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#### **LETTER**

# Synoptic and meteorological drivers of extreme ozone concentrations over Europe

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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  $100 \,\mu\mathrm{g}\,\mathrm{m}^{-3}$  (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  $\mu$ g m<sup>-3</sup> (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  $1^{\circ}$  by  $1^{\circ}$  grid cells, over the period 1998–2012. This interpolation of surface ozone concentrations provides a  $1^{\circ} \times 1^{\circ}$  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  $(1^{\circ} \times 1^{\circ})$  (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		
Су	$\sin(2\pi d/365), \cos(2\pi d/365)$	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:  $100 \, \mu \mathrm{g \, m}^{-3}$  (~50 ppb) and  $120 \, \mu \mathrm{g \, m}^{-3}$  (~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 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.

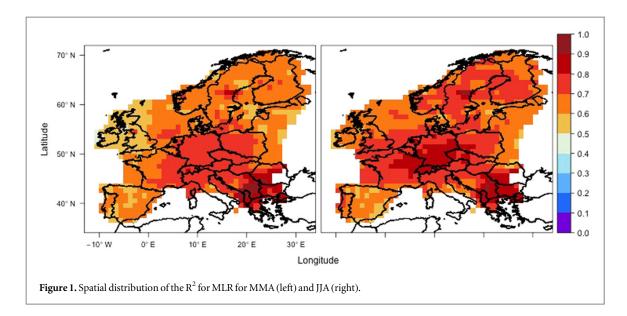
The performance of the models is higher in summer than in spring and this feature is especially observed in central and north-west Europe (figure 1). Overall, the inclusion of LO3 improves the model, which is reflected by the relative contribution to total explained variance and its relative importance 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



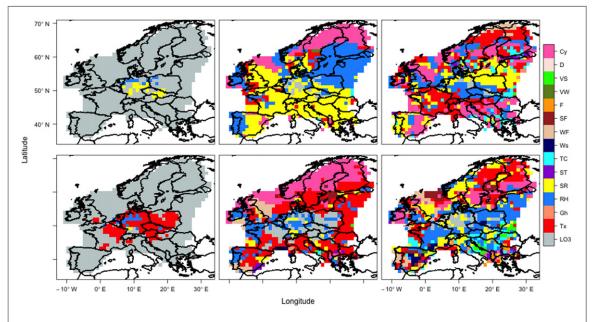


Figure 2. Spatial distribution of the most important driver (left), second most important (middle) and third most important (right) of ozone in MLR for both seasons: MAM (top) and JJA (bottom).

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 gridcells 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 $_{\rm ex50}$ ), 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.

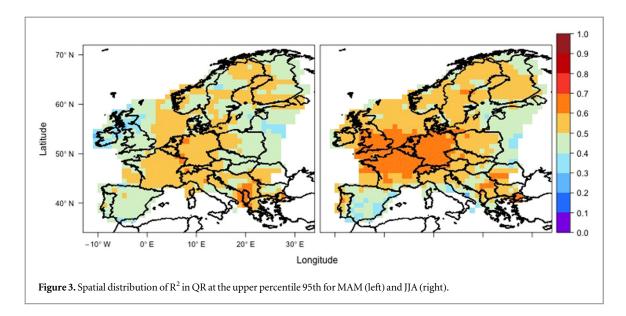
The drivers of surface ozone are identified during the model development using screening procedures that sequentially remove less significant drivers. The performance of the models is generally better in summer than spring. Geographically, the best performance is found in central and northwestern Europe (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

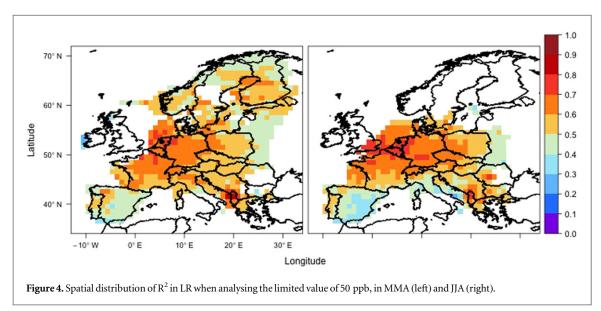
**Table 3.** Frequency (%) of selection of predictors in the QR models developed in MAM and in JJA for the 95th over all grid points.

	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 <sub>ex50</sub>	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





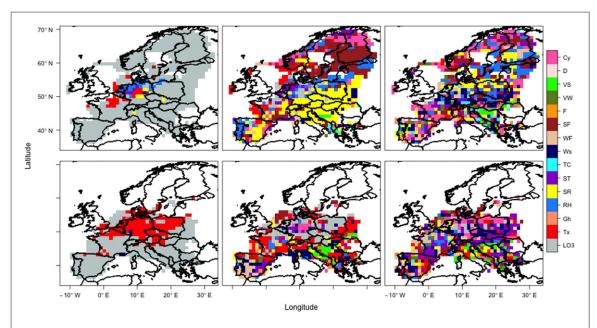
Peninsula. However, we find that meteorological drivers account for most of the explained variance of ozone in most of the grid cells over central and northwest Europe.

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 elects 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 Southcentral 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 Southcentral 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 synopticscale atmospheric circulation and ozone concentrations in metropolitan Pittsburgh, Pennsylvania *Atmos. Environ.* 26

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  ${\rm O_3}$  and  ${\rm PM_{10}}$  levels at a rural midlatitude 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  ${\rm O}_3$  and  ${\rm PM}_{10}$  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 spatiotemporal 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 11719–43
- 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