THE ROLE OF THE GLOBAL WARMING IN TROPOSPHERIC OZONE

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INTRODUCTION

The ozone concentration in a given area depends on many factors, including sunlight, meteorology, temperature and the presence of precursors as nitrogen oxides (NO_x) and volatile organic compounds (VOCs). A fraction of the emission of these last compounds comes from natural sources referred to as biogenic emissions, whereas another important part comes from anthropogenic sources as industry and traffic. Their central importance lies in that it is highly difficult to control these emissions, specially the biogenic part, mainly because they could be affected by global warming, as increased temperature accelerates both emissions and the chemical reaction involved in the formation of ozone (Tao et al., 2003). In this study, a sensitivity analysis was performed focusing on the impact of the meteorology and especially on the increment of temperature in the biogenic and traffic evaporative emissions. Attention was also paid to the kinetic of the chemical reactions which would give rise to an increment of ozone concentration (Walcek et al., 1995; Vizuete et al., 2002). Hourly simulation results generated by meteorological and air pollution models show the role of temperature in predicting ozone concentrations, hence the importance of temperature accuracy forecasted by the meteorological models.

METHODOLOGY

Hourly troposheric ozone concentrations and a new scenario were generated for the base case. This new scenario consisted of different increasing temperature hourly rates (that depended on their normalised bias) generated in order to explore the effect of temperature on a high ozone case study in Catalonia, a region located in the northeast of Spain. The time frame of the simulation spanned from 10 to 14 June 2003 and the domain under study corresponds to domain 02 showed in figure 1. Concentrations were estimated both for a base scenario, in which the adjusted temperatures and emissions where not taken into account, and a new scenario where we adjusted this variables.

Meteorological and air pollution modelling

The PSU/NCAR mesoscale model, MM5 (Grell et al., 1994), version 3.7 was used to generate meteorological fields, which were the inputs for the air pollution modelling system. Meteorological simulations were performed for four two ways nested domains (figure 1) with resolutions of 27 km, 9 km and 3 km. The coarse domain covers Southern Europe, Spain, half of France and South of Italy. An inner domain of 30x30 cells (9km) covers Catalonia while two 3 km resolution domains, the smallest ones, cover two areas whose interest lies in their high ozone level measurements. The initial and boundary conditions are updated every six hours with information obtained from the European Centre for medium Range Weather Forecast (ECMWF) model with a 0.5ºx0.5º resolution. For the three inner domains, we use a topography and land-use date base with 30'' resolution. For the two outer domains the horizontal resolution is 5'. High vertical resolution is prescribed in the ABL, 20 levels, with higher resolution (15 m approximately) on the low levels. The boundary layer processes are calculated using the MRF scheme based on Troen and Mahrt (1986).

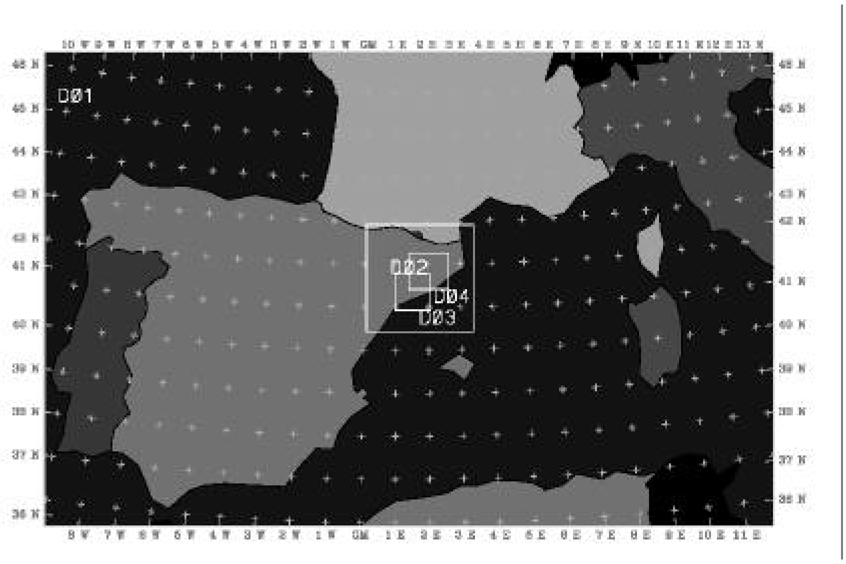

Fig. 1; Model domains

The chemical transport model used in this study is the U.S. EPA models-3/CMAQ models (Byun and Ching, 1999). This model is supported by the U.S. Environmental Agency, and is continuously under development, including a variety of the most advanced configurations and parameterizations. The air pollution modelling simulations are performed using the same domains as the meteorological model did.

Emissions model

Industry, biogenic, mobile and domestic emissions were estimated using the MECA 2006 model (Ortega et al., 2006). This model is applied over domains two, three and four while for the biggest domain the emission inventory is quantified by the top-down approach using EMEP emissions. A special module is developed in order to adapt EMEP domain (50X50 km^2) to the biggest MM5-CMAQ domain ((27x27 km^2). MECA 2006 model is developed by the authors with high spatial (3 km^2 cells) and temporal (1 hour) resolutions. It was developed into a GIS environment to estimate Catalonia's emissions during the year 2006. It includes the most important primary air pollutants (NOx, NMVOCs, CO, SO2, PM10 and PM 2.5) coming from vegetation, on-road traffic and industries and the emissions by fossil fuel consumption and domestic-commercial solvent use.

Emission scenarios

A base episode and a modified emission scenario were used to estimate ozone concentrations during a high ozone episode occurred in June 2003. Hourly gridded estimates of ozone concentrations were generated using the following scenarios:

- Scenario A: Base case, unadjusted emissions using MECA emissions model.
- Scenario B: Adjusted emissions corresponding to the hourly temperature increment.

The following question could be associated to this scenario: if temperature is increased, meteorological conditions such as cloudiness, wind's velocity and direction as well as weather patterns could be altered. Consequently, not only would temperature change but also the meteorological situation. However, in the case study this argument is not conclusive, as high ozone episodes are usually related to high temperatures which not are well predicted by meteorological models. As a matter of fact, the mean bias points out that the model tends to underestimate maximum air surface temperature up to five degrees. The situation is not the same for wind velocity and direction as the mean bias is perceptibly lower. In consequence, we assume that increasing the air surface temperature in high ozone concentration situations does not basically alter the weather pattern. Estimated ozone concentrations for each of the scenarios were compared according to several metrics (Yu et al., 2006).

RESULTS

Models performances for the base case studied.

Concerning the meteorological model, the evaluation is focused on temperature fields, as the ozone concentration sensitivity is related to this variable. The period evaluate corresponds to June episode occurred during summer 2003. Several statistics have been calculated: the mean bias (B_{MB}), the normalized mean bias (B_{NMB}), the mean absolute gross error (E_{MAGE}) and the mean normalized absolute error (E_{MNAE}), defined as

$$B_{MB} = \frac{1}{N}\sum M_i - O_i = \overline{M} - \overline{O} \qquad (1)$$

$$E_{MAGE} = \frac{1}{N}\sum \left| M_i - O_i \right| \qquad (2)$$

$$E_{RMSE} = \left[\frac{1}{N}\sum (M_i - O_i)^2 \right]^{\frac{1}{2}} \qquad (3)$$

$$B_{MNB} = \frac{1}{N}\sum \left(\frac{M_i - O_i}{O_i} \right) = \left(\frac{1}{N}\sum \frac{M_i}{O_i} - 1 \right) \qquad (4)$$

Table I provide some diurnal period air temperature performance statistics. In addition to June episode we have added in this table some others warm periods to evidence that MM5 model always tends to under-predicts higher temperatures during warmer periods. Statistics show an appreciable difference between the air temperatures forecasted by the meteorological model and those measured by local stations. Predictions always underestimated air temperatures, with differences ranging from 13% to 17%.

Table 1. Model evaluation statistics for hourly temperature (ºC) for the base case(scenario A) and warmer periods occurred during summer 2003.

Diurnal Period 09-17 UTC	B_{MNB} June	E_{MNAE} June	B_{MNB} jul	E_{MNAE} jul	B_{MNB} 1 Ag	E_{MNAE} 1 Ag	B_{MNB} 6 Ag	E_{MNAE} 6 Ag
	-0.13	0.14	-0.12	0.14	-0.17	0.17	-0.15	0.16

Ozone air quality model evaluation involves calculating the previous statistics by using only the hourly observation-prediction pairs during diurnal period or by using cutoff values in order to remove the influence of low concentrations occurring, for example, at night time. In this study the first option has been used, and a new statistic, the unpaired highest-prediction accuracy (A_u), has been added to the previous one. It is also called the unpaired peak accuracy test or unpaired peak prediction accuracy. It compares the maximum observed value across all monitors and time periods and the maximum predicted value across the entire simulation. This measure is unpaired because the peak observed and the estimated concentrations may have different locations and/or time periods. A positive A_u indicates that the model over-predicts, whereas a negative value indicates an under-prediction. It is very dependent on the location and density of the monitoring network. If a monitor is not placed in the position where the highest ozone concentration occurs and the model predicts accurately, the A_u may be deceivingly poor (Bell et al., 2004).

Table 2 provides several statistics of the model performance for the base case simulation for this study.

Table 2. Model evaluation statistics for hourly ozone concentration for the base case study, (scenario A).

Statistics	>70 µg/m^3	Max. daily values
Mean Bias B_{MB} (µg/m^3)	-23	-31
Normalized mean bias B_{NMB} (%)	-22	-27
Mean absolute gross error E_{MAGE} (µg/m^3)	24	31
Normalized mean absolute gross error E_{NMAGE} (%)	24	27
Unpaired highest prediction accuracy, A_u (%)	-14	

The modelling system performs as well or slightly worse than other modelling performance studies (Bell et al., 2004; Jimenez et al., 2006; Zhang et al., 2006). Negative bias indicates the under-prediction of the model, the same under-prediction can be observed in the negative value of the unpaired accuracy. Maximum daily normalized mean absolute gross error show some little efficiency from the model to predict maximum values.

Effects of temperature on tropospheric ozone concentration

In order to study how the underestimation of temperature can affect the model ozone predictions, a simulation with an adjusted temperature and emissions has been performed. Some statistic from the performance can be seen in Table 3.

Table 3. Model evaluation statistics for hourly ozone concentration for the case with adjusted temperature and emissions, (scenario B).

Statistics	>70 µg/m^3	Max. daily values
Mean Bias B_{MB}	-13	-14
Normalized mean bias B_{NMB} (%)	-13	-11
Mean absolute gross error E_{MAGE}	18	18
Normalized mean absolute gross error E_{NMAGE} (%)	18	16
Unpaired highest prediction accuracy, A_u (%)	-8	

Statistics show a better performance in case B than in case A. Statistics in maximum daily values have been reduced from an error of 27 % to an error of 16 %. That decreasing is not so important in the cut-off hourly validation, which means that the adjusted temperature lets the model to predict better the maximum values. However, the improvement pointed out by the statistics to increase air temperature is appreciable.

CONCLUSIONS

In this work, we have studied the effect of temperature in ozone model simulations. Temperature in the period performed have shown discrepancies of 14 % (in ºC) between predicted temperature and measurement data. These under-predictions in diurnal temperature will affect to the emissions and to the kinetics of photochemical reactions. In the period studied, the contribution from the adjusted temperature to ozone concentrations improves considerably the validation in maximum daily values. It seems that the most important contribution from temperature falls in emissions and not in kinetics, but more cases must be analysed to be able to obtain a final conclusion.

ACKNOWLEGEMENTS

This work is funded by the Spanish Government through the project CGL2006-12474-C03-02 and is also supported by the Catalan Government.

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