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Development of the high spatial resolution EMICAT2000 emission model for air pollutants from the north-eastern Iberian Peninsula (Catalonia, Spain)

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A high spatial resolution and multipurpose emission model is presented.

Abstract

Ozone (O_3) pollution episodes take place in Catalonia (NE of the Iberian Peninsula), mainly during summertime. The complex O_3 behaviour could be understood by using a Chemical Transport Model (CTM). Emission inventories provide the spatial and temporal emissions distribution of the O_3 precursors and other pollutants required by this approach. We developed the EMICAT2000 model with high spatial (cells of 1 km^2) and temporal (1 h) resolutions, to estimate the emissions during the year 2000 from Catalonia. Total annual emissions were 107 kt yr^{-1} of NO_x, 137 kt yr^{-1} of NMVOC, 267 kt yr^{-1} of CO, 65 kt yr^{-1} of SO_2, 24 kt yr^{-1} of TSP and 32 175 kt yr^{-1} of equivalent CO_2. Main NO_x sources are on-road traffic (58%) and industries (38%). Main NMVOC sources are on-road traffic (36%), vegetation (34%) and use of solvents (13%). Speciation was established according to the Carbon Bond IV mechanism. EMICAT2000 generates directly the data files required for the third generation CTM Models-3/CMAQ.

Keywords: Biogenic; Traffic; Industries; Speciation; High resolution; Catalonia

1. Introduction

Air pollutants emission inventories provide the necessary information required both for environmental policy and modelling issues (Van Aardenne, 2002). Emission inventories are the figures representing the amount of air pollutants emitted to the atmosphere from a zone (local, regional or global scales) during a specific period of time (pass, present or future), due to anthropogenic or natural activities. Emission models are the mathematical representations providing emission inventories of primary air pollutants (PAP), those emitted directly from sources.

In the policy arena, emission inventories have crucial roles and typically are required to monitor the achievement of emission trends over time, to take control of pollution actors.

Nowadays, emission inventories are under development and constant improvement worldwide. Within the ESCOMPTE program (ESCOMPTE, 2004), anthropogenic and biogenic sources from Marseille (France) were considered for a high spatial resolution (cells of 1 km^2) emission inventory. In Europe, the EMEP/CORINAIR annual emission databases (EMEP, 2004) are built with cells about 50 × 50 km^2, according to 11 source categories. At a global level, EDGAR (EDGAR,

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2004) and GEIA (GEIA, 2004) are world inventories developed with $1^\circ \times 1^\circ$ grid resolution.

Inventories for modelling demand spatial distribution and time resolution of real emissions, being one of the key elements required by a Chemical Transport Model (CTM) (Russell and Dennis, 2000). These inventories are quantified by the top–down approach (firstly describing total emissions estimated from statistical data throughout the whole region and after disaggregating them spatially and temporarily using variables assumed to have a similar distribution) or the bottom–up approach (using own information for each cell of the domain and obtaining total emissions by summing all the contributions) (Costa and Baldasano, 1996; Colvile et al., 2001).

The basic model includes the product of two variables, according to Eq. (1):

$$E_{i,j} = A_{i,j} \mathrm{EF}_{i,j} \quad (1)$$

where $E_{i,j}$ is the emission of the pollutant j due to the activity i; $A_{i,j}$ is the level of the activity i producing the pollutant j; and $\mathrm{EF}_{i,j}$ is the respective emission factor. More developed emission models integrate this basic model into more complex mathematical algorithms.

Emission inventories are estimations of real emissions. Differences between them result in a collection of inaccuracies (like the own complexity of emitter activities, poor quality of emission factors or activity data or simplification of emission models), whose assessment is considered as the uncertainty analysis (Van Aardenne, 2002).

In Catalonia (Spain), located in the NE of the Iberian Peninsula (Fig. 1), there are nowadays more than 6 millions inhabitants, and about two thirds are located in the Metropolitan Area of Barcelona (MAB). It has a complex topography, showing strong changes of relief, climate and vegetation, into a relative short territory. Catalonia industrial activity is one of the most important from Spain. Also, traffic and consumption of fossil fuels have important levels. Hence air pollutants' emissions due to anthropogenic activities are substantial, and are related with the degradation of air quality. More than 1900 ozone (O_3) exceedances in the legislative hourly concentration threshold for people information (180 $\mu g\,m^{-3}$) were recorded in the period 1991–2003, and almost 80% of these happened into the period June–August. O_3 measurements at any given location may have contributions from different sources as background concentration, long-range and regional transport, or local production. Also, O_3 could be transported from the stratosphere. This complex phenomenon may be understood by modelling.

Hourly resolution is necessary to describe the profiles of emission activities (biogenic and traffic specially) and to track the O_3 daily cycle. Also, hourly averages of the wind are enough for modelling purposes (Russell and Dennis, 2000). In region with complex topography and important variations of the land-use categories, high spatial resolution (cells of 1–2 km) should be used to adequately describe the wind fields (Salvador et al., 1999; Seaman, 2000; Jiménez et al., 2004).

Among the most important previous contributions as high spatial and temporal emission inventories, the EMITEMA-EIM emission model (Costa and Baldasano, 1996) provided anthropogenic inventories for the area of Barcelona ($39 \times 39\ km^2$) during the year 1990. This contribution covered only a small portion of industries from Catalonia. The most important industries with regard to air emissions (as oil refineries, power generation or cement production) are outside from MBA. Gómez and Baldasano (1999) presented an emission inventory due to vegetation in Catalonia for the year 1992 based on climatologic information for temperature and solar radiation as driven forces, and reported emission factors. Delgado et al. (2000) extended EMITEMA-EIM for Catalonia and presented an estimation of on-road traffic emission during the year 1994.

During last years scientific knowledge (allowing the use of better emission models) and quality of emission factors, activities and information data have improved. Also, fossil fuel consumption increased with time and the park of vehicles is under systematic evolution (more vehicles demanding more fuels, but cleaner) showing complex and dynamic traffic profiles. Hence it is necessary to have updated emission inventories for Catalonia.

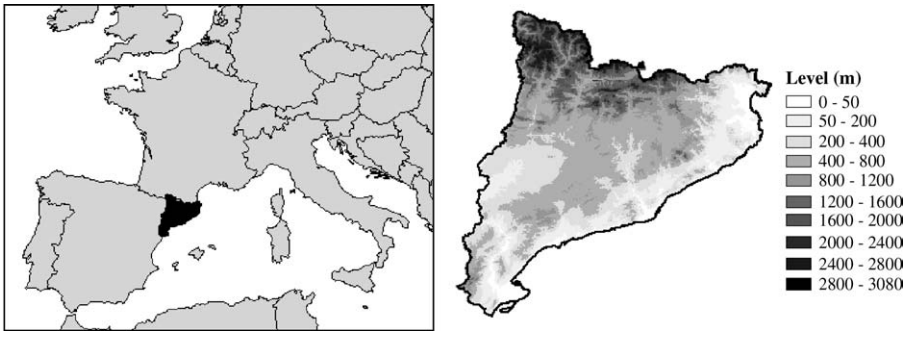

Fig. 1. Location of Catalonia.

We developed the *EMICAT2000 (Air Pollutants Emission from Catalonia during the year 2000)* emission model. We describe the approach followed, the results and model features, with regard to speciation and management of emission files to be directly used within a third generation CTM.

2. Methods

EMICAT2000 was developed giving priority to the emissions of O_3 precursors (NO_x and NMVOC), and also PAP: carbon monoxide (CO), sulphur dioxide (SO_2) and total suspended particles (TSP) were included. As GHG, carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) were included. The sources considered are: on-road traffic, industries, biogenic emission and use of fossil fuels and solvents by residential and commercial sectors.

The domain was divided in square cells of 1 km^2. The temporal base resolution is 1 h. A squared zone of 272×272 km^2 including the whole Catalan territory was selected.

2.1. On-road traffic

On-road traffic is a significant source of air pollutants. It was the most important source of VOC, NO_x and CO (56%, 74% and 99%, respectively) in the Barcelona area for year 1990 (Costa and Baldasano, 1996). In the European Union-15 States (EU 15), during the year 2000 traffic emitted 39% of NO_x, 26% of NMVOC and 62% of CO emissions (ACEA, 2004).

There are three principal methods to estimate on-road traffic emissions, which vary mainly in the way that they treat the interaction between vehicle operation and the corresponding emissions (MEET, 1999). The first one is the most straightforward and only takes into account the total number of kilometres driven by the whole fleet. This approach is normally used for top–down national yearly inventories. The second method is an average speed-dependent approach and exploits the fact that average emissions over a trip vary according to the average speed of the trip. The third approach, more complex, is called instantaneous vehicle emissions (modal modelling) and takes into account the speed variation along the trip. The potentially better description obtained by instantaneous speeds (if the information is available) is not completely exploited, since a CTM typically requires hourly integrated emission data. Hence, the average speed-dependent approach is enough for modelling purposes.

Using the bottom–up approach, we built an updated digital map of all the highways and most important roads and streets (daily average traffic (DAT) > 3000). Three kinds of emissions were included: (1) *hot exhaust emissions*, occurring under thermally stabilised engine and exhaust after treatment conditions, (2) *cold exhaust emissions*, occurring during transient thermal engine operation (cold start), and (3) *evaporative emissions*, non-exhaust VOC emission relevant for gasoline fleet. Also, we included non-exhaust particles' emissions (tire and brake wear, road abrasion).

Hot exhaust emissions were estimated using Eq. (2):

$$E_r^{i\text{hot}}(k,d) = \sum_{j=1}^{n} \text{Clf} \times \text{Crd} \times \text{DAT}_{rj}(k) L_r(k) F_j^{i\text{hot}}(s_r) \quad (2)$$

where $E_r^{i\text{hot}}(k, d)$ (expressed in g d^{-1}) is the hot exhaust emission of the pollutant i during one day (weekday or weekend) in the road section r (urban, rural or highway type) that is allocated into the cell kth, $F_j^{i\text{hot}}(s_r)$ (expressed in g km^{-1} a function of the speed s_r) is the hot emission factor of the pollutant i for the vehicle category j, $L_r(k)$ (expressed in km) is the length of the road section r, $\text{DAT}_{rj}(k)$ (expressed as number of vehicles per day) is the daily average traffic of the vehicle category j, Clf is the coefficient for daily traffic (weekday or weekend), Crd is the ratio between daily traffic for a specific month and DAT; and n (36) is the number of vehicle categories considered by EMICAT2000.

Hourly emissions were estimated applying coefficients to the daily emissions. Monthly emissions were obtained adding up the respective daily values. The same procedure was used for annual values.

Cold exhaust emissions seem to be most likely for urban driving. They occur for all vehicle categories, but emission factors can actually be reasonably estimated for gasoline and diesel passenger cars (Ntziachristos and Samaras, 2000). Hourly cold emissions were established as additional emissions using Eq. (3):

$$E_r^{i\text{cold}}(k,h) = \sum_{j=1}^{n} E_r^{i\text{hot}}(k,h)\beta(\text{ltrip},\text{at}) \left(\frac{F_j^{i\text{cold}}}{F_j^{i\text{hot}}}(\text{at}) - 1 \right) \quad (3)$$

where $E_r^{i\text{cold}}(k, h)$ (expressed in g h^{-1}) is the hourly cold emission of the pollutant i (only for urban roads), β (ltrip, at) (function of the average trip length ltrip and ambient temperature at) is the fraction of the mileage driven with cold engines or catalyst operated below the light-off temperature, $F_j^{i\text{cold}}/F_j^{i\text{hot}}(\text{at})$ is the cold over hot ratio of pollutant i emission.

Evaporative emissions were estimated by the Standard CORINAIR method (Ntziachristos and Samaras, 2000). There are three primary sources of evaporative emissions: (1) *diurnal* (*daily*) *emissions*, associated with daily variation in ambient temperature that result in vapour expansion inside the gasoline tank; (2) *hot soak emissions*, caused when a hot engine is turned off, increasing the temperature of the fuel (as into the carburettor) no longer flowing; and (3) *running losses*, that

result of vapour generated in gasoline tanks during vehicle operations.

Total daily diurnal emissions were estimated using Eq. (4):

$$E_m^{\text{devap}}(\text{daily}) = N_m\left(e_m^d(t_{\min}, t_{\max}, \text{RVP})\right) \quad (4)$$

where E_m^{devap} (daily) (expressed in g d^{-1}) is the total daily VOC diurnal evaporative emission produced by the vehicles of the category m, N_m is the number of vehicles of the fleet of Catalonia belonging to the category m, e_m^d (g VOC d^{-1}) is the diurnal emission factor (expressed in g VOC d^{-1}) which is a function of the minimum and maximum ambient temperatures ($t_{\min}$, $t_{\max}$) and the gasoline volatility (measured by its *reid vapour pressure* (RVP).

Total daily hot soak emissions were estimated using Eq. (5):

$$E_m^{\text{sevap}}(\text{daily}) = N_m\left(px_m e_m^{\text{shot}}(\text{at}, \text{RVP}) + wx_m e_m^{\text{swarm}}(\text{at}, \text{RVP})\right) \quad (5)$$

where p is the fraction of trips finished with hot engine, w is the fraction of trips finished with cold or warm engine or with catalyst below its light-off temperature, x_m is the mean number of trips of the vehicles type m, e_m^{shot} (at, RVP) (expressed in g trip^{-1}) is the emission factor for hot soak emissions, e_m^{swarm} (at, RVP) (expressed in g trip^{-1}) is the emission factor for cold and warm soak emissions.

Total hourly diurnal and soak emissions were established, dealing the daily value with averaged hourly values toward the average daily ambient temperature. Hourly diurnal emissions were spatially disaggregated according to the traffic volume of each vehicle category and population density.

Running losses emission factors were obtained using Eq. (6). Daily and hourly running losses were estimated using equivalent equations as the hot exhaust emissions.

$$F_j^{\text{revap}} = \left(px_j e_j^{\text{rhot}}(\text{at}, \text{RVP}) + wx_j e_j^{\text{rwarm}}(\text{at}, \text{RVP})\right) \quad (6)$$

where F_j^{revap} is the running losses VOC emission factor (expressed in g km^{-1}) for the vehicle category j (the same considered for cold emissions), e_j^{rhot} (at, RVP) (expressed in g km^{-1}) is the emission factor for hot running losses (g km^{-1}), and e_j^{rwarm} (at, RVP) is the warm running losses.

Daily average traffic information, vehicle park composition by type of road (differencing between weekday and weekend profiles), average speeds, temperature information, timing traffic profiles (monthly, daily and hourly) and fuel properties were collected from different sources.

Exhaust emission factors for NO_x, VOC, CO, TSP (for diesel vehicles) were obtained from the top–down European model EMEP/CORINAIR – COPERTIII (Ntziachristos and Samaras, 2000; EEA, 2003). Particles exhaust emission factors for gasoline vehicles and particles non-exhaust emission factors were obtained from CEPMEIP (2003). Full description information required by the equations, modelling hypothesis, and exploitation of results (weekday vs. weekend on-road traffic emissions) are described in Parra and Baldasano (2004), Parra (2004) and Jiménez et al. (2005).

2.2. Industrial activities

In Catalonia, industrial sector covers a wide variety and numerous production centres related with the mineral, pharmaceutical, chemical, metallurgical, textile and food industry, among others. Also, the power generation based on fossil fuels consumption represents an important component (25% for the year 2000).

Two kinds of emissions are distinguished: (1) emissions related with combustion (energy use), and (2) emissions related with the industrial processes themselves.

EMICAT2000 includes about 170 industrial centres (Fig. 2). The Tarragona industrial centre is especially important because there are allocated two refineries and numerous chemical plants, among other industrial establishments.

We included hourly real records for 12 big chimneys, which were connected to the emission network monitoring of the Environmental Department of the Catalonia Government. Other industrial emissions were estimated using the basic model; e.g. Eq. (7) is the emission model used for power generation.

$$E_{ij}(k,h) = 1000000 F_{hj} F_{dj} F_{mj} \text{PG}_j(k) \text{EF}_{ij} \quad (7)$$

where $E_{ij}(k, h)$ (g h^{-1}) is the hourly emission of the pollutant i due to the power generation of the central j, which is allocated in the kth cell; F_{hj}, F_{dj} and F_{mj} are the hourly, daily and monthly generation fractions; $\text{PG}_j(k)$ is the annual power generation (GWh yr^{-1}) and EF_{ij} is the respective emission factor (g kWh^{-1}).

The diversity and complexity of industries were reflected in the difficulty in collecting information. Available statistic data do not have the required detail. Also some production information is considered confidential and centres attitude giving information was poor.

Emission factors were chosen mainly from EEA (2003), USEPA (2003) and IPCC (1996). EMICAT2000 includes the most important centres in relation with energy consumption and O_3 precursor emissions.

2.3. Vegetation

Part of the carbon fixed by photosynthesis is emitted to the atmosphere as NMVOC, previously synthesized into the vegetal tissues through complex physiological processes. This kind of emission is the result of a

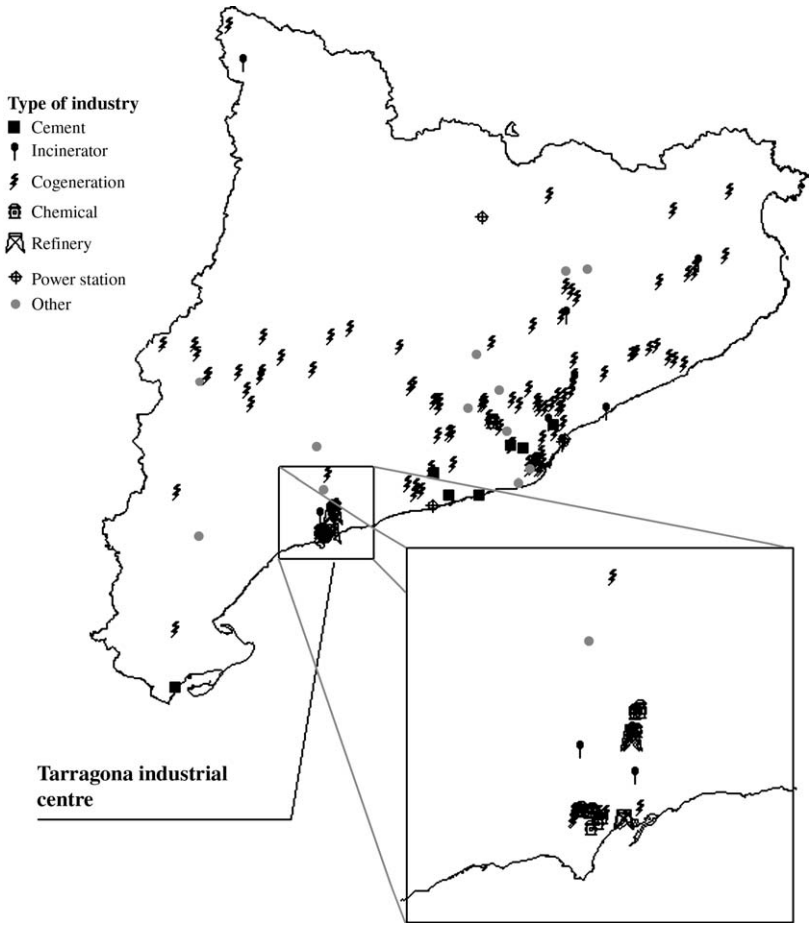

Fig. 2. Industrial sources included in EMICAT2000.

complex net of interactions including both internal (genetic and biochemical) and external factors (abiotic-temperature, solar radiation, water availability, wind, ozone and biotic-animal, plant and microorganism interactions) (Peñuelas and Llusià, 2001).

Emission model includes *isoprene*, *monoterpenes*, and *other volatile organic compounds (OVOC)*, considering the influence of temperature and solar radiation by the Guenther et al. (1993) algorithms. Emission factors by land-use categories were defined, as a result of an exhaustive selection of emission factors of the most important vegetal species. A huge database of hourly records of superficial temperature and solar radiation was incorporated.

Isoprene emission model is described by Eq. (8):

$$E_{\mathrm{iso}}(k,h) = \mathrm{EF}_j^{\mathrm{iso}}\mathrm{ECF}(T,P)\mathrm{FBD}_j A \tag{8}$$

where A is the area of each grid cell (1 km^2), FBD_j is the foliar biomass density (g m^{-2}) of the j land-use category, ECF(T,P) is the environmental correction factor (adimensional) owing to temperature and photosynthetically active radiation, $\mathrm{EF}_j^{\mathrm{iso}}$ is the standard isoprene emission factor (μg g^{-1} h^{-1}) and $E_{\mathrm{iso}}(k, h)$ is the hourly (g h^{-1}) isoprene emission.

Hourly monoterpene emissions were estimated partially by Eq. (9):

$$E_{\mathrm{mon}}(k,h) = \mathrm{EF}_j^{\mathrm{mon}} M(T)\mathrm{FBD}_j A \tag{9}$$

where $\mathrm{EF}_j^{\mathrm{mon}}$ is the standard monoterpenes emission factor (μg g^{-1} h^{-1}) and $E_{\mathrm{mon}}(k, h)$ is the hourly emission (g h^{-1}). OVOC emissions were estimated by a similar equation to the one used for monoterpenes.

Previous works (Hansen and Seufert, 1996; Atkinson and Arey, 1998; Kesselmeier et al., 1998) have elucidated the light-dependent monoterpene emissions of some Mediterranean species (*Quercus coccifera*, *Quercus ilex*). Hence, isoprene algorithm was used for monoterpene emissions from these species. Detailed explanation and results obtained from this source is described in Parra et al. (2004).

2.4. Use of fossil fuels and solvents by residential and commercial sectors

Annual total emissions were estimated using the basic emission model. Fossil fuels' emissions are based on statistical consumption. Emissions due to use of solvents were established by per capita emission factors of paint

application, glues, adhesives, household, toiletries, propellant and car care products use. Spatial allocation was made using a population density map. Temporal allocation was made by monthly, daily and hourly profiles.

3. Results and discussion

3.1. Annual emission

Table 1 shows a summary of annual emissions by sources. PAP reached 599 kt yr^{-1}, corresponding to 267 kt of CO (45%), 137 kt of NMVOC (23%), 107 kt of NO_x (18%), 65 kt of SO_2 (11%) and 23 kt of TSP (4%). On-road traffic is the most important source of CO (97%). NMVOC sources are more diverse, representing on-road traffic a 36%, 34% due to vegetation, 17% to industries and 13% to the use of solvents. Within the industrial sector, 8% corresponds to fugitive emissions from different production processes and 7% to the oil refining and olefins production.

With regard to NO_x, 58% were emitted by on-road traffic and 39% by industries. Last percentage includes 14% from power generation, 9% from cement production and 8% from oil refining.

Industries generated 94% of SO_2 emissions, belonging 44% to power generation, 39% to oil refining and 10% to olefins production.

On-road traffic produced 67% of the TSP and industry 32%.

By sources, on-road traffic emitted 65% of PAP, industry 23%, and vegetation 8%.

GHG emission was 32 175 kt yr^{-1}, belonging 63% to industry, 26% to on-road traffic and 11% to fossil fuels use by residential and commercial sectors.

Table 1 also shows the ratio of PAP to GHG. It is highest to on-road traffic (4.7%, each 100 kt of GHG emitted are associated to 4.7 kt of PAP) which resulted as the most polluted exploitation of fossil fuels. Medium percentages (0.8, 1.4, 1.8%) correspond to power generation, oil refining and olefins production. The lowest values (0.2%) correspond to cement production and fossil fuels use by residential and commercial sectors. Nevertheless, cement production ratio includes an important fraction of CO_2 ($\approx 2/3$) that is not from fuel.

Fig. 3 shows the annual CO emission distribution. Higher values are mainly located on the MAB and on

Table 1
Summary of emissions by sources in Catalonia during the year 2000

Source		Primary air pollutants (kt yr^{-1})						Greenhouse gases (kt CO_2 eq yr^{-1}) (*b*)	(*a*)/(*b*) %
		NO_x	NMVOC	CO	SO_2	TSP	Total (*a*)		
1.	Vegetation		46.9				46.9		
2.	On-road traffic	62.4	49.5	259.0	1.3	15.7	387.9	8302.0	4.7
3.	Industrial emissions	41.2	22.8	7.3	61.1	7.5	139.9	20 361.0	0.7
	Power generation	15.0	0.7	2.1	28.5	1.7	48.0	5698.0	0.8
	Cement	9.6	0.3	3.5	1.4	4.2	19.0	8477.0	0.2
	Oil refineries	8.5	6.4	1.1	25.0	0.9	41.9	2929.0	1.4
	Olefines	2.9	2.4	0.0	6.2	0.7	12.2	690.0	1.8
	Fugitives emissions		11.0				11.0		
	Other	5.2	2.0	0.6	0.0	0.0	7.8	2567.0	0.3
4.	Fossil fuels use – residential and commercial	3.3	0.2	1.1	2.3	0.3	7.2	3512.0	0.2
5.	Use of solvents		17.2				17.2		
	Total	106.9	136.6	267.4	64.7	23.5	599.1	32 175.0	1.9
	%	17.84	22.80	44.64	10.80	3.92	100		

Source		Primary air pollutants (%)						Greenhouse gases (%)
		NO_x	NMVOC	CO	SO_2	TSP	Total	
1.	Vegetation		34.33				7.83	
2.	On-road traffic	58.37	36.24	96.86	2.01	66.81	64.75	25.80
3.	Industrial emissions	38.54	16.69	2.73	94.44	31.91	23.35	63.28
	Power generation	14.03	0.51	0.79	44.05	7.23	8.01	17.71
	Cement	8.98	0.22	1.31	2.16	17.87	3.17	26.35
	Oil refineries	7.95	4.69	0.41	38.64	3.83	6.99	9.10
	Olefines	2.71	1.76	0.00	9.58	2.98	2.04	2.14
	Fugitives emissions		8.05				1.84	
	Other	4.86	1.46	0.22	0.00	0.00	1.30	7.98
4.	Fossil fuels use – residential and commercial	3.09	0.15	0.41	3.55	1.28	1.20	10.92
5.	Use of solvents		12.59				2.87	
	Total	100	100	100	100	100	100	100

the axis of highways following the coastline. Fig. 4 depicts the annual isoprene emission, which is mainly produced by shrub lands, coniferous and deciduous forest.

Table 2 shows a comparative summary of annual emissions provided by EMICAT2000 with other emission inventories. One of the elements for comparison is the emission inventory of PAP for the year 2000, obtained from the European inventory emission EMEP (EMEP, 2004). We identified the cells belonging to Catalonia from the coarser European EMEP mesh (cells about 50 km by side), which enabled us to assign the emission according to the 11 sectors of the EMEP inventories.

The annual EMICAT2000 vegetation emission (46.9 kt of NMVOC) is 10% lower than the estimation by Gómez (1998). This apparent agreement does not imply that the two estimations are equivalent. In fact, the composition in terms of isoprene, monoterpenes and OVOC, is different, therefore, involving different superficial patterns in emissions.

EMICAT2000 vegetation emission is a 52% of the EMEP sector 11 (Nature) NMVOC in the European cells where Catalonia is allocated.

In EEA (2003) a summary of vegetation emissions at a European level is included. 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 Catalonia (31 895 km^2) and Spain (504 782 km^2), we obtained 41.4 kt yr^{-1}. This value is equivalent to the annual estimation provided by EMICAT2000 (46.9 kt yr^{-1}).

With regard to on-road traffic, EMICAT2000 and the EMEP sector 7 (road traffic) estimations are equivalents. In fact the NO_x, CO and VOC emissions provided by EMICAT2000 are 74, 95 and 87%, respectively, of the EMEP values. Subsequent inclusion of other roads in EMICAT2000 (especially those with DAT less than 3000) will reduce these differences. The estimation provided by DMA (2003) was established by statistic data of fuel consumption (top–down estimation) and EMEP/CORINAIR emission factors. In comparison with EMICAT2000, NO_x estimations are equivalent but there are differences for other pollutants.

PAP emissions due to on-road traffic lowered during the 1990–2000 period. EMICAT2000 NO_x emission (62.4 kt yr^{-1}) is 67% of the 1990 value. This tendency is coherent with European emission trends during the same period, showing reductions estimated in 65–70% (Ntziachristos et al., 2002; ACEA, 2004).

EMICAT2000 CO emission (259 kt yr^{-1}) is 67% of the 1990 value, while the European reduction trend was estimated in 60% (Ntziachristos et al., 2002). VOC emission (50.5 kt yr^{-1}) is 60% of the 1990 value, almost the same as that of the European reduction trend, estimated in 58%.

Excluding CO, emissions by industrial activities are comparable with values obtained by the EMEP sectors 3

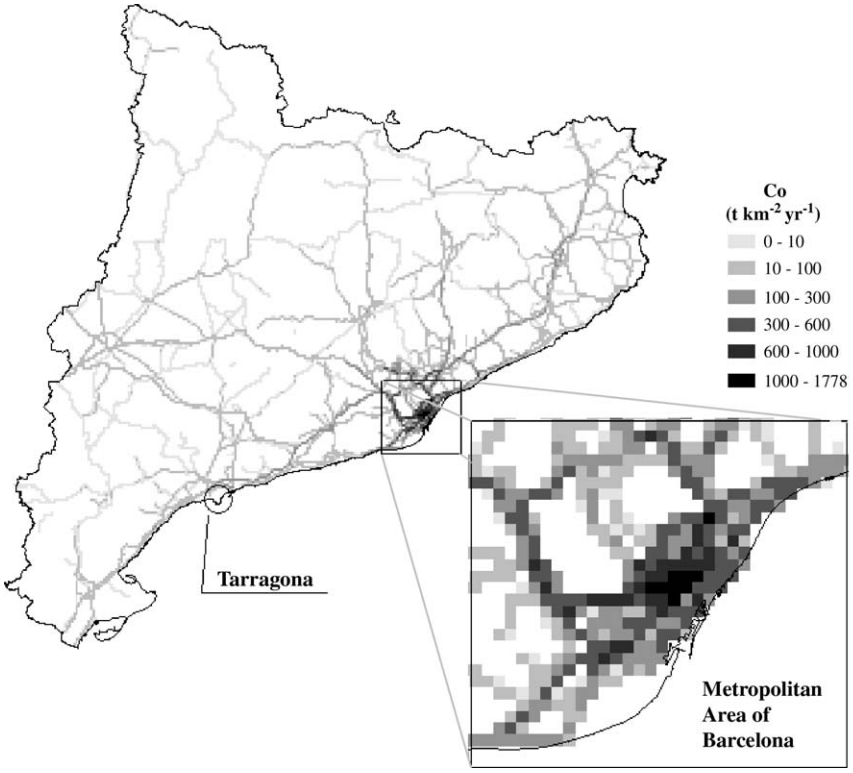

Fig. 3. CO emission distribution due to on-road traffic during the year 2000.

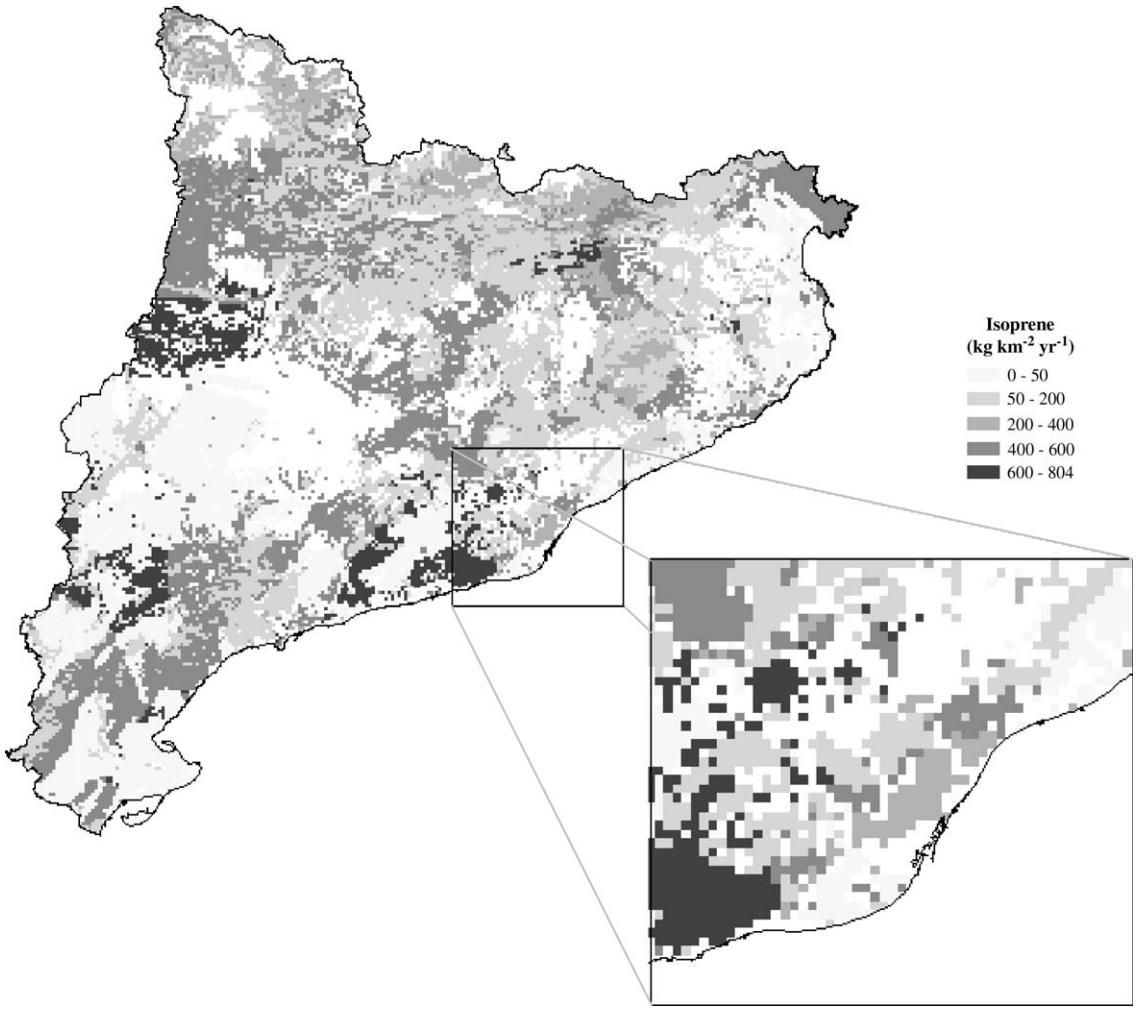

Fig. 4. Isoprene emission in Catalonia during the year 2000. Main emitter sources are shrub lands, coniferous and deciduous forest.

(industrial combustion) and 4 (production processes) for Catalonia. The lower estimation of EMICAT2000 could be explained by the relative small number of industrial centres included ($\approx 10\%$) which are the most important emitter of O_3 precursors. The large number of small centres not yet included could represent an important source of CO, although the greatest uncertainties about contribution of total CO emission exist with regard to small industries and households (ACEA, 2004).

All the sectorial annual emissions provided by EMICAT2000 agrees satisfactorily with other estimations included in this comparison.

3.2. Monthly behaviour of O_3 precursors

Higher emissions of O_3 precursors are produced during summertime (Fig. 5). NO_x increase is mainly due to higher traffic, especially by the influence of foreign vehicles during vacation period. The increase of NMVOC is explained by the highest temperature and solar radiation, implying important vegetation and on-road traffic evaporative emissions. These are the causes that most of O_3 exceedances take place during summertime in Catalonia. NO_x emission varies between 8 and 10 kt month^{-1}. The influence of on-road traffic and industries is important. NMVOC emission varies between 8 and 18 kt month^{-1}, and thus there is a great influence of the vegetation profile.

3.3. Daily speciated emissions

Inventories for modelling require disintegrated emissions (speciation) by individual compounds or species, according to their reactivity and under the approach of chemical mechanism. Jiménez et al. (2003) developed a comparative assessment of seven state-of-the-science chemical mechanisms. Almost all of them provided similar results of O_3 concentrations, although discrepancies were found for other species (such as HO_2 or HNO_3). Carbon Bond IV (CB-IV), using the least amount of reactions (hence demanding less computational time when implemented in a CTM) presented low O_3 deviation

Table 2
Comparative summary of sectorial annual emissions from EMICAT2000 with other emission inventories (kt yr^{-1})

Air pollutant	Other inventories				EMICAT2000
	Emission year and source of information				
Vegetation					
		1992 (Gómez and Baldasano, 1999)	2000 EMEP Catalonia	2000 (DMA, 2003)	
Isoprene		16.6			5.9
Monoterpenes		23.5			24.7
OVOC		10.8			16.3
Total		50.9	88.8	73.9	46.9
On-road traffic					
	1990 (DMA, 2003)	1994 (Delgado et al., 2000)	2000 EMEP Catalonia	2000 (DMA, 2003)	
NO_x	92.7	77.8	84.8	62.8	62.4
CO	385.8	283.5	274.2	366.9	259.0
COV	84.5	35.1	58.0	82.8	50.5
SO_2	12.2	6.7	2.7	12.9	1.3
TSP	11.8	4.2		10.2	15.7
CO_2 eq.				11 203	8302
Power generation					
			2000 EMEP Catalonia[a]		
NO_x			11.5		15.0
CO			1.3		2.1
NMVOC			0.4		0.7
SO_2			38.5		28.5
TSP					1.7
CO_2 eq.					5698
Industrial activities (except power generation)					
			2000, EMEP Catalonia[b]		
NO_x			30.9		26.1
CO			23.5		5.4
NMVOC			21.8		22.0
SO_2			47.2		32.7
TSP					5.9
CO_2 eq.					14 663
Use of fossil fuels in residential and commercial sectors					
				2000 (DMA, 2003)	
NO_x				2.7	3.3
CO				1.9	1.1
NMVOC				0.2	0.2
SO_2				2.4	2.3
TSP				0.3	0.3
CO_2 eq.				3693	3511

[a] Corresponds to sector 1 (*public power stations*) of the EMEP emission inventory for Catalonia.
[b] Corresponds to sector 3 (*industrial combustion*) and sector 4 (*production processes*) of the EMEP emission inventory for Catalonia.

with respect to the average values provided by the mechanisms assessed.

CB-IV is a lumped-structure condensed mechanism which uses the carbon bond approach for the lumping of organic species, and treats the reactions of four different species: (1) inorganic species, (2) explicit organic species treated explicitly due to its special importance or reactivity, (3) organic species represented by carbon surrogates, and (4) organic species represented by molecular surrogates (Gery et al., 1989). CB-IV was chosen for speciation of hourly emissions provided by EMICAT2000. Figs. 6 and 7 show the NMVOC profiles by sources.

On-road traffic speciation profiles are variable. Exhaust emissions from conventional gasoline vehicles (without catalyst), heavy gasoline duty vehicles and motorcycles produces mainly 52% of PAR (paraffin bond C–C), 21% of NR (not reactive species), 7% of TOL (toluene C_6H_4-CH_3), 6% of XYL (xylene C_6H_5-$(CH_3)_2$) and 6% of OLE (olefinic bond C=C). Exhaust emissions from gasoline vehicles with catalyst also have a wide composition, although the percentage of PAR is higher (68%) and NR lower (13%). Evaporative emissions contribute mainly as PAR (95%). On-road traffic NMVOC emissions contribute with almost all the CB-IV organic species.

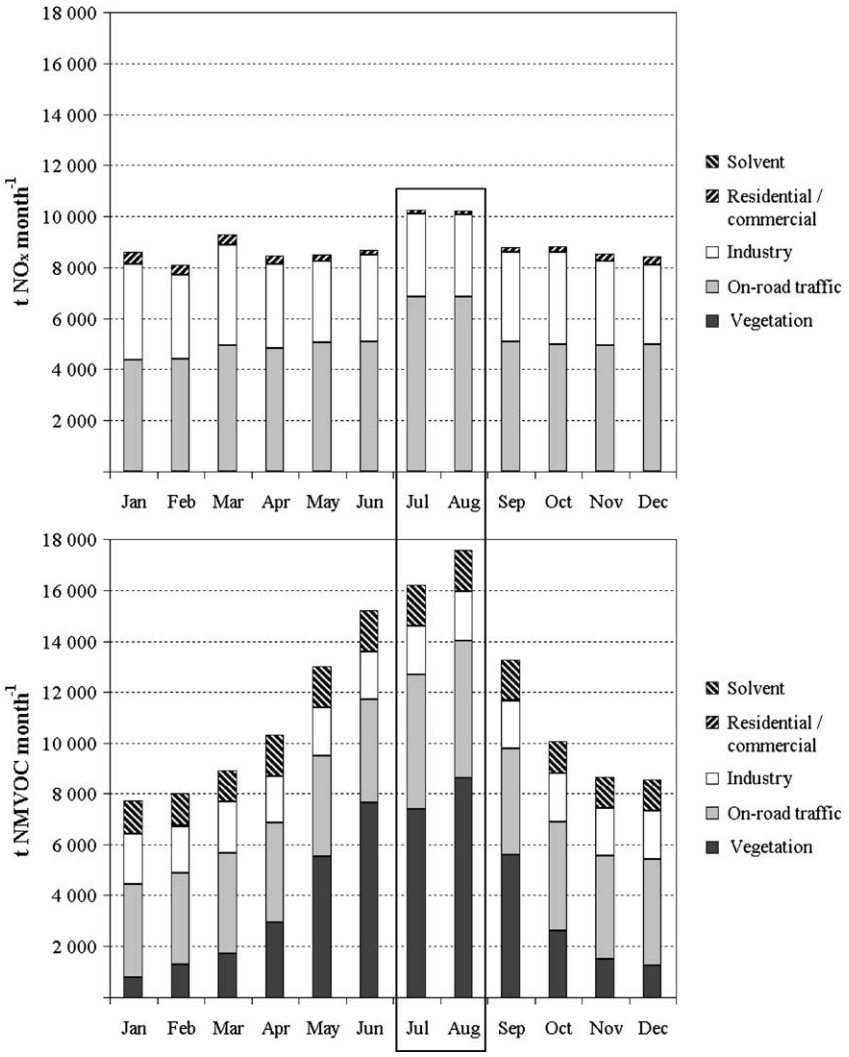

Fig. 5. Monthly emissions of NO_x and NMVOC in Catalonia during the year 2000. Emissions of O_3 precursors are higher during summertime, especially due to higher traffic flows and higher temperature and solar radiation (promoting important vegetation emissions).

Industrial profiles show a wide different range of compositions. PAR species has important contributions (between 30 and 90%) from oil refining emissions and olefins, polypropylene and ABS resins production.

Isoprene, due to its high reactivity is considered explicitly and is lumped 100% under the ISOP specie. Monoterpenes profile considered 75% as PAR, 19% as ALD2 (acetaldehyde and other heavy aldehydes) and 6% as OLE.

Table 3 shows the speciated NMVOC contribution by sectors during 14 August 2000. Table 4 presents the reaction and rate constants of CB-IV species when reacting with OH radical. These reactions have crucial roles in photochemical pollution. PAR is the main species emitted (76%), although it is the least reactive (the lowest rate constant). Sixty-five percent of PAR is emitted by on-road traffic and 28% by vegetation. The ISOP species (the highest rate constant) represents just about 1% of the NMVOC speciated emissions. Sixty-four percent and 28% were emitted by on-road traffic and vegetation, respectively. The composition of speciated emission is complex with comparative low emission of very reactive species against high emission of low reactive species.

3.4. Grid-cell size within EMICAT2000

Selection of grid-cell size is important because the characteristics of species are assumed uniform in each grid cell and chemical species are represented by concentration. Because the processes that regulate species transformations are assumed to be uniform in each grid cell, certain characteristics of sub-grid processes and their interactions may be lost as the size of the model grid cell increases.

To provide emission data with lower spatial resolution, EMICAT2000 integrates 1 km cells' emissions into squares of 2, 4 and 8 km by side. Coarse cells'

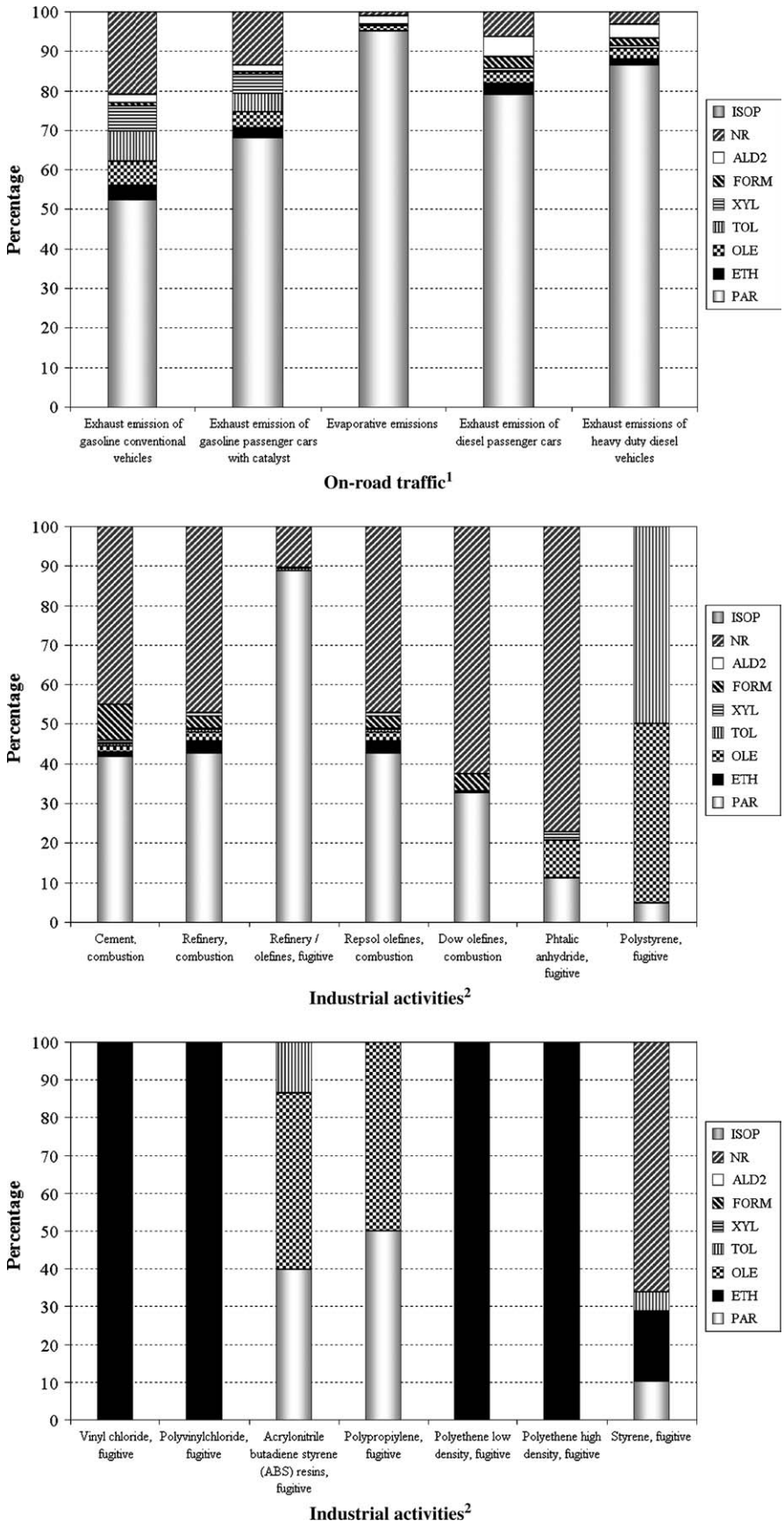

Fig. 6. NMVOC speciation profiles for on-road traffic and industrial emissions, according to the Carbon Bond IV categories. [1]On-road traffic profiles were deduced from NMVOC profiles of Ntziachristos and Samaras (2000). [2]Industrial profiles were chosen from USEPA (2004a) database.

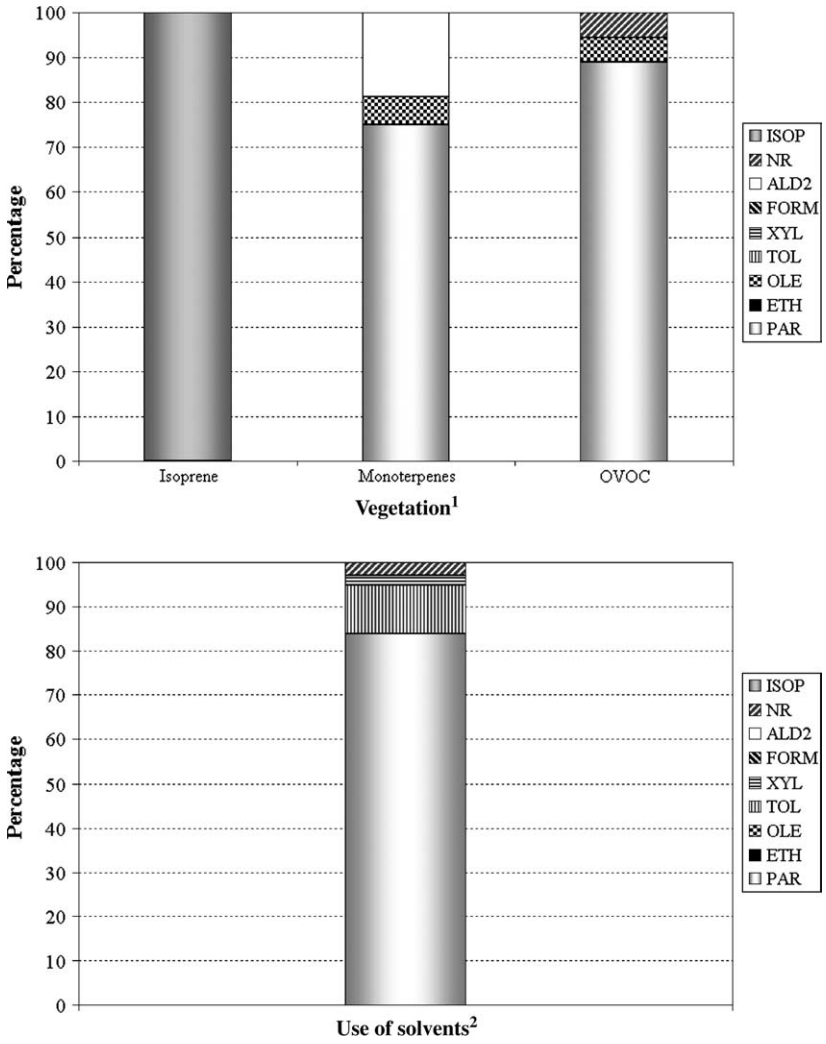

Fig. 7. NMVOC profiles speciation for vegetation and use of solvents emissions, according to the Carbon Bond IV categories. [1]Vegetation profiles were chosen from USEPA (2004b). [2]Use of solvents profile was chosen from USEPA (2004a).

Table 3
Contribution of speciated NMVOC in Catalonia during 14 August 2000

Sector	ALD2	OLE	PAR	ETH	TOL	XYL	FORM	ISOP	NR	Total	%
Carbon Bond IV category emission (kmol d^{-1})											
Vegetation	1862	1003	13 558					865	382	17 670	27.95
On-road traffic	896	1342	31 220	630	1292	1104	178		3735	40 397	63.90
Industries	2	254	1766	464	67	4	22		735	3314	5.24
Fossil fuels − residential and commercial			1						2	3	0.00
Use of solvents			1542		202	38			56	1838	2.91
Total	2760	2599	48 087	1094	1561	1146	200	865	4910	63 222	100.00
%	4.37	4.11	76.06	1.73	2.47	1.81	0.32	1.37	7.77	100.00	
Carbon Bond IV category emission (%)											
Vegetation	67.46	38.59	28.19					100.00	7.78		
On-road traffic	32.46	51.64	64.92	57.59	82.77	96.34	89.00		76.07		
Industries	0.07	9.77	3.67	42.41	4.29	0.35	11.00		14.97		
Fossil fuels − residential and commercial			0.00						0.04		
Use of solvents			3.21		12.94	3.32			1.14		
Total	100	100	100	100	100	100	100	100	100		

Carbon Bond IV species: ALD2 (acetaldehyde and other heavy aldehydes), OLE (olefinic bond C=C), PAR (paraffin bond C−C), ETH (ethene), TOL (toluene), XYL (xylene), FORM (formaldehyde), ISOP (isoprene), NR (not reactive).

Table 4
Reactivity of the Carbon Bond IV species with OH radical (USEPA, 2004b)

Reaction number	Reaction	Rate constant ($m^3\ mol^{-1}\ s^{-1}$) (293 K, 1 atm)	Rate constant ratio with PAR as reference
38	FORM + OH = HO2 + CO	1.00E−11	12
44	ALD2 + OH = C2O3	1.64E−11	20
53	PAR + OH = 0.87XO2 + 0.13XO2N + 0.11HO2 + 0.11ALD2 + 0.76ROR − 0.11PAR	8.10E−13	1
58	OLE + OH = FORM + ALD2 + XO2 + HO2 − PAR	2.90E−11	36
62	ETH + OH = XO2 + 1.56FORM + HO2 + 0.22ALD2	8.10E−12	10
64	TOL + OH = 0.08XO2 + 0.36CRES + 0.44HO2 + 0.56TO2	6.30E−12	8
70	XYL + OH = 0.7HO2 + 0.5XO2 + 0.2CRES + 0.8MGLY + 1.1PAR + 0.3TO2	2.50E−11	31
77	ISOP + OH = 0.912ISPD + 0.629FORM + 0.991XO2 + 0.912HO2 + 0.088XO2N	1.02E−10	126

emissions are obtained adding the emission values of minute cells allocated into the formers (one 8-km sided cell contains 64 1-km sided cells). Hence total emissions from different spatial resolution maps are the same. Fig. 8 shows emission maps of PAR species (12 Local Standard Time, 12 LST) for 15 August 2000. Maps made with 1 or 2 km cells depict in detail the emission configuration. The axes of the most important highways and roads are clearly defined and most of this species PAR is clearly emitted mainly from the MAB and Tarragona. Within a coarser spatial resolution, the emission features get lost. The map made with cells of 8 km, does not show clear definitions of road axis and the attenuation of emission is clearly observable in the MAB.

To appreciate the resolution level, Fig. 9 depicts the annual map emission of NMVOC from vegetation obtained with EMICAT2000, relative to the map obtained from the European database EMEP. The second map

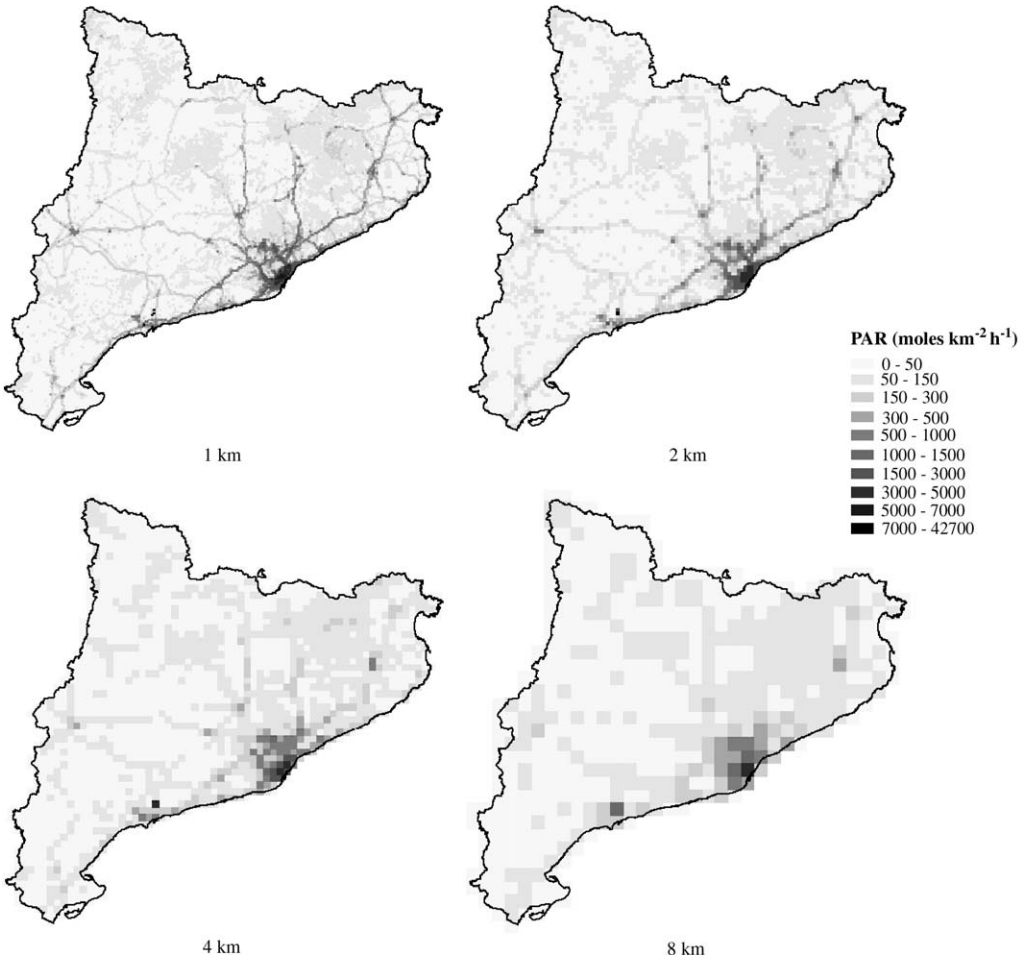

Fig. 8. PAR (paraffin bond) emissions (12 LST, 15 August 2000) in Catalonia with different spatial resolution.

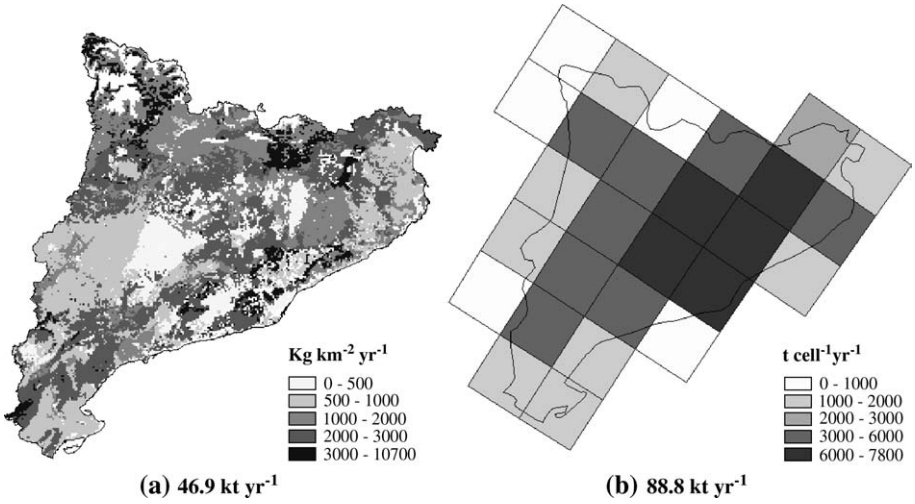

Fig. 9. NMVOC emissions for the year 2000, (a) EMICAT2000, (b) obtained from the European inventories EMEP.

shows cells with the highest values inland Catalonia, which disagrees with the result provided by EMICAT2000.

3.5. Uncertainty

Uncertainty assessment could be developed by internal approaches (using the own model and information for inventories: qualitative discussion, data quality rating, error propagation) or external approaches (using information from other studies: comparison with other inventories, forward and inverse air quality modelling).

Using an adapted method from the Data Attribute Ratings System (USEPA, 2003) for the most important emission elements, we developed a qualitative assessment of the uncertainty. We assigned numerical scores (1 for the worse representative situation, 10 for the best) to emission factors, activity data and parameters; according to three kinds of attributes, to provide an overall confidence rating for the emission inventories. Attributes considered were: (1) measurement technique (how emission factors, activities or parameters were obtained: direct measures, representative samples, small samples), (2) spatial agreement (representative level for Catalonia), and (3) temporal agreement (representative level during the year). Table 5 shows the quality uncertainty assessment obtained. The monoterpenes estimation was qualified as the more accurate (73). The highest level of uncertainty was assigned to emissions due to use of solvents.

This subjective evaluation was made mainly to establish the different quality of accuracy between the main elements of EMICAT2000 and clearly identified aspects that could be improved in the future.

Also, we developed a preliminary assessment of the uncertainty of hourly monoterpenes emission (15 August) using the Monte Carlo approach under some hypothesis (Parra et al., 2004). Working with the 95% confidence limits, we established that upper to lower emissions' ratios were between 2.3 and 4.2. The highest upper limit is 60% greater than its average value and the lowest limit is 40% of its mean value.

It was not possible to develop a quantitative assessment of uncertainty for other sources, mainly because of the non-availability of information about the statistical distribution of data and in part by the inability of commercial GIS software to store data according to their statistical distribution.

High levels of uncertainty are inherent to vegetation emission inventories. Guenther et al. (2000) mentioned

Table 5
Quality uncertainty assessment of the main emission components

Sector/emission component	Rating (points)
Vegetation	
Isoprene	53
Monoterpenes	73
OVOC	47
On-road traffic	
NO_x (hot exhaust emissions)	51
VOC (hot exhaust emissions)	51
VOC (diurnal evaporative)	41
VOC (soak evaporative)	41
VOC (running evaporative)	46
CO (hot exhaust emissions)	51
Industrial activities (primary pollutants and greenhouse gases)	
Power generation in conventional centrals	61
Cogeneration	38
Municipal waste incineration	59
Cement	56
Oil refining	41
Residential and commercial sectors	
Fossil fuels use (primary pollutants and greenhouse gases)	58
Use of solvents (VOC)	36

a factor of 3 as a reasonable estimation of the uncertainty associated with annual total NMVOC emissions. Also, a factor of 10 is mentioned that could be reached for specific times, locations and compounds. Simpson et al. (1995, 1999) indicated similar levels for Europe. Simon et al. (2001) associated annual uncertainty factors of 4 for isoprene, 5 for monoterpenes and 7 for OVOC from French ecosystems. Latter percentages could be considered as minimum factors for Catalonia.

Annual emissions by on-road traffic could have uncertainty factors until 1.5–2 (Kühlwein and Friedrich, 2000; Colvile et al., 2001), although at stretch road level it could be 10 (Colvile et al., 2001). The comparison shown in the Section 3.1 (Table 2) is a general external assessment of the accuracy (uncertainty) of EMICAT2000 annual results.

4. EMICAT2000's framework

The framework of the EMICAT2000 was developed completely into a GIS tool environment, and is shown in Fig. 10. Each emission source is built with modules for storage and management of base information, emission calculation and speciation. The emission files management module aggregates the sectorial values and generates emission files with lower spatial resolution. Its structure is flexible, allowing easily future update of emission models, emission factors, parameters or base information. It directly generates the emission files under the netCDF protocol required for the third generation Models-3/CMAQ Air Quality Modelling System, nowadays one of the most important and powerful air quality models, supported by the US Environmental Protection Agency (Byun and Ching, 1999).

5. Modelling case

To illustrate the application of EMICAT2000 for air quality modelling studies, we present the air quality simulation results over Catalonia for an episode that took place on 13–16 August, 2000. Values over the European threshold of 180 $\mu g\,m^{-3}$ for ground-level O_3 are attained. This situation is representative of an episode of photochemical pollution in the north-eastern Iberian Peninsula, since the occurrence of regional wind and pollutants re-circulations at low levels represents 45% of the yearly (and 70% of summertime days) transport patterns over the area of study (Jorba et al., 2004). The domain of study covers the domain defined for EMICAT2000. Wind fields and other meteorological information were obtained using the PSU/NCAR mesoscale model MM5.

Ground-level O_3 simulation results were compared to the measurements from 48 surface stations, located in urban, industrial and rural areas, for 1 h peaks of ozone. Table 6 collects the results of the statistical analysis using USEPA (1991) recommendations for the whole episode of 13–16 August, 2000; although there is no

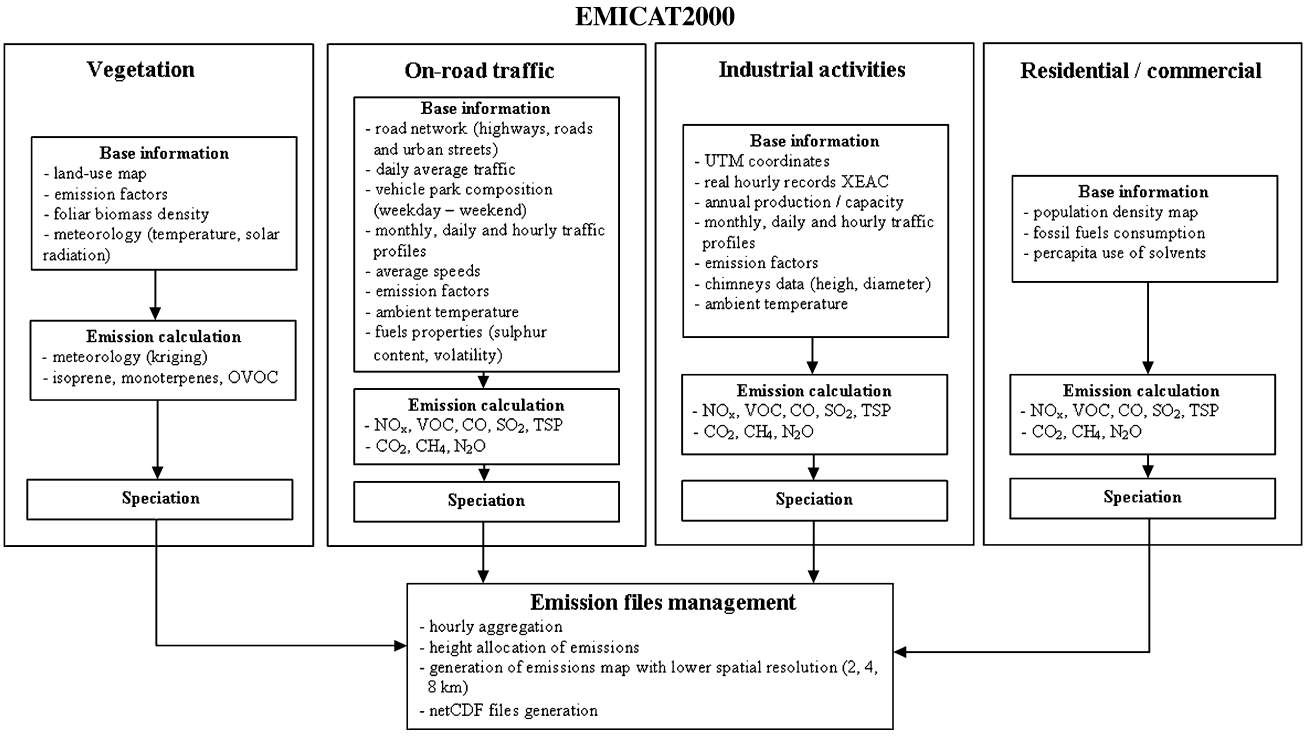

Fig. 10. EMICAT2000 emission model framework.

objective criterion set forth for a satisfactory model performance, suggested values of ±10–15% for the Mean Normalized Bias Error (MNBE), ±15–20% for the Unpaired Peak Prediction Accuracy (UPA) and +30–35% for the Mean Normalized Gross Error (MNGE) to be met by modelling simulations of O_3 have been considered for regulatory applications (Russell and Dennis, 2000).

The model results achieve the EPA goals for a discrete evaluation on all episode days. The O_3 bias is negative on each day, ranging from −2.1% on the first day of simulation until −14.3% on August 15. That suggests a slight tendency towards under prediction; however, EPA goals of ±15% are met. This negative bias may suggest that the O_3-production chemistry may not be sufficiently reactive. The modeled episode peak (189 $\mu g\,m^{-3}$) is well-captured by the model. Peak accuracy is overestimated on the first- and last-day of simulations (14.4% and 5.2%, respectively) and underestimated on the central days of the episode (−3.8% and −11.7%). The MNGE increases from August 13 until August 16 (16.8–26.7%), mainly to deviations in meteorological predictions that enlarge with the time of simulation (Jiménez et al., 2004). The uncertainty (deviation of 50% for the 1 h averages) objective for modelling of air quality assessment set in the European Directive (2003/2/CE relating to ozone in ambient air), is also achieved for the whole period of study. A more detailed description of the evaluation of this episode can be found in Jiménez and Baldasano (2004), and Jiménez et al. (2005).

The peculiar topography of the zone is one of the principal driving mechanism that contributes to the dispersion of emissions. Maximum O_3 levels are measured in Plana de Vic and the industrial zone of Tarragona. Fig. 11 shows the O_3 patterns on 15 August 2000. At night, the coast presented down-slope winds over the mountains and general offshore breeze flows. The offshore flows produce a drainage of pollutants towards the coast through the river valleys. As the day advanced, a well-developed sea-breeze regime established along all the domain. Under a weak synoptic forcing, strong insolation promotes the development of prevailing mesoscale flows associated with the local orography, while the difference of temperature between the sea and the land enhances the development of sea–land breezes (Barros et al., 2003). At noon, pollutants departing from Barcelona area and the line coast are transported inland following the breeze front, since they arrive at Vic (70-km downwind Barcelona), where wind calms and allow O_3 and its precursors to accumulate. The O_3 time series for stations of Barcelona and Vic are shown in Fig. 12.

In the southern part of the domain, the intensity of the breeze is not capable to overcome the littoral mountain range, and therefore appearing a local maximum downwind the industrial area of Tarragona before noon, as shown in Fig. 12 for the air quality station of Perafort. This pattern keeps constant during the afternoon, as seen in Fig. 11, but the breeze gains intensity and adds to the upslope winds transporting pollutants over the pre-littoral mountain ranges. In the evening, photochemical activity ceases, the see-breeze regime loses intensity and winds in the coast weaken. Inland, at the east part of the domain, strong south winds are observed, which translates into a transport of O_3 that adds to the quenching procedure of photochemical ozone by its reaction with NO emissions.

Although air quality modelling integrates uncertainties by meteorology and the CTM themselves, accurate modelling results are obtained as a consequence of the quality of emissions, which is normally considered the most important source of uncertainty (Russell and Dennis, 2000). The quality of the simulated values shown in this section (USEPA recommendations are met on every single day of simulations) and the ability to diagnose ozone dynamics over a very complex area as Catalonia is an external assessment of the accuracy of emission inventories provided by EMICAT2000.

Table 6
Statistical measures of model performance for 1 h O_3 during the episode of August 13–16, 2000

	USEPA Goal	August 13, 2000	August 14, 2000	August 15, 2000	August 16, 2000
Observed peak ($\mu g\,m^{-3}$)		157	177	189	171
Modeled peak ($\mu g\,m^{-3}$)		189	170	167	180
UPA[a] (%)	±20	14.4	−3.8	−11.7	5.2
MNBE[b] (%)	±15	−2.1	−11.0	−14.3	−5.6
MNGE[c] (%)	+35	16.8	19.8	21.7	26.7

[a] UPA: Unpaired Peak Prediction Accuracy.
[b] MNBE: Mean Normalized Bias Error.
[c] MNGE: Mean Normalized Gross Error.

6. Conclusions

We described the main elements and detailed results obtained with the high spatial (1 km^2) and temporal (1 h) resolution EMICAT2000 emission model, which provides high quality emission inventories used both for modelling and policy purposes in Catalonia (northeastern Iberian Peninsula). Uncertainty assessment was developed using both internal and external approaches. Although all sources are not included, emissions by vegetation, on-road traffic, most important industries and residential and commercial sectors define the spatial and temporal emissions pattern. Inclusion of minor sources will provide detail features.

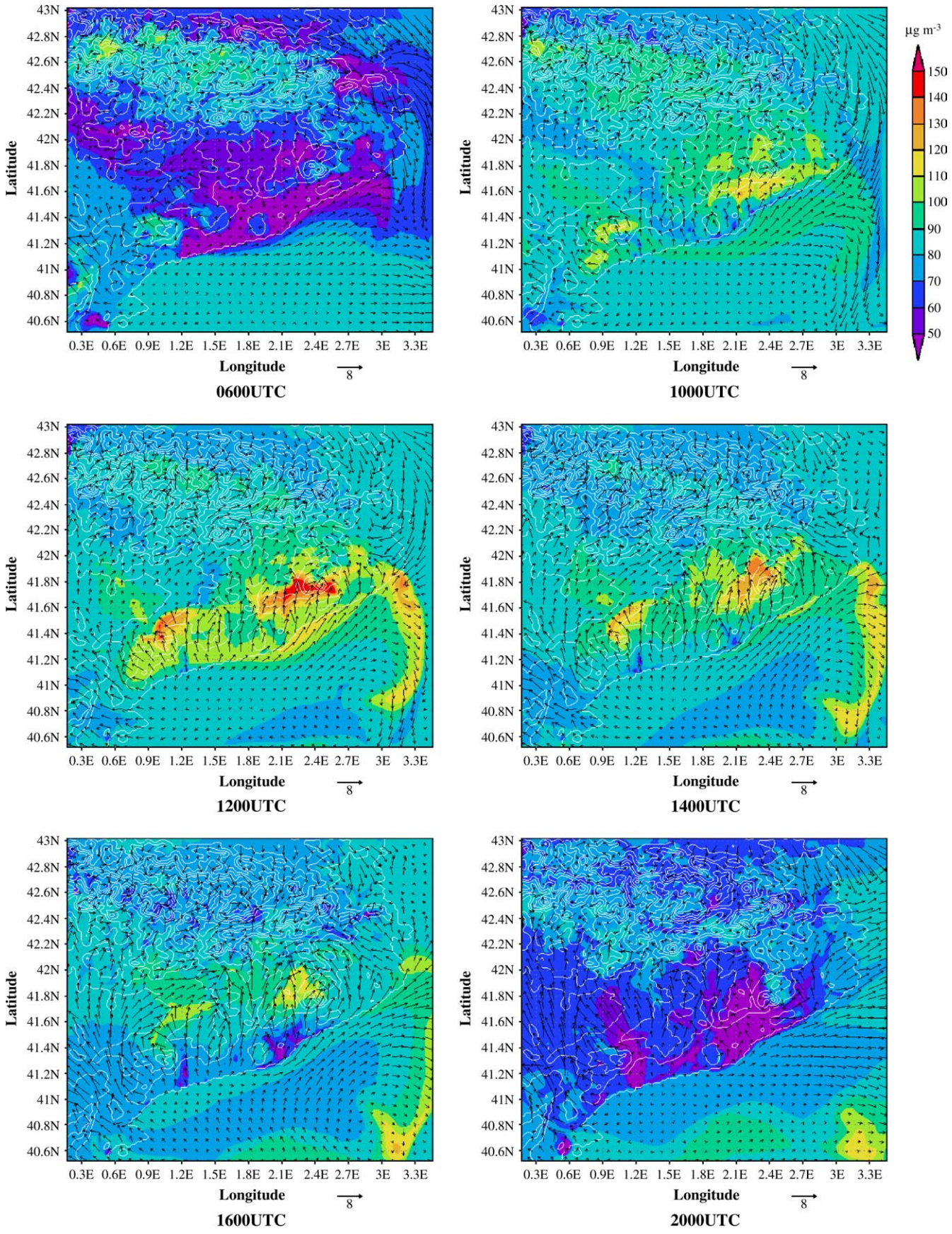

Fig. 11. O_3 concentrations ($\mu g\ m^{-3}$) and wind field vectors (m/s) at ground-level over the north-eastern Iberian Peninsula (Catalonia), simulated with EMICAT2000-MM5-Models-3/CMAQ on 15 August, 2000, at (from up-left to down-right) 0600, 1000, 1200, 1400, 1600 and 2000UTC.

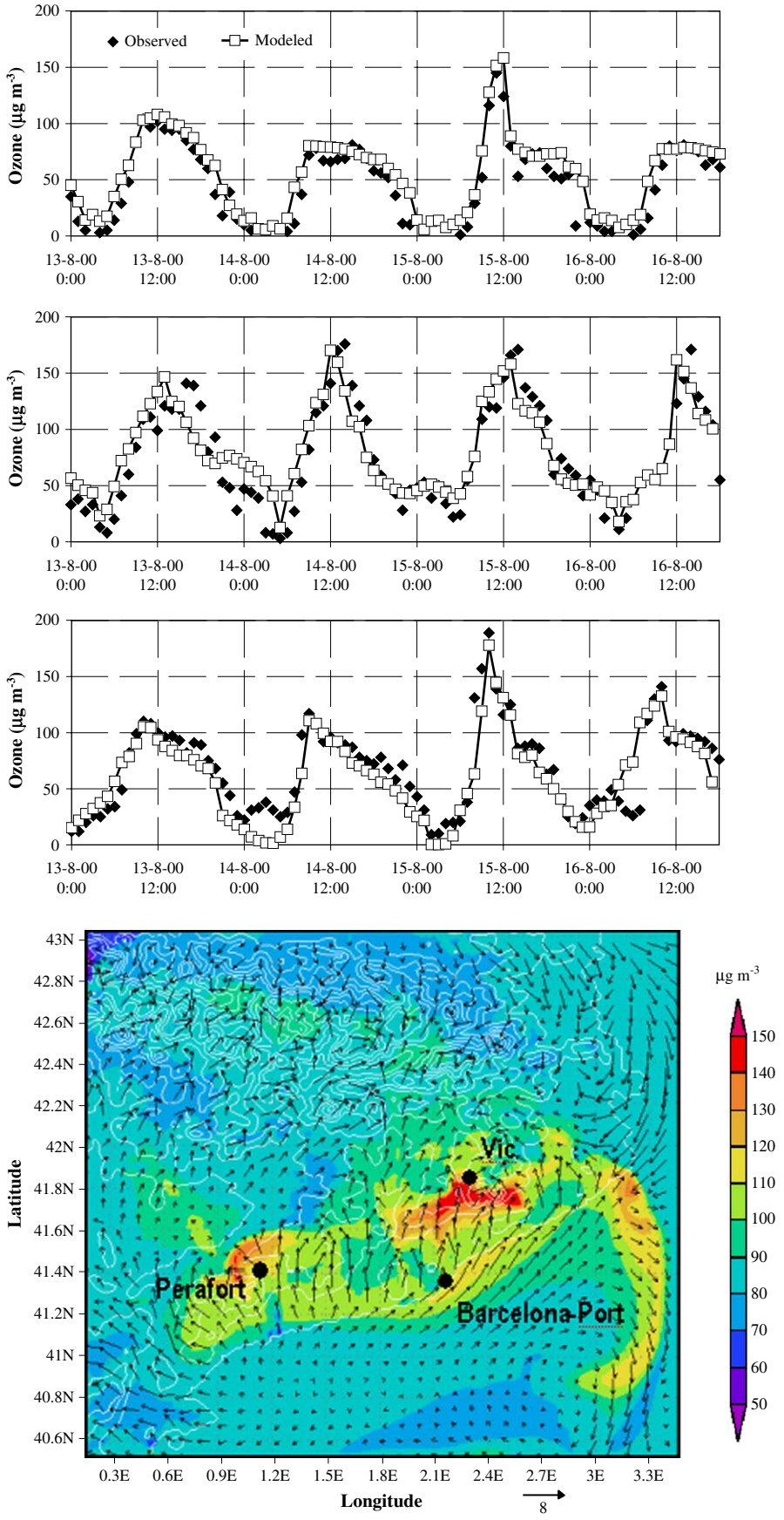

Fig. 12. Time series for measured and simulated concentrations of O_3 in the stations of Barcelona-Port (up), Vic (centre) and Perafort (down).

Application of Models-3/CMAQ requires the previous integration and treatment of emission inventories provided by different US emission models, using additional tools as MEPAP or SMOKE. It means the exploitation of this air quality model in other regions is not straight, not flexible and difficult. EMICAT2000 resolved these obstacles directly providing emission data files that are being successfully used within this CTM.

EMICAT2000 could be the basis for the forecasting of air quality in Catalonia under different emission scenarios. Also its usefulness into the framework of the European Directive (2003/2/CE relating to ozone in ambient air) could be important, especially for the data quality objectives and compilation of the results of air quality assessment by modelling.

Among the elements required for air quality modelling, high spatial emission inventories demand important resources, but the benefits and potential applications are obvious.

Acknowledgements

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