Influence of high-model grid resolution on photochemical modelling in very complex terrains

Pedro Jiménez, Oriol Jorba, René Parra and José M. Baldasano*

Laboratory of Environmental Modeling
Universitat Politècnica de Catalunya (UPC)
Av. Diagonal 647, 08028 Barcelona, Spain
Fax: +34 93340255
E-mail: pedro.jimenez-guerrero@upc.edu
E-mail: oriol.jorba@upc.edu
E-mail: rene.parra-narvaez@upc.edu
E-mail: jose.baldasano@upc.es
*Corresponding author

Abstract: The study of photochemical pollution in very complex terrains, such as the Northeastern Iberian Peninsula (NEIP), is primarily influenced by local topography. This kind of study demands a high spatial resolution. In order to illustrate the influence of the grid resolution on tropospheric ozone levels, several simulations were carried out with MM5-EMICAT2000-CMAQ model using horizontal resolutions ranging from 8, 4 and 2 km and with 6 or 16 vertical layers during an episode of photochemical pollution (13–16 August 2000). High resolutions lead to improve discrete and categorical statistical parameters when evaluating the model against meteorological and air quality data. Results show that coarser grids tend to present a homogeneous and smoothened behaviour of wind flows. Also, results do not manage to describe the particularities of the circulations of the region. Coarser simulations underestimate maximum ozone levels since the grid resolution highly influences the formation and loss processes of ozone.

Keywords: air quality modelling; model evaluation; grid resolution; very complex terrains.

Reference to this paper should be made as follows: Jiménez, P., Jorba, O., Parra, R. and Baldasano, J.M. (2005) ‘Influence of high-model grid resolution on photochemical modelling in very complex terrains’, *Int. J. Environment and Pollution*, Vol. 24, Nos. 1/2/3/4, pp.180–200.

Biographical notes: Pedro Jiménez and Oriol Jorba are PhD students in the Environmental Program at the Laboratory of Environmental Modeling (LMA), Technical University of Catalonia (UPC), Barcelona, Spain. René Parra is a post-doctoral researcher at LMA. José María Baldasano is full Professor in Environmental Science and Technology in the High Technical School of Industrial Engineers at the Technical University of Catalonia.

1 Introduction

The Northeastern Iberian Peninsula (NEIP) has a complex topography with a large coast to the Mediterranean Sea. The complex configuration of the zone comes conditioned by the presence of the Pyrenees mountain range (with altitudes over 3000 m), the influence of the Mediterranean Sea and the large valley canalisation of the Ebro River. These produce a sharp gradient in the characteristics of the NEIP. This sort of terrain induces an extremely complicated structure of the flow because of the development of large and local mesoscale phenomena that interact with synoptic flows. The characteristics of the breezes have important effects in the dispersion of pollutants emitted. In addition, the flow can be even more complex because of the nonhomogeneity of the terrain, the land-use and the types of vegetation. In these situations, the structure of the flow is extremely complicated because of the superposition of circulations of different scale. Hence, the study of photochemical pollution in complex terrains demands a high horizontal spatial resolution. The average volume defined by the model's horizontal grid spacing must be sufficiently small to allow the air quality to be reproduced accurately. The large averaging volumes used by regional models are feared to lead to unacceptable errors for many species that are formed via nonlinear chemical reactions (*e.g.*, ozone and its precursors), particularly in areas with significant chemical gradients such as cities (Russell and Dennis, 2000). On the other hand, small-scale topographical features captured by small grid spacing in the model can result in numerical instabilities that contaminate predictions. For these reasons, and since computational requirements increase markedly with the inverse of the grid spacing (for a given domain), the grid size becomes an important design decision.

Increasing spatial resolution of a meteorological model allows one to include more mesoscale motions in its numerical solution (Pielke and Uliasz, 1998). McQueen *et al.* (1995) performed a sensitivity analysis on the influence of grid size when applying an operational mesoscale numerical weather prediction model. The study showed the importance of using high-resolution grids when the model is applied to a complex terrain. Horizontal resolutions up to 1 km should be considered when simulating vertical motion associated with see breezes (Lyons *et al.*, 1995).

Jang *et al.* (1995a–b) studied the sensitivity of ozone to the horizontal grid resolution of air quality models, concluding that a coarser grid size tends to underestimate the maximum ozone levels and to overestimate minimum values, since the grid resolution highly influences the formation and loss processes of ozone, specially photochemical and vertical transport phenomena. The best agreement between the predictions and the observations was attained with horizontal resolutions of 2.5 km. Byun and Dennis (1995) revealed that a 15-layer vertical resolution with RADM model represents better the surface dry-deposition processes and surface concentration of photochemical pollutants than a 6-layer resolution, the latter being inadequate in estimating the pollutants under stable atmospheric conditions; the necessity for a high vertical resolution comes conditioned by the short scale of vertical mixing of certain gas reactions. Thunis (2001) indicated that ozone concentration fields obtained with similar meteorology and emissions but with different horizontal spatial resolutions in the Milan area produced similar O_3 surface field patterns. However, significant differences were obtained for peak concentrations (20%) between a 10- and a 50-km grid, whereas the 4- and 10-km simulation gave similar results.

In the case study of NEIP, the work of Soriano *et al.* (1997) showed that a 2-km grid resolution described correctly the forcing caused by the topography in the Barcelona Geographical Area, with no improvement in modelling results when increasing the horizontal resolution to 1 or 0.5 km. Salvador *et al.* (1999) analysed the improvement of the results when modifying the grid resolution for a domain covering the eastern Iberian Peninsula. Authors stated the enhancement in the description of meteorological phenomena in a small scale when working with resolutions of 2 km or finer. This study covers grid resolution of 6, 4, 2 and 1.5 km, obtaining significant differences in the simulation results. Nevertheless, simulations with a 1.5-km grid entail an execution time 20 times higher than the corresponding 2-km simulation.

Consequently, the precise definition of the grid resolution is important when applying an air quality model. In order to illustrate the influence of grid size on ground-level O_3, this work encloses a detailed study of the observable differences in values, location and temporal behaviour of tropospheric O_3 in the NEIP when applying a three-dimensional air quality model with different horizontal resolutions (8, 4 and 2 km). We also assess how much of the systematic model bias can be explained by changing the vertical grid resolution (6 and 16 layers covering the troposphere).

2 Methodology

Several simulations with a three-dimensional air quality model (MM5-EMICAT2000 -CMAQ) are carried out using different grid resolutions: 8 km, 4 km and 2 km. The domain of study (Figure 1) covers a squared area of 272 km × 272 km. The chemical transport model also is configured with both six and 16 vertical layers to cover the troposphere, with special emphasis in the low troposphere (4000 metres).

Figure 1 Geography of the domain and areas where photochemical pollution episodes are measured in the northeastern Iberian Peninsula (top), and model terrain data from 2 km MM5 data (bottom)

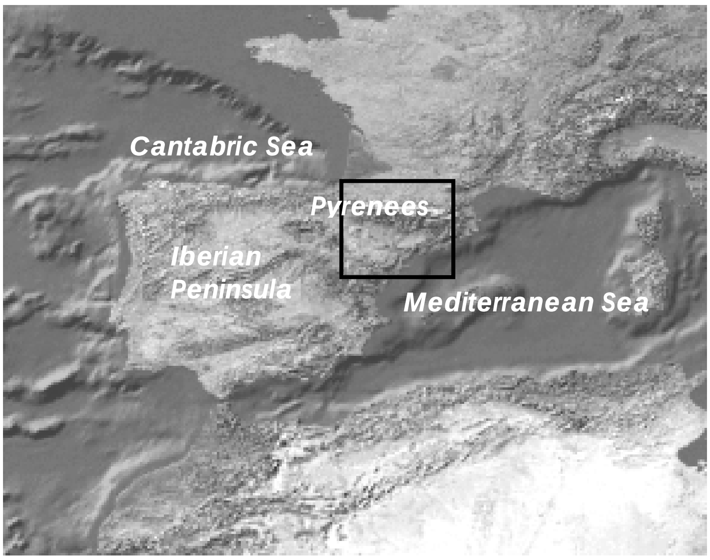
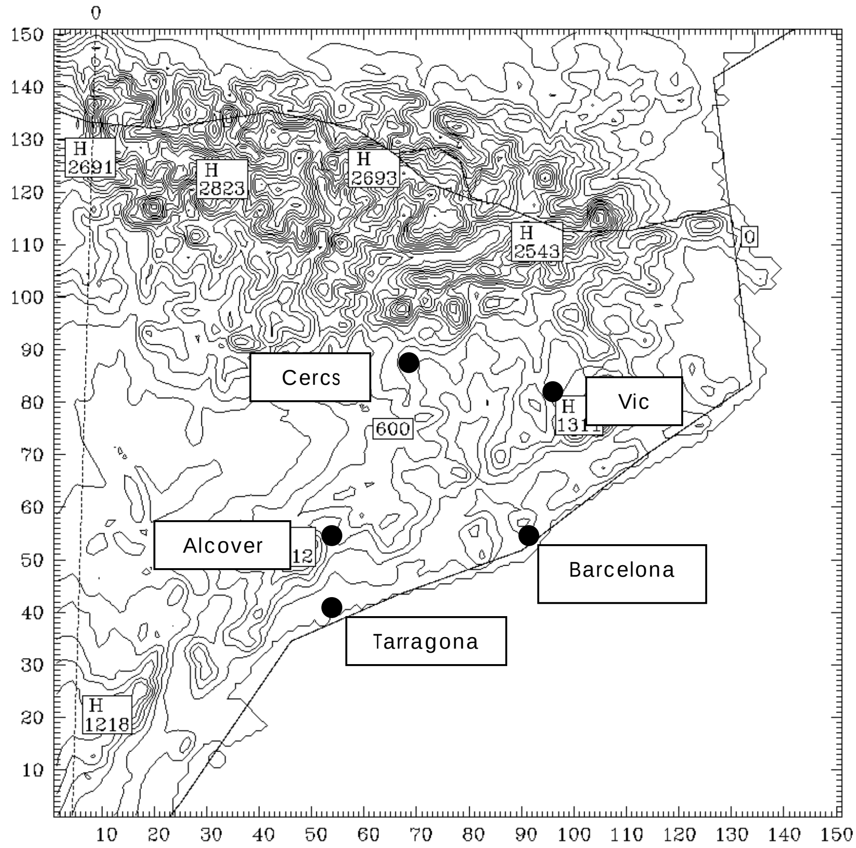


Figure 1 Geography of the domain and areas where photochemical pollution episodes are measured in the northeastern Iberian Peninsula (top), and model terrain data from 2 km MM5 data (bottom) (continued)



2.1 Models

Hourly meteorological fields were provided by MM5 mesoscale model (MMMD/NCAR, 2001). The physics options used for the simulations were the following: the MRF scheme for the PBL parameterisation, the Anthes-Kuo and Kain-Fritsch cumulus scheme (Kain and Fritsch, 1993), the Dudhia simple ice moisture scheme, the cloud-radiation scheme, and the five-layer soil model (MMMD/NCAR, 2001). MM5 produces meteorological information at 29 sigma levels. Initialisation and boundary conditions for the mesoscale model were introduced with analysis data of the European Centre of Medium-Range Weather Forecasts global model (ECMWF). Data were available at a 1-degree resolution (approximately 100-km at the working latitude) at the standard pressure levels every six hours.

The high resolution (1 h and 1 km^2) EMICAT2000 emission model (Parra, 2004) has been applied in the NEIP. As precursors of photochemical smog, the model estimates the cell emissions of nitrogen oxides (NO_x), nonmethane volatile organic compounds (NMVOCs), carbon monoxide (CO), and also Total Suspended Particles

(TSP) and sulphur dioxide (SO_2). This emission model includes the emissions from vegetation, on-road traffic, industries and emissions by fossil fuel consumption and domestic-commercial solvent use. Biogenic VOCs (BVOCs) emissions were estimated using a methodology that takes into account local vegetation data (land-use distribution and biomass factors) and meteorological conditions (surface air temperature and solar radiation from field measurements) together with emission factors for native Mediterranean species and cultures (Parra *et al.*, 2004). On-road traffic emission includes the hot exhaust, cold exhaust and evaporative emissions using the methodology and emission factors of the European model EMEP/CORINAIR–COPERTIII (Ntziachristos and Samaras, 2000) as basis, and differencing the vehicle park composition between weekdays and weekends (Parra and Baldasano, 2004). Industrial emissions include real records of some chimneys connected to the emission control net of the Environmental Department of Catalonia Government (Spain), and the estimated emissions from power stations (conventional and cogeneration units), incinerators, cement factories, refineries, olefin plants and chemical industries.

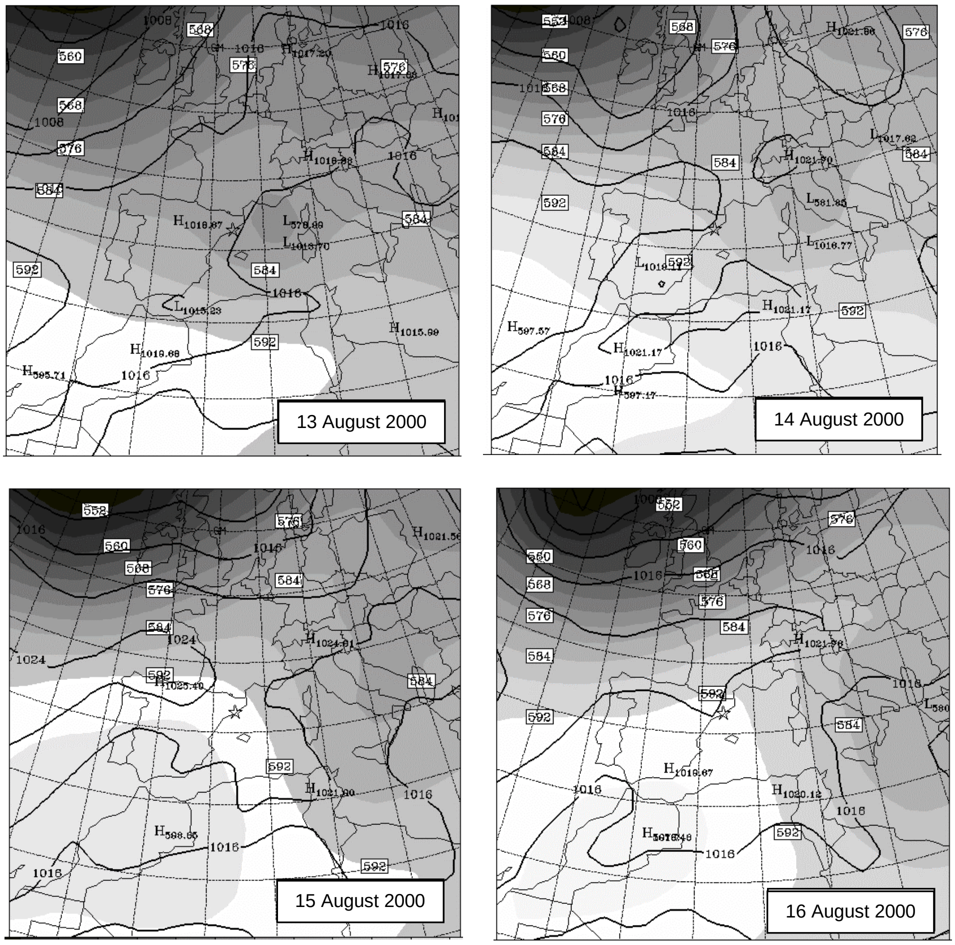
The chemical transport model used to compute the concentrations of photochemical pollutants was Models-3/CMAQ (Byun and Ching, 1999). Following the criteria of Jiménez *et al.* (2003), the chemical mechanism selected for simulations was CBM-IV (Gery *et al.*, 1989), including aerosols and heterogeneous chemistry. The algorithm chosen for the resolution of tropospheric chemistry was the Modified Euler Backward Iterative (MEBI) method (Huang and Chang, 2001). Horizontal resolutions considered were 8, 4 and 2 km. Vertical layers of the chemical transport model are defined in sigma-coordinates. The first layer considers a thickness of 150 m (for the six vertical layers case) and 36 m (for the 16 vertical layers case). Layers of both the 6-layer simulation and the 16-layer simulation were obtained by collapsing the 29 layers of the MM5 simulation.

2.2 Case study

The episode selected for performing the simulations was from 13 to 16 August 2000, which is representative of a photochemical pollution episode that covered the whole Western Mediterranean Basin. Values over the European threshold of 180 $\mu g\ m^{-3}$ for tropospheric O_3 were attained. Synoptic situation of this episode corresponds to a typical summertime low pressure gradient over the Iberian Peninsula, and this situation is related to a decrease in air quality (Jorba *et al.*, 2004). Maximum O_3 levels in the NEIP are measured downwind the city of Barcelona and in the Alcover industrial zone. The weak synoptic forcing provoked that mesoscale phenomena, induced by the particular topography of the region, would be dominant.

A high sea level pressure and almost nonexistent surface pressure gradients over the domain characterise this day (Figure 2), with slow northwesterly aloft. Under this weak synoptic forcing, strong insolation promotes the development of prevailing mesoscale flows associated with the local orography (mountain and valley breezes), while the difference of temperature between the sea and the land enhances the development of sea-land breezes (Barros *et al.*, 2003). A number of studies have shown that during this type of episodes, layering and accumulation of pollutants such as ozone and aerosols were taking place along the NEIP (Millán *et al.*, 1992; 1997). Baldasano *et al.* (1994) and Soriano *et al.* (2001) combined a numerical approach with an elastic-lidar sounding campaign to study the circulatory patterns of air pollutants over Barcelona (NEIP) in a typical summertime situation, where pollutant layers were formed by the return flow of the breeze and forcings caused by the complex orography combined with the compensatory subsidence. This was confirmed and detailed in a regular basis by Pérez *et al.* (2004).

Figure 2 Synoptic situation of 13 August 2000 to 16 August 2000 (shaded map: 00UTC 500 hPa geopotential height field [10 m], contour map: 00UTC sea level pressure field [hPa])



2.3 Evaluation of different resolutions

For the assessment of the performance of MM5 meteorological model when considering different resolutions, simulation results were compared with surface and aloft wind measurements. Validation data of 52 surface stations located across the domain and a radiosonde launched in Barcelona (in the centre of the domain in the coast) were used. Ground-level O_3 simulation results were compared with the measurements from 48 surface stations in the NEIP (Catalonia, Spain), located in both urban and rural areas. Hourly measures of ozone were utilised to indicate the performance of the chemical transport model results when employing different horizontal and vertical resolutions. The analysis of the results will consist of a statistical comparison (both discrete and categorical parameters) between the first-layer simulations results and the values measured in the air quality stations of the domain under study. Both meteorological and air quality data were provided by the Environmental Department of the Catalonia Government.

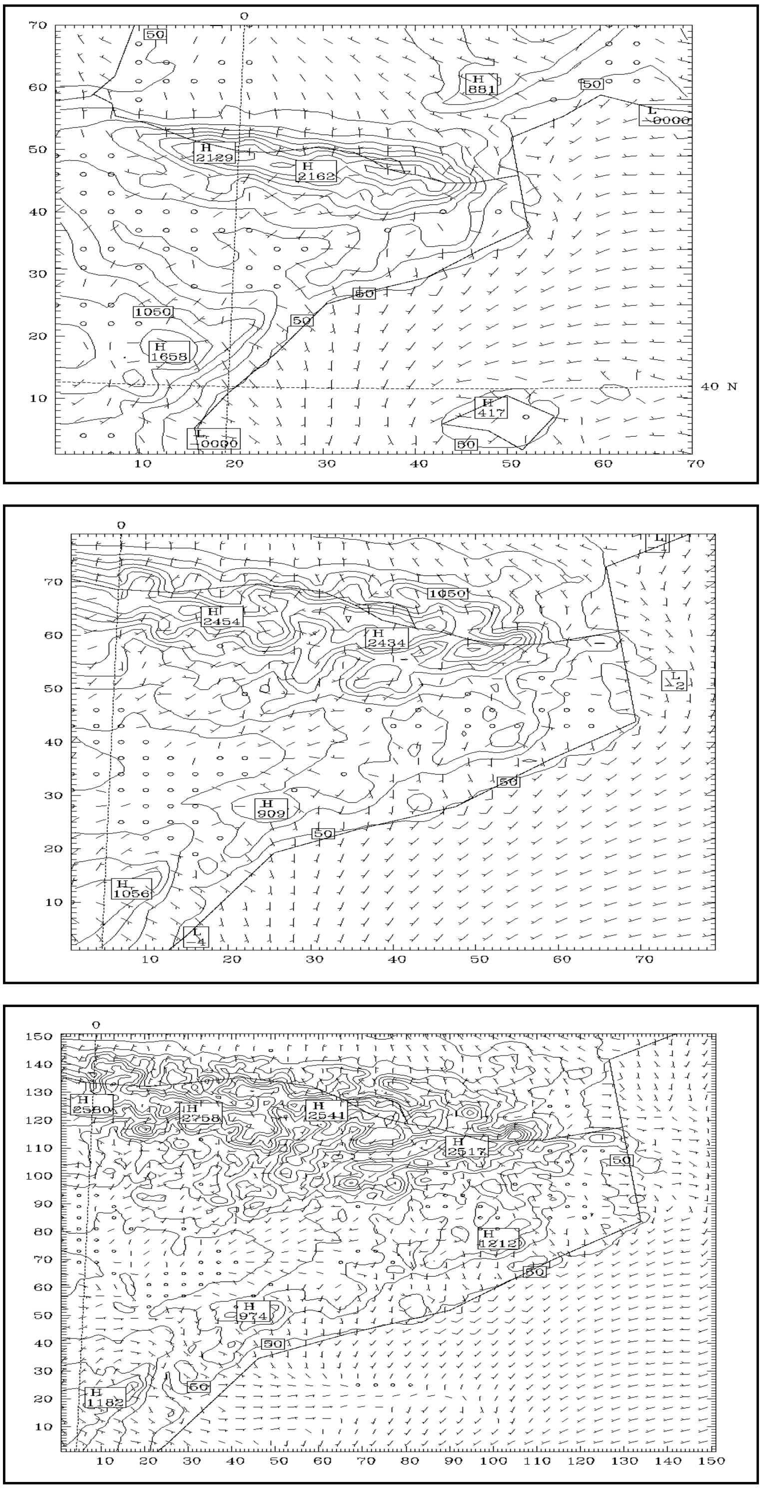
3 Influence of model resolution on wind fields

Grid size is influenced primarily by local topography (Salvador *et al.*, 1999). Topographical variations can have an important effect on mesoscale atmospheric flow and, therefore, although topography is not the only driving mechanism that contributes to the dispersion of pollutants in the given domain, it plays a major role and should be well resolved in modelling exercises. The topography of the domain of study is organised from three structural units forming a fan-shape formation with the vertex in the Empordà as shown in Figure 1:

1. Pyrenees, forming a barrier at the north of the region and expanding from east to west, constitute the first great unity
2. Central Depression stretches all interior zones
3. Mediterranean System, formed by the joint of mountain ranges and depressions parallel to the coast.

Figure 3 represents the topographic map of the domain at an 8-km, 4-km and 2-km resolution. There are several significant differences between coarse and fine topography. Nevertheless, we must bear in mind that smoothing is sometimes positive to avoid the strong topographical gradients that may not be properly resolved by the models (McQueen *et al.*, 1995). Results depict that important mesoscale phenomena within the region do not develop in the domain of study if the horizontal resolution is coarser than 4 km. Under low synoptic forcings, mountain winds develop because of the very complex topography of the region, while the difference of temperature between the sea and the land enhances the development of sea-land breezes. Clear differences are observed in the wind direction especially over the sea (Figure 3). The daily cycle of these flows constitutes an important part of the mechanism that drives the transport of air pollutants within the region.

Figure 3 Surface wind field in Catalonia for 14 August 2000 at 1200UTC. eight-kilometre grid (up), four kilometre (centre) and 2-km grid (down) [Contour lines: topography]



Results show how the see breeze development is well captured by all simulations, even though particular canalisations of the flow are only appreciable at 4- and 2-km resolutions. With lower resolutions, wind fields present a very smoothed structure. More differences appear with mountain-winds, which are not developed at 8 km. Only with a fine high resolution, the meteorological model is able to reproduce the mountain-wind system; coarser resolution simulations do not manage to describe the complexity observed in the Pyrenees, whose influence is more stressed when incrementing resolution to 2 km. The coastal mountain range orientation (SW-NE) favours a rapid solar heating of slopes and an early onset of the sea breeze aided by the upslope winds. Because of the orientation of the mountain ranges, thermal effects intensify the sea breeze. The vertical structure of the flows is also influenced by the best representation of the topography when working with high horizontal resolutions. An enhancement of vertical motions is observed; the vertical structure of the see-breeze is improved; and several orographic injections appear when increasing the resolution. Nighttime regime differs substantially in both simulations, with down-slope and down-valley winds in the Pyrenees valleys (Cerdanya) that are observable in the finest grid but are not described by the 8-km or 4-km grid.

Table 1 shows the Root Mean Square Error (RMSE) of wind speed at 10 m, for the lower, middle and upper troposphere and RMSE of wind direction at 10 m for the different horizontal resolutions. The general behaviour of the model shows a tendency to overestimate nocturnal surface winds and to underestimate the diurnal flow. A clear improvement is produced with 2-km simulation during the central part of the day. The complex structure of the see-breeze described by the 2-km simulation and the development of up-slope winds appear to agree in a higher grade with surface measurements. Statistics show how the model presents a better behaviour within the boundary layer, and major disagreement with the radiosonde appears over 1000 m AGL. At night, 8- and 4-km presents better results aloft, while at noon the high horizontal resolution simulation obtains the best statistics.

Table 1 RMSE statistic of wind speed, and wind direction for 2-, 4-, 8-km simulations at 00, 12 UTC of 13 August 2000 and 00 UTC of 14 August 2000 (surface values evaluated with 52 surface stations, aloft values evaluated with a radiosonde)

	RMSE wind speed (m/s)								
	00 UTC 13rd			*12 UTC 13rd*			*00 UTC 14th*		
	2 km	*4 km*	*8 km*	*2 km*	*4 km*	*8 km*	*2 km*	*4 km*	*8 km*
Surface 10 m	1.71	1.85	1.76	2.04	2.34	2.41	2.00	2.17	2.22
Radio < 1000 m	0.84	1.02	1.04	1.04	1.08	1.34	1.31	0.86	0.86
1000–5000 m	5.03	3.95	3.66	1.55	2.4	2.16	3.7	2.59	2.27
5000–10000 m	8.45	8.92	8.62	5.15	6.22	6.3	3.94	5.29	5.16
	RMSE wind direction (º)								
	00 UTC 13rd			*12 UTC 13rd*			*00 UTC 14th*		
	2 km	*4 km*	*8 km*	*2 km*	*4 km*	*8 km*	*2 km*	*4 km*	*8 km*
Surface 10 m	95.95	91.33	92.10	44.74	58.17	55.25	89.40	98.59	94.69

4 Importance of resolution on emissions

The main emissions sources in the NEIP are located on the coast (Parra, 2004). Total emissions of ozone precursors during 14 August 2000 were 236 t d^{-1} for NO_x and 172 t d^{-1} for VOCs. The importance of biogenic emissions is high in the area since this source represents 34% of VOCs emissions in the NEIP during this summer episode. Further, biogenic emissions is an important source of reactive compounds such as aldehydes and isoprene. Road traffic accounts for 58% of NO_x emissions and 36% of VOCs in the domain (specially olefins and aromatic compounds). Most important emitters are found along the road-axis parallel to the coast and Barcelona Geographical Area. Industrial emissions are principally located in the industrial area of Alcover, downwind the city of Tarragona, one of the most important industrial centres in Spain; and these emissions represent 39% of NO_x emissions and 17% of VOCs. Meanwhile, the use of residential and domestic solvent use provides the 13% of VOCs emissions in the area (Parra, 2004). The peculiar topography of the zone is the principal driving mechanism that contributes to the dispersion of pollutants emitted in the given domain.

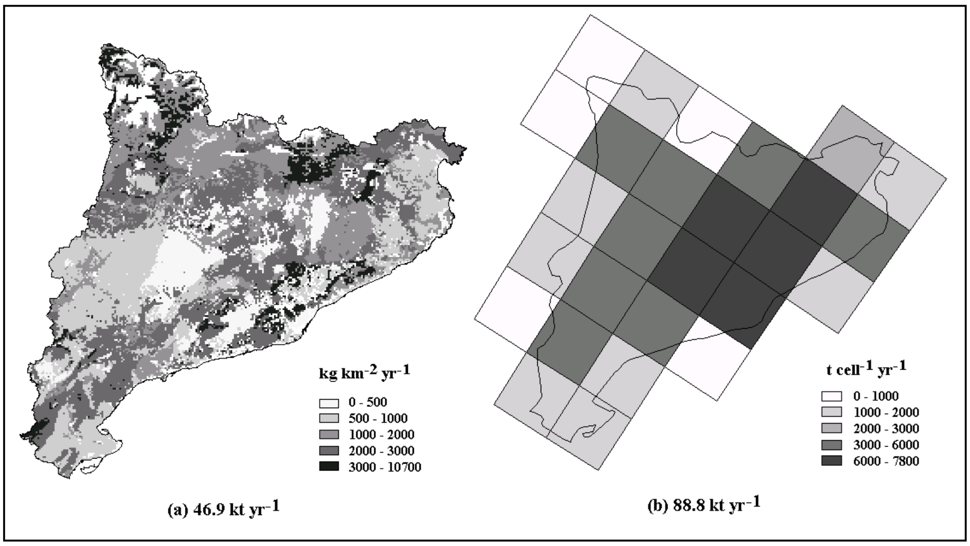
The generation of boundary conditions for the domain of the NEIP requires photochemical inputs from a coarser domain; therefore, these conditions were derived from a one-way nested simulation covering a domain of 1392 km × 1104 km centred in the Iberian Peninsula using EMEP emissions for the year 2000.[1] Cells from the European EMEP mesh have a resolution of 50 km in polar coordinates; meanwhile EMICAT2000 provides a horizontal resolution of 1 km. Table 2 shows a comparative summary of annual emissions of primary air pollutants provided by EMEP inventory and EMICAT2000 for the year 2000.

The annual EMICAT2000 vegetation emission (46.9 kt of NMVOC) is 52% lower than the estimation by EMEP (Sector 11) in an equivalent domain (Figure 4). A summary of vegetation emissions at European level is included in EEA (2003). This information was taken from Simpson *et al.* (1995) and Guenther *et al.* (1995). For Spain the total annual estimation is 657 kt yr^{-1} (21% of isoprene, 38% of monoterpenes and 41% of OVOC). Using the coarser approach of considering the surface ratio between the domain of study of the NEIP (31,895 km^2) and Spain (504,782 km^2), the result would be 41.4 kt yr^{-1}, which is an equivalent vale to the annual estimation EMICAT2000 (46.9 kt yr^{-1}), and, therefore, EMEP biogenic emissions may be overestimated in the NEIP. Regarding to on-road traffic, EMICAT2000 and the EMEP Sector 7 (road traffic) estimations are equivalent. In fact, NO_x, CO and NMVOCs emissions provided by EMICAT2000 are 74, 95 and 87%, respectively, of the EMEP values. Last, emissions by industrial activities are in the same range that values obtained by the EMEP Sectors 3 (industrial combustion) and four (production processes) for the NEIP except for the values for carbon monoxide. Small industrial centres that carry out a great uncertainty in the estimation of their emissions were not included in EMICAT2000 because of this fact, and they could contribute with a large percentage to CO emissions (Parra, 2004).

Table 2 Comparative summary of sectorial annual emissions from EMICAT2000 with EMEP European inventory (kt yr^{-1}) for the domain of the northeastern Iberian Peninsula

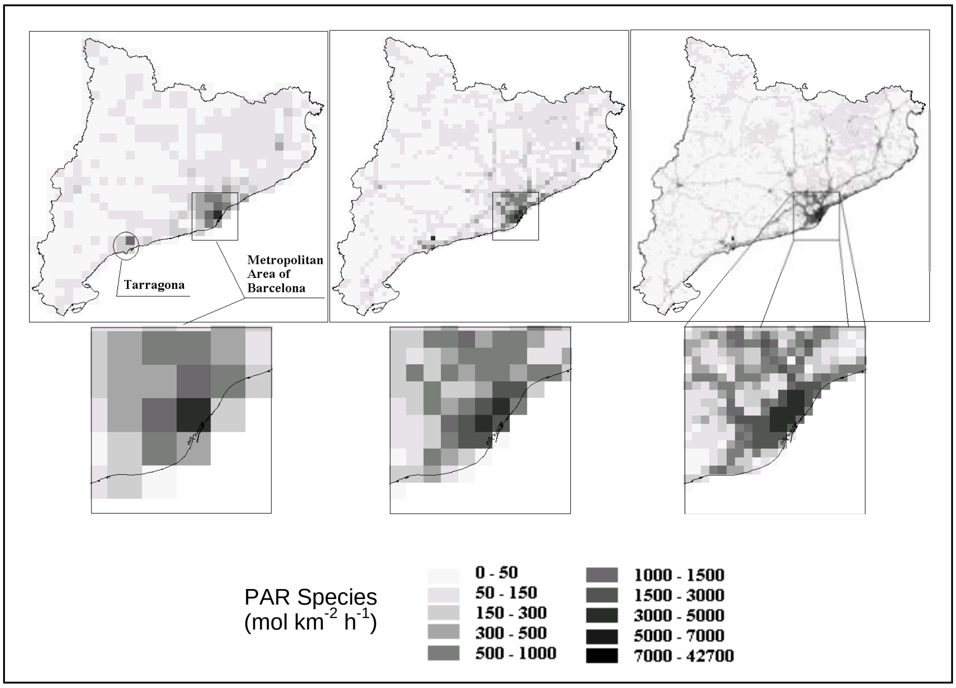
Air pollutant	*EMEP 2000 (50 km × 50 km cells)*	*EMICAT2000 (1 km × 1 km cells)*
	Vegetation	
Isoprene	–	5.9
Monoterpenes	–	24.7
Other NMVOCs	–	16.3
Total	88.8	46.9
	On-road traffic	
NO_x	84.8	62.4
CO	274.2	259.0
NMCOVs	58.0	50.5
SO_2	2.7	1.3
CO_2 eq.	–	8302
	Power generation	
NO_x	11.5	15.0
CO	1.3	2.1
NMVOCs	0.4	0.7
SO_2	38.5	28.5
CO_2 eq.	–	5698
	Industrial activities (except power generation)	
NO_x	30.9	26.1
CO	23.5	5.4
NMVOCs	21.8	22.0
SO_2	47.2	32.7
CO_2 eq.	–	14663

Figure 4 Comparison of EMICAT2000 (left) and EMEP (right) biogenic emissions estimates for year 2000



The spatial resolution on the emission of ozone precursors plays a fundamental role in the definition of emission patterns of pollutants in the NEIP. Applying a 2-km resolution depicts in detail the aforementioned emission patterns for the domain of study. The axes of the most important roads are clearly defined, as well as the metropolitan area of Barcelona and the industrial zone of Alcover. Considering a coarser resolution, these emission features smoothened, until they get lost with 8-km cells, where the attenuation of the borders of emissions do not allow a clear definition of cities, roads and other emitter areas (Figure 5).

Figure 5 Influence of the spatial resolution on NMVOCs (represented by CBM-IV species PAR) during 14 August 2000 at 1200 UTC for a resolution of 8-km (left), 4-km (centre) and 2-km (right)



5 Influence of model resolution on tropospheric ozone

The US Environmental Protection Agency has developed guidelines (US-EPA, 1991) drawn from Tesche *et al.* (1990) for a minimum set of statistical measures to be used for these evaluations where monitoring data are sufficiently dense. Those statistical figures are the following: Mean Normalised Bias Error (MNBE), Mean Normalised Gross Error for concentrations above a prescribed threshold (MNGE) and unpaired peak prediction accuracy (UPA). The values for the MNBE and UPA can be either positive or negative. Observation/prediction pairs were often excluded from the analysis if the observed concentration was below a certain cut-off; the cut-off levels varied from study to study but often a level of 120 μg m^{-3} is used (Hogrefe *et al.*, 2001), which is the criterion applied in this work.

Categorical statistics, as derived from Kang *et al.* (2003), have also been used to evaluate the different vertical and horizontal resolution, including parameters such as model accuracy (A), bias (B), Probability of Detection (POD), False Alarm Rates (FAR) and Critical Success Index (CSI). These criteria are based also upon a 120 μg m^{-3} threshold.

Table 3 collects the results of the statistical analysis; although there is no objective criterion set forth for a satisfactory model performance, suggested values of ±10–15% for MNBE, ±15–20% for the UPA and ±30–35% for the MNGE to be met by modelling simulations of O_3 have been considered for regulatory applications (Russell and Dennis, 2000). We should bear in mind, however, that a surface measurement represents a value only at a given horizontal location and height, while the concentration predicted by the model represents a volume-averaged value.

Table 3 Summary results for evaluation of ozone concentrations with different horizontal and vertical resolutions [the statistics are defined by US-EPA (1991)]

Max Obs (μg m^{-3})	*189*					
Max Sim (μg m^{-3})	*207*	*188*	*131*	*143*	*137*	*144*
	Discrete evaluation					
	2 km/ 6 layers	*2 km/ 16 layers*	*4 km/ 6 layers*	*4 km/ 16 layers*	*8 km/ 6 layers*	*8 km/ 16 layers*
MNBE (%)	–2.02	–11.38	–9.53	–16.92	–12.86	–13.12
MNGE (%)	19.72	20.89	17.42	21.61	19.27	21.01
UPA (%)	17.08	–0.76	–26.07	–19.24	–22.53	–18.49
	Categorical evaluation					
	2 km/ 6 layers	*2 km/ 16 layers*	*4 km/ 6 layers*	*4 km/ 16 layers*	*8 km/ 6 layers*	*8 km/ 16 layers*
A (%)	90.9	91.6	91.5	92.4	91.6	91.7
CSI (%)	19.0	12.5	3.2	8.9	3.2	10.0
POD (%)	26.4	14.9	3.4	9.2	3.4	11.5
B (%)	0.7	0.3	0.1	0.1	0.1	0.3
FAR (%)	59.6	56.7	72.7	27.3	70.0	56.5

Progressively increasing grid resolution from 8 km to 2 km improves the performance of all statistical parameters. The MNBE is negative for every simulation, ranging from –16.92% for 4 km/16 layers up to –2.02% for 2 km/6 layers case. The MNGE is similar in all cases studied, being within the aforementioned standards. The UPA does not greatly improve when augmenting the resolution from 8 km to 4 km, but this improvement becomes evident in the 2 km/16 layers simulation, yielding values of –0.76% for the highest vertical and horizontal resolution.

With respect to categorical forecasting, statistical parameters indicate that the accuracy (percent of forecasts that correctly predict exceedance or non-exceedance) is above 90% for every resolution, but yielding the best parameters for the 16-vertical layers resolution with respect to the inferior vertical resolution. Since this metric can be greatly influenced by the overwhelming number of non-exceedances, to circumvent this inflation the critical success index and the probability of detection are used. Both parameters perform similarly during the episode, yielding more accurate values when using a 2-km resolution (and thus, when exceedances of the 120 $\mu g/m^{-3}$ threshold taken as reference are more frequent). The value of bias (B < 1 for all simulations) indicates that exceedances are generally underpredicted for every resolution, which corresponds with the value of MNBE obtained for discrete evaluations, but this underprediction is minor for the 2 km/6 layers case (0.7) and the 2 km/16 layers case (0.3). Last, the fifth categorical parameter, the false alarm rate, indicated the number of times that the model predicted an exceedance that did not occur. This metric is higher for the simulations with inferior vertical resolution (above 70% for the 4 km/6 layers and the 8 km/6 layers cases), since of the possible influence of the vertical collapse of emissions can be high for the sum of precursor species for O_3. Nevertheless, values shown here agree with the results found by Kang *et al.* (2003).

In general, the maximum 1-hr concentration of O_3 (Figure 6) was overestimated by the 2-km resolution (Table 3), being underestimated both by the 8-km and the 4-km simulations. The error reduces substantially when incrementing the vertical grid resolution. The explanation for this behaviour is that model outputs indicate an average of the cell, diluting the very local effects of photochemical phenomena. Figure 6 depicts that an accurate definition of O_3 problem in the region is only achieved when observing the 2 km/16 layers case. Average values are underestimated by simulations when using both an 8-km, 4-km or 2-km grid size, exhibiting negative bias resolutions.

Figure 7 depicts the average values of tropospheric O_3 in the domain of study for the different horizontal and vertical resolutions. They present similarities in the prediction of the lowest values in the Barcelona Geographical Area, high values in the Pyrenees and capture O_3 depletion in the proximity of the Cercs thermal central. Nevertheless, 8-km and 4-km simulations do not show a good agreement with observations in the northeastern area of the domain, and therefore a 2-km resolution must be used. The depletion of O_3 during nighttime in Barcelona and the industrial area of Tarragona is only observed with resolutions of 4-km and finer, and is better captured with 16 vertical layers.

The effects of grid resolution on the chemistry of O_3 and NO_x are far more important than the chemistry of VOC (Jang *et al.*, 1995b). The predicted first layer O_3 and NO_x concentrations by 6- and 16-layer vertical resolutions are distinctively different for the whole domain. Ozone predictions from the 6-layer case show slightly higher values than those from the 16-layer model, meanwhile NO_x concentrations are significantly higher in the case of 16-layer vertical resolution (Figure 8). This can be explained by the fact that the VOC and NO_x emissions injected into the 6-layer model are diluted in a deeper layer compared to the same injected into the 16-layer model. Since most of the domain presents a VOC-limited sensitivity (Jiménez and Baldasano, 2004), increasing NO_x concentration in the first layer presents the contrary effect on tropospheric ozone, producing less ozone from the photochemical reaction and lowering ground-level O_3 concentrations.

Figure 6 One-hour maximum ozone levels (μg m^{-3}) in the simulation domain for an 8-km (up), 4-km (centre) and 2-km (down) horizontal resolution with six (left) and 16 (right) vertical layers for the episode of 13–16 August 2000

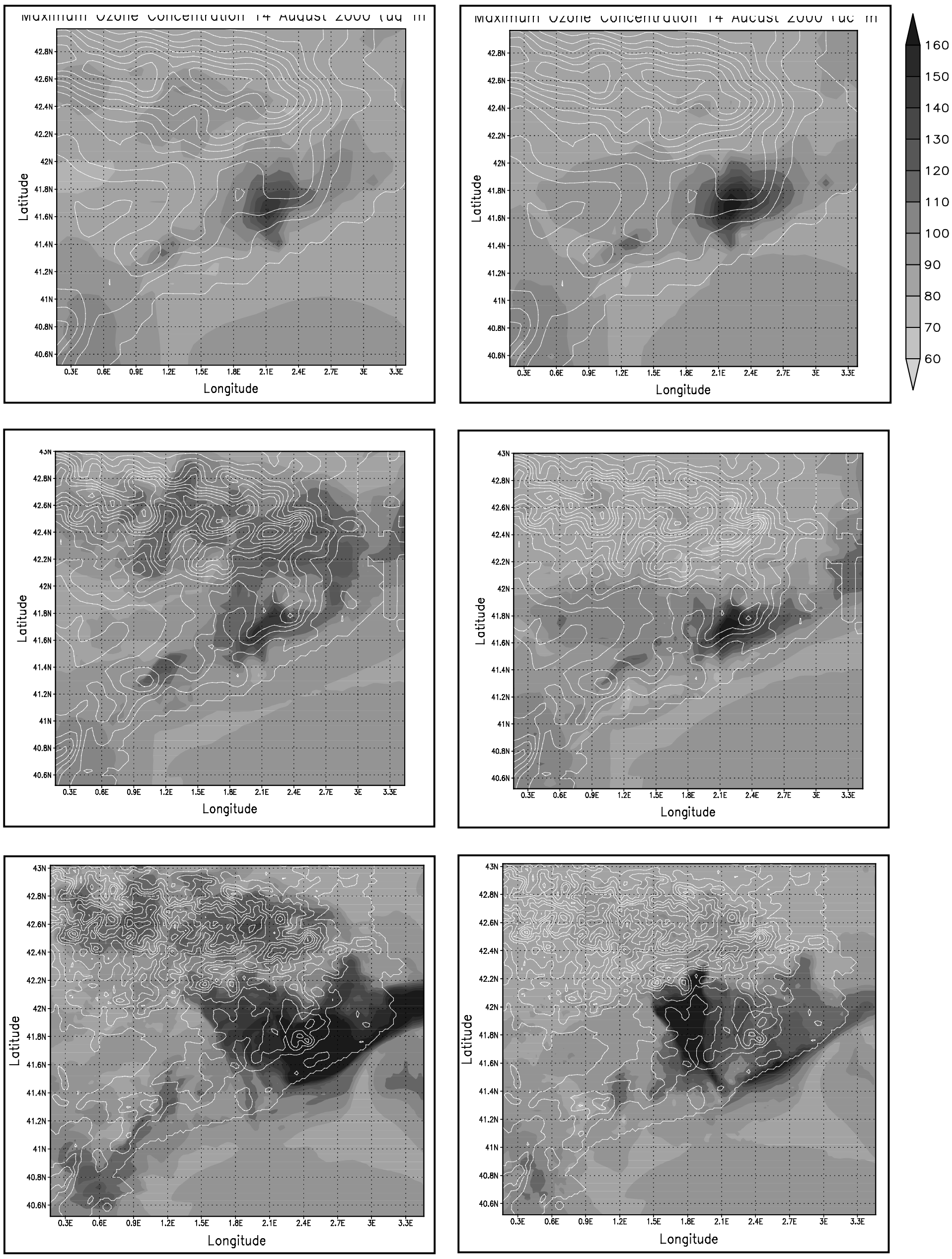


Figure 7 Average daily ozone levels (μg m^{-3}) in the simulation domain for an 8-km (up), 4-km (centre) and 2-km (down) horizontal resolution with 6 (left) and 16 (right) vertical layers for the episode of 13–16 August 2000

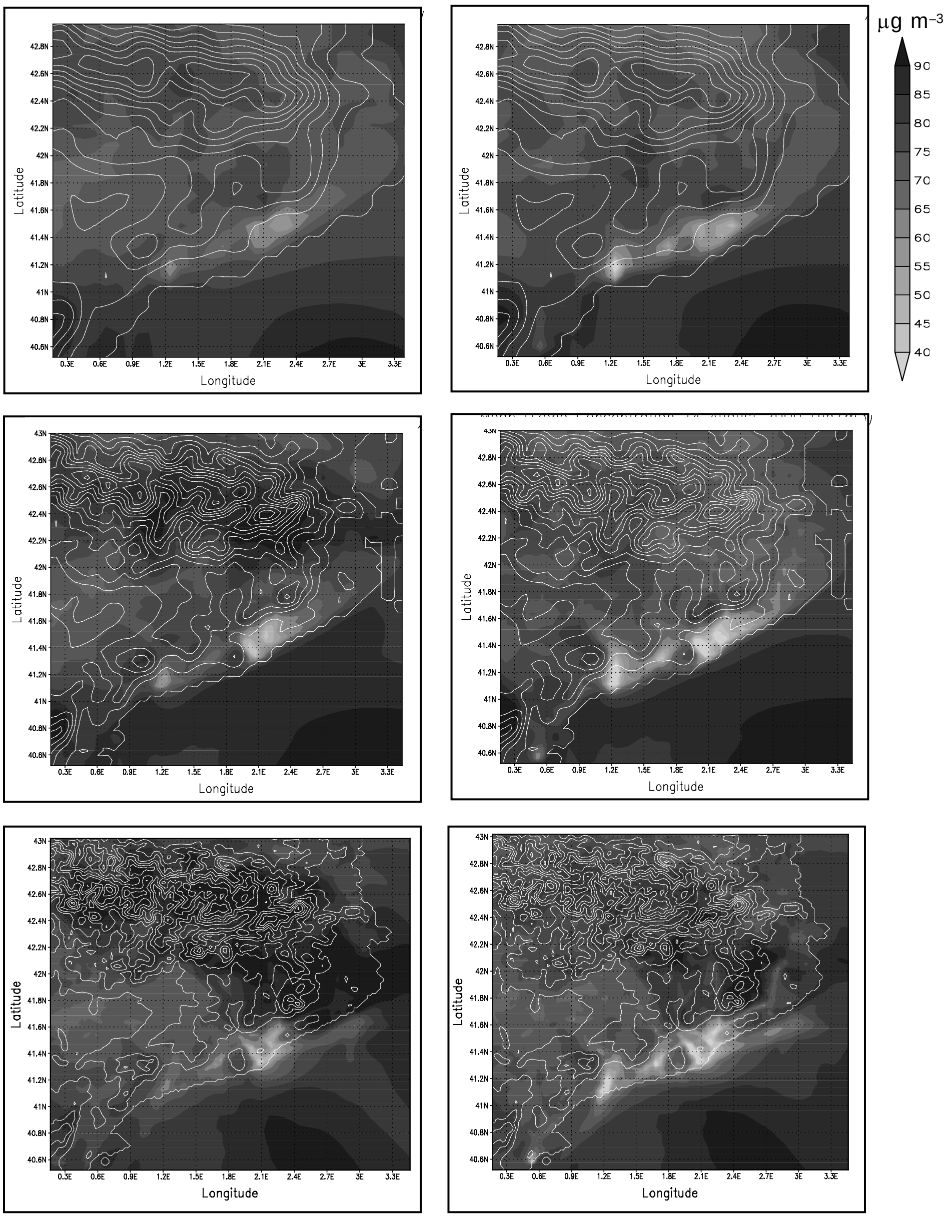
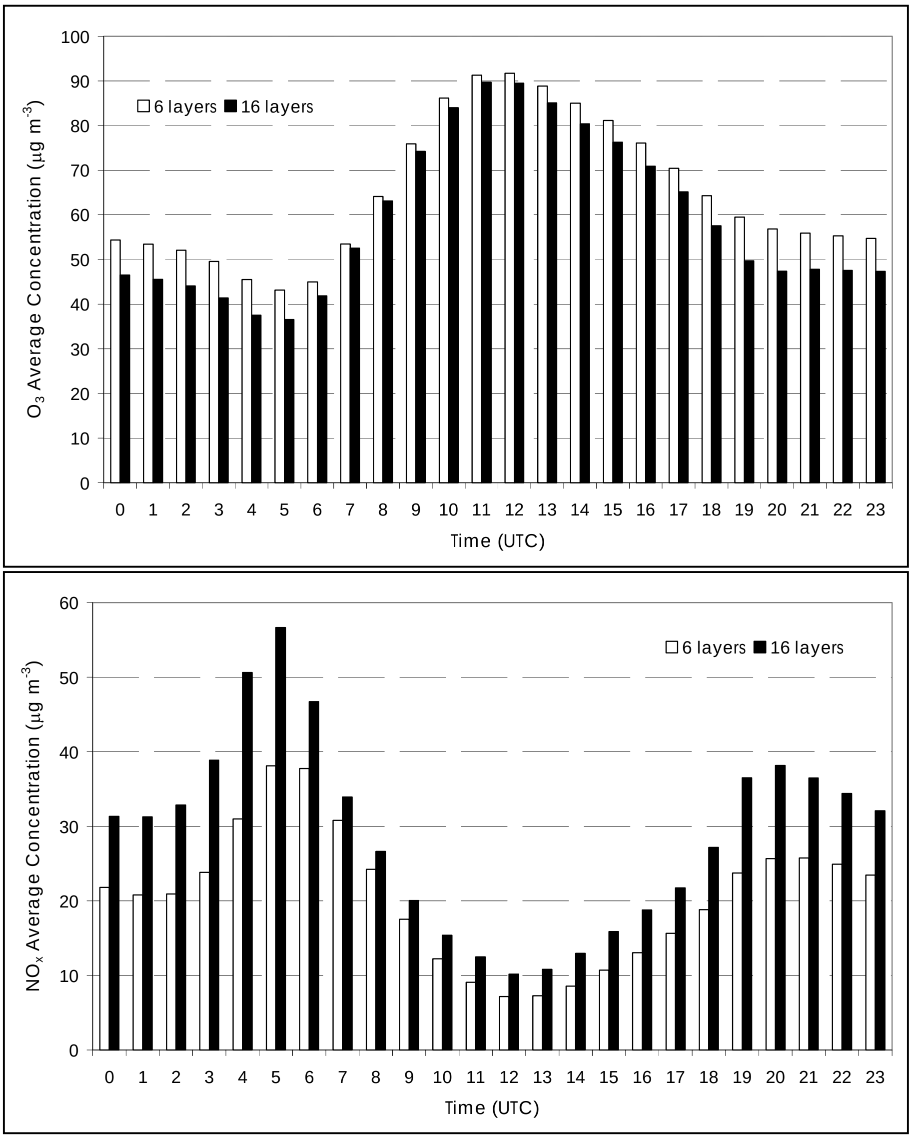


Figure 8 Average daily variation of ozone (up) and nitrogen oxides (down) for 6 and 16 vertical layers simulations in the domain of study, expressed in μg m^{-3}, for the episode of 13–16 August 2000



As shown in Table 4, coarser cell sizes are too large to correctly represent NO_x emissions, which have very distinctive grid-scale distributions. The result of photochemical reactions with diluted primary species concentrations does not give a similar dynamic range as observed ozone concentrations. In addition, performance of the model greatly improves when using 16 vertical layers instead of just six layers. The 8-km resolution tempers the concentrations of pollutants, yielding most of the values in a medium range, not capturing the extreme values (maximum and minimum levels of NO_x) that are more truthfully predicted by the finer resolution. We expect that the limited dynamic range in the daily ozone concentrations is related to the over-smoothing of NO_x emission rates in the model with coarse horizontal and vertical resolutions. NO_x species is more sensitive than ozone to model grid structure since secondary species have more horizontal homogeneity than primary species.

Table 4 Summary results for evaluation of nitrogen oxides concentrations with different horizontal and vertical resolutions [the statistics are defined by US-EPA (1991)]

Max Obs ($\mu g\ m^{-3}$)	*220*					
Max Sim ($\mu g\ m^{-3}$)	*149*	*247*	*154*	*190*	*80*	*141*
	2 km/ 6 layers	*2 km/ 16 layers*	*4 km/ 6 layers*	*4 km/ 16 layers*	*8 km/ 6 layers*	*8 km/ 16 layers*
MNBE (%)	–28.64	9.46	–23.34	–17.77	–52.32	–32.78
MNGE (%)	39.20	31.36	40.76	41.75	39.73	37.78
UPA (%)	–32.20	12.36	–30.13	–13.54	–63.86	–36.10

6 Conclusion

The effect of grid resolution on the results of an air quality model when applied to very complex terrains as the NEIP has been illustrated in this work. Simulation of the period from 13 to 16 August 2000 was used to depict the impact of grid resolution on photochemical pollution. For the complex domain studied, a clear improvement in the statistics of O_3 values has been observed when increasing model resolution from 8 km to 4 and 2 km; and from 6 to 16 vertical layers.

MM5 metorological simulations clearly show that the description of wind fields improves with finer grids. Results are very sensitive to the degree of topographical smoothing in this very complex terrain. The model is unable to reproduce the mountain-wind system with resolutions coarser than 4 km. Statistics of the 2-km simulation present the best behaviour during the development of the sea breeze, but all three simulations overestimate surface flows during the nocturnal period.

Because EMEP inventory can not be satisfactorily applied to the description of air pollutants dynamics in the NEIP because of its coarse resolution, it becomes essential to consider the emissions from this inventory in order to generate initial and boundary conditions to the domain of the NEIP through nested simulations. However, the very complex area considered in this study demands a finer resolution (1 km^2–1h). As a global amount, estimations from EMEP and EMICAT2000 are comparable and present similar results. Their main difference comes from the estimation of biogenic emissions since EMICAT2000 utilises specific emission factors for the area of study.

Outputs from the chemical transport model were sensitive to the grid size employed in the simulations, presenting a higher dependence on horizontal grid than on the vertical resolution when simulating O_3. Nevertheless, a high vertical resolution was found to be as important as horizontal resolution to properly simulate other photochemical pollutants such as NO_x. Since the dynamics of photochemical pollutants as ozone and the patterns of its precursors do not change dramatically in outline, some small-scale features appear when using a resolution of 2 km that cannot be captured with coarser horizontal resolutions. Meanwhile having a finer grid is important for addressing ozone processes in urban and industrial areas, rural areas allow larger grids to capture the nonlinearity of the ozone process.

If both discrete and categorical statistical parameters are compared with US-EPA's recommended values, a grid resolution of 2 km, both with six or 16 vertical layers, is needed when simulating photochemical pollution in the NEIP in order to ensure that results are inside the range of error tolerated. Nevertheless, the model should have enough vertical resolution (16 layers) in order to represent correctly the low-troposphere processes throughout the day.

Acknowledgements

This work was developed under the research contract REN2003–09753–C02 of the Spanish Ministry of Science and Technology. The Spanish Ministry of Education and Science is also acknowledged for the FPU doctoral fellowship held by P. Jiménez. The authors also gratefully acknowledge E. López for the implementation of EMICAT2000 into a GIS system. Stations data were provided by the Environmental Department of the Catalonia Government (Spain).

References

Baldasano, J.M., Cremades, L. and Soriano, C. (1994) 'Circulation of air pollutants over the Barcelona geographical area in summer', *Proceedings of the 6th European Symposium on Physico-Chemical Behaviour of Atmospheric Pollutants*, Report EUR 15609/1 EN: 474–479, Varese, Italy.

Barros, N., Toll, I., Soriano, C., Jiménez, P., Borrego, C. and Baldasano, J.M. (2003) 'Urban photochemical pollution in the Iberian Peninsula: the Lisbon and Barcelona Airsheds', *Journal of the Air and Waste Management Association*, Vol. 53, pp.347–359.

Byun, D.W. and Ching, J.K.S. (Eds.) (1999) 'Science algorithms of the EPA models-3 Community Multiscale Air Quality (CMAQ) modeling system', *EPA Report No. EPA-600/R-99/030*, Office of Research and Development, US Environmental Protection Agency, Washington, DC.

Byun, D.W. and Dennis, R. (1995) 'Design artifacts in Eulerian air quality models: evaluation of the effects of layer thickness and vertical profile correction on surface ozone concentrations', *Atmospheric Environment*, Vol. 29, pp.105–126.

European Environmental Agency (EEA) (2003) *EMEP/CORINAIR Emission Inventory Guidebook*, 3rd edition, September, Technical Report, Copenhagen, Denmark, No. 30.

Gery, M.W., Whitten, G.Z., Killus, J.P. and Dodge, M.C. (1989) 'A photochemical kinetics mechanism for urban and regional scale computer modeling', *Journal of Geophysical Research*, Vol. 94, No. D10, pp.12925–12956.

Guenther, A., Hewitt, C.N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W.A., Pierce, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J. and Zimmerman, P. (1995) 'A global model of natural volatile organic compound emissions', *Journal of Geophysical Research*, Vol. 100, pp.8873–8892.

Hogrefe, C., Rao, S.T., Kasibhatla, P., Hao, W., Sistla, G., Mathur, R. and McHenry, J. (2001) 'Evaluating the performance of regional-scale photochemical modeling systems: part II – ozone predictions', *Atmospheric Environment*, Vol. 35, pp.4175–4188.

Huang, H-C. and Chang, J.S. (2001) 'On the performance of numerical solvers for a chemistry submodel in three-dimensional air quality models', *Journal of Geophysical Research*, Vol. 106, pp.20175–20188.

Jang, C.J., Jeffries, H.E., Byun, D. and Pleim, J.E. (1995a) 'Sensitivity of ozone to model grid resolution – I. Application of high-resolution regional acid deposition model', *Atmospheric Environment*, Vol. 29, No. 21, pp.3085–3100.

Jang, C.J., Jeffries, H.E., Byun, D. and Pleim, J.E. (1995b) 'Sensitivity of ozone to model grid resolution – II. Detailed process analysis for ozone chemistry', *Atmospheric Environment*, Vol. 29, No. 21, pp.3101–3114.

Jiménez, P. and Baldasano, J.M. (2004) 'Ozone response to precursor controls in very complex terrains: use of photochemical indicators to assess O_3-NO_x-VOC sensitivity in the northeastern Iberian Peninsula', *Journal of Geophysical Research*, Vol. 109, D20309, doi: 10.1029/2004JD004985.

Jiménez, P., Dabdub, D. and Baldasano, J.M. (2003) 'Comparison of photochemical mechanisms for air quality modeling', *Atmospheric Environment*, Vol. 37, pp.4179–4194.

Jorba, O., Pérez, C., Rocadenbosch, F. and Baldasano, J.M. (2004) 'Cluster analysis of 4-day back trajectories arriving in the Barcelona area (Spain) from 1997 to 2002', *Journal of Applied Meteorology*, Vol. 43, No. 6, pp.887–901.

Kain, J.S. and Fritsch, J.M. (1993) 'Convective parameterisation for mesoscale models: the Kain-Fritsch scheme. The representation of cumulus convection in numerical models', in K.A. Emanuel and D.J. Raymond (Eds.) *American Meteorological Society*, p.246.

Kang, D., Eder, B.K. and Schere, K.L. (2003) 'The evaluation of regional-scale air quality models as part of NOAA's air quality forecasting pilot program', *26th Nato International Technical Meeting on Air Pollution Modeling and its Application*, Istanbul, Turkey, 26–30 May, pp.404–411.

Lyons, W.A., Pielke, R.A., Tremback, C.J., Walko, R.L., Moon, D.A. and Keen, C.S. (1995) 'Modeling the impacts of mesoscale vertical motions upon coastal zone air pollution dispersion', *Atmospheric Environment*, Vol. 29, pp.283–301.

McQueen, J.T., Draxler, R.R. and Rolph, G.D. (1995) 'Influence of grid size and terrain resolution on wind field predictions from an operational mesoscale model', *Journal of Applied Meteorology*, Vol. 34, No. 10, pp.2166–2181.

Millán, M., Salvador, R., Mantilla, E. and Kallos, G. (1997) 'Photooxidant dynamics in the Mediterranean basin in summer: results from European research projects', *Journal of Geophysical Research*, Vol. 102, pp.8811–8823.

Millán, M.M., Artiñano, B., Alonso, L., Castro, M., Fernandez-Patier, R. and Goberna, J. (1992) 'Mesometeorological cycles of air pollution in the Iberian Peninsula', *Air Pollution Research Report 44*, Commission of the European Communities, Brussels, Belgium, p.219.

MMMD/NCAR (2001) *PSU/NCAR Mesoscale Modeling System Tutorial Class Notes and User's Guide: MM5 Modeling System Version 3*.

Ntziachristos, L. and Samaras, Z. (2000) 'COPERTIII computer programme to calculate emissions from road transport. Methodology and emission factors (version 2.1), *Technical Report*, European Environment Agency, No. 49.

Parra, R. (2004) 'Development of the EMICAT2000 model for the estimation of air pollutants emissions in Catalonia and its use in photochemical dispersion models', *PhD Dissertation*, in Spanish, Polytechnic University of Catalonia, Spain.

Parra, R. and Baldasano, J.M. (2004) 'Modelling the on-road traffic emissions from Catalonia (Spain) for photochemical air pollution research. Weekday – weekend differences', *12th International Conference on Air Pollution (AP'2004)*, Rhodes, Greece.

Parra, R., Gassó, S. and Baldasano, J.M. (2004) 'Estimating the biogenic emissions of non-methane volatile organic compounds from the North Western Mediterranean vegetation of Catalonia, Spain', *The Science of the Total Environment*, Vol. 329, pp.241–259.

Pérez, C., Sicard, M., Jorba, O., Comerón, A. and Baldasano, J.M. (2004) 'Summertime re-circulations of air pollutants over the north-eastern Iberian coast observed from systematic EARLINET lidar measurements in Barcelona', *Atmospheric Environment*, Vol. 38, pp.3983–4000.

Pielke, R.A. and Uliasz, M. (1998) 'Use of meteorological models as input to regional and mesoscale air quality models – limitations and strengths', *Atmospheric Environment*, Vol. 32, No. 8, pp.1455–1466.

Russell, A. and Dennis, R. (2000) 'NARSTO critical review of photochemical models and modeling', *Atmospheric Environment*, Vol. 34, pp.2283–2324.

Salvador, R., Calbó, J. and Millán, M.M. (1999) 'Horizontal grid size selection and its influence on mesoscale model simulations', *Journal of Applied Meteorology*, Vol. 38, pp.1311–1329.

Simpson, D., Guenther, A., Hewitt, C. and Steinbrecher, R. (1995) 'Biogenic emissions in Europe 1. Estimates and uncertainties', *Journal of Geophysical Research*, Vol. 100, No. D11, pp.22875–22890.

Soriano, C., Baldasano, J.M. and Buttler, W.T. (1997) 'On the application of meteorological models and lidar techniques for air quality studies at a regional scale', in S-E. Gryning and N. Chaumeliac (Eds.) *Air Pollution Modeling and its Application XII*, Plenum Press, pp.591–600.

Soriano, C., Baldasano, J.M., Buttler, W.T. and Moore, K. (2001) 'Circulatory patterns of air pollutants within the Barcelona air basin in a summertime situation: lidar and numerical approaches', *Boundary-Layer Meteorology*, Vol. 98, No. 1, pp.33–55.

Tesche, T.W., Lurmann, F.L., Roth, P.M., Georgopoulos, P., Seinfeld, J.H. and Cass, G. (1990) 'Improvement of procedures for evaluating photochemical models', *Report to California Air Resources Board for Contract No. A832-103*, Alpine Geophysics, LLC, Crested Butte, Colorado.

Thunis, P. (2001) 'The influence of scale on modeled ground level O_3 concentrations', *Norwegian Meteorological Institute Research Report*, EMEP/MSC-W Note 2/01, Copenhagen, Denmark, No. 57, p.42.

US-EPA (1991) 'Guideline for regulatory application of the urban airshed model', *US EPA Report*, Office of Air and Radiation, Office of Air Quality Planning and Standards, Technical Support Division, Research Triangle Park, North Carolina, US, No. EPA-450/4-91-013.

Note

1 www.emep.int