Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-257, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 24 March 2017

Atmospheric Chemistry and Physics
Discussions

Advanced error diagnostics of the CMAQ and Chimere modelling systems within the AQMEII3 model evaluation framework

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Abstract. The work here complements the overview analysis of the modelling systems participating in the third phase of the Air Quality Model Evaluation International Initiative (AQMEII3) by focusing on the performance for hourly surface ozone by two modelling systems, Chimere for Europe and CMAQ for North America.

The evaluation strategy outlined in the course of the three phases of the AQMEII activity, aimed to build up a diagnostic methodology for model evaluation, is pursued here and novel diagnostic methods are proposed. In addition to evaluating the 'base case' simulation in which all model components are configured in their standard mode, the analysis also makes use of sensitivity simulations in which the models have been applied by altering and/or zeroing lateral boundary conditions, emissions of anthropogenic precursors, and ozone dry deposition.

To help understand of the causes of model deficiencies, the error components (bias, variance, and covariance) of the base case and of the sensitivity runs are analysed in conjunction with time-scale considerations and error modelling using the available error fields of temperature, wind speed, and NO_x concentration.

The results reveal the effectiveness and diagnostic power of the methods devised (which remains the main scope of this study), allowing the detection of the time scale and the fields that the two models are most sensitive to. The representation of planetary boundary layers (PBL) dynamics is pivotal to both models. In particular: *i)* The fluctuations slower than ~1.5 days account for 70-85% of the total ozone quadratic error; *ii)* A recursive, systematic error with daily periodicity is detected, responsible for 10-20% of the quadratic total error; *iii)* Errors in representing the timing of the daily transition between stability regimes in the PBL are responsible for a covariance error as large as 9 ppb (as much as the standard deviation of the network-average ozone observations in summer in both Europe and North America); *iv)* The CMAQ ozone error has a weak/negligible dependence on the errors in NO_2 and wind speed, while the error in NO_2 significantly impacts the ozone error produced by Chimere; *v)* On a continent wide monitoring network-average, a zeroing out of anthropogenic emissions produces an error increase of 45% (25%) during summer and of 56% (null) during winter for Chimere (CMAQ), while a zeroing out of lateral boundary conditions results in an ozone error increase of 30% during summer and of 180% during winter (CMAQ).

1. Introduction

The vast majority of the research and applications related to the evaluation of geophysical models make use of aggregate statistical metrics to quantify, in some averaged sense, the properties of the residuals obtained from juxtaposing observations and modelled output (typically time series of the variable of interest). This practice is rooted in linear regression analysis and the assumption of normally distributed residuals and has been proven to be reliable when dealing with simple, deterministic and low-order models. Led by the rapid pace of improved understanding of the underlying physics, the paradigm is however changed nowadays in that models have grown in complexity and nonlinear interactions and require more powerful and direct diagnostic methods (Wagener and Gupta, 2005; Gupta, et al., 2008; Dennis et al., 2010; Solazzo and Galmarini, 2016).

Evaluation of geophysical models is typically carried out under the theoretical umbrella proposed by Murphy in the early 1990s for assessing the dimensions of goodness of a forecast: consistency (*'the correspondence between forecasters' judgments and their forecasts*'), quality ('*the correspondence between the forecasts and the matching observations'*), and value ('*the incremental benefits realised by decision makers through the use of the forecasts'*) (Murphy, 1993). Since 2010, the Air Quality Model Evaluation International Initiative (AQMEII, Rao et al., 2011) has focused on the quality dimension – the one most relevant to science, according to Weijs et al. (2010) – of air quality model hindcast products, aiming at building an evalution strategy that is informative for modellers as well as to users.

Our claim is that the *value* of a model's result depends strictly on the *quality* of the model that, in turn, depends on sound evaluation. The scientific problem of assessing the *quality* of a modelling system for air quality is tackled by Dennis et al. (2010) who distinguish four complementary approaches to support model evaluation: operational, probabilistic, dynamic and diagnostic, which are also the four founding pillars of AQMEII. Several studies performed under AQMEII have focused on the operational and probabilistic evaluation (Solazzo et al., 2012a,b; Solazzo et al., 2013; Im et al., 2015a,b; Appel et al., 2012; Vautard et al., 2012) and more recently efforts have been expanded to the diagnostic aspect (Hogrefe et al., 2014; Solazzo and Galmarini, 2016; Kioutsioukis et al., 2016; Solazzo et al., 2017).

Operational metrics usually employed in air quality evalution (cfr. Simon et al., 2012 for a review) have several limitations as summarised by Tian et al. (2016): *interdependence* (they are related to each other and are redundant in the type of information they provide), *underdetermination* (they do not describe unique error features), and *incompleteness* (how many of these metrics are required to fully characterise the error?). Furthermore, they do not help to determine the *quality* problem set above in terms of diagnostic power. Gauging (average) model performance through model-to-observation distance leaves open several questions such as *a)* How much information is contained in the error? In other words, what remains wrong with our underlying hypothesis and modelling practice? *b)* Is the model providing the correct response for the correct reason? *c)* What is the degree of complexity of the system models can actually match? These questions have a straightforward, very practical impact on the use of models, the return they provide (the value) and their credibility. Answers to these questions are also relevant to the wide-spread practice of bias correction which is aimed at adjusting the model value to the observed value, rather than correcting the causes of the bias which might stem from systematic, cumulative errors.

The main aims of this study are to move towards tools devised to enable diagnostic interpretation, following the approach of Gupta et al. (2008 and 2009), Solazzo and Galmarini (2016), and Kioutsioukis et al. (2016) and to advance the evaluation strategy outlined in the course of the three phases of AQMEII. In particular, the work presented here is meant to complement the overview analysis of the modelling systems participating in AQMEII3 (summarised by Solazzo et al., 2017) by concentrating on the performance for surface ozone modelled by two modelling systems: Chimere for Europe (EU) and CMAQ for North America (NA). This study attempts to:

- Identify the time scales (or frequencies) of the error of modelled ozone;
- Attribute each type of error to processes by utilizing modelling runs with modified fluxes at the boundaries (anthropogenic emissions and deposition at the surface, and boundary conditions at the bounding planes of the domain) and breaking down the mean square error (MSE) into bias, variance and covariance. This analysis allows us to diagnose the quality of error and to determine if it is caused by external conditions or due to missing or biased parameterisations or process representations;
- Investigate the periodicity of the ozone error which can be symptomatic of recursive (either casual or systematic) model deficiencies;

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- Determine the role of the error of precursor or meteorological fields in explaining the ozone error. The significance (or the non-significance) of a correlation between the ozone error and that of one of the explanatory variables can help to understand the impact (or lack of impact) of the latter on the ozone error as well as the time-scale of the process(es) causing the error.

Among the several models participating in AQMEII3, CMAQ and Chimere have been selected as the analysis proposed in this study requires additional simulations beyond those performed by all AQMEII3 groups, which implied additional dedicated resources that were not available to all groups. This of course opens an important issue connected with the relevance of models in decision making, the adequacy of their contribution, and consequently the fact that far more resources would be required by the present complexity and state of development of modelling systems to guarantee that deeper evaluation strategies are put in place. Although only these two modelling systems are analyzed here, they represent two well-established systems that have been systematically developed over many years, are in use by a large number of research groups around the world and also have participated in the various phases of AQMEII.

The data used, model features and error decomposition methodology are summarised in section 2. Results of the aggregate time series and error decomposition analyses are presented in section 3 and results of the diagnostic error investigation through wavelet, autocorrelation, and multiple regression analysis are presented in section 4. Conclusions and final remarks are drawn in Section 5.

2. Methods

2.1 Data and models

Unless otherwise specified, analyses are carried out and results are presented for the rural receptors of three sub-regions over each continental area as shown in **Figure 1**. The three sub-regions have been selected based on similarity analysis of the observed ozone fluctuations slower than ~1.5 days. The regions where the slow fluctuations showed similar characteristics were selected through unsupervised hierarchical clustering (details in Solazzo and Galmarini, 2015). Due to the similarity of the observations within these regions which implies that they experience common physical and chemical characteristics, spatial averaging within these sub-regions was carried out.

The stations used for the analysis are part of the European (European Monitoring and Evaluation Programme: EMEP; http://www.emep.int/; European Air Quality Database AirBase; http://acm.eionet.europa.eu/databases/airbase/) and North American (USEPA Air Quality System AQS: http://www.epa.gov/ttn/airs/airsaqs/; Analysis Facility operated by Environment Canada: http://www.ec.gc.ca/natchem/) monitoring networks. Full details are given in Solazzo et al. (2017) and references therein.

Following the approach used in previous AQMEII investigations, modelled hourly concentrations in the lowest model layer (~20m for both models) and corresponding observational data are paired in time and space to provide a verification data sample $\{mod_r^t, obs_r^t;\ t=1,\dots,8760;\ r=1,\dots,n_{recs}\}$ of n_{recs} (number of monitoring stations) record of matched modelled and observational data, where the r^{th}-pair mod^{t0} and obs^{t0} is evaluated at receptor r at a given time t_0. Further, while the observations are reported at the hour at the end (for Europe) or at the beginning (for NA) of the hourly averaging window, the model values available in this study are provided instantaneously. Therefore, the modelled data were averaged between two contiguous hours and assigned to the end (or beginning) of that hour for consistency with the observations. This is of particular relevance when estimating the error due to timing of the diurnal cycle discussed in section 4.3, although for EU there is no harmonisation of time references.

For the analyses conducted in this study, the spatial average of the observed and modelled ozone time series has been carried out prior to any time aggregation, i.e. the spatial average is created by averaging the hourly values over all rural stations in each region. The analysis is restricted to stations with a data completeness percentage above 75% and located below 1000m above sea level. Time series with more than 335 consecutive missing records (14 days) have been also discarded. Missing values have not been imputed. The number of rural receptors n_{recs} for ozone is 38, 184, and 40 for EU1, EU2, and EU3 and 73, 43, and 28 for NA1, NA2, and NA3, respectively.

The configuration of the CMAQ and Chimere modelling systems for AQMEII3 is extensively discussed in Solazzo, et al. (2017) with respect to resolution, parameterisations, and inputs of emissions, meteorology, land use, and boundary conditions. For completeness a short summary is provided hereafter.

The CMAQ model (Byun and Shere, 2006) is configured with a horizontal grid spacing of 12 km and 35 vertical layers (up to 50 hPa) and uses the widely applied CB05-TUCL chemical mechanism (Carbon Bond mechanism, Whitten et al., 2010) for the representation of gas phase chemistry. Emissions from natural sources are calculated inline by the Biogenic Emissions Inventory System (BIES) model. The meteorology is calculated by the Weather Research and Forecast (WRF) model (Skamarock et al., 2008) with nudging of temperature, wind and humidity above the planetary boundary layer (PBL).

Chimere (Menut et al., 2013) is configured with a grid of 0.25 degree (~25 km x 18 km over France), 9 vertical layers (up to 500 hPa) and uses the Melchior2 chemical mechanism (Lattuati, 1997) for the representation of gas phase chemistry. Natural emissions are calculated using the MEGAN model (Guenther 2012). The hourly meteorological fields are retrieved from the Integrated Forecast System (IFS) operated by the European Centre for Medium-Range Weather Forecast (ECMWF).

Both models are widely used worldwide in a range of applications such as scenario analysis, forecasting, ensemble modelling, and model inter-comparison studies.

2.2 Sensitivity runs with CMAQ and Chimere

The Chimere and CMAQ models have been used to perform a series of sensitivity simulations aimed at a better understanding of the causes of differences between the base model simulations and observed data. In particular, the following set of sensitivity runs was performed:

- one annual run with zeroed anthropogenic emissions to provide an indication of the amount of regional ozone due to boundary conditions and biogenic emissions (referred to as '*zero Emi*');
- one annual run with a constant value of ozone (zero for NA and 35 ppb for EU) at the lateral boundaries of the model domain to provide an indication of amount of ozone formed due to anthropogenic and biogenic emissions within the domain (in addition to the constant value for EU) (referred to as '*zero BC*' and '*const BC*'). All species other than ozone had boundary condition values of zero for both NA and EU in these sensitivity simulations;
- one annual run where the anthropogenic emissions are reduced by 20%. In addition, the boundary conditions for this run were prepared from a C-IFS simulation (detail in Galmarini et al., 2017 and references therein) in which global anthropogenic emissions were also reduced by 20% (referred to as a '*20% red*');
- one run with ozone dry deposition velocity set to zero, available for the months of January and July (referred to as '*zero Dep*').

2.3 Error diagnostic metric

To aid diagnostic interpretation, the total quadratic error MSE (MSE = $E[mod\text{-}obs]^2$) is decomposed according to

$$MSE = \left(\overline{mod} - \overline{obs}\right)^2 + (\sigma_m - \sigma_o)^2 + 2\sigma_m\sigma_o(1-r) = bias^2 + var + covar \qquad \textbf{Eq 1}$$

Where σ_m and σ_o are the modelled and observed standard deviation, *var* and *covar* are the variance and covariance operators, *r* is the linear correlation coefficient, and *bias* is the time averaged offset between the mean modelled and observed ozone concentration. The MSE is a quadratic, parametric metric widely applied in many contexts and occurs because the model does not account for information that could produce a more accurate estimate. Put in an information theory context, the MSE provides a measure of the information about the observation that is missing from a Gaussian model centred at a deterministic prediction (Nearing et al., 2015). Ideally, the deviation of a perfect model from the observation should be zero or simply white noise (uncorrelated, zero mean, constant variance). Various flavours of MSE decomposition have been exploited in several geophysical contexts (Enthekabi, et al., 2010; Murphy, 1988; Wilks, 2011; Wilmott, 1981; Gupta, et al., 2009), all stemming from the consideration that the bias, the variance, and the covariance characterise different (although not complementary and not exhaustive) properties of the error – accuracy, precision, and correspondence, respectively.

The first two moments (mean and variance) relate to the systematic error (unconditional bias) and variability (variance), respectively. All other differences between the statistical properties of modelled and observed chemical species (e.g. the timing of the peaks and autocorrelation features) are quantified by the correlation coefficient, i.e. in the covariance term (Gupta et al., 2009).

The relative contribution of each of the MSE components to the overall MSE is summarised by the Theil's coefficients (Theil, 1961):

$$\begin{aligned} F_b &= \mathrm{bias}^2/\mathrm{MSE} \\ F_v &= \mathrm{var}/\mathrm{MSE} \\ F_c &= \mathrm{covar}/\mathrm{MSE} \end{aligned} \qquad \textbf{Eq 2}$$

The overall MSE suffers from the limitations of the aggregate metrics discussed in the introductory section, lacking independence and explanatory power (Tian et al., 2016). When decomposed (e.g according to **Eq 1**), however, the underdetermination issue is reduced and the MSE coefficients (**Eq 2**) do offer diagnostic aid in interpreting the modelling error (Gupta, et al., 2009).

3. Sensitivity analysis to emissions and boundary conditions perturbations

3.1. Aggregated time series of ozone

Figure 2 and **Figure 3** show monthly and diurnal curves for the base and sensitivity simulations over the three sub-regions in each continent. Results show that the monthly averaged curves of the zeroed emission runs peak in April in NA and in July in EU (May to July in EU1 are approximately the same), indicating the periods when the impact of background concentration (boundary conditions) and biogenic emissions on regional ozone is largest: springtime in NA and summer in EU. The monthly curves of 'zero BC' and 'zero Emi' for NA are anti-correlated between the months of April to July-August ('zero Emi' curve decreasing and 'zero BC' curve raising) and during autumn ('zero Emi' curve rising and 'zero BC' curve decreasing), framing the interplay among these two factors in terms of total ozone loading: boundary conditions dominating in autumn-winter and biogenic plus anthropogenic emissions are more important during spring-summer.

The daily averaged profiles of mean ozone for NA show that the observed peak (occurring between 16-18 LT in NA1 and NA2 and ~1 hour earlier in NA3) is preceded by the peak in the base run by ~1hour in NA2 and by ~2-3 hours in NA1, while the timing of the observed minimum (occurring at 8-9 am LT) is captured by the base run in NA2 and NA3 while it is preceded by the base run by ~1hour in NA1. The modelled morning transition to convective conditions is in phase with the observations except for NA1 where the modelled transition occurs one hour earlier than the observed one. The modelled afternoon transition in NA1 precedes the observed transition by 3-4 hours, possibly due to errors in the partitioning between sensible and latent surface heat flux that causes a faster-than-observed collapse of the PBL. As discussed in Appel et al. (2016), updates to the

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Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 24 March 2017

Atmospheric Chemistry and Physics
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Discussions

stomatal conductance function and the heat capacity for vegetation in WRF and the ACM2 vertical mixing scheme in both WRF and CMAQ (relative to the version of WRF and CMAQ used in the current study) lead to a change in the modelled diurnal cycle of ozone as well as other pollutants and meteorological variables. In particular, the updates lead to a delay in the evening collapse of the modelled PBL (Appel et al., 2016). The shape of the 'zero BC' curve is similar in amplitude to that of the base run, suggesting that the effect of the regional/background ozone represented through boundary conditions in a limited area model is mainly to shift the mean concentration upwards while it has no major effect on the frequency modulation. By contrast, the absence of anthropogenic emissions has a major effect of the amplitude of the signal as well as its magnitude ('zero Emi' curve). As discussed in the next section, these considerations translate into the bias and/or variance type of error due to the boundary conditions and emissions.

As for EU (**Figure 3**), the observed daily profiles in EU1 and EU2 are closely matched by the Chimere model between 11 LT and 23 LT (underestimated outside these hours), while in EU3 the daily peak (observed at 19-20 LT) is consistently occurring earlier in the model and its magnitude is overestimated. The morning transition occurs earlier in the model than the observations and follows a significant model under-prediction of nighttime and early morning ozone, due to difficulties in reproducing stable or near-stable conditions (Bessegnet et al., 2016). In EU3, the model displays the poorest performance, with significant underestimation between midnight and 9 LT (5-7 ppb) and over-estimation in daylight conditions (7-9 ppb).

As opposed to the CMAQ case for NA, the shape of the 'zero Emi' curve of Chimere closely follows the shape that of the base case (even when considering only the stations classified as 'urban', Figure S2), suggesting a bias type of error.

Due to the long time average (one year), the daily profiles displayed in **Figure 2** and **Figure 3** do not provide information about the exact timing of the minima and maxima for each season throughout the year. Figure S3 and Figure S4 report the seasonal average diurnal profiles for the model predictions and the observations (network average over all stations) and show that the timing of the ozone diurnal cycle varies seasonally.

3.2. Error decomposition

The plots in **Figure 4** (NA) and **Figure 5** (EU) show the MSE decomposition according to Eq. 1 for the summer months of June, July, and August for the base case simulation as well as the sensitivity simulations, distinguishing between daylight (from to 5am to 9pm LT) and night-time hours (the remaining hours, from 10pm to 4am LT). These plots are meant to aid the understanding of the relative impacts of potential errors in lateral boundary conditions, anthropogenic emissions, and the representation of ozone dry deposition on the total model error by comparing the magnitude and type of model error from these simulations against the model error for the base case.

The plots In **Figure 6** And **Figure 7** are complementary to Figures 4-5 and show the error decomposition for both the summer and winter season in more detail, including the error coefficients *Fb, Fv, Fc* of **Eq 2** (left vertical axis), the total MSE (right vertical axis), the sign of the bias and variance error (+/- for model over and under prediction), and the values of the correlation coefficient. Furthermore, the maps in **Figure 8** and **Figure 9** show the RMSE at the receptors for the 'base' case as well as $\Delta RMSE$, i.e. the percentage change of RMSE of the sensitivity runs with respect to the 'base' case simulation:

$\Delta RMSE = 100*(RMSE_s - RMSE_{base})/RMSE_{base}$, where the subscript *s* indicates the zeroed emission or the zeroed (constant) boundary condition simulations ($\Delta RMSE$ is measured as percentage).

The CMAQ results for NA are presented in **Figure 4**, **Figure 6**, and **Figure 8** and can be summarised as follows:

- The MSE of the base case (MSE_{base}) during summer daylight is mainly due to bias (~35% in NA1 and ~75% in NA2 and NA3) and the remaining portion is due to covariance error. The fact that there is no variance error shows that the model is able to replicate the observed 3-month averaged variability.
- The effect of zeroing the emissions of anthropogenic pollutants on the summer MSE is a rise by a factor ~2 to 4 (daylight) and by a factor ~6 to 7 during night-time in NA1 and NA2 with respect to MSE_{base}, while during night-time in NA3 the MSE stays approximately the same, indicating that the emissions have little role in determining the total error in this sub-region during night during summer. Furthermore:
  - All the error components deteriorate in the simulations with zero anthropogenic emissions except for the bias in NA3. This is particularly true for the variance, signifying the fundamental role of emissions in shaping the diurnal variation of ozone. Indeed, this suggests that the absence of a variance error in the base case (see above) is due to the correct intensity of the prescribed emissions;
  - The covariance share of the error also increases (although only slightly in NA2) for the zero emissions case, indicating that the emissions play a role in determining the timing of the modelled diurnal ozone signal, this increase is more pronounced during night-time.
- The zeroing of the input of ozone from the lateral boundaries has either no effect or only a very limited effect on the variance and covariance shares of the error, while it has a profound impact on the bias portion. This impact is approximately equal during daylight and night-time, as expected from the discussion of the daily cycle shown in **Figure 2**.
- The removal of ozone dry deposition from the model simulations (results based on July only) has the most profound impact, increasing by one order of magnitude the MSE of the base case which is approximately double the combined effect of the emissions and boundary conditions perturbation. This sensitivity gives a gross indication of the relative strength of this process vs external conditions during summer, while the 'zero BC' case has a larger effect than the 'zero deposition' case in January (not shown). Similar to the 'zero BC' case, the exclusion of ozone dry deposition from the model simulations acts as an additive term to the diurnal curve in NA1, leaving almost unaltered the shape and timing of the signal, while it impacts the variance and covariance error in the other two sub-regions.
- The instances where the '20% red' bias error is lower than the error of the base case occur when the mean ozone concentrations were overestimated in the base case (e.g. daylight for all sub-regions and NA2 and NA3 over night-time summer) as illustrated in **Figure 6**a,b.
- The maps show that there are stations where the error is reduced with zero anthropogenic emissions (e.g. a reduction of 20-30% in the south coast of the US and in the far North-east during summer, **Figure 8**d). This suggests the presence of other compensating model errors in both the base and sensitivity simulations that lead to better agreement with observations when prescribing an unrealistic emission scenario. The sources of these compensating errors need to be investigated in future work.
- The 'zero BC' run has profound negative effects over the whole continental area of NA during winter (**Figure 8**e), while the effects are smaller during summer (**Figure 8**f) especially over the southern coast due to the relatively higher importance of photochemical formation of ozone during summer.
- The error characteristics of the daily maximum 8-hour rolling mean (DM8h, **Figure 6**e) resemble those of the daylight base case (but reduced in magnitude during winter), with almost null variance error and the same sign of the bias as the base case. The error of the DM8h for the sensitivity runs is reported in Figure S5.
- On a network-wide average, removing anthropogenic emissions causes a RMSE increase of 25% during summer and of 0% (10% at 75^{th} percentile) during winter while a zeroing out of input from the lateral boundaries causes a RMSE increase of 30% during summer and of 180% during winter (median values, **Figure 8**).

The allocation of the error of the Chimere model for EU varies greatly by sub-region (**Figure 5**, **Figure 7**, and **Figure 9**):

- The summer daylight $RMSE_{base}$ ranges between ~20 ppb^2 (EU1, ~60% covariance and ~20% bias) and ~85 ppb^2 (EU3, 95% covariance). In EU3, the night-time bias of ~75% outweighs the covariance as seen in **Figure 7**a.
- Removing the anthropogenic emissions had almost no effect on the covariance share of the MSE (if not a slight reduction with respect to the base case in EU2 and EU3, and also during night-time), indicating that the error in the timing of the signal is not influenced by the emissions but rather by other processes. Moreover, the variance portion is left almost unchanged (1 ppb increase in EU1 and EU2), in contrast with the CMAQ results for NA. This would indicate that the variability of ozone concentration is hardly influenced by anthropogenic emissions in Chimere. The bias is the error component most sensitive to emissions reductions, especially in EU2 and less so in EU3. This is in line with the discussion of the daily profiles of **Figure 2**b (which showed similar shapes of for the 'zero Emi' and of the 'Base' profiles) and contrasts with the NA case where the 'zero Emi' daily profiles are flatter than the base case.
- The effect of imposing a constant ozone boundary condition value of 35 ppb (and of zero for all other species) on the model error is similar to that of removing the anthropogenic emissions as far as the total MSE and the bias of EU2 are concerned. It outweighs the latter for the total MSE, bias and variance in EU3 and covariance and night-time bias component in EU1. We can infer that the boundary conditions have a significant role in determining the timing of the ozone signal in EU1 (close to the western boundary of the domain) as the correlation coefficient degrades form 0.89 (base case) to 0.66 ('const BC') (**Figure 5** and **Figure 7**a and c). The bias staying the same in EU1 daylight summer depends on the magnitude of the constant value (35 ppb were chosen here) that is in close agreement with that of the base case while the small variance error (~2ppb) vanishing with respect to the base case might be explainable with numerical compensation.
- During summer in EU2 and EU3 changing the ozone boundary condition only influences the bias with marginal impacts on variance and covariance, while in winter (**Figure 7**c) there is also a significant reduction of the correlation coefficient, meaning that the boundary conditions modulate the timing of the signal.
- EU3 deserves special consideration as the $RMSE_{zeroEmi}$ is approximately the same as the $RMSE_{base}$, which mostly consists of covariance error during daylight and bias error during night-time. Due to the local topography, EU3 is typically characterised by stagnant conditions that are difficult to model. For example, 50% of the observed wind speed is below 1.65 ms^{-1}, while Chimere predicts 1.95 ms^{-1}. The largest impact on the total MSE is seen in the 'const BC' run and arises in the bias portion, pointing to the importance of properly characterising background (regional) concentrations.
- With respect to the base case, the DM8h (**Figure 7**e) shows a drastically reduced covariance error (the timing error is now shifted towards seasonal time scales) at the expense of an increase in variance error. The variability of the DM8h is governed by synoptic processes which are likely responsible for the variability error of the DM8h. The error of the DM8h for the sensitivity runs is reported in Figure S6.
- On a network-wide average, removing anthropogenic emission causes an RMSE increase of 45% during summer and of 56% during winter (median values, **Figure 9**c,d).
- The effect of setting the dry deposition velocity of ozone to zero (July only, **Figure 5**), increases not only the bias error but also causes large increases of the variance and covariance shares of the error. Thus in Chimere the deposition acts not only as a shifting term on the modelled concentration but it also influences the variability and timing of ozone more profoundly than for the CMAQ case examined earlier.

4. Time-scale error analysis and diagnostic

The focus of this section is ΔO_3, the time series of the deviation between the base case and observations. The nature of ΔO_3 is examined for time-frequency patterns using wavelet analysis and for error persistence using autocorrelation functions (ACF). The causes of ΔO_3 are also tentatively investigated as dependencies on other fields using multiple regression analysis combined with bootstrapping to sample the relative importance of the regression variables.

4.1. Spectral considerations

The coefficients of the ACF (Appendix 1) can be interpreted as the Fourier transform of the power spectral density. Frequency analysis of a signal is often performed by constructing the periodogram (or spectrogram, see e.g. Chatfield, 2004). This approach has proven useful when dealing with harmonic processes superimposed on a baseline signal (Mudelsee, 2014) but, at the same time, periodograms often contain high noise. Therefore, examining a signal at specific frequencies can be instructive, for instance by resorting to wavelet transform which has the further advantage of enabling a 3-dimensional time-frequency-power visualisation. Compared to a power spectrum showing the strength of variations of the signal as function of frequencies, wavelet transformation also allows the allocation of information in the physical time dimension other than phase space. Here, wavelet analysis of the periodogram of seasonal ΔO_3 is performed using the Morlet wavelet transform (Torrence and Compo, 1997).

From inspecting **Figure 10** (NA) it emerges that the highest values of spectral energies for ΔO_3 for the three sub-regions (corresponding to the 99th percentile of the spectrum) are observed for periods spanning the whole year, associated with the slow variability of the non-zero bias throughout the investigated period. Such a process is more evident in NA1 and NA2 and its magnitude is one order of magnitude (or more) of the 90th percentile value.

NA3 and to a lesser extent NA2 show a high spectral power of the error for periodicities of 1-2 months and lasting from January to May with a weaker wake extending up to the end of the year, potentially pointing to errors in the characterisation of larger-scale background concentrations associated with boundary conditions. NA3 also exhibits a high spectral power for errors associated with a periodicity of ~20 days during January-February and June-July and ~ 15 days during October and December. This may point to errors in representing the effects of changing weather regimes on simulated ozone concentrations.

Except for the long-term variations of the model error with periodicities greater than 2 months discussed above, NA1 is the only sub-region that shows only weak power associated with model errors of shorter periodicities from June to December. This suggests that fluctuations caused by variations in large scale background and changing weather patterns are better captured in this region compared to the other two sub-regions.

The energy associated with the daily error is again higher and more pronounced in NA3 than in the other sub-regions where it is most pronounced during summer (NA1) or between March to October (NA2). While during winter and autumn the daily error is likely driven by difficulties in reproducing stable PBL dynamics, during spring and summer it is also influenced by the chemical production and destruction of ozone, a process entailing NO_x chemistry, radiation, biogenic emission estimates and chemical transformation, and thus difficult to disentangle from boundary layer dynamics.

For the EU (**Figure 11**) a notable feature is the very high daily error energy in EU3 that is present throughout the year and most pronounced in summer. Such high energy suggests persistent problems in representing processes having a periodicity of one day. Further, EU3 shows an area of high energy associated with a period of one to two months and extending from February, peaking in April and May, and ending in September (mostly model underestimation, **Figure 11**c), while the error of the winter months in EU3 receives high energy from slower processes, acting on time scales of ~6 months and beyond. Considering that the EU3 region is surrounded by high mountains, tropopause folding (e.g. Bonasoni et al., 2000; Makar et al., 2010) together with the lack of modelling mechanisms for the tropopause/stratosphere exchange, could offer an explanation of the high energy of the error at long time scales (also considering that the higher level modelled by Chimere is well below the tropopause and that vertical fluxes are those prescribed by the C-IFS model). Errors in estimates of biogenic emissions also remains a plausible cause of ozone error during spring and summer months.

The similarity of the wavelet spectra for NA3 (**Figure 10**c) and EU1 (**Figure 11**a) (both regions are located on the Western edge of their domain) at the beginning of the year for periods of 1 to 2 months might be indicative of the periodicity of the bias induced by the boundary conditions. Compared to CMAQ, the error of the Chimere model is more concentrated during spring and early summer, with a periodicity of 10-20 days.

Having identified some relevant time-scales for the ΔO_3 error, in the next sections methods are proposed for its detection and quantification.

4.2. Temporal characteristics of the error of ozone

In a recent study, Otero et al. (2016) analyzed which synoptic and local variables best characterise the influence of large scale circulation on daily maximum ozone over Europe. The authors found that the 24-hour lag autocorrelation explains the majority of the variance during spring over the entire EU continent while during summer the maximum temperature is the principal explanatory variable over continental EU. Other influential variables were found to be the relative humidity, the solar radiation and the geopotential height. Camalier et al. (2007) and Lemaire et al. (2016) found that the near-surface temperature and the incoming short-wave radiation were the two most influential drivers of ozone uncertainties.

The ACF and PACF (partial autocorrelation function) of ΔO_3 (see Appendix 1 for a definition of both functions) reveals a strong periodicity for periods that are multiples of 24 hours (**Figure 15**a And **Figure 16**a) (note that the first derivative of ΔO_3 is used in this analysis to achieve stationarity). The structure of the error is such that it repeats itself with daily regularity, indicating either a systematic error in the model physics or a missing process at the daily scale, possibly related to radiation and/or PBL-related variables. While the presence of a daily periodic forcing due to the deterministic nature of day/night differences superimposed on the baseline ozone is expected, the periodicity maintained in the error structure is not and deserves further analysis.

The PACF plots confirm that the error is not simply due to propagation and memory from previous hours, but arises at 24h intervals and hence stems from daily processes. On average, for NA $corr(\Delta O_3(h), \Delta O_3(h+1))$ (i.e. the correlation between $\Delta O_3(h)$ and $\Delta O_3(h+1)$) is ~0.45, while the $corr(\Delta O_3(h), \Delta O_3(h+24))$~0.68, for any given hour h. Similarly for EU, $corr(\Delta O_3(h)$ and $\Delta O_3(h+1))$ ranges between 0.31 (EU2) and 0.54 (EU3), while $corr(\Delta O_3(h),\Delta O_3(h+24))$ ~0.70 for all sub-regions. Thus, the ozone error with a 24h periodicity has a longer memory than the error with a one hour periodicity. Since the 24h periodicity of the error is present in the entire annual time series, the periodic error is not associated with particular conditions (e.g. stability), but is rather embedded into the model at a more fundamental level. Moreover, similar periodicity is observed for the ACF of ΔWS and ΔTemp for both models (not shown), reinforcing the notion that a daily process affecting several model modules is not properly parameterised. As discussed in section 3.1, the representation of latent and sensible heat fluxes in the version of CMAQ used in this study, (i.e. the errors in the timing of the PBL collapse that has been addressed in a newer release of CMAQ) is likely (at least partially) responsible for the daily periodic error noted here. Also for Chimere the reason for the error periodicity likely lies in the PBL dynamics.

By removing the diurnal fluctuations (i.e. by screening out the frequencies between 12 hours and up to ~1.5 days by means of the Kolmogorov-Zurbenko (*kz*) filter, as described in Hogrefe et al., 2000) from the modelled and observed time series, the daily structure of the ACF disappears (**Figure 15**b and **Figure 16**b), replaced by a slow decay and negative (EU1, EU2 and partially NA1, NA2) or fluctuating (NA3, EU3) correlation values. The PACF plots in **Figure 15**b and **Figure 16**b suggest that some significant correlation persists up to ~40 hours, likely due to leakage from the removed diurnal component (as extensively discussed in several earlier works, the *kz* filter does not allow for a clear separation among components and thus some leakage is expected, see e.g. Solazzo et al. 2017).

The relative strength of the MSE for the undecomposed ozone time series and for the ozone time series with the diurnal fluctuations removed and with only the diurnal fluctuations is reported in Table 1. With the exception of NA1 and EU3, the base line error (denoted with 'noDU') accounts for ~70 to 85% of the total error, while the diurnal fluctuations (denoted with 'DU') are responsible for 10 to 23% of the total error (and even less during nighttime). The 'DU' error outweighs the 'noDU' error (67% to 26%) only in EU3, where the daily PBL issue has been pointed out in the previous section.

4.3 Covariance Error: phase shift of the diurnal cycle

This section explores the nature of the covariance error which occurs, among other reasons, when the two signals being compared are not in phase. The first and second moments of the error distribution are invariant with respect to a phase shift between the two signals (Murphy, 1995), i.e. the mean of the signal as well as the amplitude of the oscillations with respect to the mean value are not affected by a phase shift which therefore does not have an impact on the bias and variance components of the error. The correlation coefficient, on the other hand, is impacted by a lagged signal, producing a net increase of the covariance error.

The analysis of the phase lag between the daily component of the modelled and observed cycles is reported in **Figure 12** (NA) and **Figure 13** (EU), winter and summer are analysed separately.

To perform this analysis, the modelled and observed ozone time series are first filtered to isolate the diurnal component using a kz filter. Then, the cross-covariance between the two time series is calculated. The time at which the maximum covariance value occurs is taken as the phase shift between the two signals. The method has an error of ±0.5 hours.

In NA, the modelled diurnal peak occurs 1-2 hours earlier than the observed diurnal peak at many stations, and up to 3-4 hours earlier at some Canadian stations. By taking into consideration the 0.5 hour error of the estimate, the receptors at the western border (approximately corresponding to NA3) are least affected by this timing error (especially in summer **Figure 12**b), and therefore the covariance share of the error shown in **Figure 4** is not due to daily phase shift in this region but probably due to the shifting of longer (or shorter) time periods induced for example by errors in transport (wind speed and/or direction). Figures S7 in the Supplementary report the same analysis repeated for the 'zero Emi' and 'Zero BC' runs.

In the EU (**Figure 13**), no phase shift (or a phase shift compatible with the 0.5 hour estimation error) is observed in Romania, Germany and the UK during winter, while a significant phase shift (the modelled peak occurs up to 6 hours early) is observed in the North of Italy and Austria, with France and Spain oscillating between positive 3 (model delay up to 5 hours in the south of Madrid) and negative 5 and 6 hour phase shifts, with the net effect of a spatially aggregated daily cycle that is in phase with the observations (**Figure 3**b). During summer the phase shift is larger and extends also to the countries where the phase shift was null during winter. Moreover, some country-wise grouping can be detected, as for example at the border between Belgium and France, Spain and France, Finland to Sweden, possibly due to the lack of harminisation in the timing of the reporting of observational values among EU countruies (e.g. Solazzo and Galmarini, 2015). Figures S8 in the Supplementary report the same analysis repeated for the 'zero Emi' and 'Const BC' runs.

While errors in emission profiles obviously can be one cause of the phase shift and thus the covariance error of the modelled ozone signal, the representation of boundary layer processes clearly can be a factor as well. As discussed in e.g. Herwehe et al. (2011), the parameterisation of vertical mixing during transitional periods of the day can cause a time shift in the modelled ozone concentrations due to its effects on the near-surface concentrations of NO_x and ozone, which in turn affect the chemical regime and balance between ozone formation and removal.

To quantify the importance of the covariance error caused by a phase shift relative to other sources of error, **Figure 14** shows the curves of normalised MSE as the observed ozone time series is shifted with respect to itself between -10 and 10 hours. The MSE curve equals zero for a zero-hour lag and is symmetric with respect to the sign of the lag. Since this analysis compares the observed signal to itself (with varying degrees of time lags), the MSE fraction of bias and variance is zero while all of the MSE is due to the covariance.

The curves in **Figure 14** shows that a phase lag in the diurnal cycle of ±6 hour causes a MSE error in the diurnal component of magnitude *~var(obs)* (in both EU and NA), where *var(obs)* is the variance of the measured diurnal cycle (top panel). The effect on the full (undecomposed) time series is that a phase lag of ±4 (EU) and ±5-6 (NA) hour in the diurnal cycle causes a MSE error of magnitude *~var(obs)*, where in this case the variance is that of the undecomposed time series of ozone (lower panel).

Therefore, a modelled ozone peak that occurs 4 to 5 hours too early (a feature that is detected at some EU3 and Canadian stations) corresponds to a covariance error of 9.0 ppb (i.e. the standard deviation of the network-average ozone observations in summer in both EU and NA). This result also helps explain the large covariance error in EU3, which can be at least partially attributed to the large phase shift of the daily cycle.

4.4 Explaining the error of ozone

In this section a simple linear regression model for the error of ozone ΔO_3 is applied with the goal of detecting the causes of model errors on the daily and longer term scales identified in the previous section. Although a linear model is overly simplistic and other methods are available (e.g kernel smoothers), we employed the simpler approach since *i)* it is not the aim of this study to build a statistically accurate model for the model error , and *ii)* by pursuing simple reasoning we hope to identify the time scale of the error and the most likely fields causing it at that time scale. More advanced techniques are likely to overcomplicate the results and their interpretations but could be pursued in future studies.

The available regressors (explanatory variables) are the errors of the variables for which measurements have been collected within AQMEII, i.e. NO (EU only), NO_2, Temp, and WS:

$$\Delta O_3 = \beta_1 \Delta NO + \beta_2 \Delta NO_2 + \beta_3 \Delta Temp + \beta_4 \Delta WS + k \qquad \textbf{Eq 3}$$

where β_i are the coefficients of the multiple linear regression, and the intercept *k* is the portion of the ozone error not explainable by any of the regressors (the intercept). A bootstrap analysis (Mudelsee, 2014; Groemping, 2006) is used to calculate the relative importance of each error field in explaining the variance of ΔO_3 (**Figure 17** and **Figure 18**) with an uncertainty of ~5%. Since the measurements of ozone and NO_x are not always co-located with the measurements of wind speed and temperature, **Eq 3** is strictly meaningful only in a spatially-averaged sense.

None of the regressors help explain the winter ozone error of CMAQ, while ~15-20% of the ozone error variability during summer is associated with the error in temperature and, to a lesser extent, wind speed. In contrast, in Chimere the NO_2 error over EU during winter is highly correlated with the error of ozone, as is the daytime wind speed error during summer (EU1 and EU2, **Figure 16**a,b). Overall, there is no instance where the variance explained by the available variables (quantified through the coefficient of determination R^2) exceeds 0.60. There is an overwhelming daily memory of the error that can only partially be attributed to errors of the available regressor variables, pointing to the need to include additional variables in future applications of this regression analysis.

A straightforward limitation of **Eq 3** is that it assumes that successive values of the error terms are independent while in practice this is not the case (Table 2 reports the correlation coefficient of the diurnal fluctuations of the residuals, obtained by filtering out fluctuations outside faster than ~1.5 days from the measured and observed time series). Several significant collinearities can be detected (e.g between ΔWS and ΔTemp; ΔNO_2 and ΔTemp, especially in winter).

In addition to the collinearity issue, there are other endogenous variables whose error contributes to total ΔO_3 that are not part of the regression analysis, as revealed by the ACF and PACF of the first-order differentiated residuals of the regression in the last panels of each plot. Such missing variables are likely to correlate with both the dependent (ΔO_3) and the explanatory variables, an issue known as Omitted Variable Bias, e.g. Greene (1993). For instance, errors in the cloud cover and/or radiation scheme, land use masking, etc. are shared by the chemical species (ozone and its precursors) as well as by the meteorological fields. The ACF and PACF suggest that the common, omitted error of the fit propagates with daily recurrence and is not explained by the available variables, stressing the findings of the previous section and again pointing to PBL-related errors.

However, since we are not in a position to estimate the errors associated with PBL variables (radiation, temperature, turbulence) an alternate approach is to filter out the diurnal process from the modelled and observed time series and repeat the analysis based on Eq 3 (Figure S9 and Figure S10).

Table 3 reports the correlation coefficients of the residuals with the diurnal component filtered-out, and indeed the collinearity has been largely removed, especially for NA, while for EU some strong correlation persists (ΔNO_2 and ΔNO, and between ΔWS and ΔTemp in winter):

The R^2 of the regression for the 'no-DU' case drops drastically in summer (EU3 and all sub-regions in NA) as shown in Figures S9 and S10. Moreover, this analysis and its comparison to the results presented in earlier sections lead to the following conclusions:

- A strong daily error component is common to all variables investigated here.
- This error manifests itself in the correlation coefficient, thus is due to a variance/covariance type of error (otherwise, if it was a bias-type error, the R^2 would have been similar between the analysis of the signal with and without the diurnal component);
- At least in NA, the bias error discussed in section 3 cannot be explained simply in terms of the fields NO_2, Temp, and WS. Hence, the bias of the CMAQ model over the NA continent appears to be associated with processes with longer time scales, such as boundary conditions (inducing mostly bias error, as discussed in section 3), deposition, and/or transport (potential systematic errors in wind direction, for example, would likely produce a bias-type error);
- For EU1 and EU2, the error in the meteorological fields (Temp and WS) seems to explain approximately half of the summer ozone error, with a memory of up to 3-4 days (significant, although small PACF values);
- For EU3, the large error identified in section 2 and 3 is indeed dominated by daily processes. The RMSE of the observed vs modelled time series filtered to remove fluctuations faster than ~1.5 days is ~46% of the RMSE of the unfiltered time series (4.2 ppb vs 8.8 ppb, daylight summer, rural stations only). Daily variables (e.g. meteorological variables determining the heat fluxes such as temperature, radiation) and/or precursor emission are likely responsible for the error.
- The impact of ΔNO_2 and ΔNO in EU (all sub-regions, mostly daylight) and of ΔWS in EU1 (and partially EU2) on the error of ozone is similar with and without the diurnal fluctuations, indicating cross-correlation of these error fields for periods longer than one day.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-257, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 24 March 2017

Atmospheric Chemistry and Physics Discussions

5. Conclusions

This study is part of the goal of AQMEII to promote innovative insights into the evaluation of regional air quality models. This study is primarily meant to introduce evaluation methods that are innovative and that move towards diagnosing the causes of model error. It focuses on the diagnostic of the error produced by CMAQ and Chimere applied to calculate hourly surface ozone mixing ratios over North America and Europe.

We argue that the current, widespread practice (although with several exceptions) of using time-aggregate metrics to merely quantify the average distance (in a metric space) between models and observations has clear limitations and does not help target the causes of model error. We therefore propose to move towards the qualification of the error components (bias, variance, covariance) and to assess each of them with relevant diagnostic methods. At the core of the diagnostic methods we have devised over the years within AQMEII is the quality of the information that can be extracted from model and measurements to aid understanding of the causes of model error, thus providing more useful information to model developers and users than can be gained from more aggregate metric. Applying such approaches on a routine basis would help boost the confidence in using models prediction for various applications.

While remarking that the analyses carried out are not meant to compare the two models but are rather meant to show how the two models, applied to different areas and using different emissions, respond to changes, the main conclusions of this study are:

- While the zeroing/modification of input of ozone from the lateral boundaries causes a shift of the ozone diurnal cycle in both CMAQ and Chimere, the response of the two models to a modification of anthropogenic emission and deposition fluxes is very different. For CMAQ, the effect of removing anthropogenic emissions causes a shift and a flattening of the diurnal curve (bias and variance error), while for Chimere the effect is restricted to a shift. In contrast, setting the ozone dry deposition velocity to zero causes a shift (bias error) for CMAQ, while a profound change of the error structure occurs for Chimere with significant impacts not only on the bias but also the variance and covariance terms.

- On a continent wide network-average, removing anthropogenic emissions causes an error increase of 45% (25%) during summer and of 56% (null) during winter for Chimere (CMAQ), while a zeroing of ozone transport across the lateral boundaries causes an error increase of 30% during summer and of 180% during winter (CMAQ).

- Fluctuations slower than ~1.5 days account for 70-85% of the total ozone quadratic error. The partition of this error into bias, variance and covariance depends on season and region. In general, the CMAQ model suffers mostly from bias error (model overestimation during summer and underestimation during winter), while the Chimere model is rather 'centred' (i.e. almost unbiased) but suffers high covariance error (associated with the timing of the signal, thus likely to synoptic drivers)

- A recursive, systematic error with daily periodicity is detected in both models, responsible for 10-20% of the quadratic total error. For CMAQ it is likely to be associated with the timing of daily transitions in the PBL between stable and convective conditions. An indirect confirmation comes from results reported for a more recent version of CMAQ (Appel et al., 2016) which show a delay in the evening collapse of the modelled PBL that is in better agreement with observations;

- The modelled ozone daily peak accurately reproduces the observed one, although with significant exceptions in France, Italy and Austria for Chimere and with the exceptions of Canada and some areas in the eastern US for CMAQ. In these regions the peak is anticipated by up to 6 hours, causing a covariance error as large as 9 ppb;

- The ozone error in CMAQ has a weak/negligible dependence on the error of NO_2 and wind speed, while the error of NO_2 impacts significantly the ozone error produced by Chimere. On time scales longer than 1.5 days, the Chimere ozone error is significantly associated with the error in wind speed in continental Europe and the error in temperature in the Atlantic region (the UK, western France and northern Spain).

Although having exploited several evaluation frameworks over the past ten years within AQMEII (operational, diagnostic, and probabilistic) the goal of clearly associating errors to processes has not yet been achieved. As already suggested in the conclusions of the collective analysis of the AQMEII3 suite of model runs summarised by Solazzo et al. (2017), future model evaluation activities would benefit from incorporating sensitivity simulations and process specific analyses that help to disentangle the non-linearity of the many model variables, possibly by focusing on smaller modelling communities. The 'theory of evaluation' being put forward by the hydrology modelling community (Nearing et al., 2016 and references therein) may provide a template for the air quality community to further advance their model evaluation approaches.

Acknowledgments

We gratefully acknowledge the contribution of various groups to the third air Quality Model Evaluation International Initiative (AQMEII) activity. The following agencies have prepared the data sets used in this study: U.S. EPA (North American emissions processing and gridded meteorology); U.S. EPA, Environment Canada, Mexican Secretariat of the Environment and Natural Resources (Secretaría de Medio Ambiente y Recursos Naturales-SEMARNAT) and National Institute of Ecology (Instituto Nacional de Ecología-INE) (North American national emissions inventories); TNO (European emissions processing); ECMWF/MACC (Chemical boundary conditions). Ambient North American concentration measurements were extracted from Environment Canada's National Atmospheric Chemistry Database (NAtChem) PM database and provided by several U.S. and Canadian agencies (AQS, CAPMoN, CASTNet, IMPROVE, NAPS, SEARCH and STN networks); North American precipitation-chemistry measurements were extracted from NAtChem's precipitation-chemistry data base and were provided by several U.S. and Canadian agencies (CAPMoN, NADP, NBPMN, NSPSN, and REPQ networks); the WMO World Ozone and Ultraviolet Data Centre (WOUDC) and its data-contributing agencies provided North American and European ozonesonde profiles; NASA's AErosol RObotic NETwork (AeroNet) and its data-contributing agencies provided North American and European AOD measurements; the MOZAIC Data Centre and its contributing airlines provided North American and European aircraft takeoff and landing vertical profiles; for European air quality data the folowing data centers were used: EMEP/EBAS and European Environment Agency/European Topic Center on Air and Climate Change/Air Quality e-reporting provided European air- and precipitation-chemistry data. The Finnish Meteorological Institute for providing biomass burning emission data for Europe. Data from meteorological station monitoring networks were provided by NOAA and Environment Canada (for the US and Canadian meteorological network data) and the National Center for Atmospheric Research (NCAR) data support section. Joint Research Center Ispra/Institute for Environment and Sustainability provided its ENSEMBLE system for model output harmonisation and analyses and evaluation. Although this work has been reviewed and approved for publication by the U.S. Environmental Protection Agency, it does not necessarily reflect the views and policies of the agency.

Appendix 1

The autocorrelation function (ACF) is derived by the autocovariance (ACV) and expresses the correlation of a time series with its lagged version (e.g. Chatfield, 2004):

$$ACV(k) = E\{[X(t) - \mu][X(t+k) - \mu]\} = Cov[X(t), X(t+k)];$$

$$ACF(k)=ACV(k)/ACV(0)$$

At any lag k, the autocovariance coefficients c_k are given by:

$$c_k = \frac{1}{N}\sum_{t=1}^{N-k}(x_t - \overline{x})(x_{t+k} - \overline{x})$$

And, as usual, the autocorrelation coefficients are given by normalizing c_k with c_0.

The partial autocorrelation function (PACF) measures the excess of correlation between two elements of $X(t)$ lagged by s elements not accounted for by the autocorrelation of the intermediate $s-1$ elements. In other words, the ACF of $X(t)$ and $X(t+s)$ includes all the linear dependence between the intermediate $s-1$ lags. The PACF allows to investigate the direct effect of lag t on the lag $t+s$.

The advantage of using ACF and PACF is that are function of the lag *k* only (and not of the specific time *t*). This condition holds only if *X(t)* is stationary (i.e. its mean and variance do not change over time). Several tests are available to check *X(t)* for stationarity (e.g. Chatfield, 2004). Differencing the time series is typically a way to achieve stationarity.

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Tables

TABLE 1. MSE (ppb^2) of the full, undecomposed ozone time series (FT) and relative fraction of MSE of the time series derived by filtering out the diurnal fluctuations (noDU) and of the time series derived by keeping only the diurnal fluctuations (DU). The diurnal signal has been isolated by applying a filter kz(13,5). The relative fraction of noDU and of DU not adding up to 100% is because the filter allows some leakage to the nearest frequencies (see Hogrefe et al. (2000) and Solazzo and Galmarini (2016) for details). *a)* NA; *b)*EU

a)

NA1			NA2			NA3		
CMAQ MSE- Summer								
FT (ppb^2)	noDU	DU	FT (ppb^2)	noDU	DU	FT (ppb^2)	noDU	DU
28.65	40%	41%	49.12	70%	23%	79.35	84%	13%
CAMQ MSE- Winter								
86.08	94%	5%	19.27	75%	21%	61.67	74%	21%

b)

EU1			EU2			EU3		
CHIMERE MSE- Summer								
FT (ppb^2)	noDU	DU	FT (ppb^2)	noDU	DU	FT (ppb^2)	noDU	DU
20.91	85%	10%	46.19	78%	15%	125.86	26%	67%
CHIMERE MSE- Winter								
20.87	85%	12%	19.95	85%	10%	39.91	38%	59%

TABLE 2. Linear correlation coefficient between the diurnal residuals of the regressors of Eq 3. The residuals are calculated by removing from the measured and modelled time series fluctuations faster the ~1.5 days. All the correlation values are significant up to 1% significance threshold. *a)* NA; *b)* EU

a)

	Correlation among diurnal component of residuals								
	ΔNO_2			ΔTemp			ΔWS		
	NA1	NA2	NA3	NA1	NA2	NA3	NA1	NA2	NA3
	SUMMER								
ΔNO_2	1	1	1	-0.6	-0.23	-0.65	-0.19	0.46	-0.26
ΔTemp	-0.6	-0.23	-0.65	1	1	1	0.62	0.53	0.7
ΔWS	-0.19	0.46	-0.26	0.62	0.53	0.7	1	1	1
	WINTER								
ΔNO_2	1	1	1	-0.63	-0.57	-0.56	-0.55	-0.05	-0.19
ΔTemp	-0.63	-0.57	-0.56	1	1	1	-0.63	0.47	0.35
ΔWS	-0.55	-0.05	-0.19	0.49	0.47	0.35	1	1	1

b)

Correlation among diurnal component of residuals	ΔNO			ΔNO_2			ΔTemp			ΔWS		
	EU1	EU2	EU3	EU1	EU2	EU3	EU1	EU2	EU3	EU1	EU2	EU3
SUMMER												
ΔNO	1	1	1	0.05	0.68	0.48	-0.08	-0.05	-0.27	-0.07	0.11	-0.02
ΔNO_2	0.05	0.68	0.48	1	1	1	0.57	0.18	-0.27	0.51	0.38	0.26
ΔTemp	-0.08	-0.05	-0.27	0.57	0.18	-0.27	1	1	1	0.81	0.63	0.21
ΔWS	-0.07	0.11	-0.02	0.51	0.38	0.26	0.81	0.63	0.21	1	1	1
WINTER												
ΔNO	1	1	1	0.31	0.6	0.73	0.02	-0.52	-0.62	0.03	0.12	0.06
ΔNO_2	0.31	0.6	0.73	1	1	1	-0.13	-0.7	-0.7	-0.01	0.09	0.11
ΔTemp	0.02	-0.52	-0.62	-0.13	-0.7	-0.7	1	1	1	0.48	0.02	-0.01
ΔWS	0.03	0.12	0.06	-0.01	0.09	0.11	0.48	0.02	-0.01	1	1	1

TABLE 3. Linear correlation coefficient between the residuals of the regressors of Eq 3, when the diurnal fluctuations are filtered out. The residuals are calculated by removing from the measured and modelled time series fluctuations faster the ~1.5 days. All the correlation values are significant up to 1% significance threshold. *a)* NA; *b)* EU

a)

Correlation among residuals (diurnal fluctuations removed)	ΔNO_2			ΔTemp			ΔWS		
	NA1	NA2	NA3	NA1	NA2	NA3	NA1	NA2	NA3
SUMMER									
ΔNO_2	1	1	1	-0.2	-0.02	-0.26	-0.06	-0.05	-0.19
ΔTemp	-0.2	-0.02	-0.26	1	1	1	0.28	0.09	0.42
ΔWS	-0.06	-0.05	-0.19	0.28	0.09	0.42	1	1	1
WINTER									
ΔNO_2	1	1	1	-0.12	-0.42	-0.03	-0.02	-0.16	-0.11
ΔTemp	-0.12	-0.42	-0.03	1	1	1	0.54	0.34	0.13
ΔWS	-0.02	-0.16	-0.11	0.54	0.34	0.13	1	1	1

b)

Correlation among residuals (diurnal fluctuations removed)	ΔNO			ΔNO_2			ΔTemp			ΔWS		
	EU1	EU2	EU3	EU1	EU2	EU3	EU1	EU2	EU3	EU1	EU2	EU3
SUMMER												
ΔNO	1	1	1	0.22	0.71	0.69	0.12	-0.23	-0.03	0.06	-0.23	-0.08
ΔNO_2	0.22	0.71	0.69	1	1	1	-0.27	-0.41	-0.11	-0.54	-0.43	-0.01
ΔTemp	0.12	-0.23	-0.03	-0.27	-0.41	-0.11	1	1	1	0.44	0.22	0.36
ΔWS	0.06	-0.23	-0.08	-0.54	-0.43	-0.01	0.44	0.22	0.36	1	1	1
WINTER												
ΔNO	1	1	1	0.21	0.64	0.46	-0.22	-0.19	-0.02	-0.15	-0.14	-0.01
ΔNO_2	0.21	0.64	0.46	1	1	1	-0.09	-0.38	-0.35	-0.07	-0.2	-0.08
ΔTemp	-0.22	-0.19	-0.02	-0.09	-0.38	-0.35	1	1	1	0.37	-0.1	0.38
ΔWS	-0.15	-0.14	-0.01	-0.07	-0.2	-0.08	0.37	-0.1	0.38	1	1	1

FIGURES

Figure 1 Continental domains and sub-regions used for analysis. The networks of ozone receptors are also shown.

Figure 2. Average monthly and diurnal curves constructed from January – December 2010 time series of hourly ozone observations and model simulations for three North American sub-regions

Figure 3. Average monthly and diurnal curves constructed from January – December 2010 time series of hourly ozone observations and model simulations for three European sub-regions.

Figure 4 MSE decomposition for June – August hourly ozone into bias^2, variance and covariance for the three NA sub-regions. Results are presented separately for daylight hours (left) and nighttime hours (right).

Figure 5 MSE decomposition for June – August hourly ozone into bias^2, variance and covariance for the three EU sub-regions (the zero_Dep data refers to the month of July only). Results are presented separately for daylight hours (left) and nighttime hours (right)

Figure 6 CMAQ MSE breakdown for summer and winter for the base case and sensitivity simulations over NA. The error coeffcients F_b,F_v,F_c are reported on the left axis, the total MSE (ppb^2) on the right axis (red triangles). The '+' and '-' signs within the bias and variance portions of the errors indicate model over- or under-prediction of mean concentration or variance, respectively. The values in the covariance portion indicate the correlation coeffcient between modelled and observed time series. *a)* hourly time series of ozone (base case); *b)* hourly time series of '20% reduction' scenario; *c)* hourly time series of 'zero boundary conditions' scenario; *d)* hourly time series of the 'zeroed anthropogenic emissions' scenario; *e)* base case rolling average daily maximum 8-hour ozone time series. For the analysis of hourly time series in panels a) – d), results are provided separately for daytime and nighttime.

Figure 7. Chimere MSE breakdown for summer and winter for the base case and sensitivity simulations over EU. The error coeffcients F_b,F_v,F_c are reported on the left axis, the total MSE (ppb^2) on the right axis (red triangles). The '+' and '-' signs within the bias and variance portions of the errors indicate model over- or under-prediction of mean concentration or variance, respectively. The values in the covariance portion indicate the correlation coeffcient between modelled and observed time series. *a)* hourly time series of ozone (base case); *b)* hourly time series of '20% reduction' scenario; *c)* hourly time series of 'constant boundary conditions' scenario; *d)* hourly time series of the 'zeroed anthropogenic emissions' scenario; *e)* base case rolling average daily maximum 8-hour ozone time series. For the analysis of hourly time series in panels a) – d), results are provided separately for daytime and nighttime.

Figure 8. Top row: Spatial maps of RMSE (in ppb) for the base case. Middle row: Percentage RMSE changes for the zeroed emissions case with respect to the base case. Lower row: Percentage RMSE changes for the zeroed boundary condition case with respect to the base case. Left column: Winter months (DJF); Right column: summer months (JJA).

Figure 9 Top row: Spatial maps of RMSE (in ppb) for the base case. Middle row: Percentage RMSE changes for the zeroed emissions case with respect to the base case. Lower row: Percentage RMSE changes for the constant boundary condition case with respect to the base case.. Left column: Winter months (DJF); Right column: summer months (JJA).

Figure 10. Annual time series of differences between CMAQ and observed O_3 (ΔO_3, top panel) and Morlet wavelet analysis of the periodogram of ΔO_3 (lower panel) for the three NA subdomains. Black contours lines identify the 95% confidence interval. The period (in days) is reported in the vertical axis, while the quantiles of the power spectral density are measured in ppb^2. (the scale reports the quantiles of the power spectrum).

Figure 11. Same as in FIGURE 10 for Chimere over the three EU subdomains

Figure 12. Phase shift of the diurnal cycle (in hours). A positive phase shift indicates that the model peak is 'late', while a negative phase shift indicates that the modelled peak precedes the observed peak. This analysis includes urban and suburban stations in addition to rural stations.

Figure 13. As in Figure 12 for EU.

Figure 14. Normalised MSE produced by lagging the observed diurnal cycle with respect to itself. The MSE due to such a shift is entirely due to covariance error. The plots are presented for EU2 (left) and NA2 (right) for the

months of JJA. The top panel shows the impact of the phase shift on the DU component, and the lower panels show results for the undecomposed time series (FT). For EU2, a shift of ±3 hours causes an MSE of ~0.5 times the variance of the observations.

Figure 15. CMAQ model: autocorrelation (ACF) and partial autocorrelation (PACF) function for *a)* the differenced time series of residuals of ozone (mod-obs) and *b)* the differenced time series of residual of ozone obtained by filtering out the diurnal fluctuations from the modelled and observed time series. The differentiation is necessary to remove non-stationarity.

Figure 16. Chimere model: autocorrelation (ACF) and partial autocorrelation (PACF) function for *a)* the differenced time series of residuals of ozone (mod-obs) and *b)* the differenced time series of residual of ozone obtained by filtering out the diurnal fluctuations from the modelled and observed time series. The differentiation is necessary to remove non-stationarity.

Figure 17. Percentage of variance explained by the regressors (the total R^2 for the regression is reported in the title of each panel). The relative importance of each variable is assessed by using a bootstrap resampling. The plots at the bottom show the ACF and PACF of the yearly time series of residual of the fit, i.e. the portion of the ozone time series that was not captured by the linear regressions on the available variables.

Figure 18. Same as **Figure 17** for EU.

FIGURES

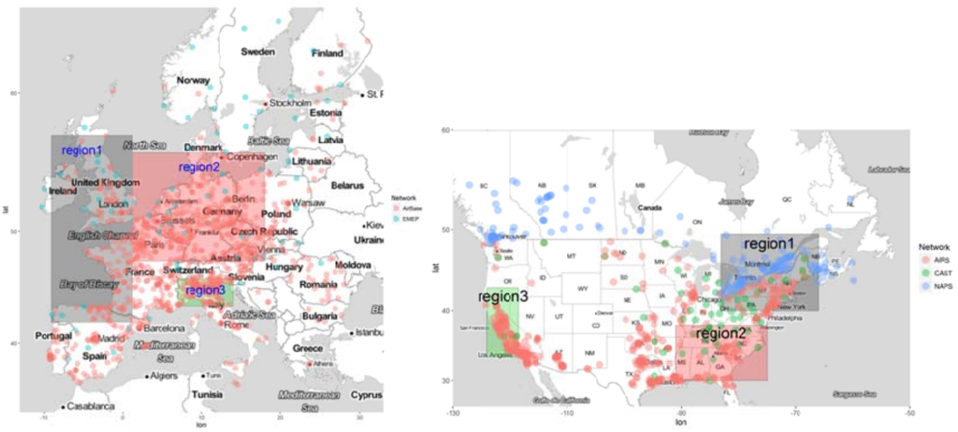

FIGURE 1. Continental domains and sub-regions used for analysis. The networks of ozone receptors are also shown

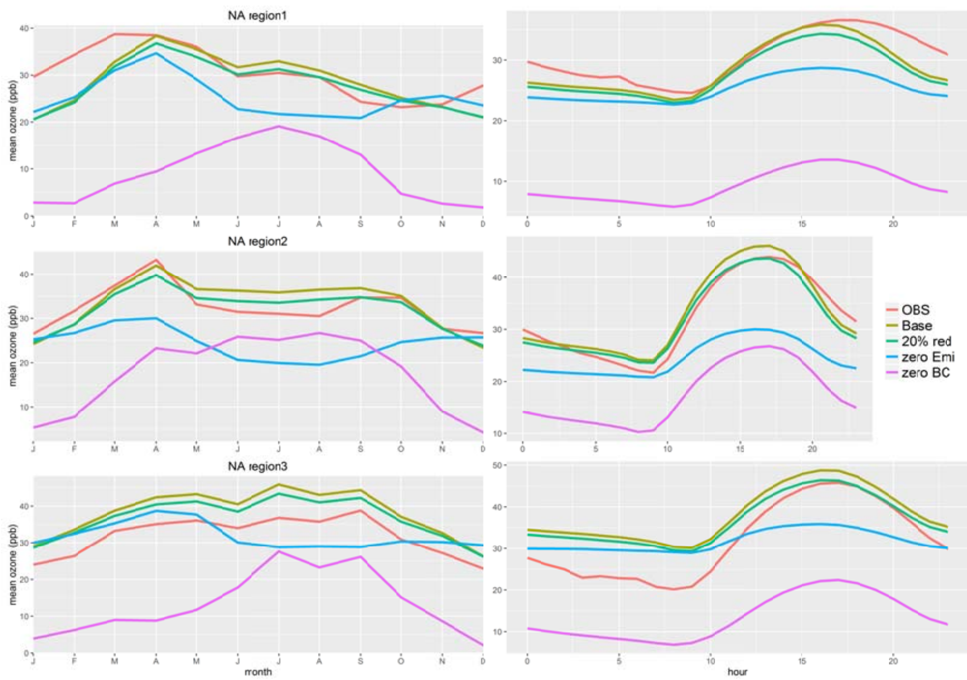

FIGURE 2. Average monthly and diurnal curves constructed from January – December 2010 time series of hourly ozone observations and model simulations for three North American sub-regions.

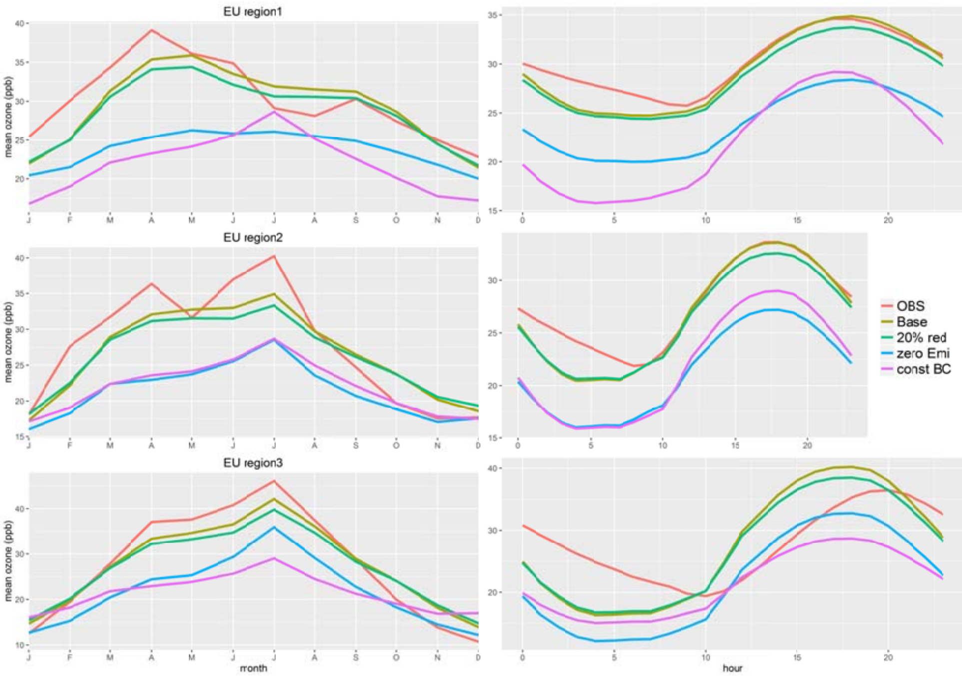

FIGURE 3. Average monthly and diurnal curves constructed from January – December 2010 time series of hourly ozone observations and model simulations for three European sub-regions.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-257, 2017
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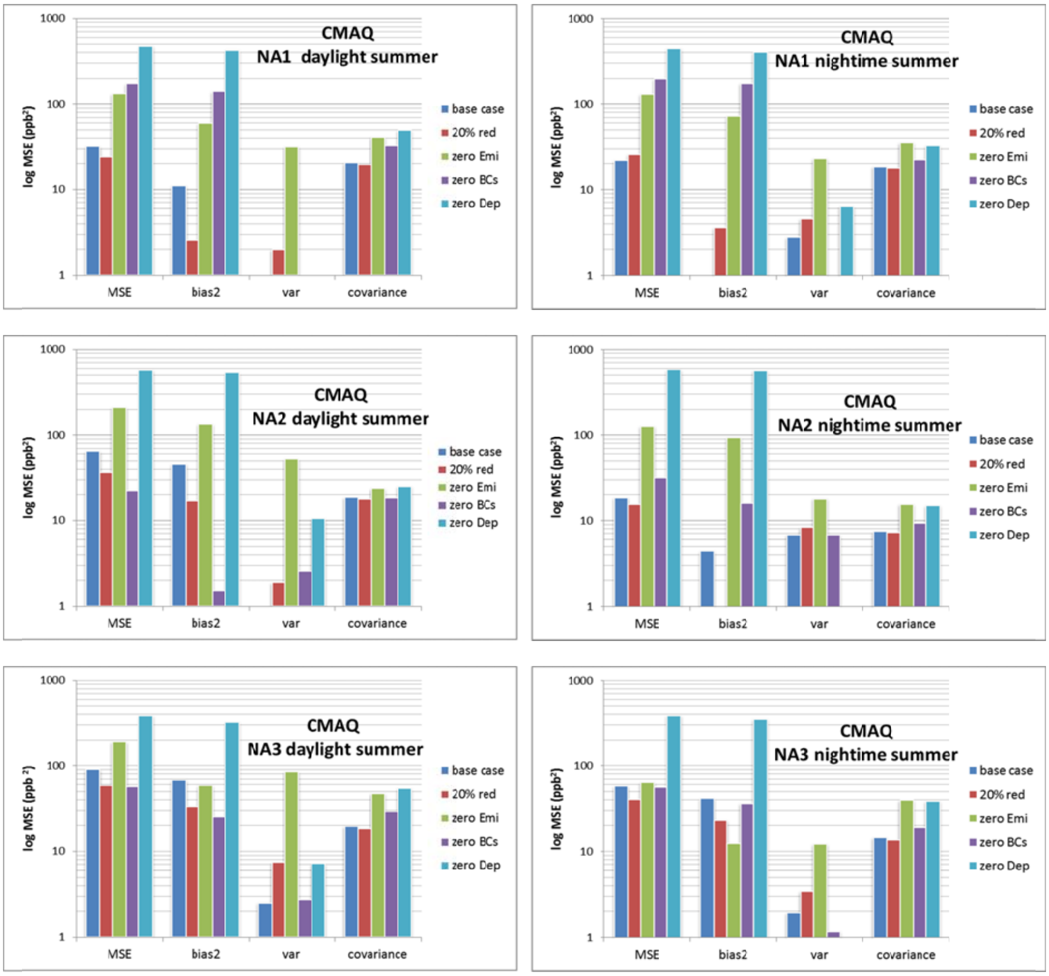

FIGURE 4. MSE decomposition for June – August hourly ozone into bias^2, variance and covariance for the three NA sub-regions. Results are presented separately for daylight hours (left) and night-time hours (right)

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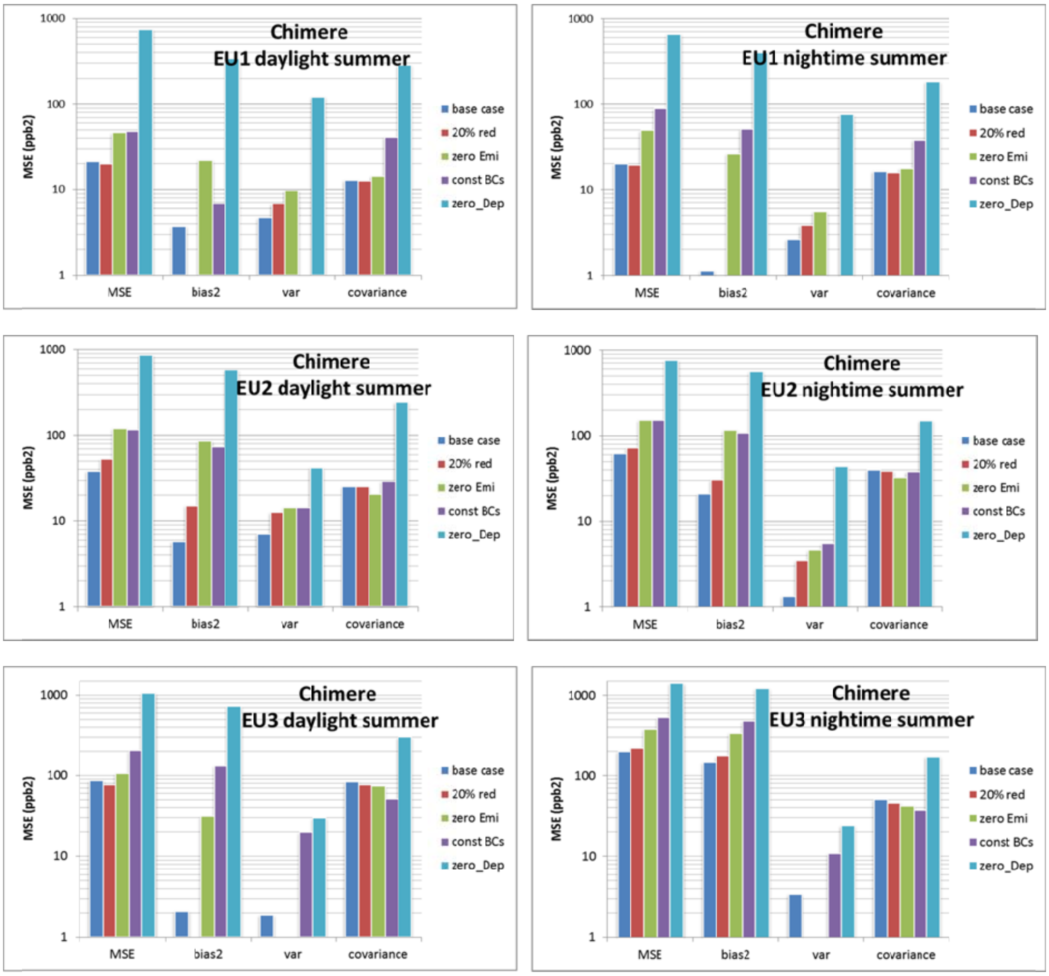
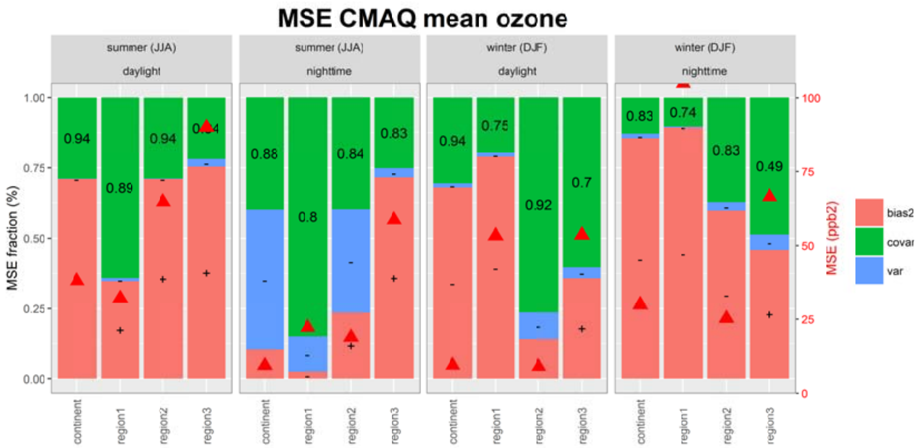
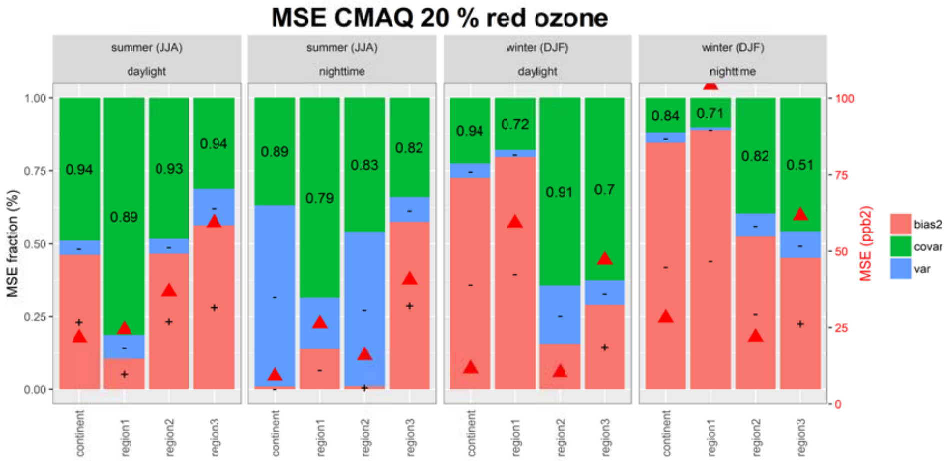


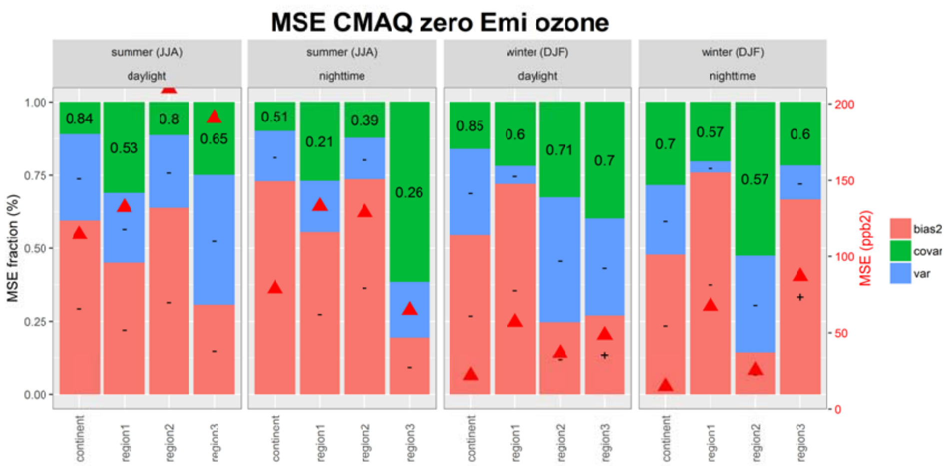
FIGURE 5. MSE decomposition for June – August hourly ozone into bias^2, variance and covariance for the three EU sub-regions (the zero_Dep data refers to the month of July only). Results are presented separately for daylight hours (left) and night-time hours (right)


MSE CMAQ mean ozone
summer (JJA) daylight
summer (JJA) nighttime
winter (DJF) daylight
winter (DJF) nighttime
MSE fraction (%)
MSE (ppb2)
bias2
covar
var
continent
region1
region2
region3

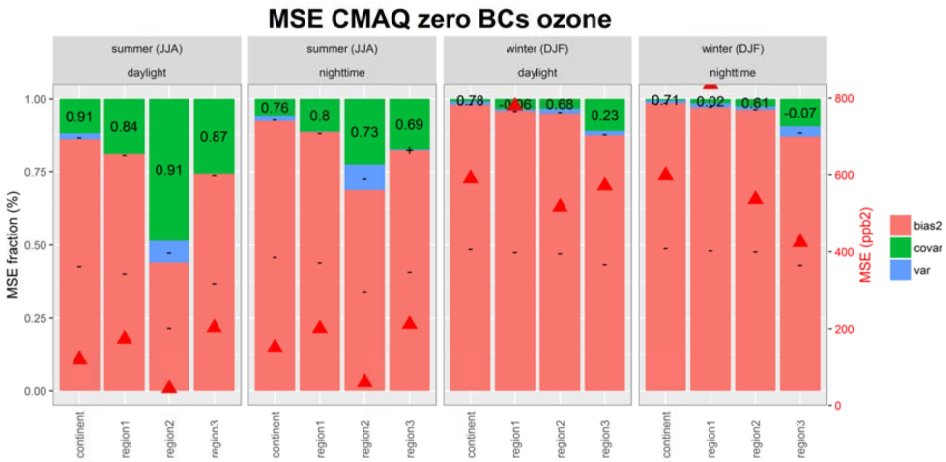
a)


MSE CMAQ 20 % red ozone
summer (JJA) daylight
summer (JJA) nighttime
winter (DJF) daylight
winter (DJF) nighttime
MSE fraction (%)
MSE (ppb2)
bias2
covar
var
continent
region1
region2
region3

b)


MSE CMAQ zero Emi ozone
summer (JJA) daylight
summer (JJA) nighttime
winter (DJF) daylight
winter (DJF) nighttime
MSE fraction (%)
MSE (ppb2)
bias2
covar
var
continent
region1
region2
region3

c)



d)

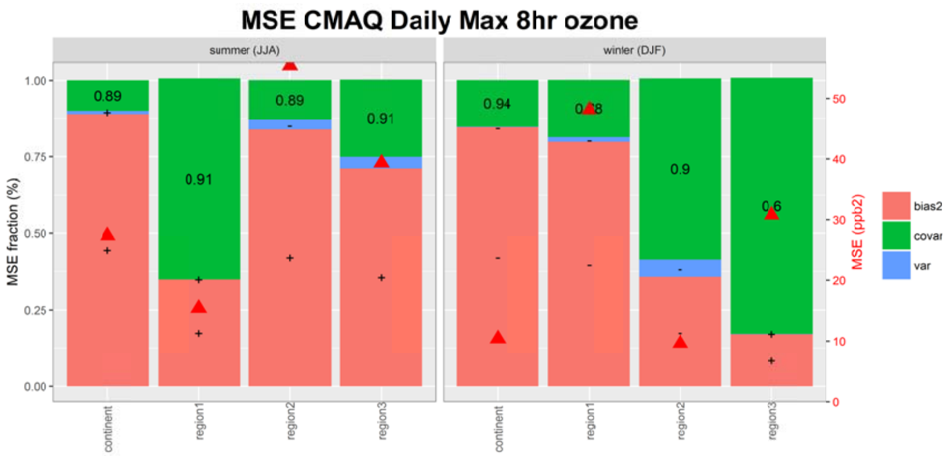
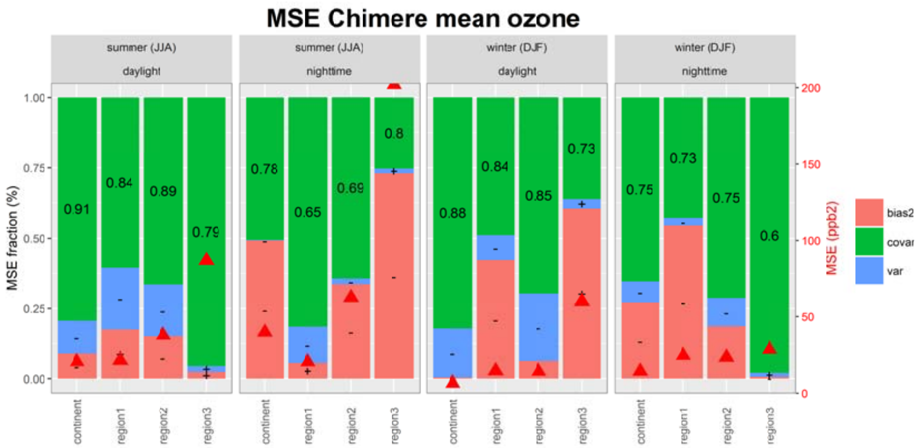
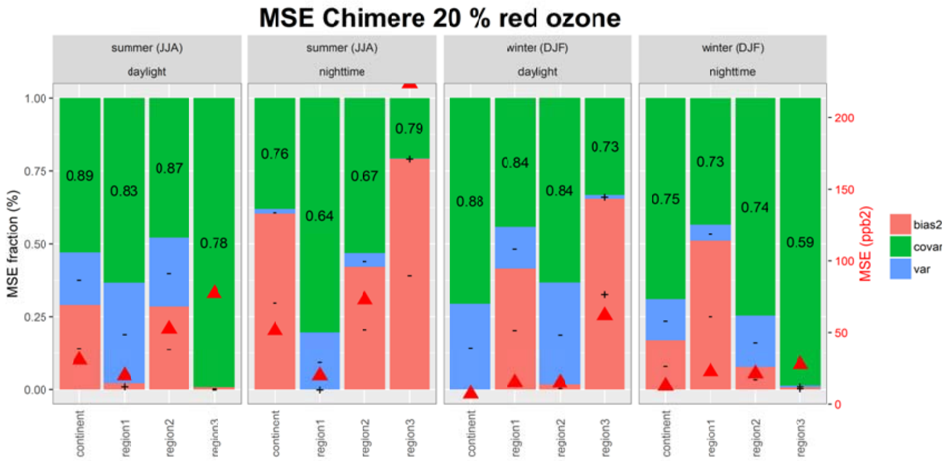


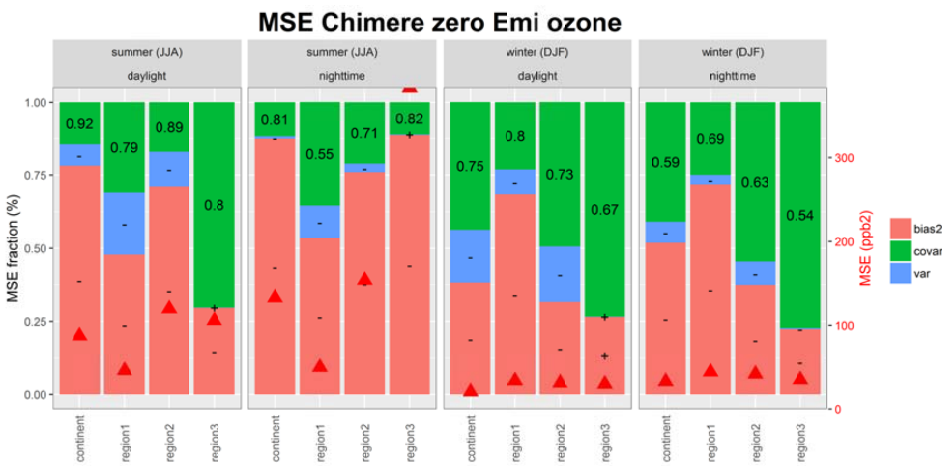
FIGURE 6. CMAQ MSE breakdown for summer and winter for the base case and sensitivity simulations over NA. The error coeffcients F_b,F_v,F_c are reported on the left axis, the total MSE (ppb^2) on the right axis (red triangles). The '+' and '-' signs within the bias and variance portions of the errors indicate model over- or under-prediction of mean concentration or variance, respectively. The values in the covariance portion indicate the correlation coeffcient between modelled and observed time series. *a)* hourly time series of ozone (base case); *b)* hourly time series of '20% reduction' scenario; *c)* hourly time series of 'zero boundary conditions' scenario; *d)* hourly time series of the 'zeroed anthropogenic emissions' scenario; *e)* base case rolling average daily maximum 8-hour ozone time series. For the analysis of hourly time series in panels a) – d), results are provided separately for daytime and nighttime. .


MSE Chimere mean ozone
summer (JJA) daylight
summer (JJA) nighttime
winter (DJF) daylight
winter (DJF) nighttime
MSE fraction (%)
MSE (ppb2)
bias2
covar
var
continent
region1
region2
region3

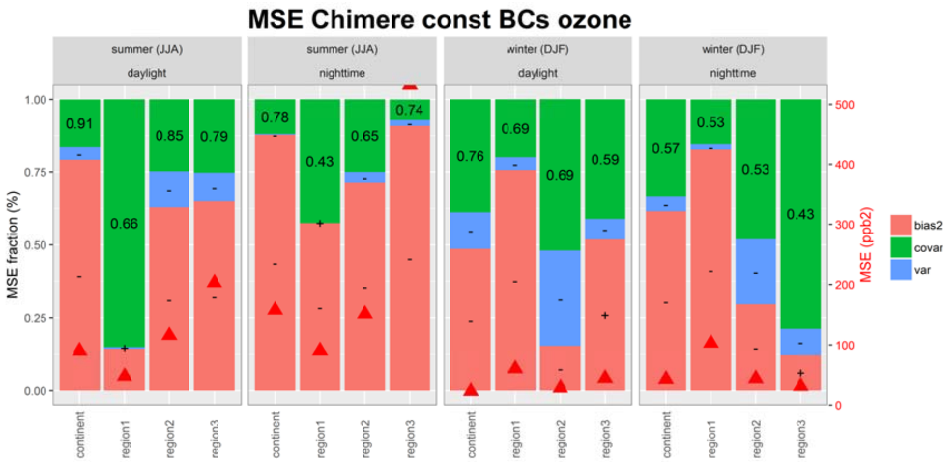
a)


MSE Chimere 20 % red ozone
summer (JJA) daylight
summer (JJA) nighttime
winter (DJF) daylight
winter (DJF) nighttime
MSE fraction (%)
MSE (ppb2)
bias2
covar
var
continent
region1
region2
region3

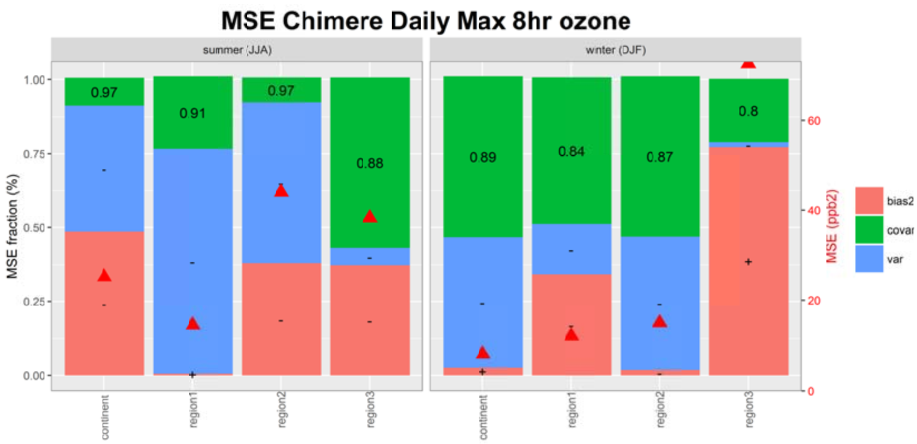
b)


MSE Chimere zero Emi ozone
summer (JJA) daylight
summer (JJA) nighttime
winter (DJF) daylight
winter (DJF) nighttime
MSE fraction (%)
MSE (ppb2)
bias2
covar
var
continent
region1
region2
region3

c)



d)



e)

FIGURE 7. Chimere MSE breakdown for summer and winter for the base case and sensitivity simulations over EU. The error coeffcients F_b,F_v,F_c are reported on the left axis, the total MSE (ppb^2) on the right axis (red triangles). The '+' and '-' signs within the bias and variance portions of the errors indicate model over- or under-prediction of mean concentration or variance, respectively. The values in the covariance portion indicate the correlation coeffcient between modelled and observed time series. *a)* hourly time series of ozone (base case); *b)* hourly time series of '20% reduction' scenario; *c)* hourly time series of 'constant boundary conditions' scenario; *d)* hourly time series of the 'zeroed anthropogenic emissions' scenario; *e)* base case rolling average daily maximum 8-hour ozone time series. For the analysis of hourly time series in panels a) – d), results are provided separately for daytime and nighttime. .

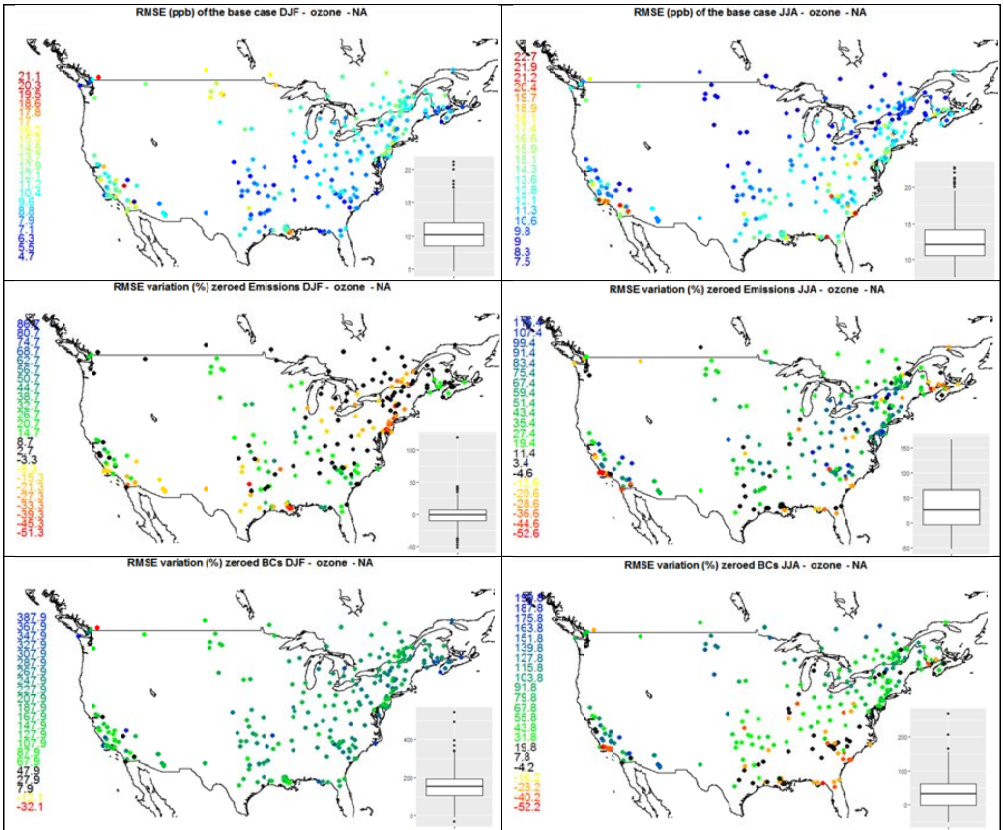
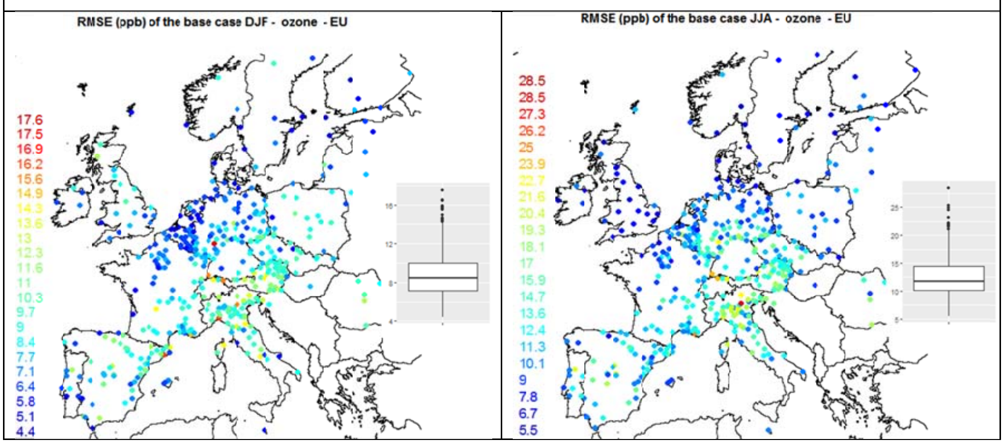


Figure 8. Top row: Spatial maps of RMSE (in ppb) for the base case. Middle row: Percentage RMSE changes for the zeroed emissions case with respect to the base case. Lower row: Percentage RMSE changes for the zeroed boundary condition case with respect to the base case. Left column: Winter months (DJF); Right column: summer months (JJA).



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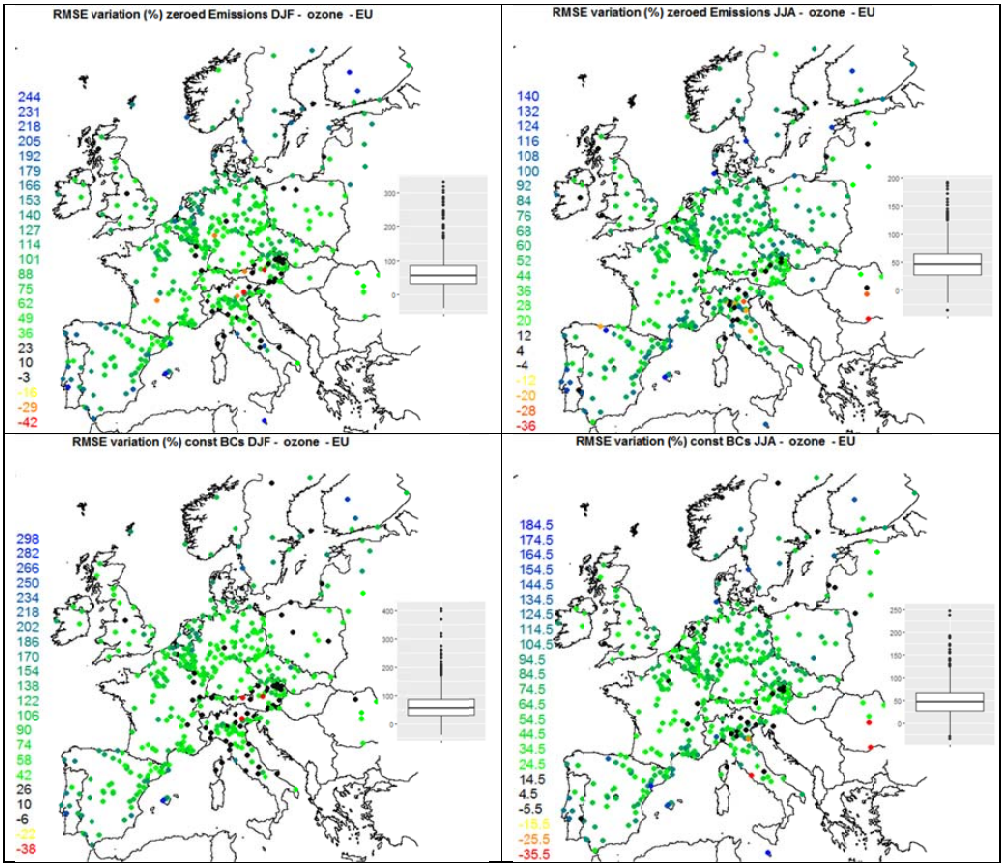

FIGURE 9. Top row: Spatial maps of RMSE (in ppb) for the base case. Middle row: Percentage RMSE changes for the zeroed emissions case with respect to the base case. Lower row: Percentage RMSE changes for the constant boundary condition case with respect to the base case. Left column: Winter months (DJF); Right column: summer months (JJA).

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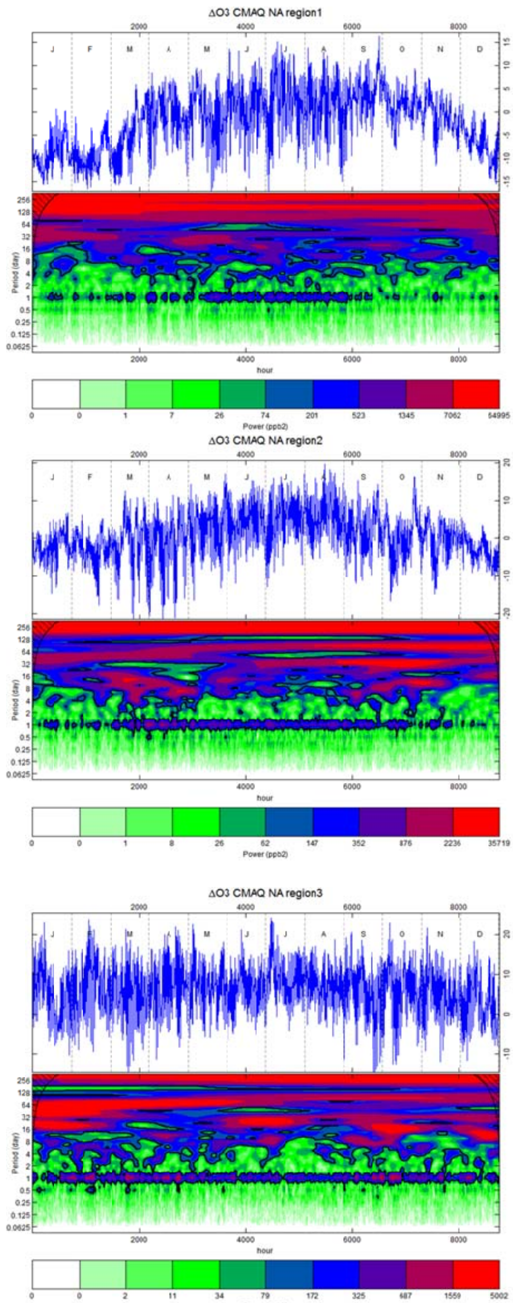

FIGURE 10. Annual time series of differences between CMAQ and observed O_3 (ΔO_3, top panel) and Morlet wavelet analysis of the periodogram of ΔO_3 (lower panel) for the three NA subdomains. Black contours lines identify the 95% confidence interval. The period (in days) is reported in the vertical axis, while the quantiles of the power spectral density are measured in ppb^2.

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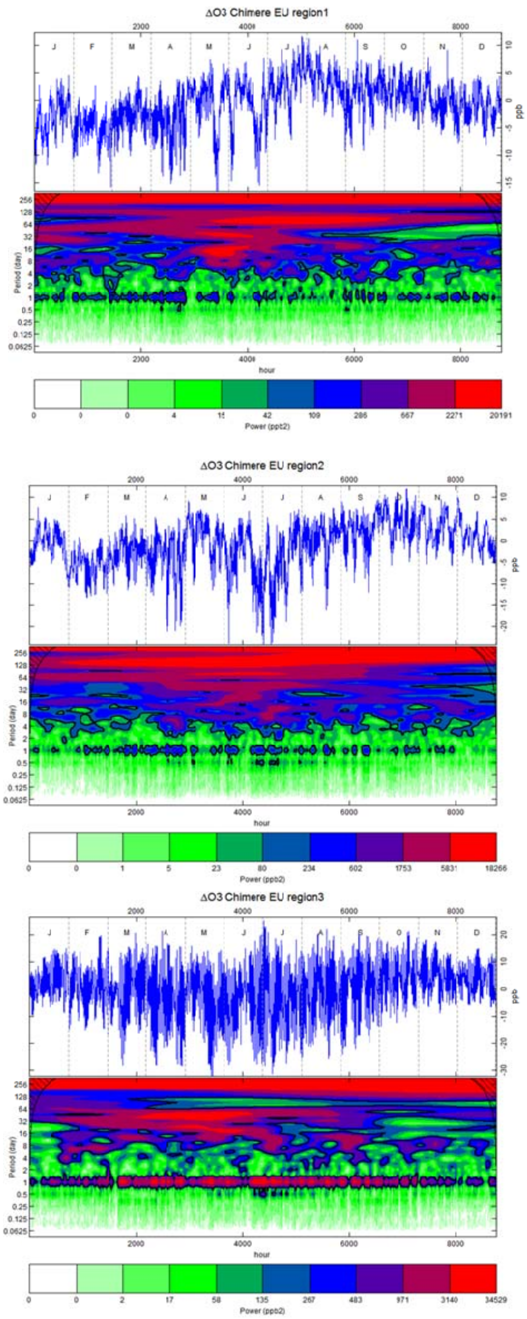


FIGURE 11. As in Figure 10 for Chimere over the three EU subdomains.

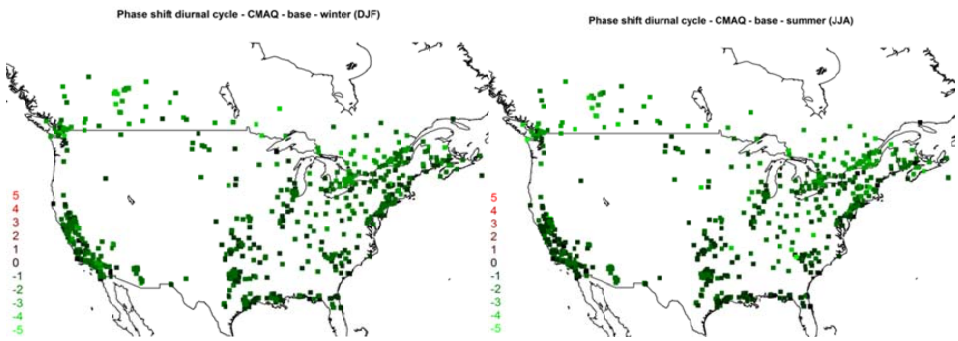


Figure 12. Phase shift of the diurnal cycle (in hours). A positive phase shift indicates that the model peak is 'late', while a negative phase shift indicates that the modelled peak precedes the observed peak. This analysis includes urban and suburban stations in addition to rural stations.

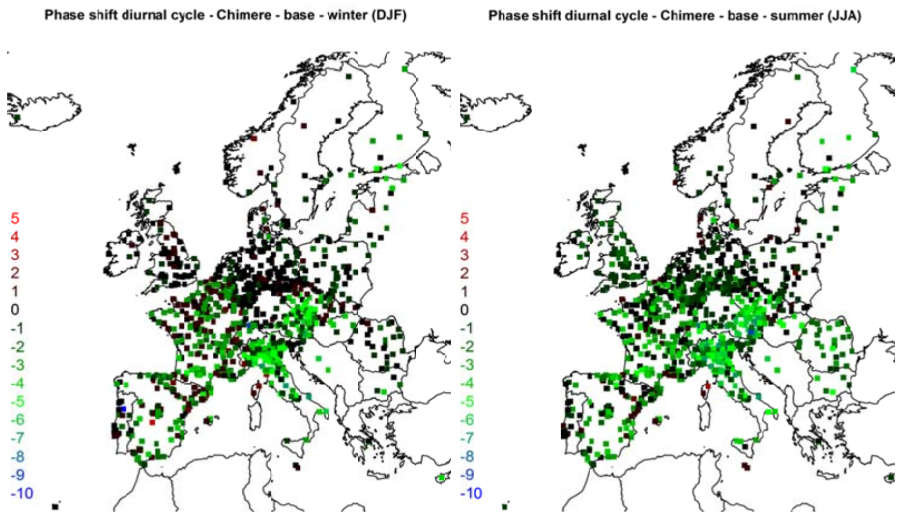


FIGURE 13. As in Figure 12 for EU.

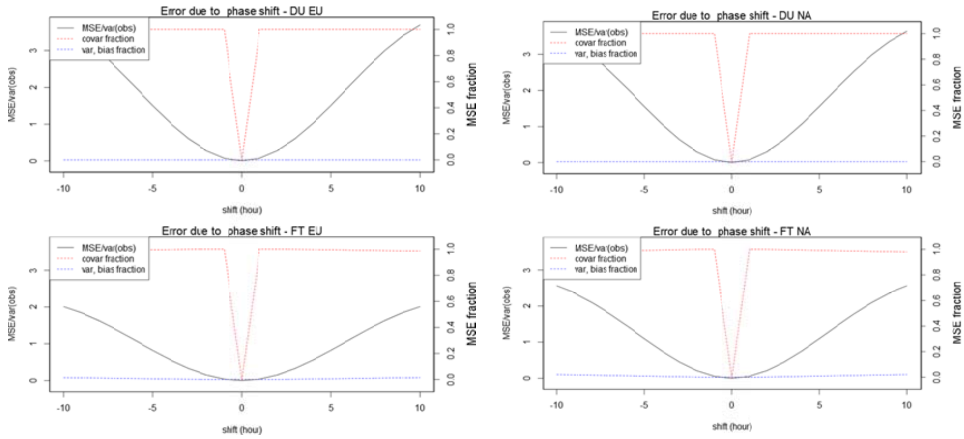
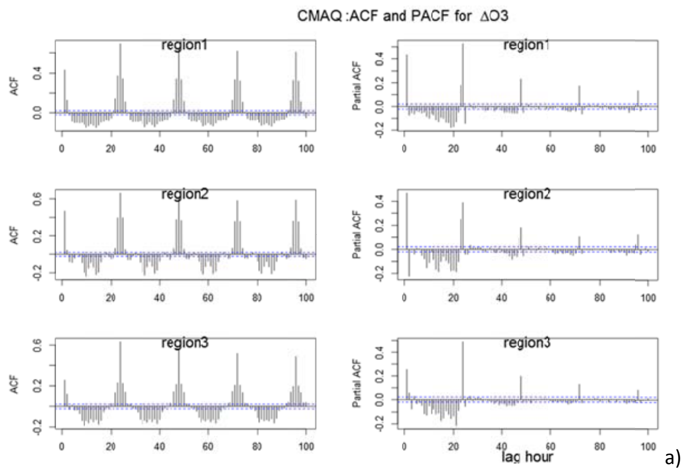
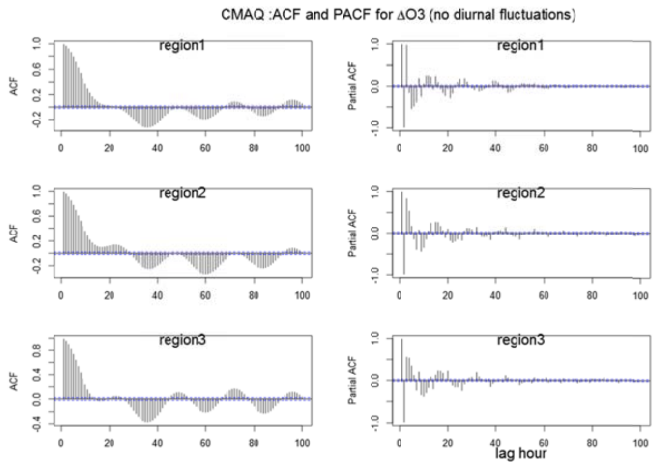


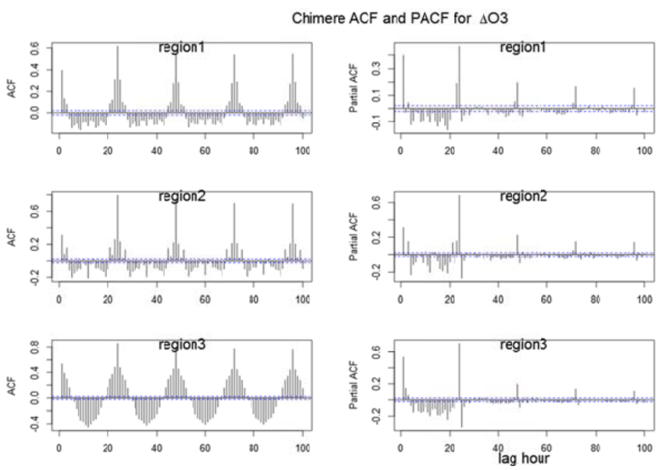
FIGURE 14. Normalised MSE produced by lagging the observed diurnal cycle with respect to itself. The MSE due to such a shift is entirely due to covariance error. The plots are presented for EU2 (left) and NA2 (right) for the months of JJA. The top panel shows the impact of the phase shift on the DU component, and the lower panels show results for the undecomposed time series (FT). For EU2, a shift of ±3 hours causes an MSE of ~0.5 times the variance of the observations.





b)

FIGURE 15. CMAQ model: autocorrelation (ACF) and partial autocorrelation (PACF) function for *a)* the differenced time series of residuals of ozone (mod-obs) and *b)* the differenced time series of residual of ozone obtained by filtering out the diurnal fluctuations from the modelled and observed time series. The differentiation is necessary to remove non-stationarity and thus to make the the acf and pacf values depending on lag only.



a)

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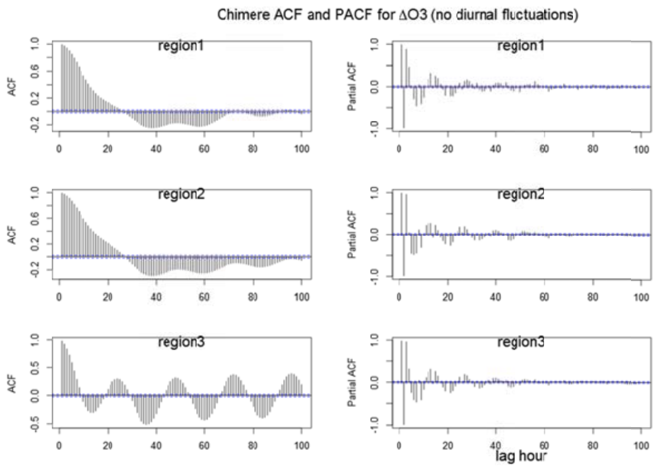
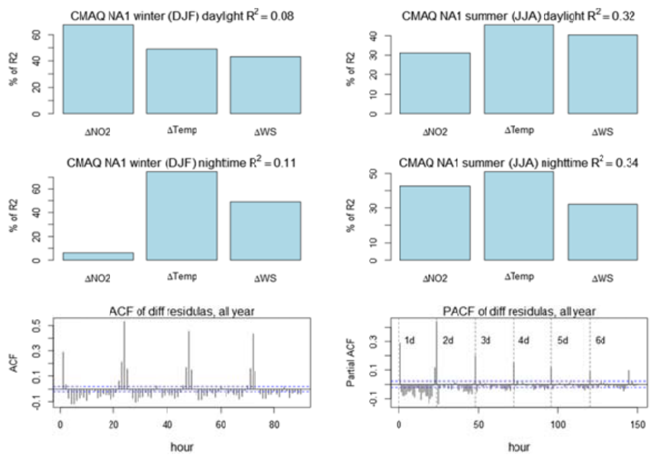
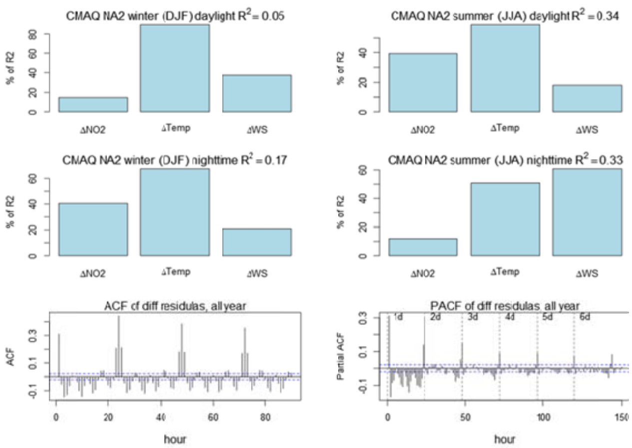


b)

FIGURE 16. Chimere model: autocorrelation (ACF) and partial autocorrelation (PACF) function for *a)* the differenced time series of residuals of ozone (mod-obs) and *b)* the differenced time series of residual of ozone obtained by filtering out the diurnal fluctuations from the modelled and observed time series. The differentiation is necessary to remove non-stationarity and thus to make the ACF and PACF values depending on lag only.





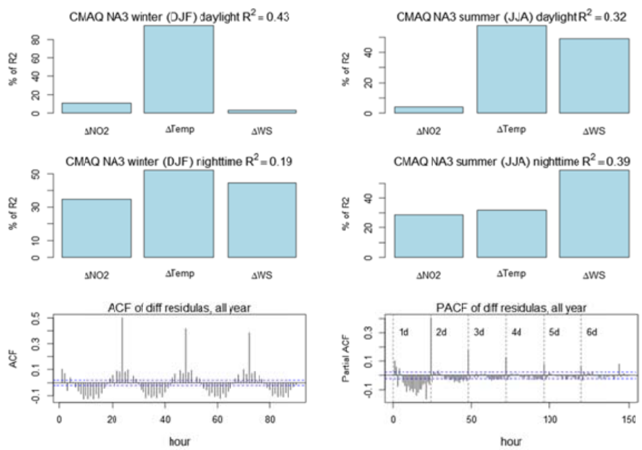


FIGURE 17. Percentage of variance explained by the regressors (the total R^2 for the regression is reported in the title of each panel). The relative importance of each variable is assessed by using a bootstrap resampling. The plots at the bottom show the ACF and PACF of the yearly time series of residual of the fit, i.e. the portion of the ozone time series that was not captured by the linear regressions on the available variables.

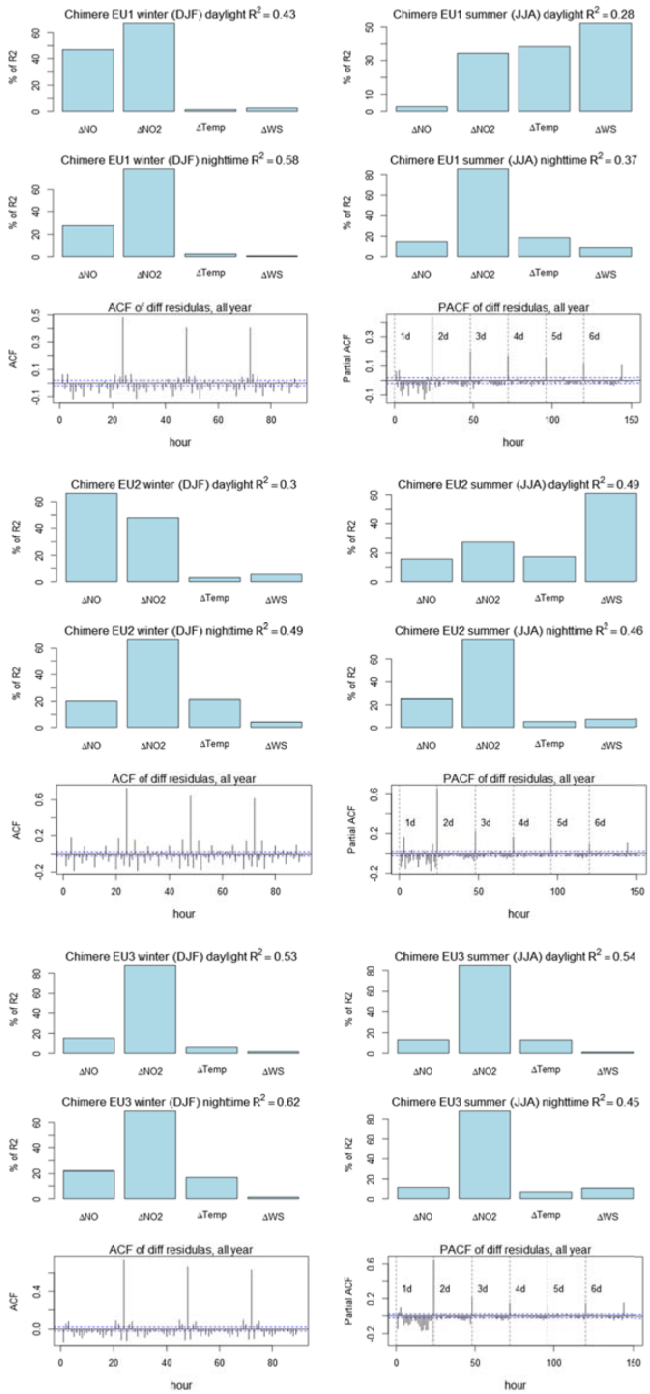


FIGURE 18. Same as Figure 17 for EU.