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AIR QUALITY SIMULATIONS FOR NORTH AMERICA WITHIN AQMEII INITIATIVE

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Abstract: In the scope of the Air Quality Model Evaluation International Initiative (AQMEII), the air quality modelling system MM5-CAMx was applied to the North American (NA) domain for calendar year 2006. The simulation domain was defined according to the spatial resolution of the emission databases provided and the common grid required by AQMEII for ensemble analysis. A Lambert Conformal Projection grid of around 5500 km by 3580 km with 24x24 km^2 horizontal resolution was defined. Meteorological inputs were developed by the application of the meteorological model MM5, run for the whole year of 2006. A spatial and temporal analysis of results based on the 2D surface fields and time series for regional monitoring stations was performed for the main gaseous pollutants. A detailed statistical analysis and evaluation against observations was carried out, considering three different sub-domains over North America, in order to comprehend the differences between the East, West and Central part. Results have shown a good agreement between observed and modelled concentrations of O_3 (especially regarding peaks) and NO_2 and a weaker performance of the air quality model for CO and SO_2. However, the model tends to underestimate O3 and overestimate NO_2 and CO at night as a consequence of meteorology (weak vertical mixing due to underestimation of the Planetary Boundary Layer (PBL) height). This work intends to be a valuable contribution to the overall AQMEII exercise since it aims to evaluate the performance of individual models to be used in the ensemble approach for the areas of interest.

Key words: *Air quality model performance, Air quality assessment, gaseous pollutants, North America.*

INTRODUCTION

Chemical transport modelling has emerged as a useful tool for air quality management and assessment at local, regional and global scales. During the last decades, substantial developments have been made in air quality models, aiming at providing researchers, decision-makers, and the general population with more reliable information on the quality of the air, and hence, acting as valuable tools to support the test and implementation of emission control strategies, air quality regulations and air quality forecast for the protection of human health (European Directive 2008/50/EC, US-EPA - www.epa.gov/air/aqmportal).

Air quality modelling applications at regional scale have evolved from the study of photochemical air pollution episodes (short-term simulations for a specific area) (e.g. Byun et al., 2007) to multi-pollutant and multiyear integrated analysis (long-term simulations of both gaseous and particulate pollutants) (e.g. Hogrefe et al., 2011). Major attention has been given to ozone (O_3) and particulate matter (PM) (e.g. Vautard et al., 2007) due to their known impacts on air quality and human health and since high levels of these pollutants have been registered in monitoring stations, even surpassing the limits regulated. Nowadays, up-to-date air quality models, used worldwide, are able to reproduce the physical processes and chemical transformations of gaseous and particulate pollutants in the atmosphere. Notwithstanding, air quality predictions still have uncertainties associated (Chang and Hanna, 2004, Borrego et al., 2008) and thus, further investigation and innovative modelling applications are needed, namely in what concerns pollutants other than O_3 and PM, as precursors of photochemical pollution or other gases like carbon monoxide (CO) and sulphur dioxide (SO_2), which are also addressed by both North American and European legislation.

The Air Quality Model Evaluation International Initiative (AQMEII) aims to build a common strategy on model development and future research priorities, and establish methodologies for model evaluation to increase knowledge on processes and to support the use of models for policy development (Rao et al., 2011). AQMEII focuses on long-term air quality simulations, for the year 2006, for North America and Europe, performed by different models, and the evaluation of results. A web-based platform for model intercomparison and multi-model ensemble analysis, the ENSEMBLE system, was developed by the Joint Research Centre (Ispra, Italy) (Galmarini et al., 2004a,b) and updated for AQMEII purposes. This tool has been used to archive and analyse, both qualitatively and quantitatively, the meteorological and air quality modelling results obtained (http://ensemble2.jrc.ec.europa.eu/cgi-bin/ensemble/).

The University of Aveiro's team has performed and evaluated meteorological and air quality simulations for the whole year of 2006, for North America. This paper presents the exploitation of results obtained by the MM5-CAMx modelling system for the gaseous pollutants covered by air quality legislation and guidelines for the protection of human health, namely O_3, NO_2, SO_2 and CO. First, an evaluation of results against observations is carried out by the analysis of time series. Daily profiles, that reveal how the model reproduces, in average, the daily evolution of air concentrations, and box plots which indicate, through statistical parameters, the variability of the hourly concentrations observed and simulated, were also built. Hence, scatter plots, that demonstrate whether the model is underestimating or overestimating the observed values, and Taylor diagram which summarizes the overall global statistical model performance, were made. Another approach to evaluate model performance is to infer the ability of the model to simulate peak concentrations, which is particularly relevant for O_3, since episodic high O_3 concentrations may cause acute health problems. Thus, a few photochemical episodes have been analysed and discussed in terms of the model capability to capture the high concentrations registered. This extended abstract briefly describes the air quality modelling application and presents the main results obtained: the evaluation with monitoring data and the episodic analysis. Finally, some conclusions of the work performed are drawn and final remarks are given, highlighting the difficulties encountered and the importance of this contribution to the integrated modelling activity and to the advances in air quality modelling research.

AIR QUALITY MODELLING APPLICATION

The MM5-CAMx air quality modelling system, composed by the MM5 meteorological model and the CAMx chemical transport model, was applied to North America. The air quality simulation domain of 3600 by 5500 km^2 with 24 km spatial resolution and 15 vertical levels was set according to the AQMEII guidelines, in order to cover the areas where monitoring data was available for model evaluation and inter-comparison.

The Comprehensive Air quality Model with extensions CAMx (ENVIRON, 2010) is a 3D Eulerian photochemical dispersion model that allows for an integrated assessment of gaseous and particulate air pollution over many scales ranging from sub-urban to continental, by solving the pollutant continuity equation for each chemical species on a system of nested three-dimensional grids. In this work, CB05 chemical mechanism was chosen. Meteorological fields, emission data and initial and boundary conditions were established to be used as input data for the CAMx model.

Meteorological inputs were generated by the application of the MM5 (Pennsylvania State University/ National Center for Atmospheric Research) Mesoscale Meteorology Model (version 3.7) (Dudhia, 1993, Grell et al., 1994), for the year 2006. MM5 was initialized, and driven, every 6 hours, by the global NCEP-FNL 3D analysis data at 1º horizontal resolution. The simulation domain of approximately 7750 by 5050 km^2, with a horizontal grid spacing of 27 km and 23 terrain following vertical levels, was defined. For cumulus parameterization, the Grell scheme was selected, and MRF was used as planetary boundary layer (PBL) type. The hourly MM5 outputs were used as meteorological inputs to the air quality model.

The USEPA has provided gridded emission databases at 12 by 12 km^2 resolution for 2006 for the North American region. It includes anthropogenic area emissions, biogenic emissions, fire emissions daily estimates and point source emissions. More details on the NA emission database can be found in the AQMEII website (http://aqmeii.jrc.ec.europa.eu/aqmeii2.htm). The sets of hourly month-specific, high-resolution (12 km) NA gridded emissions files for 2006, speciated for the CB05 chemical mechanism, were converted into the CAMx model format and aggregated to the CAMx simulation domain of 24x24 km^2 grid resolution.

Initial and boundary conditions were taken from GOCART (Ginoux et al., 2001) and LMDZ-INCA (Hauglustaine et al., 2004). Boundary conditions for the species PSO_4, NA, PCL were taken from monthly averages of the GOCART global model simulations. For the species CO, PAN, O_3, results from the global model LMDz-INCA were used. This approach has been tested and used in previous air quality modelling applications (e.g. Monteiro et al., 2007).

RESULTS AND DISCUSSION

The meteorological and air quality modelling results obtained from the MM5-CAMx simulations were then post processed through a set of post processing tools to convert them into the common grid and format required by ENSEMBLE, a domain ranging from 130 to 59.5°W and from 23.5 to 58.5°N with a horizontal resolution of 0.25 by 0.25º. AS already referred, the present paper will only tackle gaseous pollutants, focusing, as previously referred, on the regulated ones.

The air quality modelling system performance was evaluated using direct comparison with surface monitoring values for the several gaseous pollutants and also assessed in terms of air pollution episodes for the most critical pollutants. All evaluation graphics presented were produced with the ENSEMBLEv5 tool, taking advantage of its features and considering its limitations: http://ensemble2.jrc.ec.europa.eu/cgi-bin/ensemble/.

Model evaluation with observations

The evaluation exercise was performed for the gas pollutants O_3, NO_2, CO, and SO_2 and included a spatial and a temporal analysis. Taking into account the coarse spatial model resolution (0.25ºx0.25º) only regional monitoring stations were considered for the MM5-CAMx air quality results. Besides that, and due to the large size of the North American continent, the whole NA domain was split into three sub-domains – D1, D2 and D3 – covering the western, central and eastern NA respectively. The total number of used regional monitoring stations depends on the considered pollutant: a total of 297 monitoring sites for O_3, 153 for SO_2, 99 for NO_2, and 27 rural stations for CO. In terms of spatial coverage, it is relevant to comment that a more highly dense network exists at the south east and west coast of NA, which is representative of the population density and urban areas spatial pattern.

Different types of plots were computed to better compare, qualitatively and quantitatively, the modelled results with the observations for each individual sub-domain, namely: (a) averaged time series over all monitoring sites and the entire period of simulation and the respective daily average profiles, (b) box plots that quantify in terms of median and percentile sectors the modelled results, (c) scatter plots that indicate existent under or overestimations, and finally (d) Taylor diagrams that summarize all the main statistical error parameters and classify in average the model global performance for each pollutant in each sub-domain. The results for O_3 are summarized in Figure 1, considering the three different sub-domains (D1, D2 and D3), and only the summer period (April- September 2006).

There are significant similarities in the averaged model results for the three sub-domains. Model skills are similar, in average, for all the three different regions: west, central and east. The statistical analysis exhibits, in average, a correlation factor around 0.6 for all the sub-domains and a normalized Standard Deviation of 1.0. The box and scatter plots evidence a slight underestimation of the O_3 concentration values, and the comparison of the daily profiles shows that this under-prediction mainly happens during the night period (the minimum values), which indicates that model is not reproducing correctly the ozone consumption at night due to reaction with NO emissions.

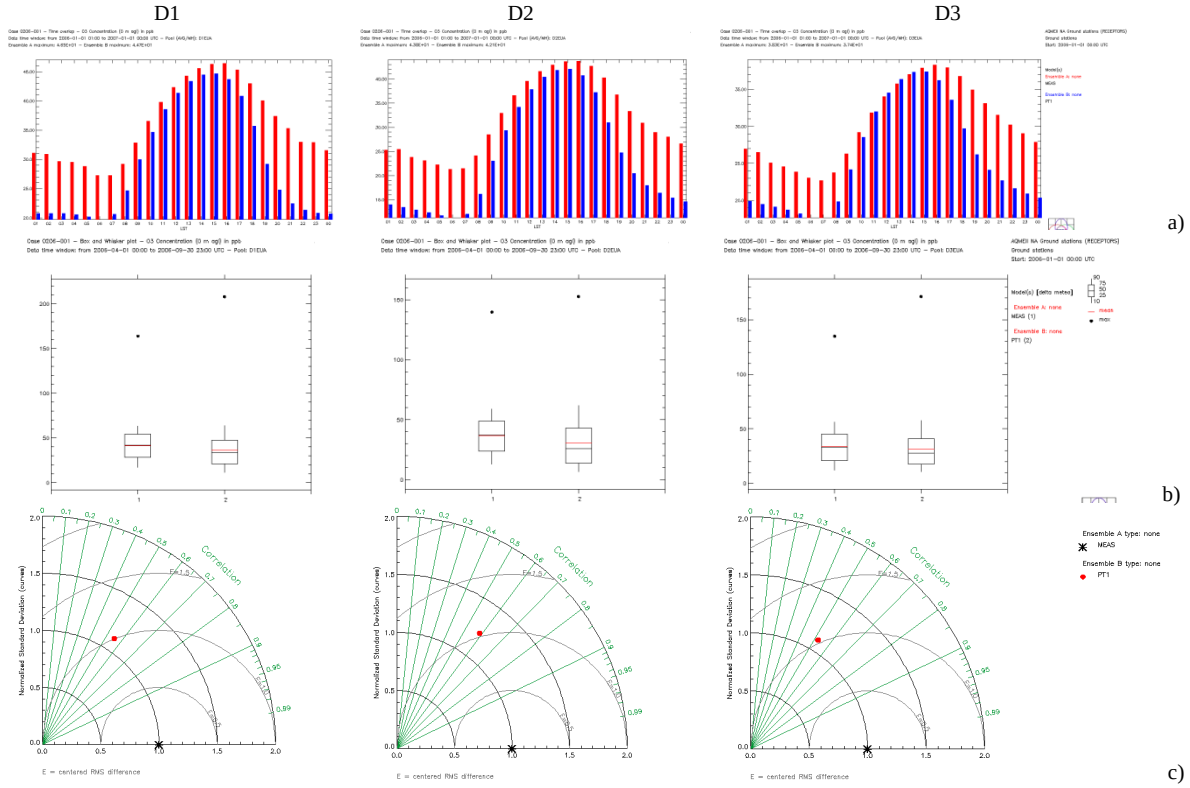

Figure 1. Evaluation of model performance for O_3, considering the summer period of 2006 (April-September): (a) daily profiles, (b) box plots and (c) Taylor diagram.

As discussed below, NO_2 and CO concentrations were over predicted at night suggesting that the meteorology provided insufficient dilution of surface emissions at night probably due to weak vertical mixing caused by an underestimation of the PBL height by the meteorological model, as reported by Vautard et al. (2001). Accordingly, ozone under prediction at night can be explained by ozone in the surface layer becoming decoupled from higher layers and suffering too much removal by deposition and reaction with surface emissions (NO and/or alkenes). In order to evaluate the model performance in terms of O_3 concentration peaks, soccer-goal plots were produced (not shown), considering only the range of O_3 observed values higher than 60 ppb and 120 ppb. For both analysed thresholds (60 and 120 ppb) the normalized mean bias and error are considered low (< 15%), which suggest a very good model prediction in terms of O_3 peak episodes. Particular high episodes will be analysed in further detail later.

Based on the analysis of results (not presented), the skill of the model performance is lower for NO_2. There is a consistent over-prediction of the NO_2 concentration values, during the entire daily profile and especially for the maximum values at night. This overestimation is constant for all the time series of the three different sub-domains. In average, the correlation factor is low (< 0.5) for all the domains, but the East domain exhibits the highest deviations (higher overestimation). As previously discussed for ozone, the main cause of NO_2 over prediction is the weak vertical mixing tending to trap surface emissions near the ground.

In contrast to O_3 and NO_2, very distinct SO_2 patterns exist among the three domains with D1 being different from D2 and D3 (Figure 2). For western NA (D1) a lower model skill was calculated, with an underestimation of the SO_2 concentrations, mainly related to the peaks of this pollutant (see time series and daily profile plots). This can suggest that the poor performance for SO_2 over the west domain may be related to the boundary layer and model resolution. High monitored SO_2 in the west results from plume impacts at monitors which occur during the day when plumes released aloft mix down to the surface. If the daytime PBL is too shallow, the plumes do not mix down to the surface, and this may explain why the model has the wrong diurnal profile. Also, model resolution of 24 km dilutes plume very quickly and the model is unable to capture the peak SO_2 events seen in the time series. In fact, a detailed analysis of the D1 domain's monitoring sites reveal that more than 20% of the available regional stations show an urban and/or industrial influence with particularly high SO_2 levels (> 100 ppb), which is not verified for the regional sites located in D2 and D3 domains.

The model results for CO (not shown) present similarities with the SO_2 plots: high underestimation over the western domain (D1) in opposition with a high overestimation on the other domains (D2 and D3). This overestimation indicates concentration of surface CO emissions near the ground due to lack of vertical mixing at day and night, because CO is overestimated during the entire day. In average, the global model skills are low for this pollutant (r<0.4 and NSD > 1.0). Nevertheless, the number of monitoring regional stations available for CO (27 for the whole NA domain) is small for this evaluation exercise, which can contribute to an inferior average skill of the model. The justification for this low model performance for both compounds can be related to the primary origin of them (both directly emitted to the atmosphere): errors on the emission inventory

(namely industrial and combustion sources) can lead to high errors on the model simulation. Besides that, both pollutants are involved in several chemistry transformations which also bring complexity to their modelling procedure.

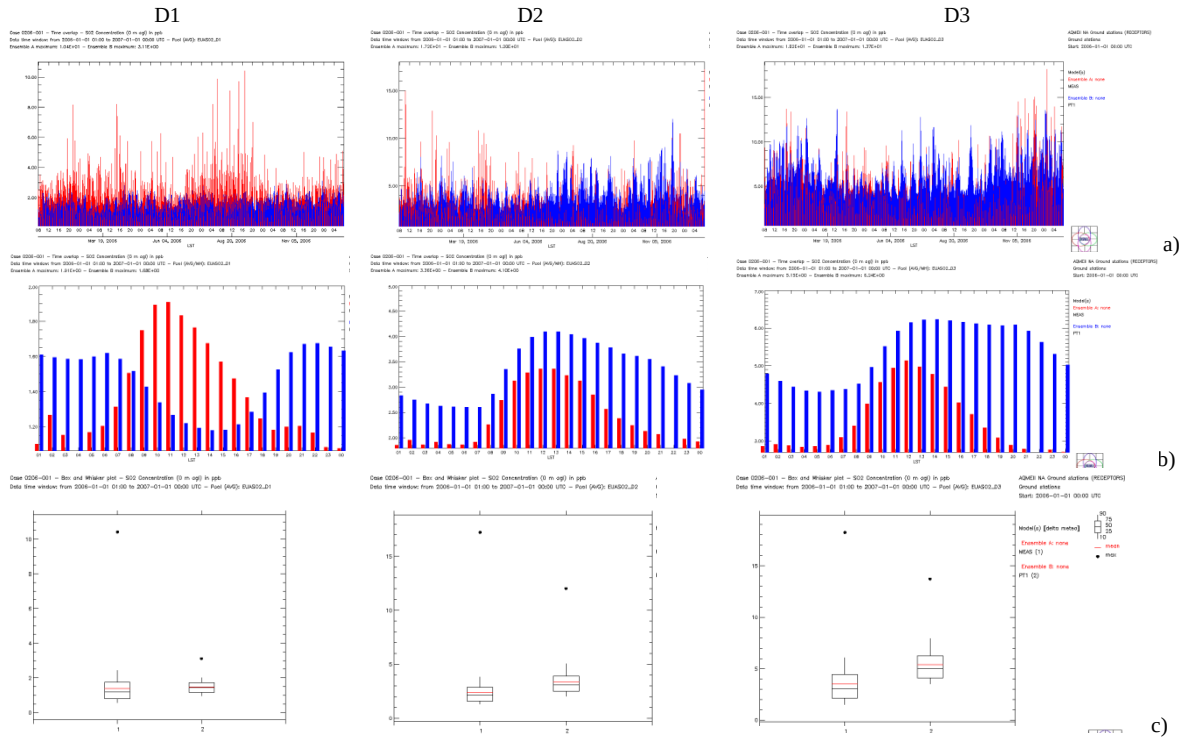

Figure 2. Evaluation of model performance for SO2, for the 2006 year: (a) time series, (b) daily profiles, and (c) box plots.

Analysis of episodes

One of the main goals of an air quality model is to predict/simulate the peak concentration of the air pollutants, in order to forecast the magnitude of air pollution episodes, responsible for the more serious and acute health problems. In this sense, different episodes of O_3 occurred during 2006 in NA are analyzed and discussed in detail. This was the only pollutant registering exceedances to the regulated limit value in 2006.

During the period 15-31 July, the highest O_3 concentration values were registered, particularly along two consecutive days (17-18th July) with hourly peaks larger than 120 ppb in a site located in the East (D3). The observed O_3 pattern is very well reproduced by the model, with the peaks being correctly simulated/predicted (Figure 3a). The same is not observed for NO_2, the model overestimated the monitoring values (Figure 3b). Nevertheless, during the specific days of the episode, the NO_2 concentrations were the same order of magnitude of the observed ones. By analysing the surface O_3 field for the 17th July at 16 UTC (not shown), it can be verified that this episode was primarily in the East, with O_3 concentrations above 100 ppb over a large area. In fact, this O_3 surface pattern is frequently observed during the summer in the Eastern US (Hegarty et al., 2007), and these episodes are more numerous when compared to the West (Cooper et al., 2005).

Another ozone episode occurred during the winter (November 2006), with very different characteristics from the latest. O_3 values above 400 ppb were monitored in one station located at 2800m altitude. Due to the absence of monitoring data before and after the peak, this episode peak may be a data error. A monitor calibration problem that was not screened out of the database should be the origin of the problem. In July 2006, also at high altitude station (1400 m), O_3 concentration values above 100 ppb were measured during several consecutive days. This is another successful example where the model was able to reproduce the high observed values of ozone during a long period of days. The daily pattern and photochemical cycle of O_3 is correctly simulated during the July month, together with the maximum concentrations of 110-120 ppb registered along the several days. Nevertheless, this example also illustrates and confirms the existence of an underestimation of the O_3 values during the night period.

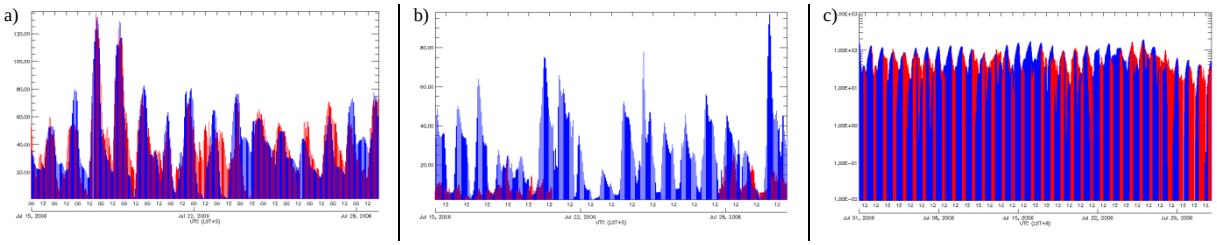

Figure 3. (a and b) Observed and modelled concentrations of O_3 and NO_2 over the period of 15-30th July, at the regional station AIRSUSNJ2CAM. (c) Observed and modelled O_3 concentrations over July, at the station AIRSUSCAICRE (red: observ, blue: modelled).

CONCLUSIONS

As one of the modelling tools participating in the AQMEII initiative, the MM5-CAMx modelling system was applied to North America, for the whole year of 2006, aiming at evaluating its ability to simulate the concentrations of gaseous atmospheric pollutants in the region.

The obtained results have shown that, regarding O_3, model performance is similar, in average, for all the three different studied regions: west, central and east of NA, with a correlation factor around 0.6. Modelling skills are lower for NO_2, exhibiting an overestimation in all sub-domains, especially for high levels of NO_2 registered at night. This is probably due to a weak vertical mixing, as a result of meteorology, and thus an insufficient dilution of surface emissions. The weakest performance of the MM5-CAMx modelling system was found for CO and SO_2, due to the observed peaks of this pollutant, especially in the west region of NA. The skill of the model to simulate peaks was also investigated. Only O_3 was addressed since it was the single pollutant registering concentrations above the limit value regulated. The analysis of three episodes has demonstrated the capacity of the model to predict the occurrence of O_3 episodes, which is important to prevent harmful effects of photochemical pollution on human health.

The results stress the importance of using emission inventories as detailed and accurate as possible and emphasize that a weak performance of the meteorological modelling has implications on the performance of the air quality model. Hence, this work highlights that additional research on the improvement of air quality modelling performance is still needed, as a multi-pollutant approach, covering not only the most studied pollutants ozone and particulate matter, but specially CO and SO_2 which are involved in both physical and chemical transformations not always well simulated by models.

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