ARAMIS a regional air quality model for air pollution management: evaluation and validation

María Rosa SOLER, Pedro GÁMEZ, Miriam OLID
Department of Astronomy and Meteorology, University of Barcelona, Martí i Franquès 1,
08028 Barcelona, Spain
rosa@am.ub.es

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Abstract
The aim of this research was to better understand the dynamics of air pollutants and to forecast the air quality over regional areas in order to develop emission abatement strategies for air pollution and adverse health effects. To accomplish this objective, we developed and applied a high resolution Eulerian system named ARAMIS (A Regional Air Quality Modelling Integrated System) over the north-east of Spain (Catalonia), where several pollutants exceed threshold values for the protection of human health. The results indicate that the model reproduced reasonably well observed concentrations, as statistical values fell within Environmental Protection Agency (EPA) recommendations and European (EU) regulations. Nevertheless, some hourly O_3 exceedances in summer and hourly peaks of NO_2 in winter were underestimated. Concerning PM10 concentrations less accurate model levels were obtained with a moderate trend towards underestimation during the day.
Key words: Air quality modelling; Model Evaluation; Pollutant exceedances; Regional scale.

Aramis un modelo regional para la gestión de la calidad del aire. Evaluación y validación

Resumen
El objetivo de este trabajo es desarrollar un modelo de predicción de la calidad del aire a escala regional con el fin de desarrollar estrategias de reducción de las emisiones y paliar los efectos adversos que los contaminantes ejercen sobre la salud. Para lograr este objetivo, se ha desarrollado y aplicado un modelo euleriano de alta resolución llamado ARAMIS (A Regional Air Quality Modelling Integrated System). ARAMIS se ha aplicado y validado en el noreste de España (Cataluña), donde periódicamente la concentración de algunos contaminantes superan los valores límite fijados para la protección de la salud humana. Los resultados de la validación indican que el modelo reproduce razonablemente bien las concentraciones observadas ya que los estadísticos caen dentro de los valores recomendados por la EPA y por la Comisión Europea, sin embargo, es importante destacar que el modelo subestima algunas superaciones de los valores máximos de la concentración de ozono en verano y concentraciones horarias máximas de dióxido de nitrógeno que se producen en invierno. En relación a la predicción de las concentraciones de partículas PM10, el modelo da peores resultados con una moderada tendencia a la subestimación durante el día.
Palabras clave: Modelización de la calidad del aire; Validación; Escala regional.

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1. Introduction

Air pollution is harmful to human health and the environment. The need to deliver cleaner air has been recognized for several decades, with action having been taken at national and international level and also through active participation. Thus, the European Commission (EC) and the U.S. Environmental Agency (US-EPA) have promoted several actions focused on establishing minimum quality standards for ambient air and tackling the problems of exceedance of several pollutants at ground level. Nevertheless, despite the considerable efforts that have been made to reduce pollutant emissions in recent decades, there are still only modest signs of reduced concentration levels for some pollutants such as ozone, nitrogen oxides and particulate matter. The nonlinearity of atmospheric chemistry, together with the multiplicity of diffuse pollutant sources, including biogenic sources which can amplify concentrations, may partly explain this behavior (Vautard et al., 2007).

As with air motion, air pollution occurs on a wide range of different spatial scales, from tiny eddies measuring centimeters or less in size to large air mass movements of continental dimensions, leading to adverse local, regional and trans-boundary environmental and health impacts. Nevertheless, local and regional emission contributions to air quality will remain dominant over the next decade, and countries and regions must continue their efforts to reduce primary pollutant emissions in a cost-effective manner.

At the national and regional level, public administrations must promote or develop i) the technical knowledge and tools necessary for a long-term strategy and ii) an integrated policy on emission regulation in order to prevent the significant negative effects of air pollution (Cuvelier et al., 2007). One effective way to conduct a detailed evaluation of the impacts of air quality policies at the regional and local scale is through measurements, but the most common procedure is to use numerical models as they can provide a more complete deterministic description of the air quality problem, including analysis of factors and causes such as emission sources and meteorological processes (Daly and Zannetti, 2007). In addition, air quality models are also becoming increasingly valuable tools for epidemiologists; many epidemiological studies have documented poor air quality as a risk factor for a variety of adverse human health outcomes.

Against this background, we developed a new air quality modelling system named ARAMIS (A Regional Air Quality Modelling Integrated System), funded by the Catalonian Government, as a tool to assess and understand the dynamics of air pollutants, to forecast air quality and to develop emission abatement strategies for air pollution and adverse health effects. ARAMIS is a high resolution system which integrates the Weather Research and Forecasting model (WRF) as a meteorological

model, the High Resolution Emission Model (HIREM) and the Models-3 Community Multiscale Air Quality Modelling System (Models-3/CMAQ) as a photochemical model. ARAMIS was developed to simulate air quality over north-east Spain at regional and local scales, since the southern Mediterranean regions as Catalonia and in particular the metropolitan area of Barcelona (BMA) (Figure 1) presents air pollution episodes. These areas exceed the air quality targets set by the European Union, specifically with regard to NO_2, PM10 and ozone (Jiménez et al., 2006a; Arasa et al., 2010; 2012). Several studies have investigated air quality concerns over Spain and selected areas (Jiménez et al., 2006a; San José et al., 2007; Jiménez-Guerrero et al., 2008; Vivanco et al., 2009; Baldasano et al., 2011) using numerical modelling, but they were conducted using different emission models. ARAMIS uses a 3 km X 3 km cell resolution to simulate the Catalonian area and 1x1 km to simulate BMA.

In this paper, we describe this new air quality system, and we evaluate its performance through comparisons with ground-based measurements obtained from the air pollution monitoring network belonging to the Environmental Department of Catalonia Government (http://www20.gencat.cat/portal/site/mediambient/). The model dynamics are assessed with the corresponding statistics, which are then compared to model performance goals and criteria.

2. Modelling approach

This section describes the models that comprise the ARAMIS modelling system

2.1 Meteorological model

Meteorological numerical simulations were performed using the Weather Research and Forecasting model (WRF-ARW), version 3.1.1 (Skamarock et al., 2008). The WRF-ARW model was configured using four nested domains with grids of 27, 9, 3 and 1 km (Figure 1a). Domains are run in one-way nesting and the first 12 h are treated as spin-up. Fields are saved every 5 min. The outermost domain, D1, covered 68 x 44 grid cells, while D2 covered 69 x 69 x 70 cells, D3 - corresponding to Catalonia - covered 93 x 93 grid cells and D4 - corresponding to the BMA - covered 93 x 114 grid cells. Initial and boundary conditions are taken for the European Centre for Medium-Range Weather Forecast global model (ECMWF) with a 0.5º x 0.5º resolution, and the boundary conditions are forced every 6 h. The physics package included the Eta surface layer scheme, which is based on Monin-Obukhov similarity theory (Monin and Obukhov, 1954), the YSU boundary layer scheme (Hong and Kim, 2008), the rapid radiative transfer model (RRTM) longwave radiation scheme (Mlawer *et al*., 1997), the Dudhia shortwave radiation scheme (Dudhia, 1989), the new Thompson microphysical scheme (Thompson *et al*., 2008), the Kain-Fritsch cumulus scheme and the Noah Land Surface scheme (Chen and Dudhia, 2001). Vertical resolution included 32 levels, 20 below (approximately) 1500 m, with the

first level at approximately 15 m and the domain top at about 100 hPa. Domains are run in one way nesting and the temporal length of meteorological simulations was 72 h, taking the first 24 h as spin-up time to minimise the effects of initial conditions. Fields are saved every 5 min and one-hour output time frequency was used in all domains and models.

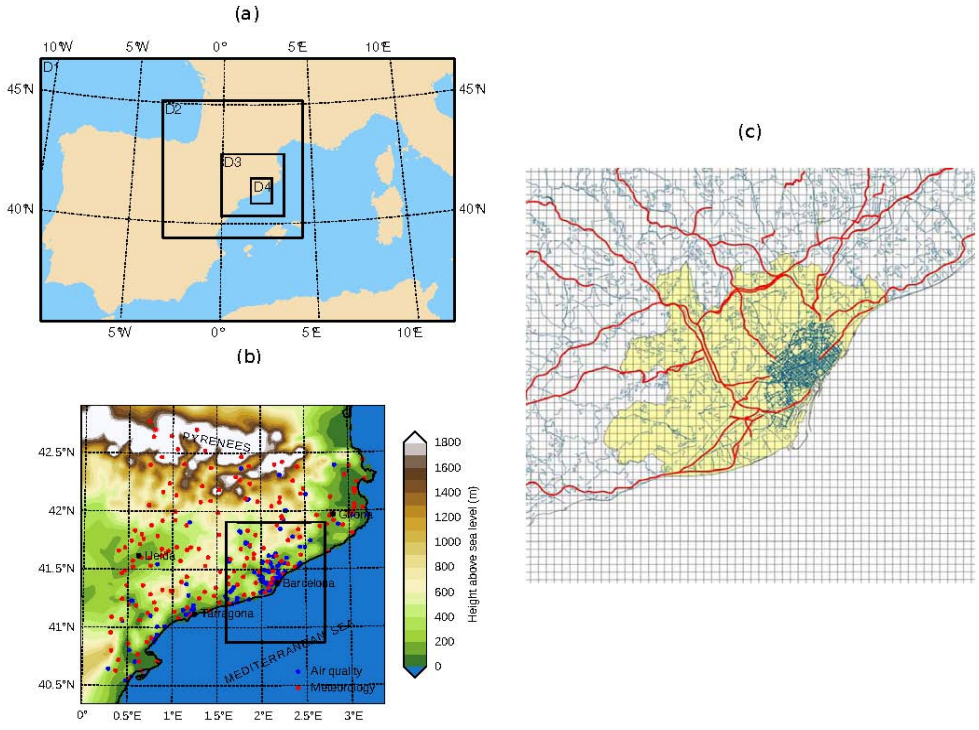

Figure 1. Model domains for WRF and ARAMIS model simulations (a). topographical features of Catalonia (domain D3) and location of measurement stations (b); black dots correspond to meteorological surface stations, while red, green and blue dots correspond to surface air quality stations (red measures both O_3 and NO_2 concentrations, green only NO_2 and blue PM10. (c) Domain D4 including the BMA (in yellow), the road network in Barcelona and the main roads and highways considered in domain D4 and implemented in HIREM emission model.

2.2 The HIREM emission model

HIREM is the new and completely updated version of the MNEQA model (Numerical Emission Model for Air Quality) (Ortega et al., 2009; Arasa et al., 2012) developed by our research group. HIREM includes emissions from natural sources, such as dust particles from erosion and hydrocarbons emitted by vegetation, as well as several anthropogenic sources, such as domestic and commercial fossil fuel uses,

domestic and commercial solvent uses, power generation plants, industrial installations and road transport. Emission methodologies are often implemented in HIREM due to the ease with which they can be revised and updated; it is also easy to combine emissions from different sources in order to study the contribution of each sector or to perform a sensitivity analysis.

The air pollutants classified as primary by HIREM are nitrogen oxide (NO_x), methane (CH_4) and non-methane volatile organic compounds (NMVOC), carbon monoxide (CO), carbon dioxide (CO_2), nitrous oxide (N_2O), sulphur dioxide (SO_2), total suspended particles (TSP) and particulate matter (PM10 and PM2.5).

As nested domains are commonly applied to air quality modelling HIREM methodology differed from one domain to another. For larger domains (D1 and D2), HIREM uses a top-down method, which incorporates pollutant emissions from EMEP/CORINAIR (2013) into the model. The top-down method consists of a disaggregation model based on Corine Land Class (CLC) 2009 soil uses, with 250-m resolution, coupled with different statistical functions, including socio-economic variables (Maes et al., 2009).

For the smaller domains such as D3 and D4, HIREM uses a bottom-up technique to calculate pollutant emissions. This approach involves working on each type of source in a particular way using local information. HIREM incorporates the industrial emissions inventory from the Catalonian Department of the Environment and residential consumption emissions which are calculated by emission factors using different residential parameters and population distributions. Natural emissions are computed in HIREM using different parameterizations (Marticorena and Bergametti, 1995; Vautard et al., 2005), whilst biogenic emissions are incorporated via the method described by Guenther et al. (1994).

One of the most important modules within the HIREM emission model is the road traffic emissions one, which includes hot, cold and evaporative emissions for some pollutants (CO, NO_x and VOC's) and also estimates particulate matter emissions from brake abrasion, tyre wear and road surface. This module uses EEA-EMEP/CORINAIR methodology based on a bottom-up approach and takes 72 vehicle categories represented by one emission factor depending on traffic speed. Principal and minor roads in Catalonia containing information about the Daily Average Traffic (DAT), length and mean velocity was provided by the Department of the Environment. In addition, the Catalonia's vehicular fleet and the hourly number of vehicles per day in 2011 measured at 72 observation points located along the principal roads were also provided. With this information we calculated different temporal profiles (monthly, daily and hourly) for different types of roads (motorway or road).

The methodology used to estimate urban emissions in Barcelona and the medium-sized cities of Catalonia was similar as mentioned above using the corresponding road network and the vehicular urban fleet provided by the Barcelona City Council and the Department of the Environment.

2.3 The photochemical model

The photochemical model used in this study to simulate pollutant dispersion was the U.S. Environmental Protection Agency (EPA) model-3/CMAQ model (Byun and Ching, 1999). From the CMAQ v4.7.1 chemical mechanisms, we use the CB-05 and associated EBI solver (Yarwood et al., 2005) including gas-phase reactions involving nitrogen pentoxide (N_2O_5) and water (H_2O), and it removes obsolete mechanism combinations in gas, aerosol (solid or liquid) and aqueous phases. In addition, we use the aerosol module, AERO4 (Bhave et al., 2005; Shankar et al., 2005), with a preliminary treatment of sea salt emissions and chemistry. For treating clouds in the model, we used the asymmetric convective module, ACM (Pleim and Chang, 1992). Boundary conditions and initial values for domain D1 came from a vertical profile supplied by CMAQ itself, whilst boundary and initial conditions for domains D2 and D3 were supplied by domain D1 and D2, respectively. The model was executed for 72 h, taking the first 24 h as spin-up time to minimize the effects of initial conditions.

3. Air quality modelling system evaluation

Models results were evaluated through comparison with a ground based measurement stations which are considered suitable to use if their data availability is at list higher or equal to 75%.

3.1. Evaluation of the meteorological model performance

Model results were evaluated from February to September 2013 by comparison with data from a set of 172 surface meteorological stations belonging to the Catalonian Meteorological Service and displayed in Figure 1b by black squares. The evaluation included several variables showed in Table 1. Temperature and humidity measured and modelled correspond to 1.5 m and 2 m a.g.l. respectively, while wind speed and direction measured and modelled correspond at 10 m a.g.l. The statistics selected for the evaluation were the root mean square error (RMSE), the mean absolute gross error (MAGE), the mean bias (MB) and the index of agreement (IOA). In order to establish a criterion for evaluating the performance of the meteorological model, we adopted the benchmarks suggested by Emery and Tai (2001) and Tesche et al., (2002) (see Table 1). Wind statistics and wind direction were calculated for wind speeds higher than 0.5 m s^{-1}, as wind direction is not reliable for lower speeds following the method described in Lee and Fernando, (2004); Soler et al., (2011).

Table 1. Statistics corresponding to WRF model evaluation for the whole studied period for Catalonian domain and quantitative performance statistics for meteorological model results

Meteorological variable	Statistic	24-h Forecast	48-h Forecast	Benchmark
Wind speed	RMSE (m s^{-1})	3.03	3.03	≤ 2
	MB (m s^{-1})	1.73	1.68	$\leq \pm 0.5$
	IOA	0.64	0.63	≥ 0.6
Wind direction	MAGE (deg)	47.28	49.14	≤ 30
	MB (deg)	2.42	2.77	$\leq \pm 10$
Temperature	MAGE (K)	1.73	1.79	≤ 2
	MB (K)	0.32	0.35	$\leq \pm 0.5$
	IOA	0.98	0.98	≥ 0.8
Specific humidity	MAGE (g Kg^{-1})	0.97	1.02	≤ 2
	MB (g Kg^{-1})	-0.12	-0.18	$\leq \pm 1$
	IOA	0.96	0.95	≥ 0.6

Meteorological prediction accuracy is also presented in Table 1. The results show that statistical values corresponding to specific humidity and temperature fell within recommendations; however, wind speed was overestimated by the model, whilst wind direction statistics showed a greater dispersion as MAGE values exceeded the benchmark value of 30º. Examining these results in greater depth, we calculated hourly evolution of the statistics corresponding to all variables evaluated. Results are presented in Figure 2. For wind speed (Figure 2a), IOA values presented very low diurnal variation, with a constant value close to 0.6, whereas MB and RMSE showed lower values during the day and higher values at night. The reason of this behaviour could be attributed to the meteorology of this area, usually characterized in late spring, summer and early autumn by antyciclonic situations with a slight pressure gradient, favouring the development of mesoscale circulations such as a sea-breeze regime on the coast and mountain winds inland. The wind speed associated with these circulation patterns was reproduced quite well by the model, although it tended to overestimate wind speed, albeit to a lesser extent during the day, with MB values between 1.0 and 2.0 m s^{-1} (Figure 2a). The hourly evolution of MAGE and MB values for wind direction is shown in Figure 2b. During diurnal hours, MAGE values decreased to values of 40 degrees, higher than the corresponding benchmark (30º); however, at night, the model did not reproduce very weak winds accurately, with MAGE values increasing to 50-60 degrees. These systematic biases in wind velocity and direction in complex topography areas have been related, at least in part, to the model's relatively smooth topography, and therefore, the effects produced by the topographic features are not well resolved in the model, producing an additional drag to that generated by vegetation. If their effects are not considered, such as is the case for WRF, it can lead to an overestimation of the wind speed and high biases in wind directions (Jiménez et al., 2010) In addition, there are other reasons responsible for the model biases that must be mentioned as i) potential errors at the synoptic scale occurring during the initialization may introduce important deviations from the

observed wind behaviour, since uncertainties at the synoptic scale can propagate to the regional scale being responsible for large misrepresentations (Jimenez et al, 2010); ii) random turbulent processes that cannot be simulated by models or to sub-grid variations in terrain and land use.

For air temperature (Figure 2c), daily evolutions are given for MAGE, MB and IOA. For most of the period studied, the MAGE value was lower than 2 degrees, whilst MB was between 1 and -1 degrees and IOA was close to 1 for the entire period. These results highlight the tendency of the WRF model to underestimate the air temperature at a height of 1.5 m during diurnal hours (Figure 2c). At night, the model slightly overestimated the temperature, resulting in a positive MB value for the entire period. This behaviour can be related to the pattern found by wind velocity, their overestimation during day time reduce air surface heating while during night time reduce air surface cooling. Lastly, Figure 2d shows hourly evolutions of MB, MAGE and IOA for specific humidity. All statistics showed scarce diurnal variation, with values of IOA close to 1 whilst MAGE and MB were close to 1 and 0.1 g kg^{-1}, respectively. This was the best variable simulated by the meteorological model.

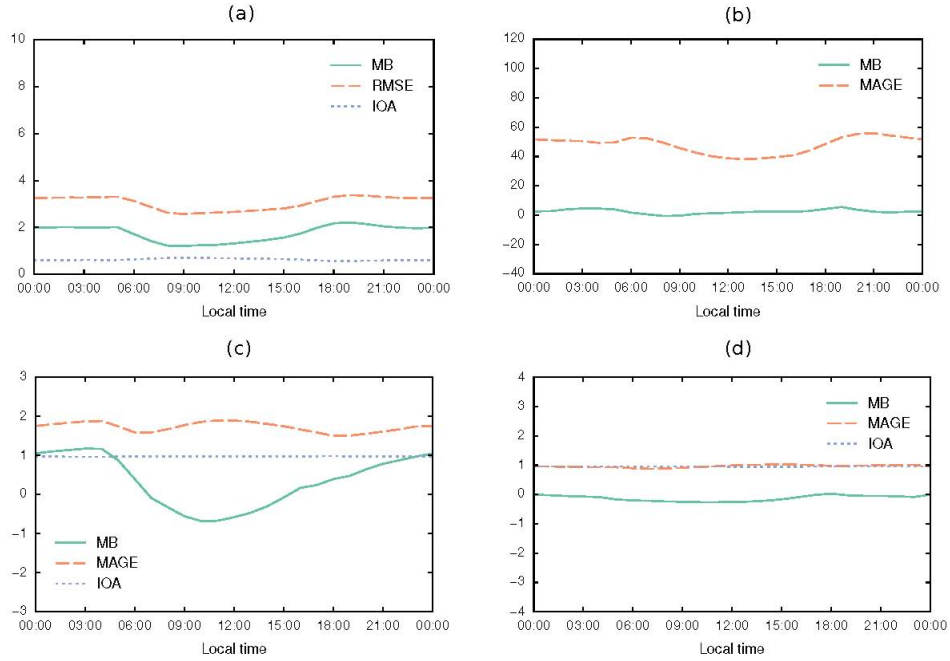

Figure 2. Time evolution of hourly statistics corresponding to: (a) wind speed measured at 10m (a.g.l.), (b) wind direction measured at 10m (a.g.l.), (c) air temperature at 1.5m (a.g.l.), (d) specific humidity at 1.5m (a.g.l.) over all measurement stations located in domain D3 (Catalonia).

The results for meteorological model performance agree with several previous studies of meteorological applications for air quality modelling, especially those based on the same area of study (Jiménez et al., 2006a; Borge et al., 2008), where classical statistics for surface fields have been reported. Meteorological predictions over complex terrain require high horizontal and vertical resolution for resolving complex mesoscale circulation patterns, especially at night where turbulent mixing is greatly reduced or even completely suppressed, or becomes intermittent (Cuxart et al., 2000; Mahrt and Vickers, 2002; Poulos et al., 2002). In addition, this stable stratification in a non-uniform terrain induces local circulations, such as drainage flows (Soler et al., 2003), and leads to several phenomena such as gravity waves, density currents (Terradellas et al., 2005; Udina et al., 2013; Ferreres et al., 2013; Soler et al., 2014), intrusions and meandering, with the frequent presence of low-level jets (Conangla and Cuxart, 2006). The misrepresentation of these effects and the local physiographical features can lead to incorrect estimations of several degrees in the temperature, in the moisture levels and errors in the wind speed and wind direction (Bravo et al., 2008).

3.2. Evaluation of the photochemical model performance

The air quality model is evaluated for the same period as before and the assessment is focused on hourly O_3, NO_2 forecast and hourly and daily PM10 predictions. Data was provided by a network of air quality surface stations: 48 measuring ozone, 67 NO_2 and 67 PM10 (31 automatic and 36 manual providing hourly and daily data respectively). All stations are displayed in Figure 1b represented by different symbols, red dots correspond to stations measuring both O_3 and NO_2 concentrations, green only NO_2 and blue PM10.

Different statistical indicators have been identified as being the most appropriate for evaluating an air quality modelling system and quantifying its performance. The most common are those proposed and recommended by the U.S.-EPA (1991, 2007), Boylan and Russell (2006) and Denby et al., (2010). In addition to those previously mentioned, we included the maximum relative directive error (MRDE) (European Commission, 2008), which is the maximum value of the RDE found at 90% of the available stations (Denby et al., 2010), the mean normalized bias error (MNBE), the mean normalized gross error (MNGE), the mean fractional bias (MFB), and the mean fractional error (MFE) (Yu et al., 2006). These two last metrics have been found to be statistically robust measures, as they are symmetric and avoid any inflation that might be caused by low values of the observed quantities. For these statistics, it is necessary to define the performance criteria goals (the level of accuracy that is considered to be close to the best a model can be expected to achieve) and the performance criteria (the level of accuracy that is considered to be acceptable for modelling applications). The performance criteria and the performance criteria goal in brackets used in this study are presented in Table 2.

Table 2. The performance criteria and the performance criteria goal in brackets for O_3, NO_2 and PM10

	O_3	NO_2	PM_{10}
MFB	≤ ±30% (15%)	≤ ±30%	≤ ±60% (30%)
MFE	≤ 45% (30%)	—	≤ 75% (50%)
MNBE	≤ ±15%	≤ ±15%	≤ ±15%
MNGE	≤ 35%	≤ 35%	≤ 35%
MRDE	≤ 50%	≤ 50%	≤ 50%
UPA	≤ ±20%	≤ ±20%	≤ ±20%

3.2.1. Ozone

The evaluation comprised a total of 48 stations measuring ozone. Statistical parameters were applied to both 1-h maximum ozone surface concentrations and maximum daily 8-h mean ozone surface concentrations (henceforth 1-h and 8-h maximum ozone concentrations). Observation–prediction pairs were often excluded from the analysis if the observed concentration was below a certain cut-off point (Russell and Dennis, 2000; Baldasano et al., 2011); since cut-off levels vary from study to study, we set a level of 40 μg m^{-3}, thus removing the influence of low concentrations such as night-time values.

The statistics compiled in Table 3 corresponding to 1-h and 8-h maximum ozone concentrations show that all statistics met the recommended performance criteria and in some cases the performance criteria goal, therefore ARAMIS model can be used for ozone modelling applications.

Table 3. Statistics obtained with ARAMIS over domain D3 at the Catalonian stations corresponding to 1-h and 8-h maximum ozone concentrations.

Filter: 40 μg/m³	1-h maximum ozone concentrations		8-h maximum ozone concentrations	
	24-h Forecast	48-h Forecast	24-h Forecast	48-h forecast
IOA	0.66	0.66	0.62	0.63
MAGE (μg/m³)	16.90	17.15	15.14	15.51
MB (μg/m³)	-3.33	1.10	-2.28	1.82
RMSE (μg/m³)	22.23	22.67	19.43	19.84
MNBE (%)	0.14	4.50	1.17	5.62
MNGE (%)	16.81	17.59	16.74	17.74
MRDE (%)	–	–	26.70	23.64
MFB (%)	-2.21	1.96	-1.11	3.09
MFE(%)	16.65	16.61	16.45	16.64
UPA (%)	-0.17	16.40	-5.66	6.33

Beside the statistical analysis, Figure 3a displays the time evolution of average hourly ozone concentrations provided by the ARAMIS model at the measurement

stations, where it can be seen that the model overestimated the ozone concentration during the night and slightly underestimated it during the day. This diurnal underestimation could be caused by: (1) sea-breeze and land-breeze circulations that are difficult to reproduce, associated with the important role of circulation patterns in photochemical simulations (Pirovano et al., 2007), (2) a tendency to underestimate ozone precursors (nitrogen oxides, carbon monoxide and volatile organic compounds) in air quality modelling systems (Russell and Dennis, 2000) and (3) uncertainty in the emissions coming from the HIREM model and in their temporal distribution. Three main sources of nocturnal overestimation might be: (1) the model does not represent nocturnal physic-chemical processes accurately enough (Jiménez et al., 2006b), (2) the HIREM emission model may not calculate night-time emissions properly, and (3) meteorological parameters, such as wind speed, wind direction and vertical mixing, are not well reproduced by the model when the synoptic forcing is weak and the ambient winds are light and variable (Bravo et al., 2008; Schürmann et al., 2009).

Time series of daily maximum 1-h and 8-h maximum ozone concentrations were presented in Figures 3b and 3c, corresponding only to today's concentration forecast as the time series corresponding to tomorrow's forecast was similar. Results show that although the mean variability and the peaks were quite well reproduced in the model, daily peaks in summer, mainly in July, were underestimated. Modelled and measured discontinuities observed in these figures are due to problems in model execution or in data availability respectively.

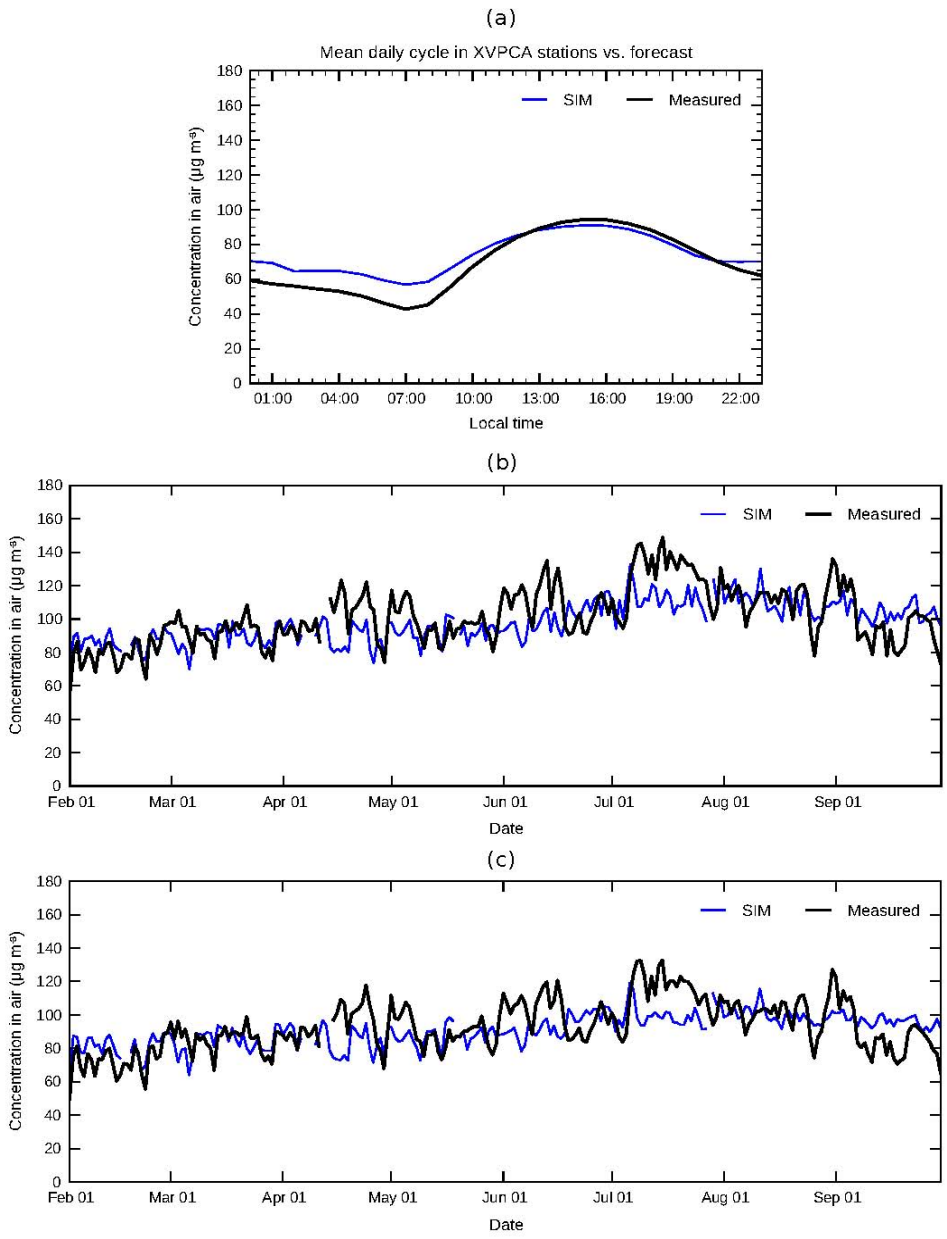

Figure 3. Modelled (blue line) and measured (black line of: (a) time evolution of averaged hourly ozone concentrations, (b) time series of daily maximum (1-h) and (c) 8-h maximum O_3 concentrations over all measurements stations located in domain D3 (Catalonia).

To end this subsection, we display the spatial distribution patterns of the average modelled 1-h and 8-h maximum ozone concentrations in Figures 4a and 4b, which are labelled in a different way according to their maximum hourly limit value or according to their 8-h maximum values. Results show that the highest mean concentrations were located inland and also over the Mediterranean Sea. The first pattern was due to the development of a sea-breeze front affecting the coastal meteorology on a daily basis, leading to low-level convergence of air pollutants over the land during the daytime, and divergence of air pollutants over the land during the night time. Therefore, the overall effect of land/ sea breeze is to enhance the air stagnant in the vertical over the coastal land (Wang et al., 2013). In Catalonia area sea breeze transports ozone and precursors (mainly NO_x) from the BMA and several industries and motorways located on the coast to inland areas (Soler et al., 2011). The strength of this on-shore flow and the complex topography of the north-west Mediterranean coast produce several pollutant injections due to topographic forcing. As the sea-breeze front advances inland and reaches the mountain ranges located nearly parallel to the coast, topographic injections occur at different altitudes (Soler et al., 2011). A number of studies have shown that during summertime, layering and accumulation of pollutants such as ozone and aerosols take place throughout eastern Spain (Pérez et al., 2004). This situation is intensified during persistent subsidence situations over the region (Millan, 2000; Jiménez et al., 2006b), intense solar radiation and high air temperatures (Ortega et al., 2011). Concerning to the second pattern, the main processes that could contribute to an increase in ozone concentration over the Mediterranean Sea include local generation due to the high amount of nitrogen oxides emitted from sea transport (Baldasano et al., 2011), low wet and dry ozone deposition, persistent subsidence over the region and photochemically aged air masses with high concentrations of O_3 and poor in photochemical precursors. These air masses are transported towards the Mediterranean area, where they sink within the anticyclone located over the Western Mediterranean Sea (Gangoiti et al., 2001).

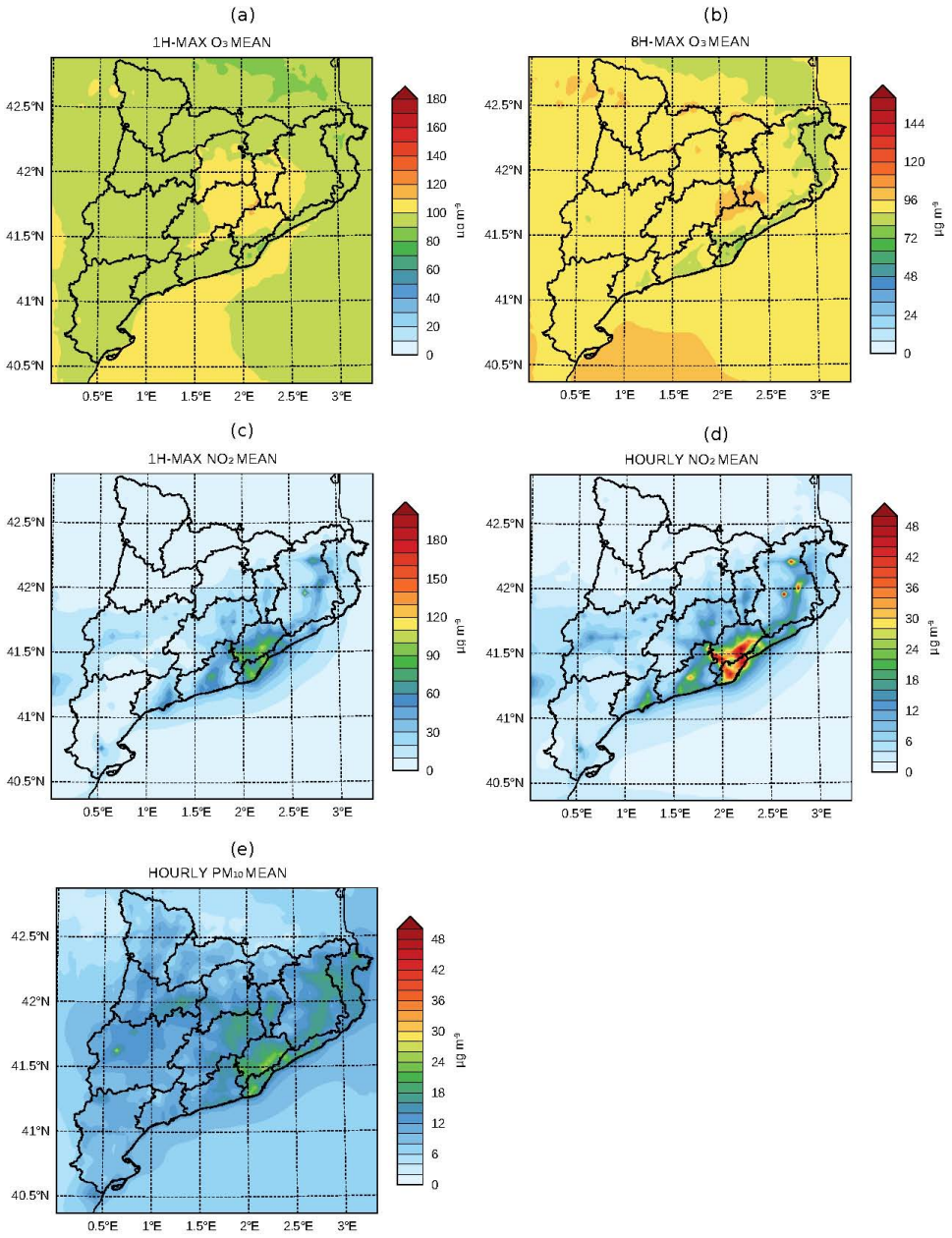

Figure 4. Averaged simulated concentrations from February to September 2013, over Catalonian domain (D3) of : (a) maximum 1-h O_3 concentrations, (b) 8-h maximum O_3 concentrations, (c) maximum 1-h NO_2 concentrations, (d) hourly NO_2 mean concentrations, (e) hourly PM10 mean concentrations.

3.2.2 Nitrogen Dioxide

The evaluation comprises 67 stations measuring NO_2 over the Catalonia area, and 22 of them are located in the BMA. In years 2012 and 2013 nine and seven stations respectively did not meet the annual threshold as they exceeded the annual limit of 40 $\mu g\ m^{-3}$, therefore, the evaluation for NO_2 concentrations was conducted for domains D3 and D4, corresponding to Catalonia and the BMA respectively. In the statistical evaluation, parameters were applied to 1 hour maximum NO_2 surface concentrations. The statistics compiled in Table 4 shows that, as in the case of ozone evaluation, they met the recommended performance criteria, although less accurate values are obtained.

Table 4. . Statistics obtained with ARAMIS for NO_2 over domains D3 and D4 and for PM10 over domain D3

	1-h maximum NO_2 concentrations (D3)		1-h maximum NO_2 concentrations (D4)		Averaged daily PM10 concentrations (D3).	
	24-h forecast	24-h forecast	24-h forecast	48-h forecast	24 h-forecast	48-h forecast
IOA	0.73	0,. 2	0.71	0.64	0.52	0.54
MAGE (μg/m³)	21.82	22.86	23.24	26.46	7.31	7.18
MB (μg/m³)	1.25	2.24	5.32	0.15	-2.25	-1.93
RMSE (μg/m³)	27.99	29.23	29.48	35.78	9,12	8.98
MNBE (%)	9.83	11.66	9.93	4.45	-2.17	-0.53
MNGE (%)	39.62	41.79	33.02	34.70	39.73	39.31
MRDE (%)	36.66	40.44	20.14	26.90	49.34	50.01
MFB (%)	-1.82	-0.99	1.06	-6.0	-13.99	-12.02
MFE (%)	36.89	38.20	28.25	32.09	40.86	39.82
UPA (%)	-17.85	-15.77	6.94	-8.15	-40.03	-32.90

For the D3 domain, the time evolution of average hourly NO_2 concentrations provided by the ARAMIS model at the measurement stations is given in Figure 5a.

The model gave quite accurate reproduction of the hourly variability, although under estimated NO_2 concentrations during the day and overestimated them at night. This behaviour was probably related to an underestimation of the emission inventory in addition to other meteorological and chemical factors. Besides, the overestimation of modelled NO_2 concentrations could be due to two main reasons: the end of the traffic activity period was not well forecasted by HIREM emission model, and probably the boundary layer height estimated by WRF model was too low. This last result is often observed in air quality models and could be due to the uncertainty of mesoscale modelling as regards accurately estimating the unstable to stable boundary layer transition (including length of time and amplitude).

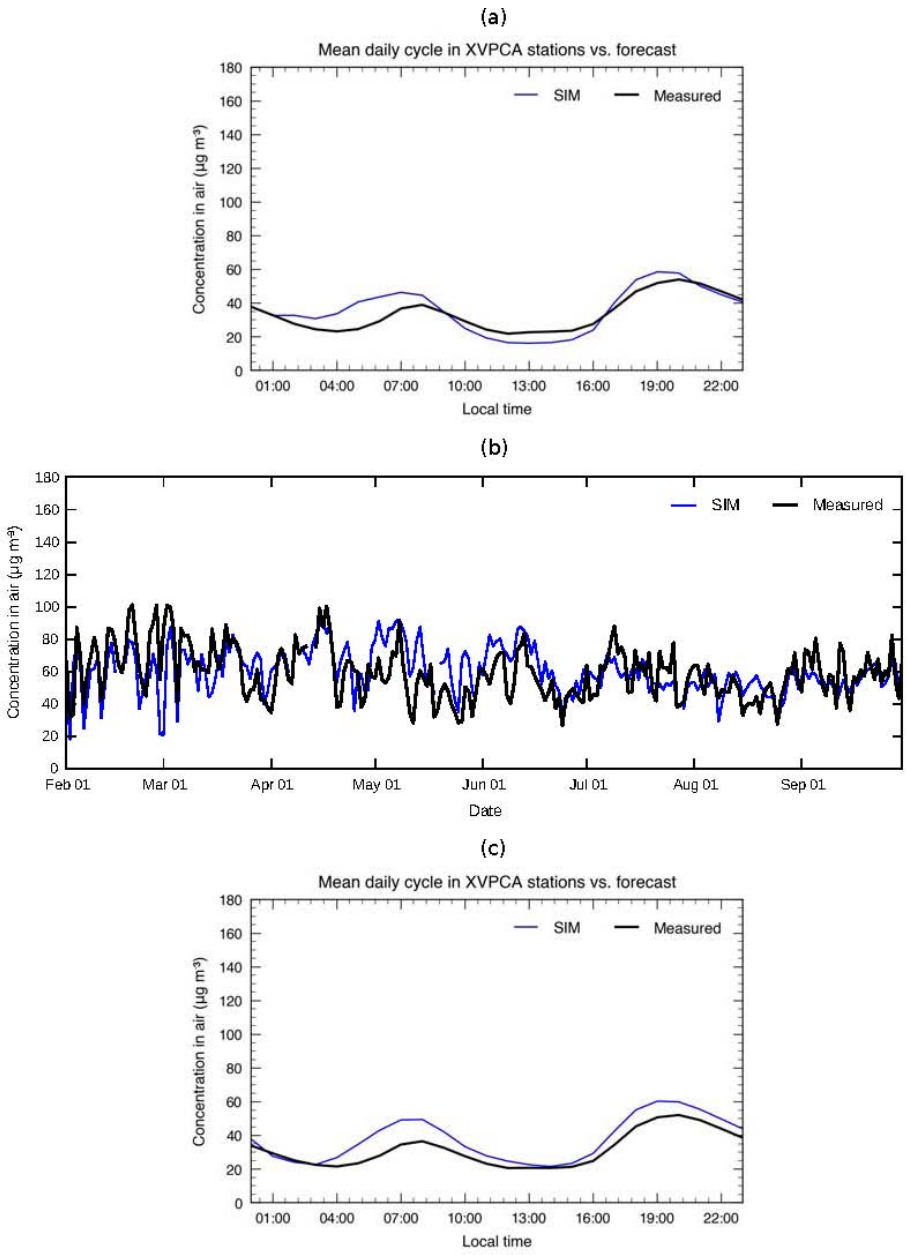

Figure 5. Modelled (blue line) and measured (black line of: (a) time evolution of averaged hourly NO_2 concentrations over all measurements stations located in domain D3 (Catalonia), (b) time series of daily (1-h) maximum NO_2 concentrations, (c) time evolution of average hourly NO_2 concentrations over all measurements stations located in domain D4 (BMA).

Figure 5b displays time series of today's daily maximum 1-h NO_2 concentrations forecasts, as the corresponding values for tomorrow's was similar. Results show that the model generally captured daily 1-h maximum concentration variations well; however, daily peaks with higher values in winter and spring related to air pollution events were underestimated. Usually, this maximum values were associated to high-pressure conditions favouring the development of local re-circulations associated with stagnant air masses.

Like the ozone analysis, the spatial distribution patterns of the average modelled 1-h maximum and hourly NO_2 concentrations are shown in Figures 4c and 4d, which are labelled in a different way according to their maximum hourly limit value or according to their mean hourly maximum values. Results show where the detailed emission implemented in the HIREM model indicates that on road traffic (Figure 1c) constituted the main source of this pollutant in the BMA, although emissions coming from industrial sources represented in Figure 4d by small dots of maxima concentrations were also noticeable.

Barcelona and the main part of the medium-sized cities are located within the domain corresponding to BMA (D4), therefore a new validation and the time evolution of average hourly NO_2 concentrations was therefore performed for this domain. The results are given in Table 4, and Figure 5c showing that the statistics met also the recommended performance criteria, and the model reproduced daily variability quite well, although it overestimated peak values.

3.2.3 Particulate Matter (PM10)

The evaluation of PM10 was performed using a total of 67 stations, 36 were manual and 31 automatic. The statistical evaluation parameters applied on a daily basis was compiled in Table 4. Results show that they almost met the recommended performance criteria, although some values ranged near or above the criteria for acceptable model performance. Besides, if we compare these results with the same statistics used to evaluate the model performance criteria for 1-h maximum NO_2 and O_3 concentrations and 8-h maximum O_3 concentrations, less accurate model levels are obtained for the PM10 concentration forecast. These results agree with those found in many studies, which have reported that models are not very accurate when simulating particulate matter over different areas (Mathias, 2008; Pay et al., 2010; Baldasano et al., 2011; Basart et al., 2012).

The time evolution of average hourly PM10 concentrations provided by the ARAMIS model at the automatic measurement stations is displayed in Figure 6a. The results show that the model tended to slightly overestimate nocturnal values and to underestimate day-time values. In addition, time series of daily average concentrations forecasted by the model shown in Figure 6b indicates quite poor reproduction of daily variability and a persistent underestimation of modelled

concentrations during summer time. As before, modelled discontinuities observed in this figure are due to problems in model execution.

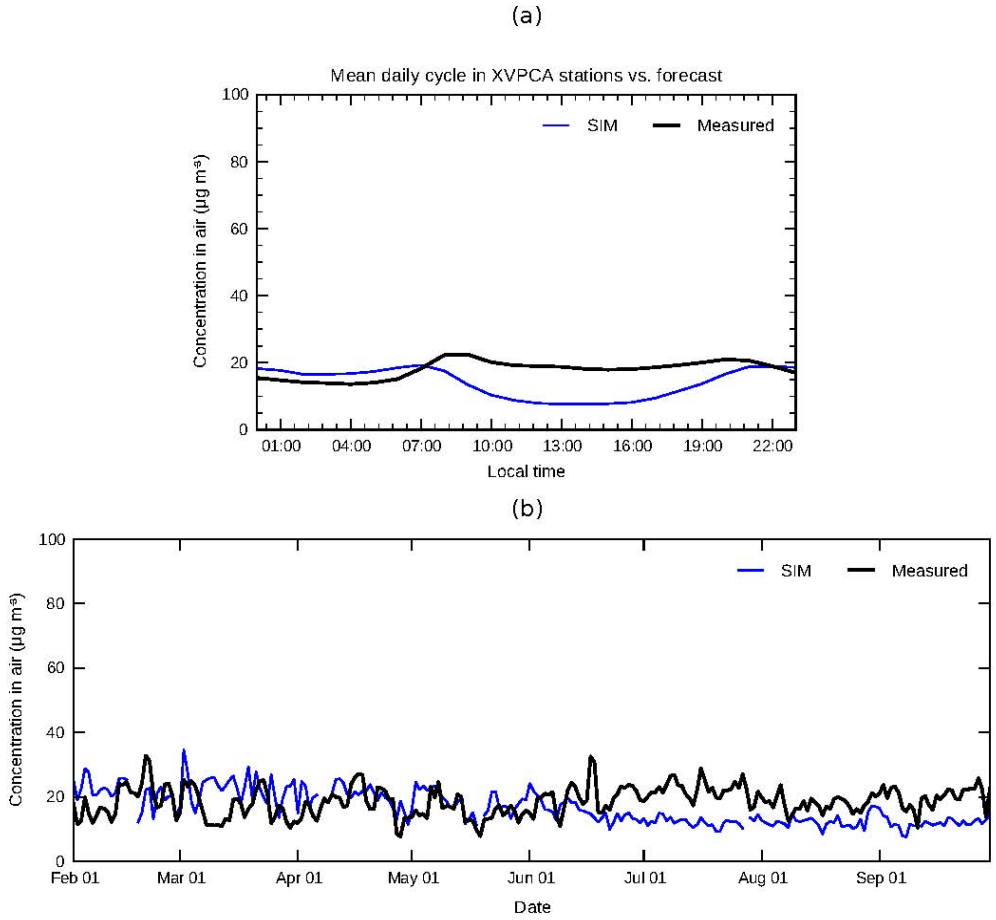

Figure 6. Modelled (blue line) and measured (black line) of: (a) time evolution of average hourly PM10 concentrations, (b) time series of daily mean PM10 concentrations over all measurements stations located in domain D3 (Catalonia).

Several factors could contribute to the underestimation of total particulate matter concentrations, of which the most important are: i) The model do not capture emission sources with high temporal variability, emissions are a key source of errors (Ritter et al., 2013); ii) a lack of fugitive dust and re-suspended matter emissions (Vautard et al., 2005; 2007), iii) a possible underestimation of primary carbonaceous particles (Schaap et al., 2004; Tsyro, 2005), iv) the inaccuracy of secondary organic aerosol (SOA) formation (Simpson et al., 2007), since much research and

development is still needed to improve secondary organic aerosol formation in chemistry transport models, v) difficulties in representing primary PM emissions such as wood burning, the building trade and other sources not considered in the emission model (Baldasano et al., 2011) and finally vi) a fraction of coarse particles is underestimated in the emission model or the assumptions and the chemical treatment in the CMAQ model aerosol module needs to be improved. Overall, a more in-depth knowledge of aerosol processes, removal, dispersion and transport processes would be necessary to improve the model's particulate matter forecast. In addition, in Mediterranean areas the parameterization of emission rates due to mineral dust must be taken into account (Büns et al., 2012). Currently this emission rates are not considered in ARAMIS but in a near future it will be incorporated to improve the simulation results.

To conclude the analysis, the spatial distribution pattern of the average modelled hourly concentrations was given in Figure 4e. Results show that as with NO_2 distribution, the highest concentrations were located in the BMA, where on-road traffic (Figure 1c) constituted the main source of this pollutant.

4. Conclusions

In this paper, we have described the evaluation and assessment of ARAMIS (WRF/HIREM/CMAQ) used to forecast air quality over regional areas in order to develop emission abatement strategies for air pollution and adverse health effects. ARAMIS was applied with high resolution over north-east Spain (Catalonia) from February to September 2013. Evaluation of the modelling results was mainly focused on the capacity of ARAMIS to reproduce the time evolution of average hourly O_3, NO_2 and particulate matter (PM10) concentrations as well as its capacity to forecast 1-h and 8-h for O_3 maximum concentrations, 1-h maximum concentration for NO_2 and daily average PM10 concentrations.

The evaluation was performed for both the WRF meteorological model and the CMAQ photochemical model using the classic approach. The results showed a good performance for WRF forecasts related to humidity and temperature, whilst worst performance was found for wind velocity, and mainly for wind direction. This result can primarily be attributed to the complex topography of the area studied which is misrepresented in the model, to random turbulent processes or to sub-grid variations in terrain and land use that cannot be simulated by the models.

To drive the evaluation of the air quality modelling system and to quantify its performance, we used the statistics and criteria proposed by the United States Environmental Protection Agency (U.S.-EPA) and European regulations. The results for O_3, NO_2 and PM10 showed that all statistics met the recommended performance criteria; the best results were found for simulated O_3 concentrations and the worst for PM10.

The time evolution of average hourly O_3, NO_2 and PM10 concentrations provided by the ARAMIS model at the measurement stations showed quite a good reproduction of hourly variability; however, the model tended to overestimate night-time concentrations and underestimate day-time ones, especially NO_2 concentrations. This behaviour has been attributed to different causes: i) the model does not represent nocturnal physico-chemical processes accurately enough, ii) the HIREM emission model may not calculate night-time emissions properly, and iii) meteorological parameters, such as wind speed, wind direction, vertical mixing and boundary layer transition were not well reproduced by the model.

Time series of daily maximum 1-h O_3, 8-h O_3 and 1-h NO_2 concentrations forecast by the model showed that mean variability was quite well reproduced by the model. However, daily ozone peaks in summer, mainly during July, and NO_2 peaks in winter and spring were underestimated. The model yielded worst results for forecast PM10 concentrations, showing quite poor reproduction of the mean daily values. As mentioned previously chemical lateral boundary conditions (LBCs) for domain D1 came from a vertical profile supplied by CMAQ itself, we believe that one way to improve pollutants variability predictions, especially ozone, would be to use a time and space variant chemical LBCs from a chemistry-climate model as showed Akritidis et al., (2013).

Results for the spatial distribution patterns of the average modelled 1-h and 8-h maximum ozone concentrations, averaged modelled 1-h maximum and hourly NO_2 concentrations and averaged modelled hourly PM10 concentrations showed that the pattern was mainly dominated by traffic emissions and therefore subject to temporal emission variations for NO_2 and PM10, whilst for O_3, in addition to precursor emission and low deposition, the pattern distribution was dominated by meteorological synoptic and mesoscale situations. Both conditions force persistent subsidence over the region and the development of local winds such as the sea breeze contributes to increasing inland ozone concentrations.

Global results of the ARAMIS evaluation showed that there are still several areas for improvement in the model, mainly related to traffic emissions, especially in small cities where these are not well characterised. However, the accurate performance of ARAMIS for O_3 and NO_2 demonstrates that the system can be implemented and evaluated operationally.

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