and Jones 2002, Tomás *et al* 2004, Grimalt *et al* 2013) for many different purposes. We offer a novel approach of the traditional objective Jenkinson and Collinson (1977) (in the following refer to as JC97) classification, by applying the scheme point-by-point (i.e., at each grid-cell) and thus, a new gridded data set of daily weather types (WT) is created.

According to the JC97 procedure, daily circulation is characterized through the use of a set of airflow indices (Lamb indices) associated to the direction, speed and vorticity of geostrophic flow (Jones *et al* 1993). Such indices of air flow computed for categorizing weather types (i.e., vorticity, strength and direction of the flow) can be used directly as predictors in a regression model (Maraun *et al* 2011, 2012) and they contain the information about the intensity of a given weather type and its subsequent relation with ozone concentrations (Hegarty *et al* 2007). As Conway *et al* (1996) point out two important advantages of using these: firstly, they provide information about the development of the circulation system without the need of separating into categories; secondly, and especially important for our statistical analysis, they are continuous variables, rather than categorical variables such as Lamb weather types. Hence, a set of airflow indices extracted from the JC97 classification is included as predictors in the model development (table 1).

2.2. Statistical model development

Multiple linear regression (MLR) is considered an effective tool to study the relationship between the predictors and the mean of the response variable, allowing identification of the main drivers of MDA8 surface ozone concentrations. MLR models and their estimation using ordinary least-squares is one of the most used techniques for statistical modelling of ozone pollutant concentrations (Thompson *et al* 2001). Furthermore, combined regression analysis and circulation-based methods have been applied in air quality research (Cheng *et al* 2007a, 2007b, Demuzere and van

Table 1. Predictors used in the regression models: local meteorological parameters, airflow indices, seasonal components and lag ozone.

Local meteorological parameters	Definition	Synoptic meteorological parameters	Definition
Tx	Maximum temperature	WF	Westerly flow
RH	Relative humidity	SF	Southerly flow
SR	Surface solar radiation	TF	Total Flow
ST	Surface thermal radiation	VW	Westerly shear vorticity
Gh	Geopotential height	VS	Southerly shear vorticity
TC	Total cloud cover	V	Total shear vorticity
Ws	Wind speed at 10 m	D	Direction of flow
Cy	$\sin(2\pi d/365)$, $\cos(2\pi d/365)$	LO3	Lag of O3 (24 h)

Lipzig 2010a, 2010b, Pearce *et al* 2011) with the advantage that this approach may reflect both local meteorological conditions and large-scale atmospheric circulation (Tang *et al* 2011). Here, we apply MLR to analyse the mean of surface ozone response.

Statistical methods such as quantile regression (QR) (Koenker and Basset 1978) expand the flexibility of both, parametric and non-parametric regression methods. For instance, QR allows the predictors to have different impacts at different points of the distribution and the robustness to departures from normality and skewed tails (Mata and Machado 1996). QR has shown its effectiveness in environmental studies where extreme values are important (Sousa *et al* 2008, Munir *et al* 2012) and for which the previous models (MLR) would fail due to their dependence on the mean (Munir *et al* 2012). Here, QR is applied to examine the effect of the meteorological drivers at the 95th percentile.

The current target values from the WHO (AQC) and the EU legislation set relevant thresholds for ozone concentrations. We use logistic regression (LR) to model the probability of ozone exceedances over these thresholds depending on the most important drivers. Logistic regression is a special case of generalized linear models (Nelder and Wedderburn 1972, McCullagh and Nelder 1989), which is a generalization of classical linear regression. It includes a static non-linear transformation (link-function) and the response is not restricted to a normal distribution (Wood 2006). Occurrences of threshold exceedance can take values of 0 (not exceeded) or 1 (exceeded), so the associated distribution for probabilities of these exceedances is the binomial distribution.

One common problem of logistic regression emerges due to an insufficient number of events (i.e., exceedance) with respect to the number of predictors. Previous studies suggest the use of 10–20 events per variable (Harrel *et al* 1985, Agresti 2007), while others concluded that only 5–10 events are sufficient (Peduzzi *et al* 1996). In our case this number of events depends on the threshold chosen for exceedance of ozone concentration: 100 μg m^{-3} (~50 ppb) and 120 μg m^{-3} (~60 ppb), motivated by WHO AQGs and EU respectively. Taking into account the above suggestions for the minimum number of events, we use 100 events at a grid cell for a logistic regression to be performed (which would cover the number of 5–10 events suggested, in this case, 17 predictors).

2.3. Selection of predictors

The choice of the input parameters and selection of the most appropriate variables is a crucial step in statistical modelling. We include some of the most commonly used parameters as potential predictors among which we systematically select: maximum temperature (Camalier *et al* 2007, Demuzere *et al* 2009), relative humidity (Dueñas *et al* 2002, Sousa *et al* 2008), total cloud cover (Bloomfield *et al* 1996), solar radiation fluxes (Chaloulakou *et al* 2003, Baur *et al* 2004), geopotential height (Camalier *et al* 2007, Porter *et al* 2015) and wind speed (Dueñas *et al* 2002). Moreover, 7 airflow indices, that add information about the relationship between ozone and prevailing synoptic conditions are also included. Additionally harmonic functions capture the effect of seasonality as in Rust *et al* (2009). Table 1 provides the list predictors used in the regression models.

The possibility of pollution episodes when levels of previous day concentrations are higher than normal has been reported by previous studies (Robeson and Steyn 1990, Ziomas *et al* 1995). Persistence of ozone (the use of values from the previous day) as used for precipitation in Rust *et al* (2013) may be a straightforward predictor that usually plays an important role to predict ozone concentrations (Barrero *et al* 2005, Banja *et al* 2012). Moreover, it has been shown that model performance increases by including persistence of air quality variables (Pérez *et al* 2000, Smith *et al* 2000, Grivas and Chaloulakou 2006). Therefore, persistent polluted episodes are accounted for by including the previous day of ozone (24 h time lag) explicitly as a predictor.

The selection of predictors is made independently for each grid-cell through a backward stepwise regression procedure. Starting with a model that includes all potential predictors, at each step the least important is sequentially removed from the regression equation according to the Akaike information criterion (AIC, Akaike 1974). In many cases predictor variables are related to each other, which leads to multicollinearity, typically resulting in underestimation of confidence intervals. A simple way to detect collinearity is to look at the correlation matrix of the predictors. In our case, we found some frequent strongest correlated pairs of predictors (e.g., total shear vorticity with both westerly and southerly components, westerly flow and direction of the flow, geopotential and total shear vorticity or relative humidity and solar radiation), which might potentially lead to unstable parameter estimates. Therefore, to deal with this situation a multicollinearity index known as variance inflation factor (VIF) is commonly used (Maindonald and Braun 2006). In our procedure particularly the, variables with a VIF above 10 are left out of the equation (Kutner *et al* 2004). After selecting the best candidates at each grid-cell independently, we assess the models performance in terms of the coefficient of determination R^2 ($0 < R^2 < 1$), with larger values indicating more variability described by the model according to their influence.

The predictor's relative importance is assessed at each grid-cell over Europe. In the case of linear regression methods, the main important predictors of the ozone are estimated using the coefficient of determination R^2, which is partitioned by averaging over orders, according to the method proposed by Lindeman *et al* (1980) (Grömping 2007). To examine the drivers of ozone exceedances, the predictors are first normalized. In QR the relative importance of the drivers is estimated by using an analysis of variance (ANOVA), which is frequently applied as a test of significance. Then, a comparison between a model with and without a predictor shows the importance of this parameter. We rank the drivers in relation to their absolute value of the significance test and their normalized coefficients. A similar process based on the absolute value of the t–statistic for each individual parameter is applied in LR.

3. Results

3.1. Drivers of maximum daily 8 h ozone

Table 2 summarizes each predictor's frequency of selection used in the MLR models for summer and spring. The screening process leaves the ozone persistence (LO3) as the most used predictor for both seasons. In summer, this is followed by the maximum temperature (Tx), the thermal surface radiation (ST), the airflow indices related to the strength of the resultant flow: southerly flow (SF) and westerly flow (WF), as well as the wind speed (Ws). The total vorticity airflow (V) is always removed due to the high correlations with its two components. The least frequently chosen predictors in summer are the total cloud (TC) and the solar radiation (SR). The results obtained for spring show that the most frequent predictors after ozone persistence are relative humidity (RH) and Tx, followed by the SF airflow index, and SR The direction of the flow (D) along with the total flow (F), show the lowest frequency of appearance.

The performance of the models is higher in summer than in spring and this feature is especially observed in central and north-west Europe (figure 1). Overall, the inclusion of LO3 improves the model, which is reflected by the relative contribution to total explained variance and its relative importance in the model (figure 2). Our results show that LO3 has a stronger influence in some specific regions. For example, we detect that the model's performance in most of south Europe improves markedly due to the effect of LO3. In particular, the increase is more pronounced over southeastern regions (i.e. Balkan Peninsula) in both seasons, whereas in some grid-cells over the southwest (e.g., Spain) there is a slight increase of the performance in spring. Models over north Europe also improve because of a larger effect of LO3, especially in summer. The relatively weak role of meteorological variables as predictors in all these regions (e.g., the Iberian Peninsula, Balkan Peninsula or Scandinavian), and the influence of persistence of ozone over those specific grid-cells, may suggest a stronger role for precursor emissions in driving ozone concentrations in these regions. However, in central Europe the models' performance is robust and it is observed that some meteorological parameters (e.g., Tx, RH or SR) play an important role in explaining most of the ozone variance. That suggests that there is a significant influence of meteorological variability in driving maximum daily ozone in this region. The mean bias has been assessed in the supplementary material, (section 3).

The spatial distribution of the first three drivers of ozone in spring and summer show the effect of the ozone persistence over most of Europe (figure 2). In general, the inclusion of the harmonic functions (Cy) reveals different regional variations of the seasonal cycle (e.g., northeast Europe). From a statistical point of view, Cy can be considered as a proxy of physical processes and thus, its dominant role in some regions might be explained by a major dependence of the Cy on other parameters (e.g., SR or Tx). Given that both variables (LO3 and Cy) are not directly meaningful physical drivers of ozone, we focus hereinafter on describing the role of the meteorological predictors as ozone drivers. Moreover, the strength of the relationship between each predictor and ozone can be interpreted in terms of the magnitude and the sign of the predictor's coefficient (not shown).

In spring, RH and SR are leading meteorological drivers of ozone over most of Europe. Tx is also another important driver, although less dominant in some places over north and central Europe. RH has a negative relationship with ozone, and it is an important driver in the northeast and in some regions in the west, specifically most of Portugal and Ireland. The impact of RH on ozone has been reported in previous studies that found strong negative correlations between relative humidity and ozone (Demuzere *et al* 2009, Dueñas *et al* 2002). Higher levels of humidity usually imply more cloudiness and instability,

Table 2. Frequency (%) of selection of predictors in the MLR models developed in MAM and in JJA over all grid points.

MLR	N° Models	Season	LO3	Tx	RH	Gh	SR	ST	Ws	TC	WF	SF	TF	VW	VS	D
	969	MAM	100	80.0	85.3	45.1	73.4	55.1	61.2	54.3	62.8	75.2	41.8	51.4	49.6	35.1
	969	JJA	100	90.5	71.5	56.2	53.3	75.7	72.9	45.1	74.9	74.7	55.3	61.9	57.0	54.8

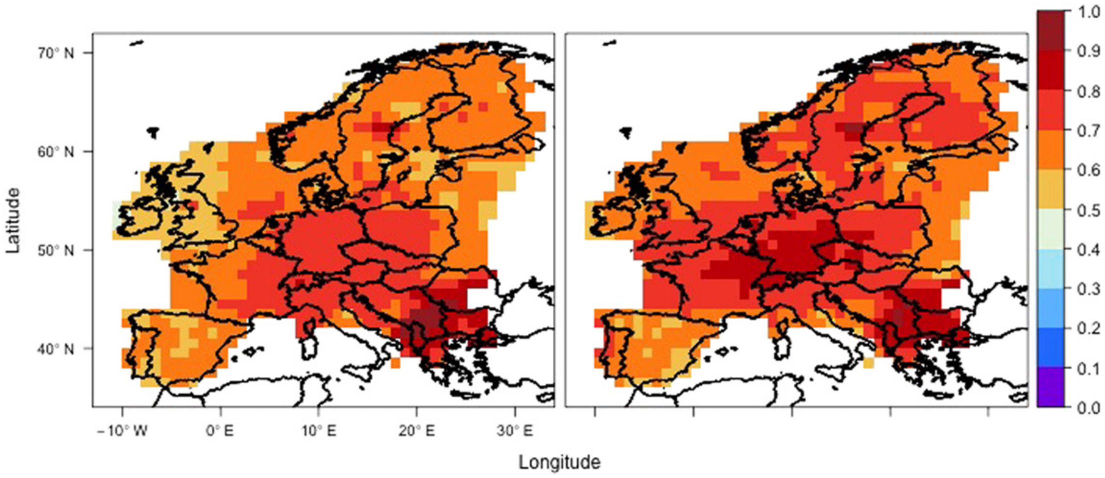

Figure 1. Spatial distribution of the R^2 for MLR for MMA (left) and JJA (right).

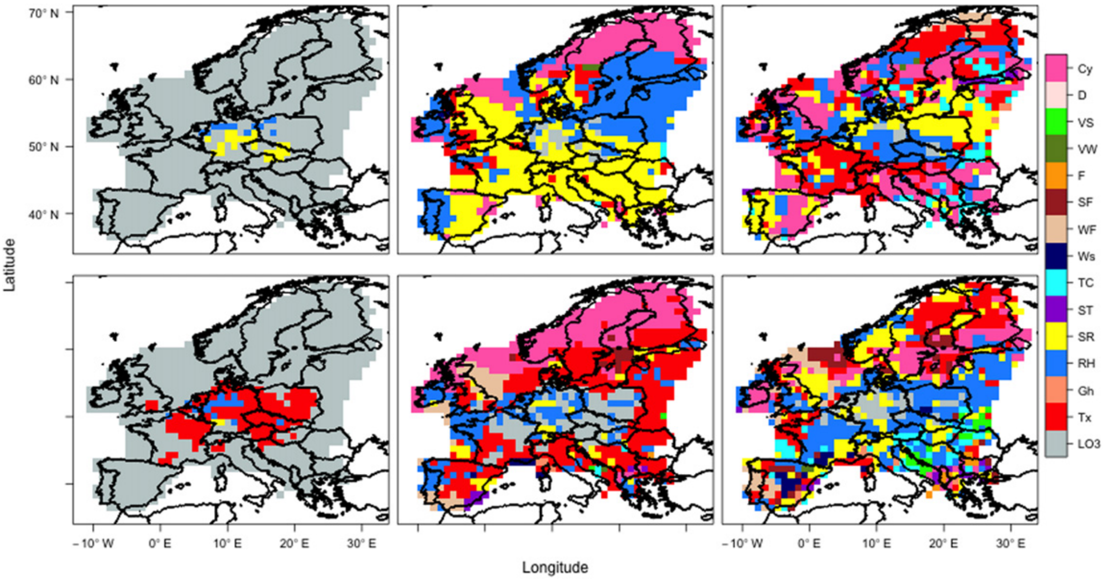

Figure 2. Spatial distribution of the most important driver (left), second most important (middle) and third most important (right) of ozone in MLR for both seasons: MAM (top) and JJA (bottom).

which suggests a reduction of ozone production (Camalier *et al* 2007, Porter *et al* 2015). A similar negative relationship is found for other meteorological variables associated with conditions of instability (TC, WF, and VW) in some specific grid-cells in the eastern regions. In contrast, SR has a positive effect on ozone and it appears as an important driver over central and south Europe.

In summer the clear dominant meteorological driver is Tx, which is positively related to ozone, especially over central Europe where it has a larger impact. Tx is also significant in the eastern and southern regions, albeit with a smaller effect. The influence of the temperature on biogenic emission has been widely investigated and in particular, the emissions of the biogenic ozone precursor isoprene increase with increasing ambient temperature (Pusede *et al* 2014). Moreover, high temperatures are usually also associated with enhanced evaporative emissions of anthropogenic VOCs (volatile organic compounds) (Ordóñez *et al* 2005). Previous studies have been established a VOC-limited regimen over those regions (Beekmann and Vautard 2010), which could explain the larger dependence of ozone on maximum temperature under specific VOC-limited conditions (Pusede *et al* 2014). In addition, the enhanced thermal decomposition of peroxyacyl nitrates (PANs) at high temperatures yields higher *in situ* ozone production, but lower downwind production (Sillman and Samson 1995). This dominance of Tx during the warmer months could be explained by its effect on ozone precursors. Other variables also play important roles in summer: for instance, RH and WF, both with a negative effect, are dominant drivers in the western regions, SR positively related to ozone in some grid-cells in southern and northern regions, or the airflow indices SF and VS with a negative effect on ozone. These results point out the main drivers of ozone are dominated by local meteorological parameters, rather than the airflow indices that define synoptic meteorological conditions.

3.2. Drivers of extreme ozone conditions

Table 3 summarizes the frequency of explanatory variables in the QR analyses of the 95th percentile of MDA8 ozone, both for spring and summer. After the screening process the LO3 is always selected as a predictor for both seasons. Tx and the airflow index SF are the most selected predictors in summer, while D and TC are those with the lowest frequency. In spring, SR and RH are the most used variables at the 95th percentile and Tx and D are the least used. In this case, less than 50% models include Tx in the predictor's subsets due to the high level of multicollinearity of Tx with the rest of the variables. Unlike in the MLR models, now the selection procedure during the spring months replaces Tx with other variables, and it does not appear to be a significant variable for modelling the high ozone percentiles.

Given that the number of exceedances depends on the chosen threshold, a different set of LR models is developed in spring and summer (table 4). Here, we specifically focus on logistic modelling for the 50 ppb limit (LR_{ex50}), for which there is a larger number of ozone exceedances over most of Europe. The results obtained with two higher limits, 55 and 60 ppb can be found in the supplementary material, (section 2).

Table 4 summarizes the frequency of appearance of individual predictors in the modelling process LR_{ex50}. LO3 is the most often selected variable in both seasons. Moreover, the screening process shows that SR, SF and RH are the most frequent used predictors in spring, whereas in summer these are Tx, Ws and ST. In general, D shows the lowest frequency of appearance in summer, whereas in spring Tx is least frequent. As in the QR analysis, the frequency of Tx considerably decreases in spring due to the multicollinearity with the rest of the variables. This result suggests that in spring Tx is less relevant for driving extreme values of ozone in many grid-cells, which differs from the result obtained when examining drivers of the whole distribution of ozone values. In that case, Tx along with RH appears to be one of the most frequent variables in spring (table 2).

The model's performance in QR at the 95th percentile shows that, in general, models perform better in summer than in spring over most of Europe (figure 3). Models over some grid-cells in west Europe (e.g., UK) show the poorest performance in spring, while in some grid-cells over southwest Europe (e.g., Spain) a decreasing performance in summer is found. The best model performance is observed in central and northwest Europe, particularly in the warmer months. Additional analysis about model performance can be found in the supplementary material (section 3). Moreover, our results confirm the role of Tx, which is the first driver of ozone extreme values in central and northwest Europe in summer (see supplementary material, figure S1).

Figure 4 depicts the performance of the logistic models regression for the threshold 50 ppb. In general, models over south Europe perform better in spring than in summer, specifically in some regions such as Spain, North Italy, or South Balkan. However, the best performance is shown in central and northwest Europe, particularly in summer. Additional measurements of the goodness of the models have been analysed (supplementary material, section 3). The influence of LO3 is mainly noticed in south and northeast Europe (figure 5). However, there are some dominant meteorological drivers of ozone exceedances above 50 ppb: Tx, SR and RH. SR and RH are dominant in spring, while Tx becomes a significant driver of extreme ozone values in summer, especially in central, northwest Europe, and also in some specific southern locations. Both parameters show up as positive drivers of ozone extremes, though the influence of Tx is slightly higher in most of the grid-cells. These results show a seasonal and regional variation of drivers of extreme ozone conditions, which are mainly dominated by local meteorological parameters (i.e., RH, SR and Tx) in some specific regions (e.g., northwest and central Europe).

4. Summary and conclusions

This study investigates the role of synoptic and local meteorological variability as a driver of surface concentrations of ozone, a toxic air pollutant. Additionally, by using a novel implementation of the JC97 classification, we are able to assess the effect of atmospheric circulation on a gridded ozone dataset. Three different regression models are employed to determine the drivers of maximum daily 8 h average ozone concentrations, as well as their extreme values as represented by their 95th percentiles, and exceedances of air quality guideline thresholds.

The drivers of surface ozone are identified during the model development using screening procedures that sequentially remove less significant drivers. The performance of the models is generally better in summer than spring. Geographically, the best performance is found in central and northwestern Europe (e.g., France, Belgium, Netherlands, Germany, Poland, Czech Republic, Austria or Switzerland). The inclusion of a one-day lag of ozone provides an additive value for predictions. Our results show that incorporating ozone persistence is particularly relevant in the southeast of Europe, especially in the Balkan

Table 3. Frequency (%) of selection of predictors in the QR models developed in MAM and in JJA for the 95th over all grid points.

QR	N° Models	Season	LO3	Tx	RH	Gh	SR	ST	Ws	TC	WF	SF	TF	VW	VS	D
	969	MAM	100	38.2	88.1	70.1	93.6	83.4	70.1	71.9	70.8	80.6	71.1	70.8	68.0	61.8
	969	JJA	100	89.2	79.9	66.9	78.0	77.4	75.4	65.2	77.3	82.7	65.6	70.7	71.1	65.0

Table 4. Frequency (%) of appearance of predictors in the LR models developed in MAM and in JJA for the selected threshold exceedances (50 ppb) over all grid-points.

LR_{ex50}	N° Models	Season	LO3	Tx	RH	Gh	SR	ST	Ws	TC	WF	SF	TF	VW	VS	D
	777	MAM	100	27.5	75.4	34.7	78.8	70.4	63.3	40.4	53.5	77.1	32.9	42.5	37.6	29.1
	530	JJA	100	81.5	59.8	38.9	47.2	74.7	76.6	35.3	67.0	63.6	42.6	40.2	57.4	37.0

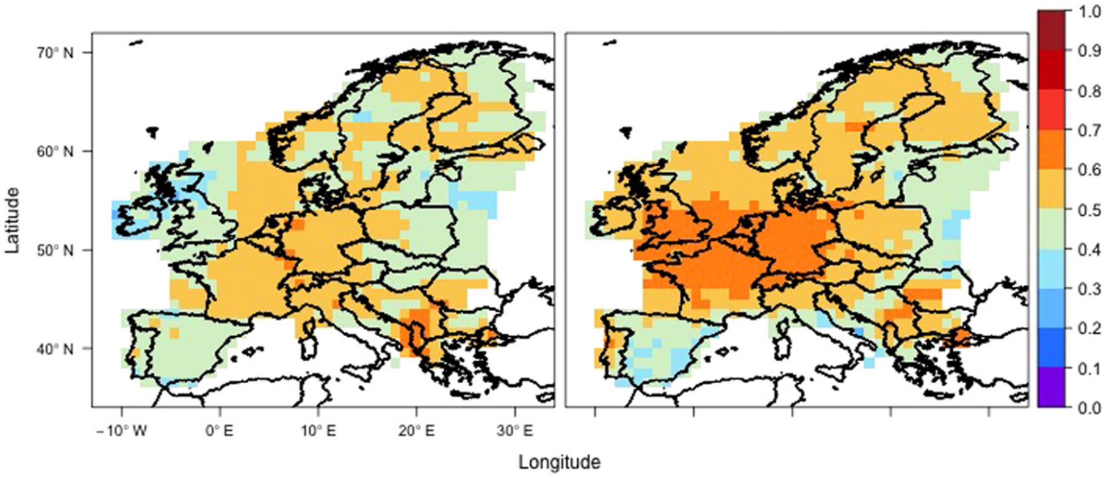

Figure 3. Spatial distribution of R^2 in QR at the upper percentile 95th for MAM (left) and JJA (right).

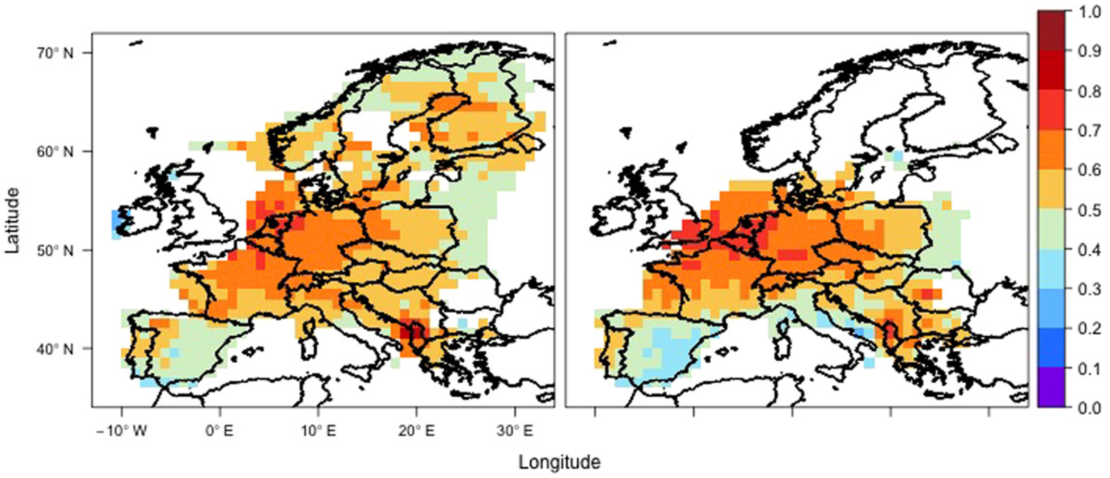

Figure 4. Spatial distribution of R^2 in LR when analysing the limited value of 50 ppb, in MMA (left) and JJA (right).

Peninsula. However, we find that meteorological drivers account for most of the explained variance of ozone in most of the grid cells over central and northwest Europe.

One of the main drivers of ozone is the daily maximum temperature, which shows a positive relationship with ozone. We identify some specific areas where ozone is particularly sensitive to maximum temperature in summer (i.e. central and northwest Europe), which we suggest could be due to the effect of temperature on emissions of VOCs in this region which previous studies have been shown to be in a VOC-limited chemical regime. Maximum temperature becomes a key driver when ozone exceeds air quality target values (50 and 60 ppb). There is also considerable regional variation of the effect of maximum temperature: in southern and northern Europe, maximum temperature also appears as a driver but with a smaller effect. Relative humidity and solar radiation, negatively and positively related to ozone, respectively, appear as other relevant drivers, particularly in spring. Our results reveal some influence of the airflow indices on ozone in specific grid-cells, which suggests that the effect of wind speed and direction plays a role in influencing surface ozone concentration only in a small number of locations in Europe.

In conclusion, this statistical analysis provides insights into the strongest meteorological drivers of ozone, which play a significant role during the warmer months. Climate change is expected to influence regional weather conditions, such as warmer temperatures or stagnant conditions, and an increase in heatwaves (Russo *et al* 2015), which will likely adversely affect ozone levels and, consequently, air quality in Europe. With the regression models developed, we are able to identify regions, which may be particularly vulnerable to increased episodes of high ozone in the future, and where special attention should be paid to mitigation strategies. Our results imply that central Europe may be especially vulnerable to such increased episodes of high ozone in the future.

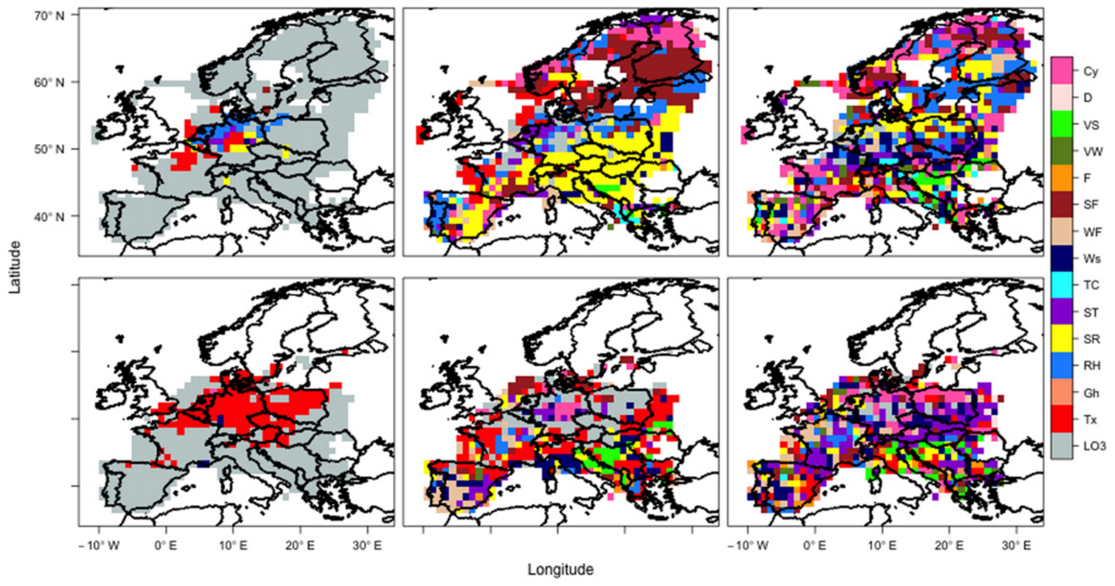

Figure 5. Spatial distribution of the most important driver (left), second most important (middle) and third most important (right) of ozone in LR for ozone exceedances (50 ppb) for both seasons: MAM (top) and JJA (bottom).

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References

Agresti A 2007 *An Introduction to Categorical Data Analysis* (Hoboken, NJ: Wiley)

Akaike H 1974 A new look at the statistical model identification *IEEE Trans. Autom. Control* **19** 716–23

Andersson C and Engardt M 2010 European ozone in a future climate: importance of changes in dry deposition and isoprene emissions *J. Geophys. Res.-Atmos.* **115** D02303

Banja M, Papanastasiou D, Poupkou A and Melas D 2012 Development of a Short-term ozone prediction tool in Tirana area based on meteorological variables *Atmos. Pollut. Res.* **3** 32–8

Barrero M A, Grimalt J O and Canton L 2005 Prediction of daily ozone concentration maxima in the urban atmosphere *Chemometr. Intell. Lab. Syst.* **80** 67–76

Baur D, Saisana M and Schulze N 2004 Modelling the effects of meteorological variables on ozone concentration-a quantile regression approach *Atmos. Environ.* **38** 4689– 4699

Beekmann M and Vautard R 2010 A modelling study of photochemical regimes over Europe: robustness and variability *Atmos. Chem. Phys. Disc.* **10** 10067–84

Bloomfield P J, Royle J A, Steinberg L J and Yang Q 1996 Accounting for meteorological effects in measuring urban ozone levels and trends *Atmos. Environ.* **30** 3067–77

Booker F L, Muntifering R, McGrath M, Burkey K, Decoteau D, Fiscus E, Manning W, Krupa S, Chappelka A and Grantz D 2009 The ozone component of global change: potential effects on agricultural and horticultural plant yield, product quality and interactions with invasive species *J. Integrative Plant Biol.* **51** 337e351

Camalier L, Cox W and Dolwick P 2007 The effects of meteorology on ozone in urban areas and their use in assessing ozone trends *Atmos. Environ.* **41** 7127–37

Chaloulakou A, Saisana M and Spyrellis N 2003 Comparative assessment of neural networks and regression models for forecasting summertime ozone in Athens *Science of the Total Environment* **313** 1–13

Cheng C S Q, Campbell M, Li Q, Li G L, Auld H, Day N, Pengelly D, Gingrich S and Yap D 2007a A synoptic climatological approach to assess climatic impact on air quality in South-central Canada: I. Historical analysis *Water Air and Soil Pollution* **182** 131–48

Cheng C S Q, Campbell M, Li Q, Li G L, Auld H, Day N, Pengelly D, Gingrich S and Yap D 2007b A synoptic climatological approach to assess climatic impact on air quality in South-central Canada: II. Future estimates *Water Air and Soil Pollution* **182** 117–30

Comrie A C 1997 Comparing neural networks and regression models for ozone forecasting *J. Air Waste Manage. Assoc.* **47** 653–63

Comrie A C and Yarnal B 1992 Relationships between synoptic-scale atmospheric circulation and ozone concentrations in metropolitan Pittsburgh, Pennsylvania *Atmos. Environ.* **26** 301–12

Conway D, Wilby R L and Jones P D 1996 Precipitation and air flow indices over the British Isles *Clim. Res.* **7** 169–83

Davies T D, Kelly P M, Low P S and Pierce C E 1992a Surface ozone concentrations in europe—links with the regional-scale atmospheric circulation *J. Geophys. Res.-Atmos.* **97** 9819–32

Davies T D, Farmer G, Kelly P M, Glover G M, Apsimon H M and Barthelmie R J 1992b Surface pressure pattern indicators of mean monthly pollutant concentrations in southern scandinavian precipitation—a test using case studies of months with high and low concentrations of nonmarine sulfate and nitrate *Atmospheric Environment Part A-General Topics* **26** 261–78

Dee D P *et al* 2011 The ERA-Interim reanalysis: configuration and performance of the data assimilation system *Quart. J. R. Meteorol. Soc.* **137** 553–97

Demuzere M, Trigo R M, Vila-Guerau de Arellano J and van Lipzig N P M 2009 The impact of weather and atmospheric circulation on O_3 and PM_{10} levels at a rural mid-latitude site *Atmos. Chem. Phys.* **9** 2695e2714

Demuzere M and van Lipzig N P M 2010a A new method to assess air quality levels using a synoptic-regression approach: I. present analysis for O_3 and PM_{10} *Atmos. Environ.* **44** 1341–55

Demuzere M and van Lipzig N P M 2010b A new method to assess air quality levels using a synoptic-regression approach: II. Future O3 concentrations *Atmos. Environ.* **44** 1356–66

Dueñas C, Fernández M C, Cañete S, Carretero J and Liger E 2002 Assessment of ozone variations and meteorological effects in an urban area in the Mediterranean Coast *Sci Total Environ* **299** 97–113

Fang Y, Naik V, Horowitz L W and Mauzerall D L 2013 Air pollution and associated human mortality: the role of air pollutant emissions, climate change and methane concentration increases from the preindustrial period to present *Atmos. Chem. Phys.* **13** 1377–94

Gardner M and Dorling S 2000 Statistical surface ozone models: an improved methodology to account for non-linear behaviour *Atmos. Environ.* **34** 21–34

Goodess C M and Jones P D 2002 Links between circulation and changes in the characteristics of Iberian rainfall *Int. J. Climatol.* **22** 1593–615

Grimalt M, Tomàs M, Alomar G, Martin-Vide J and Moreno-García M C 2013 Determination of the Jenkinson and Collison's weather types for the western Mediterranean basin over the 1948–2009 period *Temporal analysis. Atmósfera* **26** 75–94

Grivas G and Chaloulakou A 2006 Artificial neural network models for prediction of PM10 hourly concentrations, in the Greater Area of Athens, Greece *Atmos. Environ.* **40** 7-12161229

Grömping U 2007 Estimators of relative importance in linear regression based on variance decomposition *Am. Stat.* **61** 139–47

Harrel F, Lee K L, Matchar D B and Reichert T A 1985 Regression models for prognostic prediction: Advantages, problems and suggested solutions *Cancer Treatment Reports* **69** 1071–7

Hegarty J, Mao H and Talbot R 2007 Synoptic controls on summertime surface ozone in the northeastern United States *J. Geophys. Res.* **112** D14306

Holloway T, Fiore A and Galanter Hastings M 2003 Intercontinental transport of air pollution: will emerging science lead to a new hemispheric treaty? *Environ. Sci. Technol.* **37** 4535–42

Jacob D J and Winner D A 2009 Effect of climate change on air quality *Atmos. Environ.* **43** 51–63

Jenkinson A F and Collison F P 1977 An initial climatology of gales over the North Sea *Synoptic Climatology Branch Memorandum* **62** 18, UK Met Office

Jones P D, Hulme M and Briffa K R 1993 A comparison of Lamb circulation types with an objective classification scheme *Int. J. Climatol.* **13** 655–63

Koenker R and Basset G 1978 Regression quantiles *Econometrica* **46** 33–50

Kutner M H, Nachtsheim C J and Neter J 2004 Applied Linear Regression Models 4th Ed (Boston, MA: McGraw-Hill Irwin)

Lamb H H 1972 British Isles weather types and a register of daily sequence of circulation patterns *Geophysical Memoir* vol 116 (London: HMSO) pp 1861–971

Lindeman R H, Merenda P F and Gold R Z 1980 *Introduction to Bivariate and Multivariate Analysis* (Glenview: Scott, Foresman)

Linderson M L 2001 Objective classification of atmospheric circulation over southern Scandinavia *Int. J. Climatol.* **21** 155–69

Maindonald J and Braun J 2006 *Data Analysis and Graphics Using R An example-based approach* (Cambridge: Cambridge University Press)

Maraun D, Rust H W and Osborn T J 2011 The Influence of synoptic airflow on UK daily precipitation extremes: I. observed spatio-temporal relations *Clim. Dyn.* **36** 261–75

Maraun D, Osborn T J and Rust H W 2012 The influence of synoptic airflow on UK daily precipitation extremes: II. regional climate model and EOBS data validation *Clim. Dyn.* **39** 287–301

Mata J and Machado J 1996 Firm start-up size: a conditional quantile approach *European Economic Review* **40** 1305–23

McCullagh P and Nelder J A 1989 Generalized linear models *Monographs on Statistics and Applied Probability* vol 37 (London: Chapman and Hall) 2 edn

Munir S, Chen H and Ropkins K 2012 Modelling the impact of road traffic on ground level ozone concentration using a quantile regression approach *Atmos. Environ.* **60** 283–91

Nelder J A and Wedderburn R W M 1972 Generalized linear models *J. R Stat. Soc. Ser.* A **135** 370–84

Ordóñez C, Mathis H, Furger M, Henne S, Hüglin C, Staehelin J and Prévôt A S H 2005 Changes of daily surface ozone maxima in Switzerland in all seasons from 1992 to 2002 and discussion of summer 2003 *Atmos. Chem. Phys.* **5** 1187–203

Pearce J, Beringer J, Nicholls N, Hyndman R J, Uotila P and Tapper N J 2011 Investigating the influence of synoptic-scale meteorology on air quality using self-organizing maps and generalized additive modeling *Atmos. Environ.* **45** 128e–136

Peduzzi P, Concato J, Kemper E, Holford T R and Feinstein A R 1996 A simulation study of the number of events per variable in logistic regression analysis *J. Clinical Epidemiology* **49** 1372–9

Pérez P, Trier A and Reyes J 2000 Prediction of PM2.5 concentrations several hours in advance using neural networks in Santiago, Chile *Atmos. Environ.* **34** 1189–96

Porter W C, Heald C L, Cooley D and Russell B 2015 Investigating the observed sensitivities of air quality extremes to meteorological drivers via quantile regression *Atmos. Chem. Phys. Discuss.* **15** 10349–66 2015

Pusede S E *et al* 2014 On the temperature dependence of organic reactivity, nitrogen oxides, ozone production, and the impact of emission controls in San Joaquin Valley, California *Atmos. Chem. Phys.* **14** 3373–95

Robeson S M and Steyn D G 1990 Evaluation and comparison of statistical forecast models for daily maximum ozone concentrations *Atmospheric Environment* B **24** 303–12

Russo S, Sillmann J and Fischer E 2015 Top ten European heatwaves since 1950 and their occurrence in the coming decades *Environ. Res. Lett.* **10**

Rust H, Maraun D and Osborn T 2009 Modelling seasonality in extreme precipitation *Eur. Phys. J. Special Topics* **174** 99–111

Rust H, Vrac M, Sultan B and Lengaigne M 2013 Mapping weather type influence on Senegal precipitation based on a spatio-temporal statistical model *J. Climate* **26** 8189–209

Saavedra S, Rodríguez A, Taboada J J, Souto J A and Casares J J 2012 Synoptic patterns and air mass transport during ozone episodes in northwestern Iberia *Science of the Total Environment* **441** 97e110

Schnell J L *et al* 2015 Use of North American and European air quality networks to evaluate global chemistry-climate modeling of surface ozone *Atmos. Chem. Phys. Discuss.* **15** 11369–407

Sillman S and Samson P 1995 Impact of temperature on oxidant photochemistry in urban, polluted rural and remote environments *J. Geophys. Res.* **100** 497e508

Smith R L, Davis J M, Sacks J, Speckman P and Styer P 2000 Regression models for air pollution and daily mortality: analysis of data from Birmingham, Alabama *Environmetrics* **11** 719–43

Sousa S I V, Pires J C M, Mrtins F G, Pereira M C and Alvim-Ferraz M C M 2008 Potentialities of quantile regression to predict ozone concentrations *Environmetrics* **20** 147–58

Spellman G 2000 The application of an objective weather-typing system to the Iberian peninsula *Weather* **55** 375–85

Tang L, Rayner D P and Haeger-Eugensson M 2011 Have meteorological conditions reduced NO2 concentrations from local emission sources in gothenburg? *Water Air and Soil Pollution* **221** 275–86

Thompson M L, Reynolds J, Cox L H, Guttorp P and Sampson P D 2001 A review of statistical methods for the meteorological adjustment of tropospheric ozone *Atmos. Environ.* **35** 617–30

Tomás C, de Pablo F and Rivas Soriano L 2004 Circulation weather types and cloud- to-ground flash density over the Iberian Peninsula *Int. J. Climatol.* **24** 109e123

Trigo R M and DaCamara C C 2000 Circulation weather types and their impact on the precipitation regime in Portugal *Int. J. Climatol.* **20** 1559e 1581

World Health Organization WHO 2014c 2014 Air quality and health *Fact Sheet* **313** (http://who.int/mediacentre/factsheets/fs313/en/)

Wood S N 2006 *Generalized Additive Models: An Introduction with R* vol 66 (Boca Raton, FL: CRC)

Ziomas I C, Melas D, Zerefos C S, Bais A F and Paliatsos A 1995 On the relationship between peak ozone levels and meteorological variables *Fresenius Environmental Bulletin* **4** 53–8

Synoptic and meteorological drivers of extreme ozone concentrations over Europe: Supplementary Data

N Otero[1], J Sillmann[2], J L Schnell[3], H W Rust[4] and T Butler[1]

[1]Institute for Advanced Sustainability Studies e.V, Potsdam, Germany

[2]CICERO Center for International Climate and Environmental Research-Oslo, Norway

[3] Department of Earth System Science, University of California, Irvine, CA, USA

[4] Freie Universität Berlin, Institut für Meteorologie, Berlin, Germany

This is the supplementary data to the research paper "Statistical approach for assessing the influence of synoptic and meteorological conditions on surface ozone concentrations over Europe" and it provides further information about additional results obtained.

SD1. Drivers of ozone extreme conditions:

The analysis of the sensitivities of ozone extremes to the different individual predictors allows us to identify the main drivers of high ozone levels. Results of the predictor's importance show that drivers of high percentile of ozone are mainly dominated by local meteorological conditions, specifically, RH and SR in spring and Tx in summer (figure S1). SR and Tx are positively correlated with ozone, whereas RH has a negative effect on surface ozone. Tx might be considered as one of the most important driver of ozone extreme values in JJA in many places over Europe, especially over Central and North-West Europe (bottom of figure S1). This result is consistent with a recent quantile regression analysis performed over the US that showed the dominance of the temperature in the summertime (Porter *el al* 2015). Overall, the airflow indices WF, SF and VS also play a role in many grid-cells. WF and VS are in general negatively related to ozone, while SF shows a change of sign that is dependent on the grid-cell. Our findings indicate that Tx might be considered the main driver of ozone extreme values in JJA, but also in some specific grid-cells high ozone levels would be influenced by the prevailing atmospheric conditions.

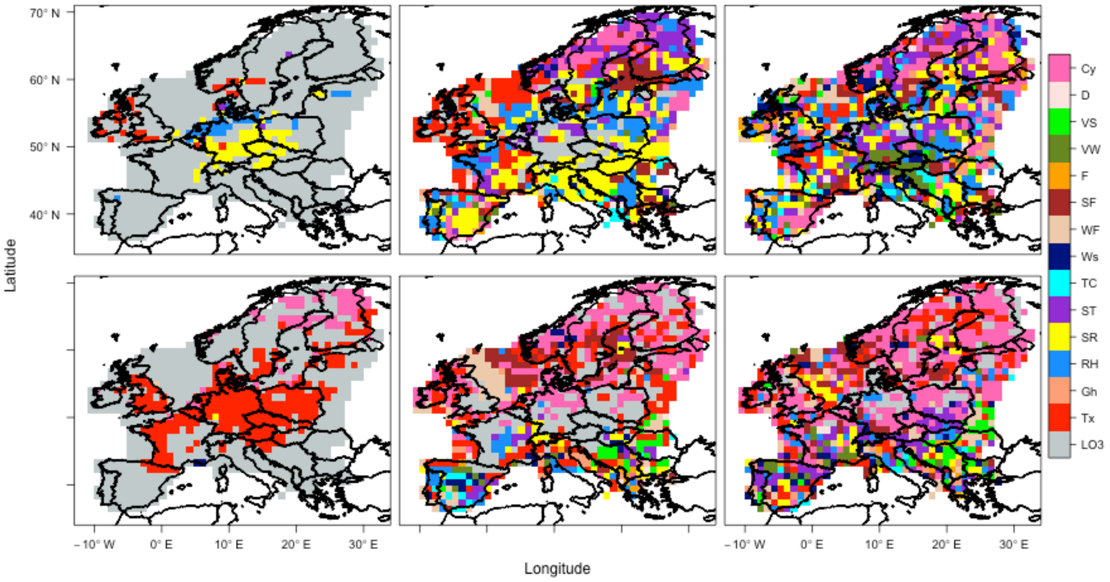


Figure S1. Spatial distribution of the most important driver (left), second most important (middle) and third most important (right) of ozone at the highest percentile (95th) in both seasons: MAM (top) and JJA (bottom).

SD2. Logistic regression:

For surface ozone a daily maximum 8-hour average threshold is specified (120 μg/m3) in the European union (EU) 2008 directive (EU, 2008c). Additional logistic regression analysis using this limit (~60ppb) and a lower one (55ppb) provide further information about drivers of ozone exceedances. In those cases, a different number of models are developed depending on the number of event of ozone exceedances. For example, in spring only a few cell-grids register more than 100 days with ozone levels above 60 ppb, which leads a total of 31 models developed, while in summer there is an increasing number of grid-cells with more episodes of high levels of ozone. The numbers of models increases when using the threshold of 55ppb (table S1).

The frequency of appearance of predictors (table S1) shows that SR, Ws, SR and RH are the most often selected predictors in spring, while Ws, Tx, and ST are more used in summer when the threshold of 55ppb is used. The frequency of appearance when the threshold is higher shows that SR, Ws and ST are most often selected predictors in spring, and Ws, Tx and ST in summer. The main difference between these frequencies is the decrease of the appearance of the RH in spring for the highest threshold. This could be explained due to the reduced number of models in spring, especially in North-West Europe where RH plays an important role.

Overall, D is the variable with lowest frequency of appearance in summer, whereas in spring Tx. This result indicates that Tx during the spring season is not a relevant variable for modelling ozone exceedances, which is in agreement with the results obtained for the logistic regression by applying lowest limit value (50ppb) and the QR analysis.

Figures S2, S3 depict the performance of the logistic models regression when applying the thresholds 55 and 60 ppb. The best model's performance is observed in summer in those models developed over

North-West and Central Europe (e.g., France, Belgium, Netherlands, Austria or Germany. On the contrary, the poorest performance is shown over South Europe, in particular over the Iberian Peninsula.

The analysis of the three most important predictors reveals that LO3 has a significant effect over South-East Europe (figures S4, S5). The results from the LR models that uses 55ppb as a threshold show that RH is a key driver of the ozone exceendances in some grid-cells over North-West and Central Europe, and SR dominates in some regions over Central and South-Europe, in spring. The analysis of the most important drivers in summer, when using both thresholds, point out Tx is a key driver over Central and North-West Europe (e.g. north of France, Belgium, Netherlands, Germany, Austria or Poland), which is in agreement with our previous results.

Additionally, and according to the results obtained from the LR using different thresholds, the top drivers of ozone exceendances are summarised in tables S3, S4 for spring and summer, respectively.

Table S1. Frequency (%) of appearance of predictors in the LR models developed in MAM and in JJA for the selected threshold exceedances (50ppb) over all grid-points.

LR_{ex55}	Nº Models	Season	LO3	Tx	RH	Gh	SR	ST	Ws	TC	WF	SF	TF	VW	VS	D
	318	MAM	100	17.9	67.0	34.6	81.8	70.8	74.5	43.5	54.4	63.2	31.4	39.3	32.4	23.3
	417	JJA	100	76.0	58.8	45.8	42.2	72.7	80.1	39.8	64.0	60.9	45.6	37.9	40.8	35.0

Table S2. Frequency (%) of appearance of predictors in the models developed in MAM and in JJA, LR_{LO3} and LR for the selected threshold exceedances (60ppb) over all grid-points.

LR_{ex60}	NºModels	Season	Lo3	Tx	RH	Gh	SR	ST	Ws	TC	WF	SF	TF	VW	VS	D
	31	MAM	100	29	48.4	51.6	74.2	61.3	67.7	51.6	29.0	51.6	9.7	38.7	29.0	35.5
	276	JJA	100	71.4	55.8	44.2	37.7	63.8	81.5	41.3	58.0	55.1	46.0	37.3	38.0	36.6

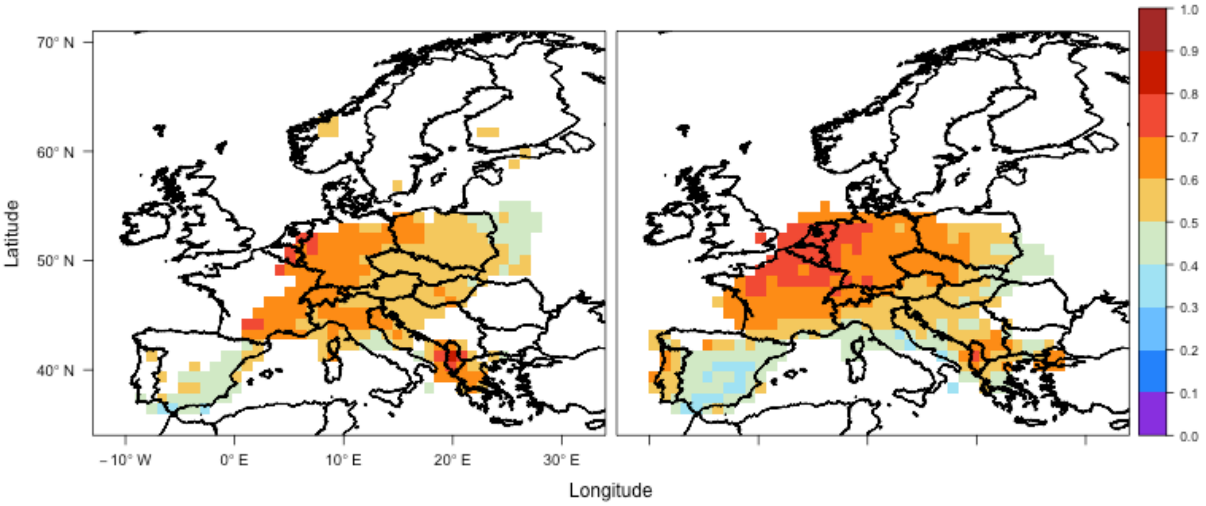


Figure S2. Spatial distribution of R^2 in the LR models when analysing the limited value of 55 ppb, in MMA (left) and JJA (right).

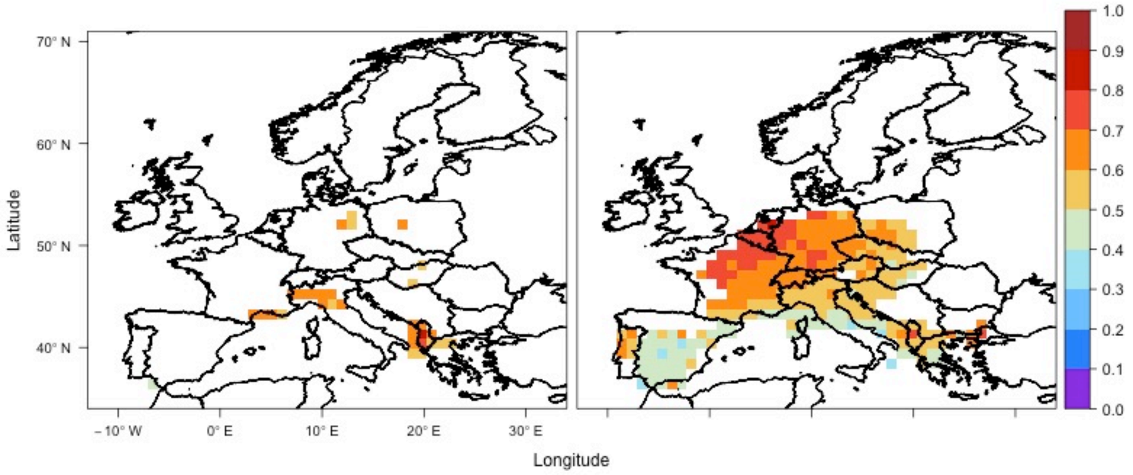


Figure S3. Spatial distribution of R^2 in the LR models when analysing the limited value of 60 ppb, in MMA (left) and JJA (right).

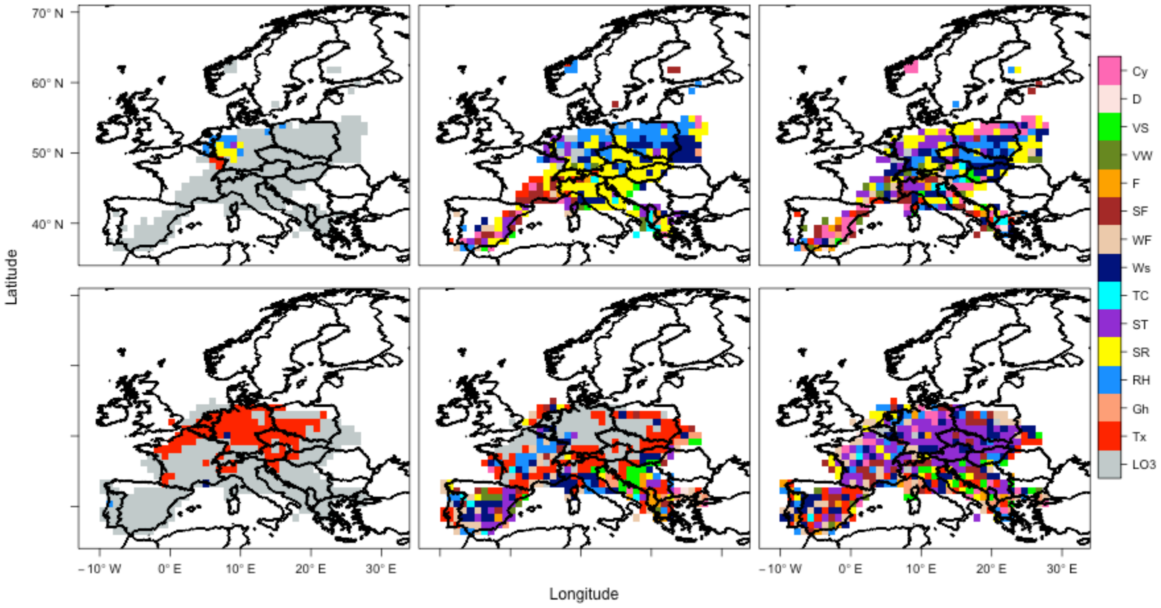


Figure S4. Spatial distribution of the most important driver (left), second most important (middle) and third most important (right) of ozone in LR for ozone exceedances (55ppb) for both seasons: MAM (top) and JJA (bottom).

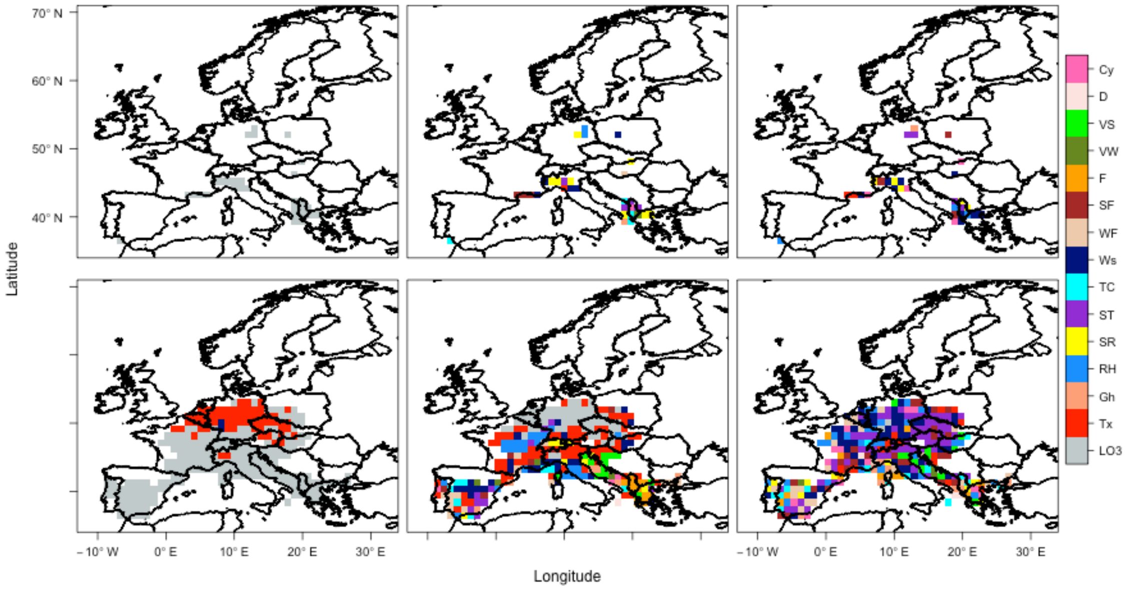


Figure S5. Spatial distribution of the most important driver (left), second most important (middle) and third most important (right) of ozone in LR for ozone exceedances (60ppb) for both seasons: MAM (top) and JJA (bottom).

Table S3. Frequency (%) of appearance of the top drivers of ozone exceedances in spring. The bold numbers in brackets indicate the number of models developed for each threshold.

Predictor	ex50 (**777**)	ex55 (**318**)	ex60 (**31**)
LO3	90.2	93.4	100
RH	3.6	3.5	
Tx	3.6	0.9	
SR	1.4	1.9	
ST	0.8	0.3	
SF	0.3		
Ws	0.1		

Table S4. Frequency (%) of appearance of the top drivers of ozone exceedances in summer. The bold numbers in brackets indicate the number of models developed for each threshold.

Predictor	ex50 (**530**)	ex55 (**417**)	ex60 (**276**)
LO3	68.5	72.7	76.8
RH		0.2	
Tx	30.0	26.6	22.5
SR			
ST			
SF	0.6		
Ws	0.7	0.5	0.7
Cy	0.1		

SD3. Analysis of mean bias:

In order to obtain a more robust measure of the model performance, we have estimated the mean bias error (MB), which can be a useful metric to determine whether predictions are over or under estimated:

$$\mathrm{MB} = \frac{1}{N} \sum f_{i - o_i} \qquad (1)$$

where f_i are the modelled and O_i are de observed values.

The mean bias has been estimated for MLR (Multiple Linear Regression) and QR (Quantile Regression) models for both spring and summer seasons. By definition, MB for QR is assessed as a local measurement at the 95th percentile. Given that LR (Logistic Regression) models estimate probabilities of success of the occurrence of ozone exceedances, in this statistical procedure we have estimated the Brier Score (BS):

$$\mathrm{BS} = \frac{1}{N} \sum (p_i - o_i)^2 \qquad (2)$$

where, p_i is the predicted probability to have the true outcome o_i (0 non-exceedance and 1 exceedance). Therefore, the BS defines the expected squared difference of the predicted probability and the response variable. The lower the Brier score of a model the better the predictive performance.

To do this, the data have been split into two subsets: 2/3 of the data used for the model training and 1/3 of the data used for validating the model through the metric proposed.

Overall, the analysis of the mean bias for MLR (figure S6) show a positive bias over most of Europe with higher values in summer, especially over East Europe. This indicates that models are in general over-estimating the observed values. In spring, the results from the MB show that models over North-West and South-East Europe under-estimate the observational dataset, while in Central and North-East Europe over-estimate ozone observations.

In the case of QR, there is a positive MB Europe (figure S7), which indicate that models over-estimate observational dataset. The highest MB is observed in spring, especially over North-West Europe and South-West (e.g. UK or Iberian Peninsula). In general a decreasing MB is observed in summer, in particular over Central Europe.

The analysis of the Brier Score in the LR models shows that the lowest values are found in North and Central Europe in spring, and in North-West and Central Europe in summer, while higher BS are observed in South Europe (figure S8). That suggests that the predictive performance of logit models is better over North-West and Central Europe

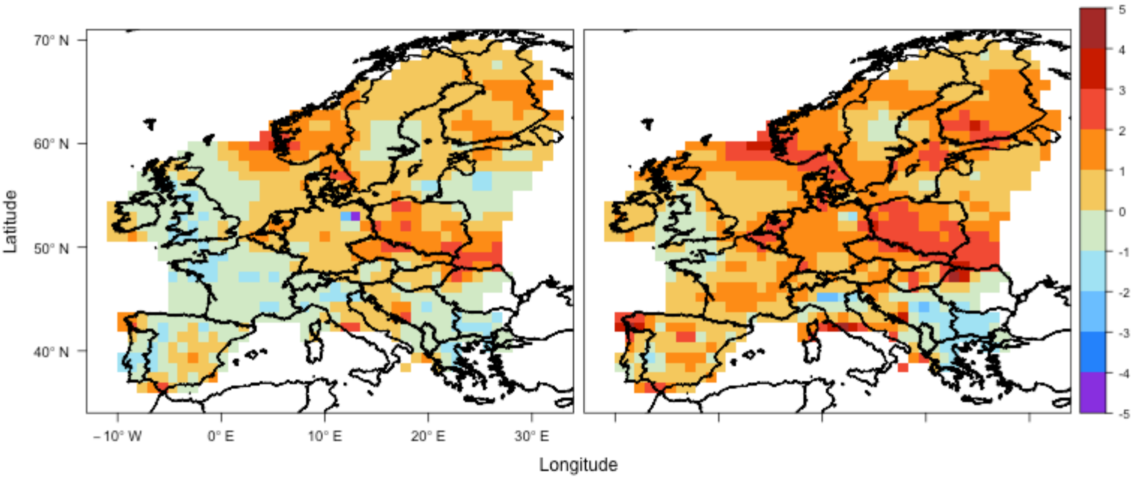


Figure S6. Mean bias(ppb) between modelled and observed values for MLR models in spring (left) and summer (right) seasons.

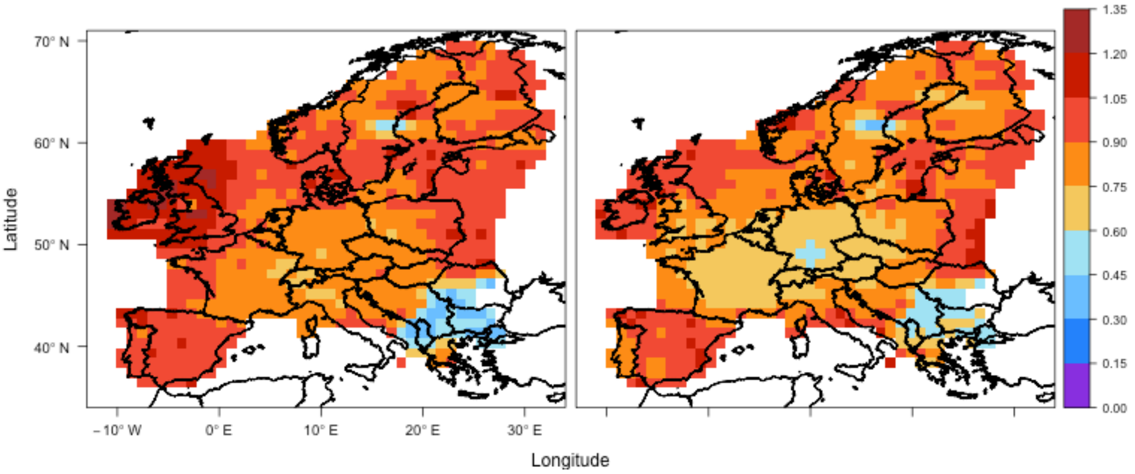


Figure S7. Mean bias(ppb) between modelled and observed values for QR models in spring (left) and summer (right) seasons.

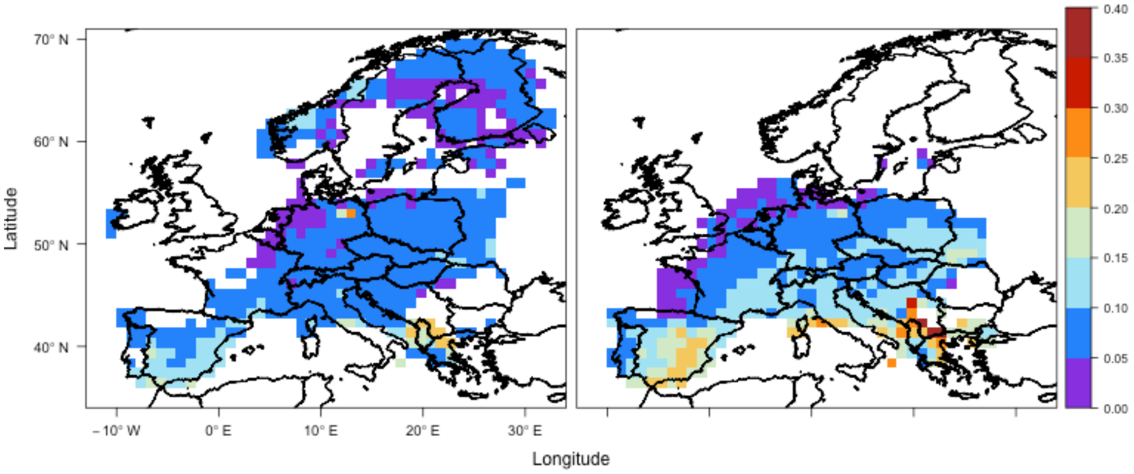


Figure S8. Brier Score for logistic regression for the threshold 55ppb in spring (left) and summer (right) seasons.

References:

EU, 2008c, Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe (OJ L 152, 11.6.2008, pp. 1–44). (http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:152:000 1:0044:EN:PDF) accessed 15 July 2014.

Porter W C, Heald C L, Cooley D, and Russell B 2015 Investigating the observed sensitivities of air quality extremes to meteorological drivers via quantile regression, *Atmos. Chem. Phys. Discuss.* **15** 14075-14109 doi:10.5194/acpd-15-14075-2015

Chapter 5

Paper III: A multi-model comparison of meteorological drivers of surface ozone over Europe

Submitted to Atmospheric Chemistry and Physics (ACP): Noelia Otero, Jana Sillmann, Kathleen A. Mar, Henning W. Rust, Sverre Solberg, Camilla Andersson, Magnuz Engardt, Robert Bergström,Bertrand Bessagnet, Augustin Colette, Florian Couvidat, Cournelius Cuvelier, Svetlana Tsyro, Hilde Fagerli, Martijn Schaap, Astrid Manders, Mihaela Mircea, Gino Briganti, Andrea Cappelletti, Mario Adani, Massimo D'Isidoro, María-Teresa Pay, Mark Theobald, Marta G. Vivanco, Peter Wind, Narendra Ojha14, Valentin Raffort and Tim Butler

A multi-model comparison of meteorological drivers of surface ozone over Europe

Noelia Otero[1,3], Jana Sillmann[2], Kathleen A. Mar[1], Henning W. Rust[3], Sverre Solberg[4], Camilla Andersson[5], Magnuz Engardt[5], Robert Bergström[5],Bertrand Bessagnet[6], Augustin Colette[6], Florian Couvidat[6], Cournelius Cuvelier[7], Svetlana Tsyro[8] , Hilde Fagerli[8], Martijn Schaap[9,3], Astrid Manders[9], Mihaela Mircea[10], Gino Briganti[10], Andrea Cappelletti[10], Mario Adani[10], Massimo D'Isidoro[10], María-Teresa Pay[11], Mark Theobald[12], Marta G. Vivanco[12] , Peter Wind[8,13], Narendra Ojha[14], Valentin Raffort[15] and Tim Butler[1,3]

[1]Institute for Advanced Sustainability Studies e.V., Potsdam, Germany

[2]CICERO Center for International Climate Research, Oslo, Norway

[3]Freie Universität Berlin, Institut für Meteorologie, Berlin, Germany

[4]Norwegian Institute for Air Research (NILU), Box 100, 2027 Kjeller, Norway
[5]SMHI, Swedish Meteorological and Hydrological Institute Norrköping, Norrköping, Sweden

[6]INERIS, Institut National de l'Environnement Industriel et des Risques, Verneuil en Halatte, France
[7]European Commission, Joint Research Centre (JRC), Ispra, Italy
[8]MET Norway, Norwegian Meteorological Institute, Oslo, Norway
[9]TNO, Netherlands Institute for Applied Scientific Research, Utrecht, the Netherlands
[10]ENE-National Agency for New Technologies, Energy and Sustainable Economic Development, Bologna, Italy
[11]Barcelona Supercomputing Center, Centro Nacional de Supercomputación, Jordi Girona, 29, 08034 Barcelona, Spain
[12]CIEMAT, Atmospheric Pollution Unit, Avda. Complutense, 22, 28040 Madrid, Spain
[13]Faculty of Science and Technology, University of Tromsø, Tromsø, Norway
[14]Max-Planck-Institut für Chemie, Mainz, Germany
[15]CEREA, Joint Laboratory Ecole des Ponts ParisTech – EDF R&D, Champs-Sur-Marne, France

Abstract. The implementation of European emission abatement strategies has led to significant reduction in the emission of ozone precursors during the last decade. Ground level ozone is also influenced by meteorological factors such as temperature, which exhibit interannual variability, and are expected to change in the future. The impacts of climate change on air quality are usually investigated through air quality models that simulate interactions between emissions, meteorology and chemistry. Within a multi-model assessment, this study aims to better understand how air quality models represent the relationship between meteorological variables and surface ozone concentrations over Europe. A multiple linear regression (MLR) approach is applied to observed and modelled time series across ten European regions in springtime and summertime for the period of 2000-2010 for both models and observations. Overall, the air quality models are in better agreement with observations in summertime than in springtime, and particularly in certain regions, such as France, Mid-Europe or East-Europe, where local meteorological variables show a strong influence on surface ozone concentrations. Larger discrepancies are found for the southern regions, such as the Balkans, the Iberian Peninsula and the Mediterranean basin, especially in springtime. We show that the air quality models do not properly reproduce the sensitivity of surface ozone to some of the main meteorological drivers, such as maximum temperature, relative humidity and surface solar radiation. Specifically, all air quality models show more limitations to capture the strength of the relationship ozone-relative humidity detected in the observed time series in most of the regions, in both seasons. Here, we speculate that dry deposition schemes in the air quality models might play an essential role to capture this relationship. We further quantify the relationship between ozone and maximum temperature (m_{o3-T}, climate penalty) in observations and air quality models. In summertime, most of the air quality models are able to reproduce reasonably well the observed climate penalty in certain regions such as France, Mid-Europe and North Italy. However, larger discrepancies are found in springtime, where air quality models tend to overestimate the magnitude of observed climate penalty.

1. Introduction

Tropospheric ozone is recognised as a threat to human health and ecosystem productivity (Mills et al. 2007). Moreover, ozone is an important greenhouse gas (IPCC, 2013). It is produced by photochemical oxidation of carbon monoxide and volatile organic compounds (VOCs) in the presence of nitrogen oxides (NOx=NO+NO2) (Jacob and Winner, 2009). While it is an important pollutant on a regional scale, due to the long-range transport effect it may also influence air quality on a hemispheric scale (Monks et al., 2015, Hedegaard et al, 2013). Moreover, its strong relationship with temperature represents a major concern, since under a changing climate the efforts on new air pollution mitigation strategies might be insufficient. This effect, referred as climate penalty (Wu et al., 2008), is expected to play an important role on future air quality (Hendriks et al. 2016). Therefore it is essential to better understand the potential implications of climate change on pollutant levels. In a comprehensive review of the existing literature about the robustness of climate penalty on Europe, Colette et al. (2015) concluded that the climate change might act against mitigation measures.

Previous studies have shown that the reduction of emissions of ozone precursors, NOx and VOCs, lead to a decrease in tropospheric ozone concentrations in Europe (Solberg et al. 2005, Jonson et al. 2006). However, there is also a large year-to-year variability due to weather conditions (Andersson et al. 2007). There is a strong correlation between ozone and temperature that has been associated with the temperature-dependent lifetime of peroxyacetyl nitrate (PAN), and also due to the temperature dependence of biogenic emission of isoprene (Sillman and Samson, 1995). Substantial increases in surface ozone have been associated with high temperatures and stable anticyclonic, sunny conditions that promote ozone formation (Solberg et al. 2008). Ozone peak concentrations are also affected by closing of the plants' stomata at very high temperatures (Hodnebrog et al. 2012). Several studies have assessed the model dependence of ozone on temperature (e.g. Steiner et al. 2006, Rasmussen et al. 2013). Recently, Coates et al. (2016) used a box model to investigate the influence of temperature and NOx on ozone production. Their analysis suggested that reductions in NOx would be required to offset additional ozone increase due to increasing temperatures under a warmer climate. An extensive review about the impacts of temperature on ozone production can be found in Pusede et al. (2015).

Previous studies have shown the importance of relative humidity on ozone pollution episodes (Camalier et al. 2007, Davies et al. 2011). Regional studies reported a negative relationship between ozone and relative humidity (Dueñas et al. 2002, Elminir 2005, Demuzere et al., 2009). Some authors attributed this negative correlation to the photolysis of ozone and subsequent loss of O1(D) to H2O (Jacob and Winner). High levels of humidity are usually related with enhanced cloud cover and thus reduced photochemistry (Dueñas et al. 2002, Camalier et al. 2007). Andersson and Engardt (2010) highlighted the importance of including meteorological dependence for dry deposition of ozone to vegetation, also incorporating soil moisture dependence. With a simple modelling approach, Kavassalis and Murphy (2017) found that the relationship ozone-relative humidity was well captured by the inclusion of the vapour pressure deficit-dependent dry deposition, indicating the relevance of detailed dry deposition schemes in the CTMs.

Increasing solar radiation leads to an increase of ozone, though with a weak effect (Dawson et al. 2007) and it has been suggested that it could reflect in part the association of clear sky with high temperatures (Ordónez et al., 2005). Then, changes in cloud cover can also affect the photochemistry of ozone production and loss (Jacob and Winner, 2009). Additionally, low wind speed is usually associated with high ozone pollution levels (Jacob and Winner, 2009).

The influence of climate change on ozone and its precursors can involve multiple processes (Colette et al, 2015). A common approach to study the impact of climate change on air quality requires the use of air quality models that aim to represent dynamic and chemical processes in the atmosphere. The relevance of climate change for future European air quality has been assessed in several studies that also reflect differences depending on the modelling system and future emissions scenarios adopted for each study (e.g. Lagner et al. 2005, Meleux et al. 2007, Anderson and Engardt, 2010).

Air quality models can be divided into two categories: offline chemistry transport models (CTMs) in which the model chemistry runs using meteorological data as input, and online models that allow coupling and integration of chemistry with some of the physical components to various degrees (Baklanov et al. 2014). Differences between offline and online modelling approaches can be fairly small or significant, depending on the level of the model complexity and simulated variables (Zhang, 2008). The large number and complex interactions between meteorology and chemistry in the atmosphere influence the ability of the model to represent observed situations (Kong et al. 2014). Due to assumptions, parametrizations and simplifications of processes, the models themselves are subject to large uncertainties (Manders et al. 2012), which have been reflected in some regional differences in the magnitude of surface ozone response to projected climate change (Andersson and Engardt, 2010). Thus, model biases when compared to observations still remain a concern, especially in terms of the response of air quality under future climate (Fiore et al. 2009, Rasmussen et al. 2012). Comparisons between model outputs and measurements of available observational dataset assess the reliability of air quality models, and they are essential to quantify the models ability to reproduce observations.

The EURODELTA project was initiated by the Task Force on Measurement and Modelling and the Joint Research Centre of the European Commission to provide a benchmark for the EMEP model in order to assess its relevance for policy support (Colette et al.2017a). These multi-model exercises contribute to further improving modelling techniques and understanding the associated uncertainties in the models performance. Previous exercises have evaluated the performance of chemistry transport models for future European air quality (e.g. van Lon et al. 2007, Thunis et al. 2008). Recently, Bessagnet et al. (2016) presented an intercomparison and evaluation of chemistry transport model performance with a joint analysis of some meteorological fields. They highlighted the limitations of models to simulate meteorological variables, such as wind speed and planetary boundary layer height. Particularly, in the case of ozone, they showed the importance of boundary conditions on model calculations. Within this framework, the ongoing Eurodelta-Trends (EDT) exercise (Colette et al. 2017a) builds upon this tradition and focuses on the context of air quality trends modelling. This exercise has been designed to better understand the evolution of air pollution and its drivers over the last two decades (1990-2010) by the use of state-of-the-art air quality models. The EDT project will allow the evaluation of the skill of regional air quality models and quantification of the role of the different key driving factors of surface ozone, such as emissions changes, long-range transport and meteorological variability. One of the main goals of the EDT project is to assess the efficiency of mitigation strategies for improving air quality (more details can be found in Colette et al. 2017a).

Quantification and isolation of the effects of meteorology on ozone is a challenge, due to the complex interrelation between ozone, meteorology, emissions and chemistry (Solberg et al. 2015). There is a large number of representative studies in the literature that have established the relationship between surface ozone concentrations and meteorological variables using statistical modelling techniques (e.g. Bloomfield et al. 1996, Chaloukau et al 2003, Barrero et al. 2005, Ordóñez et al., 2005, Camalier et al., 2007, Seo et al., 2014, Porter et al. 2015, Otero et al., 2016). Most of these works examined the impact of meteorology on ozone pollution levels through observational datasets. Only a few studies, to our knowledge, examined the statistical relationship between surface ozone and meteorological parameters from models.

Davis et al. (2011) developed regression models to analyse the observed and modelled relationship between meteorology and surface ozone across the Eastern of U.S. They found that the Community Multiscale Air Quality (CMAQ) model did not capture the effect of temperature and relative humidity on daily maximum 8-h ozone and it generally underestimated the observed sensitivities to both meteorological variables, especially in the northeast. Rasmussen et al. (2012) examined the ozone-temperature relationship in a coupled chemistry-climate model and they found that the model underestimated the effect of temperature on ozone over the Mid-Atlantic. Lemaire et al. (2016) proposed a combined statistical and deterministic approach to assess the air quality response to projected climate change. Based on a data set from a deterministic climate and chemistry models, they identified the two major drivers of surface ozone over eight European regions, selected from a set of potential predictors that reached the highest correlations with ozone. Afterwards they built statistical models consisting of generalized linear models, which could be used to predict air quality.

Given that meteorology plays an essential role for surface ozone concentrations, it might be a considerable source of uncertainties in model outputs. The present study, thus, aims to provide a simple method to examine the influence of meteorological variability on modelled surface ozone concentrations over Europe. Specifically, our analysis focuses on the ozone season (April to September) over the years 2000-2010. The choice of this period is mainly motivated by the availability of the observational dataset from Schnell et al. (2014, 2015) (see section 2.1). Within the EDT framework, a recent report has presented the main findings on the long-term evolution of air quality (Colette et al. 2017b). Part of these results was obtained from the analysis of the 1990s (1990-2000) and 2000s (2000-2010) separately. Consistently, we decided to focus on the second decade, for which the interpolated dataset of observed on maximum daily 8-hourly mean ozone (MDA8 O3) used in this study was available. Similarly to Otero et al. (2016), we apply a multiple linear regression approach to examine the meteorological influence MDA8 O3. Statistical models are developed separately for observational datasets and air quality models, with the primary focus on examining the relationship between MDA8 O3 and potential meteorological drivers in the air quality models and comparing these with the corresponding relationships determined from observed data. Therefore, this study offers a method of model evaluation capable of understanding the discrepancies between air quality models and observations in terms of representing the relationship to meteorological input variability.

The present paper is structured as follows. Section 2 describes the observational data as well as the air quality models studied here. The methodology and the design of the statistical models are introduced in section 3. Section 4 discusses the results and the summary and conclusions are discussed in section 5.

2. **Data**

2.1. **Observations**

This study uses gridded MDA8 O3 concentrations created with an objective-mapping algorithm developed by Schnell et al. (2014). They applied a new interpolation technique over hourly observations of stations from the European Monitoring and Evaluation Programme (EMEP) and the European Environment Agency's air quality database (AirBase) to calculate surface ozone averaged over 1° by 1° grid cells. Recently, Otero et al. (2016) used this dataset for examining the influence of synoptic and local meteorological conditions over Europe. This interpolated product offers a possibility to establish a direct comparison between observations and CTMs. However, it must be acknowledged that for some areas with a low number of stations (i.e. the southeastern or northeastern European regions) the values interpolated into the 1x1 degree grid cells may not be representative of such large scales. A complete description of this process can be found in Schnell et al. (2014, 2015). The gridded dataset covers a total of 15-years (1998-2012), but here we use a common period of 11-years for both observations and CTMs (2000-2010).

This study investigates the observed influence of meteorological variables on MDA8 O3, based on the ERA-Interim reanalysis product provided by the European Centre for Medium-Range Weather Forecasts (ECMWF) at 1ºx1º resolution (Dee et al. 2011). Meteorological reanalyses products are essentially model simulations constrained by observations and they have been widely validated against independent observations. Daily mean values are calculated as the mean of the four available time steps at 00, 06, 12, and 18UTC for 10m wind speed components (u and v) and 2m relative humidity. Maximum temperature is approximated by the daily maximum of those time steps, while daily mean surface solar radiation is obtained from the 3-hourly values provided for the forecast fields.

2.2. **Chemistry Transport Models (CTMs)**

A set of state-of-the-art air quality models participating in the EDT exercise is used here: LOTOS-EUROS (Schaap et al., 2008, Manders et al. 2017), EMEP/MSC-W (Simpson et al., 2012), CHIMERE (Mailer et al., 2017), MATCH (Robertson et al., 1999), MINNI (Mircea et al., 2016) and WRF-Chem (Grell et al. 2005, Mar et al. 2016). The domain of the CTMs extends from 17ºW to 39.8ºE and from 32ºN to 70ºN and it follows a regular latitude-longitude projection of 0.25x0.4 respectively. The main features of the CTM setup are largely constrained by the EDT experimental protocol (e.g. meteorology, boundary conditions, emissions, resolution, see Colette et al. 2017a for further details). For instance, the boundary conditions were defined from climatology of observational data for most of the experiments of the EDT exercise (included the data used here). However, the representation of physical and chemical processes and the vertical distribution differ in the CTMs, as well as the vertical distribution of model layers (including altitude of the top layer and derivation of surface concentration at 3m height in the case of EMEP, LOTOS-EUROS and MATCH). Moreover, there were no specific constrains imposed on biogenic emissions (including soil NO emissions), which are represented by most of the models using an online module (Colette et al. 2017a). Since we aim here to compare the modelled relationship between meteorology and surface ozone, prescribing common features in the CTMs is particularly an advantage to identify potential sources of discrepancies.

Only one of the participating CTMs included online coupled chemistry/meteorology (WRF-Chem), while all the rest of the models used are offline. The CTMs were forced by regional climate model simulations using boundary conditions from the ERA-Interim global reanalysis (Dee et al., 2011). Most of these offline CTMs used the same meteorological input data, with a few exceptions. Three of them (EMEP, CHIMERE and MINNI) used input meteorology from the Weather Research and Forecast Model (WRF) (Skamarock et al. 2008). LOTOS-EUROS and MATCH used the input meteorology produced by RACMO2 (van Meijgaard, 2012) and HIRLAM (Dahlgren et al. 2016), respectively. Unlike the rest of the regional climate models, RACMO2 used in the EDT exercise excluded nudging towards ERA-Interim, which might have some impact in the meteorological fields generated by RACMO2. As mentioned, WRF-Chem couples the meteorology simulations online with chemistry. The meteorology used to drive WRF-Chem (initial and lateral boundary conditions and the application of limited four-dimensional data assimilation; see Colette et al GMD 2017a) is the same WRF meteorology from Skamarock et al. (2008) used as input for the EMEP, CHIMERE, and MINNI runs. Table 1 summarises the CTMs and the corresponding sources of meteorological input data used here. It is important to highlight that though WRF-Chem is not strictly a CTM, in order to avoid confusion with the statistical models developed in this study, we refer to all the air quality models considered (offline and online models) as CTMs hereafter. As with the observations, CTMs and their meteorological counterpart were interpolated to a common grid with 1° x 1° horizontal resolution. The use of a coarser resolution could have an impact in some regions with a complex orography where airflow is usually controlled by mesoscale phenomena (e.g. see-breeze and mountain-valley winds) or in regions characterized by high emissions densities (Schaap et al., 2015, Gan et al. 2016). In such cases the use of a finer grid could be beneficial to capture the variability of local processes.

A set of meteorological parameters was selected from the meteorological input data for the regression analyses. Similarly to the procedure with ERA-Interim, daily means are obtained from the available time steps every 3 hours in the case of WRF and RACMO2, and every 6 hours for HIRLAM for the following variables: 10m wind speed components, 2m relative humidity and surface solar radiation. Maximum temperature is also approximated by the daily maximum of those time steps.

3. **Multiple Linear regression model**

Summertime usually brings favourable conditions for high tropospheric ozone concentrations, such as air stagnation due to high-pressure systems, warmer temperatures, higher UV radiation, and lower cloud cover (Dawson et al. 2007). As stated above, the impact of meteorology on ozone concentration has been addressed through a wide variety of statistical methods in the literature. This study attempts to better understand how CTMs represent the influence of meteorology on ozone. To this aim, we use a multiple linear regression approach that can provide useful information of sensitivities in the distribution of ozone concentration as a whole (Porter et al., 2015).

A total of five meteorological predictors (Table 2) are selected based on the existing literature that has shown their strong influence on ozone pollution. (e.g. Bloomfield et al. 1996, Barrero et al. 2005, Camalier et al. 2007, Dawson et al. 2007, Rasmussen et al. 2012, Davis et al. 2011, Doherty et al., 2013, Otero et al. 2016). Moreover, it has been shown that the occurrence of air pollution episodes might increase when the pollution levels of the previous day are higher than normal (Ziomas et al. 1995). Then, apart from the meteorological predictors, we add the effect of the lag of ozone (MDA8 from the previous day) in order to examine the role of ozone persistence. Additionally, we include harmonic functions that capture the effect of seasonality as in Rust et al. al (2009) and Otero et al. (2016), which is referred as "day" in the MLRs (see Table 2).

For this study, we divide the European domain into 10 regions: England (EN), Inflow (IN), Iberian Peninsula (IP), France (FR), Mid-Europe (ME), Scandinavia (SC), North Italy (NI), Mediterranean (MD), Balkans (BA) and Eastern Europe (EA). These regions are based on those defined in the recent ETC/ACM Technical Paper (Colette et al. 2017b). For our study, we further subdivide the original Mediterranean region (MD) into a region covering the Balkans (BA), due to the strong influence of the ozone persistence on MDA8 O3 over this particular region as noted previously in Otero et al. (2016). Figure 1 shows the spatial coverage of each region and Table 3 lists their coordinates. As shown Otero et al. (2016), the relative importance of predictors in the MLRs shows distinct seasonal patterns. Then, multiple linear regression models (MLR, hereafter) are developed for each region for two seasons: springtime (April-May-June, AMJ) and summertime (July-August-September, JAS). These seasons differ from the meteorological definition, but cover the period when surface ozone typically reaches its highest concentrations (i.e. April-September). Since the observations did not cover exactly the whole European domain as CTMs, we applied an observational-mask to use the same number of grid-cells for CTMs and observations. Data used to estimate parameters of the MLR were spatially averaged over each region. Thus, we compare MLRs developed separately for CTMs and observations at each region and season. The observational dataset contains the gridded MDA8O3 and the meteorology input from ERA-Interim, while the dataset for the CTMs contains the MDA8O3 from each one of them along with the corresponding meteorological input (e.g. LOTOS and RACMO2, CHIMERE and WRF) (see table 1).

A MLR is built to describe the relationship between MDA8 O3 (predictand) and a set of covariates (or predictors) describing seasonality, ozone persistence and the influence of meteorological fields (table 2). A data series y_t, t= 1,..N (e.g. observations or CTM simulations) for a given region and season is conceived as a Gaussian random variable Y_t with varying mean μ_t and homogeneous variance σ^2. The mean μ_t is described as a linear function of the covariates, i.e.

$$Y_t \sim \mathcal{N}(\mu_t, \sigma^2),$$
$$\mu_t = \beta_0 + \beta_{sin} sin\left(\frac{2\pi}{365.25} d_t\right) + \beta_{cos} cos\left(\frac{2\pi}{365.25} d_t\right) + \beta_{lag} y_{t-1} + \sum_{K=1}^{K} \beta_k \, x_{t,k} \quad (1)$$

with t indexing daily values and d_t referring to the day in the year associated with the index t. β_0 is a constant offset, β_{sin} and β_{cos} are the first order coefficient of a Fourier series (e.g. Rust et al. 2009, 2013, Fischer et al. 2017), β_{lag} describes the persistence with respect to the previous day concentration y_{t-1} ; if t is the first day in the late summer season (JAS, July 1st), y_{t-1} is the concentration of June 30th. Further regression coefficients β_k describe the linear relation to potential meteorological drivers (see table 2). For covariates standardized to unit variance, the regression coefficients (β) are standardised coefficients giving the change in the predictand with the covariate in units of covariate standard deviation.

Following the same strategy as used in Otero et al. (2016), the MLRs are developed through several common steps: 1) starting with the full set of potentially useful components in the predictor, a stepwise backward regression using the Akaike Information Criterion (AIC) as a selection criterion removes successively those components in the predictor, which contribute least to the model performance; and 2) a multi-collinearity index known as variance inflation factor (VIF, Maindonald and Braun 2006) is used to detect multi-collinearity problems in the predictor (i.e. high correlations between two or more components in the predictor). Components with a VIF above 10 are left out of the predictor (Kutner et al 2004).

The statistical performance of each MLR (built separately from observations and CTMs) is assessed through the adjusted coefficient (R^2) and the root mean square error (RMSE). The R^2 estimates the fraction of total variability described by the MLR and the RMSE gives the average deviation between model and observation obtained in the MLR. We also examine the relative importance of the individual components in the predictor. According to the method proposed by Lindeman et al (1980), the relative importance of each predictor is estimated by its contribution to the R^2 coefficient (Grömping 2007). We assess the sensitivities of ozone to the predictors through the standardised coefficients obtained from the regression. These coefficients indicate the changes in the ozone response to the changes in the predictors, in terms of standard deviation. Thus, for every standard deviation unit increase (decrease) of a specific predictor, the predictand (MDA8 O3) will increase (decrease) the amount indicated by its coefficient in standard deviation units,. The use of standardised coefficients allows us to establish a direct comparison in the influence of individual predictors. The effect of seasonality introduced by the harmonic functions (namely, “day”, table 2) is kept in the MLRs (Eq. 1) for its usefulness in improving the power of the regression analysis, however further explanation about the effect of the predictors focuses on the rest of the variables.

4. Results and discussion

4.1. CTM performance by region

We compare the seasonal cycle of observations and CTMs through the time series of daily averaged values of MDAO8 O3 from observations and CTMs for the whole period (i.e. April-September, 2000-2010) spatially averaged over each region. Furthermore, correlation coefficients between both CTMs and observations at each region and season are used to quantify the CTM performance.

4.1.1. Seasonal cycle of MDA8 O3

We examine the ozone seasonal cycle represented by both the observational and modelled dataset. Figure 2 depicts daily averages during 2000-2010 of MDA8 O3 at each region for the CTMs and observations. In general, all CTMs are biased high compared with observations. CTM results are visually closer to observations in the northwestern regions (i.e. IN, EN and FR), while the spread becomes larger over the southern and southeastern regions (i.e. BA, NI, MD). The IN, EN and SC regions show the highest observed concentrations in the starting months (AMJ), which is not generally well captured by most of the CTMs, and they show a more flat timeline (e.g. LOTOS, MATCH, CHIMERE or WRF-Chem). For example, in the SC region, some of the CTMs underestimate the ozone concentrations in AMJ (i.e. WRF-Chem, CHIMERE and MINNI). The rest of the regions show the highest observed concentrations in JAS, which is generally overestimated by the CTMs. Models show discrepancies when compared to each other and to observations, and in some regions we find substantial differences. Larger discrepancies are found in the southern regions, such as IP, MD and BA, where the models show a considerable spread. There, the CTMs are not able to capture the variability of MDA8 O3 and they exhibit a different behaviour when compared to each other. For instance, the EMEP model shows a peak of ozone levels in April, while CHIMERE and MINNI show a peak in July. Overall LOTOS shows a relatively constant positive bias in all regions, more evident in the MD and NI regions. WRF-Chem tends to underestimate the ozone concentrations at the start of the seasonal period in some regions (e.g. SC, ME, EN, or EA).

CTM assessments have been presented in early EURODELTA exercises, although with a different set up for different purposes, which makes it difficult to establish a direct comparison on the performance of the models. For instance, Colette et al. (2017b) reported systematic differences among some models (i.e. CHIMERE, EMEP and LOTOS) when examining the long-term mean ozone concentration during the whole period of 1990-2010. Bessagnet et al. (2016) showed that most of the models in their study, (e.g. CHIMERE, LOTOS, or MINNI among others) overestimated the ozone concentrations in the selected study period. Specifically, they found a larger spread during nighttime than daytime, which was suggested to be related to the vertical mixing, given that most of the models shared the same meteorology but different vertical resolution and boundary conditions.

4.1.2. Correlation coefficients between modelled and observed time series

The correlation coefficients between the observed and modelled values of MDA8 O3 at each region and in each season are shown in Fig. 3. Overall, MDA8 O3 from the CTMs is better correlated with observations in JAS than in AMJ in the regions ME, NI, EA and EN. As expected from inspection of the average time series (Fig. 2), the lowest correlations between models and observations are found in BA, especially in AMJ for all models. In particular, EMEP is negatively correlated with observations over this region. As mentioned above, the larger discrepancies between CTMs and observations found over BA might be attributed to a low density of observation sites from which the interpolated dataset is derived, resulting in a lower quality or higher uncertainties of such product (Schnell et al. 2014). The highest correlations in AMJ are obtained at the following regions: ME; FR; NI; and EN for most of the models, except for EMEP for which the highest correlation with observations was found in IN and SC. The WRF-Chem model also shows a different behaviour in terms of the correlation coefficient with higher values in NI, MD and IP, and very low and negative correlations (-0.02) in SC. In general, the models that are most closely correlated with observations are MATCH, MINNI and CHIMERE, while LOTOS and WRF-Chem show the lowest correlations. In the case of LOTOS, it could be partially due to the use of a different set-up of the RACMO2 model, without nudging towards ERA-Interim (section 2.2). These correlations reflect the patterns represented by the seasonal cycle described above.

4.2. MLR performance

Figures 4 and 5 depict the statistical performance of each MLR in terms of R^2 and RMSE (respectively) at the different regions for both seasons, AMJ and JAS. The R^2 values indicate that all MLRs models (based on both observations and CTMs) are able to explain more than 60% of the MDA8 O3 variance in all regions. Overall, the MLRs show a stronger fit in JAS than in AMJ in most of the regions, with the exception of SC and IN that, in general show lower values of R^2 in JAS than in AMJ (Fig. 4). The MLRs appear to perform better in certain regions such as NI, ME, FR or EA, while the poorest statistical performance is found in IN and EN. The results obtained from the CTM-based MLRs show a similar performance to the observation-based MLRs in most of the regions. The lowest RMSE values for most of the MLR are found in SC ranging between 1 and 3 ppb, while EN shows the largest RMSE values, especially for the MLR built from WRF-Chem (Fig. 5). The MLRs from MATCH and CHIMERE show the lowest RMSE values (1-3ppb) suggesting the best statistical fit from a predictive point of view.

Both R^2 and RMSE metrics indicate that the statistical performance of MLRs for observations and CTMs show distinct variations between seasons and regions. Overall, better performances are found in JAS and in some regions (i.e. ME, NI, or FR) where MLRs are able to describe more than the 80% of the variance in CTMs and observations. This could be attributed to the major role of meteorology in summer influencing local photochemistry processes of ozone production, while in spring long range transport plays a stronger role (Monks, 2000, Tarasova et al. 2007). As it includes the bias, the RMSE reveals more differences among the MLRs when compared to each other (e.g. larger errors for WRF-Chem or LOTOS when compared to MATCH or CHIMERE). However, it is interesting that in general all MLRs show a similar tendency when evaluating the statistical performance, which indicate that observations-based and CTMs-based MLRs present a similar statistical performance for modelling MDA8 O3. The ability of the CTMs to reproduce the influence of meteorological drivers on MDA8 O3 is discussed in more detail below.

4.3. Effects of drivers of ozone concentrations

The analysis of the influence of the predictors in the MLRs reveals distinctive regional patterns in both observation-based and CTM-based MLRs. In agreement with Otero et al. (2016), here we also find that the regions geographically located towards the interior (including central, western and eastern regions) appear to be more sensitive to the meteorological predictors, especially in JAS. On the contrary, a minor meteorological contribution is found in the regions over the northernmost and southernmost edges, implying that non-local processes play a stronger role. Considering such similarities, in the following, the regions: EN, FR, ME, NI and EA are referred as the internal regions, while the rest of the regions: IN, SC, IP, MD and BA, are referred as the external regions (see Fig. 1).

4.3.1 Relative importance

Figure 6 depicts the relative importance of the predictors for the observation-based and CTM-based MLRs in the internal regions (Fig. 1). Here, a larger meteorological influence (i.e., the predictors other than LO3 and day) can be seen in JAS compared to AMJ in all of these regions. In general, the dominant meteorological drivers from the observation-based MLRs in these internal regions are RH and Tx. The contribution of RH is evident in AMJ (e.g. ME, or EA), while Tx is clearly dominant in JAS. SSRD is also a key driver of MDA8 O3 and generally, the wind factors (W10m and Wdir) appear to have a minor contribution.

Despite the CTM-based MLRs being able to capture the meteorological predictors, we observe discrepancies among the internal regions when compared to the observation-based MLR. The inter-model differences in terms of the relative importance of predictors are greater in AMJ than in JAS. For instance, the contribution of the LO3 is overestimated by most of CTMs, specifically WRF-Chem that shows a larger sensitivity to LO3 in both seasons over all of these regions. Similarly, EMEP also shows a larger contribution of LO3 than the rest of the CTMs, particularly in AMJ. Substantial differences are found in the influence of RH when comparing the observation-based and the CTMs-based models. The CTMs do not capture the relative importance of the RH well, especially in AMJ. In general, the CTMs driven by WRF meteorology show a slightly larger contribution of RH in most of the cases, although we notice that there are also some differences among the models that share the same meteorology. CTMs do capture the relative importance of Tx in all regions, but overall they overestimate it, as they also show for SSRD. Here, we find discrepancies when comparing the contribution of predictors in the statistical models from CTMs driven by the same meteorology (e.g. EMEP and WRF-Chem when compared to CHIMERE and MINNI). The largest differences among the CTMs are found for WRF-Chem, which tends to underestimate the contribution of the meteorological drivers in most of the regions. Interestingly, as mentions in Section 2, this is the only online coupled model participating in EDT.

Figure 7 presents the relative importance of individual predictors in the MLRs developed at the external regions (Fig. 1) for both seasons. The observation-based MLRs show that the main driving factor is LO3 in AMJ, while the effect of meteorological drivers becomes stronger in JAS. RH presents a larger contribution in some regions (e.g. IN, IP or SC) in AMJ and Tx in JAS (e.g. IN, IP, SC and BA). The contribution of wind components, Wdir and W10m, is mainly reflected in both seasons in the western regions (i.e. IN and IP) and in MD, respectively.

Overall, all CTMs show this tendency, although there are substantial differences when comparing the individual drivers' contribution in the observation-based and CTM-based MLRs, particularly in AMJ (Fig. 7). CTMs do not capture the contribution of LO3 reflected by the observation-based MLRs. As in the previous analysis (section 4.1) the largest discrepancies are found in BA, where observation-based MLR shows that most of the variability of ozone would be explained by LO3. On the contrary the CTM-based MLRs underestimate the contribution of LO3 and overestimate the meteorological effect in terms of larger contribution of Tx, SSRD and RH (e.g. LOTOS, CHIMERE and MINNI). The contribution of RH is underestimated by the CTMs in most of the regions, (except in BA). On the contrary, the relative importance of SSRD is overestimated in some regions (e.g. IP, IN or MD) and Tx (IN, SC), in particular for the CTMs driven by WRF. Overall, CTMs show the observed contribution of W10m and Wdir in both seasons, although with some inconsistences among the regions and CTMs.

Our results indicate that the relative importance of meteorological factors is stronger in the internal regions (Fig.6) than in the external regions (Fig.7), which could be partially attributed to a larger variability of most of the meteorological fields in internal regions (Fig. S1). The external regions are also more likely to be influenced by the lateral boundary conditions applied by each CTM. In addition, in some external regions (e.g. IP or MD), as mentioned in section 2, the use of a coarser grid in some regions might be insufficient to capture mesoscale processes, such as land-sea breezes, which also control

MDA8 O3 concentrations (Millán et al. 2002). Moreover, we observe that meteorology becomes more important in summer, when local photochemistry processes are dominant. In general, CTMs show this tendency, but limitations to reproduce the effect of some meteorological drivers are found. Specifically, while CTMs tend to overestimate the contribution of Tx, and SSRD, they underestimate the relative importance of RH, which is also reflected in the correlations coefficients between predictand the predictors (Figs. S2, S3).

4.3.2 Sensitivity of ozone to the drivers

We assess the sensitivities of MDA8 O3 to the drivers through their standardised coefficients obtained in the MLR (Section 3). These coefficients provide further information about the changes of MDA8 O3 due to effect of each driver. Figures 8 and 9 depict the values of the main driving factors obtained in the MLR for the internal and the external regions (respectively): LO3, Tx and RH. Similarly to those patterns described by the relative importance of drivers, we observe that the ozone response to LO3 is stronger in AMJ than in JAS: the corresponding standardised coefficients are always positive and generally higher in AMJ. The observed sensitivities to LO3 are smaller in the internal regions (Fig. 8), being particularly dominant in the external regions (Fig. 9). Overall, most of the CTMs reflect a similar tendency. However, there are evident differences among observations and CTMs when comparing the values of the standardised coefficients, specifically in some regions such as BA or MD. When comparing the ozone responses of the CTMs to LO3, we observe that in most of the regions MATCH and MINNI show values closest to observations, while WRF-Chem shows a large sensitivity to LO3.

Correlations between MDA8 O3 and Tx are strong, especially in the internal regions in JAS (Fig. S2). Overall, we show that the CTMs appear to capture the observed effect of Tx better in JAS than in AMJ in most of the regions. The highest sensitivities to Tx are found in some internal regions such as ME, NI, FR and EN, which is also shown in the CTMs. However, we see that most of the CTMs tend to overestimate the effect of Tx. Moreover, distinct sensitivities to Tx are shown by models that share the same meteorology (i.e. CHIMERE, EMEP, MINNI and WRF-Chem). In particular, the MINNI and CHIMERE models show higher Tx sensitivities when compared to the rest of the CTMs. While MINNI model presents the highest sensitivities to Tx in spring, specifically in EN and FR, EMEP shows smaller values and it underestimates the correlations between Tx and MDA8 O3 (Figs. S2, S3).

The slope of the ozone-temperature relationship (m_{O3-T}) has been used in several studies to assess the ozone climate penalty (eg. Bloomer et al., 2009, Steiner et al., 2010, Rasmussen et al., 2012, Brown-Steiner et al. 2015) in the context of future air quality. Thus, we additionally analyse the relationship ozone-temperature in order to provide insight into the ability of CTMs to reproduce the observed m_{O3-T}. Similarly as in previous work (Brown-Steiner et al. 2015), the slopes are obtained from a simple linear regression using only Tx (without the influence from other predictors) and they are used to quantify such relationship in both seasons, AMJ and JAS.

Figures 10 and 11 illustrate the m_{O3-T} for the internal and the external regions respectively. The observed m_{O3-T} is larger in JAS than in AMJ. In AMJ, it ranges between -0.45 and 1.15 ppbK^{-1} with the largest values found in ME, NI and MD. In JAS, the observed climate penalty is of the order of 1-2.7 ppbK^{-1} with the largest values in EN, FR, ME, NI, and MD. CTMs show a better agreement with observations in JAS than in AMJ. CTMs tend to overestimate the climate penalty in AMJ in most of the regions, with some exceptions, such as EMEP and MATCH that systematically underestimate the slopes. Also, CTMs are generally better in simulating the observed $m_{O3\text{-}T}$ in the internal regions compared to the $m_{O3\text{-}T}$ in the external regions, where in general CTMs appear to overestimate the climate penalty in both seasons. Using this metric, we identify some regions particularly sensitive to temperature, with larger values of $m_{O3\text{-}T}$ (e.g. EN, ME, FR, NI or MD). Through a multi-model assessment, Colette et al. (2015) showed a significant summertime climate penalty in southern, western and central European regions (e.g. EA, IP, FR, ME or MD) in the majority of the future climate scenarios used. Our study shows that most of the CTMs confirm the observed climate penalty in JAS in such regions in the near present, although we found that most of the CTMs overestimate the climate penalty in AMJ, especially in the external regions.

We see a stronger effect of RH in AMJ than in JAS in the observations compared with the CTMs (Figs. 8 and 9), with the greatest impact in the internal regions (e.g. EA, ME, NI, FR and EN). The CTMs show this tendency slightly in some regions (e.g. ME, FR or EN), but differences become evident when compared to the observed values and overall they underestimate the effect of RH. As mentioned, CTMs underestimate the strength of the relationship between ozone-RH (Figs. S2, S3). This general lack of sensitivity to RH could also partially explain the tendency for all CTMs to show a high bias in simulated ozone compared with observations (Fig. 2). Among the possible reasons for this inconsistency, we hypothesize that it can be related to the fact that ozone removal processes can be associated to higher relative humidity levels during thunderstorm activity on hot moist days, which might not be well captured by CTMs. Furthermore, the documented impacts of ozone dry deposition suggest that it may also play a role in explaining the problems that CTMs show to reproduce the observed relationship ozone-relative humidity.

High SSRD levels favour photochemical ozone formation and it is usually positively correlated to ozone. In this case, CTMs also present some limitations to capture this effect and they overestimated the sensitivities of ozone to SSRD (Figs. S4, S5). For example, the observations show lower and surprisingly negative effect of SSRD. Although the correlations between SSRD and ozone are positive (see Fig. S2, S3), the presence of other predictors in the regression may reverse the sign of the estimated coefficient. The CTMs show a stronger sensitivity of ozone to SSRD and they overestimate its influence on surface ozone. Similarly, the sensitivities to Wdir and W10m are also overestimated by the CTMs, especially in AMJ (Figs. S4, S5).

Our analysis suggests that CTMs present more limitations to reproduce the influence of meteorological drivers to MDA8 O3 concentrations in the external regions than in the internal regions, particularly in AMJ. Moreover, we find the largest discrepancies in BA, where models show the poorest seasonal performance and correlation coefficients (Figs. 2 and 3, respectively), probably due a low quality of the observational dataset.

Furthermore, LO3 is the main driver over most of the external regions and explains a large proportion to the total variability of MDA8 O3, while meteorological factors play a smaller influence. Lemaire et al. (2016) found a very low performance (based on R^2) over the British Isles, Scandinavia and the Mediterranean using a different statistical approach that only included two meteorological drivers. They attributed this low skill to the large influence over those regions of long-range transport of air pollution (Lemaire et al. 2016). Our results confirm the small influence of the meteorological drivers over those regions and the strong influence of the ozone persistence. Moreover, in the case of the external regions of northern Europe, it could also be explained due to the dominance of transport processes such as the stratospheric-tropospheric exchange or long-range transport from the European continent, rather than local meteorology, particularly in AMJ (Monks, 2000, Tang et al. 2009, Andersson et al. 2009).

Previous work pointed out that local sources of NOx and biogenic VOC (ozone precursors) are important factors of summertime ozone pollution in the Mediterranean basin (Richards et al. 2013). Moreover, some studies suggested that the local vertical recirculation and accumulation of pollutants play an important role in ozone pollution episodes in this region: during the nighttime the air masses are held offshore by land-sea breeze, creating reservoirs of pollutants that are brought the following day (Millán et al. 20002, Jiménez et al. 2006, Querol et al. 2017). All of these factors (e.g. local emissions as well as local and large-scale processes) control the ozone variability, which might explain the smaller influence of local meteorological factors shown in this study over the Mediterranean basin when compared to meteorological influence in the internal regions. Thus, we may hypothesize that the strong impact of LO3 observed in the external regions over southern Europe (i.e. IP, MD, BA) could be partially due to the role of vertical accumulation and recirculation of air masses along the Mediterranean coasts as a result of the mesoscale phenomena, which is enhanced by the complex terrains that surround the Basin. Other important factor for the strong impact of LO3 observed is the slow dry deposition of ozone on water that would favour the ozone persistence in southern Europe.

Overall we conclude that CTMs capture the effect of meteorological drivers better in the internal regions (EN, FR, ME, NI and EA), where the influence of local meteorological conditions is stronger. The major effect of meteorological parameters found in the internal European regions might be also attributed to the fact that overall the variability of meteorological conditions is larger in those regions (Fig. S1). We also find differences among the CTMs driven by the same meteorology. As mentioned in the introduction, Bessagnet el al. (2016) suggested that the spread in the model results could partly explained by the differences in the vertical diffusion coefficient and the planetary boundary layer, differently diagnosed in each of the CTMs. Our results also indicate that even though models share the same meteorology (considering the prescribed requirements defined by the EDT exercise) they show discrepancies when compared to each other, which could be attributed other sources of uncertainties (such as physical and chemical internal process in the CTMs). The NMVOC and NO_x emissions from the biosphere are critical in the ozone formation. Since biogenic emissions were not specifically prescribed, which have a strong dependence on temperature and solar radiation, discrepancies in the CTMs performances, (e.g. different sensitivities to Tx) might be expected. Furthermore, we notice that the CTMs do not reproduce consistently the regional ozone-temperature relationship, which is a key factor when assessing the impacts of climate change on future air quality.

5. Summary and conclusions

The present study evaluates the capability of a set of Chemical Transport Models (CTMs) to represent the regional relationship between daily maximum 8-hour average ozone (MDA8 O3) and meteorology over Europe. Our results show systematic differences between the CTMs in reproducing the seasonal cycle when compared to observations. In general, they tend to overestimate the MDA8 O3 in most of the regions. In the western and northern regions (i.e. Inflow, England and Scandinavia), some models did not capture the high ozone levels in spring (e.g. CHIMERE, MINNI and WRF-Chem), while in other southern regions (e.g. Iberian Peninsula, Mediterranean and Balkans) they overestimated the ozone levels in summer (e.g. LOTOS, CHIMERE). Of the CTMs, MATCH and MINNI were the most successful in capturing the observed seasonal cycle of ozone in most regions. All CTMs revealed limitations to reproduce the variability of ozone over the Balkans region, with a general overestimation of the ozone concentrations, considerably larger during the warmer months (July, August). As reflected in the results, a limitation of the interpolated observational product used here is that in some regions (e.g. southern Europe) it has a lower quality due to a reduced number of stations (section 2.1).

The MLRs performed similarly for most of the CTMs and observations, describing more than 60 % of the total variance of MDA8 O3. Overall, the MLRs perform better in JAS than in AMJ, and the highest percentages of described variance were found in Mid Europe and North Italy. This could be attributed to local photochemical processes being more important in JAS, and is consistent with a stronger influence of long-range transport in AMJ.

The effects of predictors revealed spatial and seasonal patterns, in terms of their relative importance in the MLRs. Particularly, we noticed a larger local meteorological influence in the regions located towards the interior, here termed as the internal regions (i.e. England, France, Mid-Europe, North Italy and East-Europe). A minor local meteorological contribution was found in the rest of the regions, referred as the external regions (i.e. Inflow, Iberian Peninsula, Scandinavia, Mediterranean and Balkans). The CTMs are in better agreement with the observations in the internal regions than in the external regions, where they were not as successful in reproducing the effects of the ozone drivers. Overall, the different behaviour in the MLRs developed in the external regions could be attributed to (i) a larger influence of dynamical processes rather than local meteorological processes (e.g. long range transport in the northern regions) (ii) a stronger impact of the boundary conditions (iii) the use of a coarser grid that might be insufficient to capture mesoscale processes that also influence MDA8 O3 (e.g. sea-land breezes in the southern regions).

We found substantial differences in the sensitivities of MDA8 O3 to the different meteorological factors among the CTMs, even when they used the same meteorology. As Bessagnet et al. (2016) point out, the differences amongst CTMs could be partly attributed to some other diagnosed model variables (e.g. vertical diffusion coefficient and boundary layer height, as well as vertical model resolution). To assess the effect of such potential sources of uncertainties, further investigations would be required. Moreover, variations in the sensitivity of ozone to meteorological parameters could depend on differences in the chemical and photolysis mechanisms and the implementation of various physics schemes, all of which differ between the CTMs (see Colette et al. 2017a). Specifically, the discrepancies found in the sensitivities of MDA8 O3 to maximum temperature might be also attributed to biogenic emissions not prescribed in the models. This was particularly reflected in the analysis of the slopes ozone-temperature ($m_{O3\text{-}T}$) to assess the climate penalty, which differed between CTMs and regions when compared to the observations in both seasons. Most of the CTMs confirm the observed climate penalty in JAS, but with larger discrepancies in the external regions than in the internal regions. Furthermore, CTMs tend to overestimate the climate penalty in AMJ (particularly in the external regions).

Our results have shown that CTMs tend to overestimate the influence of maximum temperature and surface solar radiation in most of the regions, both strongly associated with ozone production. None of the CTMs captured the strength of the observed relationship between ozone and relative humidity appropriately, underestimating the effect of relative humidity, a key factor in the ozone removal processes. We speculate that ozone dry deposition schemes used by the CTMs in this study may not adequately represent the relationship between humidity and stomatal conductance, thus underestimating the ozone sink due to stomatal uptake. Further sensitivity analyses would be recommended for testing the impact of the current dry deposition schemes in the CTMs.

Data availability

The data are available upon request from the corresponding author.

Acknowledgments

We acknowledge Jordan L. Schnell for providing the interpolated dataset of MDA 8 O3. Modelling data used in the present analysis were produced in the framework of the EURODELTA-Trends Project initiated by the Task Force on Measurement and Modelling of the Convention on Long Range Transboundary Air Pollution. EURODELTA-Trends is coordinated by INERIS and involves modelling teams of BSC, CEREA, CIEMAT, ENEA, IASS, JRC, MET Norway, TNO, SMHI. The views expressed in this study are those of the authors and do not necessarily represent the views of EURODELTA-Trends modelling teams.

List of Tables:

CTM	**Meteorology**	**Coupling**
LOTOS-EUROS	RACMO2	Off-line
MATCH	HIRLAM	Off-line
EMEP	WRF	Off-line
CHIMERE		Off-line
MINNI		Off-line
WRF-Chem		On-line

Table 1. List of the chemistry-transport models used in the study, their corresponding meteorological driver and chemistry/meteorology coupling.

Predictor	Definition
LO3	Lag of O3 (24 h)
Tx	Maximum temperature
RH	Relative humidity
SSRD	Surface solar radiation
Wdir	Wind direction
W10m	Wind speed
day	$\sin(2\pi d_t/365.25)$, $\cos(2\pi d_t/365.25)$

Table 2. List of the predictors used in the multiple linear regression analysis: meteorological parameters, lag of O3 (24h, previous day) and the seasonal cycle components.

Region	Acronym	Coordinates (longitude, latitude)

England	EN	5W-2E, 50N-55N
Inflow	IN	10W-5W, 50N-60N, and 5W-2E, 55N-60N
Iberian Peninsula	IP	10W-3E, 36N-44N
France	FR	5W-5E, 44N-50N
Mid-Europe	ME	2E-16E, 48N-55N
Scandinavia	SC	5E-16E, 55N-70N
North Italy	NI	5E-16E, 44N-48N
Balkans	BA	18E-28E, 38N-44N
Mediterranean	MD	3E-18E, 36N-44N
Eastern Europe	EA	16E-30E, 44N-55N

Table 3. List of the regions with the short name and the coordinates.

List of Figures:

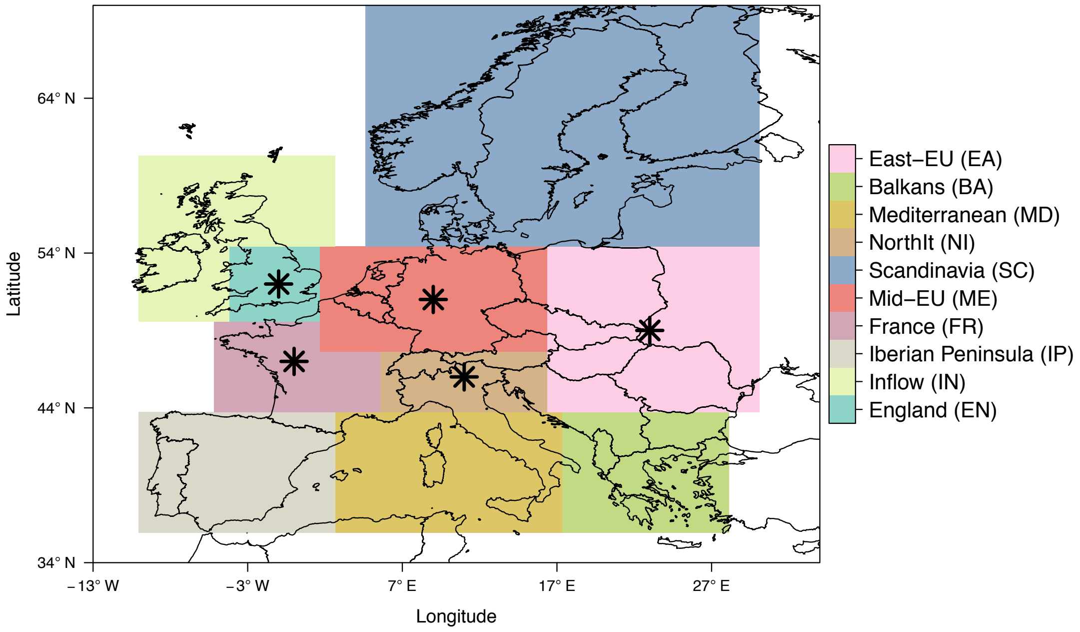


Figure 1. Map of the regions considered in the study. Regions indicated with a black star are referred to the internal regions in the text. The rest of regions are referred to the external regions of the European domain.

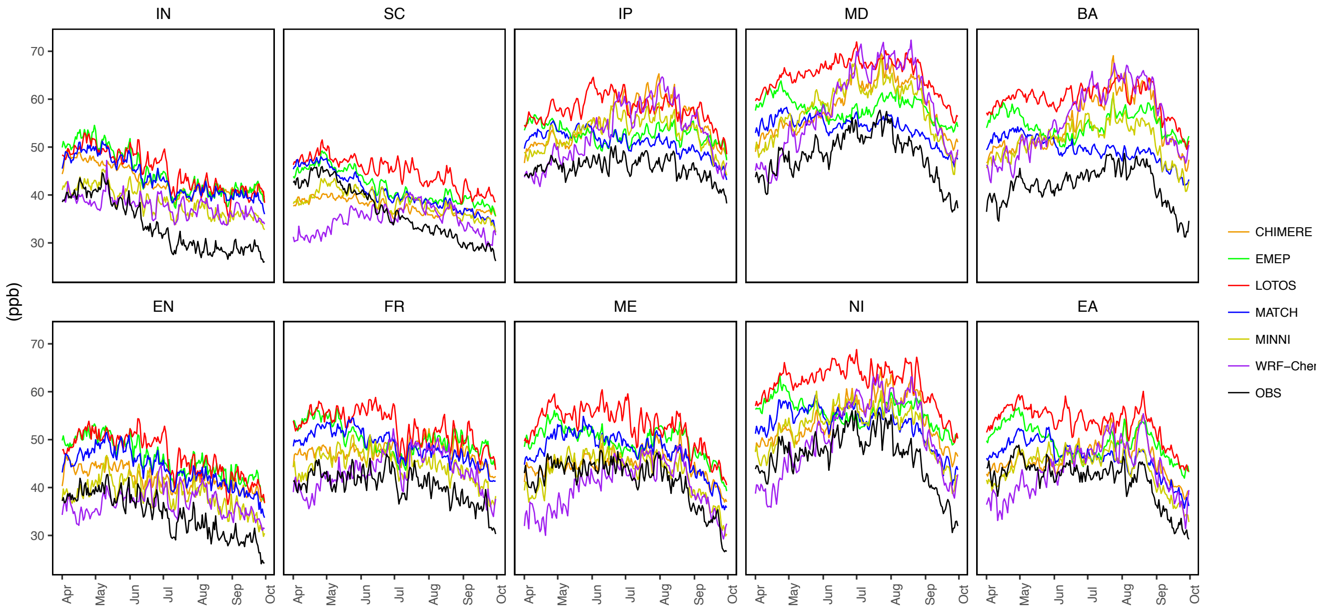
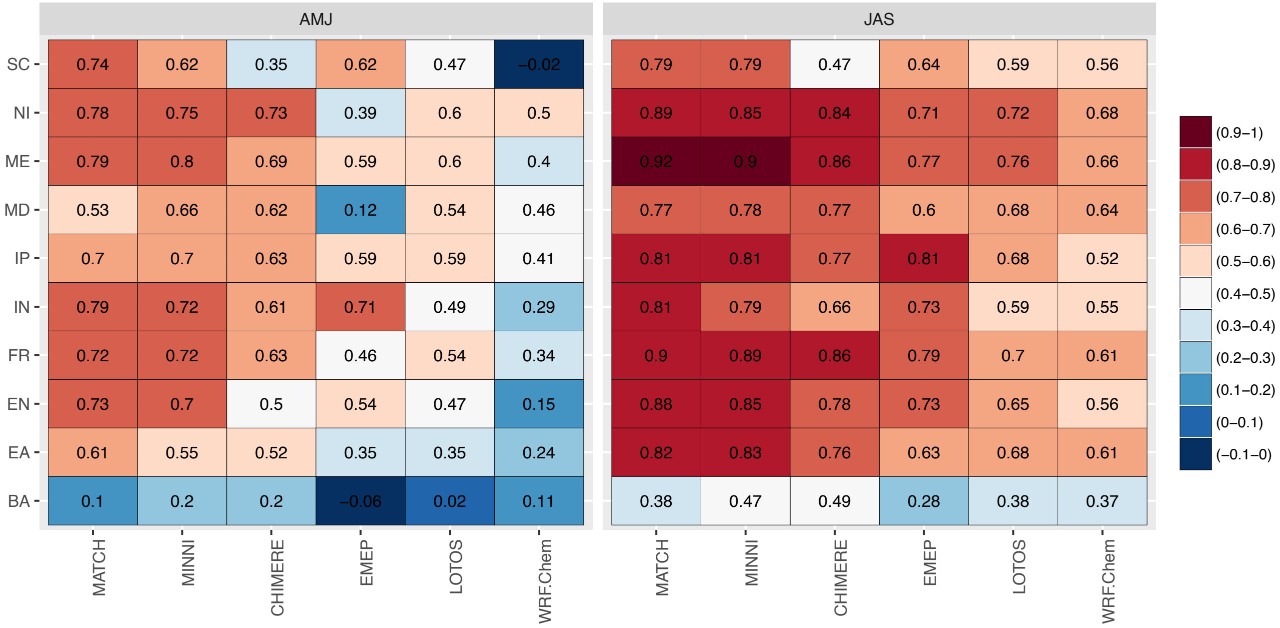
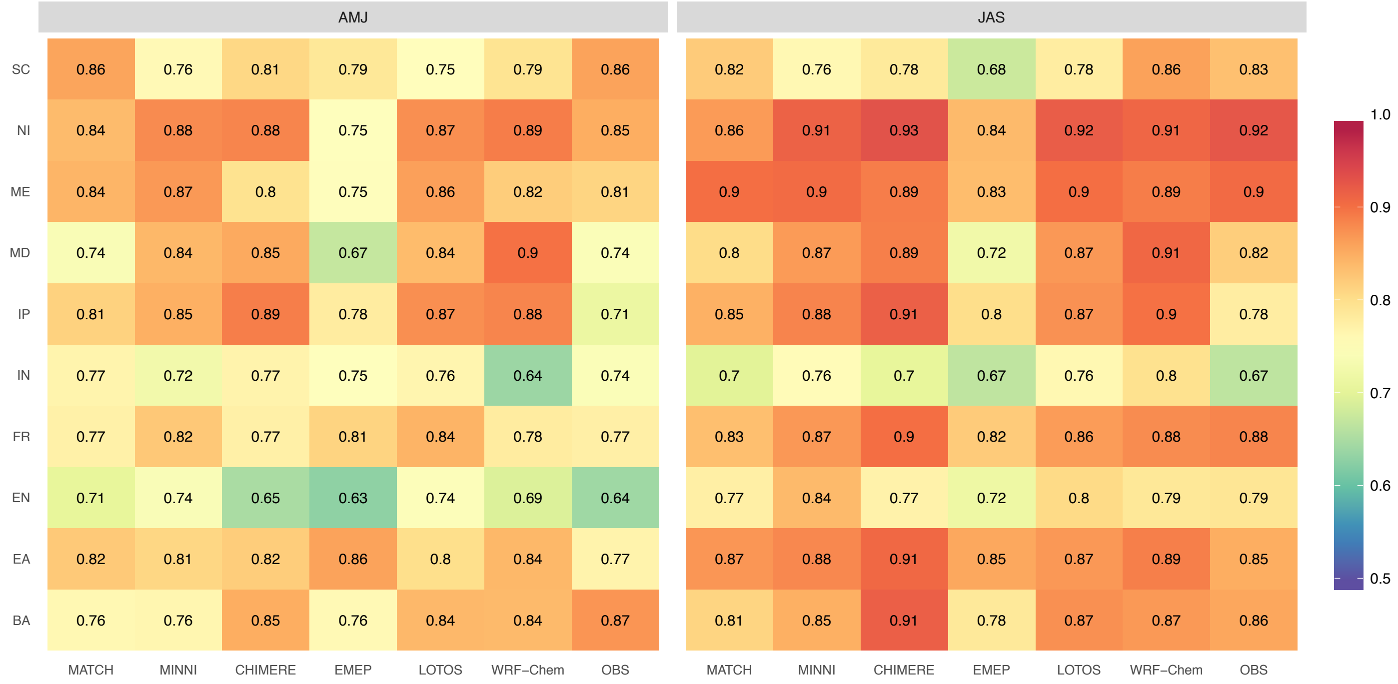


Figure 2. Time series of daily averages of MDA8 O3 during the ozone season (April-September) for the period of study (2000-2010) at each subregion.



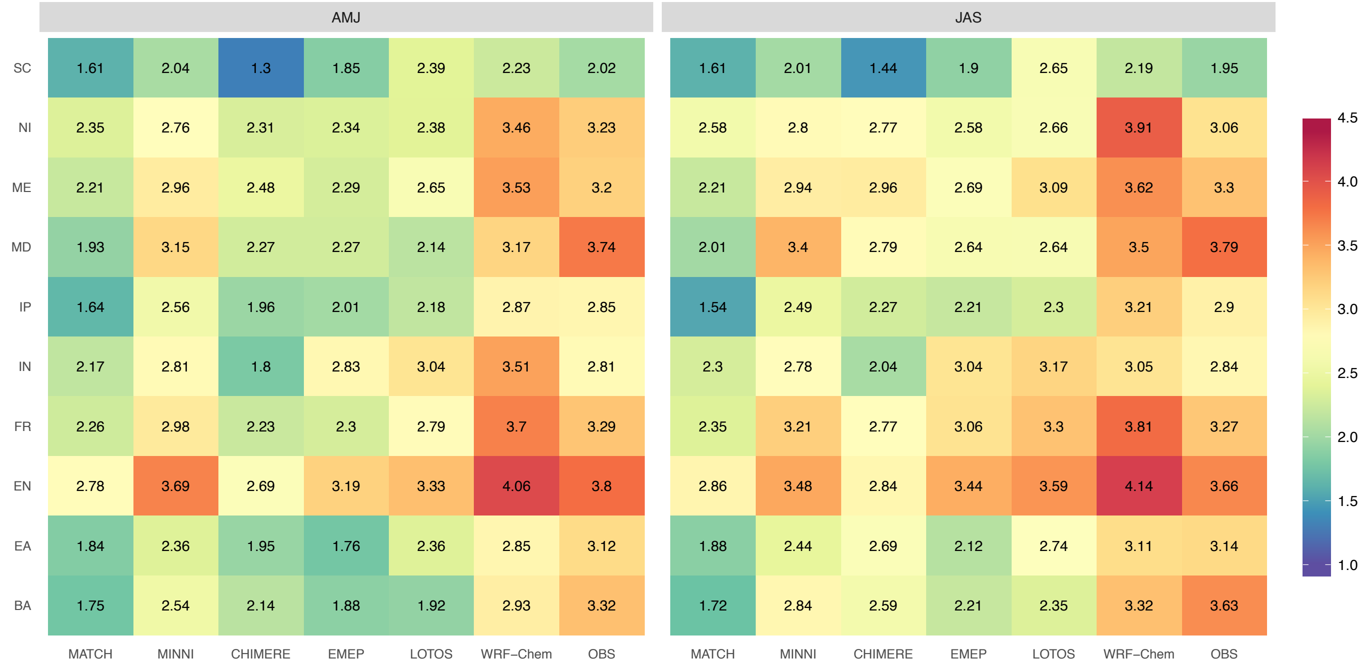
	AMJ						JAS					
	MATCH	MINNI	CHIMERE	EMEP	LOTOS	WRF.Chem	MATCH	MINNI	CHIMERE	EMEP	LOTOS	WRF.Chem
SC	0.74	0.62	0.35	0.62	0.47	−0.02	0.79	0.79	0.47	0.64	0.59	0.56
NI	0.78	0.75	0.73	0.39	0.6	0.5	0.89	0.85	0.84	0.71	0.72	0.68
ME	0.79	0.8	0.69	0.59	0.6	0.4	0.92	0.9	0.86	0.77	0.76	0.66
MD	0.53	0.66	0.62	0.12	0.54	0.46	0.77	0.78	0.77	0.6	0.68	0.64
IP	0.7	0.7	0.63	0.59	0.59	0.41	0.81	0.81	0.77	0.81	0.68	0.52
IN	0.79	0.72	0.61	0.71	0.49	0.29	0.81	0.79	0.66	0.73	0.59	0.55
FR	0.72	0.72	0.63	0.46	0.54	0.34	0.9	0.89	0.86	0.79	0.7	0.61
EN	0.73	0.7	0.5	0.54	0.47	0.15	0.88	0.85	0.78	0.73	0.65	0.56
EA	0.61	0.55	0.52	0.35	0.35	0.24	0.82	0.83	0.76	0.63	0.68	0.61
BA	0.1	0.2	0.2	−0.06	0.02	0.11	0.38	0.47	0.49	0.28	0.38	0.37

Figure 3. Correlation coefficients between observed and modelled MDA8 O3 for spring (AMJ) and summer (JAS) for the period of study (2000-2010) at each region (rows) and models (columns, ordered by highest correlation values).



	AMJ							JAS						
	MATCH	MINNI	CHIMERE	EMEP	LOTOS	WRF–Chem	OBS	MATCH	MINNI	CHIMERE	EMEP	LOTOS	WRF–Chem	OBS
SC	0.86	0.76	0.81	0.79	0.75	0.79	0.86	0.82	0.76	0.78	0.68	0.78	0.86	0.83
NI	0.84	0.88	0.88	0.75	0.87	0.89	0.85	0.86	0.91	0.93	0.84	0.92	0.91	0.92
ME	0.84	0.87	0.8	0.75	0.86	0.82	0.81	0.9	0.9	0.89	0.83	0.9	0.89	0.9
MD	0.74	0.84	0.85	0.67	0.84	0.9	0.74	0.8	0.87	0.89	0.72	0.87	0.91	0.82
IP	0.81	0.85	0.89	0.78	0.87	0.88	0.71	0.85	0.88	0.91	0.8	0.87	0.9	0.78
IN	0.77	0.72	0.77	0.75	0.76	0.64	0.74	0.7	0.76	0.7	0.67	0.76	0.8	0.67
FR	0.77	0.82	0.77	0.81	0.84	0.78	0.77	0.83	0.87	0.9	0.82	0.86	0.88	0.88
EN	0.71	0.74	0.65	0.63	0.74	0.69	0.64	0.77	0.84	0.77	0.72	0.8	0.79	0.79
EA	0.82	0.81	0.82	0.86	0.8	0.84	0.77	0.87	0.88	0.91	0.85	0.87	0.89	0.85
BA	0.76	0.76	0.85	0.76	0.84	0.84	0.87	0.81	0.85	0.91	0.78	0.87	0.87	0.86

Figure 4. Coefficients of determination (R^2) for each CTM-based (ordered as in Fig.3) and observation-based MLR in spring (AMJ) and summer (JAS).



	AMJ							JAS						
	MATCH	MINNI	CHIMERE	EMEP	LOTOS	WRF–Chem	OBS	MATCH	MINNI	CHIMERE	EMEP	LOTOS	WRF–Chem	OBS
SC	1.61	2.04	1.3	1.85	2.39	2.23	2.02	1.61	2.01	1.44	1.9	2.65	2.19	1.95
NI	2.35	2.76	2.31	2.34	2.38	3.46	3.23	2.58	2.8	2.77	2.58	2.66	3.91	3.06
ME	2.21	2.96	2.48	2.29	2.65	3.53	3.2	2.21	2.94	2.96	2.69	3.09	3.62	3.3
MD	1.93	3.15	2.27	2.27	2.14	3.17	3.74	2.01	3.4	2.79	2.64	2.64	3.5	3.79
IP	1.64	2.56	1.96	2.01	2.18	2.87	2.85	1.54	2.49	2.27	2.21	2.3	3.21	2.9
IN	2.17	2.81	1.8	2.83	3.04	3.51	2.81	2.3	2.78	2.04	3.04	3.17	3.05	2.84
FR	2.26	2.98	2.23	2.3	2.79	3.7	3.29	2.35	3.21	2.77	3.06	3.3	3.81	3.27
EN	2.78	3.69	2.69	3.19	3.33	4.06	3.8	2.86	3.48	2.84	3.44	3.59	4.14	3.66
EA	1.84	2.36	1.95	1.76	2.36	2.85	3.12	1.88	2.44	2.69	2.12	2.74	3.11	3.14
BA	1.75	2.54	2.14	1.88	1.92	2.93	3.32	1.72	2.84	2.59	2.21	2.35	3.32	3.63

Figure 5. Root mean square errors (RMSE) for each CTM-based (ordered as in Fig.3) and observation-based MLR at each region, in spring (AMJ) and summer (JAS).

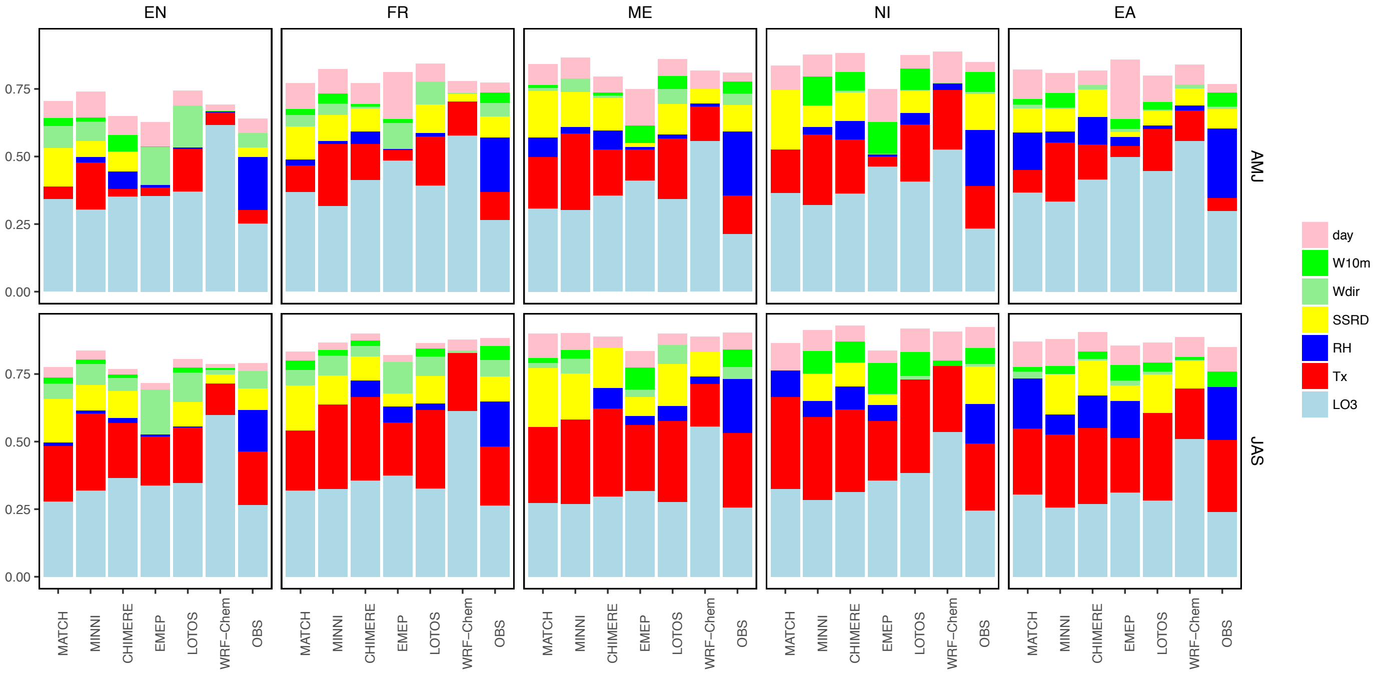


Figure 6. Proportion of each predictor to the total explained variance for each CTM-based (ordered as in Fig.3) and observation-based MLR in AMJ (top) and JAS (bottom) for the internal regions: England (EN), France (FR), Mid-Europe (ME), North Italy (NI) and East-Europe (EA).

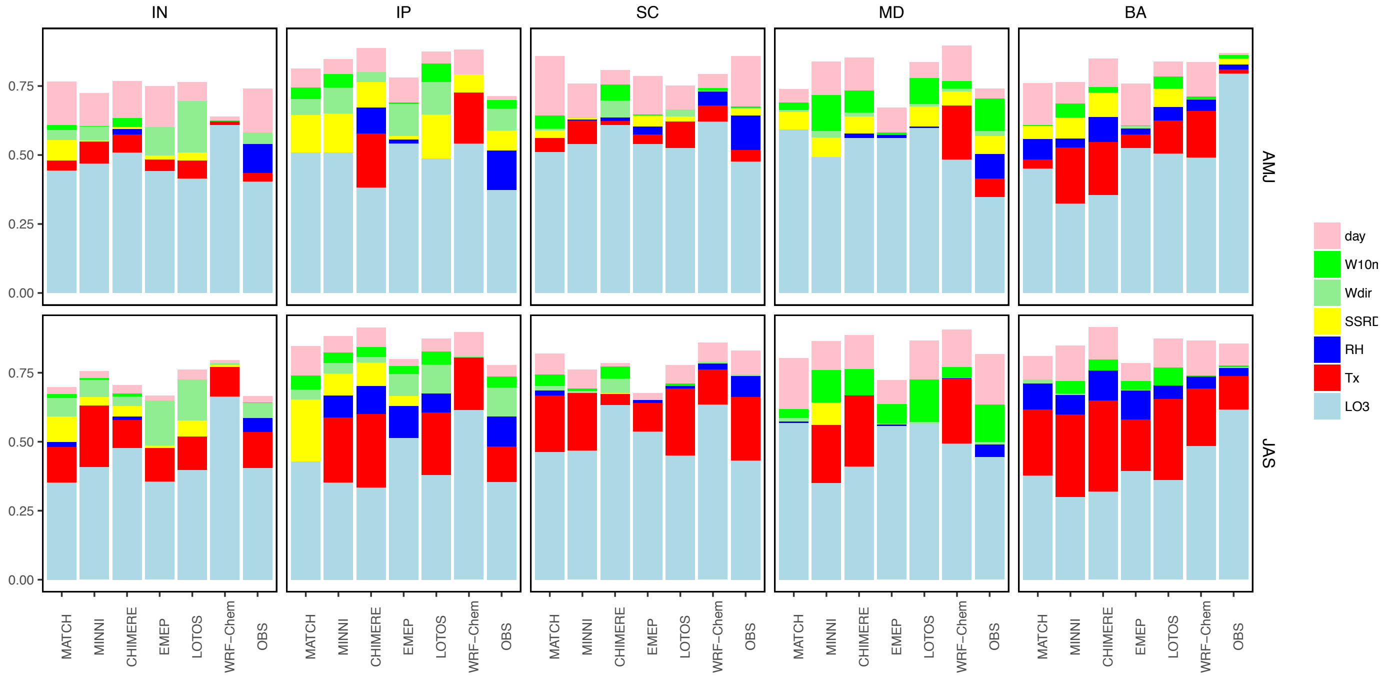


Figure 7. Proportion of each predictor to the total explained variance for each CTM-based (ordered as in Fig.3) and observation-based MLR in AMJ (top) and JAS (bottom) for the external regions: Inflow (IN), Iberian Peninsula (IP), Scandinavia (SC), Mediterranean (ME) and Balkans (BA).

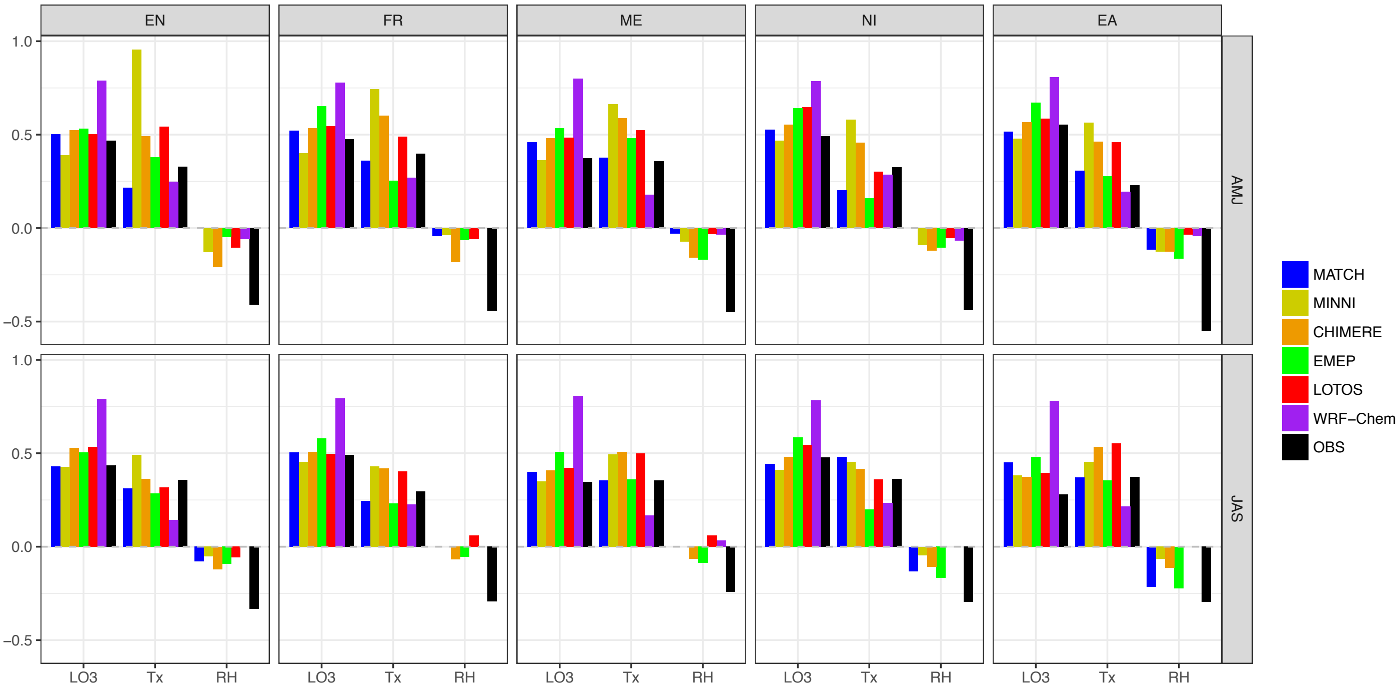


Figure 8. Standardised coefficients values of the main key-driving factors (LO3, Tx and RH) for each CTM-based (ordered as in Fig.3) and observation-based MLR in AMJ (top) and JAS (bottom) and for the internal regions: England (EN), France (FR), Mid-Europe (ME), North Italy (NI) and East-Europe (EA).

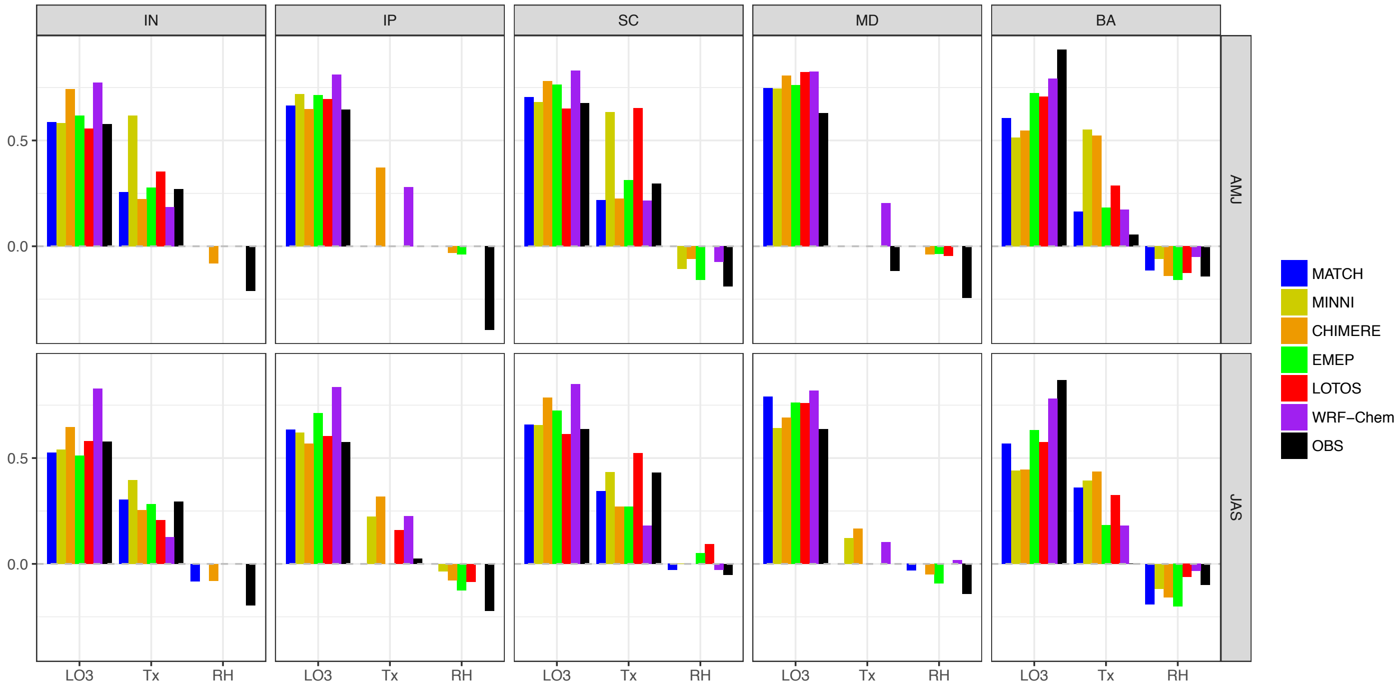


Figure 9. Standardised coefficients values of the main key-driving factors (LO3, Tx and RH) for each CTM-based (ordered as in Fig.3) and observation-based MLR in AMJ (top) and JAS (bottom) and for the external regions: Inflow (IN), Iberian Peninsula (IP), Scandinavia (SC), Mediterranean (ME) and Balkans (BA).

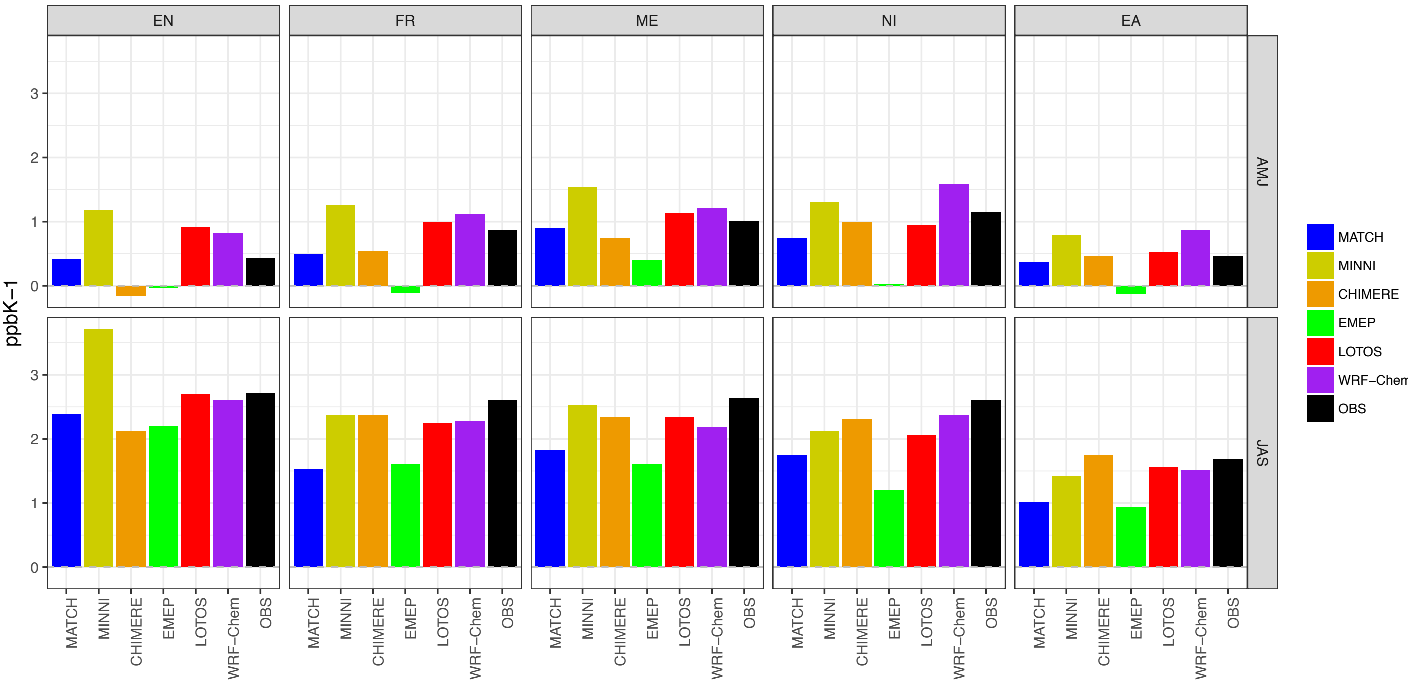


Figure 10. Slopes (m_{O3-T}; $ppbK^{-1}$) obtained from a simple linear regression to estimate the relationship ozone-temperature for each CTM-based (ordered as in Fig.3) and observation-based MLR in AMJ (top) and JAS (bottom) and for the internal regions: England (EN), France (FR), Mid-EU (ME), North Italy (NI), East-EU (EA).

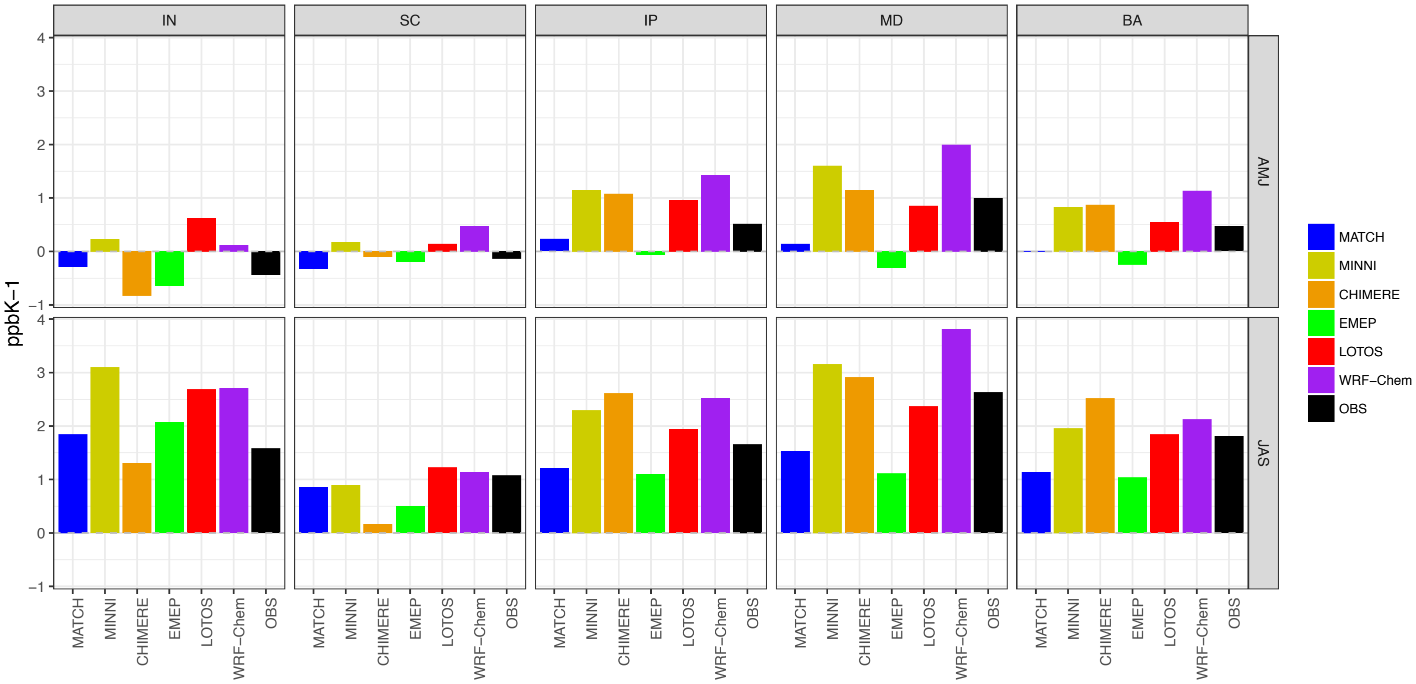


Figure 11. Slopes (m_{O3-T}; ppbK^{-1}) obtained from a simple linear regression to estimate the relationship ozone-temperature for each CTM-based (ordered as in Fig.3) and observation-based MLR in AMJ (top) and JAS (bottom) and for the external regions: Inflow (IN), Iberian Peninsula (IP), Scandinavia (SC), Mediterranean (ME) and Balkans (BA).

References

Andersson, C., J. Langner, and R. Bergström: Interannual variation and trends in air pollution over Europe due to climate variability during 1958–2001 simulated with a regional CTM coupled to the ERA-40 reanalysis, Tellus, Ser. B, 59, 77–98, 2007.

Andersson C., Bergström R., Johansson C., Population exposure and mortality due to regional background PM in Europe–Long-term simulations of source region and shipping contributions, Atmospheric Environment, 43, 22, 3614-3620, http://dx.doi.org/10.1016/j.atmosenv.2009.03.040, 2009.

Andersson, C. and Engardt, M.: European ozone in a future climate: Importance of changes in dry deposition and isoprene emissions, J. Geophys. Res.-Atmos., 115, D02303, doi:10.1029/2008jd011690, 2010.

Baklanov, A., Schlünzen, K., Suppan, P., Baldasano, J., Brunner, D., Aksoyoglu, S., Carmichael, G., Douros, J., Flemming, J., Forkel, R., Galmarini, S., Gauss, M., Grell, G., Hirtl, M., Joffre, S., Jorba, O., Kaas, E., Kaasik, M., Kallos, G., Kong, X., Korsholm, U., Kurganskiy, A., Kushta, J., Lohmann, U., Mahura, A., Manders-Groot, A., Maurizi, A., Moussiopoulos, N., Rao, S. T., Savage, N., Seigneur, C., Sokhi, R. S., Solazzo, E., Solomos, S., Sørensen, B., Tsegas, G., Vignati, E., Vogel, B., and Zhang, Y.: Online coupled regional meteorology chemistry models in Europe: current status and prospects, Atmos. Chem. Phys., 14, 317–398, doi:10.5194/acp-14-317-2014, 2014.

Barrero M A, Grimalt J O and Canton L.: Prediction of daily ozone concentration maxima in the urban atmosphere Chemometr. Intell. Lab. Syst. 80 67–76, 2005.

Bessagnet, B., Pirovano, G., Mircea, M., Cuvelier, C., Aulinger, A., Calori, G., Ciarelli, G., Manders, A., Stern, R., Tsyro, S., García Vivanco, M., Thunis, P., Pay, M.-T., Colette, A., Couvidat, F., Meleux, F., Rouïl, L., Ung, A., Aksoyoglu, S., Baldasano, J. M., Bieser, J., Briganti, G., Cappelletti, A., D'Isidoro, M., Fi- nardi, S., Kranenburg, R., Silibello, C., Carnevale, C., Aas, W., Dupont, J.-C., Fagerli, H., Gonzalez, L., Menut, L., Prévôt, A. S. H., Roberts, P., and White, L.: Presentation of the EURODELTA III intercomparison exercise – evaluation of the chemistry transport models' performance on criteria pollutants and joint analysis with meteorology, Atmos. Chem. Phys., 16, 12667–12701, doi:10.5194/acp-16-12667-2016, 2016.

Bloomfield P J, Royle J A, Steinberg L J and Yang Q: Accounting for meteorological effects in measuring urban ozone levels and trends Atmos. Environ. 30 3067–77, 1996 .

Bloomer, B. J., Stehr, J. W., Piety, C. A., Salawitch, R. J., and Dickerson, R. R.: Observed relationships of ozone air pollution with temperature and emissions, Geophys. Res. Lett., 36, L09803, doi:10.1029/2009gl037308, 2009.

Brown-Steiner, B., Hess, P. G., and Lin, M. Y.: On the capabilities and limitations of GCCM simulations of summertime regional air quality: A diagnostic analysis of ozone and temperature simulations in the US using CESM CAM-Chem, Atmos. Environ., 101, 134–148, doi:10.1016/j.atmosenv.2014.11.001, 2015.

Camalier, L., Cox, W., and Dolwick, P.: The effects of meteorology on ozone in urban areas and their use in assessing ozone trends, Atmos. Environ., 41, 7127–7137, 2007.

Chaloulakou A, Saisana M and Spyrellis N: Comparative assessment of neural networks and regression models for forecasting summertime ozone in Athens Science of the Total Environment 313 1–13, 2003.

Coates, J., Mar, K. A., Ojha, N., and Butler, T. M.: The influence of temperature on ozone production under varying NOx conditions – a modelling study, Atmos. Chem. Phys., 16, 11601-11615, https://doi.org/10.5194/acp-16-11601-2016, 2016.

Colette, A., Andersson, C., Baklanov, A., Bessagnet, B., Brandt, J., Christensen, J., Doherty, R., Engardt, M., Geels, C., Giannakopoulos, C., Hedegaard, G., Katragkou, E., Langner, J., Lei, H., Manders, A., Melas, D., Meleux, F., Rouil, L., Sofiev, M., Soares, J., Stevenson, D., Tombrou-Tzella, M., Varotsos, K., and Young, P.: Is the ozone climate penalty robust in Europe?, Environ. Res. Lett., 10, 084015, doi:10.1088/1748-9326/10/8/084015, 2015.

Colette, A., Andersson, C., Manders, A., Mar, K., Mircea, M., Pay, M.-T., Raffort, V., Tsyro, S., Cuvelier, C., Adani, M., Bessagnet, B., Bergström, R., Briganti, G., Butler, T., Cappelletti, A., Couvidat, F., D'Isidoro, M., Doumbia, T., Fagerli, H., Granier, C., Heyes, C., Klimont, Z., Ojha, N., Otero, N., Schaap, M., Sindelarova, K., Stegehuis, A. I., Roustan, Y., Vautard, R., van Meijgaard, E., Vivanco, M. G., and Wind, P.: EURODELTA-Trends, a multi-model experiment of air quality hindcast in Europe over 1990–2010, Geosci. Model Dev. Discuss., https://doi.org/10.5194/gmd-2016-309, accepted, 2017a.

Colette, A., Solberg, S., Beauchamp, M., Bessagnet, B., Malherbe, L., and Guerreiro, C.: Long term air quality trends in Europe: Contribution of meteorological variability, natural factors and emissions, ETC/ACM, Bilthoven, 2017b.

Comrie A. C. : Comparing neural networks and regression models for ozone forecasting J. Air Waste Manage. Assoc. 47 653–63, 1997.

Dahlgren, P., Landelius, T., Kållberg, P., and Gollvik, S.: A high-resolution regional reanalysis for Europe. Part 1: Three-dimensional reanalysis with the regional HIgh-Resolution Limited-Area Model (HIRLAM), Quarterly Journal of the Royal Meteorological Society, 142, 2119-2131, 10.1002/qj.2807, 2016.

Davis, J., Cox, W., Reff, A., Dolwick, P.: A comparison of Cmaq-based and observation-based statistical models relating ozone to meteorological parameters. Atmospheric Environment 45, 3481e3487. http://dx.doi.org/10.1016/ J.Atmosenv.2010.12.060, 2011.

Dawson, J.P., Adams, P.J., Pandis, S.N.: Sensitivity of ozone to summertime climate in the Eastern USA: a modeling case study. Atmos. Environ. 41, 1494– 1511, 2007.

Dawson, J. P., Racherla, P. N., Lynn, B. H., Adams, P. J., and Pan- dis, S. N.:

Simulating present-day and future air quality as cli- mate changes: model evaluation, Atmos. Environ., 42, 4551– 4566, doi:10.1016/j.atmosenv.2008.01.058, 2008.

Dee D P et al. :The ERA-Interim reanalysis: configuration and performance of the data assimilation system Quart. J. R. Meteorol. Soc. 137 553–97, 2001.

Doherty, R. M., Wild, O., Shindell, D. T., Zeng, G., MacKenzie, I. A., Collins, W. J., Fiore, A. M., Stevenson, D. S., Dentener, F. J., Schultz, M. G., Hess, P., Derwent, R. G., and Keating, T. J.: Impacts of climate change on surface ozone and intercontinental ozone pollution: a multi- model study, J. Geophys. Res.-Atmos., 118, 3744–3763, doi:10.1002/jgrd.50266, 2013.

Elminir H.K.: Dependence of urban air pollutants on meteorology, Science of The Total Environment, 350,225-237, http://dx.doi.org/10.1016/j.scitotenv.2005.01.043, 2005.

Fischer M., Rust H.W., and Ulbrich U.: Seasonality in extreme precipitation – using extreme value statistics to describe the annual cycle in german daily precipitation. Meteorol. Z. accepted, 2017.

Fiore, A. M., Dentener, F. J., Wild, O., Cuvelier, C., Schultz, M. G., Hess, P., Textor, C., Schulz, M., Doherty, R. M., Horowitz, L. W., MacKenzie, I. A., Sanderson, M. G., Shindell, D. T., Stevenson, D. S., Szopa, S., Van Dingenen, R., Zeng, G., Atherton, C., Bergmann, D., Bey, I., Carmichael, G., Collins, W. J., Duncan, B. N., Faluvegi, G., Folberth, G., Gauss, M., Gong, S., Hauglustaine, D., Holloway, T., Isaksen, I. S. A., Jacob, D. J., Jonson, J. E., Kaminski, J. W., Keating, T. J., Lupu, A., Marmer, E., Montanaro, V., Park, R. J., Pitari, G., Pringle, K. J., Pyle, J. A., Schroeder, S., Vivanco, M. G., Wind, P., Wojcik, G., Wu, S., and Zuber, A.: Multimodel estimates of intercontinental source-receptor relationships for ozone pollution, J. Geophys. Res.-Atmos., 114, D04301, doi:10.1029/2008jd010816, 2009.

Gan C., Hogrefe C., Mathur R., Pleim J., Xing J., Wong D., Gilliam R., Pouliot G., Wei C.: Assessment of the effects of horizontal grid resolution on long-term air quality trends using coupled WRF-CMAQ simulations, Atmospheric Environment, 132, 207-216,1352-2310,https://doi.org/10.1016/j.atmosenv.2016.02.036., 2016.

Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled “online” chemistry within the WRF model, Atmospheric Environment, 39, 6957-6975, http://dx.doi.org/10.1016/j.atmosenv.2005.04.027, 2005.

Grömping U. : Estimators of relative importance in linear regression based on variance decomposition Am. Stat. 61 139–47, 2007.

Hedegaard, G. B., Christensen, J. H., and Brandt, J.: The relative importance of impacts from climate change vs. emissions change on air pollution levels in the 21st century, Atmos. Chem. Phys., 13, 3569-3585, doi:10.5194/acp-13-3569-2013, 2013.

Hendriks C., Forsell N., Kiesewetter G., Schaap M., Schöpp W.: Ozone concentrations and damage for realistic future European climate and air quality scenarios, Atmospheric Environment, 144, 208-219, http://dx.doi.org/10.1016/j.atmosenv.2016.08.026, 2016.

Hodnebrog, Ø., Solberg, S., Stordal, F., Svendby, T. M., Simpson, D., Gauss, M., Hilboll, A., Pfister, G. G., Turquety, S., Richter, A., Burrows, J. P., and Denier van der Gon, H. A. C.: Impact of forest fires, biogenic emissions and high temperatures on the elevated Eastern Mediterranean ozone levels during the hot summer of 2007, Atmos. Chem. Phys., 12, 8727-8750, https://doi.org/10.5194/acp-12-8727-2012, 2012

Hogrefe, C., Biswas, J., Lynn, B., Civerolo, K., Ku, J. Y., Rosenthal, J., Rosenzweig, C., Goldberg, R., and Kinney, P. L.: Simulating regional-scale ozone climatology over the eastern United States: model evaluation results, Atmos. Environ., 38, 2627–2638, 2004.

IPCC, Climate Change 2013, the Physical Science Basis. Working Group I contribution to the fifth assessment report of the Intergovernmental Panel on Climate Change, Cambridge University Press, 2013.

Jacob, D. J. and Winner, D. A.: Effect of climate change on air quality, Atmos. Environ., 43, 51–63, doi:10.1016/j.atmosenv.2008.09.051, 2009.

Jonson, J. E., Simpson, D., Fagerli, H., and Solberg, S.: Can we explain the trends in European ozone levels?, Atmos. Chem. Phys., 6, 51–66, doi:10.5194/acp-6-51-2006, 2006.

Kavassalis, S. C., and J. G. Murphy: Understanding ozone-meteorology correlations: A role for dry deposition, Geophys. Res. Lett., 44, 2922–2931, doi:10.1002/2016GL071791, 2017.

Kong, X., Forkel R., Sokhi R. S., Suppan P., Baklanov A., Gauss M., Brunner D., Barò R., Balzarini A., Chemel C., Curci G., Jiménez-Guerrero P., Hirtl M., Honzak L., Im U., Pérez J. L., Pirovano G., San Jose R., Schlünzen K. H. , Tsegas G., Tuccella P., Werhahn J., Žabkar R., Galmarini S.: Analysis of Meteorology-Chemistry Interactions During Air Pollution Episodes Using Online Coupled Models within AQMEII Phase-2, Atmospheric Environment, http://dx.doi.org/10.1016/j.atmosenv.2014.09.020., 2014.

Kutner M. H., Nachtsheim C. J. and Neter J.: Applied Linear Regression Models 4th Ed (Boston, MA: McGraw-Hill Irwin) 2004.

Langner, J., Bergstro ̈m R. and Foltescu V.: Impact of climate change on surface ozone and deposition of sulphur and nitrogen in Europe, Atmos. Environ., 39, 1129–1141, doi:10.1016/j.atmosenv.2004. 09.082, 2005.

Lelieveld, J., and P. J. Crutzen: Influence of cloud and photochemical processes on tropospheric ozone, Nature, 343, 227–233, 1990.

Lemaire, V. E. P., Colette, A. and Menut, L.: Using statistical models to explore ensemble uncertainty in climate impact studies: the example of air pollution in Europe, Atmos. Chem. Phys., 16, 2559-2574, doi:10.5194/acp-16-2559-2016, 2016.

Lindeman R.H. , Merenda P. F. and Gold RZ: Introduction to Bivariate and Multivariate

Analysis. Scott, Foresman, Glenview, IL, 1980.

Linderson M. L.: Objective classification of atmospheric circulation over southern Scandinavia Int. J. Climatol. 21 155–69, 2001.

Mailler, S., Menut, L., Khvorostyanov, D., Valari, M., Couvidat, F., Siour, G., Turquety, S., Briant, R., Tuccella, P., Bessagnet, B., Colette, A., Létinois, L., Markakis, K., and Meleux, F.: CHIMERE-2017: from urban to hemispheric chemistry-transport modeling, Geosci. Model Dev., 10, 2397-2423, https://doi.org/10.5194/gmd-10-2397-2017, 2017.

Maindonald J, Braun J: Data analysis and graphics using R: an example-based approach.Cambridge (United Kingdom), Cambridge University Press, 2006.

Manders, A. M. M., van Meijgaard, E., Mues, A. C., Kranenburg, R., van Ulft, L. H., and Schaap, M.: The impact of differences in large-scale circulation output from climate models on the regional modeling of ozone and PM, Atmos. Chem. Phys., 12, 9441–9458, doi:10.5194/acp-12-9441-2012, 2012.

Manders, A. M. M., Builtjes, P. J. H., Curier, L., Denier van der Gon, H. A. C., Hendriks, C., Jonkers, S., Kranenburg, R., Kuenen, J., Segers, A. J., Timmermans, R. M. A., Visschedijk, A., Wichink Kruit, R. J., Van Pul, W. A. J., Sauter, F. J., van der Swaluw, E., Swart, D. P. J., Douros, J., Eskes, H., van Meijgaard, E., van Ulft, B., van Velthoven, P., Banzhaf, S., Mues, A., Stern, R., Fu, G., Lu, S., Heemink, A., van Velzen, N., and Schaap, M.: Curriculum Vitae of the LOTOS-EUROS (v2.0) chemistry transport model, Geosci. Model Dev. Discuss., https://doi.org/10.5194/gmd-2017-88, in review, 2017.

Mar, K. A., Ojha, N., Pozzer, A. and Butler, T. M.: Ozone air quality simulations with WRF-Chem (v3.5.1) over Europe: model evaluation and chemical mechanism comparison, Geosci. Model Dev., 9, 3699- 3728, 10.5194/gmd-9-3699-2016, 2016.

Meijgaard, E. v., van Ulft, L. H., Lenderink, G., de Roode, S. R., Wipfler, L., Boers, R., and Timmermans, R. M. A.: Refinement and application of a regional atmospheric model for climate scenario calculations of Western Europe, KvR 054/12, 44, 2012.

Meleux, F., Solmon, F., and Giorgi, F.: Increase in summer European ozone amounts due to climate change, Atmos. Environ., 41, 7577–7587, doi:10.1016/j.atmosenv.2007.05.048, 2007.

Millán, M. M., Sanz, M. J., Salvador, R., and Mantilla, E.: Atmospheric dynamics and ozone cycles related to nitrogen deposition in the western Mediterranean, Environ. Pollut., 118, 167–186, 2002.

Mills, G., Hayes, F., Jones, M. L. M., and Cinderby, S.: Identifying ozone-sensitive communities of (semi-)natural vegetation suitable for mapping exceedance of critical levels, Environ. Pollut., 146, 736–743, doi:10.1016/j.envpol.2006.04.005, 2007.

Mircea, M., Grigoras, G., D'Isidoro, M., Righini, G., Adani, M., Briganti, G., Ciancarella, L., Cappelletti, A., Calori, G., Cionni, I., Cremona, G., Finardi, S., Larsen, B. R., Pace, G., Perrino, C., Piersanti, A., Silibello, C., Vitali, L., and Zanini, G.: Impact of grid resolution on aerosol predictions: a case study over Italy, Aerosol and Air Quality Research, 1253–1267, doi: 10.4209/aaqr.2015.02.0058, 2016.

Monks, P. S.: A review of the observations and origins of the spring ozone maximum, Atmos. Environ., 34, 3545–3561, 2000.

Monks, P. S., Archibald, A. T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K. S., Mills, G. E., Stevenson, D. S., Tarasova, O., Thouret, V., von Schneidemesser, E., Sommariva, R., Wild, O., and Williams, M. L.: Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer, Atmos. Chem. Phys., 15, 8889-8973, https://doi.org/10.5194/acp-15-8889-2015, 2015.

Ordóñez, C., Mathis, H., Furger, M., Henne, S., Hoglin, C., Staehelin, J., Prevot, A.S.H.: Changes of daily surface ozone maxima in Switzerland in all seasons from 1992 to 2002 and discussion of summer 2003. Atmos. Chem. Phys. 5, 1187– 1203, 2005.

Porter W C, Heald C L, Cooley D and Russell B 2015 Investigating the observed sensitivities of air quality extremes to meteorological drivers via quantile regression Atmos. Chem. Phys. Discuss. 15 10349–66, 2015.

Pusede S E et al. : On the temperature dependence of organic reactivity, nitrogen oxides, ozone production, and the impact of emission controls in San Joaquin Valley, California Atmos. Chem. Phys. 14 3373–95, 2014.

Querol, X., Gangoiti, G., Mantilla, E., Alastuey, A., Minguillón, M. C., Amato, F., Reche, C., Viana, M., Moreno, T., Karanasiou, A., Rivas, I., Pérez, N., Ripoll, A., Brines, M., Ealo, M., Pandolfi, M., Lee, H.-K., Eun, H.-R., Park, Y.-H., Escudero, M., Beddows, D., Harrison, R. M., Bertrand, A., Marchand, N., Lyasota, A., Codina, B., Olid, M., Udina, M., Jiménez-Esteve, B., Soler, M. R., Alonso, L., Millán, M., and Ahn, K.-H.: Phenomenology of high-ozone episodes in NE Spain, Atmos. Chem. Phys., 17, 2817-2838, https://doi.org/10.5194/acp-17-2817-2017, 2017.

Rasmussen, D. J., Fiore, A. M., Naik, V., Horowitz, L. W., McGinnis, S. J., and Schultz, M. G.: Surface ozone-temperature relationships in the eastern US: A monthly climatology for evaluating chemistry-climate models, Atmos. Environ., 47, 142–153, doi:10.1016/j.atmosenv.2011.11.021, 2012.

Robertson, L., Langner, J., and Engardt, M.: An Eulerian Limited-Area Atmospheric Transport Model, Journal of Applied Meteorology, 38, 190-210, 1999.

Rust H, Maraun D. and Osborn T.: Modelling seasonality in extreme precipitation Eur. Phys. J. Special Topics 174 99–111, 2009.

Rust H. W., Vrac M., Sultan B., and Lengaigne M.: Mapping weather-type influence on Senegal precipitation based on a spatial-temporal statistical model. J. Climate, 26:8189–8209. ISSN 0894-8755. URL http://dx.doi.org/10.1175/JCLI-D-12-00302.1.1. 2013

Schaap, M., Timmermans, R. M. A., Roemer, M., Boersen, G. A. C., Builtjes, P., Sauter, F., Velders, G., and Beck, J.: The LOTOS-EUROS model: description, validation and latest developments, International Journal of Environment and Pollution, 32, 270-290, 2008.

Schaap M., Cuvelier C, Hendriks C., Bessagnet B., Baldasano J.M., Colette A., Thunis P., Karam D., Fagerli H., Graff A., Kranenburg R., Nyiri A., Pay M.T., Rouïl L., Schulz M., Simpson D., Stern R., Terrenoire E., Wind P.: Performance of European chemistry transport models as function of horizontal resolution, Atmospheric Environment, 112, 90-105, http://dx.doi.org/10.1016/j.atmosenv.2015.04.003, 2015.

Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X. Y., Wang, W., and Powers, J. G.: A Description of the Advanced Research WRF Version 3, NCAR, 2008.

Schnell, J. L., Holmes, C. D., Jangam, A., and Prather, M. J.: Skill in forecasting extreme ozone pollution episodes with a global atmospheric chemistry model, Atmos. Chem. Phys., 14, 7721–7739, doi:10.5194/acp-14-7721-2014, 2014.

Schnell, J. L., Prather, M. J., Josse, B., Naik, V., Horowitz, L. W., Cameron-Smith, P., Bergmann, D., Zeng, G., Plummer, D. A., Sudo, K., Nagashima, T., Shindell, D. T., Faluvegi, G., and Strode, S. A.: Use of North American and European air quality networks to evaluate global chemistry–climate modeling of surface ozone, Atmos. Chem. Phys., 15, 10581-10596, doi:10.5194/acp-15-10581-2015, 2015.

Seo, J., Youn, D., Kim, J. Y., and Lee, H.: Extensive spatiotemporal analyses of surface ozone and related meteorological variables in South Korea for the period 1999–2010, Atmos. Chem. Phys., 14, 6395-6415, https://doi.org/10.5194/acp-14-6395-2014, 2014.

Sillman, S. and Samson, P.J: Impact of temperature on oxidant photochemistry in urban, polluted rural and remote environments. Journal of Geophysical Research 100: doi: 10.1029/94JD02146. issn: 0148-0227, 1995.

Solberg, S., R. G. Derwent, Ø. Hov, J. Langner, and A. Lindskog: European abatement of surface ozone in a global perspective, Ambio, 34, 47–53, 2005.

Solberg, S., Hov, Ø., Sovde, A., Isaksen, I. S. A., Coddeville, P., De Backer, H., Forster, C., Orsolini, Y., and Uhse, K.: European surface ozone in the extreme summer 2003, J. Geophys. Res.Atmos., 113, D07307, doi:10.1029/2007jd009098, 2008.

Solberg, S., Colette, A., and Guerreiro, C. B. B.: Discounting the impact of meteorology to the ozone concentration trends. ETC/ACM, NILU, INERIS. https://doi.org/10.13140/rg.2.2.15389.92649, 2016.

Steiner, A.L., Tonse, S., Cohen, R.C., Goldstein, A.H., Harley, R.A.: Influence of future climate and emissions on Regional air quality in California. Journal of Geophysical Research-Atmospheres 111, 2006.

Vautard, R., Honore, C., Beekmann, M., and Rouil, L.: Simulation of ozone during the August 2003 heat wave and emission control scenarios, Atmos. Environ., 39, 2957–2967, doi:10.1016/j.atmosenv.2005.01.039, 2005.

Tang L., Chen D.L., Karlsson P.E., Gu Y.F., Ou T.H. Synoptic circulation and its influence on spring and summer surface ozone concentrations in Southern Sweden Boreal Environment Research, 14, 889-902, 2009.

Tarasova, O. A., Brenninkmeijer, C. A. M., Jöckel, P., Zvyagintsev, A. M., and Kuznetsov, G. I.: A climatology of surface ozone in the extra tropics: cluster analysis of observations and model results, Atmos. Chem. Phys., 7, 6099–6117, doi:10.5194/acp-7-6099-2007, 2007.

Thompson M L, Reynolds J, Cox L H, Guttorp P and Sampson P D: A review of statistical methods for the meteorological adjustment of tropospheric ozone Atmos. Environ. 35 617–30, 2001.

van Loon, M., Vautard, R., Schaap, M., Bergström, R., Bessagnet, B., Brandt, J., Builtjes, P. J. H., Christensen, J. H., Cuvelier, C., Graff, A., Jonson, J. E., Krol, M., Langner, J., Roberts, P., Rouil, L., Stern, R., Tarrasón, L., Thunis, P., Vignati, E., White, L., and Wind, P.: Evaluation of long-term ozone simulations from seven regional air quality models and their ensemble, Atmospheric Environment, 41, 2083-2097, 2007.

Wu, S., Mickley, L. J., Leibensperger, E. M., Jacob, D. J., Rind, D., and Streets, D. G.: Effects of 2000–2050 global change on ozone air quality in the United States, J. Geophys. Res.-Atmos., 113, D18312, doi:10.1029/2007JD009639, 2008.

Zhang, Y.: Online-coupled meteorology and chemistry models: his- tory, current status, and outlook, Atmos. Chem. Phys., 8, 2895– 2932, doi:10.5194/acp-8-2895-2008, 2008.

Supplementary material

A multi-model comparison of meteorological drivers of surface ozone over Europe

Noelia Otero[1,3], Jana Sillmann[2], Kathleen A. Mar[1], Henning W. Rust[3], Wenche Aas[4], Camilla Andersson[5], Magnuz Engardt[5], Bertrand Bessagnet[6], Giancarlo Ciarelli[7], Augustin Colette[6], Florian Couvidat[6], Kees Cuvelier[8], Philippe Thunis[8], Hilde Fagerli[9], Martijn Schaap[10,3], Astrid Manders[10], Mihaela Mircea[9], Maria-Teresa Pay[12], Mark Thobald[12], Valentin Raffort[11], Yelva Roustan[11], , Svetlana Tsyro[6], Marta G. Vivanco[12], Peter Wind[11,13] and Tim Butler[1,3]

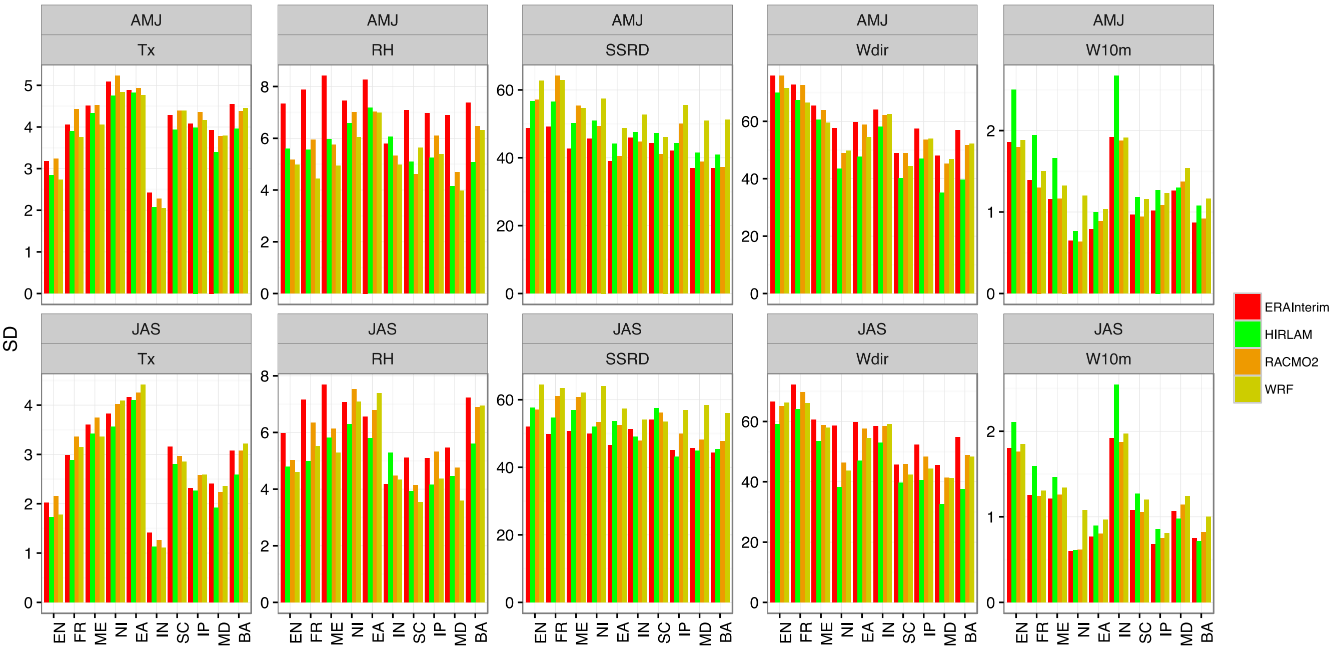


Figure S1. Standard deviations of the meteorological predictors: Maximum temperature (Tx), relative humidity (RH), solar radiation (SSRD), wind direction (Wdir) and wind speed-10m (W10m). Standard deviations are computed for each season, AMJ (top) and JAS (bottom), and for each region: England (EN), France (FR), Mid-EU (ME), NI (North Italy), EA (East-EU), IN (Inflow), SC (Scandinavia), IP (Iberian Peninsula), MD (Mediterranean) and Balkans (BA).

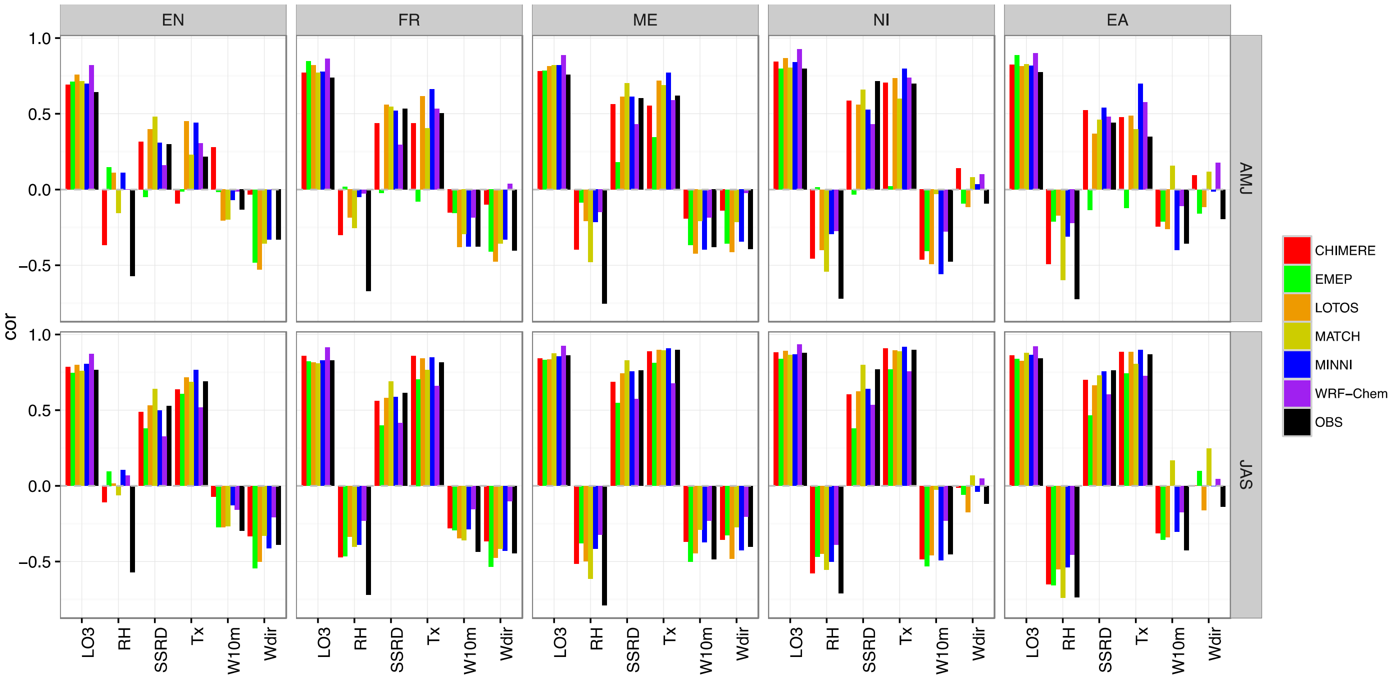


Figure S2. Correlation coefficients between MDA8 O3 and each potential predictor used in the MLR. Correlations are computed for each season, AMJ (top) and JAS (bottom), and for internal regions: England (EN), France (FR), Mid-EU (ME), NI (North Italy), EA (East-EU).

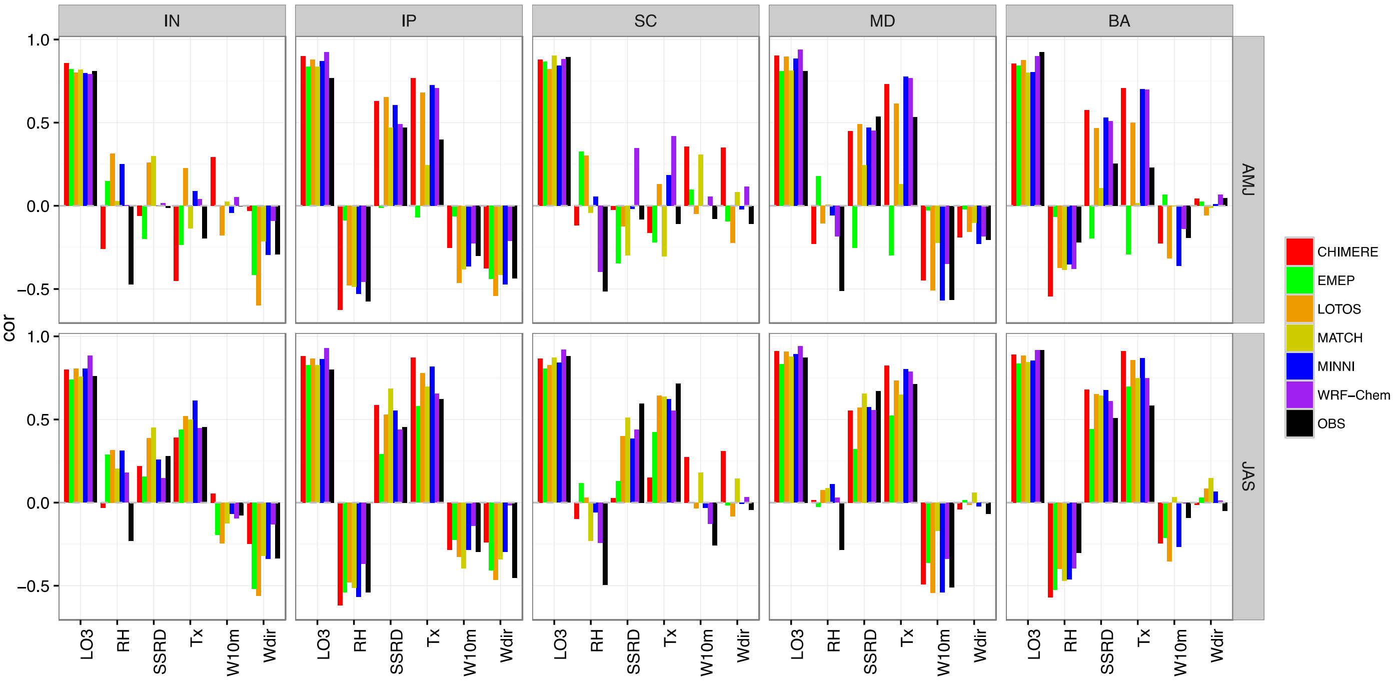


Figure S3. Correlation coefficients between MDA8 O3 and each potential predictor used in the MLR. Correlations are computed for each season, AMJ (top) and JAS (bottom), and for external regions: IN (Inflow), SC (Scandinavia), IP (Iberian Peninsula), MD (Mediterranean) and Balkans (BA).

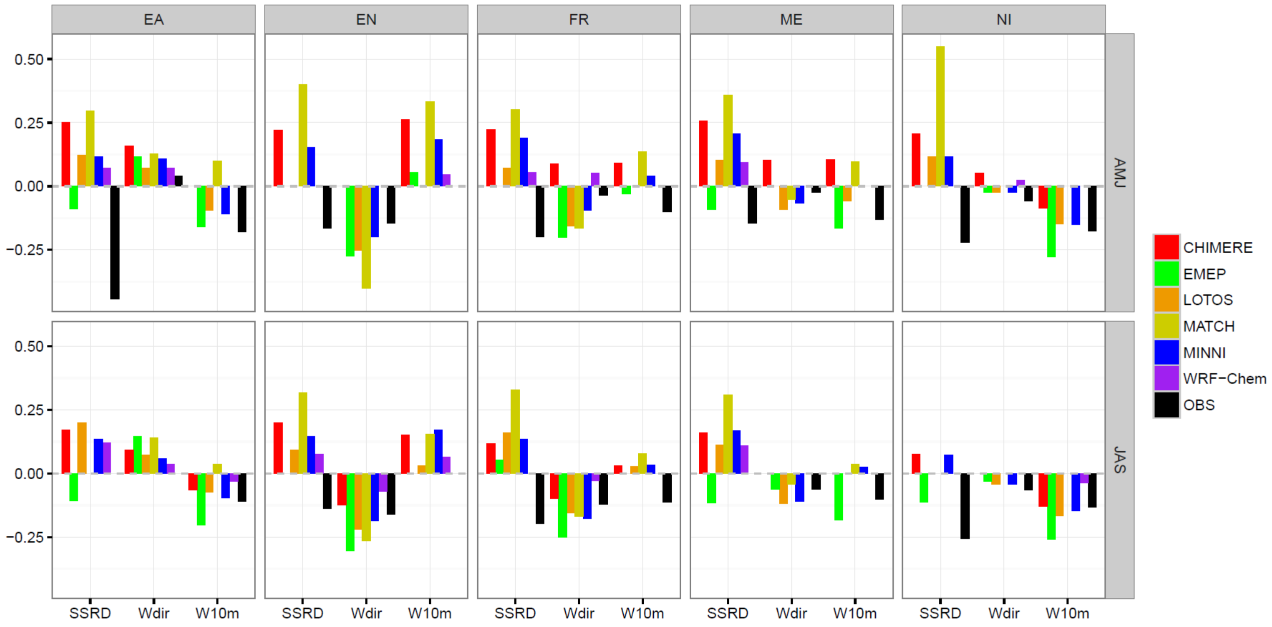


Figure S4. Standardised coefficients values of the rest of the meteorological predictors (SSRD, Wdir and W10m) for each CTM-based and observation-based MLR in AMJ (top) and JAS (bottom) and for the internal regions: England (EN), France (FR), Mid-Europe (ME), North Italy (NI) and East-Europe (EA).

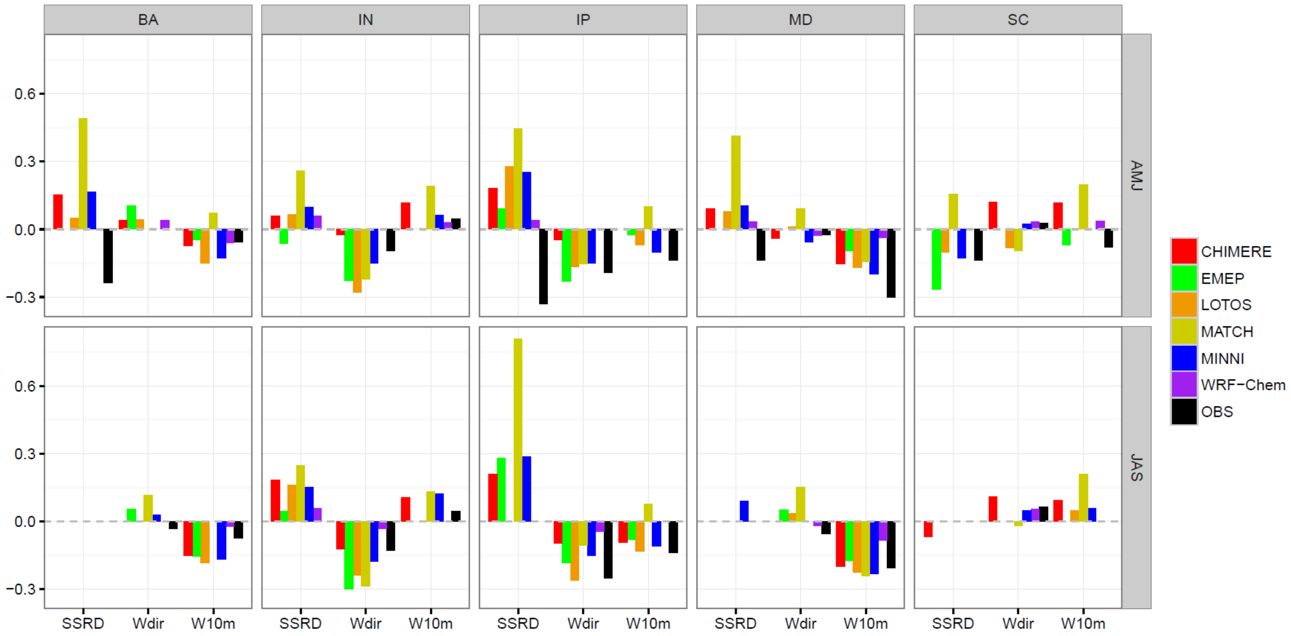


Figure S5. Standardised coefficients values of the rest of the meteorological predictors (SSRD, Wdir and W10m) for each CTM-based and observation-based MLR in AMJ (top) and JAS (bottom) and for the external regions: Inflow (IN), Iberian Peninsula (IP), Scandinavia (SC), Mediterranean (ME) and Balkans (BA).

Chapter 6

Summary and Outlook

6.1 General summary

This dissertation has investigated the influence of synoptic conditions on surface ozone concentrations, indirectly by analysing the links between synoptic patterns and maximum temperatures, and directly by using of the airflow indices derived from the classification of weather types. In addition, the influence of local meteorological conditions has been assessed. Several research questions were raised (section 1.5) and they have been addressed in the previous chapters. In the following, the main conclusions obtained for each set of the research questions are summarised.

Do state-of-the-art climate models realistically reproduce the occurence of weather types over Europe? Based on their representation, what are the expected changes in the frequency of weather types under future climate projections? How do synoptic patterns relate with temperature anomalies in the present and future climate conditions?

As shown in previous works (e.g. Kenawy and McCabe, 2016; Lorenzo et al., 2011; Perez et al., 2014) atmospheric circulation patterns can be an useful tool for evaluating climate model simulations and their projections under future climate conditions. The first study of this dissertation assesses the capability of models to reproduce the occurrence of weather types, which is essential to provide a greater confidence when examining changes in large-scale circulation under future climate, and particularly the implications on future air quality. The multi-model ensemble (MME) mean represents reasonably well the main features of large-scale atmospheric circulation, in terms of relative frequencies of WT, when compared to the ERA-Interim in the near-present (1986-2005). However, notable seasonal and regional discrepancies between the MME mean and ERA-Interim are found for certain weather types, such as a significant model underestimation of Low-Flow days in summer over South Europe and an overestimation of Westerlies in winter over Central

Europe. Similarly, in winter the MME overestimates the occurrence of Cyclonic days in Northwest and Central-East Europe, while Anticyclonic days are underestimated.

The projected future changes of WT frequencies using the emission scenario RCP8.5 show that the changes in the occurrence frequency for the late twenty-first century (2081-2100) would have a major impact for some weather types over South Europe. Specifically, Anticyclonic days would increase throughout the year (except in summer) over the Mediterranean basin. Consistently with previous studies (e.g Donat et al., 2010; van Ulden and van Oldenborgh, 2006), our results suggest an increase of the occurrence of Westerlies mostly in winter over Central Europe, which are associated with positive anomalies of maximum temperatures. Similarly, in summer and autumn an increase of Low Flow conditions, associated with warmer temperatures, is found over South Europe, especially over the Mediterranean basin. Particularly, such increasing Low Flow days might lead to an increase in the occurrence of stagnant conditions (i.e. weak circulation, low wind), favouring episodes of air pollution.

Finally, the contribution to the projected temperatures of frequency-related changes is considerably small. Hence, changes in European temperatures must be mainly driven by changes in the weather types themselves (within-type variations), likely affected also by global warming.

How do regression-based models represent the influence of synoptic and local meteorological conditions on surface ozone over Europe? What are the main drivers of the variability of surface ozone over Europe and how do they influence the observed variability?

To address this question, three regression methods were applied in order to examine the drivers of maximum daily 8-hour average ozone (MDA8 O_3) concentrations, when considering (i) ozone means, (ii) the high percentile of the ozone distribution, and (iii) ozone exceedances based on the air quality guideline thresholds. While most of the previous studies have analysed the relationship between ozone and particular meteorological and/or synoptic conditions at specific locations, this study provides a comprehensive analysis of the spatial influence on ozone over the whole European domain.

The combination of the airflow indices and meteorological variables within the regression-synoptic technique shows that local meteorological conditions have a larger influence on ozone than synoptic conditions. The results reveal distinct seasonal and regional patterns in terms of statistical performance and the dominance of ozone's drivers. For example, the statistical models perform better in summer (June-July-August, JJA) than in spring (March-

April-May, MAM) and particularly over northwestern, central and southeast regions. In the latter, the inclusion of the ozone persistence (ozone concentrations from the pervious day) as a predictor, improves significantly the statistical model performance, while the meteorological drivers account for most of the explained variance of ozone in most of the grid cells over central and northwest Europe.

Regarding the relative importance of drivers, the results show that the ozone persistence is the main driver over the southern regions, especially in MAM, where the climatic factors (airflow indices and meteorological variables) have a minor effect. It must be highlighted that over those regions, in particular over the Mediterranean basin, the meteorology driving ozone variability is markedly influenced by the complexity of the orography and mesoscale processes (such as strong land-sea breezes) (e.g. Querol et al., 2017; Richards et al., 2013). However, these effects are not well captured by the regression models, in which the ozone persistence has the largest contribution, explaining most of the proportion of ozone variability over the southern European regions. Maximum temperature is the most dominant driver in JJA in some regions (e.g. central and northwest Europe), where MDA8 O_3 is particularly sensitive to maximum temperature. This could be due to the effect of temperature on emissions of VOCs in those regions, in which previous studies showed a VOC-limited regimen (Beekmann and Vautard, 2010). With a minor, but also significant contribution, relative humidity and solar radiation are found key drivers in specific grid-points over the western and central regions in spring. The role of these drivers over central locations is reflected in the different statistical models used in this study.

Since climate change is expected to alter regional meteorological conditions, the analyses presented in the first two studies (Chapter 3 and 4) provide further insights into the impacts of the key meteorological drivers of ozone over Europe, especially during the warmer months. In particular, these results point out the vulnerability of central European regions to future episodes of ozone pollution under a warmer climate.

Are AQ models able to capture the basic relationship between meteorological conditions and ozone? Do AQ capture the drivers of ozone derived from observations? In particular, do they represent realistically the climate penalty?

Following a similar procedure than in the previous study, a MLR is built separately for time series from observations (observation-based) and air quality models (CTM-based) over ten European regions. In this case, the seasons differ from the meteorological definition used in the previous study, but cover the period when ozone typically reaches its highest concentrations(i.e. April-September). Here, the seasons are defined as: spring (April-May-June, AMJ) and summer (July-August-September, JAS).

Similarly to the results described above, this study also reveal differences in terms of statistical performance and ozone's drivers between seasons and regions. Most of the MLRs perform better in JAS than in AMJ, which could be attributed to the major role of meteorology in JAS influencing local photochemistry processes of ozone production, while in AMJ long range transport plays a stronger role (Monks, 2000; Tarasova et al., 2007). Certain regions such as, England, France, Mid-Europe, North Italy and East-Europe (here referred to as internal regions), show a strong influence of local meteorological conditions. On the contrary, in the rest of the regions, Inflow, Scandinavia, Iberian Peninsula, Mediterranean and Balkans (referred to as external) the local meteorological factors do not appear to play a significant role.

Overall, CTMs are in better agreement with the observations in the internal regions than in the external regions, where they present more discrepancies with the observations. The differences in the external regions might be due to several reasons, such as, (i) a larger influence of dynamical processes, (ii) a stronger influence of boundary conditions applied to the CTMs, and (iii) the interpolation of a coarser grid to capture mesoscale processes that influence MDA8 O_3 (e.g. sea-land breezes). In some cases (i.e. Balkans), it is important to highlight that such differences might be also caused by a lower density of observing sites from which the interpolated product used here is derived, which can result in a such product (Schnell et al., 2014).

The analysis of the drivers' contribution and sensitivities in the observed-based MLRs shows that the dominant drivers in the internal regions are maximum temperature and relative humidity. While the contribution of relative humidity is stronger in AMJ, the influence of maximum temperature becomes stronger in JAS, which is reflected in the values of the standardised coefficients obtained in the MLRs. On the contrary, the ozone persistence (ozone from the previous day) has the largest contribution in the external regions, especially in AMJ, when MDA8 O_3 shows the highest sensitivities to the ozone persistence. The CTMs are able to reproduce the same tendency as the observations regarding the influence of maximum temperature and ozone persistence, although with discrepancies among the regions (and CTMs), especially in the external regions, as mentioned before. All of the CTMs analysed in this study present deficiencies to capture the strength of the relationship ozone-relative humidity. Here, we speculate that the impacts of ozone dry deposition may play a role in explaining the problems of CTMs to reproduce the relationship ozone-relative humidity detected in the observational dataset.

Finally, the assessment of the slopes of relationship the ozone-temperature (climate penalty) reveals differences among the CTMs and regions when compared to the observations. As expected, the largest differences are found in the external regions, where in some cases, CTMs differ in both magnitude and sign, mostly in AJM. While, most of the CTMs consistently show the climate penalty in JAS, they tend to overestimate it in AMJ (especially in the external regions).

6.2 Outlook

The work documented in this dissertation provided a comprehensive characterisation of the influence of large-scale atmospheric circulation and local meteorological conditions on European ozone concentrations. As stated in the introduction, GCMs and AQ models are essential tools to investigate the impacts of climate change on air pollution. Then, evaluating the models' performances is required to provide greater confidence in their use. By applying two different statistical approaches, this study also offers a multi-model evaluation within the framework of global and regional modelling. Based on the methods presented here and the main findings described in the previous chapters, a number of recommendations and suggestions can be drawn for future work, which are detailed below.

The new JC extended version has been successfully applied over the whole European domain providing an effective tool to evaluate GCMs and to examine potential changes in the frequencies of weather types under future scenarios of climate change. In general, GCMs are able to represent realistic synoptic patterns when comparing to reanalysis datasets. However, GCMs show some regional and seasonal limitations in reproducing the frequencies of some weather types. Special attention must be focused on those patterns related to warmer temperatures and indirectly affecting air quality. For instance, GCMs show serious limitations in reproducing low flow conditions in summer and autumn over South Europe (e.g. the Mediterranean Basin). This could be partially attributed to a misrepresentation of mean sea-level pressure giving rise to an underestimation of low flow conditions. It is essential to improve the representation of large-scale circulation variables (e.g. mean sea level pressure) in GCMs to reproduce realistic synoptic patterns.

The analysis of the relationship between weather types and temperature anomalies provides valuable information in the context of air quality, identifying synoptic situations that usually favour episodes of ozone pollution (e.g. warmer temperatures under low flow or anticyclonic days in summer). Our results indicate that the projected changes in European temperatures can be attributed to changed characteristics in the patterns themselves (within-type variations), rather than due to changes in the frequencies of weather types. Quantifying the contribution of both non-dynamic (including physical processes) and dynamic mechanisms in the within-type variations was beyond of the scope of this study.

However, a forthcoming study could adopt a more sophisticated approach to investigate the contribution of the within-type variations and their impacts in projected future temperatures, which are expected to influence future air quality.

As shown in previous studies (e.g. Kyselý, 2007), the persistence of certain circulation patterns is associated with surface air temperature anomalies, and the occurrence of episodes of extreme temperature are pronounced under persistent circulation. A recent study (Schnell and Prather, 2017) showed that extreme temperatures and air pollution (specifically, O_3 and PM_{10}) episodes are often time-space overlapped. Then, another possible direction for exploiting the usefulness of the extended JC classification developed in this study, could be focused on investigating the overlap between the occurrence of persistent weather types and specific episodes of temperature extremes and air pollution. Indeed, this approach can be applied to other pollutants, such as NO_2 or PM_{10} that have been linked to certain prevailing atmospheric conditions in previous works (e.g. Grundström et al., 2015; Pleijel et al., 2016; Pope et al., 2014).

Based on our findings, the synoptic-regression approach used in this research could be improved and expanded in various ways. For instance, it has been shown that the inclusion of airflow indices might improve the statistical regression models in some locations, but in general they did not show a strong influence on daily ozone concentrations. This points out the limitations of using daily values of airflow indices within our strategy, since the results indicated that they are not able to capture short-term (day-to-day) fluctuations of ozone concentrations, which are more influenced by local meteorological parameters. Possibly the effect of circulation patterns on ozone concentrations could be better captured by using a different time-scale (i.e. monthly or seasonal datasets). Previous studies have shown the link between the inter-annual variability of weather types and ozone concentrations, specifically over Northern regions (e.g. Pleijel et al., 2016; Tang et al., 2009). Thus, it would be interesting to examine the influence of the variability of weather types on ozone at monthly/seasonal (or year-to-year) along with local meteorological conditions. Furthermore, given the close relationships between meteorological and synoptic conditions, another way to expand the synoptic-regression technique can be by adding interaction terms in the general equation (see Chapter 2). The interaction terms would represent the combined effect of the predictors (e.g. frequencies of WT with meteorological variables, such maximum temperature, relative humidity or wind).

The regression methods offer a simple alternative to assess the relationship between ozone and meteorological conditions, in observations and also in AQ model simulations. Nevertheless, the ozone variability can be modulated by others factors, such as photochemical reactions, which are not only controlled by meteorological conditions, but also by emission of ozone precursors. Thereby, this approach can be expanded with the inclusion of ozone precursor datasets (i.e. VOC or NOx). In addition, it is worthy to mention that impacts of changes in land use can influence ozone concentrations through ozone precursors (e.g. VOC or NO) emitted from certain vegetation species and also deposition of ozone under future warmer conditions (Tai et al., 2013). Therefore, further analyses could also consider the effects of future changes in land use that may drive local increases or decreases in ozone pollution.

An issue identified in this procedure is the use of a coarser grid (1°x1°), which might be insufficient to capture some mesoscale processes, (e.g. land-sea breezes). Thus, it would be highly recommended to test this method with a finer grid in order to assess the role of the model resolution. In that sense, it is also recommended the use of data from monitoring sites (without previous interpolation processes).

Ultimately, by identifying some limitations in the ability of current AQ models to reproduce the relationship between models and certain meteorological variables, this dissertation can serve as a basis for future model developments. For instance, one the main findings from the multi-model evaluation is that the AQ models do underestimate the strength of the relationship between ozone and relative humidity detected in the observational dataset. As previous studies suggest (e.g. Kavassalis and Murphy, 2017), we speculate that dry deposition schemes in the models might be playing an important role for representing the observed relationship between ozone and relative humidity. In this sense, this work provides useful information pointing out that future model developments could be focused on sensitivity analyses on the current dry deposition schemes to better understand the impacts of dry deposition on the ozone-relative humidity relationship.

Bibliography

S.A. Abdul-Wahab, C.S. Bakheit, and S.M. Al-Alawi. Principal component and multiple regression analysis in modelling of ground-level ozone and factors affecting its concentrations. *Environmental Modelling and Software*, pages 1263–1271, 2005.

H. R. Anderson, A. Ponce de Leon, J. M. Bland, J. S. Bower, and D. P. Strachan. Air pollution and daily mortality in london: 1987-92. *Br. Med.*, 312(665-9):193–217, 1996. doi: https://doi.org/10.1136/bmj.312.7032.665.

H. R. Anderson, R. G. Derwent, and J. Stedman. Health effects of climate change in the uk. london: Uk department of health. *Air pollution and climate change*, pages 193–217, 2001.

C. Andersson and M. Engardt. European ozone in a future climate: Importance of changes in dry deposition and isoprene emissions. *J. Geophys. Res. Atmos.*, 115(D02303), 1996. doi: 10.1029/2008jd011690,2010.

M. Ashmore. Assessing the future global impacts of ozone on vegetation. *Plant. Cell. Environ.*, 28:949–964, 2005.

A. Baklanov, K. Schlünzen, P. Suppan, J. Baldasano, D. Brunner, S. Aksoyoglu, G. Carmichael, J. Douros, J. Flemming, R. Forkel, S. Galmarini, M. Gauss, G. Grell, M. Hirtl, S. Joffre, O. Jorba, E. Kaas, M. Kaasik, G. Kallos, X. Kong, U. Korsholm, A. Kurganskiy, J. Kushta, U. Lohmann, A. Mahura, A. Manders-Groot, A. Maurizi, N. Moussiopoulos, S. T. Rao, N. Savage, C. Seigneur, R. S. Sokhi, E. Solazzo, S. Solomos, B. Sørensen, G. Tsegas, E. Vignati, B. Vogel, and Y. Zhang. Online coupled regional meteorology chemistry models in europe: current status and prospects. *Atmos. Chem. Phys.*, 14:317–398, 2014. doi: https://doi.org/10.5194/acp-14-317-2014.

M. A. Barrero, J. O. Grimalt, and L. Canton. Prediction of daily ozone concentration maxima in the urban atmosphere. *Chemometr. Intell. Lab. Syst*, 80:67–76, 2005.

R. G. Barry and A. H. Perry. *Synoptic climatology*. Methuen, London, 1973.

D. Baur, M. Saisana, and N. Schulze. Modelling the effects of meteorological variables on ozone concentration-a quantile regression approach. *Atmos. Environ.*, 38:4689–4699, 2004.

C. Beck and A. Philipp. Evaluation and comparison of circulation type classifications for the european domain. *Physics and Chemistry of the Earth*, 35:374–387, 2010.

M. L Bell, A. McDermott S. L. Zeger, and J. M. Samet F. Dominici. Ozone and short-term mortality in 95 us urban communities, 1987–2000. *JAMA*, 292:2372–2378, 2004.

M. L. Bell, R. D. Peng, and F. Domenici. The exposure-response curve for ozone and risk of mortality and the adequacy of current ozone regulations. *Environ. Health Perspect.*, 114:532–536, 2006.

B. J. Bloomer, J. W. Stehr, C. A. Piety, R. J. Salawitch, and R. R. Dickerson. Observed relationships of ozone air pollution with temperature and emissions. *Geophys. Res. Lett.*, 36:L09803, 2009. doi: 10.1029/2009gl037308.

P. J. Bloomfield, J. A. Royle, L. J. Steinberg, and Q. Yang. Accounting for meteorological e!ects in measuring urban ozone levels and trends. *Atmos. Environ.*, 30:3067–3077, 1996. doi: 10.1029/2009gl037308.

M. Brauer, M. Amann, R. T. Burnett, A. Cohen, F. Dentener, M. Ezzati, S. B. Henderson, M. Krzyzanowski, R. V. Martin, R. Van Dingenen, A. van Donkelaar, and G. D. Thurston. Exposure assessment for estimation of the global burden of disease attributable to outdoor air pollution. *Environ. Health Perspect.*, 46:652–660, 2012. doi: 10.1021/es2025752.

R. T. Burnett, S. Cakmak, J. R. Brook, and D. Krewski. The role of particulate size and chemistry in the association between summertime ambient air pollution and hospitalizations for cardiorespiratory diseases. *Environ. Health Perspect.*, 105:614–20, 1997.

L. Camalier, W. Cox, and P. Dolwick. The effects of meteorology on ozone in urban areas and their use in assessing ozone trends. *Atmos. Environ.*, 41:7127–7137, 2007.

M. Canynová and R. Huth. Atmospheric circulation in uence on climatic trends in europe: an analysis of circulation type classifications from the cost733 catalogue. *Int. J. Climatol.*, 36:2743–2760, 2016.

D. C. Carslaw, S. D. Beevers, and J. E. Tate. Modelling and assessing trends in traffic-related emissions using a generalized additive modelling approach. *Atmos. Environ.*, 41: 5289–5299, 2007.

A. Chaloulakou, D. Assimakopoulos, and T. Lekkas. Forecasting daily maximum ozone concentrations in the athens basin. *Science of the Total Environment*, 56:97–112, 1999.

A. Chaloulakou, M. Saisana, and N. Spyrellis. Comparative assessment of neural networks and regression models for forecasting summertime ozone in athens. *Science of the Total Environment*, 313:1–13, 2003.

W. L. Chameides and J. C. G. Walker. A photochemical theory for tropospheric ozone. *Science of the Total Environment*, 78:8751–8760, 1973.

R. E. Chandler and H. S. Wheater. Analysis of rainfall variability using generalized linear models - a case study from the west of ireland. *Water Resour. Res.*, 38(10), 2002.

D. Chen. Monthly circulation climatology for sweden and its application to a winter temperature case study. *Int. J. Climatol.*, 20:1067–1076, 2000.

Z. H. Chen, S. Y. Cheng, J. B. Li, X. R. Guo, W. H. Wang, and D. S. Chen. Relationship between atmospheric pollution processes and synoptic pressure patterns in northern china. *Water, Air, & Soil Pollution*, 42:6078–6087, 2008.

C. S. Cheng, M. Campbell, Q. Li, A. Auld, N. Day, D. Pengell, S. Gingrich, and D. Yap. A synoptic climatological approach to assess climatic impact on air quality in south-central canada. part i: Historical analysis. *Water, Air, & Soil Pollution*, 182:131–148, 2007.

J. Coates, K. A. Mar, N. Ojha, and T. M. Butler. The influence of temperature on ozone production under varying nox conditions in a modelling study. *Atmos. Chem. Phys.*, 16: 11601–11615, 2016. doi: https://doi.org/10.5194/acp-16-11601-2016.

A. Colette, B. Bessagnet, R. Vautard, S. Szopa, S. Rao, S. Schucht, Z. Klimont, L. Menut, G. Clain, F. Meleux, G. Curci, and L. Rouïl. European atmosphere in 2050, a regional air quality and climate perspective under cmip5 scenarios. *Atmos. Chem. Phys.*, 13: 7451–7471, 2013. doi: 10.5194/acp-13-7451-2013.

A. Colette, C. Andersson, A. Baklanov, B. Bessagnet, J. Brandt, J. Christensen, R. Doherty, M. Engardt, C. Geels, C. Giannakopoulos, G. Hedegaard, E. Katragkou, J. Langner, H. Lei, A. Manders, D. Melas, F. Meleux, , L. Rouil, M. Sofiev, J. Soares, D. Stevenson, M. Tombrou-Tzella K. Varotsos, and P. Young. Is the ozone climate penalty robust in europe? *Environ. Res. Lett.*, 10:084015, 2015. doi: doi:10.1088/1748-9326/10/8/084015.

A. C. Comrie. An enhanced synoptic climatology of ozone using a sequencing technique. *Phys. Geogr.*, 13:53–65, 1992.

A. C. Comrie. Comparing neural networks and regression models for ozone forecasting. *J. Air Waste Manage. Assoc.*, 47:653–63, 1997.

O.R. Cooper, D. D. Parrish, J. Ziemke, N. V. Balashov, M. Cupeiro, I. E. Galbally aand S. Gilge, L. Horowitz, N. R. Jensen, J. F. Lamarque, V. Naik, S. J. Oltmans, J. Schwab, D. T. Shindell, A. M. Thompson, V. Thouret, Y. Wang, and R. M. Zbinden. Global distribution and trends of tropospheric ozone: An observation-based review. *Elem. Sci. Anth.*, 2:000029, 2014. doi: 10.12952/journal.elementa.000029.

N. Cortesi, R. M. Trigo, J. C. González-Hidalgo, and A. M. Ramos. Modelling monthly precipitation with circulation weather types for a dense network of stations over iberia. *Hydrol. Earth Syst. Sci.*, 17:665–678, 2013. doi: 10.5194/hess-17-665-2013.

P. J. Crutzen. Photochemical reactions initiated by and influencing ozone in the unpolluted troposphere. *Tellus*, 26:47–57, 1974.

T. D. Davies, P. M. Kelly, P. S. Low, and C. E. Pierce. Surface ozone concentrations in europe – links with the regional-scale atmospheric circulation. *Geophys. Res. Atmos.*, 97:9819–9832, 1992.

J. Davis, W. Cox, A. Reff, and P. Dolwick. A comparison of cmaq-based and observation-based statistical models relating ozone to meteorological parameters. *Atmos. Environ.*, 45(20):3481–3487, 2011. doi: 10.1016/j.atmosenv.2010.12.060.

J. M. B. Davis, B. K. Eder, D. Nychka, , and Q. Yang. Modeling the effects of meteorology on ozone in houston using cluster analysis and generalized additive models. *Atmos. Environ.*, 32(14/15):2505–2520, 1998.

R. E. Davis and D. A. Gay. A synoptic climatologicalanalysis of air quality in the grand canyon national park. *Atmos. Environ.*, 27A:713–727, 1993.

J.P. Dawson, P. J. Adams, and S. N. Pandis. Sensitivity of ozone to summertime climate in the eastern usa: a modeling case study. *Atmos. Environ.*, 41:1494– 1511, 2007.

U. Dayan and I. Levi. Relationship between synoptic-scale atmospheric circulation and ozone concentrations over israel. *J. Geophys. Res*, 107(D24):4813, 2002. doi: 10.1029/2002JD002147.

U. Dayan, A. Tubi, and I. Levi. On the importance of synoptic clas- si cation methods with respect to environmental phenomena. *Int. J. Climatol.*, 32:681–694, 2012.

M. Demuzere and N. P. M. van Lipzig. A new method to estimate air–quality levels using a synoptic–regression approach. part i: Present–day o3 and pm10 analysis. *Atmos. Environ.*, 44:1341–1355, 2010.

M. Demuzere, J. Vila-Guerau de Arellano R. M. Trigo, and N. P. M. van Lipzig. The impact of weather and atmospheric circulation on o3 and pm10 levels at a rural mid-latitude site. *Atmos. Chem. Phys.*, 9:2695e2714, 2009.

M. Demuzere, P. Kassomenos A., and Philipp. The cost733 circulation type classification software: an example for surface ozone concentrations in central europe. *Theor. Appl. Climatol.*, 105:143–166, 2011. doi: 10.1007/s00704-010-0378-4.

K.L. Denman, G. Brasseur, A. Chidthaisong, P. Ciais, P.M. Cox, R. E. Dickinson, D. Hauglustaine, C. Heinze, E. Holland, D. Jacob, U. Lohmann, S. Ramachandran, P. L. da Silva Dias, S. C. Wofsy, and X. Zhang. *Couplings Between Changes in the Climate System and Biogeochemistry. In: Climate Change 2007: The Physical Science Basis.Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change[Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M.Tignor and H.L. Miller (eds.)].* ambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

T. M. Dessouky and A. F. Jenkinson. An objective daily catalogue of surface pressure, ow and vorticity indices for egypt and it's use in monthly rainfall forecasting. *Meteorol. Res. Bull.*, 11:1–25, 1977.

R. M. Doherty, O. Wild, D. T. Shindell, G. Zeng, I. A MacKenzie, W. J. Collins, D. S. Stevenson A. M. Fiore and, F. J. Dentener, M. G. Schultz, P. Hess, R. G. Derwent, and T. J. Keating. Impacts of climate change on surface ozone and intercontinental ozone pollution: A multi-model study. *J. Geophys. Res. Atmos.*, 118:3744–3763, 2013. doi: 10.1002/jgrd.50266.

R. Dole, M. Hoerling, J. Perlwitz, J. Eischeid, P. Pegion, T. Zhang, X. W. Quan, T. Xu, and D. Murray. Was there a basis for anticipating the 2010 russian heat wave? *Geophys. Res. Lett.*, 38:L06702, 2011. doi: 10.1029/2010GL046582.

P. Domonkos, J. Kyselý, K. Riotrowicz, P. Petrovic, and T. Likso. ariability of extreme temperature events in south-centraleurope during the 20th century and its relationship withlarge scale circulation. *Int. J. Climatol.*, 23):987–1010, 2003.

M. Donat, G. C. Leckebusch, J. G. Pinto, and U. Ulbrich. Examination of wind storms over central europe with respect to circulation weather types and nao phases. *Int. J. Climatol.*, 30(9):1289– 1300, 2010. doi: 10.1002/joc.1982.

C. Dueñas, M. C. Fernández, S. Cañete, J. Carretero, and E. Liger. Assessment of ozone variations and meteorological effects in an urban area in the mediterranean coast. *Science of The Total Environment*, 38299(Issue1):97–113, 2011. doi: http://dx.doi.org/10.1016/S0048-9697(02)00251-6.

EEA. *The European environment — state and outlook 2010:synthesis*. European Environment Agency,Copenhagen, 2010.

EEA. *Air quality in Europe - 2015 report,Technical Report 5/2015*. European Environmental Agency, 2015.

EEA. *Air quality in Europe - 2016 report,Denmark*. European Environmental Agency, 2016.

H. K. Elminir. Dependence of urban air pollutants on meteorology. *Science of The Total Environment*, 350:225–237, 2005.

M. Engardt, R. Bergström, and C. Andersson. Climate and emis- sion changes contributing to changes in near-surface ozone in europe over the coming decades:results from model studies. *Ambio.*, 38:452–458, 2009.

EU. *Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on Ambient Air Quality and Cleaner Air for Europe*. 2008.

J. Fiala, L. Cernikovsky, F. de Leeuw, and P. Kurfuerst. Air pollution by ozone in europe in summer 2003. overview of exceedances of ec ozone threshold values during the summer season april–august 2003 and comparisons with previous years. *Eur. Environ. Agency Copenhagen*, EEA Topic Rep. 3/2003, 2003.

A. H. Fink, T. Brucher, A. Kruger, G. C. Leckebusch, J. G. Pinto, and U. Ulbrich. The 2003 european summer heatwaves and drought—synoptic diagnosis and impacts.weather. *Weather*, 59:209–216, 2004.

A. H. Fink, T. Brucher, A. Kruger, G. C. Leckebusch, J. G. Pinto, and U. Ulbrich. The 2003 european summer heatwaves and drought—synoptic diagnosis and impacts.weather. *Changes in atmospheric constituents and in radiative forcing. In: Solomon, S. (Ed.), Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, 59: 209–216, 2007.

G. Flato, J. Marotzke, B. Abiodun, P. Braconnot, S.C. Chou, W. Collins, P. Cox, F. Driouech, S. Emori, V. Eyring, C. Forest, P. Gleckler, E. Guilyardi, C. Jakob, V. Kattsov, C. Reason, and M. Rummukainen. Evaluation of climate models. in climate change 2013: The physical science basis. contribution of working group i to the fifth assessment report of the intergovernmental panel on climate change. t.f. stocker, d. qin, g.-k. plattner, m. tignor, s.k. allen, j. doschung, a. nauels, y. xia, v. bex, and p.m. midgley. *Eds. Cambridge University Press*, 59:741–882, 2013. doi: 10.1017/CBO9781107415324.020.

H. Flocas, A. Kelessis, C. Helmis, M. Petrakakis, M. Zoumakis, and K. Pappas. Synoptic and local scale atmospheric circulation associated with air pollution episodes in an urban mediterranean area. *Theor. Appl. Climatol.*, 95:265–277, 2009. doi: 10.1007/s00704-008-0005-9.

J. Fuhrer. Ozone risk for crops and pastures in present and future climates. *Naturwissenschaften*, 96:173–194, 2009. doi: 10.1007/s00114-008-0468-7.

R. García-Herrera, D. Paredes, R. M. Trigo, and I. F. Trigo. The outstanding 2004-2005 drought in the iberian peninsula: associated atmospheric circulation. *Journal of Hidrometeorology*, 8:483–498, 2007.

M. Gardner and S. Dorling. Statistical surface ozone models: an improved methodology to account for non-linear behaviour. *Atmos. Environ.*, 34:21–34, 2000.

F. Giorgi. Climate change hot-spots. *Geophysical Research Letters*, 33(8), 2006. doi: 10.1029/2006GL025734.

F. Giorgi and F. Meleux. Modelling the regional effects of climate change on air quality. *C. R. Geoscience*, 339:721–733, 2007.

C. M. Goodess and P. D. Jones. Links between circulation and changes in the characteristics of iberian rainfall. *Int. J. Climatol.*, 22:1593–1615, 2002.

M. Grimalt, M. Tomàs, G. Alomar, J. Martín-Vide, and M. C. Moreno-García. Determination of the jenkinson and collison's weather types for the western mediterranean basin over the 1948–2009 period. *Temp. Anal. Atmos.*, 26(1):75–94, 2012.

M. Grundström, L. Tang, M. Hallquist, H. Nguyen, D. Chen, and H. Pleijel. Influence of atmospheric circulation patterns on urban air quality during the winter. *Atmos. Pollut. Res.*, 6:278e285, 2015.

M. Hart, R. De Dear, and R. Hyde. A synoptic climatology of trop- ospheric ozone episodes in sydney, australia. *Int. J. Climatol.*, 26:635–1649, 2006. doi: doi:10.1002/joc.1332.

T. J. Hastie and R. J.Tibshirani. *Generalized Additive Models*. Chapman & Hall, 1990.

J. Hegarty, H. Mao, and R. Talbot. Synoptic controls on summertime surface ozone in the northeastern united states. *J. Geophys. Res.*, 112:D14306, 2007. doi: 10.1029/2006JD008170.

Ø. Hodnebrog, S. Solberg, F. Stordal, T. M. Svendby, D. Simpson, M. Gauss, A. Hilboll, G. G. Pfister, S. Turquety, A. Richter, J. P. Burrows, and H. A. C. Denier van der Gon. Impact of forest fires, biogenic emissions and high temperatures on the elevated eastern mediterranean ozone levels during the hot summer of 2007. *Atmos. Chem. Phys.*, 12:8727–8750, 2012. doi: https://doi.org/10.5194/acp-12-8727-2012.

D. E. Horton, C. B. Skinner Sand D. ingh, and N.S. Diffenbaugh. Occurrence and persistence of future atmospheric stagnation events. *Nature Climate Change*, 4:698–703, 2014. doi: 10.1038/nclimate2272.

J. Hsu and M. J. Prather. Stratospheric variability and tropospheric ozone. *J. Geophys. Res. Atmos.*, 114:D06102, 2009. doi: 10.1029/2008jd010942.

HTAP. *Hemispheric Transport of Air Pollution*. UNECE, Geneva, 2010.

Y. Huang, F. Dominici, and M. L. Bell. Bayesian hierarchical distributed lag models for summer ozone exposure and cardiorespiratory mortality. *Environmetrics*, 16:547–562, 2005.

M. Hulme and E. Barrow. *Climate of the British Isles: present, past and future*. Routledge, London, 1997.

R. Huth. Circulation classification scheme applicable in gcm studies. *Theoretical and Applied Climatology*, 67:1–18, 2000.

R. Huth, C. Beck, A. Philipp, M. Demuzere, Z. Ustrnul, M. Cahynova, J. Kysely, and O. E. Tveito. Classifications of atmospheric circulation patterns. *Annals. NY Acad. Sci.*, 1146: 105–152, 2008.

R. Huth, C. Beck, and O. E. Tveito. Preface of classifications of atmospheric circulation patterns – theory and applications. *Physics and Chemistry of the Earth*, 35:307–308, 2010.

IPCC. *The Physical Science Basis*. Cambridge University Press, Cambridge, 2013.

I. S. A. Isaksen, C. Granier, G. Myhre, T. K. Berntsen, S. B. Dalsoren, M. Gauss, Z. Klimont, R. Benestad, P. Bousquet, W. Collins, T. Cox, V. Eyring, D. Fowler, S. Fuzzi, P. Jockel, P. Laj, U. Lohmann, M. Maione, P. Monks, A. S. H. Prevot, F. Raes, A. Richter, B. Rognerud, M. Schulz, D. Shindell, D. S. Stevenson, T. Storelvmo, W. C. Wang, M. van Weele, M. Wild, and D. Wuebbles. Atmospheric composition change: Climate-chemistry interactions. *Atmos. Environ.*, 43:5138–5192, 2009. doi: 10.1016/j.atmosenv.2009.08.003.

D. J. Jacob and D. A. Winner. Effect of climate change on air quality. *Atmos. Environ.*, 43: 51–63, 2009. doi: 10.1016/j.atmosenv.2008.09.051.

D. J. Jacob, J. A. Logan, G. M. Gadner, R. M. Yevich, C. M. Spivakovsky, and S. C. Wofsy. Factors regulating ozone over the united states and its export to the global atmosphere. *Geophys. Res.*, 98:14,817–14,826, 1993.

A. F. Jenkinson and F. P. Collison. *An initial climatology of gales over the North Synoptic climatology*, volume 62. Branch Memorandum, UK Met. Office, Bracknell, 1977.

M. Jerrett, R. T. Burnett, C. A. Pope, K. Ito, G. Thurston, D. Krewski, Y. L. Shi, E. Calle, and M. Thun. Long-term ozone exposure and mortality. *N. Engl. J. Med.*, 360:1085–1095, 2009. doi: 10.1056/NEJMoa0803894.

C.E. Johnson, W. J. Collins, D. S. Stevenson, and R. G. Derwent. The relative roles of climate and emissions changes on future oxidant concentrations. *J. Geophys. Res.*, 104: 18,631–18,645, 1999.

P.D. Jones, M. Hulme, and K. R. Briffa. A comparison of lamb circulation types with an objective classification scheme. *Int. J. Climatol.*, 13:655–663, 1993.

P.D. Jones, C. Harpham, and K. R. Briffa. Lamb weather types derived from reanalysis products. *Int. J. Climatol.*, 33:1129–1139, 2013. doi: 10.1002/joc.3498.

G. Kallos and R. A. Pielke. Synoptic and mesoscale weather conditions during air pollution episodes in athens, greece. *Bound. Layer Meteorol.*, 62:163–184, 1993.

P. A. Kassomenos. Synoptic circulation control on wild reoccurrence. *Phys. Chem. Earth*, 35:544–552, 2010.

P. A. Kassomenos, A. Gryparis, E. Samoli, K. Katsougianni, S. Lykoudis, and H. Flocas. tmospheric circulation types and daily mortality in athens, greece. *Environ. Health Perspect.*, 109:591–596, 2001.

S. C. Kavassalis and J. G. Murphy. Understanding ozone-meteorology correlations: A role for dry deposition. *Geophys. Res. Lett.*, 44:2922–2931, 2017. doi: 10.1002/2016GL071791.

A. M. El Kenawy and M. F. McCabe. Future projections of synoptic weather types over the arabian peninsula during the twenty-first century using an ensemble of cmip5 models. *Theor. Appl. Climatol.*, 2016. doi: 10.1007/s00704-016-1874-y.

R. Koenker. *Quantile regression*, volume 38 of *Economic Society Monograph*. Cambridge University press, 2005.

R. Koenker and G. Basset. Regression quantiles. *Econometrica*, 46:33–50, 1978.

J. Kyselý. Implications of enhanced persistence of atmospheric circulation for the occurrence and severity oftemperature extremes. *Int. J. Climatol.*, 65:689–695, 2007.

A. A. Lacis, D. J. Wuebbles J. A., and Logan. Radiative forcing of climate by changes in the vertical distribution of ozone. *J. Geophys. Res.*, 95:9971–9981, 2015. doi: 10.1029/JD095iD07p09971.

J. F. Lamarque, P. Hess, L. Emmons, L. Buja, W. Washington, and C. Granier. Tropospheric ozone evolution between 1890 and 1990. *J. Geophys. Res. Atmos*, 110:D08304, 2005. doi: 10.1029/2004jd005537.

J. Langner, R. Bergström, and V.Foltescu. Impact of climate change on surface ozone and deposition of sulphur and nitrogen in europe. *Atmos. Environ.*, 39:1129–1141, 2005.

E. M. Leibensperger, L.J. Mickley, and D. J. Jacob. Sensitivity of u.s. air quality to midlatitude cyclone frequency and implications of 1980–2006 climate change. *Atmos. Chem. Phys.*, 8:7075–7086, 2008.

J. Lelieveld and F. J. Dentener. What controls tropospheric ozone? *J. Geophys. Res.Atmos.*, 105:3531–3551, 2000. doi: 10.1029/1999jd901011.

V. E. P. Lemaire, A. Colette, and L. Menut. Using statistical models to explore ensemble uncertainty in climate impact studies: the example of air pollution in europe. *Atmos. Chem. Phys.*, 16:2559–2574, 2016. doi: 10.5194/acp-16-2559-2016,2016.

M. Leśniok, A. Malarzewski, and T. Niedźwiedź. Classification of circulation types for southern poland with an application to air pollution concentration in upper silesia. *Physics and Chemistry of the Earth, Parts A/B/C*, 35(9):516–522, 2010. doi: http://dx.doi.org/10.1016/j.pce.2009.11.006.

M. N. Lorenzo and J. J. Taboada. Influences of atmospheric variability on freshwater input in galician rias in winter. *Journal of Atmosphere and Ocean Sciences*, 10(4):377–387, 2005.

M. N. Lorenzo, J. J. Taboada, and L. Gimeno. Links between circulation weather types and teleconnection patterns and their influence on precipitation patterns in galicia (nw spain). *Int. Climatol.*, 28(11):1493–1505, 2008.

M. N. Lorenzo, A. M. Ramos, and J. J. Taboada. Changes in present and future circulation types frequency in northwest iberian peninsula. *PLoS One*, 6(1):e16201, 2011. doi: 10.1371/journal.pone.0016201.

Convention LRTAP. *Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads and Levels; and AirPollution Effects, Risks and Trends*. 2010. URL http://icpmapping.org/Latest_update_Mapping_Manual.

D. Maraun, F. Wetterhall, A. M. Ireson, R. E. Chandler, E. J. Kendon, M. Widmann, S. Brienen, H. W. Rust, T. Sauter, M. Themessl, V. K. C. Venema, K. P. Chun, C. M. Goodess, R. G. Jones, C. Onof, M. Vrac, and I. Thiele-Eic. Precipitation downscaling under climate change: Recent developments to bridge the gap between dynamical models and the end user. *Reviews of Geophysics*, 48(3), 2010. doi: 10.1029/2009RG000314.

J. Mata and J. Machado. Firm start-up size: a conditional quantile approach. *European Economic Review*, 40:1305–23, 1996.

F. Meleux, F. Solmon, and F. Giorgi. Increase in summer european ozone amounts due to climate change. *Atmos. Chem. Phys.*, 41:7577–7587, 2007.

L. J. Mickley, D. J. Jacob, B. D. Field, and D. Rind. Effects of future climate change on regional air pollution episodes in the united states. *Geophys. Res. Lett.*, 31:L24103, 2004. doi: doi:10.1029/2004GL021216.

G. Mills, A. Buse, B. Gimeno, V. Bermejo, M. Holland, L. Emberson, and H. Pleijel. A synthesis of aot40-based response functions and critical levels of ozone for agricultural and horticultural crops. *Atmos. Chem. Phys.*, 41:2630–2643, 2007.

G. Mills, H. Pleijel, S. Braun, P. Büker, V. Bermejo, E. Calvo, H.Danielsson, L. Emberson, I. González Fernández, L. Grünhage, H. Harmens, F. Hayes, P. E. Karlsson, and D. Simpson. New stomatal flux-based critical levels for ozone effects on vegetation. *Atm. Envir.*, 45:5064–5068, 2011.

P. S. Monks. A review of the observations and origins of the spring ozone maximum. *Atmos. Environ.*, 34:3545–3561, 2000.

P. S. Monks, A. T. Archibald, A. Colette, O. Cooper, M. Coyle, R. Derwent, D. Fowler, C. Granier, K. S. Law, G. E. Mills, D. S. Stevenson, O. Tarasova, V. Thouret, E. von Schneidemesser, R. Sommariva, O. Wild, and M. L. Williams. Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. *Atmos. Chem. Phys.*, 15:8889–8973, 2015. doi: https://doi.org/10.5194/acp-15-8889-2015.

K. M. Mortimer, L. M. Neas, D. W. Dockery, S. Redline, and I. B. Tager. The effect of air pollution on inner-city children with asthma. *European Respiratory Journal*, 19: 699–705, 2002. doi: 10.1183/09031936.02.00247102.

S. Munir, H. Chen, and K. Ropkins. An investigation into the association of ozone with traffic-related air pollutants using a quantile regression approach. *Environmental Health and Biomedicine Brebbia*, 34:21–32, 2011.

S. Munir, H. Chen, and K. Ropkins. Modelling the impact of road traffic on ground level ozone concentration using a quantile regression approach. *Atmos. Environ*, 60:283–291, 2012.

G. Myhre, D. Shindell, F. M. Bréon, W. Collins, J. Fuglestvedt, J. Huang, D. Koch, J. F. Lamarque, D. Lee, B. Mendoza, T. Nakajima, A. Robock, G. Stephens, T. Takemura, and H. Zhang. Anthropogenic and natural radiative forcing, in: Climate change, the physical science base, contribution of working group 1 to the fifth assessment report of the intergovernmental panel on climate change. 2013.

J. A. Nelder and R. W. M. Wedderburn. Generalized linear models. *Stat. Soc. Ser.*, 135: 370–384, 1972.

C. G. Nolte, A. B. Gilliland, C. Hogrefe, and L. J. Mickley. Linking global to regional models to assess future climate impacts on surface ozone levels in the united states. *J. Geophys. Res.*, 113:D14307, 2008.

G. P. O'Hare and R. Wilby. A review of ozone pollution in the united kingdom and ireland with an analysis using lamb weather types. *The Geographical Journal*, 161:1–20, 1995.

C. Ordóñez, H. Mathis, M. Furger, S. Henne, C. Hoglin, J. Staehelin, and A. S. H. Prevot. Changes of daily surface ozone maxima in switzerland in all seasons from 1992 to 2002 and discussion of summer 2003. *Atmos. Chem. Phys.*, 5:1187– 1203, 2005.

C. Ordóñez, D. Brunner, J. Staehelin, P. Hadjinicolaou J. Pyle, M. Jonas, H. Wernli, and A. Prévôt. Strong influence of lowermost stratospheric ozone on lower tropospheric background ozone changes over europe. *Geophys. Res. Lett.*, 34:L07805, 2007. doi: 10.1029/2006GL029113.

D. Paredes, R. M. Trigo, R. García-Herrera, and I. F. Trigo. Understanding precipitation changes in iberia in early spring: weather typing and storm-tracking approaches. *J. Hydrometeor*, 7:101–113, 2006.

D. D. Parrish, K. S. Law, J. Staehelin, R. Derwent, O. R. Cooper, H. Tanimoto, A. Volz-Thomas, S. Gilge, H. E. Scheel, M. Steinbacher M., and E. Chan. Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes. *Atmos. Chem. Phys.*, 12:11485–11504, 2012. doi: 10.5194/acp-12-11485-2012.

D. D. Parrish, K. S. Law, J. Staehelin, R. Derwent, O. R. Cooper, H. Tanimoto, A. Volz-Thomas, S. Gilge, H. E. Scheel, M. Steinbacher M., and E. Chan. Lower tropospheric ozone at northern midlatitudes: Changing seasonal cycle. *Geophys. Res. Lett.*, 40: 1631–1636, 2013. doi: 10.1002/grl.50303.

J. Perez, M. Menendez, and I. Losada. Evaluating the performance of cmip3 and cmip5 global climate models over the northeast atlantic region. *Clim. Dyn.*, 43 (9-10):2663–2680, 2014.

A. Peters, D. W. Dockery, J. Heinrich, and H. Wichmann. Medication use modifies the health effects of particulate sulfate air pollution in children with asthma. *Environ. Health Perspect.*, 105:430–435, 1997.

A. Philipp, J. Bartholy, C. Beck, M. Erpicum, P. Esteban, X. Fettweis, R. Huth, P. James, S. Jourdain, F. Kreienkamp, T. Krennert, S. Lykoudis, S. Michalides, K. Pianko, D. Rasilla-Álvarez, R. Schiemann, A. Spekat, and F. S. Tyvios. Cost733cat – a database of weather and circulation type classifications. *Phys. Chem. Earth*, 35:360 – 373, 2010.

H. Pleijel, M. Grundström, G. P. Karlsson, P. E. Karlsson, and C. Deliang. A method to assess the inter-annual weather-dependent variability in air pollution concentration and deposition based on weather typing. *Atmospheric Environment*, 126:200–210, 2016. doi: http://dx.doi.org/10.1016/j.atmosenv.2015.11.053.

R. J. Pope, N. H. Savage, M. P. Chipperfield, S. R. Arnold, and T. J. Osborn. The influence of synoptic weather regimes on uk airquality: analysis of satellite column no2. *Atmos. Sci. Lett.*, 15:211–217, 2014. doi: 10.1002/asl2.492.

W. C. Porter, C. L. Heald, D. Cooley, and B. Russell. Investigating the observed sensitivities of air-quality extremes to meteorological drivers via quantile regression. *Atmos. Chem. Phys.*, 15:10,349–10,366, 2015. doi: 10.5194/acp-15-10349-2015.

P. Post, V. Truija, and J. Tuulik. Circulation weather types and their in uence on temperature and precipitation in estonia. *Boreal Env. Res.*, 7:281–289, 2002.

X. Querol, G. Gangoiti, E. Mantilla, A. Alastuey, M. C. Minguillón, F. Amato, C. Reche, M. Viana, T. Moreno, A. Karanasiou, I. Rivas, N. Pérez, A. Ripoll, M. Brines, M. Ealo, M. Pandolfi, H. K. Lee, H. R. Eun, Y. H Park, M. Escudero, D. Beddows, R. M. Harrison, A. Bertrand, N. Marchand, A. Lyasota, B. Codina, M. Olid, M. Udina, B. Jímenez-Esteve, M. R. Soler, L. Alonso, M. Millán, and K. H. Ahn. Phenomenology of high-ozone episodes in ne spain. *Atmos. Chem. Phys.*, 17:2817–2838, 2017. doi: https://doi.org/10.5194/acp-17-2817-2017.

P. N. Racherla and P. J. Adams. Sensitivity of global tropospheric ozone and fine particulate matter concentrations to climate change. *J. Geophys. Res.*, 11:D24103, 2006.

H. Radjavi and P. Rosenthal. *Climate Change 2007 – The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. Chapter 1, His- torical Overview of Climate change, in: Intergovernmental Panel on Climate Change Climate change: The Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the IPCC*. Cambridge University Press, Cambridge, 2007.

D. J. Rasmussen, J. L. Hu, A. Mahmud, and M. J. Kleeman. The ozone-climate penalty: Past, present, and future. *Environ. Sci. Technol.*, 47:14258–14266, 2013. doi: 10.1021/es403446m.

N. A. D. Richards, S. R. Arnold, M. P. Chipperfield, G. Miles, A. Rap, R. Siddans, S. A. Monks, and M. J. Hollaway. The mediterranean summertime ozone maximum: global emis-sion sensitivities and radiative impacts. *Atmos. Chem. Phys.*, 13:2331–2345, 2013. doi: 10.5194/acp-13-2331-2013.

A. Russo, R. Trigo, H. Martins, and M. T. Mendes. No2, pm10 and o3 urban concentrations and its association with circulation weather types in portugal. *Atmos. Environ.*, 89:768–785, 2014.

S. Russo, J. Sillmann, and E. Fischer. Top ten european heatwaves since 1950 and their occurrence in the coming decades. *Environ. Res. Lett.*, 10, 2015.

S. Saavedra, A. Rodríguez, J. J. Taboada, J. A. Souto, and J. J. Casares. Synoptic patterns and air mass transport during ozone episodes in northwestern iberia. *Science of the Total Environment*, 441:97e110, 2012.

A. Santurún, J. C. Gónzalez-Hidalgo, A. Sanchez-Lorenzo, and M. T. Zarrabeitia. Surface ozone concentration trends and its relationship with weather types in spain (2001–2010). *Atmos. Environ.*, 101:10–22, 2014.

U. Schlink, S. Dorling, E. Pelikan, G. Nunnari, G. Cawley, H. Junnien, M. Creig, R. Foxall, and K. Eben. A rigorous inter-comparison of ground-level ozone predictions. *Atmos. Environ.*, 37:3237–3253, 2003. doi: 10.1016/S1352-2310(03)00330-3.

J. L. Schnell and M.J. Prather. Co-occurrence of extremes in surface ozone, particulate matter, and temperature over eastern north america. *Proc.of the National academy of science of United Stated of America*, 114:2854–2859, 2017. doi: 10.1073/pnas.1614453114.

J. L. Schnell, C. D. Holmes, A. Jangam, and M. J. Prather. Skill in forecasting extreme ozone pollution episodes with a global atmospheric chemistry model. *Atmospheric Chemistry and Physics*, 14(15):7721–7739, 2014. doi: 10.5194/acp-14-7721-2014.

J. H. Seinfeld and S. N. Pandis. *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*. John Wiley M. L. & Sons Inc, New York,second edition, 2006.

D. Shindell, G. Faluvegi, A. Lacis, J. Hansen, R. Ruedy, and E. Aguilar. Role of tropospheric ozone increases in 20th century climate change. *J. Geophys. Res. Atmos.*, 111: L04803, 2006. doi: 10.1029/2005jd006348.

D. Shindell, J. C. I. Kuylenstierna, E. Vignati, R. van Dingenen, M. Amann, Z. Klimont, S. C. Anenberg, N. Muller, G. Janssens-Maenhout, F. Raes, J. Schwartz, G. Faluvegi, L. Pozzoli, K. Kupiainen, L. Höglund-Isaksson, L. Emberson, D. Streets, V. Ramanathan, K. Hicks, N. T. K. Oanh, G. Milly, M. Williams, V. Demkine, and D. Fowler. Simultaneously mitigating near-term climate change and improv- ing human health and food security. *Science*, 335:183–189, 2012. doi: 10.1126/science.1210026.

S. Sillman. The relation between ozone, no and hydrocarbons in urban and polluted rural environments. *J. Geophys. Res.*, 33:1821–1845, 1999. doi: http://dx.doi.org/10.1016/S1352-2310(98)00345-8.

S. Sillman and P. J. Samson. Impact of temperature on oxidant pho- tochemistry in urban, polluted rural and remote environments. *J. Geophys. Res.*, 100(D6):11497–11508, 1995. doi: 10.1029/94JD02146.

R. A. Silva, J. J. West, Y. Zhang, S. C. Anenberg, J. F. Lamarque, D. T. Shindell, W. J. Collins, S. Dalsoren, G. Faluvegi, G. Folberth, L. W. Horowitz, T. Nagashima, V. Naik, S. Rumbold, R. Skeie, K. Sudo, T. Takemura, D. Bergmann, P. Cameron-Smith, I. Cionni, R. M. Doherty, V. Eyring, B. Josse, I. A. MacKenzie, D. Plummer, M. Righi, D. S. Stevenson, S. Strode, S. Szopa, and G. Zeng. Global premature mortality due to anthropogenic outdoor air pollution and the contribution of past climate change. *Environmental Research Letters*, 8(3):034005, 2013.

S. Solberg, Ø. Hovo., A. Søvde A., I. S. A. Isaksen, P. Coddeville, H. De Backer, C. Forster, Y. Orsolini, and K. Uhse. European surface ozone in the extreme summer 2003. *J. Geophys. Res.*, 113:D07307, 2008. doi: 10.1029/2007JD009098.

S.I.V. Sousa, F.G. Martins, M.C. Pereira, and M.C.M. Alvim-Ferraz. Prediction of ozone concentrations in oporto city with statistical approaches. *Chemosphere*, 64(7):1141–1149, 2006. doi: 10.1016/j.chemosphere.2005.11.051.

S.I.V. Sousa, J. C. M. Pires, F.G. Martins, M.C. Pereira, and M.C.M. Alvim-Ferraz. Potentialities of quantile regression to predict ozone concentrations. *Chemosphere*, 20: 147–158, 2009.

G. Spellman. The use of an index-based regression model for precipitation analysis on the iberian peninsula. *Theor. Appl. Climatol.*, 66:229–239, 2000.

A. L. Steiner, S. Tonse, R. C. Cohen, A. H. Goldstein, and R. A. Harley. Influence of future climate and emissions on regional air quality in california. *J. Geophys. Res.*, 111: D18303, 2006.

D. S. Stevenson, F. J. Dentener, M. G. Schultz, K. Ellingsen, T. P. C. van Noije, O. Wild, G. Zeng, M. Amann, C. S. Atherton, N. Bell, D. J. Bergmann, I. Bey, T. Butler, J. Cofala, W. J. Collins, R. G. Derwent, R. M. Doherty, J. Drevet, H. J. Eskes, A. M. Fiore, M. Gauss, D. A. Hauglustaine, L. W. Horowitz, I. S. A. Isaksen, M. C. Krol, J. F. Lamarque, M. C. Lawrence, V. Montanaro, J. F. Muller, G. Pitari, M. J. Prather, J. A. Pyle, S. Rast, J. M. Rodriguez, G. Sanderson, N. H. Savage, D. T. Shindell, S. E. Strahan, K. Sudo, and S. Szopa. Multimodel ensemble simulations of present-day and near-future tropospheric ozone. *J. Geophys. Res. Atmos.*, 111:D08301, 2006. doi: 10.1029/2005jd0063383.

D. S. Stevenson, P. J. Young, V. Naik, J. F. Lamarque, D. T. Shindell, A. Voulgarakis, R. B. Skeie, S. B. Dalsoren, G. Myhre, T. K. Berntsen, G. A. Folberth, S. T. Rumbold, W. J. Collins, I. A. MacKenzie, R. M. Doherty, G. Zeng, T. P. C. van Noije, A. Strunk, D. Bergmann, P. Cameron-Smith, D. A. Plummer, S. A. Strode, L. Horowitz, Y. H. Lee, S. Szopa, K. Sudo, T. Nagashima, B. Josse, I. Cionni, M. Righi, V. Eyring, A. Conley, K. W. Bowman, O. Wild, and A. Archibald. Tropospheric ozone changes, radiative forcing and attribution to emissions in the atmospheric chemistry and climate model intercomparison project (accmip). *J. Geophys. Res.*, 13:3063–3085, 2013. doi: 10.5194/acp-13-3063-2013.

A. P. Tai, L. J. Mickley, C. L. Heald, and S. Wu. Effect of co2 inhibition on biogenic isoprene emission: implications for air quality under 2000 to 2050 changes in climate, vegetation, and land use. *Geophys. Res. Lett.*, 40:3479–83, 2013.

L. Tang, P. E. Karisson, Y. Gu, and D. Chen. Synoptic weather types and long-range transport patterns for ozone precursos during high-ozone events in southern sweden. *Ambio.*, 38:459e464, 2009.

L. Tang, D. P. Rayner, and M. Haeger-Eugensson. Have meteorological conditions reduced no2 concentrations from local emission sources in gothenburg? *Water Air and Soil Pollution*, 221:275–286, 2011.

O. A. Tarasova, C. A. M. Brenninkmeijer, P. Jöckel, A. M. Zvyagintsev, and G. I. Kuznetsov. A climatology of surface ozone in the extra tropics: cluster analysis of observations and model results. *Atmos. Chem. Phys.*, 7:6099–6117, 2007. doi: 10.5194/acp-7-6099-2007.

L.H. Tecer, F. Ertürk, and O. Cerit. Development of a regression model to forecast ozone concentration in istanbul city, turkey. *Fresenius Environmental Bulletin*, 12:1133–1143, 2003.

M. L. Thompson, J. Reynolds, L. H. Cox, P. Guttorp, and P. D. Sampson. A review of statistical methods for the meteorological adjustment of tropospheric ozone. *Atmos. Environ.*, 35:617–630, 2001.

R. M. Trigo and C. C. Dacamara. Circulation weather types and their influence on the precipitation regime in portugal. *Int. J. Climatol.*, 20(13):1559–1581, 2000.

M. C. Turner, M. Jerrett, C. A. Pope, D. Krewski, S. M. Gapstur, W. R. Diver, B. S. Beckerman, J. D. Marshall, J. Su, D. L. Crouse, and R. T. Burnett. Long-term ozone exposure and mortality in a large prospective study. american journal of respiratory and critical care medicine. *American journal of respiratory and critical care medicine*, 193(10):1134–1142, 2016. doi: 10.1164/rccm.201508-1633OC.

A. P. van Ulden and G. J. van Oldenborgh. Large-scale atmospheric circulation biases and changes in global climate model simulations and their importance for regional climate scenarios. *Atmos. Chem. Phys.*, 6:863–881, 2006.

K. V. Varotsos, M. Tombrou, and C. Giannakopoulos. Statistical estimations of the number of future ozone exceedances due to climate change in europe. *J. Geophys. Res. Atmos.*, 118, 2013. doi: 10.1002/jgrd.50451.

R. Vautard, P. Yiou, F. D'Andrea, N. de Noblet, N. Viovy, C. Cassou, J. Polcher, P. Ciais, M. Kageyama, and M. Fan. Summertime european heat and drought waves induced by wintertime mediterranean rainfall. *Geophys. Res. Lett.*, 34:L07711, 2013. doi: 10.1029/2006GL028001.

S. M. Vicente-Serrano and J. I. López-Moreno. The influence of atmospheric circulation at different spatial scales on winter drought variability through a semi-arid climatic gradient in northeast spain. *Int. J. Climatol.*, 26:1427–1453, 2006. doi: 10.1002/joc.1387.

S. M. Vicente-Serrano, J. I. López-Moreno, A. Drumond, L. Gimeno, R. Nieto, and E. Morán-Tejeda. Effects of warming processes on droughts andwater resources in the nw iberian peninsula (1930-2006). *Climate Res.*, 48:203–212, 2011. doi: 10.3354/cr01002.

E. von Schneidemesser, P. S. Monks, J. D. Allan, L. Bruhwiler, P. Forster, D. Fowler, A. Lauer, W. T. Morgan, P. Paasonen, M. Righi, K. Sindelarova, and M. A. Sutton. Chemistry and the linkages between air quality and climate change. *Chem. Rev.*, 115: L07711, 2015. doi: 10.1021/acs.chemrev.5b00089.

S. Wagg, G. Mills, F. Hayes, S. Wilkinson, D. Cooper W. J., and Davies. Reduced soil water availability did not protect two competing grassland species from the negative effects of increasing background ozone. *Environ. Pollut.*, 165:91–99, 2012. doi: 10.1016/j.envpol.2012.02.010.

WHO. *Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide*. 2005.

WHO. *Review of Evidence on Health Aspects of Air Pollution-REVIHAAP Project. Technical Report*. Regional Office for Europe, Copenhagen, Denmark, 2013.

R. L. Wilby, C. W. Dawson C. W., and E. M. Barrow. Sdsm – a decision support tool for the assessment of regional climate change impacts. *Environmental Modelling and Software*, 17:147–159, 2002.

R. L. Wilby, S. P. Charles, E. Zorita, B. Timbal, P. Whetton, and L. O. Mearns. The guidelines for use of climate scenarios developed from statistical downscaling methods.supporting material of the intergovernmental panel on climate change. *prepared on behalf of Task Group on Data and Scenario Support for Impacts and Climate Analysis (TGICA)*, 2004.

E. K. Wise. Climate-based sensitivity of air quality to climate change scenarios for the southwestern united states. *Int. J. Climatol.*, 29(1):87–97, 2009. doi: 10.1002/joc.1713.

S. N. Wood. *Generalized Additive Models:An Introduction with R*, volume 66. Chapman & Hall/CRC,Boca Raton, Florida, 2006.

S. Wu, L. J. Mickley, E. M. Leibensperger, D. J. Jacob, D. Rind, and D. G. Streets. Effects of 2000–2050 global change on ozone air quality in the united states. *J. Geophys. Res.Atmos.*, 113:D18312, 2008. doi: 10.1029/2007JD009639.

Z. Yan, S. Bate., R. Chandler, V. Isham, and H. Wheater. An ana-lysis of daily maximum windspeed in northwestern europe using gen-eralized linear models. *J. Clim.*, 15: 2073–2088, 2002.

B. Yarnal. *Synoptic climatology in environmental analysis*. Belhaven Press, London, 1993.

B. Yarnal, A. C. Comrie, B. Frakes, and D. P. Brown. Developments and prospects in synoptic climatology. *Int. J. Climatol.*, 21:1923–1950, 2001.

P. J. Young, A. T. Archibald, K. W. Bowman, J. F. Lamarque, V. Naik, D. S. Stevenson, S. Tilmes, A. Voulgarakis, O. Wild, D. Bergmann, P. Cameron-Smith, I. Cionni, W. J. Collins, S. B. Dalsoren, R. M. Doherty, V. Eyring, G. Faluvegi, L. W. Horowitz, B. Josse, Y. H. Lee, I. A. MacKenzie, T. Nagashima, D. A. Plummer, M. Righi, S. T. Rumbold, R. B. Skeie, D. T. Shindell, S. A. Strode, K. Sudo, S. Szopa, and G. Zeng. Preindustrial to end 21st century projections of tropospheric ozone from the atmospheric chemistry and climate model intercomparison project (accmip). *Atmos. Chem. Phys.*, 13:2063–2090, 2013. doi: 10.5194/acp-13-2063-2013.

Appendix A

Contribution to Paper I

The main topic of this study was formulated by Jana Sillmann and Tim Butler. The implementation and development of the extended version of the JC classification was performed by myself. The literature review, all post-processing of model and reanalyses datasets were conducted by my self. Discussion and interpretation of the results were done with Jana Sillmann and Tim Butler, who also aided to improve the manuscript initially prepared by myself.

Contribution to Paper II

The statistical design for this study was conducted by myself in discussions with Tim Butler and Jana Sillmann. The improvements in the statistical modelling approach were done in discussions with Henning Rust, who also provided the support for the correct statistical model development. The observational dataset for maximum daily 8h average of ozone was provided by Jordan L. Schnell. All processing data, including the statistical models and graphical plotting were mainly performed by myself. Discussions and interpretation of the main results were done during the evaluation meetings with Mark Lawrence, Peter Builtjes and Uwe Ulbrich, among others. I prepared the manuscript, which was improved with the aid of the rest of the co-authors, Jana Sillmann, Henning Rust, Jordan Schnell and Tim Butler.

Contribution to Paper III

The topic of this study was mainly formulated by Tim Butler and myself. The European modelling group from the EDT multi-model chemistry transport experiment facilitated the model outputs. As part of the modelling group, Kathleen A. Mar and I contributed with the WRF-Chem simulations. The model setup was previously defined and developed by Kathleen A. Mar, who provided me the support and aid to run the model simulations by myself used in this study. The literature studies, post-processing of model outputs (including plotting of figures and the analyses) were mainly conducted by myself. I prepared the manuscript that was further improved by the co-authors.

Appendix B

List of publications

- **Otero, N.**, et al. “A model comparison of meteorological drivers of surface ozone over Europe“, 2017 *submitted to ACP*.

- Colette, A., Andersson, C., Manders, A., Mar, K., Mircea, M., Pay, M.-T., Raffort, V., Tsyro, S., Cuvelier, C., Adani, M., Bessagnet, B., Bergström, R., Briganti, G., Butler, T., Cappelletti, A., Couvidat, F., D’Isidoro, M., Doumbia, T., Fagerli, H., Granier, C., Heyes, C., Klimont, Z., Ojha, **Otero, N.**, Schaap, M., Sindelarova, K., Stegehuis, A. I., Roustan, Y., Vautard, R., van Meijgaard, E., Vivanco, M. G., and Wind, P.: EURODELTA-Trends, a multi-model experiment of air quality hindcast in Europe over 1990–2010, *Geosci. Model Dev.*, 10, 3255-3276, https://doi.org/10.5194/gmd-10-3255-2017, 2017

- **Otero, N.**, Sillmann, J. and Butler, T. “Assessment of an extended version of the Jenkinson-Collison classification on CMIP5 models over Europe." *Clim Dyn* doi:10.1007/s00382-017-3705-y, 2017

- **Otero, N.**, Sillmann, J., Schnell, J. L., Rust, H. W., and Butler, T. “Synoptic and meteorological drivers of extreme ozone concentrations over Europe." *Environ* Res. Lett., 11, 024005, doi:10.1088/1748-9326/11/2/024005, 2016

Conferences contribution

Presentations

- Otero, N., Sillmann, J. and Butler, T. “A model comparison of meteorological drivers of surface ozone over Europe.” *18th Task Force on Measurement and Modelling Meeting*, Prague, 2017.

Posters

- Otero, N., Sillmann, J. and Butler, T. “Assessment of an extended version of the Jenkinson-Collison classification on CMIP5 models over Europe.” *AGU Fall meeting*, 2016
- Otero, N., Sillmann, J., Schnell, J. L., Rust, H. W., and Butler, T. “Synoptic and meteorological drivers of extreme ozone concentrations over Europe.” *The EGU General Assembly*, 2015
- Otero, N., Sillmann, J. and Butler T. “Effect of “low-wind” circulation types on air pollution conditions in present and future climate.” *REKLIM Conference 2014 “Our future – our climate” Berlin*, 2014
- Otero, N., Sillmann, J. and Butler T. “Effect of “low-wind” circulation types on air pollution conditions in present and future climate PhD-Conference on Earth System Science”, *Jena*, 2014

Declaration

I hereby declare that except where specific reference is made to the work of others, the contents of this dissertation are original and it is the result of my own work. This dissertation has not been submitted in whole or in part for consideration for any other degree or qualification in this, or any other University.

Noelia Otero Felipe
February 2018