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UNIVERSITAT POLITÈCNICA DE CATALUNYA
Programa de doctorado en Ingeniería Ambiental

PhD Thesis

Assessing variations in urban air quality when introducing on-road traffic management strategies by means of high-resolution modelling. Application to Barcelona and Madrid urban areas.

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Directors:
Dr. Pedro Jiménez Guerrero
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Barcelona, December 2008

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Summary

The urban air pollution affects human health, causes damage to ecosystems and transboundary air pollution. The southern Mediterranean region and specifically the Iberian Peninsula are commonly affected by highly polluted episodes. The high O_3 levels, and specifically the NO_2 and PM concentrations in urban areas, are of special concern, frequently exceeding the European air quality targets. On road traffic is the main source of anthropogenic emissions in the urban environment. Different strategies addressed to reduce this contribution are being currently implemented and tested. The quantitative assessment of their effects in advance is fundamental to help decision makers. Air quality modelling is the most suitable tool to perform this kind of evaluations.

This PhD Thesis proposes the use of the mesoscalar WRF-ARW/HERMES/CMAQ modelling system with high resolution to test in advance such strategies for on-road traffic emissions abatement. It focuses on the two most populated urban areas of Spain, Barcelona and Madrid, which constitute a typically coastal and continental environment. The 17-18 June, 2004 is chosen as the study case. It fits in a poor air quality situation in both areas but also in an usual traffic circulation pattern (working days). Several feasible short-term strategies are selected and implemented in the emission model in the most realistic way as possible. They include: use of alternative fuels, such as natural gas or biodiesel, introduction of new technologies, such as hybrid electric vehicles, and urban management schemes, such as speed circulation limit. They are evaluated in terms of emissions, fuel consumption and air quality changes in the urban areas and at a regional scale, over the North-eastern and Central Iberian Peninsula.

The effects of these strategies depend on the study areas. The factors that condition these differences are mainly: (1) the specific vehicle fleet composition, having Barcelona a larger proportion of diesel and commercial vehicles and lower quantity of passenger cars than Madrid; (2) the different contributions of the activity sectors to anthropogenic emissions, which in Barcelona and the North-eastern Iberian Peninsula reflect a heavier industrial activity against Madrid and the Central Iberian Peninsula region; (3) the different contributions of atmospheric processes leading to the concentration of pollutants; their quantification leads to the observance of characteristic transport patterns of a coastal area and very complex terrains in Barcelona, and a much simpler behaviour in Madrid, a typically continental area; (4) the chemical sensitivity regime also differs, which particularly affects the O_3 response to NO_x abatement strategies.

The effect of all the tested strategies in urban NO_2, SO_2 and PM_{10} concentrations is positive, being lower than in the base case. Nevertheless the extent of this effect largely depends on the affected fleets and on the urban area of application. The introduction of natural gas vehicles proved to be specifically effective in reducing SO_2 and PM_{10} concentrations in Barcelona and Madrid areas. The introduction of biodiesel as a fuel would slightly affect the urban air quality levels, improving mainly the SO_2 levels. The use of hybrid cars affects the NO_x emissions considerably, reducing NO_2 urban levels. Moreover in Madrid it causes a noticeable reduction in the local O_3 concentrations. The Barcelona photochemical regime involves local O_3 concentrations increase when reducing NO_x emissions.

The introduction of an 80 km h^{-1} speed limit in the Barcelona area reduces NO_2 and PM_{10} levels, specifically in the zones affected by the measure. The inclusion in the model of hourly speed data from measurement campaigns instead of the previously constant speed considered, allowed assessing the gains of such a measure in a more realistic manner, taking into account real circulation patterns and the congestion effect.

The effects of the selected strategies are always positive in downwind areas, even in terms of O_3 concentration.

The application of high resolution modelling proved to be a useful tool to quantitatively asses the effect of management strategies. The detailed emissions inventories and the availability of emission factors for new technology vehicles or alternative fuels are key factors to this kind of developments.

Resumen

La contaminación del aire urbano tiene efectos negativos en la salud humana, los ecosistemas y se asocia con el transporte de contaminantes a larga distancia. La región del sur del Mediterráneo y en concreto la Península Ibérica se ven sometidos frecuentemente a episodios de contaminación fotoquímica. Los niveles de O_3 troposférico y, en zonas urbanas, las concentraciones de NO_2 y material particulado, superan frecuentemente los niveles establecidos por la legislación europea.

El tráfico rodado constituye la mayor fuente de emisiones antropogénicas en el entorno urbano. Actualmente, se están ensayando distintas alternativas para reducir su contribución. El pronóstico cuantitativo de sus efectos es fundamental y proporciona la base para la toma de decisiones. La herramienta más adecuada para llevar a cabo este tipo de evaluaciones es la modelización atmosférica.

Esta tesis propone el uso del modelo mesoescalar WRF-ARW/HERMES/CMAQ con alta resolución para pronosticar el efecto de distintas estrategias de reducción de emisiones de tráfico. Se centra en las dos mayores ciudades de España: Barcelona y Madrid, representativas de un entorno costero y un entorno continental. Como caso de estudio se ha seleccionado el 17 y 18 de Junio de 2004, que se corresponde simultáneamente con altos niveles de contaminación y con un patrón de circulación de tráfico habitual (días laborables)

Se han seleccionado distintas estrategias realizables a corto plazo, que incluyen: el uso de combustibles alternativos: como gas natural o biodiesel, la introducción de nuevas tecnologías en vehículos, como el uso de vehículos híbridos, o sistemas de planificación urbana, como la introducción de un límite de velocidad. Se han evaluado en términos de cambio de emisiones, consumo de combustible y calidad del aire, no sólo en las zonas urbanas si no a escala regional (en el Noreste y Centro de la Península Ibérica)

Los efectos de dichas estrategias dependen de la zona de aplicación. Los factores principales que condicionan dichas diferencias son: (1) la composición específica de la flota, teniendo la de Barcelona mayor número de vehículos pesados diesel y menor número de turismos que la de Madrid, (2) el peso de los distintos sectores de actividad en el balance de emisiones total, que en Barcelona y el noreste peninsular refleja una mayor actividad industrial frente a Madrid y el centro de la Península, (3) las distintas contribuciones de los procesos atmosféricos a la concentración final de contaminantes, la cuantificación de dichos procesos permite definir patrones de circulación característicos de zonas costeras y con una orografía muy compleja en el área de Barcelona, mientras que en Madrid, el comportamiento es más simple, (4) el régimen de sensibilidad química, que es diferente en ambas ciudades, determina la respuesta del O_3 troposférico a la disminución de emisiones de NO_x.

El efecto de las estrategias estudiadas es positivo en términos de concentración de NO_2, SO_2 y PM_{10}, siendo éstas menores que en el escenario base (sin cambios). Sin embargo, el alcance depende en gran medida de las flotas específicas afectadas y del área urbana que se considere. La introducción de vehículos a gas natural constituye una medida eficaz para reducir los niveles de SO_2 y PM_{10} en las ciudades. El uso de biodiesel B20 disminuye fundamentalmente la concentración de SO_2, aunque puede conllevar un ligero incremento de

concentración de NO_2. El uso de vehículos híbridos reduce fundamentalmente las emisiones de NO_x, lo que produce una disminución de los niveles de NO_2 urbanos. En Madrid este hecho tiene efectos positivos en la concentración de O_3 local, sin embargo en Barcelona la disminución de emisiones de NO_x supone un incremento del O_3 local en todos los casos.

La limitación de velocidad a 80 km h^{-1} en el área de Barcelona reduce los niveles de NO_2 y PM_{10}, sobre todo en las zonas directamente afectadas por la medida. La introducción en el modelo de velocidades de circulación horarias, en lugar de la velocidad constante previamente considerada, ha permitido determinar estos cambios en calidad del aire de manera más precisa, teniendo en cuenta patrones de circulación reales y el efecto de la congestión.

En general las estrategias seleccionadas tienen efectos positivos en zonas a sotavento de las ciudades, incluso en el caso del O_3.

La aplicación de modelización atmosférica con alta resolución es una herramienta útil para determinar cuantitativamente los efectos de estrategias de reducción de emisiones de tráfico. Los inventarios de emisiones detallados y la disponibilidad de factores de emisión para nuevas tecnologías o combustibles alternativos son un factor clave para este tipo de desarrollos.

Publications related to this Thesis

International Journals included in the Science Citation Index (SCI)

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Acronyms

a.g.l.: above ground level

AOPII: Auto-Oil II Programme

AQS: Air Quality Station

ARW: Advanced Research WRF

BC: base case

BCN: Barcelona

BSC-CNS: Barcelona Supercomputing Center – Centro Nacional de Supercomputación

CAFE: Clean Air For Europe

CBIV: Carbon Bond IV (chemical mechanism)

CIP: Central Iberian Peninsula

CLRTAP: Convention on Long-range Transboundary Air Pollution

CMAQ: Community Multiscale Air Quality model

EC: European Commission

EEA: Environmental European Agency

EMEP: European Monitoring and Evaluation Programme

ENGVA: European Natural Gas Vehicles Association

US-EPA: Environmental Protection Agency of United States

ETBE: ethyl-tert-butil-ether

EU: European Union

EZM: Eumac Zooming Model

FAME: fatty acid methyl ester

GHG: greenhouse gas

HERMES: High Elective Resolution Modeling Emissions System

HDV: heavy duty vehicle

IEA: International Energy Agency

IIASA: International Institute for Applied Systems Analysis

IP: Iberian Peninsula

IPR: Integrated Process Rate

MAD: Madrid

MNBE: Mean Normalized Bias Error

MNGE: Mean Normalized Gross Error

NCEP: National Centers for Environmental Prediction

NEIP: North-eastern Iberian Peninsula

NEC: National Emissions Ceilings

NG: Natural Gas
NGV: Natural Gas Vehicle
LDV: light duty vehicle
PBL: Planetary Boundary Layer
RADM: Regional Acid Deposition Model
RAINS: Regional Air Pollution Information and Simulation
UNECE: United Nations Economic Commission for Europe
UPA: Unpaired Peak Accuracy
UTC: Coordinated Universal Time
WHO: World Health Organization
WRF: Weather Research and Forecasting
XVPCA: Xarxa per la Vigilancia I Previssió de la Contaminació Atmosférica (Surveillance and precaution against atmospheric pollution network, from the Generalitat de Catalunya Government)

1 Introduction

1.1 Air quality in Europe. Urban air pollution

Atmospheric pollution is the environmental factor with the largest impact on human health and is responsible for the largest number of diseases related to the environment (EEA, 2005a). A recent report of the European Environmental Agency points out that despite the substantial body of international and national legislation and significant reductions in the emissions of some common pollutants, poor air quality is still causing hundreds of thousands of premature deaths in Europe every year and continues to damage crops and ecosystem health (EEA, 2007a). Specifically fine particulates concentrations, ground-level O_3, and acidification and eutrophication of ecosystems are a major concern. Tropospheric ozone and particulate matter affect particularly the human health (EC, 2005; WHO, 2004). Fine particles (diameter less than 2.5 µm) are associated with increased mortality, especially from cardiovascular and cardiopulmonary diseases (EEA, 2005a; Pope and Dockery, 2006).

The tropospheric ozone (O_3) levels are still a problem in southern European countries especially during summer. In Spain exceedances of both the alert (180 µg m^{-3} 1-hr average) and the information threshold (120 µg m^{-3} 8-hr average) are registered (EEA, 2007b). Concerning particulate matter several studies indicate that the European air quality targets are not accomplished (Ziomas et al., 1998; Dueñas et al., 2002; Palacios et al., 2002; Ribas and Peñuelas, 2004).

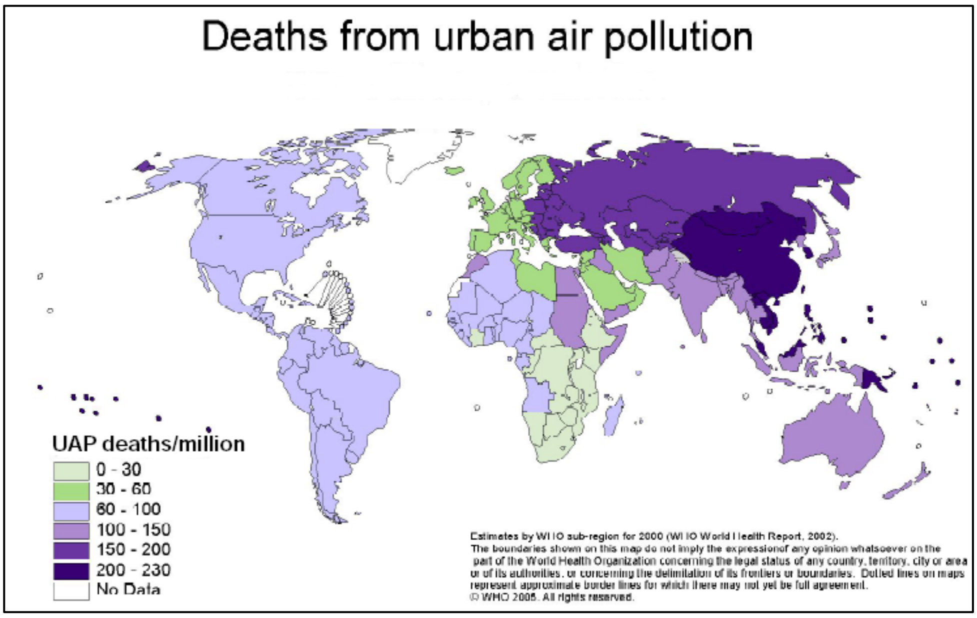
The World Health Organization (WHO) estimates that urban air pollution kills some 800.000 million people annually (WHO, 2002, Figure 1.1). In the urban environment, where 80% dwellers in Europe live, the major problems are related to fine particulate matter –$PM_{2.5}$- and nitrogen oxides concentrations -NO_x - (EEA, 2006a). The levels of urban pollutants depend on the local emissions, the meteorological conditions and the transport patterns, the local topography, the reactivity of the urban airshed and the background pollutants level or the transboundary pollutants transport (Vignati et al., 1996; Carruthers et al., 1999; Thunis et al., 2007).

The largest urban areas of Spain, Barcelona and Madrid (Figure 1.2), frequently exceed the daily PM_{10} target for human health protection (50 µg m^{-3}). I.e. this threshold was exceeded for the Barcelona area at 14 air quality stations during the year 2007 (Servei Vigilanciai i Control del Aire, 2008) and in the Madrid area from January 2008 to March 2008, exceedances of this limit were registered for almost all stations (D.G. Medio Ambiente, 2008). Moreover, the 40 µg m^{-3} limit that will be in practice at 2010 is not being accomplished in the urban areas (OSE, 2007). The effects of short term exposure to these PM levels cause approximately 1.4 premature deaths by 100.000 inhabitants (estimated for Bilbao, Madrid and Sevilla in 2005). Moreover, recent estimates indicate that the reduction of the $PM_{2.5}$ urban levels could prevent 3777 deaths a year in Barcelona, Bilbao, Madrid and Sevilla (OSE, 2007).

Additionally, the NO_2 EU thresholds are also exceeded both in Barcelona and Madrid. During the year 2007 exceedances of the annual limit for human health protection (40 µg m^{-3}) were registered in 6 of the 13 air quality stations in the Barcelona area (Servei Vigilanciai i Control del Aire, 2008). And in Madrid during the period from January to March, 2008, four air quality stations had already registered exceedances of this annual limit (D.G. Medio Ambiente, 2008).

In the specific area of the conurbations the O_3 EU thresholds for human health protection are rarely exceeded (Servei Vigilanciai i Control del Aire, 2008; A.G. Medio Ambiente, 2008), but the urban air pollution plume is on the origin of exceedances occurring in downwind areas. The CO, SO_2, benzene and Pb levels are below the legislation thresholds in both urban areas (Servei Vigilanciai i Control del Aire, 2008; A.G. Medio Ambiente, 2008).

As well as a source of local pollution, urban activities contribute to transboundary pollution and greenhouse gases (GHG) concentration increase (Fenger, 1999; Baldasano et al., 2003).



Estimates by WHO sub-region for 2000 (WHO World Health Report, 2002).
The boundaries shown on this map do not imply the expressionof any opinion whatsoever on the part of the World Health Organization concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. Dotted lines on maps represent approximate border lines for which there may not yet be full agreement.
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Figure 1.1 Estimated deaths caused by urban air pollution worldwide in 2000 (WHO, 2002)

Figure 1.2 Episodes of photochemical and particulate matter pollution in Barcelona (left) and Madrid cities (right)

1.1.1 Air pollution origin. On-road traffic as a source of atmospheric pollutant emissions

The last report of the Environmental Ministry of Spain indicated that during 2006 in Spain 1.2 million t y^{-1} of SO_x were emitted, the 72.8% came from the power generation sector, which also contributed with a 21.0% of total emitted NO_x. These compounds emissions accounted for 1.6 million t during that year, being the main emitter on-road traffic, that contributed with a 31.1% of the total amount. This sector was also the largest contributor to CO emissions (34.6% of 2.7 million t y^{-1} emitted) and the second emitter of PM_{10} (20.9% of 0.2 million t y^{-1}). The largest fraction of PM_{10} emissions comes from other types of transport (26.5%). Concerning NMVOCs emissions, biogenic (49.3%) and solvents use (19.6%) contributed the most to the 2.5 million t y^{-1} emitted (NEI, 2006). In conclusion, the largest fractions of atmospheric pollutants emissions have an anthropogenic origin (Figure 1.3), but also natural sources, such as biogenic or mineral dust have to be considered.

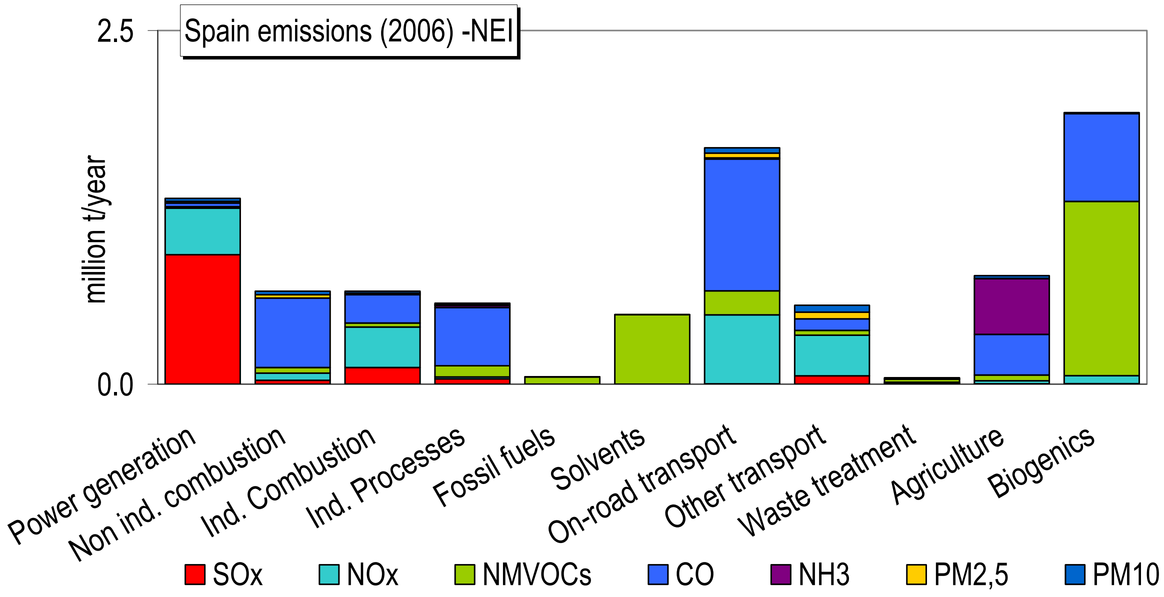

Figure 1.3. Pollutants mass emissions by sector for Spain, 2005. Source: National Emissions Inventory (2008).

In urban areas, the transportation sector, especially on-road transport, is the main emission source (Costa and Baldasano, 1996; Colvile et al., 2001; Ghose et al., 2004, Parra et al., 2006; Baldasano et al., 2008a).

The NO_x are mainly originated by combustion at high temperatures. The on-road traffic mainly emits NO (≈90%), which oxidizes in the atmosphere to form NO_2 and other oxidized compounds (see *section 1.4* for details). The NO_2 urban levels can be attributed to the regional background level, the urban background level, the directly emitted amount, and the chemically formed fraction (Palmgren et al., 1996).

The urban PM_{10} levels come from long range transport (usually secondary aerosols, nitrates and sulphates, and biogenic origin aerosols), from tailpipe emissions from on-road transport (mainly elemental carbon and organic carbon particulates) and brake wear or road dust resuspension (Lenschow et al., 2001; Abu-Allaban et al., 2003). The tailpipe emissions of diesel fuelled vehicles are normally higher than those attributed to petrol fuelled ones. Specifically in the urban Iberian Peninsula environments on-road transport, industrial sources and natural sources (such as the mineral dust outbreaks) are the main contributors to PM_{10} concentrations (Querol et al., 2004).

The SO_x emissions are mainly due to the combustion of fossil fuels containing sulphur, therefore power generation plants and on-road transport are the main sources of these pollutants. The sulphur content is higher in diesel than in gasoline, therefore the SO_x emissions of the diesel fuelled vehicles are usually larger. Nevertheless, the efforts done in the last decades in reducing the sulphur content on fossil fuels lead to low levels of SO_2, which currently does not involve a problem in urban areas (OSE, 2007).

The NMVOCs origin largely depends on the characteristic of the urban area, being the main source of these compounds on-road transport in cities such as Martorell (Spain) (Baldasano et al., 1998), or Borough (UK) (Crabbe et al., 1999), but having also the industrial activities or domestic heating important contributions in other urban areas (Cirillo et al., 1996). These pollutants emissions are mainly associated to the petrol fuelled vehicles, due to the larger volatility of this fuel respect to diesel (Palmgren et al., 1999). Biogenic sources are also important contributors to NMVOCs emissions.

In conclusion, the on-road transport is an important contributor to a broad range of pollutants emissions in the urban areas. The emitted amounts depend on the vehicle fuel, the cubic capacity of the engine, the age and weight of the vehicle and the circulation speed. The development of strategies to abate the emissions of this sector is crucial in order to improve the urban air quality. Despite the fact that fuel specifications as well as the end-of-pipe technologies have reduced the emissions per kilometre by several orders of magnitude for some pollutants during last decades (with the exception of CO_2), increased travel demand and traffic congestion have severely offset the expected beneficial effect on air quality (Panis et al., 2006).

1.2 Air quality related legal framework

The processes leading to final concentrations of pollutants in the atmosphere are complex. Emissions rates, gas phase and heterogeneous chemical reactions and transport patterns affect the final levels. The efforts to improve air quality conditions focus on anthropogenic emissions reductions, designed to attaint fixed goals based on population and ecosystems exposure. The European Union (EU) develops a legal framework related with air quality that all the member countries have to accomplish. The European initiatives have historically focused on different hot spots: acidification, eutrophication, etc.; but they have been recently reoriented to an integrated approach, considering different hot spots at once. The air pollution related directives can be classified in two groups: (1) pollutants emissions control and (2) air quality targets.

1.2.1 Pollutants emissions control in Europe

In order to control the air pollutants emissions the EU establishes a series of directives. Among others in 2001 the EU approved the National Emission Ceilings –NEC- directive (2001/81/EC) that set upper limits for each Member State for the total emissions in 2010 of the four pollutants responsible for acidification, eutrophication and ground-level ozone pollution (SO_2, NO_x, VOCs and ammonia). Parallel to the development of the EU NEC Directive, the EU Member States together with Central and Eastern European countries, the United States and Canada have negotiated a "multi-pollutant" protocol under the Convention on Long-Range Transboundary Air Pollution framework (CLRTAP, developed by the UNECE in 1979), the so-called Gothenburg protocol, agreed in November 1999, which sets equal or lower emissions ceilings.

On the other hand, some European directives are oriented to reduce the emissions of specific activities, such as large power plants (2001/80/EC) or waste incineration (2000/76/EC); or specific pollutants, such as VOCs (1999/13/EC, 94/63/EC) or SO_x, indirectly limited by controlling the sulphur content in liquid fuels (1999/32/EC).

The emissions of motor vehicles have originally been regulated by Directive 70/220/EEC (light duty vehicles - LDV) and 88/77/EC (heavy duty vehicles - HDV) and amendments to those directives. The proposed limits have been tightened since then, resulting in the Euro limits (see Table 1-1)

Table 1-1 European limits for on-road vehicles emissions

Limit	Year of implementation	Vehicles affected	European Directive
Euro 1	1993	LDV	91/441/EEC 93/59/EEC
Euro 2	1996	Cars	94/12/EC; 96/69/EC
Euro 3	2000	All	98/70/EC
Euro 4	2005	All	98/70/EC; 2002/80/EC
Euro 5	2008	All	2005/55/EC; 2005/78/EC; COM (2005) 683*
Euro 6	2014	LDV	COM (2005) 683*

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* This proposal has been formally accepted by the Council on May, 2007.

1.2.2 European air quality targets

The emission control strategies intend to achieve acceptable air quality levels. The WHO recommended air quality thresholds to the human health protection are usually taken as a basis for the regulatory measures of a large number of countries around the world, among them the European countries.

Table 1-2 European air quality standards and WHO guidelines to human health protection.

Pollutant		Limit value	Date	Reference
O_3	WHO	100 $\mu g\ m^{-3}$ 8-hr average		2005
	EU / BOE	120 $\mu g\ m^{-3}$ 8-hr average (1)	2010	Dir 2002/3/EC. RD 1796/2003
		180 $\mu g\ m^{-3}$ 1-hr average (2)		
		240 $\mu g\ m^{-3}$ 1-hr average (3)		
PM_{10}	WHO	50 $\mu g\ m^{-3}$ 24-hr average		2005
	EU /BOE	50 $\mu g\ m^{-3}$ 24-hr average(4)	2005	Dir 1999/30/CE RD 1073/2002
		40 $\mu g\ m^{-3}$ Annual mean		
		50 $\mu g\ m^{-3}$ 24-hr average(5)	2010	
		20 $\mu g\ m^{-3}$ Annual mean (6)		
SO_2	WHO	20 $\mu g\ m^{-3}$ 24-hr average		2005
	EU / BOE	350 $\mu g\ m^{-3}$ 1-h average (7)	2005	Dir 1999/30/CE RD 1073/2002
		125 $\mu g\ m^{-3}$ 24-hr average (8)		
NO_2	WHO	200 $\mu g\ m^{-3}$ 1-hr average		2005
		40 $\mu g\ m^{-3}$ Annual average		
	EU	200 $\mu g\ m^{-3}$ 1-hr average (9)	2010	Dir 1999/30/CE RD 1073/2002
		40 $\mu g\ m^{-3}$ Annual mean		
Pb	WHO	0,5 $\mu g\ m^{-3}$ Annual mean		
	EU / BOE	0,5 $\mu g\ m^{-3}$ Annual mean	2005	Dir 1999/30/CE. RD 1073/2002
C_6H_6	WHO	--	--	--
	EU/BOE	5 $\mu g\ m^{-3}$ Annual mean	2010	Dir 2000/69/CE RD 1073/2002
CO	WHO	10 mg m^{-3} 8-hr average		
	EU/ BOE	10 mg m^{-3} 8-hr average	2005	Dir 2000/69/CE RD 1073/2002

(1). It won't be surpassed more than 76 times during 3 years

(2) Population information threshold

(3) Alert threshold

(4) It won't be surpassed more than 35 times a year.

(5) It won't be surpassed more than 7 times a year.

(6) Under revision

(7) It won't be surpassed more than 24 times a year.

(8) It won't be surpassed more than 3 times a year.

(9) It won't be surpassed more than 18 times a year

Under the framework of the CLRTAP several protocols to abate specific air pollution problems were put into practice (i.e. acidification or haze control protocols). Lately the EU policies have a multi-pollutant multi-effect approach. The most recent initiative is the Clean Air For Europe (CAFE) program launched in 2001. CAFE pretends to establish a long-term, integrated strategy to tackle air pollution and to protect against its effects on human health and the environment, under the framework of the 6th Environment Action Program. CAFE addresses health and environmental problems related to fine particles, ground-level O_3, acidification and eutrophication (EEA, 2007b).

The air quality targets currently established by the EU are shown in Table 1-2. The recently approved Directive 2008/50/CE, which has to be transposed to national legislation of the member states before the 11 June, 2010, compiles all these air quality thresholds and it adds a limit for $PM_{2.5}$ concentration. This is defined as an annual average of 25 µg m^{-3} to be achieved in 2015 and 20 µg m^{-3} in 2020.

1.3 Strategies to reduce on-road traffic contribution to urban air pollution

The European policies reflect the growing interest of developed countries in improving their air quality by testing different kind of initiatives (Nagl et al., 2006). The main tools for air quality management continue being the use of policies (firstly adopted by the Alkali Acts in UK, 1863-1874), the economic coercion and incentives to behaviour changes (from the Clean Air Acts of the 50's and 60's); and the reductions on emissions thanks to technological improvements in vehicles and industries (Langston, 1990)

As aforementioned, some of the efforts in emissions reduction focus on on-road transport, because of being a large contributor to primary air pollutants, especially on urban areas. Transport is today fuelled to a very large extent by oil. This situation has implications for energy consumption, greenhouse gas emissions and air quality. Moreover, on-road transport causes noise pollution and congestion.

Currently, the strategies applied to reduce traffic impacts on urban areas have mainly two orientations: (1) the reduction of km travelled (minimization of the number of vehicles and/or the distance travelled per vehicle); and (2) the reduction of unitary emissions by vehicle.

In the former group the public transport improvement, both from the infrastructural and service point of view, parking places restriction, roads construction to high occupation vehicles (taxis, buses or private cars with a high occupation rate), and fitting out cycle paths stand out. Also the introduction of taxes to use urban infrastructures contributes to reduce the congestion in urban zones (EC, 2001a). Parking taxes are the simplest example, but some cities are testing more elaborated formulas, like London, which has introduced an urban toll system with electronic car identification and electronic taxes collection system (Beevers and Carslaw, 2005).

In the second case, the introduction of alternative fuels and new technology vehicles or the circulation speed variation are the main paths to achieve reductions on emissions by vehicle. The EU White Paper on transport policy (EC, 2001a) points out that the urban transport provides a useful market for expanding the use of alternative energies. Several European cities have taken this path: Paris, Florence, Stockholm, Luxembourg, Barcelona or Madrid, have yet natural gas, biofuel or hydrogen fuel cell urban buses. Gradually, private cars and trucks could turn to substitution fuels. The EU Green Paper towards a European strategy for the security of energy supply (EC, 2000) lays down the foundations to the energetic diversification. The Proposal for an Action Plan and two European Directives (EC, 2001b) pretends to introduce up to a 20% of alternative energies in transport at in 2020. It bets for biofuels in the short term (EC, 2006), natural gas in the medium-term and fuel cells or hydrogen internal combustion engines in the long term.

1.3.1 Reduction of the km travelled (minimization of the number of vehicles and/or the distance travelled per vehicle)

The management options addressed to reduce the km travelled affect the final user behaviour. A brief summary of this kind of strategies is reflected in Table 1-3

The public transport improvement is fundamental in order to reduce the private transport, but seems to be insufficient itself to decrease the number of vehicles circulating by urban areas. The strategies that administrations have to follow to implement a competitive and efficient public transport net are out of the scope of this work, but they require necessarily of economic investments in infrastructures and the development of an accessible and competitive transport network.

Table 1-3 Urban on-road transport management strategies (1)

Strategies addressed to reduce the number of vehicles circulating in urban areas
1. Public transport improvement
2. Urban tolls
3. Car sharing promotion
4. Parking taxes
5. Circulation restrictions to specific type of vehicles in sensitive areas or in specific days
6. Urban planning

Concerning economical coercion measures, the payment of parking taxes is the most common of those applied. Its main purpose is to reduce the number of vehicles circulating, especially on city centres. The next step in this kind of strategies is the urban toll, which is based on the payment of a tax by the drivers to have access to an area or the whole conurbation. This strategy was firstly adopted in Singapore in 1975 with the main objectives of reducing air pollution and traffic congestion in the city. This system proved to be efficient, combined with circulation licenses by areas schemes and public transport improvements (Chin and Koh, 1989; Chin, 1996). From the 80's to the 90's reductions on NO_x were observed, meanwhile CO and smoke levels remained practically invariable. In 1998 the system was improved by introducing an Electronic Road Pricing System much more selective, taking into account vehicles types and maximum congestion hours. This renewed urban toll achieved reductions on traffic flows of a 20% and increased average circulation speed on a 33% (Tuan Seik, 2000).

The urban tolls have been implemented in different cities around the world. Oslo and other Norwegian cities have urban tolls since the 90's, Toronto was the first conurbation in establishing a fully electronic toll system in 1997, Melbourne has adopted this system in 2000 and in London it works since the year 2003. In the latter case reductions on NO_x achieve 12.0% and on PM_{10}, 12.9%, from 2002 to 2003 in the affected area (Beevers and Carslaw, 2005)

Finally, the prohibition of specific kind of vehicles or the circulation during critical days is the most drastic of the measures that could be adopted, which is used in very polluted urban areas, such as Mexico City. Additional strategies such as bike roads construction or car sharing promotion would improve the effects of the aforementioned measures.

On the long term, urban planning taking into account an integrated urban plan, infrastructures and public transport management seems to be the best option to reduce atmospheric pollution and congestion (Joumard et al., 1996). The worldwide example in this sense is the Curitiba city in Brazil. This kind of planning involves taking into account the existing urban structures, therefore the ideal case of study of Curitiba is not always achievable; nevertheless improvements in current urban planning can be done: ring roads and high speed roads construction, public transport improvement, or designing of business and commercial areas in order to reduce daily trips.

The efforts to solve air quality problems introducing these kinds of measures could not always achieve the desirable effects. They seem to be effective in reducing the vehicles number circulating in the affected areas, but may produce an increase in traffic density in adjacent roads and the effects of the subsequent circulation speed changes could not always be positive in terms of vehicles emissions. Even if the vehicle technology and unitary emissions are not the unique factor that sets final emissions, but also traffic density plays an important role (Mitchell et al., 2005; Irving and Moncrieff, 2004), it has to be considered that the cleaner and more efficient technologies introduction normally involve benefits not only in air quality terms but also in greenhouse gases emissions budget (Fenger, 1999). In any case, the success of this kind of policies depends on the characteristics of the conurbation and the air quality effects may be studied for each particular case

1.3.2 Reduction on unitary emissions by vehicle

The reduction on unitary emissions by vehicle can be achieved: (1) by **changing circulation speed** - Table 1-4, column A -, or (2) by **changing the vehicle technology or introducing alternative fuels** - Table 1-4, column B–

The strategies addressed to produce changes in circulation speed could have both positive and negative effects on air quality. The hot emissions of vehicles vary as a function of the speed, presenting normally a U shaped function with a minimum at medium speeds (i.e. CO emission factors for petrol and diesel cars are minimum at around 70 km h^{-1}, see Figure 1.4). Nevertheless this variation depends not only on the type of vehicle, but also on the specific pollutant that is considered (Figure 1.4) and therefore it does not exist a unique optimal circulation speed. Meanwhile the measures addressed to change circulation speed also pretend to reduce congestion, so that they would limit the acceleration and deceleration effect on emissions

Table 1-4 Urban on-road transport management strategies (2)

Strategies addressed to reduce unitary emissions by vehicle		
(A) Changing circulation speed	(B) Introducing alternative fuels or new technologies	
1. Ring roads and highways construction	1. Low sulphur fuels	5. Particulate filters
2. Specific speed limits by roads or city areas	2. Natural gas	6. Catalyst systems
	3. Biofuels	7. Hybrid vehicles
	4. Hydrogen	8. Electric vehicles
		9. Fuel Cells

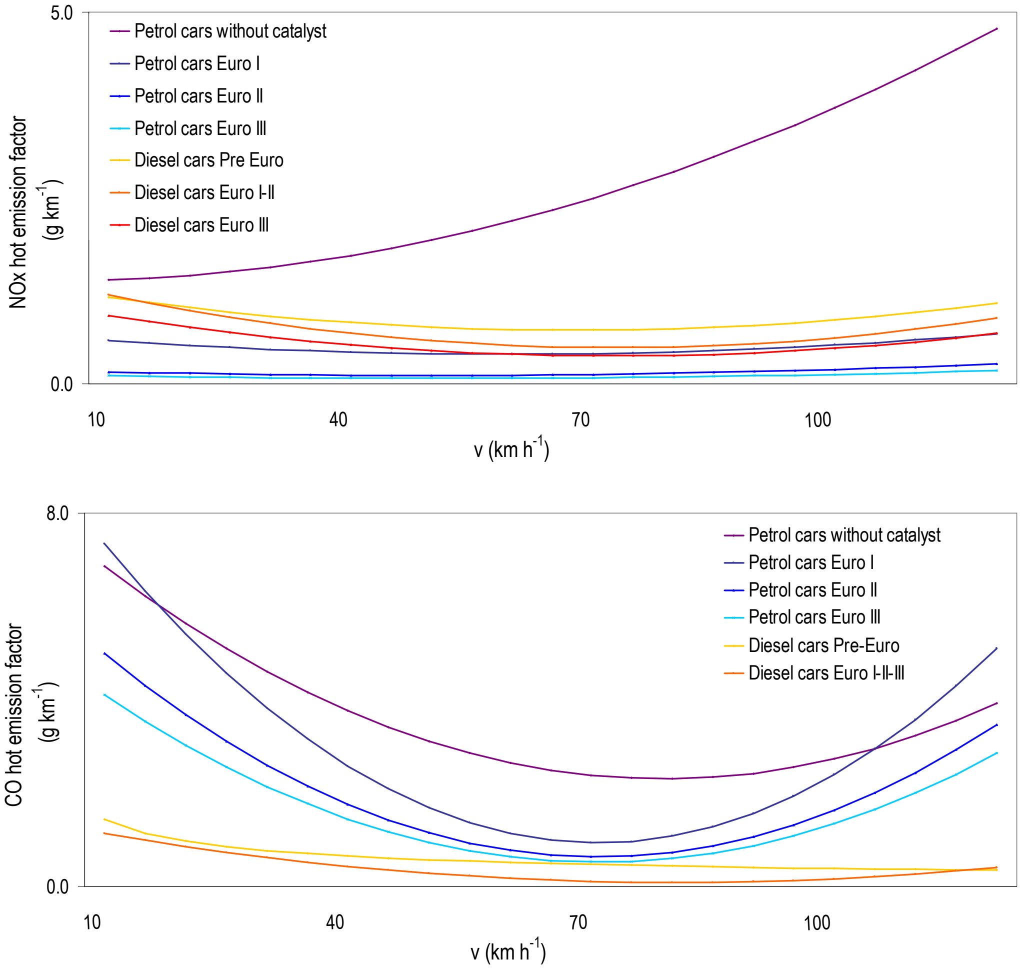


Figure 1.4 Example of NO_x (up) and CO (down) hot emission factors for petrol and diesel passenger cars accomplishing different emissions standards (Ntziachristos and Samaras, 2000)

The speed limits can be applied in the short term and they are being widely used nowadays. For example in cities such as Barcelona, where different neighbourhoods are obliged to run at speeds lower than 30 km h^{-1} and the highway access to the urban area are limited to 80 km h^{-1}.

In the medium or long term, decongestion and circulation speed changes can be achieved by ring roads or highways construction. This kind of strategies normally involve reductions on emissions during an initial period (Monzón and Villanueva, 1996; Baldasano and Soriano, 1999), nevertheless the growing number of vehicles (Figure 1.5) and the increased activity promoted by new infrastructures appearance makes the increase in roads capacity insufficient for long periods of time. (i.e. the Rondas ring-roads implementation in Barcelona had positive effects in air quality and emissions in the urban area from 1992 to 1996, when they reached the saturation (Baldasano and Soriano, 1999)), moreover the effects of this kind of measures are normally positive in city centres, but not always in the surrounding areas (i.e. the M40 implementation in Madrid involved environmental benefits in the city centre and a global reduction on emissions due to decongestion, but the emissions moved to surrounding areas (Monzón and Villanueva, 1996))

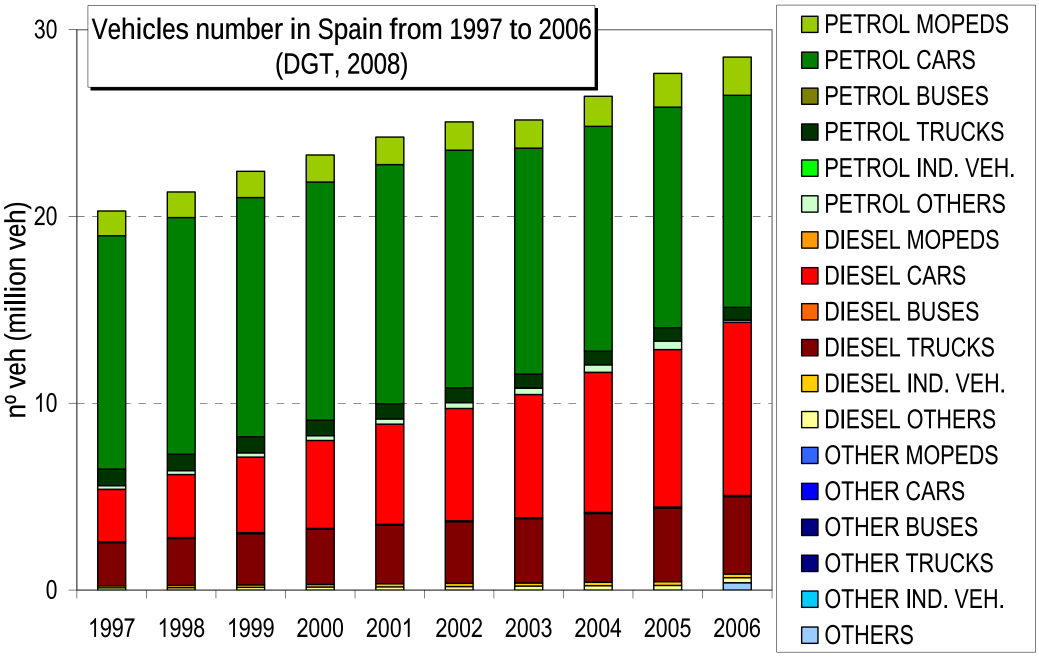


Figure 1.5 Evolution of the number of vehicles in Spain from 1997 to 2006 (source: DGT, 2008)

The strategies directly applied to vehicles, such as technological improvements in existing vehicles, new technology vehicles introduction or alternative fuels use, are the most efficient in order to reduce air pollutants emissions and fuel consumption (Jorgensen et al., 1993).

The technological improvements have been put into practice during last 20 or 25 years, starting with the three ways catalysts for gasoline vehicles, the oxidizing catalyst for diesel vehicles, the turbo compressor system or the direct injection. They have augmented the energy efficiency of the engines, reducing the tailpipe emissions.

For example the particulate matter filters introduction in diesel vehicles, reduced the finest fraction of particulates with an average efficiency of 90.0 to 99.9% (Joumard, 2005). Nevertheless, in some cases the new technologies increase some specific pollutant emissions; e.g. diesel oxidation catalyst systems or particulate filters may produce a growth in NO_2 emissions (Carslaw et al., 2007) and the introduction of Euro 3 and Euro 4 standards with three ways catalysts in gasoline passenger cars involves NH_3 emissions being larger than those of NO-NO_2 (Heeb et al., 2008). Therefore these changes have implications on air quality, in this specific case affecting photochemical ozone formation and related irritants, changing the oxidative strength of the atmosphere, as well as the urban aerosol compositions. The trend in newer technologies involves the introduction of electric devices, combining them with internal combustion engines, which is known as hybrid technology, or alone, in the electric vehicles.

The hybrid engines present different configurations; normally they recover the braking energy to produce electricity that is stored in the batteries. It is also possible to charge the vehicle batteries by driving at the optimal speed, which guarantees the maximum engine efficiency. They reduce emissions by saving fossil fuel consumption; nevertheless their behaviour depends largely on driving conditions (Jeanneret et al., 1999; Joumard, 2005) and it is usually optimum at urban driving.

The pure electric vehicles involve zero tailpipe emissions, nevertheless indirect emissions on the energy production sites have to be taken into account and they will depend on the electrical mix generation of the specific country or region. Currently they are not very extended due to the high costs of use, their limited autonomy and relatively low efficiency (Joumard, 2005), together with the low lifetime of the batteries.

In parallel to these advances alternative fuels are being used or tested nowadays. The natural gas, compressed or liquefied, can be currently used for a large range of vehicles types. Its main advantage is the reduction on particulate matter, NMVOCs and sulphur compounds emissions. Worldwide Argentina and Brazil are the countries which account for the largest natural gas vehicles fleets, and in the European context Italy stands out. In Spain natural gas urban buses and garbage trucks are already circulating in several cities, such as Barcelona and Madrid.

The biofuels provide an alternative to fossil fuels of vegetal origin. Bioethanol (mainly ETBE, ethyl-tert-butil-ether) and biodiesel (mainly FAME, fatty acid methyl ester) can be used as substitutes or additives for gasoline and diesel fossil, respectively, involving lower emissions of sulphur compounds and particulates. Their main advantage is the reduction of the introduction of new carbon atoms in the global carbon budget. Nevertheless they are being cause of controversy because of their implications in biodiversity loss or even in the food security supply (FAO, 2007).

Compressed hydrogen can be used as a fuel in internal combustion engines, which involves a large reduction on NO_x emissions, particulates and VOCs and reduces to zero those of CO. This kind of technology is still in development. Nevertheless, the use of hydrogen in vehicles seems to be oriented to fuel cells. A fuel cell is a device that transforms directly the chemical energy in electrical energy, by means of an electrolytic process. They are also in a development stage and are being tested i.e. in Europe under the CUTE project, which has implemented fuel cell urban buses propelled with hydrogen in different European cities, such as Madrid and Barcelona. The reaction product is water vapour; therefore the pollutant emissions attributed to this kind of vehicles would be just those concerning the hydrogen production, transport and supply. The well to wheel studies are the most adequate to define the environmental impacts of this kind of technologies, i.e. if the hydrogen is produced from natural gas in the EU, the reduction in final emissions is around 30% respect to traditional fuel use (Joumard, 2005).

In the next 25 years the continuous growth of the vehicles use could cancel the effects of technological improvements; therefore an exclusively technological answer to environmental problems related with transport could not be expected (Cannibal and Lemon, 2000). On the other hand, the new technologies and alternative fuels introduction may have secondary impacts, such as unexpected changes in emissions composition (Richter and Williams, 1998; Fenger, 1999).

The main conclusion is that it does not exist a universal strategy that could be applied to different geographical locations expecting similar results and to achieve effective reductions in traffic derived emissions. Combinations of measures may be applied, focused on reducing both the number of vehicles circulating and the unitary emissions by vehicle, in the short, medium and long-term.

1.4 Tropospheric chemistry

The final concentrations of pollutants within an area depend not only on the primary pollutants emitted amounts and the transport patterns due to meteorological conditions, but also on the chemical transformations that take place in the low troposphere. This section provides a brief description of secondary pollutants formation in the troposphere; it is not on the scope of this chapter to thoroughly analyze each process leading to atmospheric pollutants formation or disappearance. A deeper analysis of gas phase chemical processes and heterogeneous chemistry can be found in Atkinson (2000), Jacob (2000) and Meng et al. (1997). Note that some uncertainties concerning the kinetics of some reactions and certain reaction paths persist until nowadays, this work does not pretend to deep in these uncertainties, but to have them present in order to better understand the results.

In summary, the main products of gas-phase atmospheric chemistry are ozone and oxidized compounds. Atmospheric oxidation proceeds via chains of radical reactions, which can be long and complex in the case of organic compounds. The predominant hydrocarbon in the atmosphere is methane, but a large number of other hydrocarbons and organic compounds with anthropogenic and biogenic origin are present. Photochemical ozone results from the interaction of emissions of nitrogen oxides (NO_x) and non-methane volatile organic compounds (NMVOCs).

In a clean atmosphere, the O_3 is photochemically formed from the photolysis of NO_2 at wavelengths lower than 424 nm (reaction [1.1]) and because O_3 reacts rapidly with NO (reactions [1.2] and [1.3]) these reactions result in a photoequilibrium between NO, NO_2 and O_3 with no net formation or loss of O_3. However, the degradation reactions of VOCs lead to the formation of intermediate $RO_2\cdot$ and $HO_2\cdot$ radicals. These $HO_2^{\cdot}$ and $RO_2^{\cdot}$ radicals react with NO, converting NO to NO_2, which then photolyzes to form O_3 (Figure 1.6), this process results in net formation of O_3 (Jacobson, 1999; Atkinson, 2000):

$$NO_2 + h\nu \rightarrow NO + O^{\cdot} \qquad [1.1]$$

$$O^{\cdot} + O_2 + M \rightarrow O_3 + M \qquad [1.2]$$

$$O_3 + NO \rightarrow NO_2 + O_2 \qquad [1.3]$$

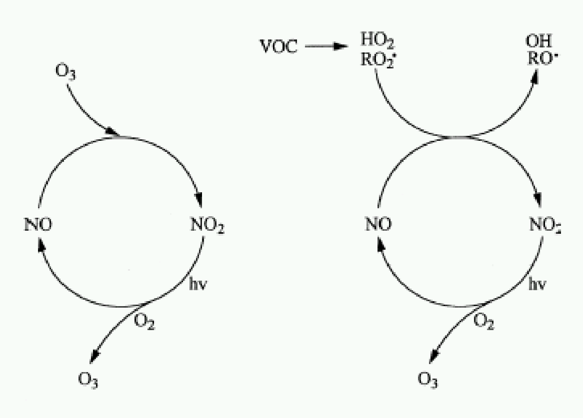


Figure 1.6 Schematics of the reactions involved in NO-to-NO_2 conversion and O_3 formation in absence of VOCs (right) and in the presence of VOCs (left) (Atkinson, 2000)

In the lower troposphere, chemical reactions of anthropogenic and biogenic VOCs and anthropogenic NO_x emissions dominate over those of methane and its degradation products. Chemistry of the troposphere is significantly complicated because of the presence of many volatile organic compounds of various classes: alkanes, alkenes, alkynes, aromatics, aldehydes, ketones, alcohols, etc. and the complexity in the chemistry of these organic species, which in some cases involves uncertainties related with kinetics or reaction paths.

Radicals have a decisive role in tropospheric chemistry because they are very reactive species that rapidly attack different atmospheric pollutants, since they own a free electron in their structure that can be transferred to other species in chemical reactions. Most important radicals are oxygen atoms ($O\cdot$), hydroxyl ($OH\cdot$), hydroperoxyl ($HO_2^{\cdot}$), peroxyalkyl ($RO_2\cdot OH$), alkyl ($RC\cdot H_2$) and alcoxy ($RO^{\cdot}$), however hydroxyl radical dominates atmospheric photochemistry, both in a polluted or clean troposphere, initiating chain reactions when attacking VOCs and CO. On the other hand, the methane and CO reactions are the dominant loss processes for the OH radical in the clean troposphere.

Among these reaction chains, the role of alkenes is outstanding. They are important contributors to overall ozone formation, because of the double bonded carbon atoms. Photolysis and the initial reactions of many VOCs with OH radicals and NO_3 radicals lead to the formation of alkyl or substituted alkyl ($RO^{\cdot}$) radicals, and the reactions of O_3 with alkenes and other VOCs containing "C=C" bonds lead to the formation of organic peroxy ($RO_2^{\cdot}$) radicals. A generalized tropospheric degradation scheme which is applicable for most VOCs (Figure 1.6) involves the intermediate organic radicals. Generation of the organic peroxy radicals occurs largely by the attack of the hydroxyl radical ($OH^{\cdot}$) on hydrocarbons. The main steps of this process are the formation of the hydroperoxyl radical and conversion of NO to NO_2.

On the other hand, ozone photolysis produces an excited singlet $O(^1D)$ oxygen atom (reaction [1.4]) that reacts with water vapour to form hydroxyl radicals - reaction [1.5] - (Wayne, 2000).

$$O_3 + h\nu \rightarrow O(^1D) + O_2 \qquad [1.4]$$

$$O(^1D) + H_2O \rightarrow 2\ OH^{\cdot} \qquad [1.5]$$

$OH^{\cdot}$ and $HO_2^{\cdot}$ act as catalysts in these reaction cycles, they are not consumed and the cycles occurs until a termination reaction takes place, the most common is the elimination of $OH^{\cdot}$ by reaction with NO_2 to form gaseous nitric acid (reaction [1.6]).

$$OH^{\cdot} + NO_2 + M \rightarrow HNO_3 + M \qquad [1.6]$$

Peroxy Acetyl Nitrates (PAN) and analogues are formed during degradation of aldehydes, by the reaction of alkyl and acyl peroxy radicals ($RO_2\cdot$) with NO_2 and it is also originated by the ketones degradation reactions. The PAN molecule serves as a temporary reservoir for peroxyacetyl radical and NO_2, because the formation reaction reverses thermally (reaction [1.7]), having PAN molecule a lifetime of 45 minutes at 298 K.

$$CH_3C(O)O_2^{\cdot} + NO_2 \leftrightarrow CH_3C(O^{\cdot})O_2NO_2 \qquad [1.7]$$

In addition, the heterogeneous and aqueous reactions in which aerosols (liquid and solid particles) and clouds take part can affect the O_3 and oxidants concentrations in several ways, promoting the generation and disappearance of hydroxyl radicals and NO_x, direct O_3 looses and halogen radicals production (Jacob, 2000).

Therefore O_3 and PM are chemically coupled, and this coupling is of profound importance in understanding processes that control the levels of both (Meng et al., 1997).

Condensed water is the predominant form of suspended matter in the troposphere, and clouds the most abundant form of condensed water (Wayne, 2000). Moreover, among aerosols sulphates (SO_4), nitrates (NO_3), ammonia (NH_4), organic compounds and crustaceous material are present, that reach the solid or liquid phase by different processes (Figure 1.7)

Heterogeneous and clouds chemistry is linked with radicals chemistry, i.e. the $HO_2^{\cdot}$ is scavenged by cloud droplets because of acid-base dissociation. In fact, the secondary aerosols formation depends intimately on the presence of ozone precursors, essentially NO_x and VOCs (Meng et al., 1997). The VOCs and NO_x chemistry is controlled by the hydroxyl radical ($^{\cdot}OH$) which levels depend on the VOCs composition and the VOCs/NO_x ratio (Bowman and Seinfeld, 1994). The cyclic system of $^{\cdot}OH$-$HO_2^{\cdot}$ transformation generates tropospheric O_3 and H_2O_2, which act as oxidants of the SO_2 forming sulphates The hydroxyl levels set the oxidizing rates of SO_2 and NO_2 to sulphuric and nitric acid, respectively, which are the gaseous precursors of sulphate and nitrate. Besides the hydroxyl radical and O_3 can attack the hydrocarbon chains (RH, with R larger than 7 C) producing the secondary organic aerosols formation.

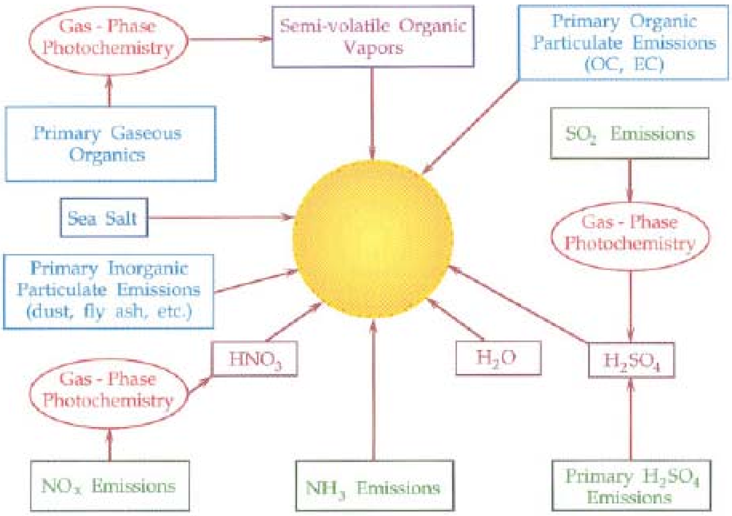


Figure 1.7 Routes of incorporation of chemical species into atmospheric particulate matter (Meng et al., 1997).

1.5 Atmospheric modelling as a management tool

There is a growing interest in evaluating the impacts of the environmental management strategies. Nowadays assessing the most suitable strategies for on-road traffic emissions abatement in urban areas is of special concern, and constitutes a complex problem due to the non linearity of the chemical and atmospheric processes leading to final concentrations of pollutants.

Currently, the main tools addressed to urban air quality management are: (1) **Air quality stations data analysis**; (2) **Emissions estimation and analysis** and (3) **Atmospheric modelling**. Though their individual analysis has been used in the past for air quality management purposes, these are complementary tools that have to be considered as a whole. This allows a better evaluation and understanding of the effects of the emissions abatement strategies. In some cases the air quality models are combined with models that provide different indicators, such as population exposure, water or soil quality effect, externalities estimation, etc. These models are commonly known as **integrated evaluation models**.

1.5.1 Air quality stations data analysis

It is fundamental to know the real efficiency of the control measures put into practice. The most accurate tool to this purpose is the data analysis from air quality stations. This analysis provides real variations in air quality due to the application of the management strategy (Chin, 1996; Beevers and Carslaw; 2005, Bono et al., 2001). Its main limitations are the lack of data, the low density of the air quality stations nets, and the fact that it is an evaluation tool “a posteriori”, it doesn’t allow us to know the air quality variations before implementing the emissions abatement measure.

1.5.2 Emissions estimation

The emissions estimation is essential to evaluate the effect of a management strategy introduced. It could be used as an evaluation parameter itself (Baldasano and Soriano, 1999; Burón et al., 2005; Faiz et al., 2006). Nevertheless due to the non-linearity of the atmospheric chemistry and the transport processes that occur in the atmosphere it is not possible to establish a direct relationship between emissions reductions and local air quality improvement.

1.5.3 Atmospheric modelling

Atmospheric modelling, used as a management tool, permits to evaluate the possible initiatives in advance, helps decision making, and it could be also used to evaluate measures already in practice. The complexity of tropospheric chemistry and atmospheric processes leading to final pollutants concentrations makes atmospheric modelling the most suitable tool to assess the effects of emissions abatement strategies.

The Lagrangian or Gaussian models use is limited nowadays, in spite of being largely used in the past due to their simplicity and low computational requirements. The increase in computational capacity and the larger database capacity and availability during last years, together with advances in the knowledge and understanding of atmospheric processes lead to the extended use of the Eulerian grid modelling systems. Currently the most advanced are the 3rd generation systems, which are three-dimensional, non-hydrostatic, Eulerian grid models. They fundamentally include three modules: (1) an emissions model, which describes the spatio-temporary distribution of emissions both natural and anthropogenic, (2) a meteorological model, which describes the state and evolution of the atmosphere in which these emissions are introduced, and (3) a chemical transport model, which describes the chemical transformations that take place for this specific emissions and meteorological situation (Seinfeld, 1988; Russell and Dennis, 2000).

Eulerian grid models are the most efficient method to simulate atmospheric dynamics, photochemistry and secondary aerosols formation. Used as management tools are versatile and permit a detailed definition of reduction scenarios. Their main limitations were the large databases and the computational capacity they needed (Seinfeld, 1988), but advances had been done in this field.

In order to study urban air quality local to regional models are the most adequate, nevertheless if the O_3 dynamics have to be captured is necessary to apply a regional scale (Moussiopoulos, 1996). The regional component plays an important role in local or urban air quality, especially in complex topography areas, such as coastal regions (Fenger, 1999). Different studies in Mediterranean regions showed that urban areas can be affected significantly by emission sources located hundreds of km far away (Kallos, 1998; Jiménez et al., 2006). It is essential to adequately define not only the working scale but also the spatial resolution, which may be as fine as possible (Reis et al., 2000) in order to better define the high pollutants concentrations of in urban areas and control the effects of reducing emissions. Finally, an accurate definition of the meteorological conditions and atmospheric dispersion mechanisms is also important to set urban air quality (Giovannoni et al., 1995; Vignati et al., 1996).

The Eulerian grid models have been widely used to perform management tasks in urban areas, such as the Mexico City; where the application of mesoscalar atmospheric models plays a fundamental role among the analysis, management and decision tools implemented to improve air quality conditions and help decision makers (Streit and Guzmán, 1996).

In the European context Eulerian grid models have been also widely used as management tools. From the sensitivity studies performed to assess the effects of hypothetical reductions on emissions (i.e. evaluation of the effects reductions on NO_x and VOCs emissions (Simpson, 1995)), to the emissions scenarios evaluation, which are performed from a more realistic point of view (Simpson and Andersson-Sköld, 1997). Also predictions of future emission scenarios have been tested (Reis et al., 2000). To do so, a wide range of models were used, i.e. the EMEP (European Monitoring and Evaluation Program) three-dimensional oxidation model (Jonson, 1999; Simpson and Jonson, 2000; Reis et al., 2000), the mesoscalar Eumac Zooming Model –EZM- (Vinuesa et al., 2001), the CHIMERE multi-scale model or the CAMx Comprehensive Air Quality Model –CAMx-, both used in the CityDelta Project (Cuvelier et al., 2007) or the Models 3 Community Multiscale Air Quality – CMAQ - model (Jiménez and Baldasano, 2004).

One of the most ambitious studies related with on-road traffic management and air quality in Europe was performed under the Auto-Oil II program (AOPII, 2000). It intended to make an assessment of the future trends in emissions and air quality and to establish different policy options for on-road traffic, technologically possible and cost-effective, in order to attain the EU15 air quality objectives for 2010. It took into account 10 European cities, among them Madrid. It projected that in broad terms applying the Auto-Oil I initiative measures the emissions from road transport would fall by 70-80% between 1995 and 2010 and indeed fall further beyond, as Euro 4 technologies continued to penetrate the market from 2005 onwards. It pointed that similar reductions could not be expected for CO_2 emissions from road transport, which were expected to be 10-15% higher in 2010 than in 1995. The remaining air quality challenges were related to exceedances of the NO_2 and PM_{10} objectives in urban areas, and VOCs emissions reductions. It proposed different measures both from the environmental and the economical point of view: the introduction of tighter emission standards for motorcycles, the promotion of enhanced environmentally friendly (EEV) passenger cars, changes in fuel specifications for gasoline and diesel (though not on sulphur or aromatics in gasoline which had been fixed for 2005), promotion of alternative fuels in captive fleets, local measures including parking charges, differentiated road pricing, improved infrastructure, public transport priority, improved logistics for freight, national scrappage schemes, fiscal instruments including fuel duty increases or replacement of registration or circulation taxes with fuel duty increases.

The CityDelta project is other European initiative that deserves mention; it was designed to evaluate the impact of emission reduction strategies on air quality at the European continental scale and in European cities. It focused on O_3 and PM comparing different models and configurations (Cuvelier et al., 2007). The main conclusions are the need for using fine scales to define urban air quality changes achieved by emissions reductions and that the elaboration of efficient emission control strategies requires a correct knowledge of the respective contributions of local and regional sources to urban air pollution levels. The regional background levels of O_3 and PM play a significant role, since they determine the part of the pollution that cannot be regulated at the local scale (Thunis et al., 2007).

An alternative approach to these kind of studies is the Integrated Resources Management concept, which has its origins in the 70's decade, and from ending 80's is used to describe forms of using and managing the environment and natural resources in order to reach a sustainable development (Irving and Moncrieff, 2004). It takes into account different criteria, in Europe is widely used: the CLRTAP (Convention on Large Range Transboundary Air Pollution) of the UNECE (United Nations Economic Commission for Europe), the RAINS- Regional Air Pollution Information and Simulation- (establishes strategies for the acid rain reduction in Europe), the ASAM focused on reducing acidification, euthrophication and O_3 excess in Europe (Mediavilla-Sahagún and ApSimon, 2003) could be understood as integrated evaluation strategies. Under the integrated evaluation models concept it is possible to find a wide range of systems that combine the estimation of more than one type of variables for the strategies evaluation to provide a solution for a specific problem. They can combine: external costs estimation, impacts on climate, transport mobility, air quality index, greenhouse gas emissions, precipitation, prediction of land use, transport and energy consumption or even human health effects or population exposure (Irving and Moncrief, 2004; Affum et al., 2003; Oudinet et al., 2005). Their main advantage is that they permit to evaluate the strategies from different points of view. Nevertheless large databases are needed and due to the large number of variables involved they entail also a large number of uncertainties (Cocks et al, 1998). The input data variation can led to significantly different optimal strategies prediction, therefore integrated evaluation models outputs may be always reviewed critically and not being accepted without questioning.

1.6 Objectives

The main objective of this PhD thesis is to **explore the capabilities of an atmospheric modelling system to assess the air quality effects of different strategies for on-road traffic emissions abatement in urban areas.**

Barcelona and Madrid, the two largest conurbations of Spain, are chosen as study areas. They are characterized by frequently suffering air pollution episodes. Moreover they constitute a typically coastal (Barcelona) and continental (Madrid) conurbation, located in the Northeastern and Central Iberian Peninsula, which allows comparing the response to the same strategy in those environments.

The improvement of urban air quality conditions is of special concern nowadays, the high pollutants levels and the large population exposed makes the selected areas ideal for the objectives of this study. A representative case of study is selected for the 2004 year, taking into account poor air quality conditions in these regions and avoiding distorting factors in the traffic circulation patterns (working days).

The WRF-ARW/HERMES/CMAQ modelling system accomplishes the requirements to perform this assessment. It is a third generation, Eulerian, three-dimensional model, representative of the state-of-the-art in air quality modelling, which can be applied with high temporal and spatial resolution to the study areas. Moreover the HERMES emissions model is specifically developed for Spain and takes into account on-road traffic emissions for Barcelona and Madrid conurbations, which permits the design of emissions scenarios in detail. A deeper description of HERMES can be found in Baldasano et al. (2008a).

These particularities allowed:

1) To characterize the atmospheric behaviour differences between a coastal and a continental environment in the Iberian Peninsula. The quantification of the contribution of atmospheric processes to the concentration of main gaseous pollutants provides the basis for establishing the causes and dynamics of air pollution in the studied regions. This constitutes a valuable information to better define strategies to improve the air quality in these areas.
2) To compile and discuss the currently available options to abate on-road traffic emissions, proposing several alternatives. The tested strategies are defined taking into account their feasibility in the short term and the current planning, from the European context to the local councils. The final selected strategies are oriented to reduce unitary emissions by vehicle and represent the short term most adopted schemes:

   a. Alternative fuels use: natural gas and biodiesel introduction.
   b. New technologies introduction: hybrid vehicles use.
   c. Circulation speed change: introduction of a 80 km h^{-1} speed limit, specifically planned for the Barcelona Metropolitan Area

3) To compare the strategies for on-road traffic emissions abatement in urban areas in terms of air quality, defining what the best options are. Their effects are evaluated during the vehicles use phase and mainly concern:
   a. Fuel consumption change
   b. Primary pollutants emissions variation
   c. Air quality levels change (not only in the urban areas, but also in the regions affected by the urban plumes).
4) To evaluate the different response to the same strategy for on-road traffic emissions abatement of both studied areas, taking into account particularities such as the main emission sectors, the specific vehicles fleet composition or the atmospheric behaviour.
5) To improve the on-road traffic emissions module of the HERMES model.
   a. Adding new vehicle categories to the urban vehicle fleet composition.
   b. Including the most suitable emission factors for the new technology vehicles or alternative fuels, previously analyzing the quality of the information to be included.
   c. Changing the speed representation in some roadways, from a constant circulation speed to an hourly circulation speed, which is evaluated in terms of air quality levels assessment.

1.7 Scope and structure of the document

This document presents the PhD Thesis "Assessing variations in urban air quality when introducing on-road traffic management strategies by means of high-resolution modelling. Application to Barcelona and Madrid urban areas."

Chapter 1 has described the current air quality situation around Europe, focusing on urban air pollution conditions. The southern Mediterranean region frequently experiences serious photochemical pollution episodes, normally associated to summertime meteorological conditions. Barcelona and Madrid are the largest conurbations of Spain and perfectly suit as cases of study. They permit to compare the response to strategies for on-road traffic emissions abatement in a coastal and a continental environment. As case of study two summertime polluted days of 2004 were selected, the 17-18, June, which are representative of poor air quality conditions in the study areas and fit in usual circulation patterns (working days).

This chapter also summarizes the current European legal framework to improve air quality conditions, centring the discussion in the study areas, which are directly affected by the European plans and programmes.

The on-road transport sector is the largest contributor to anthropogenic emissions in urban areas. The technological and planning options currently available to reduce its impact can be categorized in: (1) reducing the km travelled (minimizing the number of vehicles circulating and/or the distance travelled by vehicle) or (2) reducing the unitary emissions by vehicle.

The parking taxes, urban tolls, car sharing promotion, public transport improvements or urban planning schemes are found in the first group.

The **alternative fuels use**, **new technology vehicles** or the **circulation speed changes** are found in the second group. These were selected as cases of study. At least a representative scenario of each potential option was chosen, taking into account their feasibility in the short term. The scenarios design searches for a realistic implementation, considering market penetration of new vehicles, alternative fuels currently promoted or strategies already put into practice in the study areas.

The most suitable tool to evaluate the effects of these strategies is air quality modelling. As already stated, the third generation Eulerian three dimensional modelling systems are the most advanced tools in air quality modelling and allow to define the effects of management strategies in advance, taking into account not only emitted pollutants, but also specific meteorological conditions, tropospheric chemistry and topographic features that condition the final concentrations over an area. They are versatile and accomplish all the requirements for this study. Therefore the **WRF-ARW/HERMES/CMAQ modelling system** perfectly fits in this framework.

A deep description of the methodology and the modelling system used is provided on **Chapter 2**. In addition, the model is used to characterize the defined domains and to compare the atmospheric behaviour of the North-eastern Iberian Peninsula, where Barcelona is located, with the Central Iberian Peninsula, where Madrid is, by means of the Process Analysis tool implemented in the CMAQ photochemical model. It reflects the main differences in atmospheric dynamics between the coastal and continental environment and quantifies the different contributions to gas-phase concentrations of pollutants. This analysis is fundamental to design strategies for abating pollution and to better understand the effects of the on-road traffic management options.

The introduction of **alternative fuels** is currently taking place in the Barcelona and Madrid urban areas, moreover is promoted by the European Union legal framework, which specifically proposes the **natural gas** and **biofuels** use in the short term. Therefore these two options were selected as cases of study and several feasible scenarios of introduction of natural gas and biodiesel in the conurbations were designed.

Chapter 3 discusses the effects on urban air quality of substituting the oldest petrol and diesel vehicles of Barcelona and Madrid by **natural gas vehicles**. Seven scenarios affecting specific vehicle types were designed. The on-road traffic emissions module of HERMES is deeply described in this chapter to make more comprehensible the implementation of the new scenarios. The fuel consumption, emissions variation and air quality changes due to the natural gas fleets introduction is analyzed.

The bioethanol gasoline blends and the biodiesel diesel fossil blends are the most common biofuels used in the transport sector. This work is centred in evaluating their effects in air quality during the use phase.

The requirements of specific input data make the bioethanol scenarios design unviable with the available information. **Chapter** 4 considers the introduction of **biodiesel** as a fuel, both in Barcelona and Madrid. Two scenarios are designed taking into account different penetration rates. Their effects in fuel consumption, primary pollutants emissions and air quality are described.

Concerning **new technology vehicles**, the **hybrid vehicles** represent the first step in the path to the fuel cells or the pure electric vehicles, which are nowadays still in a development stage. Mainly the petrol hybrid electric cars are already being introduced in the markets. **Chapter 5** describes the implications on fuel consumption, emissions and air quality of the introduction of hybrid cars in Barcelona and Madrid. Two scenarios are defined taking into account an optimistic and a pessimistic market share during a 5 years period.

Finally the **modification of the circulation speed** in urban areas would lead to changes in the air pollutants emissions. One of the most used options to induce these variations is introducing speed limits. The planned strategy of the Generalitat de Catalunya government, which has imposed in January 2008 a 80 km h^{-1} speed limit in the Barcelona metropolitan area is tested in terms of air quality. The on-road traffic emissions are highly sensitive to speed changes. The availability of hourly variable speed data from measurement campaigns allowed improving the HERMES emissions model estimates, which for this particular case took into account the traffic congestion effect. **Chapter** 6 describes the improvement in air quality assessment achieved with the new data introduced in the HERMES emissions model. Also the 80 km h^{-1} scenario implementation and the main changes in fuel consumption, emissions and air quality levels are discussed.

Chapter 7 reflects the main conclusions of the present work.

The Chapters 2, 3 and 6 are presented as they have been submitted to an international SCI journal and include their own introduction, specific methodology, results, discussion and conclusions. Because part of the methodological aspects and data are common in different Chapters, some minor adjustments have been performed with respect to the journal version in order to avoid unnecessary repetitions. The Chapters 4 and 5 were adapted to the same format. Chapter 5 was presented in the HARMO12 conference in October 2008, and it is published as an extended abstract in the Proceedings of the HARMO 12 conference (Croatian Meteorological Journal, Vol. 42).

All references are compiled in a separated chapter: **Chapter** 8.

More details about the studied regions, the measured air quality levels, the meteorological conditions and the selected episode to perform this study can be found in the **Annex 1**.

2 WRF-ARW/HERMES/CMAQ modelling system. Quantification of the origins of atmospheric pollutants concentrations in the Northeastern and Central Iberian Peninsula during the 17-18 June, 2004 episode.

Published as: *Gonçalves, M., Jiménez, P., Baldasano, J.M., 2008. Contribution of atmospheric processes affecting the dynamics of air pollution in south-western Europe during a typical summertime photochemical episode. Atmospheric Chemistry and Physics Discussions 8, 18457-18497.*

2.1 Introduction

Atmospheric chemistry transport model simulations suggest that summertime O_3 is enhanced in the entire Mediterranean troposphere, contributing substantially to the radiative forcing of climate and air quality issues (Lawrence et al., 1999; Hauglustaine and Brasseur, 2001; Jiménez et al., 2006). Specifically the south-western Europe and the Iberian Peninsula (IP) frequently experience photochemical pollution episodes during summertime, in which O_3 and PM_{10} exceedances of the European air quality targets are registered. Atmospheric modelling is a fundamental tool to assess the air quality levels in these situations. Nevertheless, the air quality models usually provide the net concentration of pollutants, without the capabilities of understanding and isolating the atmospheric processes involved, which would explain the reasons for a model's predictions and show the relative importance of each process (Jeffries and Tonnesen, 1994; Jang et al., 1995a, 1995b). For the Mediterranean area, these issues have been studied by measurement campaigns, but also combining both modelling and experimental techniques (e.g. Millán et al., 1996; 1997; Lelieveld et al., 2002; Lawrence et al., 2003; Roelofs et al., 2003; Cros et al., 2004; Dufour et al., 2005; Jonson et al., 2006; among others). Currently, some Eulerian grid models, such as the Models-3 Community Multiscale Air Quality Model - CMAQ (Gipson, 1999), permits to assess the contribution of different atmospheric processes to the net concentrations of pollutants.

This analysis is useful not only for a better understanding of the atmospheric behaviour of an specific area, i.e. New York (Jang et al., 1995a, 1995b) or the Puget Sound Region –USA- (Jiang et al., 2002); but also for assessing which are the models or the models' configurations presenting better skills (Jeffries and Tonnesen, 1994; O'Neil and Lamb, 2005). The peer-reviewed works applying the process analysis focus mainly on episodic events (San José et al., 2002; O'Neil and Lamb, 2005), although annual (Zhang et al., 2006) and even climatologic simulations (Hogrefe et al., 2005) have been also performed.

This work pretends to assess and quantify the contribution of different atmospheric processes to the concentration of O_3 and its precursors in south-western Europe by means of the Integrated Process Rate (IPR) analysis tool available in the CMAQv4.6 model. In order to highlight the different behaviour of atmospheric dynamics in a coastal environment and an inland-continental zone, the Northeastern and Central Iberian Peninsula domains (NEIP and CIP, respectively) have been selected. These areas show different topographic patterns, the former being a coastal region characterized by a very complex terrain and the latter a continental inner region with a much simpler topography, which brings different locally-driven flows. Moreover, they house the largest Spanish urban areas, Barcelona and Madrid, and therefore photochemical pollution episodes are of special concern by their direct effects on population.

2.2 Methods

2.2.1 Modelling system

The WRF-ARW/HERMES/CMAQ modelling system is applied with high spatial (1 km^2) and temporal resolution (1 hr) to the study areas. The fine scale used is essential in complex terrains such as those studied (Jiménez et al., 2005a; 2006).

The Weather Research and Forecasting (WRF) Model (Michalakes et al., 2005; Skamarock et al., 2005) provides the meteorology parameters as inputs to CMAQ. The initial and boundary conditions are obtained from the National Centers for Environmental Prediction (NCEP) reanalysis data (available at the standard pressure levels for every 6 hours with 0.5 x 0.5 degree resolution). Four nested domains are defined over each study area (Table 2-1), covering the final domains (D4) (Figure 2.1): the NEIP (322 x 259 km^2) and the CIP (181 x 214 km^2), respectively. 33 sigma vertical layers cover the troposphere (up to 50 hPa ≈ 20 km), with 12 layers under the PBL.

Table 2-1. Summary of the defined domains and the WRF-ARW meteorological model configuration used

Domains definition	Physical parameterizations
Four one way nested domains D1-European domain 55x55 cells of 54 km D2- Iberian Peninsula domain 94x82 cells of 18 km D3- Iberian Peninsula Zone: 104x103 cells of 6 km D4a- Northeastern IP domain 322x259 cells of 1 km D4b- Central IP domain 181x214 cells of 1 km (Schematic representation in Figure 2.1)	ARW dynamical core Yonsei University PBL scheme Kain-Fritsch (D1-D2) and explicit (D3-D4) cumulus scheme Single Moment 3 class microphysics' scheme RRTM for long-wave radiation scheme and Dudhia for short-wave scheme Noah Land Surface Model.

The High Elective Resolution Emission Modelling System (HERMES) has been developed for Spain with a high spatial (1 km^2) and temporal (1 hr) resolution (Baldasano et al., 2008a). This model focuses on the estimation of gas and particulate matter pollutants, including the ozone precursors. HERMES considers the emissions from the following sources: (1) power generation plants, (2) industrial installations, (3) domestic and commercial fossil fuel use, (4) domestic and commercial solvents use, (5) road transport, (6) ports, (7) airports and (8) biogenic emissions; using a bottom-up approach except for the domestic and commercial fossil fuel use, where a top-down approach was adopted and regional emissions were allocated to fine grid cells by surrogate indexes. It follows the methodologies and criteria of previous emission models developed for the eastern Iberian Peninsula: EMICAT2000 (Parra et al., 2004; 2006) and EMIVAL (Arévalo et al., 2004). The reference year chosen is 2004, since it is the most recent year in which all the data needed for the HERMES development are available.

HERMES generates results according to the European Environmental Agency's Selected Nomenclature for Air Pollution (SNAP). Furthermore, HERMES has the capability of presenting results according to individual installations, industrial activities, land use classification or type of pollutant or process (fugitive, evaporative, hot or cold emissions).

The chemistry transport model used to compute the concentrations of photochemical pollutants is CMAQ (Byun and Schere, 2006). The initial and boundary conditions were derived from a one-way nested simulation covering a domain of 1392 x 1104 km^2 centred in the Iberian Peninsula, that used EMEP emissions for 2004 and disaggregated to 18 km. A 48-hour spin-up was performed to minimize the effects of initial conditions for the final domains. The chemical mechanism selected for the simulations following the criteria of Jiménez et al. (2003) was Carbon Bond IV (CBMIV) (Gery et al., 1989) including aerosols and heterogeneous chemistry. The CMAQ model configuration uses the Yamartino-Blackman Cubic scheme (YAM) (Yamartino, 1993) for the horizontal and vertical advection and transport scheme and the Eddy diffusion scheme for the vertical and horizontal diffusion. NO_x and volatile organic compounds (VOC) speciation of HERMES emissions are detailed in Parra et al. (2006). The vertical and horizontal resolution of the final domains (D4) (Figure 2.1) are the same than those used in the meteorological simulation (1 km^2, 33 vertical layers) and the species concentration and atmospheric processes contributions are estimated hourly.

The high resolution employed and the huge number of variables involved in the atmospheric integrations (not only the pollutants concentrations, but also the process analysis) require of high-performance computing. The availability of the MareNostrum supercomputer hold in the BSC-CNS, together with the advances in the parallelization of air quality model codes, have allowed these high-resolution simulations.

2.2.2 Integrated process rate (IPR)

The contributions of different processes are assessed by means of the Integrated Process Rate (IPR) analysis available in the CMAQv4.6 model. It provides the effects of all the physical processes and the net effect of chemistry on model predictions, which is the fraction of the mass or concentration in each model cell (or group of cells) in terms of the process that gave rise to this portion of the mass or concentration. More details can be found in Jeffries and Tonnesen (1994); Jang et al. (1995a, 1995b) and Gipson (1999). The processes contributions are estimated for each cell (1 km^2) on a hourly basis, providing the gas-phase chemical contribution, the net transport (advection and convection), vertical and horizontal diffusion, emissions, dry and wet deposition and clouds interactions contributions to net pollutants concentrations. The main assumptions of the modelling system are summarized below in order to clarify the subsequent discussion of results (Gipson, 1999):

(1). **Net transport:** It considers horizontal and vertical advection sum, which is the transport of pollutants due to the mean wind fields. The advection scheme used is globally mass-conserving. The horizontal advection is estimated by the YAM scheme, deriving a vertical velocity component at each grid cell that satisfies the mass continuity equation using the driving meteorology model's air density. The vertical advection module works with no mass-exchange boundary conditions at the bottom or top of the model. In this work the net effect of horizontal and vertical advection is considered to obtain comparable contributions with the rest of the processes. As already pointed out by Jiang et al. (2003), in some cases it could be more meaningful to aggregate the contributions of several processes instead of considering each of them separately.

(2). **Gas phase chemistry:** CBIV mechanism is used with the Euler Backward Iterative (EBI) (Hertel et al., 1993) solver.

(3). **Diffusion:** The diffusion involves sub-grid scale turbulent mixing of pollutants. Horizontal diffusion is estimated by a diffusion coefficient based on local wind deformation. Vertical diffusion is estimated using the Eddy diffusivity theory.

(4). **Dry deposition:** The deposition process is simulated as a flux boundary condition that affects the concentration in the lowest layer.

(5). **Clouds chemistry and wet deposition:** Clouds play a key role in aqueous chemical reactions, vertical mixing of pollutants and removal of pollutants by wet deposition. They also indirectly affect the concentration of pollutants by altering the solar radiation, which, in turn, affects photochemical pollutants, such as ozone, and the flux of biogenic emissions. CMAQ models sub-grid convective precipitating clouds, sub-grid non-precipitating clouds, and grid-resolved clouds. The cloud module vertically redistributes pollutants for the sub-grid clouds, calculates in-cloud and precipitation scavenging, performs aqueous chemistry, and accumulates wet deposition amounts. The used scheme is a RADM based cloud processor that uses the asymmetric convective model to compute convective mixing.

(6). **Aerosols:** The third generation aerosols module is used, which takes chemical species concentrations and reactivity rates from the chemistry solvers and primary particulate concentrations from the emissions processor to compute fine and coarse particulate concentrations. The emissions from different sources are estimated with HERMES for primary aerosols, not taking into account marine aerosols. The deposition velocity for particles is estimated from the aerosol size distribution, which is calculated from the mass and number concentration for each of the modes considered: Aitken ($0< \Phi < 0.1$ μm), accumulation ($0.1< \Phi < 2.5$ μm), and coarse (2.5 μm $< \Phi < 10$ μm)

The contribution of processes in this work is analyzed in several ways:

(1) **Analysis focused on model evaluation**. The contribution of processes to the concentration of gaseous pollutants in the lowest vertical level is assessed for specific points in the domains, coincident with the location of characteristic air quality stations (AQS): rural background AQS and urban background AQS. The contributions are obtained as concentration (μg m^{-3}) variation for the surface cell. The weighted contributions are also obtained by equation [2.1], where: $\%PC_i$ is the relative contribution of the i process to the net fluxes from or to the specific cell or domain (including chemical contributions) and PC_j is the contribution of each process, being j = gas-phase chemistry, transport (horizontal plus vertical advection), emissions, horizontal and vertical diffusion, dry deposition, clouds processes and aerosols processes.

$$\%PC_i = \frac{PC_i}{\sum_j \mathrm{abs}(PC_j)} \cdot 100 \qquad [2.1]$$

Note that the sum of $\%PC_i$ for all i is not 100%, being the sum of abs($\%PC_i$) exactly 100%.

(2) **Analysis of the dynamics of pollutants during the episode**. The processes are analyzed for the selected domains, up to the selected top of the atmosphere (33 sigma vertical layers up to 50 hPa) depicting contribution maps.

(3) **Analysis of specific locations with high O_3 levels during the episode**. Finally, the contribution of the different processes is assessed for four selected subdomains (20x10 km^2) in which maximum O_3 concentrations occur during the episode. Positive and negative contributions expressed as pollutants density under the PBL (height obtained from the model) in g m^{-2} are assessed for O_3, NO_x and NMVOCs in each subdomain.

2.2.3 Domains description

The coastal domain (NEIP) covers 83398 km^2 and is located in the Mediterranean littoral (Figure 2.1-left). It is characterized by a very complex terrain, dominated in the northern part by the Pyrenees mountains (up to 3400 m), and with two mountain chains parallel to the coast: the Pre-coastal (1000-1500 m height) and Coastal chains (500 m average height). The Ebro valley is located in the Southern region and acts as important channel for local wind flows. The Central Plateau covers the inland area. The major pollutants emission sources are the urban areas of Barcelona, accounting for 3.1 million inhabitants, and Tarragona, which is located in a densely industrialized area, and the road network connecting the Iberian Peninsula with France, all of them located along the coastal axis. Also inland some industrial areas have an important contribution to pollutants emissions in the area (i.e. the Cercs power plant). The HERMES model estimates 299 t d^{-1} of NO_x emitted during the episode, being a 57% produced by on-road traffic, which constitutes the main source of primary pollutants in the region (Costa and Baldasano, 1996; Parra et al., 2006), contributing also with 24% of the 692 t d^{-1} of NMVOCs emitted in the coastal domain. In this particular case 48% comes from biogenic sources. The industrial and power generation sectors are the main emitter of SO_2 and primary particles, accounting for 94% and 71% of the mass emissions (125 t d^{-1}, 45 t d^{-1}), respectively.

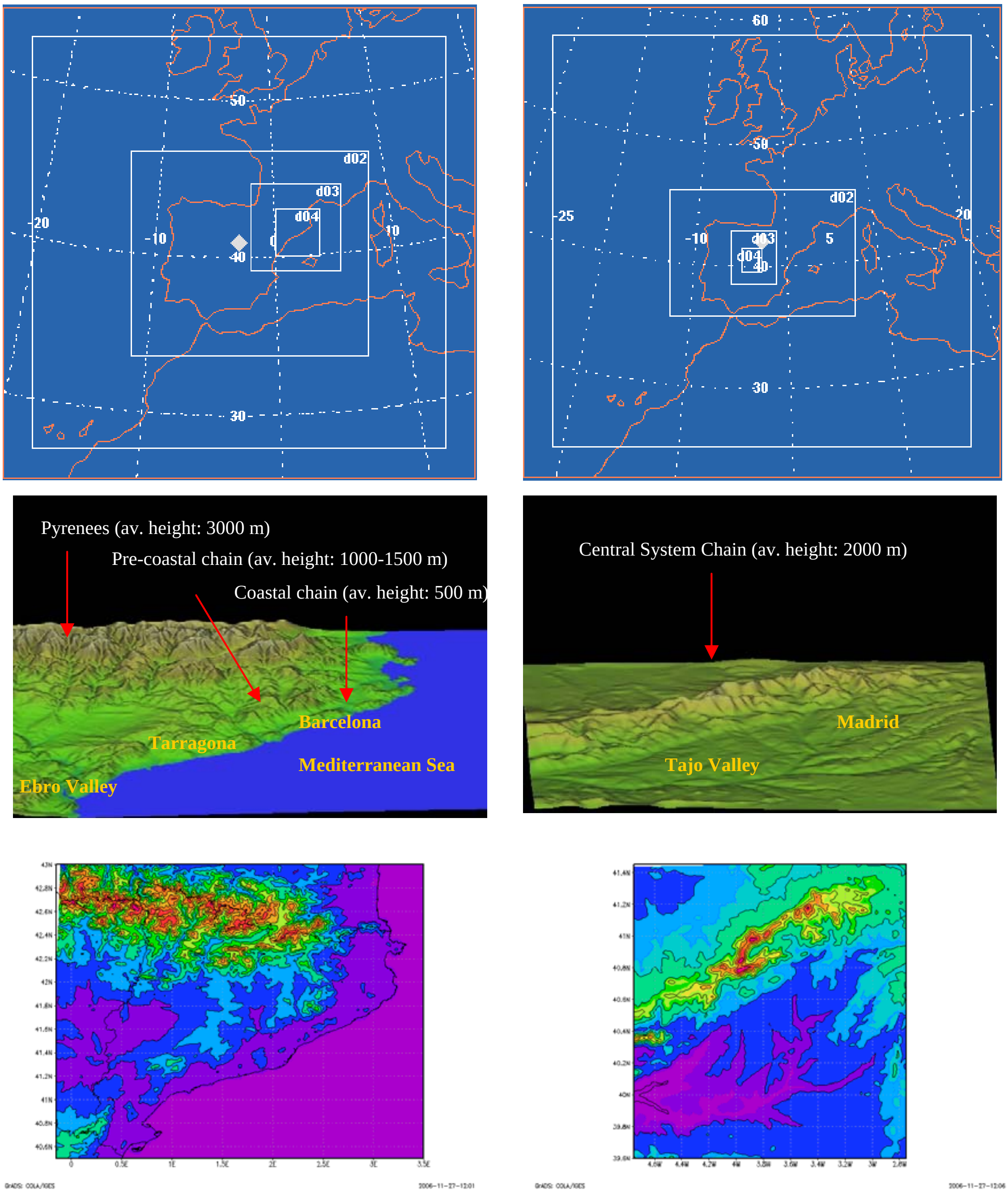


Figure 2.1. Selected domains and topography of the final study areas (D4): Northeastern IP-322 x 259 km^2 -(left) and Central IP -181 x 214 km^2- (right) domains

The continental domain (CIP) domain covers an area of 38734 km^2 in the centre of the Iberian Peninsula (Figure 2.1-right). The main topographic features in the region are the Central System located in the north-western area of the domain, with summits reaching 2500 m, and the Tajo valley in the Southern area. Both clearly affect the wind flows during the episode, characterized by the dominance of mesoscale phenomena. The main emission sources in the region are the Madrid urban area, the Spanish administrative capital, which accounts for 5.8 million inhabitants; the road network that connects Madrid with surrounding conurbations and several industrial sources, mainly located in the south-western part of the domain. The HERMES emissions model estimates indicate that on-road traffic accounts for 75% of total NO_x emissions within the region (231 t d^{-1}), and 19% of NMVOC (790 t d^{-1} in the whole area), being the biogenic sources the main contributors to these primary pollutants (64%). Industrial and power generation emissions of SO_2 and primary particulate matter account for 90% and 65% of the total emissions, 66 t d^{-1} and 36 t d^{-1}, respectively.

2.2.4 Episode selection

The 17-18 June, 2004 episode is characterized by a western recirculation in the synoptic scale, a typical summertime situation in south-western Europe since these conditions dominate 45% of the annual and 78% of the summertime transport patterns over the coastal Mediterranean areas (Jorba et al., 2004) and 36% of the annual and 45% of the summertime situations in the central-continental areas of the Iberian Peninsula. They are frequently associated with local-to-regional episodes of air pollution related to high levels of O_3 during summer (e.g. Toll and Baldasano, 2000; Barros et al., 2003; Ortega et al., 2004; Taghavi et al., 2004; Cousin et al., 2005; Coll et al., 2005; Jiménez et al., 2006; among others), being the study case one of the most polluted episodes of 2004 in the considered areas. The weak synoptic forcing and the stagnant conditions dominate the Iberian Peninsula. Therefore the mesoscale phenomena induced by the particular topography of the regions control the superficial wind flows. In these conditions the sea breezes and mountain-valley winds, and the development of the Iberian thermal low are characteristic features of the region (Jorba et al., 2004). The main processes inland are the convective circulations developed by the surface heating and the formation of compensatory subsiding flows in coastal areas (Millán et al., 1997; Pérez et al., 2004).

More details on the episode selection and the air quality conditions of the study areas can be found in *Annex 1*.

2.3 Results and discussion

2.3.1 Summary of the model evaluation for the 17-18 June, 2004 episode

The surface level pollutants concentrations predicted by the WRF-ARW/HERMES/CMAQ model are evaluated against hourly data from 45 AQS (provided by the Environmental Departments of the Catalonia and Madrid Governments, Spain). The evaluation results for O_3 accomplish the recommendations of the European Union, set by the European Directives 1999/30/EC, 2002/3/EC and 2008/50/EC (uncertainty of 50% for the air quality objective for modelling assessment methods). The US Environmental Protection Agency guidelines (US-EPA, 1991; 2007) recommend the use of different statistical parameters and combination of methods to assess a model performance, among them the mean normalized bias error (MNBE), which corresponds to the average difference between the modelled and the observed concentrations (eq. [2.2]); the mean normalized gross error (MNGE), which corresponds to the absolute values for the differences between modelled and observed concentrations (eq. [2.3]); and the unpaired peak prediction accuracy (UPA), which corresponds to the differences in modelled and observed peak concentrations (eq. [2.4]).

$$\mathrm{MNBE}\,(\%) = \frac{(\mathrm{MOD}-\mathrm{OBS})}{\mathrm{OBS}}100 \qquad [2.2]$$

$$\mathrm{MNGE}\,(\%) = \frac{\mathrm{abs}(\mathrm{MOD}-\mathrm{OBS})}{\mathrm{OBS}}100 \qquad [2.3]$$

$$\mathrm{UPA}(\%) = \frac{\max(\mathrm{MOD})-\max(\mathrm{OBS})}{\max(\mathrm{OBS})}100 \qquad [2.4]$$

Where MOD are the concentrations estimated by the model and OBS are the monitored concentrations.

The averaged MNBE for O_3 is 7.95%; the MNGE, 25.89% and the UPA, -0.39% (Table 2-2). Moreover, the NO_2, SO_2 and PM_{10} surface levels estimated agree with observations, with values for these statistical parameters lower or around 50%. The largest deviations are observed in the NO_2 predictions accounting for a 50.92% MNGE, nevertheless the reliability of the modelling system is acceptable being MNBE and the UPA -12.68% and -28.35%, respectively; and the average absolute error and the mean bias accounting for 27 µg m^{-3} and -16 µg m^{-3}, respectively.

The O_3 concentrations are overpredicted by the model both in the NEIP and CIP domains (Table 2-2), being the average normalized bias 5.85% in the former and 12.98% in the latter. Previous studies (Jiménez et al., 2006; Jiménez-Guerrero et al., 2008; Baldasano et al., 2008b) point that under low pressure gradient situations mesoscale models tend to underestimate daytime wind flows in coastal areas, which would favour the O_3 accumulation. The O_3 peaks are overestimated in the CIP (UPA: 11.02%), while an underprediction is assessed for the NEIP domain (UPA: -5.18%). The flow patterns in both domains differ, presenting the NEIP a very complex behaviour, with layering of pollutants and multiday accumulations that may be not accurately reproduced by the model, causing this underprediction in peak concentrations.

NO_2 concentrations are overpredicted in the CIP domain (positive bias of 5.89%), which could be attributed to the weaknesses of the model to represent wind flows under this low pressure gradient situation. The uncertainties related to the emissions account and the atmospheric chemistry representation in the CMAQ model could also play an important role. The underprediction in the coastal domain (MNBE: -27.12%) could be related with inaccuracies in the modelled chemical behaviour, linked to O_3 chemistry that may be causing also the high levels estimated for this pollutant. The NO_2 peak concentrations are underestimated in both domains (UPA: -26.3% in the CIP domain and -23.73% in the NEIP domain). Jiménez-Guerrero et al. (2008) run the WRF-ARW/HERMES/CMAQ model for the NEIP indicating an underprediction of NO_2 due to the relatively low vertical resolution in the lower troposphere that could generate an artificial vertical exchange between the boundary layer and the free troposphere, enhancing the NO_x venting. In this case, doubling the vertical resolution (33 vertical layers versus 16 in the mentioned study) leads to the same problem. This fact suggests that discrepancies between modelled and measured levels do not only depend on transport patterns, but also on the chemical behaviour represented by the model and on the emissions estimates.

Table 2-2. Summary of validation results for the 17-18 June, 2004. The statistical parameters are estimated for the 45 AQS stations of the XVPCA and Comunidad de Madrid air quality networks and separately for the NEIP and CIP domains as an average for the 48 hr simulated.

45 air quality stations (AQS) data	Bias (µg m^{-3})	Error (µg m^{-3})	MNBE (%)	MNGE (%)	UPA (%)
O_3	4.9	25.6	7.95%	25.89%	-0.39%
NO_2	-16.0	27.0	-12.68%	50.92%	-28.35%
SO_2	-8.9	11.0	-35.76%	47.95%	-35.33%
PM_{10}	-12.1	16.9	-15.86%	38.59%	-48.42%
Coastal Domain (NEIP) (33 AQS)	Bias (µg m^{-3})	Error (µg m^{-3})	MNBE (%)	MNGE (%)	UPA (%)
O_3	2.3	25.7	5.85%	26.65%	-5.18%
NO_2	-26.7	34.5	-27.12%	51.82%	-31.87%
SO_2	-10.0	12.9	-32.77%	49.95%	-29.23%
PM_{10}	-3.3	16.1	15.19%	50.65%	-14.82%
Central Domain (CIP) (12 AQS)	Bias (µg m^{-3})	Error (µg m^{-3})	MNBE (%)	MNGE (%)	UPA (%)
O_3	11.1	25.4	12.98%	24.06%	11.02%
NO_2	-2.2	17.3	5.89%	49.78%	-23.83%
SO_2	-6.8	7.1	-41.73%	43.94%	-47.53%
PM_{10}	-14.4	17.1	-23.62%	35.57%	-56.82%

The underestimation in SO_2 concentrations in both domains (MNBE of -32.77% and -41.73% in the NEIP and CIP domains, respectively) and in peak levels (UPA of -29.23% and -47.53% in the NEIP and CIP domains, respectively) could reflect a light underestimation on sulphur oxides emissions in the regions. On average, the PM_{10} concentrations are also slightly underpredicted by the model (MNGE of -15.86% and UPA -48.42%), which could be related not only with the inaccuracies in representing accumulation and transport patterns during this low gradient pressure situation, but also to the limitations related to the emissions model (HERMES) that does not take into account natural sources of primary particulates such as erosive or saltation processes or marine aerosols (Vautard et al., 2005a).

The contribution of processes in the first simulated layer (0-38 m agl.) provides useful information about the performance of the model for predicting surface O_3 concentrations. The surface O_3 concentration in rural background stations of the coastal domain (such as Agullana, Ponts and Rubí) and the continental domain (i.e. SM Valdeiglesias, Guadarrama and Buitrago de Lozoya) is overestimated during the morning and underestimated in the afternoon (the average bias is negative for the costal domain stations, -8.1%, -7.1% and -11.8%, respectively, and -8.01%, 10.27% and 2.21% for the continental domain AQS aforementioned).

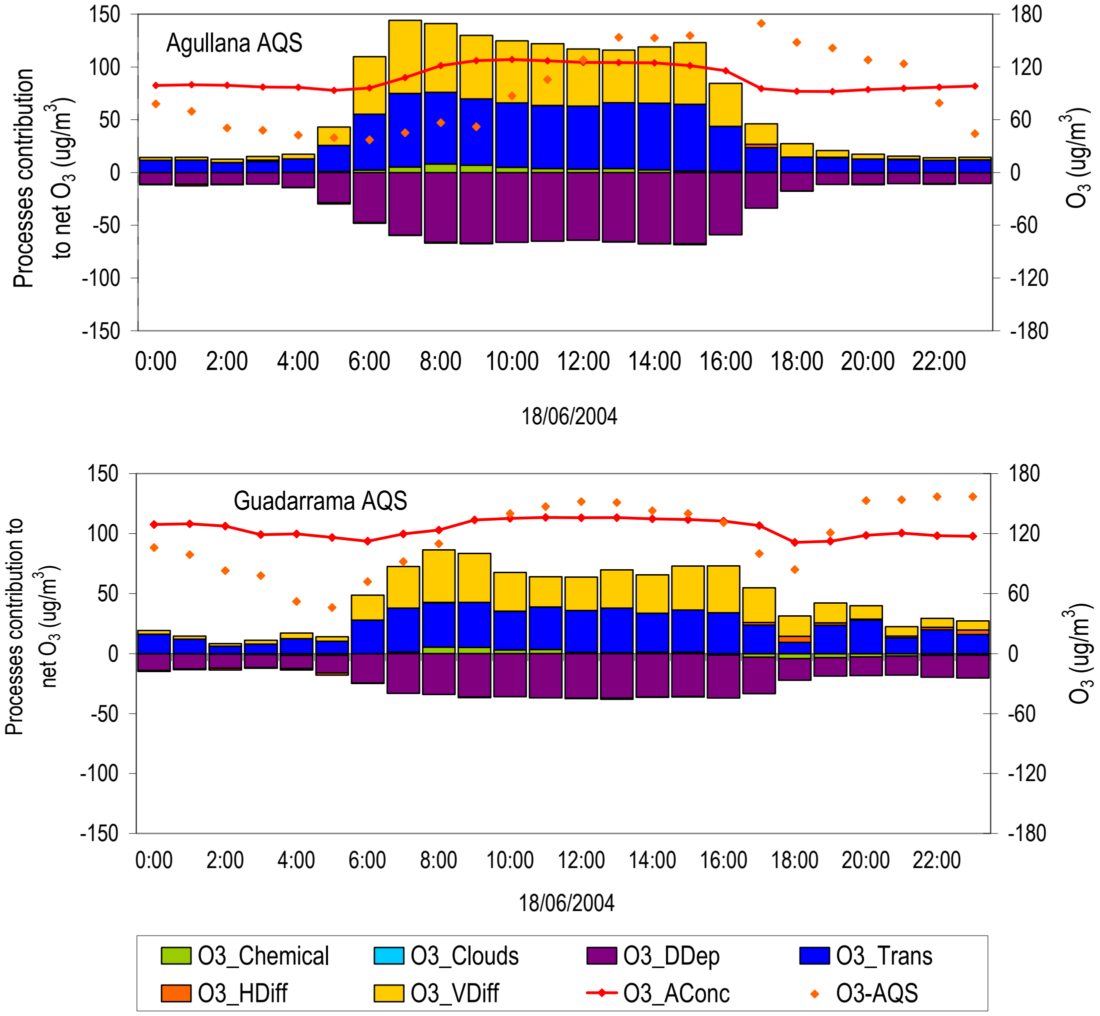


Figure 2.2. Atmospheric processes contribution to net O_3 concentration in the 1st simulated vertical layer (0-38 m agl.) – solid bars-, modelled (red dots) and measured (orange dots) O_3 hourly average concentration in rural background air quality stations of the NEIP (up-Agullana AQS) and the CIP domain (down-Guadarrama AQS) during the 18 June, 2004.

The representative sites selected reflect a common behaviour (Figure 2.2) independently of their location may mean that a slight deviations modelled advection fluxes. The vertical diffusion accounts for 33% of the net processes contribution to the surface cells, but is compensated by 32% removal of dry deposition. The processes controlling the total change in these cells' concentration are the net transport term (horizontal + vertical advection, accounting on average for 8%) and chemical production (7%). If the vertical transport (diffusion + dry deposition) is considered as a whole, the transport weight in the change of concentration involves 48% and chemistry 37% of positive contribution. The largest deviations in model predictions on the coastal domain stations occur specifically in the 1600 to 2000 UTC period, when the daytime breezes decrease and the land breezes and night-time winds start developing. The surface winds during this period are overestimated by the meteorological model which in turn favours pollutants venting in the chemistry transport model simulation. In the CIP concentrations are underestimated from 1000 UTC, but maximum deviations are found in the 1900-2200 UTC period.

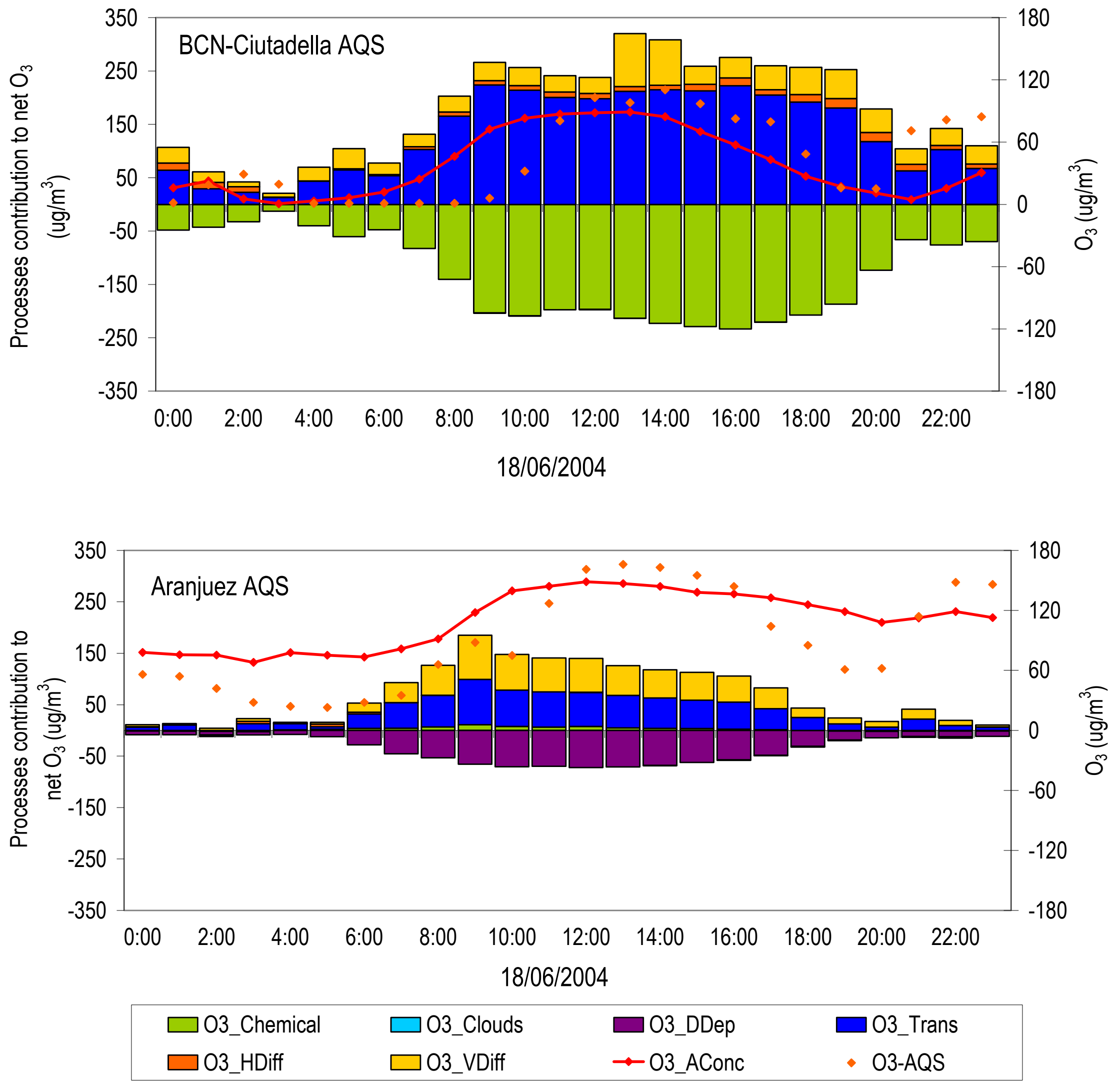


Figure 2.3. Atmospheric processes contribution to net O_3 concentration in the first simulated vertical layer (0-38 m agl.) –solid bars-, modelled (red dots) and measured (orange dots) O_3 hourly average concentration in urban background air quality stations of the NEIP (BCN-Ciutadella AQS) and the CIP domain (down-Aranjuez AQS) during the 18 June, 2004.

Previous works confirm the trend of the mesoscale meteorological models, and specifically the WRF-ARW, to underpredict the land breezes and underestimate the advective flows in the latter hours of the day (Jiménez-Guerrero et al., 2008), which causes overprediction in O_3 concentrations during night-time. The average processes contributions to net O_3 concentrations assessed during this period indicates that net transport accounts for 56% and chemical destruction for -26%, when considering the combined effect of vertical processes (vertical diffusion + dry deposition).

In the urban background stations locations (such as Barcelona-Ciutadella and Tarragona in the NEIP, or Aranjuez and Chapineria in the CIP) the modelled O_3 has a similar behaviour (Figure 2.3). Albeit during the morning there is an overestimation trend, during the afternoon and night-time the model predictions are frequently below the measured levels.

The NEIP domain chemical regime involves O_3 photochemical destruction in the first layer accounting for -46% of net night-time fluxes and -44% of net daytime fluxes on average when considering the vertical fluxes as a whole (being the sum of vertical diffusion and dry deposition a positive term that accounts for 10% and 13%, respectively). In the CIP the periods of O_3 net formation occur in the first vertical layer during daytime, not existing in the urban background stations of the NEIP. On average the net effect of daytime gas phase chemistry accounts for -5% of total fluxes of O_3. The main positive contributions come from net transport, involving 86% of net concentration variation in the surface cells. During night-time main sinks of O_3 are chemical destruction (-27%) and the vertical flux term (vertical diffusion plus dry deposition, that combined remove -12% of net O_3 fluxes). In the coastal domain stations the chemical destruction of O_3 is the main removal process even during daytime, probably due to the O_3 titration by NO_x emissions. On-road traffic plays an important role in this sense being the major NO_x emitter the Barcelona area.

This behaviour is also reproduced by the model in specifically urban stations such as Barcelona-Eixample, Constantí or L'Hospitalet (NEIP) and Majadahonda, Coslada or Leganés (CIP) where main contributions to surface O_3 come from transport and vertical diffusion, and the main sinks are gas-phase chemistry and dry deposition.

2.3.2 Pollutants dynamics in the coastal and central south-western Mediterranean

The results of the photochemical simulations for the coastal and central-continental domains show the maximum O_3 concentrations occurring in downwind areas from Barcelona (NEIP) and Madrid (CIP) conurbations after the maximum photochemical activity hours (Figure 2.4). During the day the increase of the solar radiation and the temperature (reaching 30-35ºC in both domains) promote the high levels of O_3, exceeding in some cases the population information threshold (180 μg m^{-3}). The gas-phase chemistry accounts in these cases for 50 to 75 µg m^{-3} in the first layer cells. In the urban areas and main roads, as main sources of NO_x, chemical destruction of O_3 occurs up to -200 µg m^{-3} per hour during the traffic circulation peaks. The vertical gradients of O_3 concentration generated in these areas, larger during daytime, increase the contribution of diffusion processes to ground-level O_3 (up to 200 µg m^{-3} h^{-1} fluxes, mainly from upper vertical layers).

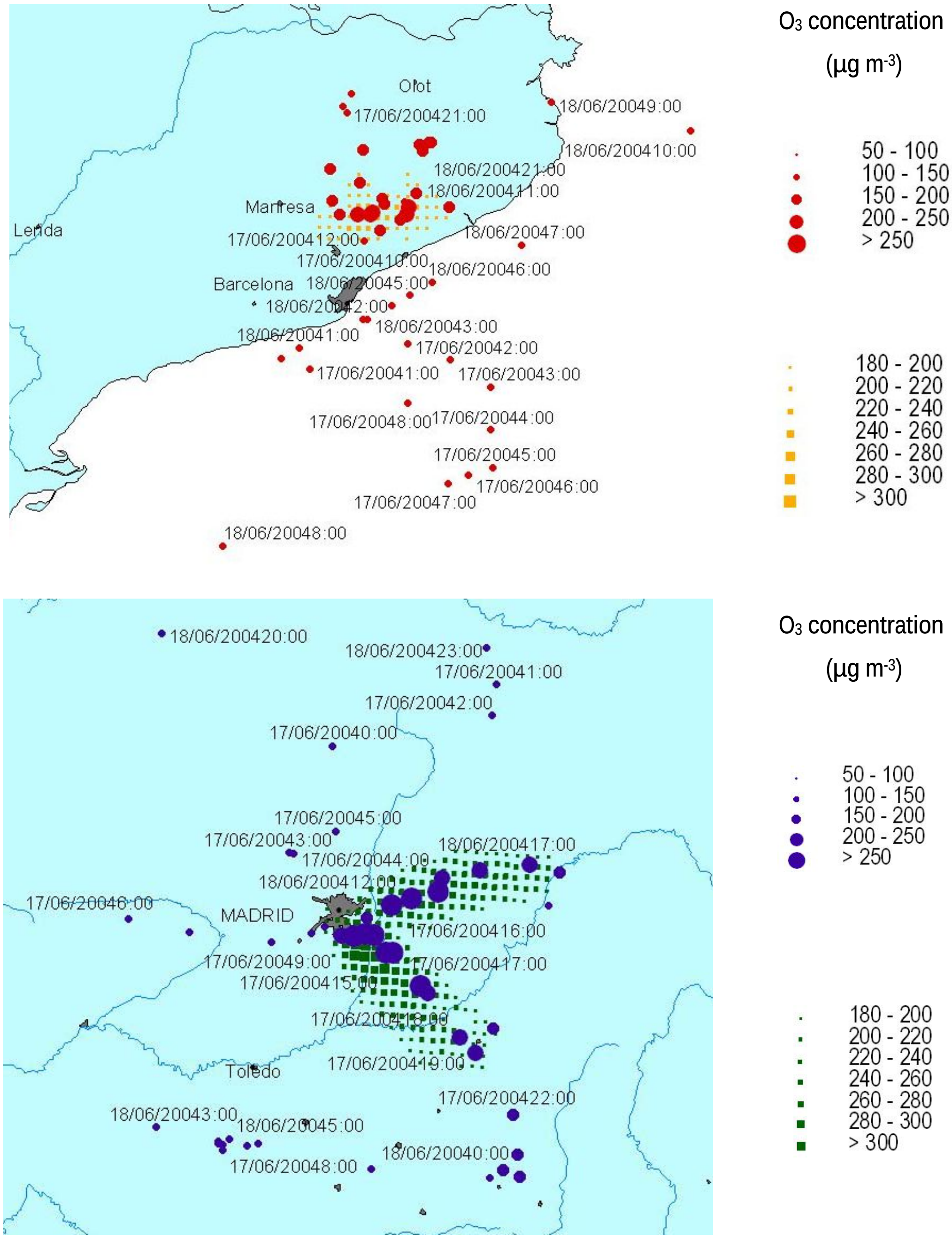


Figure 2.4. Hourly maximum O_3 surface concentration location (circles) in the coastal –NEIP- (up) and the central –CIP- (down) domains during the 17-18 June, 2004 episode, simulated by WRF-ARW/HERMES/CMAQ. Squares: concentrations above the EU human health protection limit (180 µg m^{-3}, Dir 2002/3/EC).

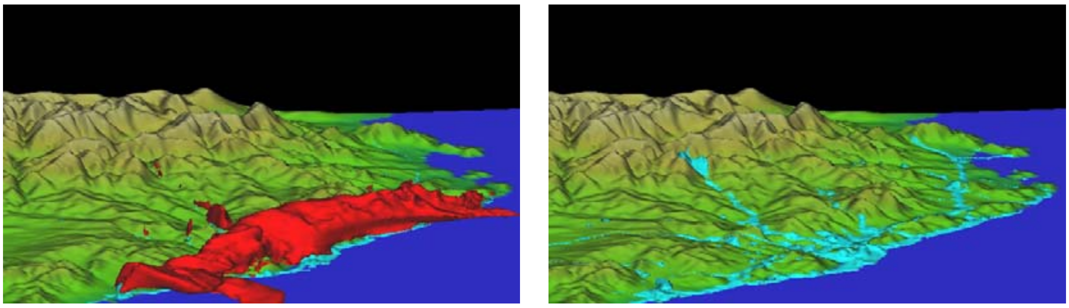
Surface O_3 concentrations are larger in the continental than in the coastal domain. While in the NEIP the breezes and mountain-valley winds regime involves the accumulation and recirculation of pollutants aloft, in the CIP the main flows are dominated by thermal phenomena that transport pollutants within the convective recirculation cell. The PBL height reaches its maximum at midday, being higher in the central-continental domain –around 3200 m agl- than in the coastal domain –maximum around 2000 m agl-, due to the lamination of the PBL growth by the Mediterranean Sea breezes.

The emitted NO_x and NMVOCs in the central-continental domain during the morning react forming O_3, the surface heating generates a convective cell and the photochemical pollution plume rises, reaching 2500-3000 m agl (Figure 2.5 and 2.6) and moves downwind, affecting the southern area during the 17 June and the Eastern area on the 18 June. The maximum surface O_3 concentrations occur at midday (from 1300 to 1600 UTC of 17 June and 1200 to 1400 UTC of 18 June). During the last hours of the day the convective cell weakens and falls over the Madrid downwind areas, the PBL height reduction involves high O_3 surface concentrations even when the photochemical production diminishes. At night-time, the O_3 generation ceases, and the concentration of photochemical pollutants decays. The katabatic winds dominant in the Central system area enhance the effect of the wind shift, being the main flows channelled by the Tajo valley towards the south-western area (hourly maximum concentrations in this period are located in this area, see Figure 2.4).

The coastal domain behaviour differs, being more favourable to several days photochemical episodes, due to the reservoir layers formed over the Mediterranean Sea during night-time (Figure 2.5 and 2.6). Specifically, the main emission sources are located in the coastal area. When the mesoscale phenomena dominate the pollutants transport, the breeze regime development in the first hours of the day transports the primary pollutants inland. The littoral mountain chain acts as a barrier at dawn, recirculating the pollutants towards the Mediterranean Sea; these return flows are enhanced by the anabatic winds development. As the day advances the sea breeze regime develops and reaches the Pre-littoral, higher than the previous (1000-1500 m), producing a second recirculation flow in altitude. On the other hand, the river valleys and main roads act channelling the pollutants flow inland. The pollution plume reaches flat inland areas, where it accumulates during the afternoon and dusk, because of the Pyrenees barrier cutting the flows to the Northern area. In fact maximum O_3 surface concentrations during the episode are reached in the 1200-1400 UTC period in north-eastern areas from the Barcelona conurbation and maximum hourly concentrations in the 1800-2100 UTC period are found in the northern domain (in the Pre-Pyrenees region) (Figure 2.4). When the mesoscale wind regime shifts, katabatic winds and land breezes involve a return flow of the pollutants plume over the Mediterranean Sea. Moreover, previous studies (e.g. Jiménez et al., 2006) have detected a layer of pollutants outbreak in altitude with peninsular origin (3000-3500 m agl) that contributes in this kind of episodes to the high concentration of pollutants in the area.

On the other hand, the urban plume reactivity differs, being in the Madrid airshed more favourable to O_3 formation in situ, while in Barcelona O_3 titration by NO_x emissions involves a sink of O_3 in the surface layer, formation occurs mainly in upper levels and in areas located around 100 km far away from the city (Figure 2.5).

In summary, the main contributors to O_3 surface concentrations are the net transport and vertical diffusion, while the main sink is dry deposition. The wet deposition and cloud processes are negligible during the episode, due to the low cloudiness and the absence of precipitation, typical in a summertime period such as the one studied. The aerosols interaction with photochemical gaseous pollutants is also negligible. The chemical contribution differs depending on the location and the NO_x-VOCs ratio. The process analysis indicates that maximum chemical production of O_3 does not occur in the first vertical layer of the model, but in layers aloft during morning and midday (Figure 2.5). This effect will be deeply discussed in the next section. The horizontal and vertical advective transport set the areas that will be affected by these pollutants, being the transport patterns much simpler in the central domain than in the coastal area, where stratified layers and accumulation of pollutants are frequent during summertime episodes with stagnant conditions, as shown in Figure 2.6. The process analysis confirms the results of previous experimental and modelling studies carried out in these areas (e.g. Baldasano et al., 1994; Millán et al., 1997; Martín et al., 2001a; 2001b; Soriano et al., 2001; Pérez et al., 2004; Jiménez et al., 2006).



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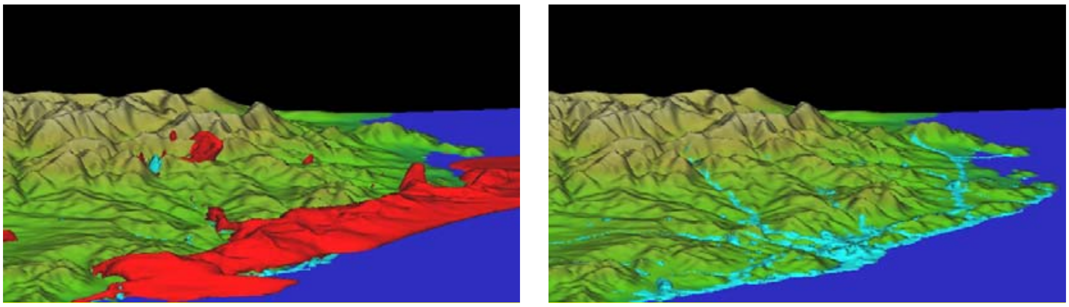

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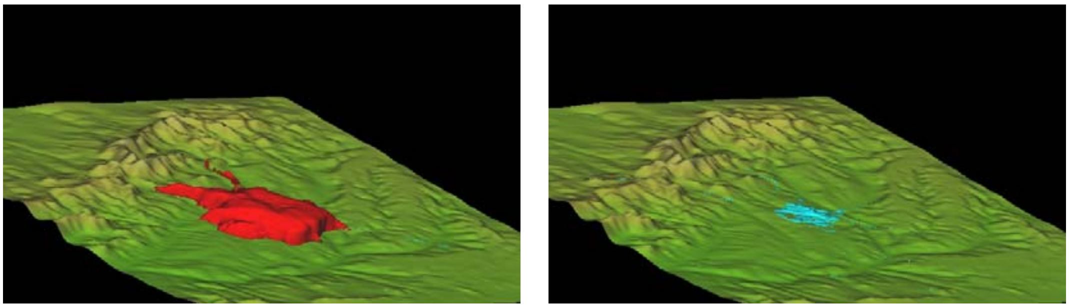

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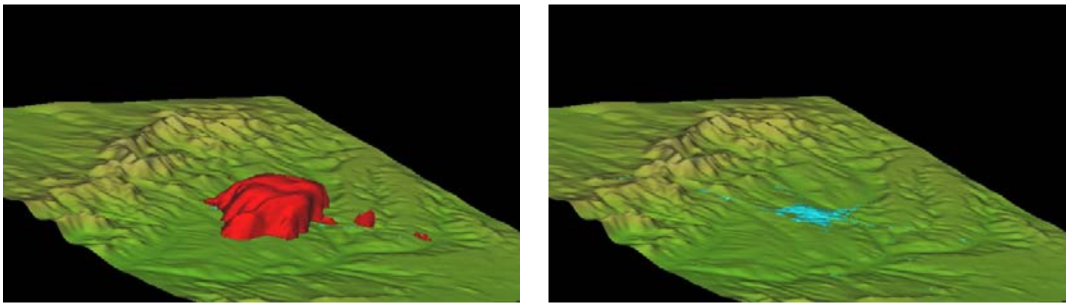
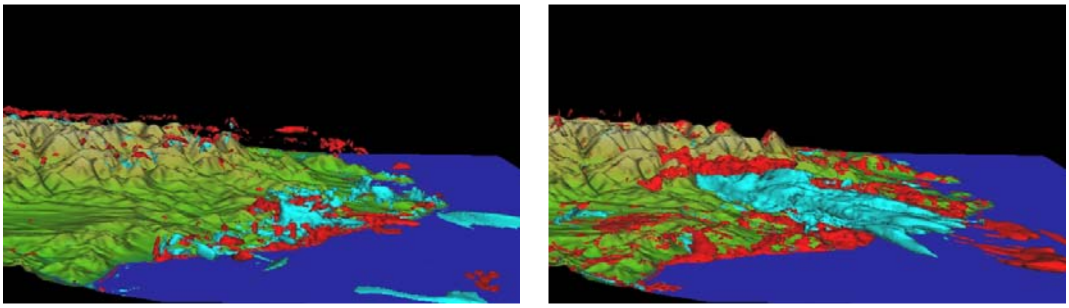


18/06/2004 1000 UTC 18/06/2004 1800 UTC

Figure 2.5. 10 µg m^{-3} isosurface of chemical production (red) and destruction (blue) of O_3 for the NEIP (up) and CIP (down) domains at 1000 and 1800 UTC of the 17-18, June, 2004.



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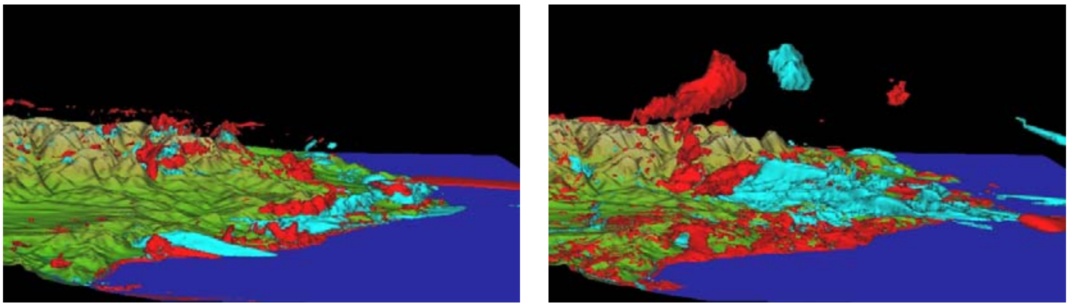

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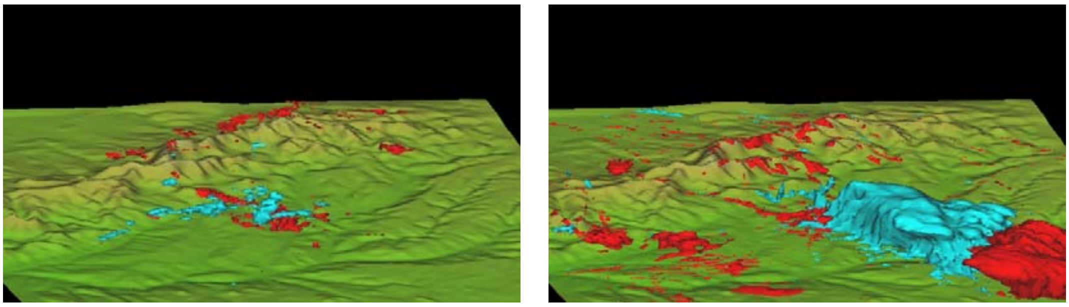

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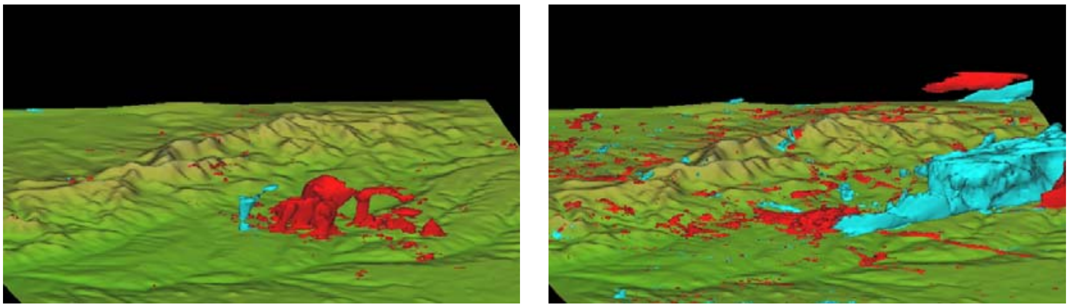


18/06/2004 1000 UTC 18/06/2004 1800 UTC

Figure 2.6. 10 µg m^{-3} isosurface of O_3 net transport (horizontal + vertical advection) -positive contribution in red and negative contribution in blue- for the NEIP (up) and CIP (down) domains at 1000 and 1800 UTC of the 17-18 June, 2004.

2.3.3 Processes contribution to O_3-NO_x-NMVOCs concentrations

In order to define the origins of maximum O_3 concentrations in the coastal and continental domains, two subdomains (20x10 km^2) –MAX1 and MAX2- are defined considering the areas where maximum concentrations are estimated by the model on the first and second day of the episode, respectively (Figure 2.7). The O_3, NO, NO_2 and NMVOC density (g m^{-2}) in the atmospheric column under the PBL is assessed together with the contributions of different processes leading to these levels. All values are obtained as an hourly average for the whole subdomain areas (200 km^2). The night-time modelled PBL height (Figure 2.7) is near surface level (around 20 m agl), then the pollutants density and processes contribution are negligible versus those of daytime periods.

The O_3 maximum concentrations during the 17 June occur in the MAX1 domains. In the costal-MAX1 the horizontal advective transport to the area constitutes the major positive contribution to net O_3 until 1600 UTC (0.042 g m^{-2} h^{-1} on average from 900 to 1600 UTC). There is also a significant vertical advective transport removing O_3 from the PBL (-0.052 g m^{-2} h^{-1}). Later on, until 1900 UTC, the horizontal advective flows remove O_3 from this area (0.057 g m^{-2} h^{-1} on average), due to the transport of the pollutants plume towards the Mediterranean Sea. The O_3 chemical production from 700 UTC to 1700 UTC is the second source of importance (0.019 g m^{-2} h^{-1}). The continental-MAX1 domain presents a similar behaviour, being the horizontal advective transport the most important positive contribution to net O_3 until 1500 UTC, on average 0.042 g m^{-2} h^{-1} (Figure 2.8). Then, the wind shift involves a negative contribution of horizontal advection until 1900 UTC (-0.079 g m^{-2} h^{-1}). In this case there are no important vertical injections, being the contributions of vertical advection almost negligible compared to horizontal advection and chemical production (i.e. vertical transport causes a removal of -0.006 g m^{-2} h^{-1} on average in the 1100 to 1500 UTC period). The complex topography in the coastal area explains, as already commented, the vertical transport flows of pollutants caused by the breeze regimes mechanically enhanced by the mountain chains located along the coast. Meanwhile, in the continental domain the vertical flows promote the transport of pollutants under the PBL; these are thermally produced and they have no topographic forcing involving injections in upper levels.

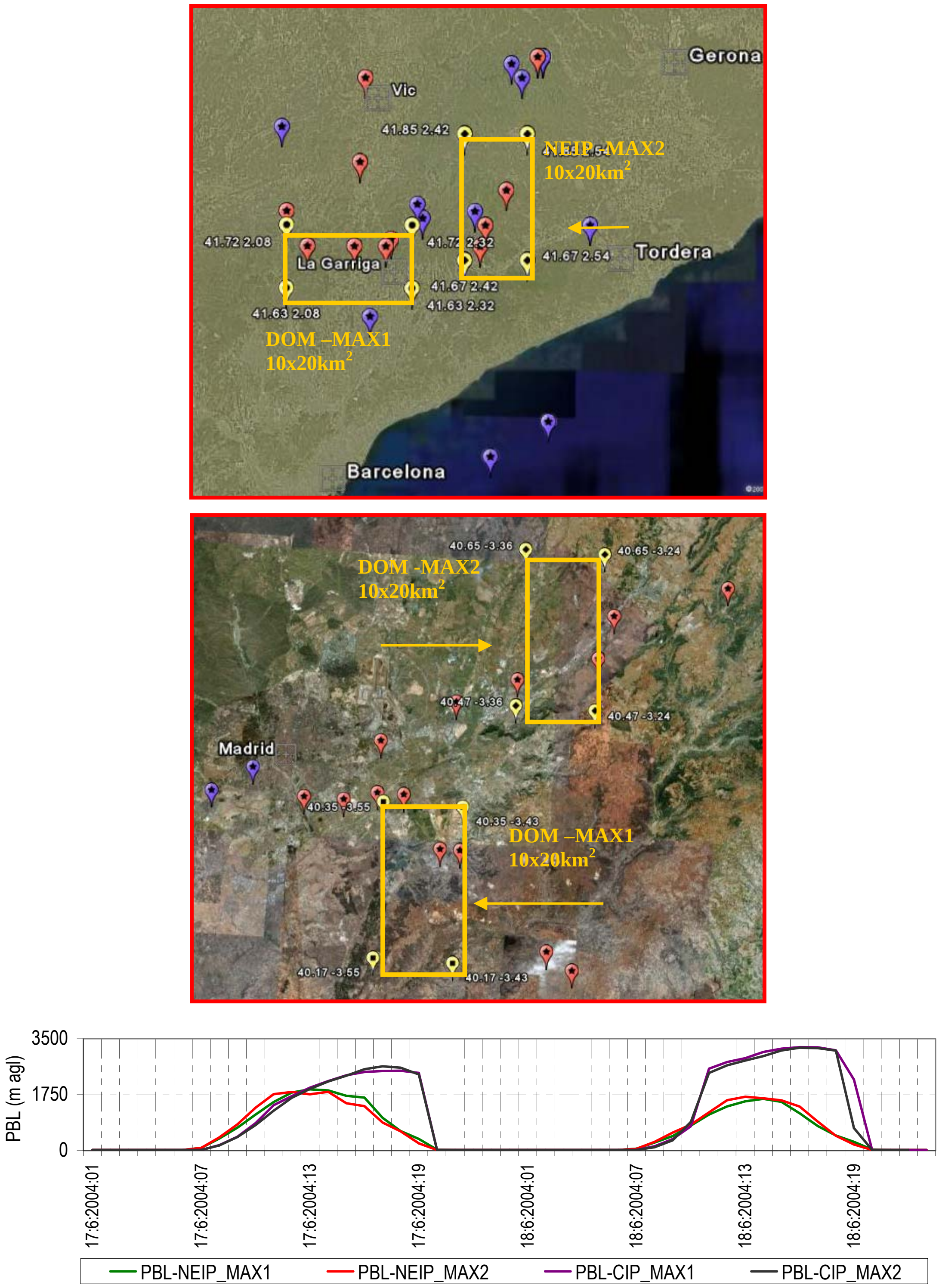


Figure 2.7. Subdomains MAX1 and MAX2 (in yellow) selected in the NEIP (up) – MAX1:LAT:41.63-41.72, LON:2.08-2.32; MAX2: LAT:41.67-41.85, LON:2.42-2.54 - and CIP domains (down) – MAX1:. LAT:40.17-40.35, LON:-3.55-(-3.43); MAX2: LAT:40.47-40.65; -3.36-(-3.24)-. Maximum hourly concentrations above 180 µg m^{-3} in red, concentrations below this limit in purple. Evolution of the modelled PBL height in the NEIP and CIP subdomains

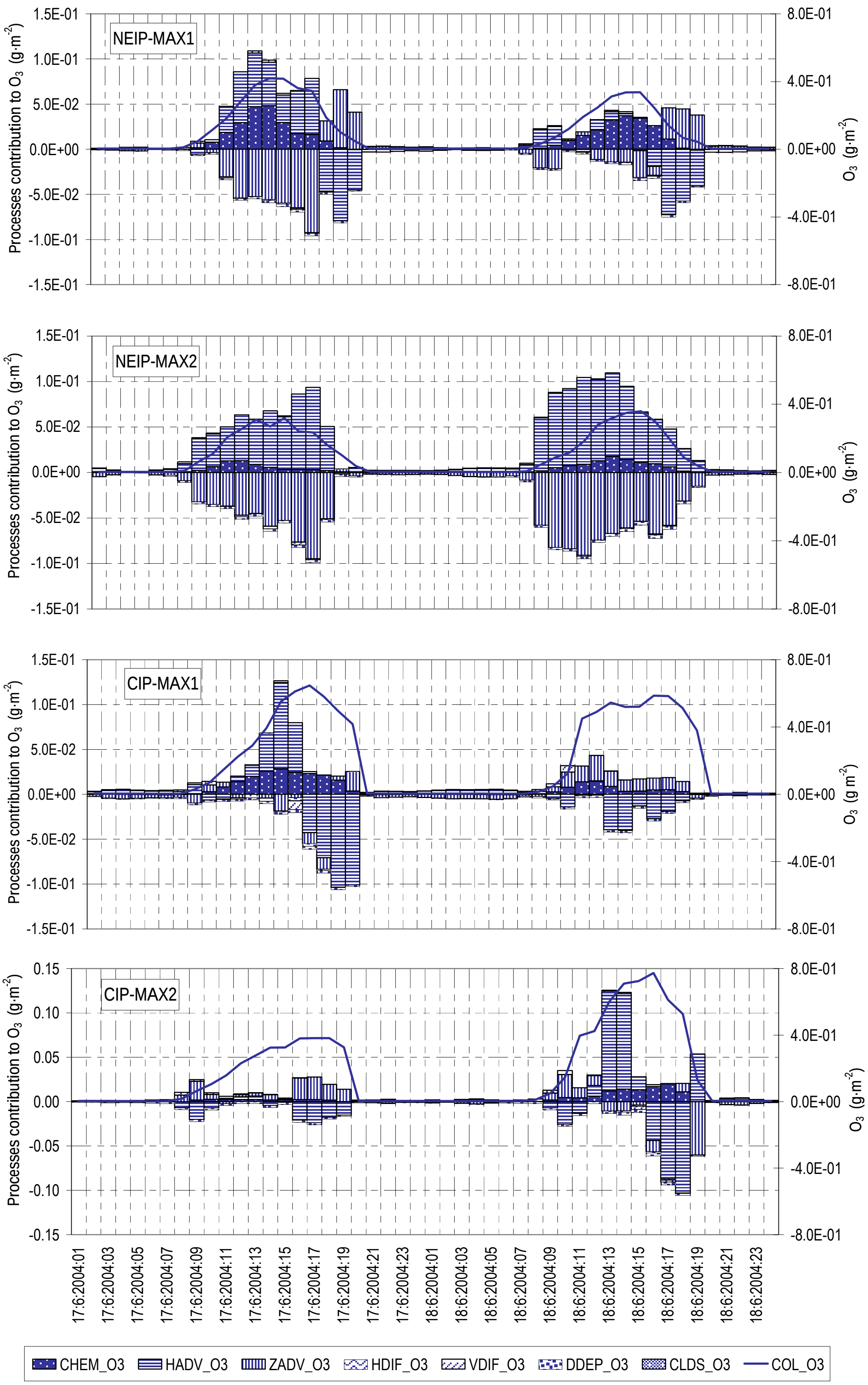


Figure 2.8. Atmospheric processes contribution to net O_3 density (g m^{-2}) under the modelled PBL during the 17-18 June, 2004. Averaged contributions for the NEIP-MAX1 and MAX2 domains, and CIP-MAX1 and MAX2 domains.

On the other hand, the MAX2 subdomains stand for the locations of maximum O_3 concentrations during the second day of the episode. As discussed for the first day, the contributions to net O_3 density are mainly transport; the continuous horizontal advective flows involve high O_3 densities in the coastal-MAX2 subdomain (horizontal transport contributes on average with 0.064 g m^{-2} h^{-1} during the 800 to 1900 UTC period). Once again, the vertical injections are the major sink of O_3 in the region (-0.062 g m^{-2} h^{-1} on average). Chemical production in this case is lower than in areas nearer from the Barcelona urban area, such as MAX1 domain (0.009 g m^{-2} h^{-1} on average), and the O_3 concentrations estimated by the model are clearly affected by horizontal advection. The continental-MAX2 domain is characterized by an important horizontal advection during the central hours of the day, from 1200 to 1500 UTC contributing with 0.062 g m^{-2} h^{-1} on average; then the horizontal flows remove O_3 from the domain (-0.078 g m^{-2} h^{-1} until 1800 UTC). The relatively high O_3 density observed from 1400 to 1600 UTC is due to the chemical production (0.015 g m^{-2} h^{-1} from 1300 to 1800 UTC) and the larger integration height (PBL height 3134 to 3220 m agl as observed in Figure 2.7) considered.

The NEIP subdomains (coastal) are characterized by the low weight of emissions in the NO_x budget under the PBL (Figure 2.9) when compared to CIP (continental). Specifically in the NEIP-MAX1, the horizontal advection is the main source of NO-NO_2 until 1600 UTC (3.4 10^{-4} and 0.0022 g m^{-2} h^{-1}, respectively) and chemical reaction involves the main sink of these oxides (-4.4 10^{-4} and -0.0016 g m^{-2} h^{-1}, respectively). From then on the horizontal transport removes NO_2 and vertical transport from layers above the PBL contributes positively until 1800 UTC. While NO continues oxidizing and chemical destruction is still the main sink of this compound, from 1800 to 2000 UTC net chemical production of NO_2 in situ is observed (2.07 10^{-4} g m^{-2} h^{-1}). The minimum NO_x density observed on 17 June around 1500-1600 UTC reflects the direct effect of the peaks of traffic emissions during the early morning and the afternoon in closer areas (such as Barcelona city), which is the main origin of the advected NO_x.

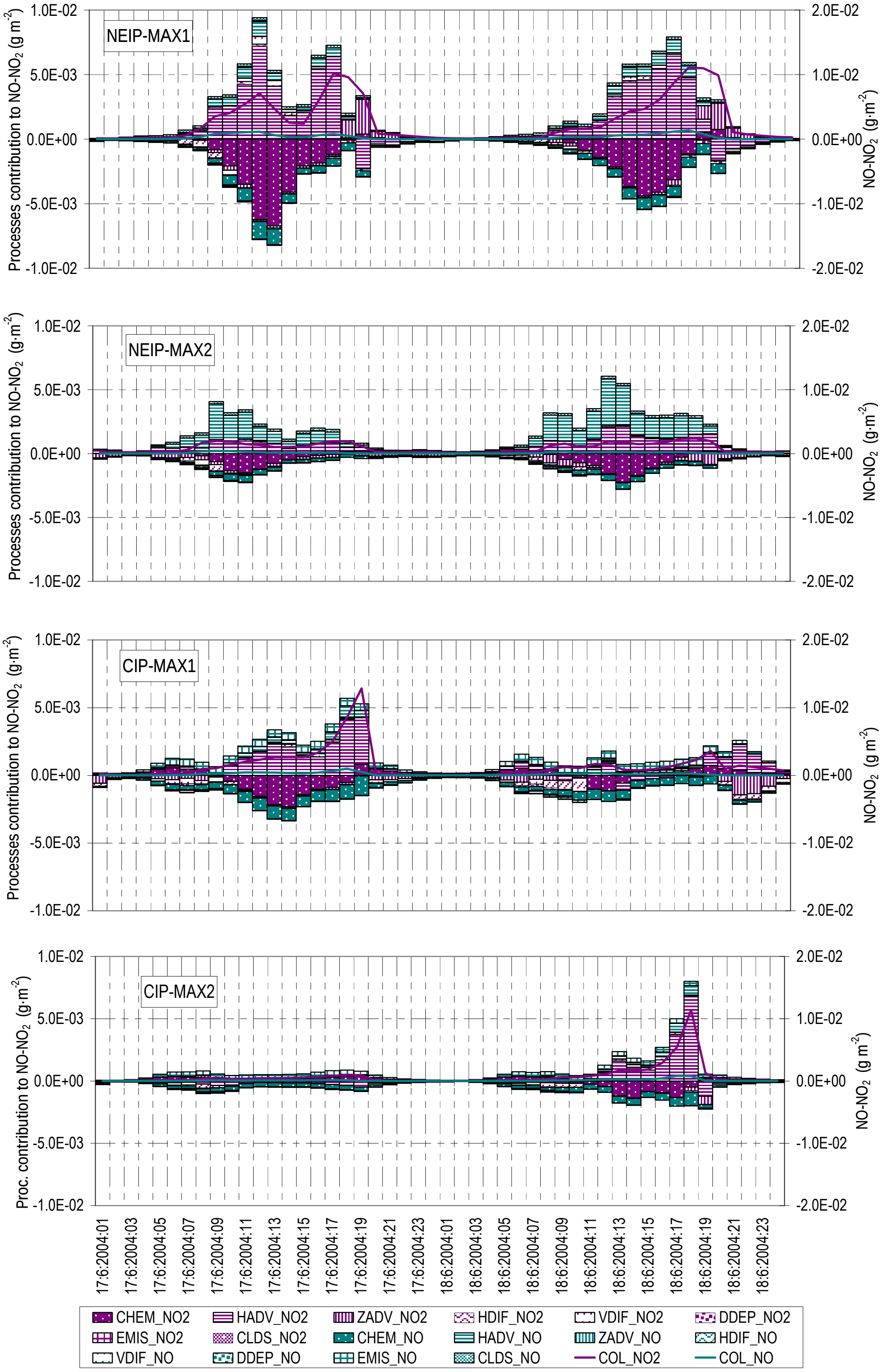


Figure 2.9. Atmospheric processes contribution to net NO$_X$ density (g m^{-2}) under the modelled PBL during the 17-18 June, 2004. Averaged contributions for the NEIP-MAX1 and MAX2 domains, and CIP-MAX1 and MAX2 domains.

The CIP subdomains present a similar behaviour concerning NO_x (Figure 2.9), being the major source the horizontal advection from surrounding areas (2.0 10^{-4} g m^{-2} h^{-1} for NO and 0.0011 g m^{-2} h^{-1} for NO_2 until 1900 UTC) and the main sinks the gas-phase chemical reactions (-5.8$10^{-4}$ g m^{-2} h^{-1} and -4.2 10^{-4} g m^{-2} h^{-1}, respectively). Nevertheless the emissions of NO have limited importance in these subdomains, and specifically in the CIP-MAX1 domain, where NO emissions contribute with an approximately constant density, from 400 UTC to 1900 UTC (4.9 10^{-4} g m^{-2} h^{-1}). The increase in NO_2 density during the 1500-1800 UTC period due to the advective transport and the lower chemical destruction may be related to the high O_3 density observed. The NO_x levels in the central-MAX2 domain are low during the 17 June and increase considerably by horizontal advection during the 18 June. In the early morning (until 1000 UTC) losses of NO_x by vertical diffusion (-2.8 10^{-5} g m^{-2} h^{-1} for NO and -1.2 10^{-4} g m^{-2} h^{-1} for NO_2) to upper layers are estimated by the model.

In the coastal-MAX1 domain the NMVOC levels are relatively high (Figure 2.10). Horizontal advection until 1700 UTC (contributing on average with 7.5 10^{-5} g m^{-2} h^{-1}) and vertical advection from upper layers until 1900 UTC (3.6 10^{-5} g m^{-2} h^{-1}) are their main sources. The daytime average contribution of emissions is low and accounts for 1.2 10^{-5} g m^{-2} h^{-1}. During the morning, until 1000 UTC, some NMVOCs are injected in layers above the PBL by vertical diffusion (-1.5 10^{-5} g m^{-2} h^{-1}) and vertical advective processes (-2.5 10^{-5} g m^{-2} h^{-1} on average until 1600 UTC), involving a sink of these organic compounds, together with chemical reaction (-1.3 10^{-5} g m^{-2} h^{-1} on average during daytime). Nevertheless, the net transport (advection and diffusion) involves a positive contribution during daytime on average of 3.8 10^{-5} g m^{-2} h^{-1}. From 1100 UTC the vertical diffusion contributes positively to the NMVOCs density under the PBL, nevertheless its contribution is almost negligible compared to the net transport by advection. The NEIP-MAX2 behaviour is more homogeneous, being the horizontal advective flows the main contributors to net NMVOCs density under the PBL (5.6 10^{-5} g m^{-2} h^{-1}) and vertical advection the main sink (-3.8 10^{-5} g m^{-2} h^{-1}), together with chemical reaction (-6.5 10^{-6} g m^{-2} h^{-1}) and vertical diffusion specially during the morning (-1.3 10^{-5} g m^{-2} h^{-1} until 1100 UTC).

The horizontal advection affecting the CIP-MAX1 domain during 17 June involves the largest contribution to net NMVOCs density under the PBL until 1600 UTC (Figure 2.10, average contribution of 1.27 10^{-4} g m^{-2} h^{-1}), then the winds shift removes these compounds by the same process. The chemical destruction reaches its maximum during the 1400-1500 UTC, accounting for -2.0 10^{-5} g m^{-2} h^{-1}. Vertical advection and diffusion remove NMVOCs from the PBL, but in a lower extent. The specific study area is not characterized by a high emissions rate; therefore the contribution of this process is almost negligible compared to transport processes. The CIP-MAX2 presents a similar behaviour during the 18 June, when it is directly affected by the horizontal advective flows. The winds shift occurs one hour earlier, being the horizontal transport the main sink of NMVOCs in the area from 1600 UTC (-6.5 10^{-5} g m^{-2} h^{-1}). The maximum removal by chemical destruction also occurs in the 1400-1500 UTC period (-1.7 10^{-5} g m^{-2} h^{-1}).

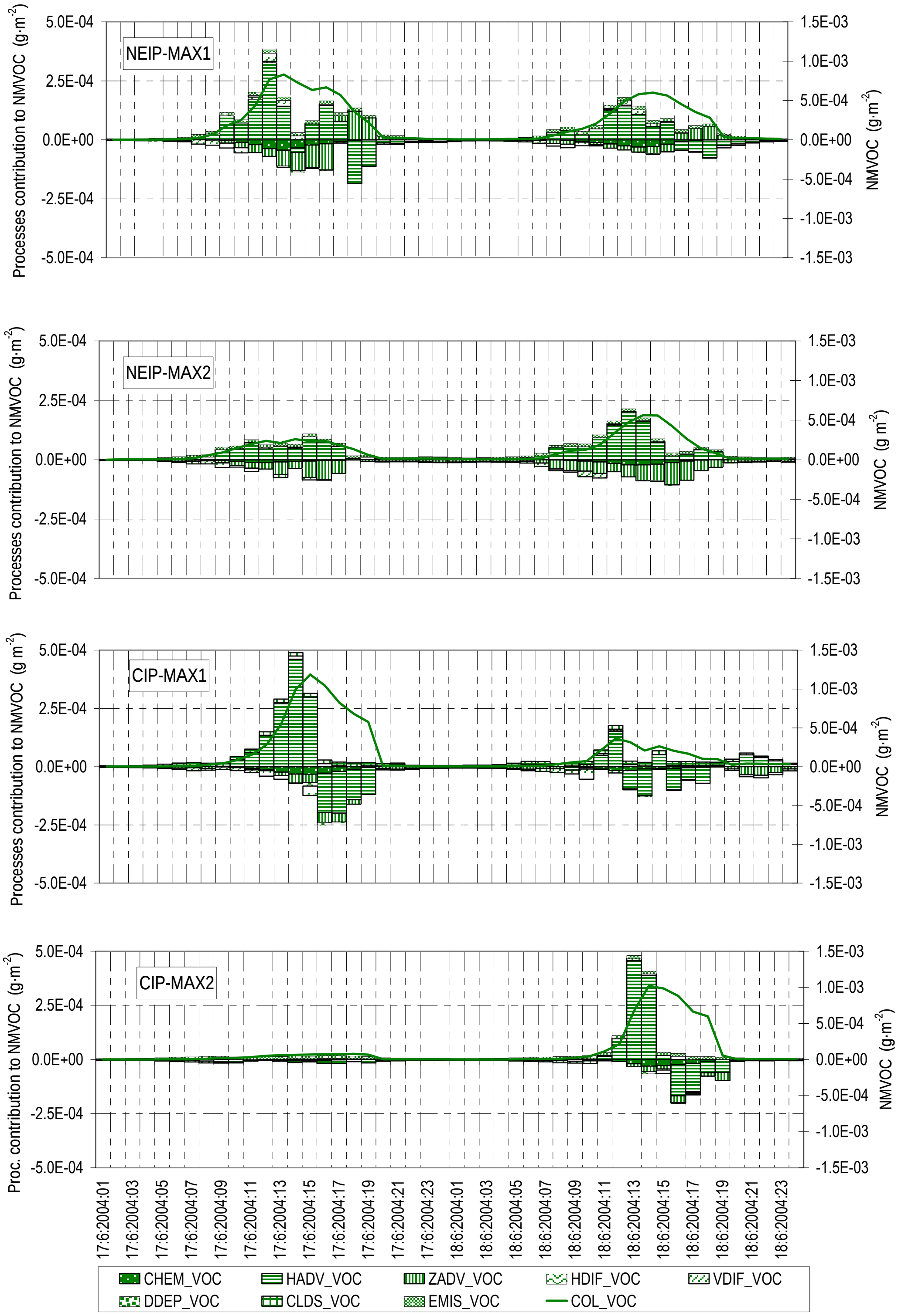


Figure 2.10. Atmospheric processes contribution to net NMVOC density (g m^{-2}) under the modelled PBL during the 17-18 June, 2004. Averaged contributions for the NEIP-MAX1 and MAX2 domains, and CIP-MAX1 and MAX2 domains.

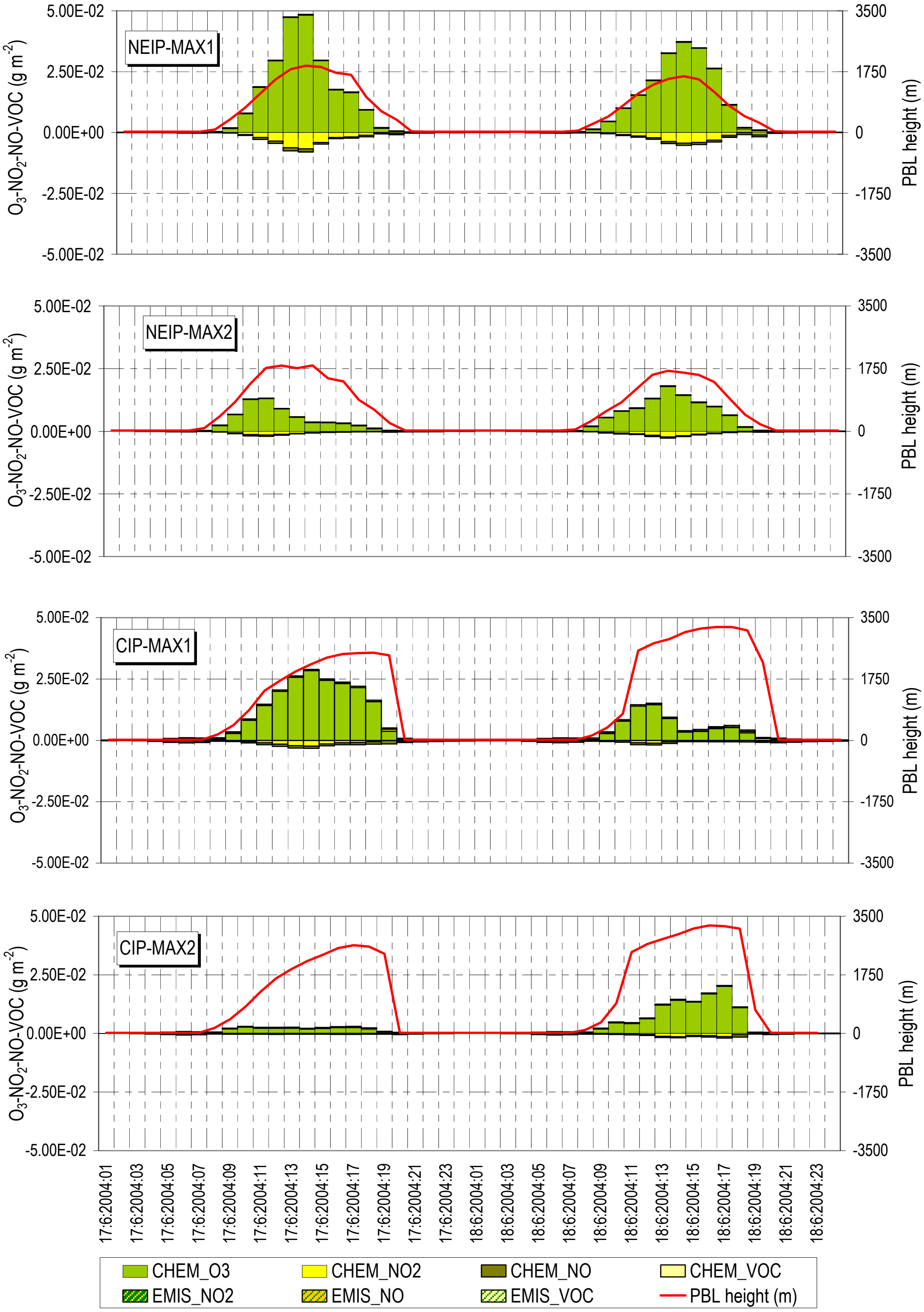


Figure 2.11. Gas-phase chemistry and emissions contributions to net NO_x-NMVOCs-O_3 density (g m^{-2}) under the modelled PBL height (in red) during the 17-18 June, 2004 for the NEIP-MAX1 and MAX2 domains, and the CIP-MAX1 and MAX2 domains.

Differences in chemical behaviour are observed for the studied domains (Figure 2.11). The coastal domains present similar chemical behaviours; the NMVOCs are the only locally emitted precursors, which react with transported NO_x generating O_3 by gas-phase chemistry. The CIP-MAX1 domain includes emitters of NO_x and NMVOCs, which react forming O_3, nevertheless the O_3 chemistry does not involve destruction of NMVOCs in the same magnitude as the coastal domains. In absence of solar radiation, when the O_3 stopped forming, the NO oxidation continues producing net NO_2 (1800-1900 UTC period). In the CIP-MAX2 domain the emissions are an important source of NMVOCs. These are chemically destroyed in the 1000 to 1900 UTC period on 17 June, being the generation of O_3 by chemistry relatively low. The low contribution of net transport of NO_x and the absence of emissions of this compounds in the area may be the main causes for the low O_3 production. On the second day of the episode the horizontal advection directly affect the MAX2 involving a much higher net chemical production of O_3, being the chemical oxidation of NMVOCs lower..

2.4 Conclusions

The Integrated Process Rate implemented in the CMAQ model was applied to obtain quantitative information about atmospheric processes affecting the concentration of pollutants in typical coastal and continental environments located in south-western Europe: the Northeastern Iberian Peninsula (NEIP) and Central Iberian Peninsula (CIP) domains, respectively. A representative summertime photochemical pollution episode characterized by stagnant conditions over the area was selected.

The model performance agrees with European Directives recommendations, nevertheless specifically in background air quality stations tends to overestimate the O_3 morning concentrations and underestimate the O_3 levels during the afternoon. The findings of this work together with previous studies results allow us to depict the main causes for these deviations, being aware that there is not an unique reason for a model failure in predictions. The chemical destruction may be underpredicted during night-time, which favours the high estimated morning concentrations. Moreover the overpredicted flows during the afternoon and dusk and night-time could cause an enhanced venting of pollutants. The mesoscale meteorological models have shown inaccuracies in predicting wind flows during the dominant stagnant conditions; which together with the underestimations in the emissions account play a fundamental role in these deviations. NO_2 is clearly linked to O_3, with both inaccuracies in transport and chemical behaviour being on the origin of the errors in the simulated concentrations. On the other hand, O_3 peaks and NO_2 underprediction in the coastal domain may be related to difficulties for the model to define the accumulation layers formed and characteristic recirculation of pollutants due to the very complex terrain. In the case of SO_2 and PM10 a slight underestimation of emissions could be the main cause for the underestimation of these pollutants.

Applying the Integrated Process Rate tool to the first vertical layer simulated provides information about the surface concentration of pollutants estimated by the model and permits to test the mass consistency. In order to perform a deeper study of the contributions of main atmospheric processes leading to the levels of these pollutants the vertical component has to be considered. In this work the whole atmospheric column under the PBL is selected, reflecting that chemistry and transport patterns vary with height.

The process analysis indicates that the maximum concentration of photochemical pollutants occur due to transport phenomena. Specifically in the coastal domain (NEIP), the high surface O_3 levels are not produced in situ, but come from horizontally advected flows during the morning and gas-phase chemical contributions occurring aloft. During the last hours of the day vertical advective flows inject the pollutants in layers above the PBL, which accumulate due to the stagnant conditions over the region and contribute positively to surface concentrations the next simulated day. The central-continental domain (CIP) behaviour slightly differs, with horizontal advection being also the main contributor to O_3 surface concentrations, but having the chemical formation some importance at low levels. The transport patterns differ between the coastal and the continental area, where the O_3 precursors are homogeneously distributed in the whole atmospheric column under the PBL and vertical injections of pollutants above the PBL are less frequent.

The dry deposition is an important sink for pollutants in the lowest layer of the model, coupled in the present simulation with vertical diffusion flows. In spite of the stagnant conditions dominating during the episode, the diffusive processes contributions to net pollutants concentrations under the PBL are relatively low, and in particular the horizontal diffusion is negligible compared to other atmospheric processes. Vertical diffusion compensates the loss of O_3 in surface layers due to NO titration, contributing positively to net O_3 concentrations in the road network and urban areas. The O_3 peaks at surface level are higher in the CIP domain, mainly due to the much simpler transport pattern compared to the coastal region, which involves that almost all the chemically produced O_3 under the PBL contributes to surface concentrations when the convective cell weakens. Controlling emissions of precursors in both domains is a decisive factor in order to abate photochemical pollution episodes during summertime. The emitted NMVOCs have mainly a biogenic origin and therefore policies focused on reducing NO_x emissions must be addressed in these regions. Finally, the cloud processes, wet deposition and heterogeneous chemistry contributions are negligible during the whole episode, characterized by a high solar radiation and no precipitation or cloudiness.

This work explores the possibilities of applying process analysis to high resolution simulations proving that it could be useful not only to better evaluate the simulation results, but also to perform more accurate source apportionment of pollutants over a region.

3 Natural gas vehicles use in urban areas. Effects on emissions and air quality: application to Barcelona and Madrid Greater Areas (Spain)

Corresponding to: Gonçalves, M., Jiménez, P. and Baldasano, J.M., 2008. *Emissions variation in urban areas resulting from the introduction of natural gas vehicles: application to Barcelona and Madrid Greater Areas (Spain) The Science of the Total Environment (under review)* and Gonçalves, M., Jiménez, P., Baldasano, J.M., 2009. *High resolution modelling of the effects of alternative fuels use on urban air quality: Introduction of natural gas vehicles in Barcelona and Madrid Greater Areas (Spain) The Science of the Total Environment 407, 776-790. doi:10.1016/j.scitotenv.2008.10.017*

3.1 Introduction

Improving air quality in urban areas is nowadays an important environmental challenge (Fenger, 1999; Baldasano et al., 2003). On-road traffic is the largest contributor to pollutants emissions in urban areas (Costa and Baldasano, 1996; Colvile et al., 2001; Querol et al., 2001; Artiñano et al., 2004; Ghose et al., 2004) and it remains a key target for public health action in Europe (Künzli et al., 2000). The southern Mediterranean region frequently register exceedances of the European air quality targets, particularly concerning PM_{10} and O_3 (Jiménez et al., 2006). Additionally high NO_2 levels are registered in conurbations (EEA, 2006a). Therefore different strategies for the abatement of on-road traffic emissions are currently being tested (Nagl et al., 2006).

In the European context the alternative fuels use is being promoted. Specifically, the White Paper on transport policy (EC,2001a) points out that the urban transport is already providing an useful market for expanding the use of alternative energies. Several European cities have taken this path: Paris, Florence, Stockholm, Luxembourg, Barcelona and Madrid have yet natural gas (NG), biofuel or hydrogen fuel cell urban buses. Gradually, private cars and trucks could turn to substitution fuels. The EU Green Paper towards a European strategy for the security for the Energy supply (EC, 2000) lays down the foundations to the energetic diversification. The European Commission (EC, 2001a; 2001b; 2007) proposes the introduction up to 20% of alternative energies in transport at 2020. It bets for biofuels in the short term, natural gas in the medium term and fuel cells or hydrogen internal combustion engines in the long term.

Air quality modelling associated with emissions scenarios has become an important tool for assessing the effects of these strategies on advance, providing pollutants concentration variation (Schell et al., 2002; Ponche and Vinuesa, 2005; Vautard et al., 2005b; Mediavilla-Sahagún and ApSimon, 2006; Reis et al., 2006). In this sense, several studies have been performed related to the impacts of alternative fuel use such as ethanol or compressed natural gas on air quality, human health or related costs (e.g. Asia-Beijing (Cheng et al., 2007), USA-Albuquerque (Gaffney et al., 1997), USA-nationwide (Jacobson, 2007) and urban areas of the United States (Cohen et al., 2003)).

This work assesses the impacts on emissions and air quality of a vehicle fleet change to set out the real efficacy of the introduction of natural gas as an alternative fuel. The study cases focus on the most populated cities of Spain: Barcelona and Madrid, during a typical summertime polluted episode of the year 2004. The scenarios proposed are based on the substitution of specific groups of vehicles: public transport fleet, freight transport fleet and private cars. Finally, a combined scenario is also evaluated. This study analyses the variation on emissions of primary pollutants: carbon monoxide (CO), nitrogen oxides (NO_x), sulphur dioxide (SO_2), non-methane volatile organic compounds (NMVOCs) and PM_{10}. $PM_{2.5}$ emissions are set separately, due to its effects related to human health. The impact on urban air quality of these scenarios is assessed by means of the WRF-ARW/HERMES/CMAQ modelling system in terms of ozone (O_3), nitrogen dioxide (NO_2), SO_2 and PM_{10} concentration variation.

3.2 Methods

The selected days: 17-18 June, 2004 are described in detail in *section 2.2.4* of *Chapter 2* and *Annex 1* of this document. The criteria followed for its selection were: (1) poor air quality conditions, set as worst case scenario to analyze the effects of emissions change in air quality; and (2) an usual traffic circulation pattern, skipping weekends or holidays, in order to obtain representative results (working days).

3.2.1 Particularities of the on-road transport module in the HERMES emissions model

A deep description of the WRF-ARW/HERMES/CMAQ modelling system is provided in the *section 2.2.1* of *Chapter 2*. This section summarizes the particularities concerning the on-road traffic emissions module and the changes performed to implement the new scenarios.

The high resolution (1 km^2, 1 hr) HERMES emissions model specific for the Iberian Peninsula (Baldasano et al., 2008a) is used to evaluate the change in traffic emissions for each scenario. The traffic emissions module of HERMES considers fundamentally a bottom-up approach and takes into account 72 diesel and petrol vehicles categories (including Euro II and Euro III emission standards) according to COPERT III - EMEP/CORINAIR methodology (Ntziachristos and Samaras, 2000; EEA, 2006b); divided by fuel type, vehicle weight, age of the vehicle and cubic capacity; each of them with its specific emissions factors, defined as a function of the circulation speed. The emissions account in HERMES traffic module considers hot exhaust, cold exhaust and evaporative emissions. It also estimates PM produced by brakes abrasion, tire wear and pavement erosion. The vehicular fleet is defined for Spain, and specifically for Barcelona and Madrid areas (Figure 3.2), using the year 2004 data provided by the national traffic management organism of Spain (Dirección General de Tráfico), and distributed in the 72 previous mentioned categories. The model includes the definition of the road network, dividing it in stretches (inside the 1 km^2 cells) with specific temporary disaggregating profiles (distinguishing day-type: weekday-holiday, and month), average speed circulation, daily average traffic (number of vehicles per day), stretch length, route type (highway, road or urban) and circulation zones for Barcelona and Madrid. This information covers 67% of the intercity roads length for the whole national territory and 80-85% of the total traffic volume. Moreover the available information covers the total road network for Barcelona and Madrid Greater Areas, which involves the 50% of the national urban traffic volume

The mobility zones in the Barcelona and Madrid urban permit to distinguish vehicles included in the same category but differenced by their activity, i.e. taxis from private cars or heavy duty vehicles from buses. The metropolitan transport organizations provide mobility data for the greater areas of Madrid and Barcelona (Table 3-1). According to them five areas are defined in Barcelona (Figure 3.1), resulting in a different distribution by vehicle activity. The taxis circulation is enhanced in the downtown area (zone 1. Main roads), being 18.0% of the total volume of vehicles, while the heavy duty vehicles proportion is lower (14.8%). The ring-roads (zone 3. Ronda de Dalt and zone 4. Ronda Litoral) and the industrial area of Zona Franca (zone 5) are characterized by lower ratio of taxis (6.3%, 4.5%, respectively) and a higher ratio of heavy duty vehicles (21.7% and 23.4% in the ring roads and 30.7% in the Zona Franca). In Madrid the mobility data result in four zones definition (Figure 3.1), consisting of approximately concentric areas from the downtown. Applying the mobility data, the taxis percentage in the downtown achieves 20.3%, being just 6.0% in the outskirts. The heavy duty vehicles vary from 5.8% (zone 2) to 8.9% (zone 4), differing considerably from the Barcelona mobility patterns, partially due to the historically larger industrial activity in this later city.

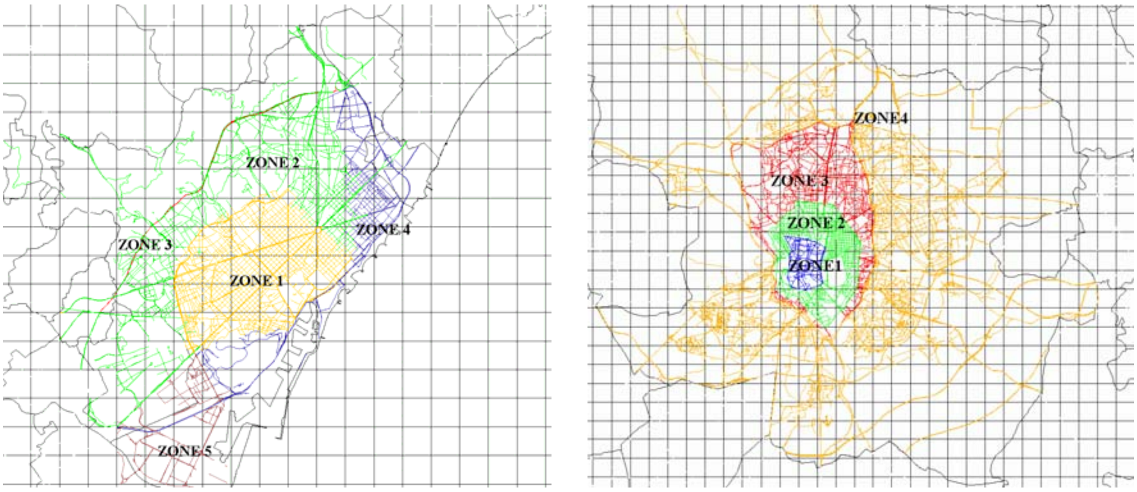


Figure 3.1 Circulation zones distribution for Barcelona (left) and Madrid (right).

The WRF-ARW/HERMES/CMAQ is applied with high spatial (1 km^2) and temporal (1 hr) resolution. The use of fine scale was demanded by the need for assessing the subtle air quality variations in urban areas, as shown in the CityDelta project experience (Cuvelier et al., 2007; Thunis et al., 2007) and others (e.g. Vivanco et al., 2009); and in order to describe the transport and transformation of pollutants, as well as the dynamics on an hourly basis in very complex terrains as the study areas (Jiménez et al., 2005a; 2005b; 2006). The base case is defined taking into account 2004 data and the emissions scenarios designed intend to be as feasible as possible, defining the vehicle fleet groups susceptible of change and considering the substitution of the oldest diesel and petrol vehicles by NGV.

Four nested domains were defined for the simulations (see *section 2.2.3* in *Chapter 2*), centring the final domains (D4) in Barcelona and Madrid. They cover the Northeastern Iberian Peninsula (322 x 259 km^2) and the Central Iberian Peninsula (181 x 214 km^2) respectively, to assess not only the effects in urban areas, but also to detect the urban plume behaviour. The high resolution requires of high-performance computing. The availability of the MareNostrum supercomputer hold in the BSC-CNS, together with the advances in the parallelization of air quality model codes, have allowed the high-resolution simulations and the large number of scenarios.

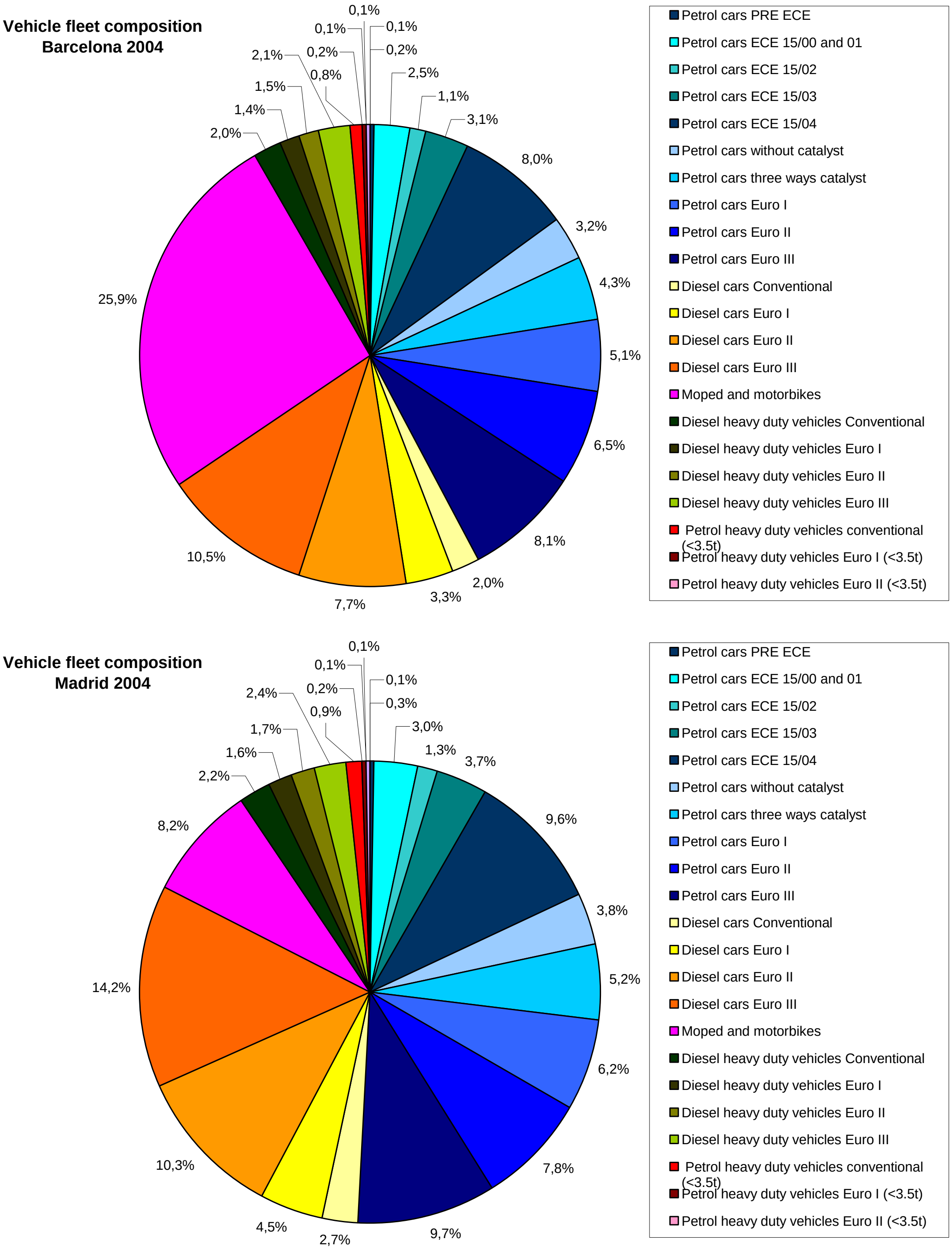


Figure 3.2. Vehicle fleet composition in 2004 in Barcelona (up) and Madrid (down): different categories of petrol cars (blue), diesel cars (orange), moped and motorbikes (pink), categories of diesel HDV (green) and petrol HDV (red).

Table 3-1 Vehicles distribution in each circulation zone in Barcelona (up) and Madrid (down), differentiating taxis from private cars and buses (urban buses and coaches) from HDV

ZONES IN BARCELONA	Mopeds	Private cars	Taxis	Buses	HDV
Zone 1. Main roads	25.80%	39.00%	18.00%	2.40%	14.80%
Zone 2. Access roads	5.10%	59.70%	10.00%	1.50%	23.70%
Zone 3. Ronda de Dalt	6.40%	63.60%	6.30%	2.00%	21.70%
Zone 4. Ronda Litoral	2.30%	65.80%	6.30%	2.20%	23.40%
Zone 5. Zona Franca	2.00%	60.40%	4.50%	2.40%	30.70%
ZONES IN MADRID [(a)]	LDV (including mopeds and motorbikes)		Taxis	Buses	HDV
Zone 1. Inside the 1st ring road	67.70%		20.30%	3.80%	8.20%
Zone 2. Between the 1st and the 2nd ring road	75.98%		15.83%	2.42%	5.77%
Zone 3. Between the 2nd ring road and the M30	81.40%		9.70%	2.30%	6.60%
Zone 4. Outside the M30	82.60%		6.00%	2.50%	8.90%

(a) Each zone includes the external ring-road.

3.2.2 Implementation of the scenarios in the traffic emissions module

In order to evaluate the change in emissions due to the introduction of natural gas vehicles (NGV) there are two requirements: (1) estimation of the speed-dependant emission factors for different categories of NGV (cars, light duty vehicles-LDV-, and heavy duty vehicles-HDV-); (2) definition of the vehicle number variation in each scenario.

3.2.2.1 Emission factors

Natural gas can be used as a fuel for different vehicle technologies and with different operation systems (mainly on stoichiometric mode for low emissions or on lean mode for higher efficiency (Samaras and Zierock, 2008)). Therefore a wide range of emission factors can be found for this alternative fuel use, which hardens the selection process. A review and comparative analysis of the most suitable emission factors for NGV is carried out. To achieve the objectives of this work: (1) speed-dependant emission factors are required; (2) European emission factors are preferred conditioned by the study area; and (3) all the emission factors may preferably come from the same source to guarantee homogeneity.

Currently, the pollutant emissions for different technology NGV can be estimated by means of (1) non speed-dependant bulk emission factors (Calais and Sims, 2000; Coroller and Plassat, 2003; Kremer, 1999; Eudy, 2000; Nylund et al., 2004, 2005; Rabl, 2002; Ristovski et al., 2004; Samaras and Zierock, 2007); (2) emissions correction factors that can be applied to diesel or petrol emission factors (Hickmann, 1998; Samaras, 1998; IEA, 1999; ENGVA, 2006).

The peer reviewed literature reflects that currently the natural gas use focus on urban areas. Specifically, it is being used as a fuel in the urban buses fleet of different European cities (Ntziachristos and Samaras, 2005), which enlarges the available data corresponding to this kind of vehicles. The emissions standards accomplished by these vehicles vary, so do the methods for the emissions assessment, resulting in a wide dispersion of the bulk emission factors found (Table 3-2). There is an acceptable agreement in VOCs and NOx emission factors for CNG Euro II HDV (6.08 g km^{-1} and 13.72 g km^{-1} on average, respectively, being the standard deviation of the compiled values 1.57 and 3.28), nevertheless the CO and PM emission factors present a larger dispersion. The reliability of the Euro III to V HDV emission factors is lower, exceeding in some cases the standard deviation values the average emission factor, i.e. the Enhanced Environmentally Friendly Heavy Duty Vehicles (EEV HDV) estimations indicate an emission factor of 1.54 g CO km^{-1}, and the standard deviation is 1.81 (Table 3-2).

The available emission rates of NG light duty vehicles are sparse (Table 3-2). Fundamentally provided by the European Natural Gas Vehicles Association (ENGVA) and obtained by testing specific type of vehicles (i.e.: Euro IV Ford Transit vans), these emission factors are not complete enough to extract conclusions. Although the data dispersion is lower than in the HDV case, this fact is attributed to the common origin of main part of the data. Considering this partial information the NG-LDVs would emit 0.47 g km^{-1} of CO and 0.04 g km^{-1} of NMVOCs on average (the standard deviation for these estimates is 0.25 and 0.02, respectively). The peer reviewed values are coherent, being lower for LDV than for HDV.

Ristovski et al. (2004) provide emission factors for petrol cars fitted to operate either with petrol or compressed NG. The parameters established in their study (a correlation between speed circulation and emission factors to some discrete values) are not comparable to those obtained from the other sources. Furthermore, the dual fuelled vehicle studied does not fit in the parameters of this work. Nevertheless the revision of NGV emission factors they perform agree with our results, obtaining very large variations in emission levels for different vehicles. This study remarks that the technology and condition of the vehicles is an important determinant of the emissions levels, but assures that the compressed natural gas engines present lower emissions of toxic compounds than petrol or diesel equivalent engines.

The second group of references proposes the use of emission correction factors. That is, a factor relating the emissions of the alternative fuel vehicle, like NGV, to an existing technology or fuel, like diesel or petrol vehicles (Table 3-3). With the exception of the ENGVA data, the sources do not provide correction factors for all pollutants in this study (NO_x, NMVOCs, CO, SO_2 and PM_{10}) for HDV, LDV and cars, although they are an useful reference. The correction factors for NG LDV with respect to petrol LDV agree for NMVOCs and NO_x, being on average 2% and 45% of the correspondent petrol vehicle, respectively (deviations of 2% and 13%). Larger discrepancies are found in the methane estimation, being the total emitted VOCs 145% of the petrol LDV on average (standard deviation of 45%).The reliability of CO correction factors is even lower (average factor: 38%; standard deviation 14%). For PM there is no emission correction factor applicable to petrol LDV.

The correction factors compiled for HD-NGV are referred to diesel HDV. The emission standards accomplished by the reference vehicles are unknown, except for the ENGVA provided values, which are related to diesel Euro III vehicles. The values acceptably agree for PM and VOCs levels, being the deviations larger for CO (170% of standard deviation of the sample which average correction factor is 165%) and NO_x corrections. They provide larger VOCs emissions for the HD-NGV than those of the diesel vehicles, mainly due to the CH_4 contribution, being the reported NMVOCs lower in all cases. The PM emissions for a HD-NGV are on average the 11% of the same vehicle propelled by diesel (accounting the standard deviation for 4%).

The previous analysis reflects the lack of information concerning emission factors for natural gas vehicles and the dispersion of the collected data. The need for using speed dependant emission factors invalidates the bulk emission factors. Moreover the peer reviewed correction factors provide incomplete data or values referred to unknown technologies. For all these reasons and to guarantee the homogeneity the reduction factors provided by ENGVA (2006) shown in Table 3-3 (in yellow) are used. They represent the emissions reduction of each pollutant for a NGV referred to a diesel Euro III vehicle of the same category, distinguishing cars and LDV from HDV. These are in accordance with the MEET project conclusions in terms of NGV emissions, that is, CO emissions are significantly reduced with the use of NG (a NGV emits 53% to 58% of a diesel vehicle emission according to ENGVA), NO_x is generally reduced (16% or 18% of the diesel vehicles NO_x emissions). Considering that the VOCs emitted by a NGV are mainly CH_4 (95%) (Rabl, 2002) it would be possible to estimate CH_4 emissions.

Table 3-2 Emission factors for compressed NG (CNG) HDV and LDV. Average values and standard deviation of data obtained from different sources

Emission factors for CNG Heavy duty vehicles (g km^{-1})						
Emission standard	CO	VOCs	NMVOCs	NOx	PM	Source
Euro I CNG	8.4	7.0		16.5	0.02	Samaras and Zierock, 2007
Euro II CNG	4.00	7.00		17.00	0.01	Nylund and Erkkilä, 2005 (a)
Euro II CNG	5.40	8.40		14.80	0.04	Coroller and Plassat, 2003
Euro II CNG	0.60	5.30		13.50	0.03	Coroller and Plassat, 2003
Euro II CNG	12.00	5.00		8.30	0.03	Coroller and Plassat, 2003
Euro II CNG	2.7	4.7		15	0.01	Samaras and Zierock, 2007
AverageEII (g km^{-1})	*4.94*	*6.08*		*13.72*	*0.024*	
Standard deviation	*4.32*	*1.57*		*3.28*	*0.01*	
Unknown	0.66		2.75	9.87	0.05	Beer et al., 2000
Unknown	0.03	0.38		1.73	0.01	ENGVA, 2006 (b)
Euro III CNG(1)	0.20	1.00		10.00	0.01	Nylund and Erkkilä, 2005
Euro III CNG	0.38	1.17	0.03	16.92	0.01	ENGVA, 2006 (b)
Euro III CNG	1.0	1.33		10.00	0.01	Samaras and Zierock, 2007
Average (g km^{-1})	*0.45*	*0.97*	*1.39*	*9.70*	*0.02*	
Standard deviation	*0.38*	*0.42*	*1.92*	*5.38*	*0.02*	
Euro V CNG (1)	1.00	1.00		3.00	0.01	Nylund and Erkkilä, 2005
EEV (Euro IV-V)	0.13	1.78	0.04	10.80	0.02	ENGVA, 2006 (b)
EEV (Euro IV-V)	0.85	0.36	0.02	3.73	0.01	ENGVA, 2006 (b)
EEV (Euro IV-V)	4.72	2.11	0.04	3.89	0.01	ENGVA, 2006 (b)
EEV (Euro IV-V)	1.0	1.0		2.5	0.005	Samaras and Zierock
Average (g km^{-1})	*1.54*	*1.25*	*0.03*	*4.78*	*0.01*	
Standard deviation	*1.81*	*0.70*	*0.01*	*3.41*	*0.01*	
Emission factors for NG light duty vehicles (g km^{-1})						
Emission standard	CO	VOCs	NMVOCs	NOx	PM	Source
Unknown	0.78	0.09	0.05	0.02		ENGVA, 2006 (b,c)
Unknown	0.65	0.09	0.06	0.02		ENGVA, 2006 (b,c)
Euro IV	0.40	0.07		0.01		ENGVA, 2006 (b,c)
Euro IV	0.37	0.06		0.02		ENGVA, 2006 (b,c)
Unknown	0.14		0.02	0.08		Kremer, 1999
Average (g km^{-1})	*0.47*	*0.08*	*0.04*	*0.03*		
Standard deviation	*0.25*	*0.02*	*0.02*	*0.03*		

(a) More than 200 tests carried out within 34 individual buses (included diesel buses not shown here)

(b) The European Natural Gas Vehicles Association (ENGVA) provided several sources of information concerning the emission factors of natural gas vehicles.

(c) Tests results sheet for a Ford Transit van.

Table 3-3 Emission correction factors for NG-LDV respect to petrol LDV and diesel LDV and correction factors for NG-HDV with respect to diesel vehicles. The finally selected correction factors are highlighted in yellow.

NGV category	Reference vehicle	Correction factor (CF) (a) $CF=EF_{NG}/EF_{reference\text{-}vehicle}$					
		CO	VOCs	NMVOCs	NO_x	PM	Source
NG-LDV	Petrol car with TWC (b)	0.38	1.81	0.13	0.37		Samaras, 1998
	Petrol LDV with TWC	0.52	1.80	0.11	0.61		Samaras, 1998
	Petrol LDV	0.24	1.02		0.39	0.00	IEA. 1999.
	Average	*0.38*	*1.54*	*0.12*	*0.45*		
	Standard deviation	*0.14*	*0.45*	*0.02*	*0.13*		
	Euro III diesel cars and LDV (< 7.5t)	0.53		0.44	0.18	0.05	ENGVA, 2006
NG-HDV	Diesel HDV	0.46	3.40		0.58	0.09	Samaras, 1998.
		3.60	3.98		0.25	0.15	IEA, 1999.
		0.90	6.00		0.30	0.08	Rabl, 2002.
	Average	*1.65*	*4.46*		*0.38*	*0.11*	
	Standard deviation	*1.70*	*1.36*		*0.18*	*0.04*	
	Euro III diesel HDV	0.58		0.11	0.16	0.12	ENGVA, 2006

(a). SO_2 emission factor is considered as zero, because of the low sulphur content of NG (less than 10 ppb).

(b).TWC: three ways catalyst.

Nowadays, the speed-dependant correlation for the NGV emission factors is not available. Usually the emission correction factors proposed by the different authors (Hickmann, 1998, Samaras, 1998, IEA, 1999) are referred to the average speed of the analysis cycle. Considering that Friedrich and Bickel (2001) propose a constant reduction factor for the NG bus emissions with respect to diesel bus emissions that can be used in all the speed range, the emission reduction factors (Table 3-3) are used in all the speed range to estimate the NGV emissions. However, we should bear in mind that Samaras (1998) warns against the use of these correction factors for the whole speed range.

This ratio is applied to cold and hot emissions since it is a relationship between total exhaust emissions and emission factors. Non-combustion particle emissions are independent of the vehicle change, because it is considered that both the new and the old vehicles have the same weight and that the driving conditions are essentially analogous.

The use of speed dependant emission factors provides a more realistic representation of the vehicles behaviour. The final estimations of emission factors for NGV are compared with the peer-reviewed factors when possible. These are found in the same order of magnitude, especially for PM (Figure 3.3) and CO (Figure 3.4). Nevertheless the speed dependency shows that at low speed considering bulk emission factors would underestimate the natural gas vehicles emissions, while at high circulation speeds they would produce an overestimation of emissions. The NO_x emission factors for NG-HDV behave in an analogous way, nevertheless the U shaped speed dependency for LDV is not captured by the constant emission factors (Figure 3.5).

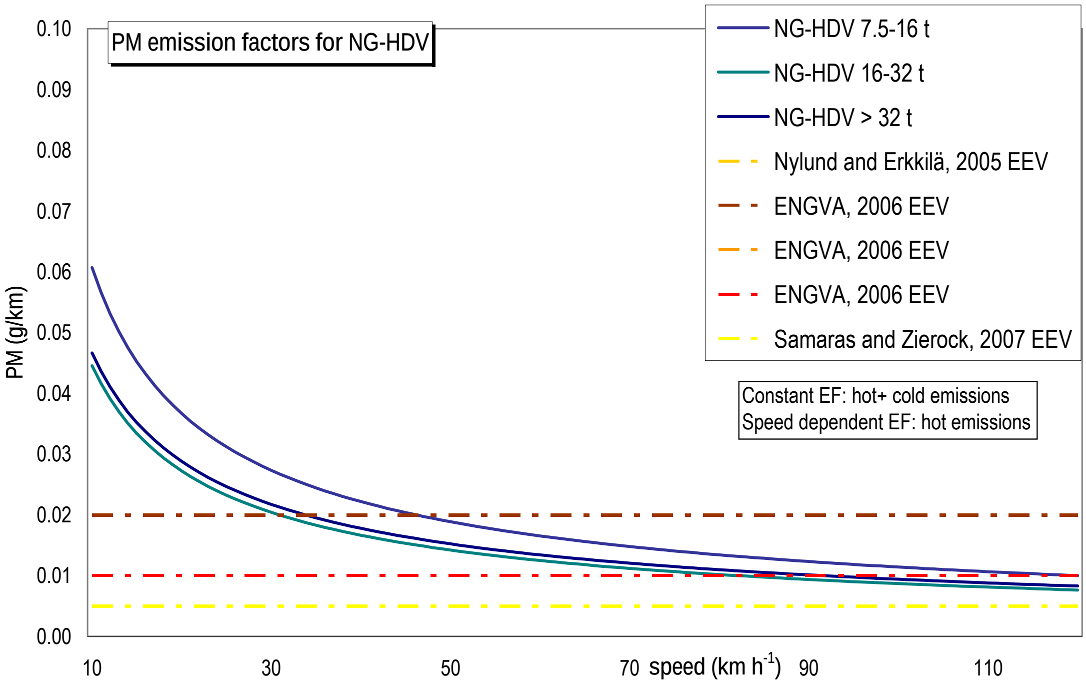


Figure 3.3 PM emission factors estimated for natural gas heavy duty vehicles –NG-HDV-. The dotted lines present the peer-reviewed constant emission factors and the solid lines those estimated by the chosen correction factors (ENGVA, 2006) applied to the correspondent diesel emission factors (EEA-EMEP CORINAIR, 2000)

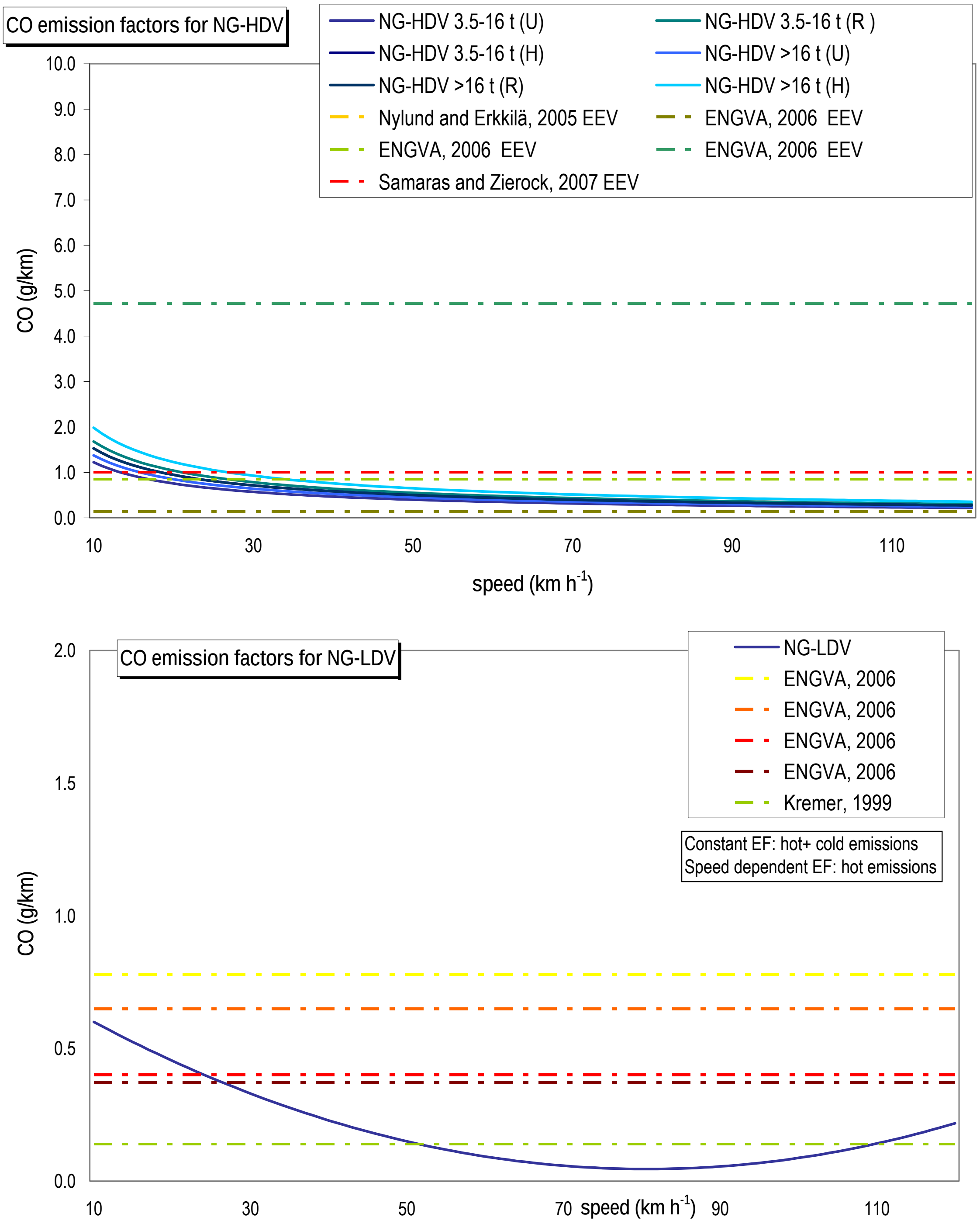


Figure 3.4. CO emission factors estimated for natural gas heavy duty vehicles –NG-HDV- (up) and light duty vehicles –NG-LDV- (down). The dotted lines present the peer-reviewed constant emission factors and the solid lines those estimated by the chosen correction factors (ENGVA, 2006) applied to the correspondent diesel emission factors (EEA-EMEP CORINAIR, 2000).

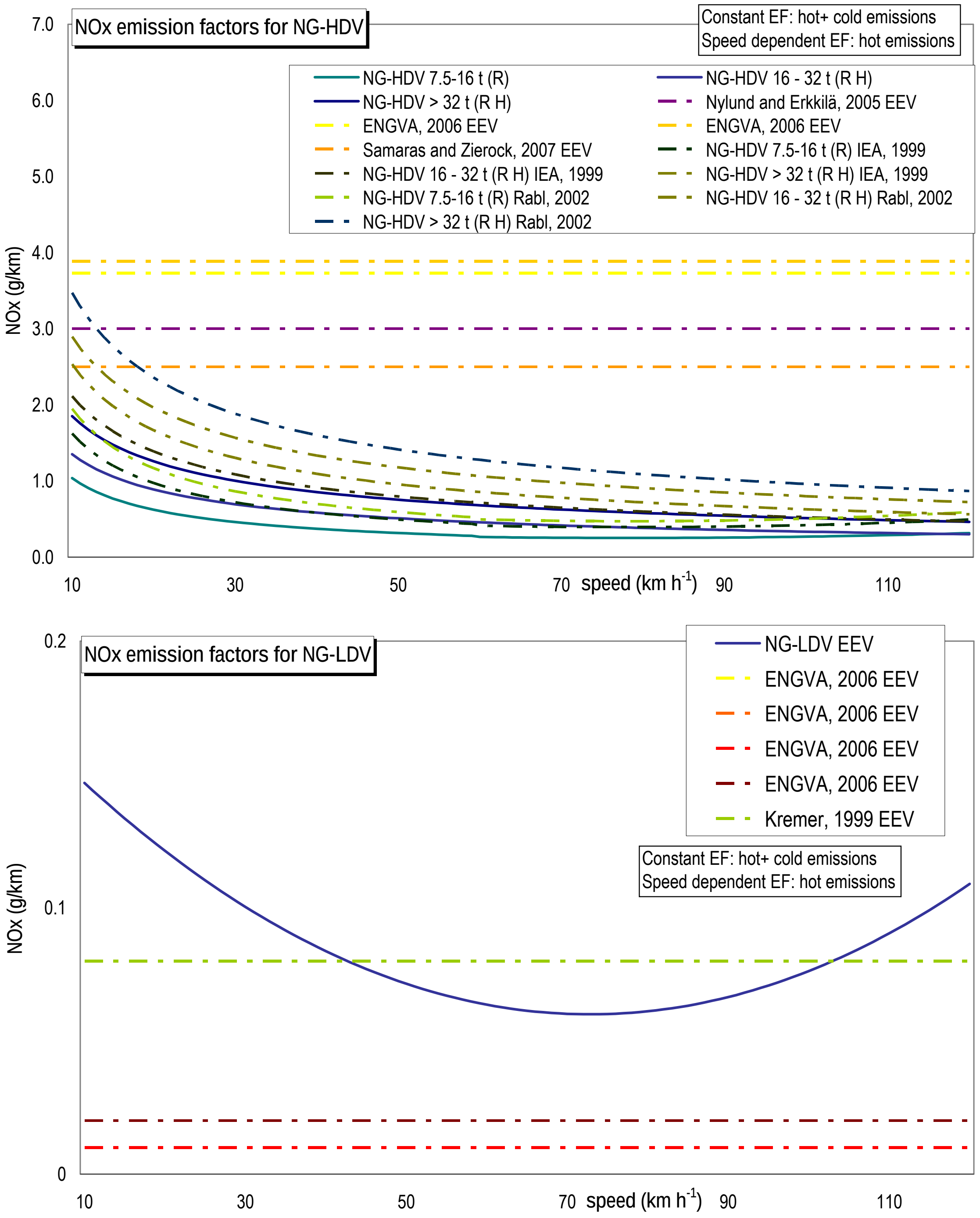


Figure 3.5 NO$_x$ emission factors estimated for natural gas heavy duty vehicles –NG-HDV- (up) and light duty vehicles –NG-LDV- (down). The dotted lines present the peer-reviewed constant emission factors and the estimations done with the correction factors from Samaras (1998), IEA (1999) and Rabl (2002).The solid lines present the emission factors estimated by the chosen correction factors (ENGVA, 2006) applied to the correspondent diesel emission factors (EEA-EMEP CORINAIR, 2000)

3.2.2.2 Vehicle fleet variations

The base case scenario (BC) is based on the vehicle fleet composition for the year 2004 in Barcelona and Madrid cities (Spain). Specifically the emissions are calculated with HERMES for the 18 June, 2004. To implement the different scenarios the vehicle fleet composition in the urban areas is changed according to the type and percentage of NGV introduced in each case. The reduction scenarios proposed are:

(E1) Scenario 1. Transformation to NGV of 100% of urban buses fleet;

(E2) Scenario 2. Transformation to NGV of 50% of taxis fleet;

(E3) Scenario 3. Transformation to NGV of 50% of intercity buses fleet;

(E4) Scenario 4. Transformation to NGV of 50% of light commercial vehicle fleet;

(E5) Scenario 5. Transformation to NGV of 10% of private cars fleet;

(E6) Scenario 6. Transformation to NGV of 100% of heavy duty freight transport vehicle fleet;

(E7) Scenario 7. Combined scenario

The vehicle fleet composition circulating in the urban zones is modified to introduce these scenarios adding natural gas vehicles categories (Table 3-4 and Table 3-5). The same percentage of the oldest diesel and petrol vehicles is removed.

Substituting urban buses (E1), intercity buses (E3) and heavy duty freight transport vehicles (E6) do not involve a vehicle fleet change larger than 1.5% of the vehicles circulating either in Barcelona or in Madrid in any zone.

Changing the 50% of the taxis removes the pre-Euro I and a fraction of the Euro I diesel cars from the greater areas. 9.0% of NG cars are introduced in Barcelona access roads (zone 2) and 10.1% in Madrid downtown (zone 1)

When transforming the light commercial vehicles (E4), the largest variations in vehicle fleet are estimated in the industrial and port zone of Barcelona (zone 5, 14.7%) and in outskirts of Madrid (zone 4, 4.3%). This fleet renewal involves removing all the petrol and diesel Conventional commercial light vehicles (lower than 3.5 t) and partially those included in the Euro I category in Barcelona. In Madrid not only all the diesel and petrol commercial vehicles lower than 3.5 t, but also some of the diesel Conventional vehicles from 3.5 t to 7.5 t and of the petrol vehicles weighting more than 3.5 t have been changed. Expected changes in emissions in this scenario will be much higher in Barcelona than in Madrid, due to the vehicle fleet composition and the higher weight of light commercial vehicles in the former.

Table 3-4 Percentage of the vehicles changed in each scenario for Barcelona, differentiated by circulation zone and type of vehicle.

Reduction scenario	Previous vehicle category	ZONE 1	ZONE 2	ZONE 3	ZONE 4	ZONE 5
Scenario 1. NG urban buses	Heavy duty diesel vehicle 3.5-7.5t	0.04%	0.06%	0.05%	0.06%	0.06%
	Heavy duty diesel vehicle 7.5-16t	0.50%	0.80%	0.67%	0.73%	0.80%
	Heavy duty diesel vehicle 16-32t	0.19%	0.30%	0.25%	0.28%	0.30%
TOTAL		0.73%	1.16%	1.07%	0.97%	1.16%
Scenario 2. NG taxis	Diesel cars	5.00%	9.00%	3.15%	3.15%	2.25%
TOTAL		5.00%	9.00%	3.15%	3.15%	2.25%
Scenario 3. NG intercity buses	Heavy duty diesel vehicle 7.5-16t	0.33%	0.53%	0.49%	0.44%	0.53%
	Heavy duty diesel vehicle 16-32t	0.05%	0.09%	0.08%	0.07%	0.09%
TOTAL		0.39%	0.62%	0.57%	0.52%	0.62%
Scenario 4. NG light commercial vehicles	Heavy duty diesel and petrol vehicles <3.5	11.12%	6.95%	10.98%	10.19%	14.41%
	Heavy duty diesel and petrol vehicles 3,5-7.5t	0.21%	0.13%	0.21%	0.19%	0.28%
TOTAL		11.34%	7.08%	11.19%	10.38%	14.69%
Scenario 5. NG cars	Petrol and diesel cars	5.97%	3.90%	6.58%	6.36%	6.04%
TOTAL		5.97%	3.90%	6.58%	6.36%	6.04%
Scenario 6. NG heavy duty freight transport vehicles	Heavy duty diesel vehicle 7.5-16t	0.76%	0.48%	0.75%	0.70%	0.99%
	Heavy duty diesel vehicle 16-32t	0.20%	0.13%	0.20%	0.18%	0.26%
	Heavy duty diesel vehicle >32t	0.06%	0.04%	0.06%	0.06%	0.08%
TOTAL		1.03%	0.64%	1.01%	0.94%	1.33%
Scenario 7. NG urban and intercity buses, cars, taxis, commercial vehicles, heavy duty freight transport vehicles	Petrol and diesel cars	10.97%	12.90%	9.73%	9.51%	8.29%
	Diesel and petrol HDV <3,5t	11.12%	6.95%	10.98%	10.19%	14.41%
	Diesel and petrol HDV 3,5-7,5t	0.25%	0.20%	0.27%	0.25%	0.34%
	Diesel HDV 7.5-16t	1.59%	1.81%	1.97%	1.81%	2.32%
	Diesel HDV 16-32t	0.44%	0.51%	0.55%	0.51%	0.65%
	Diesel HDV >32t	0.06%	0.04%	0.06%	0.06%	0.08%
TOTAL		24.45%	22.40%	23.57%	22.31%	26.09%

Table 3-5 Percentage of the vehicles changed in each scenario for Madrid, differentiated by circulation zone and type of vehicle.

Reduction scenario	Previous vehicle category	ZONE 1	ZONE 2	ZONE 3	ZONE 4
Scenario 1. NG urban buses	Heavy duty diesel vehicle 3.5-7.5t	0.10%	0.07%	0.06%	0.07%
	Heavy duty diesel vehicle 7.5-16t	1.17%	0.75%	0.71%	0.77%
	Heavy duty diesel vehicle 16-32t	0.05%	0.03%	0.03%	0.04%
TOTAL		1.33%	0.85%	0.80%	0.87%
Scenario 2. NG taxis	Diesel cars	10.15%	7.92%	4.85%	3.00%
TOTAL		10.15%	7.92%	4.85%	3.00%
Scenario 3. NG intercity buses	Heavy duty diesel vehicle 7.5-16t	1.02%	0.65%	0.62%	0.67%
	Heavy duty diesel vehicle 16-32t	0.22%	0.14%	0.13%	0.14%
TOTAL		1.24%	0.79%	0.75%	0.81%
Scenario 4. NG light commercial vehicles	Heavy duty diesel and petrol vehicles <3.5	3.74%	2.63%	3.01%	4.06%
	Heavy duty diesel and petrol vehicles 3,5-7.5t	0.18%	0.13%	0.15%	0.20%
TOTAL		3.92%	2.76%	3.16%	4.26%
Scenario 5. NG cars	Petrol and diesel cars	6.16%	6.91%	7.40%	7.51%
TOTAL		6.16%	6.91%	7.40%	7.51%
Scenario 6. NG heavy duty freight transport vehicles	Heavy duty diesel vehicle 7.5-16t	0.26%	0.19%	0.21%	0.29%
	Heavy duty diesel vehicle 16-32t	0.07%	0.05%	0.06%	0.08%
	Heavy duty diesel vehicle >32t	0.02%	0.02%	0.02%	0.02%
TOTAL		0.36%	0.25%	0.29%	0.39%
Scenario 7. NG urban and intercity buses, cars, taxis, commercial vehicles, heavy duty freight transport vehicles	Petrol and diesel cars	16.31%	14.83%	12.25%	10.51%
	Diesel and petrol HDV <3,5t	3.74%	2.63%	3.01%	4.06%
	Diesel and petrol HDV 3,5-7,5t	0.29%	0.20%	0.21%	0.27%
	Diesel HDV 7.5-16t	2.45%	1.58%	1.54%	1.73%
	Diesel HDV 16-32t	0.34%	0.22%	0.22%	0.25%
	Diesel HDV >32t	0.02%	0.02%	0.02%	0.02%
TOTAL		23.15%	19.47%	17.25%	16.84%

The substitution of 10% of private cars is mainly reflected in the Dalt ring road of Barcelona (zone 3, 6.6%), and in Madrid outskirts (zone 4, 7.5%), being larger in last case. This scenario affects private cars, both petrol and diesel. It involves changing all diesel Conventional and all petrol pre-ECE, ECE-15/01 and ECE-15/02 cars. In Madrid the petrol ECE-15/03 are also partially modified.

Finally the combined scenario is estimated as the addition of the changes performed in all previous scenarios, changing a percentage of vehicles of 26.1% in Barcelona and 23.1% in Madrid.

The proposed scenarios are comparable (Table 3-6) with the European scheduled plan (EC, 2001a) in terms of fuel consumption. The substitution of 100% of urban buses (E1), 50% of intercity buses (E3) or 100% of heavy duty freight transport (E6) are comparable to the 2% proposed for 2010 (values ranging from 1.4% to 3.2%). The transformation of 50% of taxis (E2) or 10% of private cars (E5) are close to the 2015 proposal (5% of fuel substitution) with 3.1 - 6.8% of current fuel substituted by NG. Finally the change of 50% of commercial light vehicles involves the transformation of 6.0% - 12.9% which is near to the 10% proposed to 2020. The fuel change in the combined scenario is around 20% (10%) in Barcelona (Madrid) larger than the most optimistic ratio of introduction proposed by the EU, despite its interest lays on representing the aggregated effect.

Table 3-6 Fuel consumption –FC- (gasoline- diesel in t d^{-1}) and % substituted by NG in the proposed scenarios.

SCENARIO	Barcelona		Madrid	
	FC [(a)] (t d^{-1})	% change	FC[(a)] (t d^{-1})	% change
Base case scenario. 2004 vehicle fleet composition	1376		3607	
Scenario 1. NG urban buses	1332	3.2	3504	2.9
Scenario 2. NG taxis	1313	4.6	3496	3.1
Scenario 3. NG intercity buses	1353	1.7	3503	2.9
Scenario 4. NG light commercial vehicles	1198	12.9	3391	6.0
Scenario 5. NG cars	1310	4.8	3362	6.8
Scenario 6. NG heavy duty freight transport vehicles	1333	3.1	3557	1.4
Scenario 7. Combined scenario	958	30.4	2777	23.0

[(a)] Estimated gasoline and diesel consumption in each scenario.

3.3 Emissions analysis

The study areas are centred in the two largest and most populated cities of Spain, where problems related with poor air quality and traffic derived emissions are of special concern. These are: the Spanish administrative capital, Madrid, located in the Central Iberian Peninsula, where a domain is defined covering 373 km^2 and accounting for 2.840 million inhabitants; and the second largest conurbation of Spain, Barcelona, located in the northern Mediterranean coast, where a 130 km^2 area is defined including 1.770 million inhabitants (Figure 3.6).

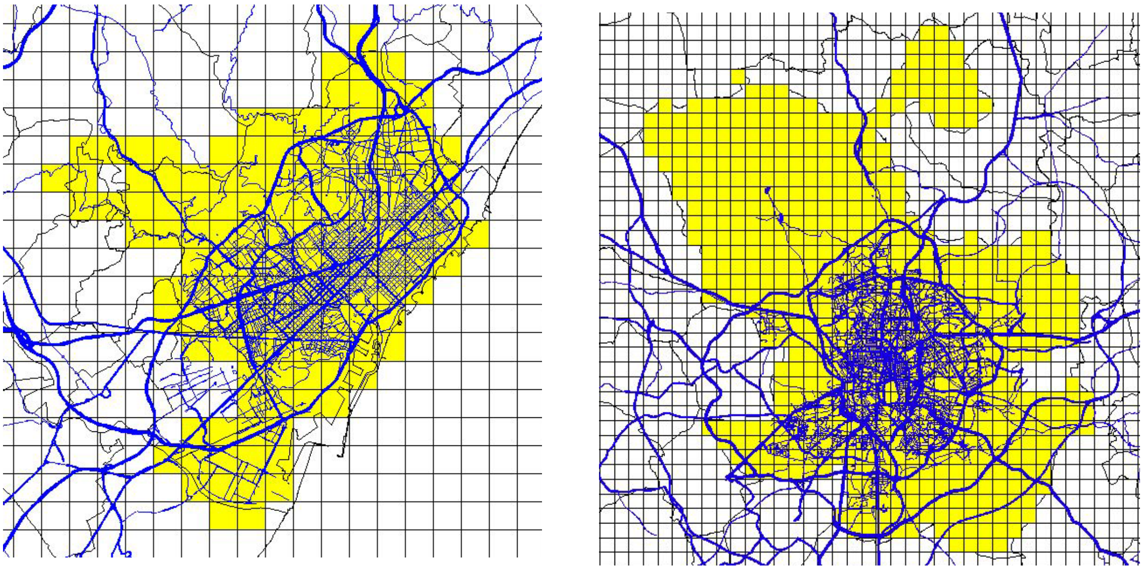


Figure 3.6. Areas defined to assess the emissions variation over Barcelona area (right) and Madrid area (left) in yellow.

The vehicles fleet in Barcelona is lower than 1 million vehicles and the city has a historically intense industrial activity, while Madrid involves over 1.7 million of circulating vehicles and the industrial activity in the city and the surrounding areas has been traditionally devoted to the services sector (Artiñano et al., 2004). This is reflected in the emissions account for the base case scenario: the 86% of the total mass emissions (NO_x, NMVOCs, CO, SO_2 and PM_{10}) in Barcelona come from on-road traffic, face to the 93% in Madrid. The weight of industrial emissions is higher in Barcelona (7%) than in Madrid (1%).

The on-road traffic is mainly responsible for NO_x emissions (81% in Barcelona, 94% in Madrid), CO emissions (98% in Barcelona, 100% in Madrid) and NMVOCs (77% in Barcelona and 79% in Madrid) -Figure 3.7-. The industrial component importance in Barcelona is also reflected in the SO_2 emissions (53%). In Madrid the traffic and energy consumption of domestic and commercial sector substitute the industry as main SO_2 emitter (contributing by 53% and 42%, respectively). Concerning $PM_{10,}$ the industrial sector contribution is also important in Barcelona (35%), while in Madrid it is almost negligible (2%). Then the measures for on-road traffic control emissions are expected to be more effective in reducing emissions in Madrid than in Barcelona.

On-road traffic emits mainly fine particulate matter. It is responsible for 2.0 t.d^{-1} of $PM_{2.5}$ emissions in Barcelona and 4.2 t d^{-1} in Madrid, accounting for more than 85% of the PM_{10} in both cities (2.2 t d^{-1} and 4.8 t d^{-1}, respectively). The total $PM_{2.5}$ emitted are 2.6 t d^{-1} in Barcelona and 4.6 t d^{-1} in Madrid. $PM_{2.5}$ disaggregation is estimated in base of the chemical mechanism Carbon Bond IV (Gery et al., 1989). Major fractions are elemental carbon –PEC- (47% weight in Barcelona and 58% weight in Madrid) and organic carbon –POA- (35% in Barcelona and 37% in Madrid), which are the main compounds emitted in motor vehicles exhaust (Hildemann et al., 1991; Gillies and Gertler, 2000)

The HERMES traffic module calculations of PM coarse (PM with aerodynamic diameter ranging between 2.5 and 10 µm) emissions do not depend on the fuel type used by vehicles, therefore in the framework of this study this kind of particulate emissions does not change either in quantity or in composition, because the circulation model and the typology of vehicles are invariable among scenarios.

Figure 3.8 depicts the specific weighted change in traffic emissions for each scenario, where the sum of all scenarios (E7) involves the largest relative variation in all compounds emissions. Both for Barcelona and Madrid it reduces $PM_{2.5}$ (from 40% to 46% reduction in combined scenario), SO_2 (from 27% to 35% reduction) and NO_x (from 27% to 35%).

The substitution of the 100% of the urban buses (E1), of the 50% of the intercity buses (E3) and of the 100% of HDV (E6) are negligible in terms of emissions change (Table 3-7 and Table 3-8). None of them involves a fleet change larger than 1.5% neither in Barcelona nor in Madrid. The diesel HDVs implicated are large contributors to SO_2 and PM emissions. Then the NG urban buses reduce mainly SO_2 (4.2% in Barcelona and 3.9% in Madrid with respect to total SO_2 traffic emissions) and $PM_{2.5}$ (3.5% in Barcelona and 3.3% in Madrid), but also NO_x (3.6% in Barcelona and 2.7% in Madrid). The impact of substituting the freight transport vehicles in Barcelona (E6) reduces SO_2 by 4.1%. The relevance of this scenario in Madrid is much lower (1.9%), because the vehicle fleet is mainly formed by cars (50% are petrol cars and 32% diesel cars, Figure 3.2). Regarding E3, changes are more significant in Madrid, due to the larger number of intercity buses, making the SO_2 emissions from traffic reduce up to 4.0%. While in Barcelona the traffic emissions do not decrease over 2.2% for any pollutant.

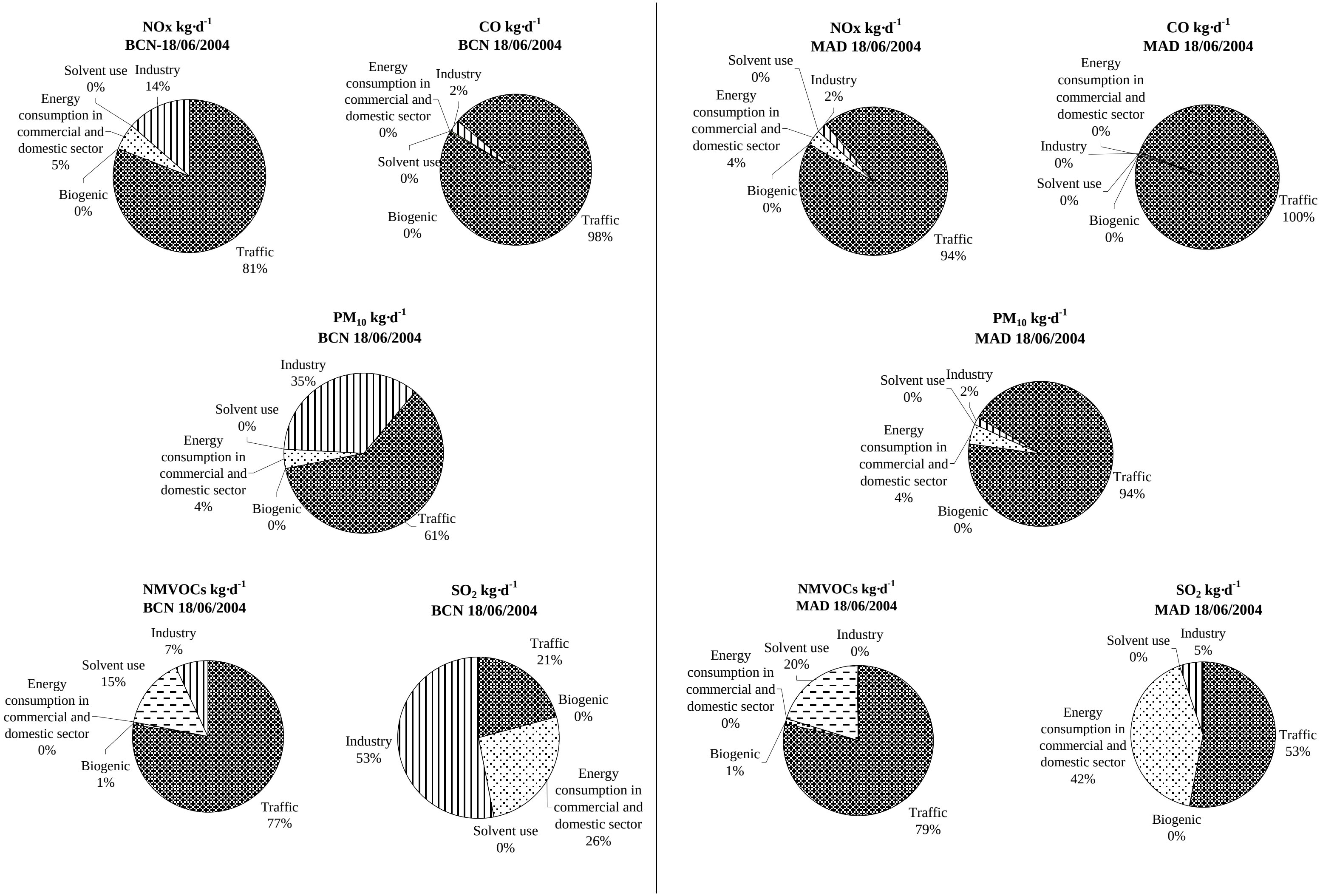
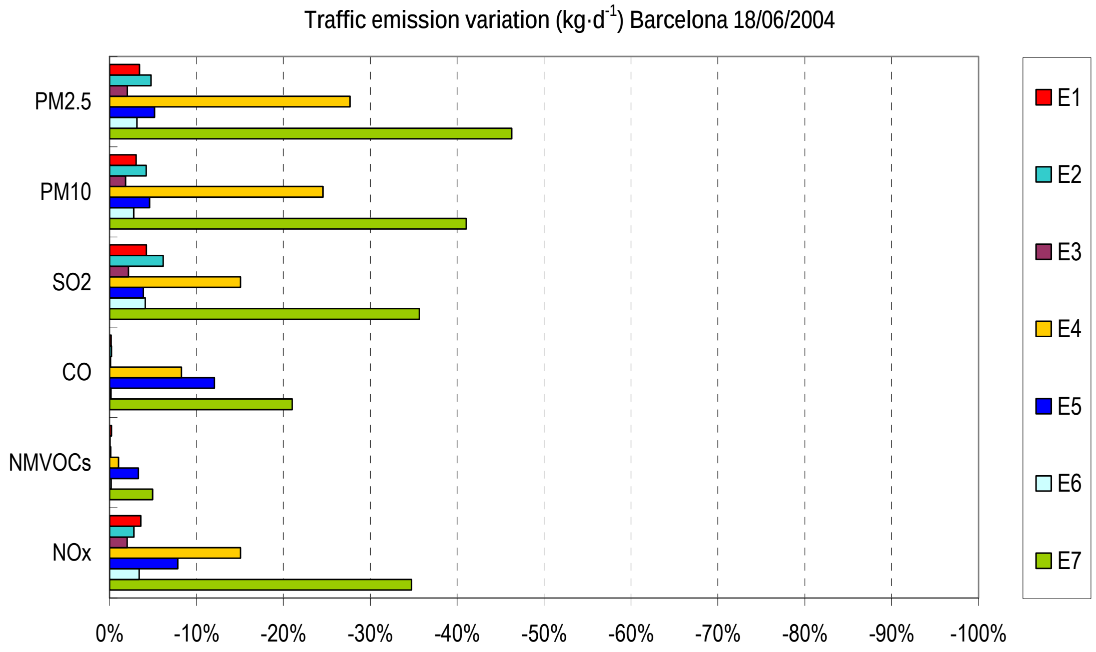


Figure 3.7 Source apportionment for Barcelona greater area emissions (left) and for Madrid greater area emissions (right) on the 18 June, 2004



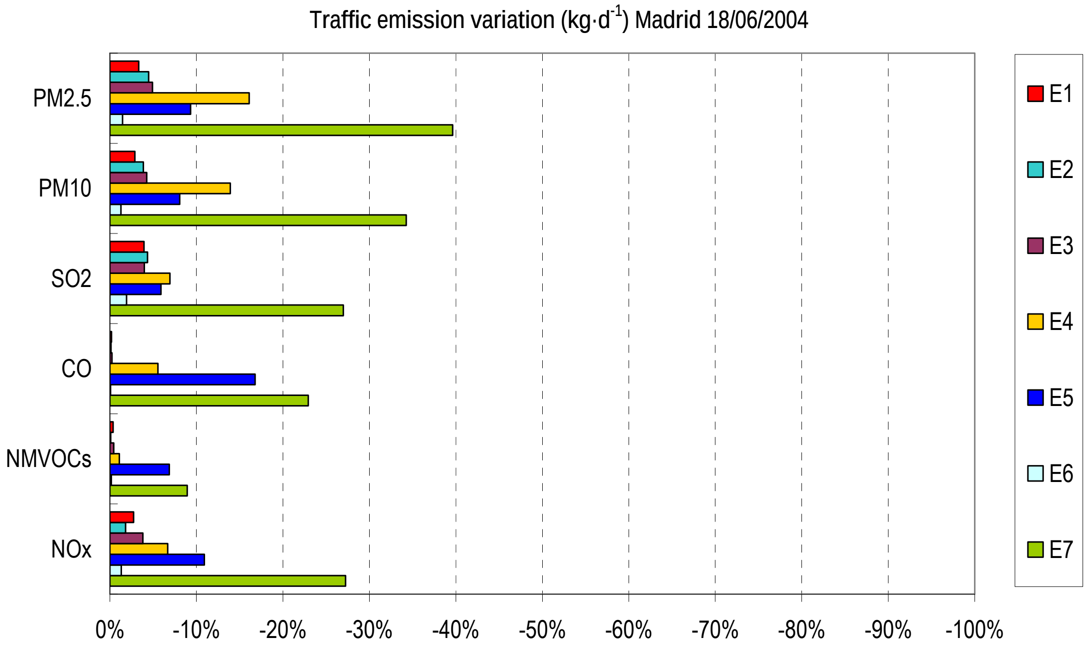


Figure 3.8 Traffic emissions variation in the different scenarios respect to the base case scenario for Barcelona greater area (up) and Madrid greater area (down). Scenarios BC, E1-E7 detailed on section 3.2.2.2.

The most effective individual scenario in reducing NO_x emissions in Barcelona is E4, changing the 50% of light commercial vehicles (reduction in NO_x traffic emissions of 15.1%)-Table 3-7; Table 3-8 and E5 in Madrid, changing 10% of private cars (reduction in NO_x traffic emissions of 10.9%)-Table 3-8-. The vehicle fleet composition is different in both cities (Figure 3.2). Meanwhile the Madrid vehicle fleet is mainly composed by private cars and taxis, the weight of commercial light vehicles fraction is important in Barcelona. This type of vehicles is mainly petrol or diesel LDV (less than 7.5 t) with a large contribution to NO_x, SO_2 and PM emissions, especially the diesel ones (Moussiopoulos, 2003). In Madrid the large number of vehicles involved (both petrol and diesel cars) explains the reduction in emissions.

The substitution of the oldest petrol and diesel private cars (E5) involves the largest reductions in NMVOCs and CO traffic emissions (3.3% and 12.1% reduction in Barcelona and 6.9% and 16.8% reduction in Madrid, respectively (Figure 3.8)).

SO_2 and $PM_{2.5}$ reductions are mainly noticeable when transforming to NG 50% of the commercial light vehicles (E4). In Barcelona area the SO_2 from on-road traffic is reduced by 15.1% and in Madrid by 6.9%. Meanwhile, due to the large contribution of industrial emissions in Barcelona area, if all emission sectors are considered, the reduction in SO_2 emissions accounts for 3.1%. $PM_{2.5}$ from on-road traffic is reduced by 27.7% in Barcelona and by 16.1% in Madrid.

Table 3-7 Emissions reduction from on-road traffic (kg d^{-1}) for each scenario in Barcelona. Further definition of the scenarios on section 3.2.2.2

Emissions from road traffic in Barcelona						
Pollutant (kg d^{-1})	NO_x	NMVOCs	CO	SO_2	PM_{10}	Total
Base case scenario (BC)	23949	72740	116162	736	7356	223244
%	NOx	NMVOCs	CO	SO_2	PM_{10}	
E1-BC	- 3.6%	- 0.2%	- 0.2%	- 4.2%	- 3.1%	
E2-BC	- 2.8%	- 0.1%	- 0.2%	- 6.2%	- 4.2%	
E3-BC	- 2.0%	- 0.1%	- 0.1%	- 2.2%	- 1.8%	
E4-BC	- 15.1%	- 1.0%	- 8.3%	- 15.1%	- 24.5%	
E5-BC	- 7.8%	- 3.3%	- 12.1%	- 3.9%	- 4.6%	
E6-BC	- 3.4%	- 0.2%	- 0.2%	- 4.1%	- 2.8%	
E7-BC	- 34.7%	- 5.0%	- 21.0%	- 35.6%	- 41.0%	

Table 3-8 Emissions reduction from road traffic (kg d^{-1}) for each scenario in Madrid. Further definition of the scenarios on section 3.2.2.2.

Emissions from road traffic in Madrid						
Pollutant (kg d^{-1})	NO_x	NMVOCs	CO	SO_2	PM_{10}	Total
Base case scenario (BC)	66700	95767	297574	1832	18238	485432
%	NO_x	NMVOCs	CO	SO_2	PM_{10}	
E1-BC	-2.7%	-0.4%	-0.2%	-3.9%	-2.9%	
E2-BC	-1.8%	-0.1%	-0.1%	-4.3%	-3.9%	
E3-BC	-3.8%	-0.4%	-0.2%	-4.0%	-4.3%	
E4-BC	-6.7%	-1.1%	-5.5%	-6.9%	-13.9%	
E5-BC	-10.9%	-6.9%	-16.8%	-5.9%	-8.1%	
E6-BC	-1.3%	-0.1%	-0.1%	-1.9%	-1.3%	
E7-BC	-27.3%	-8.9%	-22.9%	-27.0%	-34.3%	

The substitution of 50% of taxis by NG cars (E2) involves a change in vehicle fleet from 2.2% up to 10.1%, depending on the circulation zone and the city, while the transformation of 10% of private cars changes the vehicles circulating from 3.9% up to 7.5%. Differences on the results in emissions variation (larger in E5 than in E2 in all cases), indicate that smaller changes in the vehicle fleet affecting the whole greater areas can be more effective than deeper changes focused on most reduced zones, that is, emission reduction strategies involving just a change in an area of the city may not have the expected result. On the other hand, the fuel substitution estimated for E2 is 3.8% and for E5, 5.8% on average, clearly affecting the emissions differences between scenarios.

3.4 Validation of the base case simulation

Air quality surface station hourly data (provided by the Environmental Departments of the Catalonia and Madrid Governments, Spain) averaged over the domains of study are used to evaluate the performance of WRF-ARW/HERMES/CMAQ predicting ground-level O_3, NO_2, SO_2 and PM_{10} during the episode of 17-18 June, 2004.

The European Directives 1999/30/EC, 2002/3/EC and 2008/50/EC assume an uncertainty of 50%, defined as the maximum error of the measured and calculated concentration levels, for the air quality objective for modelling assessment methods. In addition, the US Environmental Protection Agency (US-EPA) has recently developed new guidelines (US EPA, 2007) that indicate that it is inappropriate to establish a rigid criterion for model acceptance or rejection. However, in the EPA guide for the 1-hour ozone attainment demonstrations (US-EPA, 1991), several statistical goals were identified for operational model performance. Although there is no criterion for a "satisfactory" model performance, US-EPA (1991, 2005) 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 for concentrations above a prescribed threshold (MNGE) to be met by modelling simulations of O_3, to be considered for regulatory applications.

The statistical values obtained as a result of the evaluation (Table 3-9) meet the uncertainty objectives set by the European Directives (e.g. the average MNGE for selected air quality stations is 15% for O_3 predictions, 28% for NO_2 and SO_2 estimations and 26% for PM_{10}). They confirm the need for working with fine grids in areas where the influence of on-road traffic is important; it becomes essential for addressing air quality processes in urban and industrial areas, whereas for rural areas larger grids may be allowed, for example, to capture the non-linearity of the O_3 chemical formation as a function of precursor concentrations (Jang et al., 1995; Jiménez et al., 2005b).

Table 3-9 Summary of the model evaluation for the 17-18 June, 2004 episode. Average statistical parameters for selected AQS in the study areas and during the 48 hr simulated period.

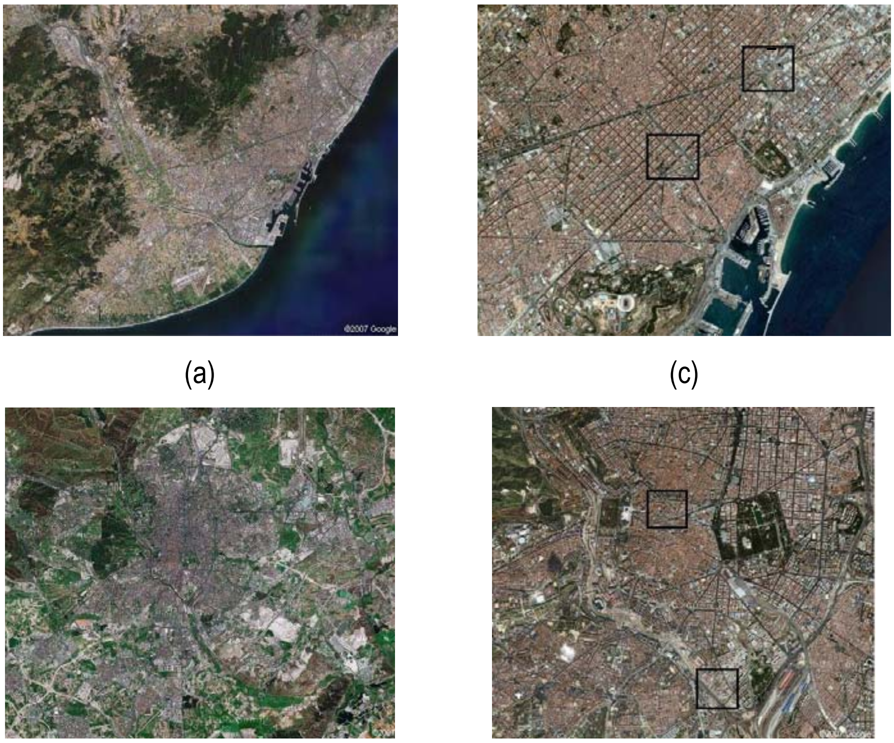
	MNBE (%)	*MNGE (%)*	*UPA (%)*
O_3	-4%	15%	-9%
NO_2	-6%	28%	-4%
SO_2	-12%	28%	-12%
PM_{10}	-3%	26%	-9%

3.5 Quantifying air quality variations

The simulations provide the hourly and average concentrations for all species considered in each cell of both Northeastern and Central Iberian Peninsula domains (see Chapter 2). In order to assess the influence in Barcelona and Madrid, the analysis focuses on these areas, defining three domains in each of them (Figure 3.9): (1) two domains covering the greater areas in order to identify the average behaviours (Barcelona area, Madrid area); (2) two smaller domains (4 km^2) over downtown areas to determine the direct effect on the city dwellers (Barcelona-Downtown, Madrid-Downtown); (3) finally, two domains (4 km^2) corresponding to the areas with the largest traffic density to estimate the largest changes achieved in cities (Barcelona-Gloriès, Madrid-Legazpi).

To compare the numerical results with European air quality targets (see Chapter 1) the maximum hourly concentrations for O_3, NO_2, SO_2 and PM_{10}, the O_3 8-hr average concentration and the daily average NO_2, SO_2 and

PM_{10} concentrations are estimated. Also the variations in maximum and hourly concentration for each domain and scenario are calculated.



(a) (c)

(b) (d)

Figure 3.9. Domains defined to the results analysis: (a) Barcelona area (41º15’N - 41.º36’N; 1º50’E - 2º27’E); (b) Madrid area (40º09’ N - 40º33’ N; 3º54’ W - 3º27’ W); (c) Barcelona-Downtown (41º23’21’’N - 41º24’36’’N; 2º10’05’’ E -2º11’24’’ E) and Barcelona-Gloriès (41º24’12’’N-41º25’12’’N; 2º11’13’’E - 2º12’36’’E); (d) Madrid-Downtown (40º25’04’’ N - 40º26’08’’ N; 3º41’59’’ W-3º40’48’’ W) and Madrid-Legazpi (40º23’28’’N - 40º24’33’’N; 3º41’42’’ W - 3º40’12’’ W)

3.6 Impacts of NGV on urban air quality: comparison among scenarios

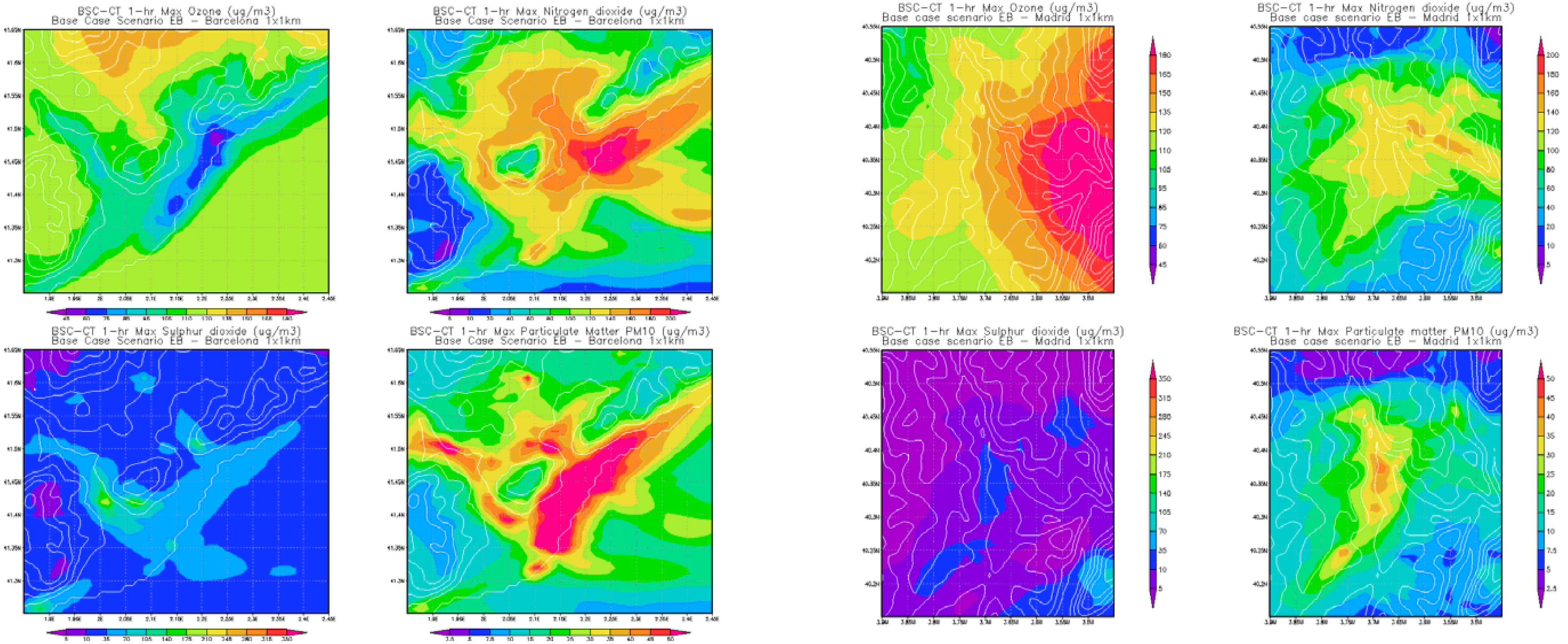
The base case scenario simulation for 17-18 June, 2004 indicates that the NO_2, SO_2 and PM_{10} concentrations are lower in Madrid than in Barcelona. Nevertheless, modelled O_3 values are higher in Madrid. For instance, the O_3 8-hr average concentration is 76.9 µg m^{-3} in Barcelona and 86.0 µg m^{-3} in Madrid area (Table 3-10)

Both in Barcelona and Madrid areas the O_3 concentration is lower in downtown than in downwind areas (Figure 3.10), due to the higher concentrations of NO that acts as an O_3 sink and to the depletion of radicals via HNO_3 formation by NO_2 consumption (Atkinson, 2000). Moreover the mobility data considered in both cities to estimate on-road traffic emissions distinguished four concentric zones in Madrid and five in Barcelona from the city centre. In these zones the circulation of specific vehicle types is enhanced face to others, according to their activity. The gradient of O_3 concentrations over the greater areas suggests that NO_x emissions are larger in the downtown due to a larger volume of traffic and to its specific typology: mainly petrol and diesel cars and light duty commercial vehicles.

The on-road traffic highly influences urban air pollution, resulting i.e. the highest NO_2 concentrations in the base case scenario along the road axis (Figure 3.10). In Barcelona, NO_2 concentrations downtown are especially remarkable, exceeding in some cases the EU-limit for 1-hour average concentration (200 µg m^{-3}). Also PM_{10} and SO_2 show higher concentrations downtown than in the surrounding areas, but the SO_2 concentrations remain low in all cases (1-hr maximum concentration lower than 350 µg m^{-3} – limit for 1-hr average concentration set by the EU-, and 24-hr average concentration lower than 125 µg m^{-3} – limit for 24-hr average concentration). Problems related to air quality in Madrid downtown are mainly associated to NO_2 and PM_{10}, but the concentrations do not exceed the European targets in any case.

The NGV scenarios result in an increase of urban O_3 levels. This variation is larger in the combined scenario, E7 (Figure 3.11), when the changes in the vehicle fleet are more pronounced (up to 26.1% of vehicles substituted in Barcelona and up to 23.1% in Madrid), which has a direct effect on emissions variation, see Table 3-7 and Table 3-8 (achieving reductions i.e. in NO_x traffic derived emissions of -34.7% and -27.3%, respectively). The 8-hr average concentration reaches 77.9 µg m^{-3} in the Barcelona area domain (+1.3%) and 88.1 µg m^{-3} in the Madrid area (+2.5%). This effect can be locally more important, i.e. for the Barcelona Downtown area increases of 7.8% are registered (79.3 µg m^{-3} (+7.8%)) and the highest concentration is estimated for Madrid-Legazpi: 147.0 µg m^{-3} (+2.4%).

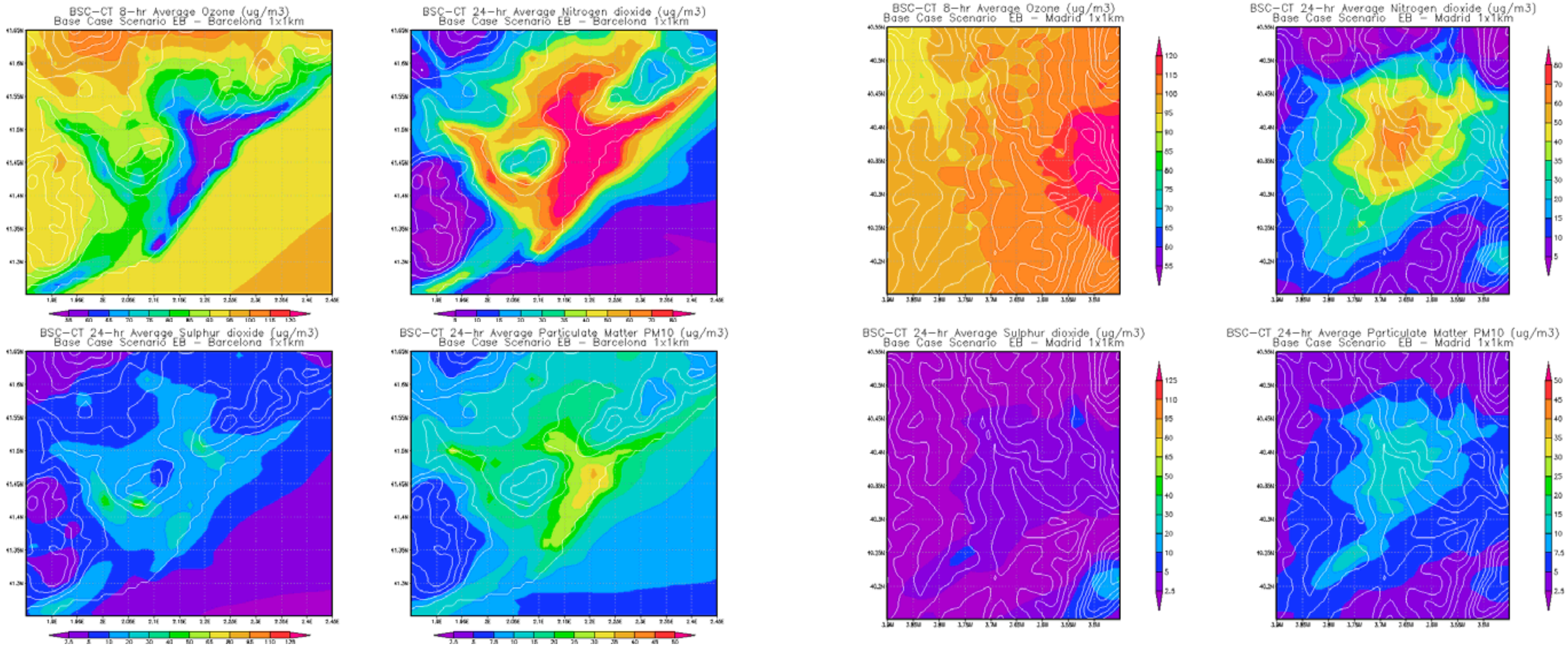
The introduction of NGV promotes the reduction of the NO_x concentration in cities, making O_3 concentration rise in almost all cases (Table 3-10), this behaviour is characteristic of a VOC-sensitive area, usually produced in conditions with relatively low VOCs and high NO_x (Jiménez and Baldasano, 2004; Sillman and He, 2002). Similar studies (Reis et al., 2000; Guariso et al., 2004) have reported analogous results locally in VOC-controlled areas when reducing NO_x emissions. In the case of Madrid the results suggest that it could be under a transition sensitivity regime, depending on the ratio of NO_x/VOCs emissions reductions in each scenario the response of the O_3 concentration presents a different trend. For example when transforming the 10% of the private cars to NG (E5) the O_3 maximum hourly concentration in Madrid-Downtown and Madrid-Legazpi domains slightly decreases (-1.0% and -2.1%).



(a)

(b)

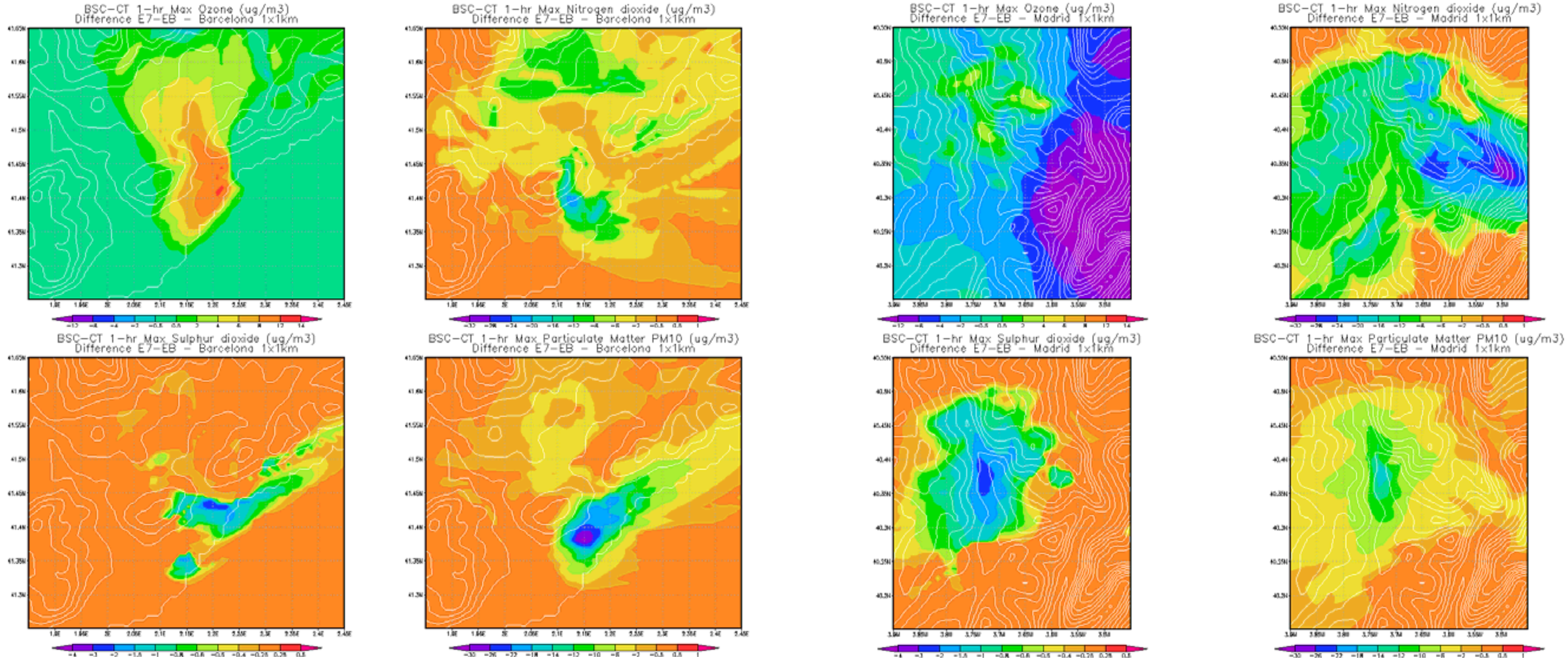
Figure 3.10. Maximum hourly concentration of O_3, NO_2, SO_2 and PM_{10} for the base case scenario in the Barcelona area (a) and in the Madrid area (b); and 8-hr O_3 average concentration and 24-hr NO_2, SO_2 and PM_{10} average concentration for the base case scenario in the Barcelona area (c) and the Madrid area (d) during the episode (17-18 June, 2004).



(c)

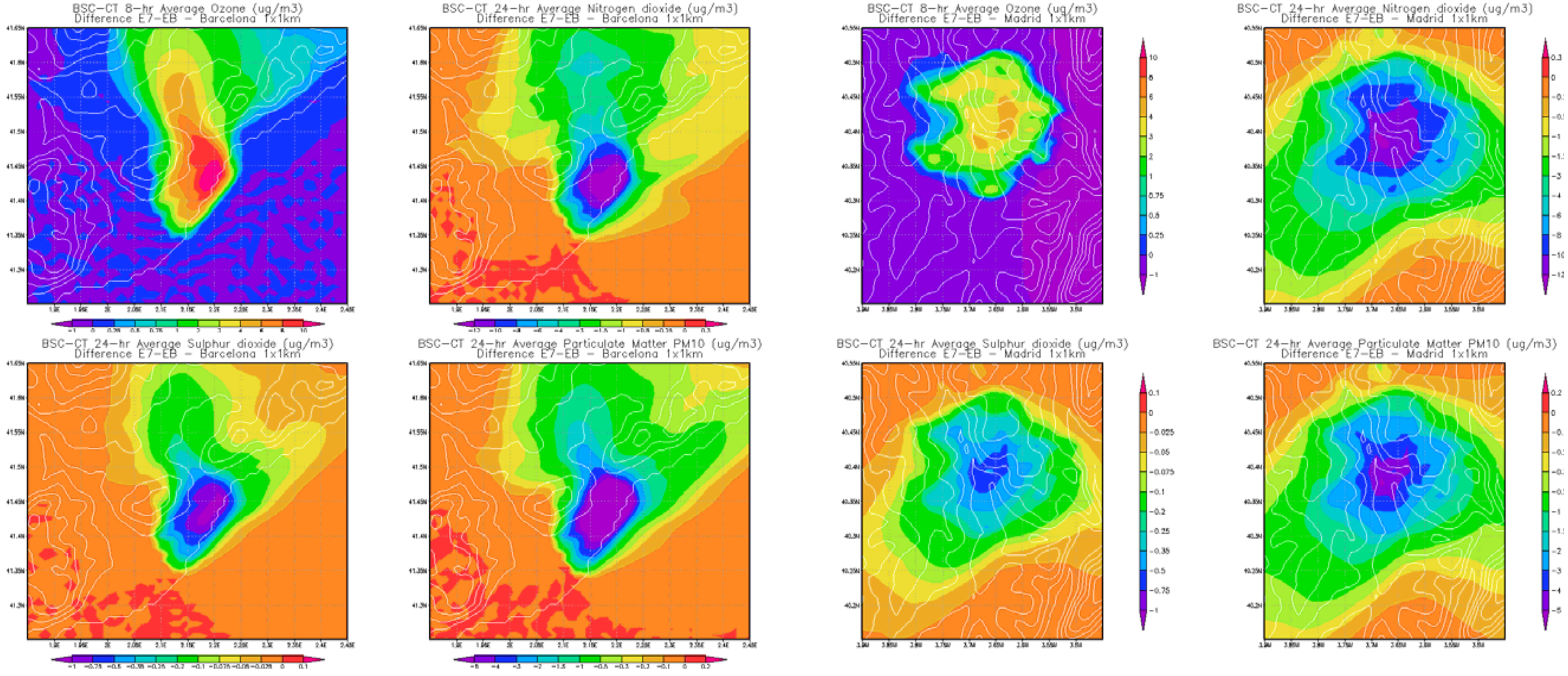
(d)

Figure 3.10. Continued.



(a) (b)

Figure 3.11. Difference in maximum hourly concentration of O_3, NO_2, SO_2 and PM_{10} in Barcelona area (a) and Madrid area (b); and difference in 8-hr O_3 average concentration, and 24-hr NO_2, SO_2 and PM_{10} average concentration in Barcelona area (c) and Madrid area (d) between the combined scenario and the base case scenario (µg m^{-3}) during the episode (17-18 June, 2004).



(c) (d)

Figure 3.11. Continued.

The O_3 urban peaks increase with decreasing NO_x emissions for Barcelona area (Figure 3.12), except when introducing 50% of NGV as commercial light vehicles (E4). In this case a slight reduction in the maximum concentration is observed (around 0.5%), which does not affect the trend in 8-hr average concentration (Table 3-10). On the other hand, for the Madrid area, a slight reduction on NO_x emissions (up to 0.04 kmol d^{-1}, Figure 3.12) involves higher O_3 peaks, but for largest changes on NO_x emissions, the O_3 peaks locally decrease, being up to 3.7% lower in case of changing 10% of private cars by NGV (E5) or up to 5.4% in case of introducing the combined scenario (E7). Moreover a decrease of O_3 8-hr average concentrations when introducing in E5 and E7 occurs in downwind areas of the Central Iberian Peninsula domain.

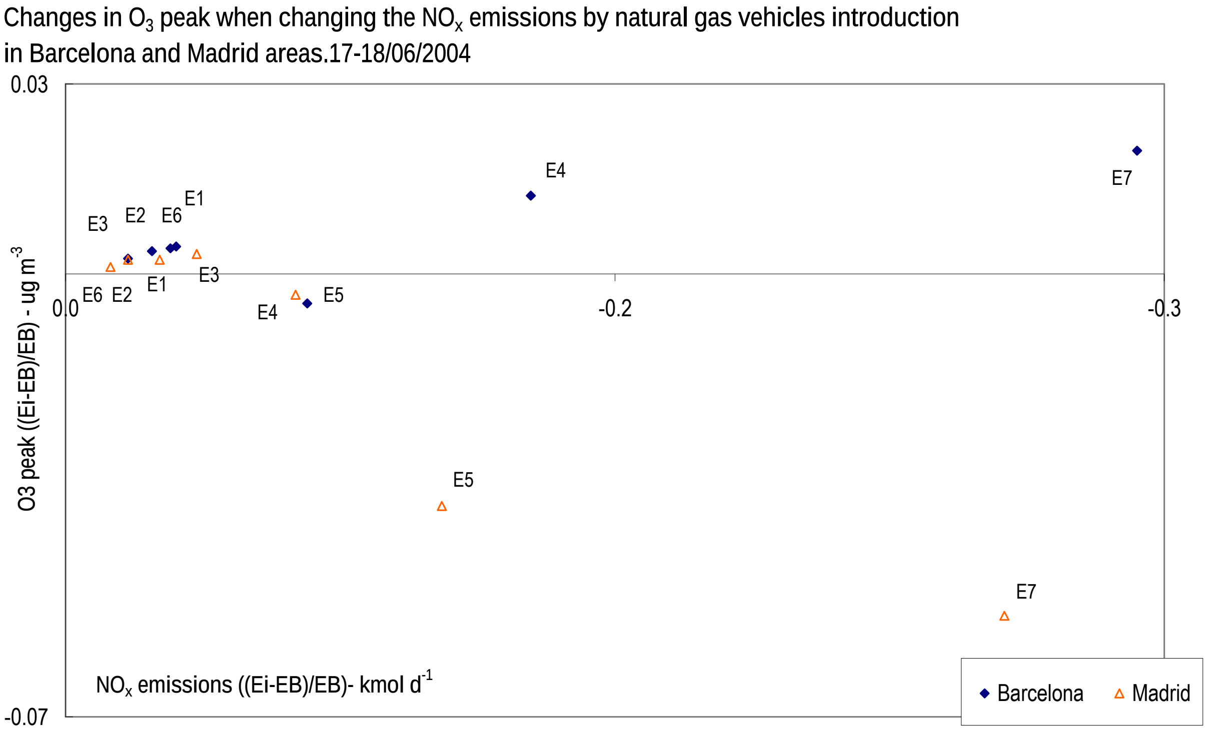


Figure 3.12. Changes in O_3 peak concentration in the Barcelona and Madrid urban areas when changing the NO_x emissions by NGV introduction. EB: Base case scenario, Ei: i scenario, with i=1…7.

The effects on air quality of introducing NGV are positive in both urban areas in terms of NO_2, SO_2 and PM_{10} concentrations, reducing both average and maximum hourly values in all scenarios tested. The E7 involves the largest reductions in all cases (Figure 3.11). The NO_2 24-hr average concentration in Barcelona reduces up to 6.1% (32.9 µg m^{-3}, Table 3-10) from the base case scenario (35.0 µg m^{-3}). In Madrid the same scenario involves a reduction of 20.6% (17.7 µg m^{-3}; 22.2 µg m^{-3} in the base case; Table 3-10). The largest change is observed in Madrid-Legazpi domain where NO_2 24-hr average concentration decreases by 23.2%, from 63.6 µg m^{-3} to 48.8 µg m^{-3}.

Table 3-10. 8-hr O_3 and 24-hr NO_2 average concentration in Barcelona and Madrid areas. Variation among scenarios.

	Barcelona area			Madrid area		
	O_3 8-hr average concentration			O_3 8-hr average concentration		
	Conc (µg m^{-3})	ΔConc (µg m^{-3})	Variation (%)	Conc (µg m^{-3})	ΔConc (µg m^{-3})	Variation (%)
Base case	76.9	-	-	86.0	-	-
Scenario 1	77.0	0.09	0.12%	86.2	0.21	0.24%
Scenario 2	77.0	0.07	0.09%	86.1	0.13	0.15%
Scenario 3	77.0	0.05	0.07%	86.3	0.28	0.33%
Scenario 4	77.3	0.40	0.52%	86.5	0.52	0.60%
Scenario 5	77.1	0.19	0.25%	86.8	0.83	0.97%
Scenario 6	77.0	0.09	0.11%	86.1	0.10	0.12%
Scenario 7	77.9	0.97	1.26%	88.1	2.12	2.47%
	NO_2 24-hr average concentration			NO_2 24-hr average concentration		
	Conc (µg m^{-3})	ΔConc (µg m^{-3})	Variation (%)	Conc (µg m^{-3})	ΔConc (µg m^{-3})	Variation (%)
Base case	35.0	-	-	22.2	-	-
Scenario 1	34.8	-0.20	-0.56%	21.8	-0.44	-1.98%
Scenario 2	34.9	-0.15	-0.42%	22.0	-0.28	-1.24%
Scenario 3	34.9	-0.11	-0.32%	21.6	-0.61	-2.73%
Scenario 4	34.1	-0.89	-2.54%	21.1	-1.12	-5.04%
Scenario 5	34.6	-0.46	-1.30%	20.4	-1.82	-8.20%
Scenario 6	34.8	-0.19	-0.55%	22.0	-0.21	-0.95%
Scenario 7	32.9	-2.15	-6.13%	17.7	-4.58	-20.59%

The PM_{10} average reduction over Barcelona is 6.6% (9.7 µg m^{-3}; 10.4 µg m^{-3} in the base case; Table 3-11) and over Madrid is 14.9% (4.2 µg m^{-3}; 4.9 µg m^{-3} in the base case; Table 3-11 but the introduction of E7 affects particularly the maximum hourly concentration in Madrid-Downtown, where a reduction of 42.8% is estimated (from 33.7 μg m^{-3} to 19.3 μg m^{-3}).

The effects in SO_2 concentrations are more noticeable in Madrid than in Barcelona (up to 8.7% reduction in E7 for the 24-hr average concentration, Table 3-11) reflecting the different composition of the emission sources between cities. While Madrid is characterized by a commercial and tertiary industrial activity in Barcelona a heavier industrial component is present, that has a direct effect on the weight of traffic contribution to SO_2 emissions (estimated in a 53% of total SO_2 in mass for Madrid and 21% for Barcelona) and indirectly involves that a traffic emissions abatement strategy could be more effective in reducing SO_2 concentrations in Madrid than in Barcelona.

Introducing a 50% of NG commercial light vehicles is the most effective scenario in reducing NO_2 in Barcelona (-2.5% on 24-hr average concentration; Table 3-10), nevertheless in Madrid better results are achieved when changing the 10% of the private cars fleet (-8.2% on 24-hr average concentration; Table 3-10). The vehicle fleet of Madrid city is mainly composed of diesel and petrol private cars and taxis (82% of the total number of vehicles face to a 66% in Barcelona), which determines the important effect on the air quality of this scenario, according also with the estimated emissions reductions (15% reduction in NO_x traffic emissions in Barcelona and 10% reduction in Madrid, Table 3-7-Table 3-8).

In order to reduce SO_2 and PM_{10} concentrations, the most effective measure involves the substitution of the oldest diesel and petrol light commercial vehicles, which are important contributors to the emission of these species. The major changes on air quality are registered both in the downtown and in the outskirts with an important industrial activity, which steps up this kind of vehicles circulation.

The substitution of the whole urban bus fleet in both cities, the 50% of the intercity buses or the 100% of the heavy duty freight transport vehicles by NGV is negligible in terms of air quality (<3% variation in the studied ground level of pollutants in almost all cases) due to the relatively low number of vehicles involved (<1.5% of the total vehicle fleet in cities) and the low fuel substitution associated, from 1.4 to 3.2% of conventional fuels changed by NG. The diesel heavy duty vehicles substituted are usually large contributors to PM_{10} and SO_2 emissions, so that the largest effects are noticed over these pollutants concentrations (e.g. the introduction of 50% of NG intercity buses involves by -6.1% change in PM_{10} maximum hourly concentration in Madrid-Downtown).

The variations in pollutants concentrations when transforming the 50% of the taxis to NG cars are not remarkable; however they are more important in Madrid than in Barcelona area. All taxis substituted are considered as diesel cars, so that the effects are mainly noticed in terms of SO_2 and PM_{10} concentrations, e.g. in Madrid-Downtown the SO_2 maximum hourly concentration reduces by 4.6% (from 10.1 μg m^{-3} in the base case to 9.6 μg m^{-3}) and PM_{10} maximum hourly concentration 6.1% (from 33.7 μg m^{-3} to 31.7 μg m^{-3}).

Table 3-11. 24-hr SO_2 and PM_{10} average concentration in Barcelona and Madrid areas. Variation among scenarios.

	Barcelona area			Madrid area		
	SO_2 24-hr average concentration			SO_2 24-hr average concentration		
	Conc (µg m^{-3})	ΔConc (µg m^{-3})	Variation (%)	Conc (µg m^{-3})	ΔConc (µg m^{-3})	Variation (%)
Base case	7.7	-	-	1.5	-	-
Scenario 1	7.7	-0.01	-0.18%	1.5	-0.02	-1.28%
Scenario 2	7.7	-0.02	-0.25%	1.5	-0.02	-1.30%
Scenario 3	7.7	-0.01	-0.09%	1.5	-0.02	-1.29%
Scenario 4	7.7	-0.05	-0.65%	1.4	-0.03	-2.28%
Scenario 5	7.7	-0.01	-0.17%	1.4	-0.03	-1.88%
Scenario 6	7.7	-0.01	-0.18%	1.5	-0.01	-0.63%
Scenario 7	7.6	-0.12	-1.52%	1.3	-0.13	-8.67%
	PM_{10} 24-hr average concentration			PM_{10} 24-hr average concentration		
	Conc (µg m^{-3})	ΔConc (µg m^{-3})	Variation (%)	Conc (µg m^{-3})	ΔConc (µg m^{-3})	Variation (%)
Base case	10.4	-	-	4.9	-	-
Scenario 1	10.4	-0.05	-0.48%	4.9	-0.06	-1.21%
Scenario 2	10.4	-0.07	-0.66%	4.8	-0.08	-1.64%
Scenario 3	10.4	-0.03	-0.29%	4.8	-0.09	-1.79%
Scenario 4	10.0	-0.42	-3.99%	4.6	-0.31	-6.24%
Scenario 5	10.4	-0.08	-0.74%	4.7	-0.17	-3.49%
Scenario 6	10.4	-0.05	-0.44%	4.9	-0.03	-0.54%
Scenario 7	9.7	-0.69	-6.60%	4.2	-0.73	-14.92%

3.7 Conclusions

Nowadays, testing urban air quality management strategies and evaluating them in terms of emissions variation is a major concern. These plans are mainly focused on reducing on-road traffic emissions. In fact at present European large cities are trying out different strategies, among others the introduction of alternative fuels like natural gas.

Several realistic vehicle fleet variation scenarios of implementation of NGV are studied in the two main cities of Spain (Barcelona and Madrid greater areas) by using the HERMES emission model. Feasible changes on vehicle fleets have been introduced in the different scenarios: urban and intercity buses, taxis, light commercial and heavy duty freight transport vehicles and private cars.

Both cities, Madrid and Barcelona, differ in size, number of inhabitants and economic activities shape; and therefore in the contribution of activity sectors to atmospheric emissions. Madrid shows a larger contribution of on-road transport to emissions than Barcelona (93% versus 86% when primary pollutants are considered for a polluted summertime episode of the year 2004), because the number of vehicles circulating in the former is almost twice as large, and Barcelona area has a heavier industrial activity contributing to the emission of pollutants.

The changes in on-road transport emissions mainly depend on the specific vehicle fleets involved and the vehicle fleet composition of the study areas. The largest variations occur for the combined scenario tested (E7), when up to 26% of the vehicle fleet is transformed in Barcelona and up to 23% in Madrid. Ozone precursors and primary pollutants emissions decrease up to 38.4 t d^{-1} in Barcelona and 98.8 t d^{-1} in Madrid. NGV are useful to reduce SO_2 and PM emissions, especially when substituting old commercial LDV.

The decrease estimated in PM is mainly due to the reduction in the fine fraction emissions (lower than 2.5 µm), composed fundamentally by elemental and organic carbon. This fraction decreases by 27.7% in Barcelona and 16.1% in Madrid when changing 50% of light commercial vehicles. The origin of PM coarser fraction from road traffic is erosion or wearing processes, so it remains unaffected in all the considered scenarios.

The reduction of NO_x emissions from on-road transport in Barcelona should manage the substitution of the oldest commercial light vehicles (E4), while in Madrid the transformation of 10% of the oldest private cars to natural gas is more effective. This fact is attributed to the different vehicle fleet composition, characterised by a larger percentage of petrol and diesel cars in Madrid than in Barcelona.

Nevertheless, even considering the introduction of NGV accomplishing the EEV standards, the conventional fuel substitution has to reach certain critical values (around 4%) for being effective in the reduction of emissions. Collateral impacts of using NGV such as the construction of facilities for gaseous fuel supply and the possible increase in the GHG outcome by the methane emissions increase have to be considered by the policy makers.

The base case simulation results indicate that the air quality improvement strategies in Barcelona and Madrid areas must be addressed mainly to reduce NO_2 and PM_{10} ground levels, which are larger in downtown areas due to the pervasive traffic emissions. O_3 levels could generate air quality problems, involving exceedances of the European targets in downwind areas, but not in the conurbations. Despite the SO_2 concentrations are higher in downtown areas than outskirts, they do not involve problems related to human health.

As shown, the largest reductions are achieved in the combined scenario and the individual scenarios proving to be more effective in reducing NO_2, SO_2 and PM_{10} concentrations are the transformation of 50% of commercial light vehicles (involving approximately the 10% of current fuel substitution by NG of the EU proposal to 2020) and the 10% of private cars (around the 5% of current fuel substitution by NG proposed to 2010 by the EU). The latter is especially remarkable when considering NO_2 reductions in Madrid area.

The efficacy of the tested measures depends not only on the number and category of the vehicles substituted, but also on the specific characteristics of the conurbation. Madrid is a more favourable scenario to introduce traffic management strategies than Barcelona, where other emission sources must be controlled in addition, especially when referring to the decrease of PM_{10} and SO_2 levels. On the other hand, except for O_3, the levels of pollutants are higher in Barcelona, so that it becomes essential to introduce air quality management strategies in this area.

The simulation system WRF-ARW/HERMES/CMAQ proves to be a suitable tool for the management and assessment of urban air quality especially when applied with high spatial and temporal resolution. Specifically, the HERMES emission model permits to assess emission variation scenarios based in real changes of application and it provides the base to design scenarios by changing the vehicle fleet composition in urban areas of Spain. The available information about emission factors for new technology vehicles or alternative fuels is sparse. Efforts may be done in a future in order to enlarge current databases and provide speed dependant emission factors. These would necessarily have to reflect the changes in vehicles fleet composition that are already taking place, specially in urban areas. The improvement of air quality conditions in main cities of Spain makes the evaluation of environmental abatement strategies crucial. Currently some of these strategies are focused on traffic emissions reduction; and the introduction of alternative fuels like NG seems to be a path to reduce primary pollutants emissions on large urban areas and ameliorate air quality specially in terms of SO_2 and PM_{10}.

4 Urban air quality variations by biodiesel use. Barcelona and Madrid cases.

4.1 Introduction

The effect of on-road traffic emissions on air quality and greenhouse gases outcome, the security for energy supply and the rising prices of oil and their derivatives are the main reasons for developing and developed countries to bet for the biofuels use. Specifically the European Union is promoting the use of biofuels. It pretends to achieve by 5.75% of the on-road transport energy consumption coming from biofuels in 2010, and 10% in 2020 (COM(2001)547; COM(2006)845; COM(2008)19). Their main advantage is that they have the potential to leapfrog a number of traditional barriers to entry faced by other alternative fuels because they are liquid fuels, largely compatible with current vehicles and blendable with current fuels (IEA, 2004).

Nowadays, liquid biofuels are commonly blended with fossil fuels, being the most used the ethanol - gasoline and the fatty acid methyl esters (FAME) - diesel fossil blends.

In most countries, the efforts to introduce ethanol into the transport fuel market has focused on low-percentage blends, such as ethanol E10, 10% ethanol to 90% gasoline volumetric blend (sometimes known as “gasohol”). The main reason is that nearly all recent-model fossil gasoline vehicles are fully compatible with them. These kind of blends are being currently used in the United States, Canada, Australia, Brazil and many European countries.

On the other hand, FAME is generally accepted to be fully blendable with conventional diesel, except for certain considerations when using high-percentage biodiesel blends or neat (pure) biodiesel. Another type of biodiesel, synthetic diesel fuel produced from biomass gasification and Fischer-Tropsch synthesis, is even closer in composition to conventional diesel fuel and blendability is a non-issue. Therefore biodiesel can be easily used in existing diesel engines in its pure form (B100) or in virtually any blend ratio with conventional diesel fuels. Germany, Austria and Sweden have promoted the use of 100% pure biodiesel in trucks with only minor fuel system modifications. In France, biodiesel is often blended at 5% in standard diesel fuel and at 30% in some fleet applications. In Italy, it is commonly blended at 5% in standard diesel fuel. Lower-level (20% or less) biodiesel blends can be used as a direct substitute for diesel fuel in virtually all heavy-duty diesel vehicles without any adjustment to the engine or fuel system (EC, 1998; Lindhjem and Pollack., 2003). Therefore the 80% diesel fossil – 20% biodiesel blend (B20) is accepted to be fully compatible with current diesel engines and constitutes one of the most used biodiesel-diesel blends. For these reasons the biodiesel B20 blend is selected to carry out this study. The availability of data concerning changes on primary pollutants emissions caused by the biofuel introduction also determined this decision. The available data for bioethanol use are sparse or incomplete for the objectives of this work, and mainly is addressed to the GHG emissions outcome during the life cycle of the fuel (e.g. Lechón et al., 2005).

The WRF-ARW/HERMES/CMAQ model is used to assess the effects of the use of biodiesel B20 as a fuel in Barcelona and Madrid vehicle fleets. Two different penetration scenarios were defined: (B1) the 10% of the oldest petrol and diesel cars are substituted by new Euro III biodiesel (B20) fuelled cars; and (B2) the B20 biodiesel blend is used in all the diesel vehicle fleets of Barcelona and Madrid, without fleet renewal. Their effects are analyzed for the 17-18 June, 2004 episode in the Northeastern (NEIP) and Central Iberian Peninsula (CIP) domains, which are centred in the Barcelona and Madrid urban areas.

4.2 Methods

The studied days: 17-18 June, 2004, and the reasons for their selection are described in detail in *section 2.2.4* of *Chapter 2* and *Annex 1* of the present document.. A deep description of the WRF-ARW/HERMES/CMAQ modelling system used and the domains selected is provided in the *section 2.2.1* of *Chapter 2*. It has to be noted that the HERMES model was updated during the execution of this work. From now on ports and airports emissions are included in simulations. *Section 4.3.3* provides a brief description of the air quality modelling evaluation against air quality stations data when including these changes. The particularities concerning the on-road traffic emissions module can be found in the *section 3.2.1* of *Chapter 3*. Here the changes performed to implement the new scenarios are shown.

The scenarios design in the HERMES on-road traffic module requires introducing new emission factors for each new included vehicle category. The B1 scenario includes Euro III B20 fuelled cars and the B2 scenario includes B20 fuelled cars, light duty and heavy duty vehicles from pre-ECE and conventional to Euro III emission standards.

4.2.1 Emission factors for biodiesel blends

The emissions change of a fossil diesel vehicle when fuelled with biodiesel depends on several factors:

(1) the biodiesel percentage in the blend, being the most common those blends with 20% vol. of biodiesel (B20) or the pure biodiesel (B100);

(2) the characteristics and composition of the fossil diesel included in the blend (e.g. the sulphur concentration of the fossil fuel would affect the SO_2 emissions of the blend);

(3) the engine technology and depuration systems;

4) the vehicle category (weight, age, cubic capacity);

(5) the origin of the biodiesel: soybean, rapeseed, or animal fats etc.

In spite of the variability introduced by these factors, some common trends are found (Booz-Allen; 1994; Sharp, 1994; Schumacher et al., 1995; Sharp, 1998; Shennan et al., 1998, Lindhjem and Pollack, 2002; Knothe 2006; NBB, 2006; Ropkins et al., 2007; Samaras and Zierock, 2007). Most of the peer reviewed studies focus on heavy duty vehicles, both urban buses and trucks. They agree in indicating particulate matter, CO and VOCs reductions, around 5%, 9% and 12%, respectively, when introducing B20 instead of pure diesel fossil. Concerning NO_x emissions some discrepancies are found, although most studies indicate an increase of these emissions when introducing biodiesel. The average increase estimated from the literature is around 4% (Table 4-1). The variability of the conditions in the tests, types of vehicles and composition of the base fuels is in the origin of the dispersion of the sampled data, the standard deviations are often larger than the average values. This is also reflected in the B100 compiled emissions, the average PM emissions are around 57% lower, the CO around 36% and the VOCs around 39% respect to the same fossil diesel vehicle, but the standard deviation for CO emissions data are 40% of the average value estimated. The effect on NO_x emissions would be a 10% increase (Table 4-1).

Table 4-1 Summary of emissions variation (%) for HDV when using biodiesel B20 or B100 as a fuel respect to the diesel fossil use. Average values and standard deviations. The characteristics of vehicles and references are specified below.

B20 (%)													Av.	StDev
NO_x	1.8	3.5	2.4	2.0	2.0	2.0	-0.3	9.0	-1.0	11.3	24.3	-12.8	3.7	8.7
PM	-13.6	-15.0	-8.9	-10.1	-12.0	-10.1	9.4	-33.2	11.9	-29.0	-14.6	61.9	-5.3	24.7
CO	-9.2	-9.0	-13.1	-11.0	-12.0	-21.1	10.6	-11.2	22.8	-17.3	-21.5	-11.1	-8.6	12.8
VOCs	-7.3	-15.0	-17.9	-21.1	-20.1	-11.0	-0.7	16.3	-3.5	-31.5	-32.3	-4.7	-12.4	13.7
B100 (%)													Av.	StDev
NO_x	8.9	9.0	13.5			10.0							10.4	2.2
PM	-68.1	-47.0	-55.3			-48.0							-54.6	9.7
CO	-46.2	-20.0	-42.7			-77.0							-46.5	23.4
VOCs	-36.7	-17.0	-63.2			-48.0							-41.2	19.5
	(1)	*(2)*	*(3)*	*(4)*	*(5)*	*(6)*	*(7a)*	*(7b)*	*(7c)*	*(8a)*	*(8b)*	*(8c)*		

(1) Shenann et al., 1998. 4 stroke engine. Urban bus. Biodiesel from soybean oil

(2) EEA-EMEP/CORINAIR, 2007. HDV

(3) Lindhjem and Pollack, 2002. Diesel fossil with S content < 500 ppm

(4) EPA, 2002. HDV

(5) NBB,2006. HDV

(6) Knothe et al., 2006 - 6 cylinder 14 L HD diesel engine equipped with ERC-exhaust gas recirculation 2003; Soybean based biodiesel combined with an average diesel fossil

(7) Howes and Rideout, 1995a. Urban transit bus powered by a 1988 DDE CII 6V92TA engine. Soybean based biodiesel combined with a low sulphur diesel fossil.
   a. Central business district emissions cycle
   b. Arterial emissions cycle
   c. NY City bus composite emissions cycle

(8) Howes and Rideout, 1995b. Urban transit bus powered by a 1981 DDC 8V71 engine. Soybean based biodiesel combined with a low sulphur diesel fossil.

The EEA-EMEP/CORINAIR Emissions Inventory Guidebook (Samaras and Zierock, 2007) provides emission change for three types of vehicles: cars, light duty vehicles and heavy duty vehicles, both for B20 and B100 blends. Their reported data are based on literature review and correspond mainly to rapeseed methyl esters biodiesel. These were found to be the most suitable emission factors for the objectives of this study. Mainly because the EEA-EMEP/CORINAIR methodology takes into account the European context, both in the vehicles typology (Euro classification) and the biodiesel origin: rapeseed based biodiesel predominates in Europe (BRAC, 2006). Additionally it provides the emissions change for several kind of vehicles and a wide range of pollutants (Table 4-2).

Table 4-2 shows the differences in emissions caused by different fuel blends on fossil diesel corresponding to a Euro III vehicle or engine technology. The effect of biodiesel on other technologies may vary but the extent of the variation is difficult to estimate in the absence of detailed literature data. With regard to NO_x, CO_2 and CO, any effect of technology should be negligible, given the marginal effect of biodiesel on these pollutants in general. The effect of biodiesel on PM for different technologies is more difficult to assess. For older diesel technologies with no advanced combustion concepts and aftertreatment systems, biodiesel may lead to a higher reduction than the one shown for Euro III. For more recent technologies, with ultra high pressure combustion and aftertreatment, the biodiesel effect is difficult to predict. (Samaras and Zierock, 2007).

Table 4-2 Biodiesel blend effect in the diesel vehicles. Values for Euro III standard (EEA-EMEP/CORINAIR methodology, Samaras and Zierock, 2007, except for SO_2)

Pollutant	*Vehicle type (Euro III)*	*B20 (%)*	*B100 (%)*
CO_2	Car	-2.0	
	LDV	-1.5	
	HDV	0	0.1
SO_2	Car, LDV, HDV	-20	-100
NO_x	Car	1	
	LDV	2	
	HDV	3.5	9
PM	Car, LDV	-20	
	HDV	-15	-47
CO	Car	-5	
	LDV	-6	
	HDV	-9	-20
NMVOCs	Car	-10	
	LDV	-15	
	HDV	-15	-17

In the framework of this study the values shown in Table 4-2 are used for technologies up to Euro III, but not newer because the vehicle fleet composition corresponds to 2004 and the newest category implemented in

HERMES is Euro III. The assumption of the same reductions for vehicles older than Euro III involves probably an overestimation of PM emissions, this will be taken into account in the results discussion. The CH_4-NMVOCs ratio was considered equal to those of the fossil diesel, and the NH_3 and N_2O emissions were considered equivalent to those of diesel fossil because of the lack of specific data. The SO_2 emissions are supposed to be reduced proportionally to the biodiesel content in the blend because the biodiesel does not content sulphur. Some studies indicate an increase in the fuel consumption when introducing biodiesel in the blend, due to the discrepancies in the values found the same fuel consumption as for the diesel fossil vehicles is considered. The emissions variations are considered to be constant in the whole speed range.

4.3 Results and discussion

4.3.1 Fuel consumption

The European 2010 proposal for the biofuels use was 5.75% in the transport sector (COM(2001)547). This goal was reviewed in 2006, and assumed as a very optimistic objective. In case that all member countries reach their objectives for 2010 the biofuels consumption in Europe will be 5.45% (COM(2006)845). The conclusions of this revision indicate that the European Commission still bets for the biofuels use, and the 10% goal for 2020 is maintained, indicating that incentives for biofuels use have to be set.

The B1 biodiesel scenario designed involves the substitution of 0.9% and 1.2% of fossil fuels in Barcelona and Madrid, respectively (Table 4-3). The fuel substituted in the B2 scenario accounts for 10.5% and 9.9% in Barcelona and Madrid, respectively.

The substitution of the B1 scenario remains below the national and European goals for the biofuels introduction. It changes the oldest gasoline and diesel vehicles by new Euro III cars that are fuelled with B20. This scenario reduces the overall fuel consumption by means of a vehicle fleet renewal (by 0.5% in Barcelona and 0.7% in Madrid). These particularities will be taken into account in the results discussion. The change of some gasoline vehicles by newer biodiesel ones has direct implications in SO_2 emissions, because of the largest sulphur content of the diesel fossil respect to gasoline.

The B2 scenario agrees with the European Union proposals for 2020, that substitutes all the diesel fossil consumption in the urban areas by B20, without vehicle fleet renewal. As already commented the biofuel consumption was considered invariable respect to the diesel fossil, therefore the global fuel consumption remains in the same values, 1,2 t d^{-1} in Barcelona and 3.6 t d^{-1} in Madrid.

Table 4-3. Fuel consumption in the base case, the B1 and the B2 scenarios in the Northeastern Iberian Peninsula, the Central Iberian Peninsula, Barcelona and Madrid areas as estimated by HERMES for the 17-18 June, 2004. Percentage of pure biodiesel used based on total fuel consumption.

Fuel consumption	NEIP					BCN				
t d^{-1}	Fossil fuel	Petrol	Diesel*	Biodiesel	%	Fossil fuel	Petrol	Diesel*	Biodiesel	%
Base Case	7764	3951	3813	0	0.0%	1248	556	692	0	0.0%
B1 scenario	7746	3906	3840	11	0.1%	1231	513	718	11	0.9%
B2 scenario	7628	3951	3677	136	1.8%	1117	556	561	131	10.5%
Fuel consumption	CIP					MAD				
t d^{-1}	Fossil fuel	Petrol	Diesel*	Biodiesel	%	Fossil fuel	Petrol	Diesel*	Biodiesel	%
Base Case	8725	4402	4323	0	0.0%	3607	1799	1809	0	0.0%
B1 scenario	8653	4232	4421	45	0.5%	3537	1632	1905	44	1.2%
B2 scenario	8360	4402	3958	364	4.2%	3250	1799	1452	357	9.9%

(*) Includes the fossil fuel amount blended with biodiesel

4.3.2 Emissions variation

The HERMES emissions model provides the emissions account for the domains of interest and the fuel consumption estimates during the 17-18 June episode. From now on the 18 June is taken as a basis for all calculations and provided data in order to simplify the analysis of the behaviour during the episode (analogous results were obtained for the first day, 17 June).

The on-road traffic sector is the main contributor to NO$_x$ (57%), NMVOCs (43%) and CO (87%) emissions in the NEIP domain, being also the second source of primary particulate matter (27% of PM$_{10}$ and 32% of PM$_{2.5}$), only overpassed by the combustion in manufacturing industry processes (Figure 4.1). A 21% of these emissions are specifically associated to the Barcelona urban area (corresponding to a defined domain over the greater area of 130 km^2, which .accounts for 1.8 million inhabitants, see *Figure 3.6* on *Chapter 3* for details).

The pollutants emissions pattern in the CIP domain is almost analogous (Figure 4.1), being on-road transport the main emitter of NO$_x$ (74%) and CO (95%). The particulate matter origin is also mainly attributed to the industrial combustion (63% of PM$_{10}$ and 54% of PM$_{2.5}$), but the on-road transport involves the emission of a 42% of the total emitted PM2.5. Moreover, the metropolitan area of Madrid is larger than Barcelona, and the traffic in the conurbation accounts for a 27% of total pollutants emissions (taking into account a 373 km^2 domain defined over the urban area, which accounts for 2.8 million inhabitants, see *Figure 3.6* on *Chapter 3* for details). The largest amount of emitted NMVOCs is attributed in the region to the biogenic emissions (54%).

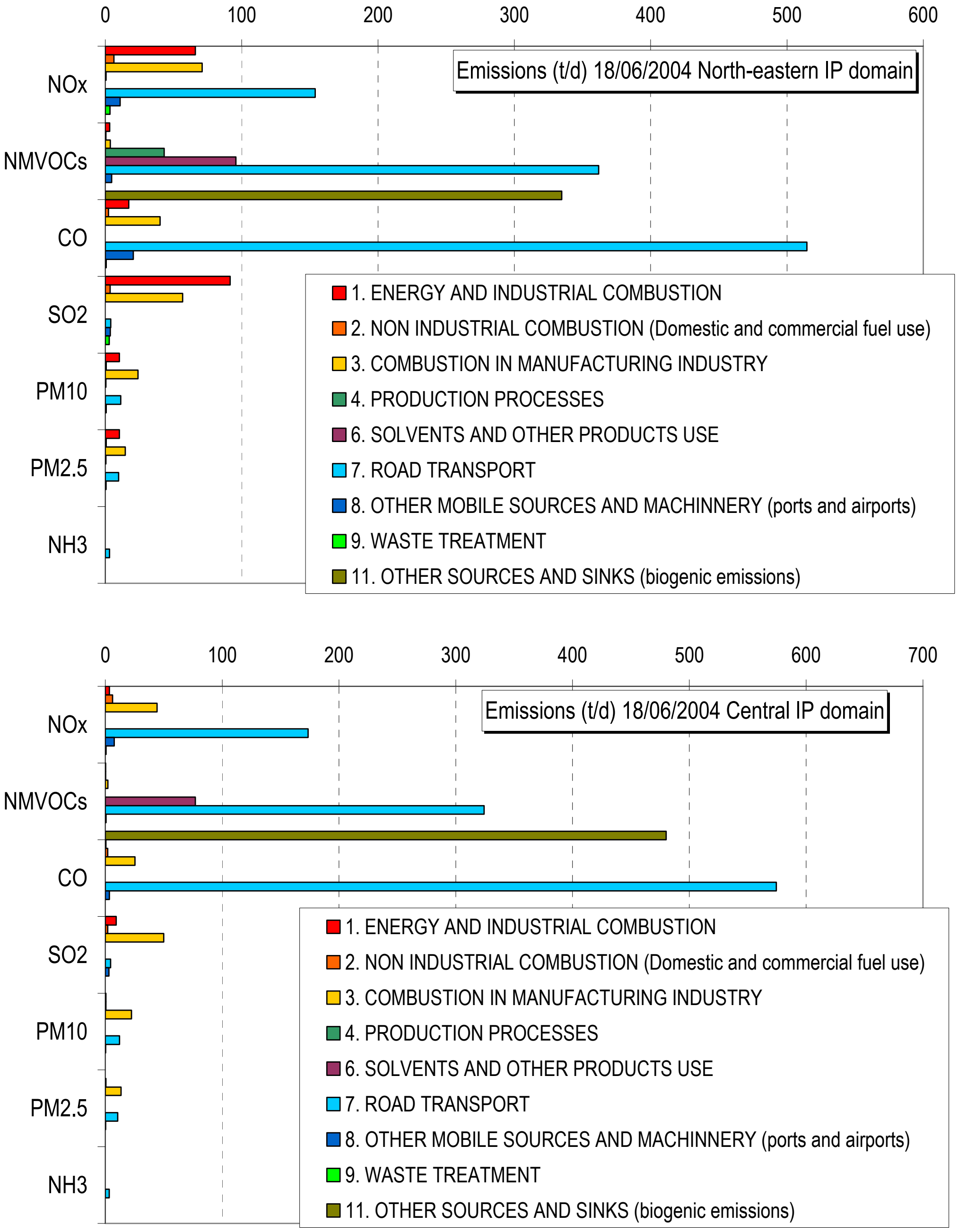


Figure 4.1. Emissions account (t d^{-1}) for the base case scenario provided by HERMES emissions model for the Northeastern IP domain (up) and the Central IP domain (down) during the 18 June; divided by activity sector.

The legislation introduced during last decades to reduce the sulphur content on fuels for on-road transport involved that the emissions of SO_2 attributed to this sector considerably decrease, being in the NEIP domain a 4% and in the CIP domain a 6% of the totally emitted. Concerning this pollutant, efforts have to be done to reduce the industrial combustion sector (60% and 73% in NEIP and CIP) contributions, and specifically in the NEIP also those of energy production (24%).

The substitution of 10% of the oldest diesel and petrol cars in Barcelona and Madrid by newer B20 fuelled cars (B1) reduces NO_x, NMVOCs, CO and PM_{10} emissions in both urban areas (Table 4-4). These changes are attributed both to the fleet renewal and the fuel change. The NO_x emissions decrease in spite of the assumed increase in the emission factor respect to the diesel fossil. The renewal of the vehicle fleet introducing Euro III vehicles compensates the effect of increasing NO_x due to the biodiesel introduction. The oldest gasoline cars substitution has also an aggregated effect in the NMVOCs and CO emissions, traditionally larger in gasoline vehicles, because of being a more volatile fuel and presenting a more inefficient combustion. The PM emissions are commonly larger when using diesel fuelled vehicles, the reductions observed here are not only attributed to the biodiesel introduction, but also to the vehicle fleet renewal, which considers the introduction of vehicles accomplishing Euro III standards.

Table 4-4. On-road traffic emissions variation when introducing the B1 and B2 scenarios of biodiesel use in the Northeastern and Central Iberian Peninsula during the 18, June, 2004.

On-road traffic emissions 18/06/2004 Barcelona												
T d^{-1}	NO_x		NMVOCs		CO		SO_2		PM_{10}		$PM_{2.5}$	
BC	21.3		88.1		104.4		0.7		2.0		1.8	
B1	20.1		85.7		91.6		0.7		1.9		1.7	
B2	21.5		88.0		104.1		0.6		1.9		1.6	
Diff.	NO_x		NMVOCs		CO		SO_2		PM_{10}		$PM_{2.5}$	
	t d^{-1}	%	t d^{-1}	%	t d^{-1}	%	t d^{-1}	%	t d^{-1}	%	t d^{-1}	%
BC-B1	1.2	5.5%	2.5	2.8%	12.8	12.3%	-0.01	-0.8%	0.1	3.2%	0.1	3.7%
BC-B2	-0.2	-1.0%	0.2	0.2%	0.3	0.3%	0.1	14.0%	0.1	7.4%	0.1	8.4%
On-road traffic emissions 18/06/2004 Madrid												
T d^{-1}	NO_x		NMVOCs		CO		SO_2		PM_{10}		$PM_{2.5}$	
BC	66.7		134.5		297.6		1.8		4.9		4.2	
B1	61.3		128.0		248.2		1.8		4.6		4.0	
B2	67.2		134.1		296.8		1.6		4.4		3.7	
Diff.	NO_x		NMVOCs		CO		SO_2		PM_{10}		$PM_{2.5}$	
	t d^{-1}	%	t d^{-1}	%	t d^{-1}	%	t d^{-1}	%	t d^{-1}	%	t d^{-1}	%
BC-B1	5.4	8.1%	6.5	4.8%	49.3	16.6%	-0.02	-0.9%	0.2	4.4%	0.2	5.0%
BC-B2	-0.5	-0.7%	0.3	0.3%	0.8	0.3%	0.3	13.8%	0.5	10.4%	0.5	12.0%

The designed B1 scenario in Barcelona accounts for 5.5%, 2.8%. 12.3% and 3.7% lower on-road traffic emissions of NO_x, NMVOCs, CO and $PM_{2.5}$ than the base case. In Madrid the observed reductions are larger, due to the largest proportion of cars in this city vehicle fleet. B1 involves 8.1%, 4.8%, 16.6% and 5.0% lower emissions of NO_x, NMVOCs, CO and $PM_{2.5}$ than the base case. The SO_2 emissions increase in both urban areas, this is related with the substituted fraction of gasoline (which S content is 0.015 ppm) by diesel fossil (which S content is 0.035 ppm). In Barcelona area the B1 scenario emits 0.8% more SO_2 than the base case and in Madrid 0.9%.

When fuelling all the diesel vehicle fleet of Barcelona and Madrid with B20 (B2), the NO_x emissions increase by the biodiesel effect, being 1.0% larger in Barcelona and 0.9% in Madrid. NMVOCs and CO emissions, which are traditionally low for diesel vehicles, are 0.2% to 0.3% lower. The largest reductions are observed in SO_2 and $PM_{2.5}$, being 14% and 8.4% lower in Barcelona, and 13.8% and 12% lower in Madrid. The ratio of diesel to petrol consumption in Barcelona is higher than Madrid, therefore this scenario is expected to have larger implications in the former on-road traffic emissions.

4.3.3 Air quality simulation evaluation

Air quality surface station hourly data (from 46 air quality stations of the Environmental Departments of the Catalonia and Madrid Governments, Spain) averaged over the domains of study are used to evaluate the performance of WRF-ARW/HERMES/CMAQ predicting ground-level O_3, NO_2, SO_2 and PM_{10} during the case study of 17-18 June, 2004. The evaluation results accomplish the recommendations of the European Directives 1999/30/EC; 2002/3/EC and 2008/50/EC for the air quality objective for modelling assessment methods (they assume a 50% Mean Normalized Gross Error – MNGE - for hourly average O_3, and a 50-60% for hourly average NO_2 and SO_2 concentrations, see Table 4-5).

Table 4-5. Summary of the model evaluation for NEIP and CIP domains, during the 17 -18 June, 2004. The statistical parameters are estimated for the 48 hr period in AQS from the XVPCA and Comunidad de Madrid networks.

Model evaluation	Number of stations	N data	MNBE(%)	MNGE(%)	UPA(%)
O_3	46	1222	-10%	25%	-16%
NO_2	30	1258	-31%	55%	-26%
SO_2	23	481	-43%	55%	-35%
PM_{10}	12	512	-49%	56%	-48%

The slight underestimation of the air quality concentrations (being both the MNBE and the UPA negative for all pollutants and ranging from -10% to -49%), does not have an unique origin. The meteorological model fairly represents the situations of low pressure gradient (Jiménez-Guerrero et al, 2008), as this case of study. Moreover there are still some uncertainties concerning photochemistry (Sillman et al., 1998; Jiménez and Baldasano, 2004) in the low troposphere, and the HERMES emissions model frequently underestimates primary particulates, because it does not take into account erosive or saltation processes and marine aerosols (Vautard et al., 2005a).

4.3.4 Effect on air quality of the biodiesel use

The maximum O_3 levels occur in downwind areas from the Barcelona and the Madrid conurbations, the main NO_x emitters in the studied regions (Figure 4.2). Values above the population information threshold of 120 µg m^{-3} for the 8-hr average concentration (Dir 2002/3/CE) occur in the northern area of the NEIP and in the southern area from Madrid city in the CIP.

In the NEIP, a part from the Barcelona urban area, the Tarragona industrial region (located in the Mediterranean coast, about 100 km to the south of Barcelona) constitutes an important focus of pollutants emissions. Both condition the location of the worst air quality levels in the region during stagnant situations, such as this of 18 June, being in these areas the NO_2 and PM10 daily average concentrations above 80 µg m^{-3} and 45 µg m^{-3}, respectively (Figure 4.2, Figure 4.3). The highest SO_2 concentrations occur over Tarragona area (up to 125 µg m^{-3}), being also important the Barcelona area levels (from 30 to 65 µg m^{-3}) and these achieved in the north-western area of the domain, over the location of the Cercs power plant, which uses coal as a fuel (Figure 4.2).

In the CIP the NO_2, SO_2 and PM_{10} average concentrations are lower than in the NEIP. The highest levels of daily average NO_2 and PM_{10} occur in the Madrid urban region (up to 60 µg m^{-3} and 30-35 µg m^{-3} , respectively) (Figure 4.3), and are associated to on-road traffic emissions. The SO_2 highest levels, up to 30 µg m^{-3}, concentrate over the south-western region of the domain, where some manufacturers and industries are located (Figure 4.2).

The air quality effect of the planned biodiesel scenarios concentrate over the urban areas. The main air quality problems there are mainly related to NO_2 and PM_{10} concentrations, specifically in Madrid area the O_3 levels are also high (Figure 4.3). Two domains are defined over each conurbation to quantify the effects of the proposed scenarios on O_3, NO_2, SO_2, PM_{10} and $PM_{2.5}$ average concentrations. The Barcelona and Madrid greater areas domains (40x40 km^2) provide a measure of the average effect in the areas where the changes are implemented. Additionally two smaller domains (2x2 km^2) are defined over downtown (Barcelona-downtown and Madrid-downtown), expecting to define the effects in those areas where most population will be affected by air quality changes (see *Figure 3.9* on *Chapter 3 for details*).

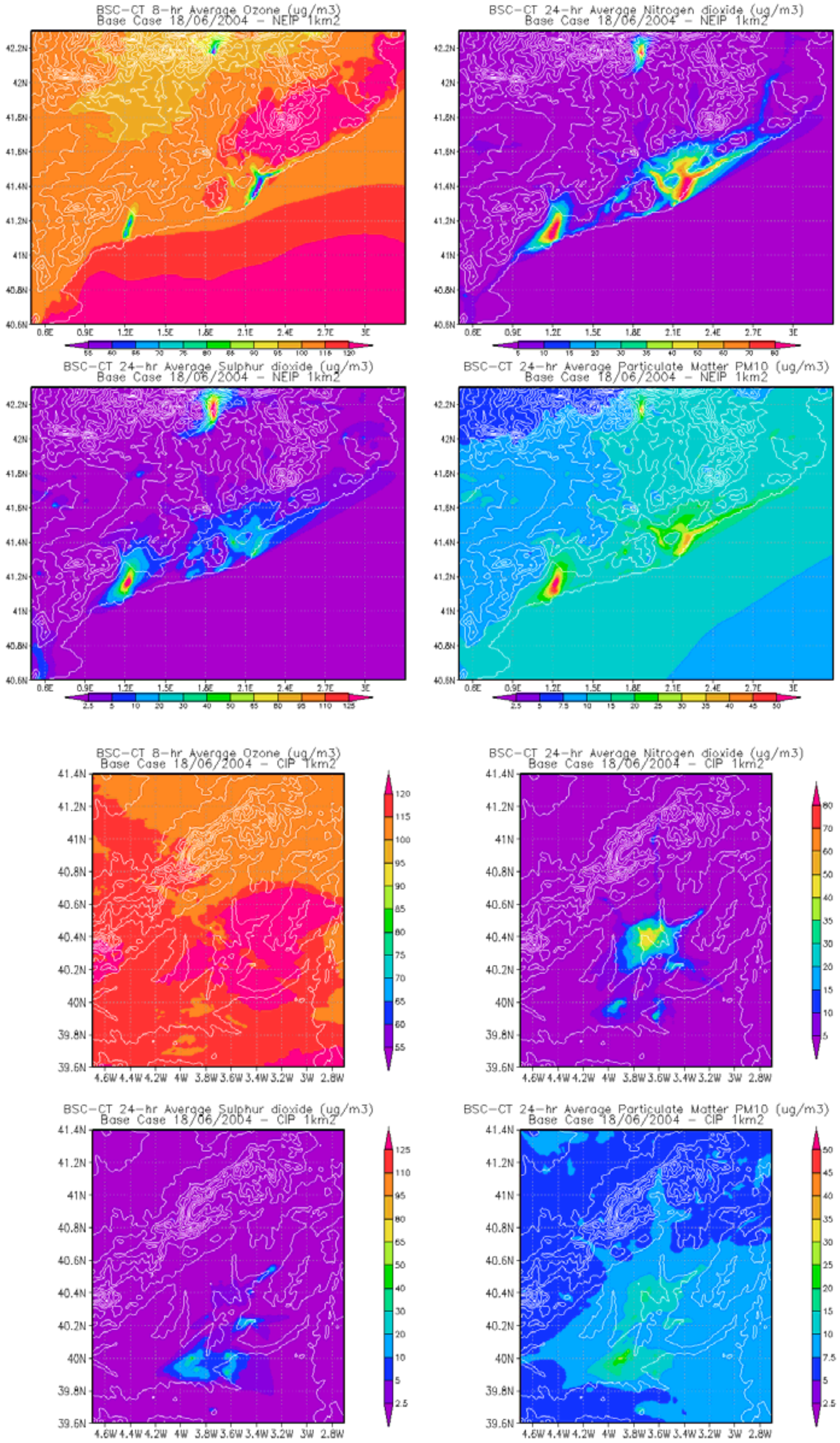


Figure 4.2. 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentration for the NEIP (up) and CIP (down) domains, during the 18 June, 2004.

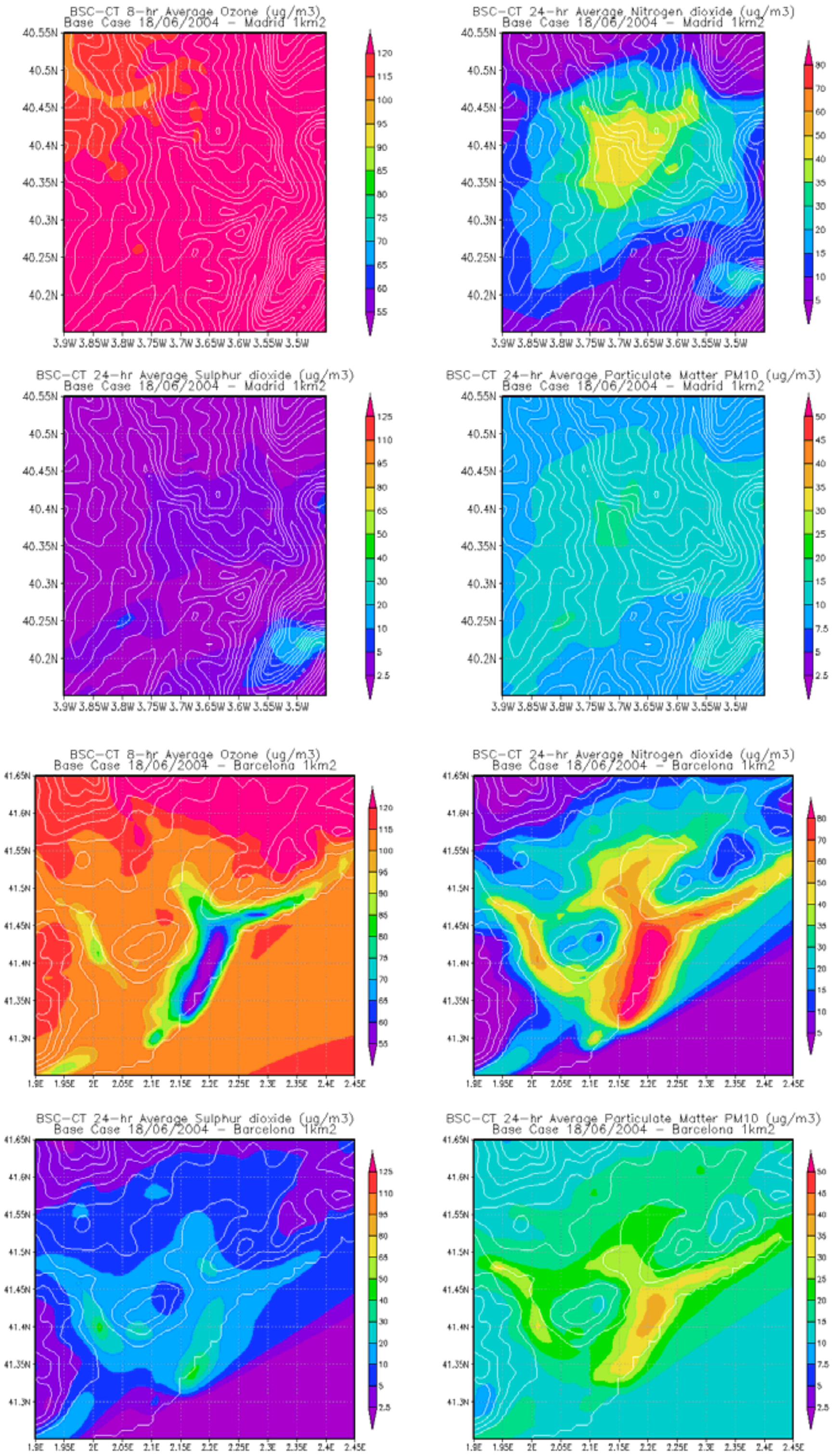


Figure 4.3. 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentration for the Barcelona (up) and Madrid (down) areas, during the 18 June, 2004.

The substitution of 10% of the oldest petrol and diesel cars by B20 fuelled Euro III cars slightly reduces the NO_2 and PM_{10} concentrations (Figure 4.4, Figure 4.5), being reduced by 1.5% and 0.2% the 24-hr average concentrations in Barcelona and by 3.7% and 0.8% in Madrid, respectively (Table 4-6, Table 4-7) The maximum reductions are observed in downtown areas. The Madrid NO_2 levels are specifically affected by this scenario, observing reductions on maximum concentrations around 6 $\mu g\ m^{-3}$ (Figure 4.5), and 24-hr average concentration decreases by 5.2% (Table 4-7). The overall effect in $PM_{2.5}$ concentration is also larger in Madrid, decreasing by 1.2%, face to 0.3% in Barcelona greater area (Table 4-6, Table 4-7). Nevertheless the Barcelona downtown average concentration is specifically reduced by 1.4%,

The effect in O_3 concentration differs between Barcelona and Madrid, due to the different photochemical sensitivity regime they present. The B1 scenario decreases 8-hr average O_3 concentration up to 0.5 $\mu g\ m^{-3}$ in Madrid (reductions on maximum concentrations up to 4 $\mu g\ m^{-3}$ are observed), which represents 0.4% reductions. On the contrary the hourly maximum O_3 concentration in Barcelona increases up to 4 $\mu g\ m^{-3}$. Specifically in downtown the 8-hr average concentration rises 2 $\mu g\ m^{-3}$ (3.2%), due to the NO_x emissions reduction in a VOCs controlled area. These differences on the chemical regime are also in the origin of the different O_3 levels found in the conurbations in the base case, which are higher in Madrid. The effect in downwind O_3 concentrations is positive both in the NEIP and CIP domains (up to 2.5 $\mu g\ m^{-3}$ reductions on 8-hr average concentration, not shown).

The SO_2 emissions increase does not have a remarkable effect on air quality levels of this pollutant, being on average 0.01% and 0.11% higher the concentrations in Barcelona and Madrid greater areas (Table 4-6, Table 4-7).

When the whole diesel vehicle fleet of Barcelona and Madrid is fuelled with B20 the effect on SO_2 and PM_{10} concentrations is larger than in case of substituting and renewing the 10% of the cars fleet (Figure 4.6; Figure 4.7) Hourly maximum concentrations of SO_2 and PM_{10} decrease up to 1.0 $\mu g\ m^{-3}$ and 4.0 $\mu g\ m^{-3}$, respectively, in Barcelona and in a larger extent in Madrid (up to 1.5 $\mu g\ m^{-3}$ and 5.0 $\mu g\ m^{-3}$). The changes concentrate in downtown areas, where the SO_2 24-hr concentration decreases by 1.5% and 6.9% in Barcelona and Madrid, respectively, and the $PM_{2.5}$ 24-hr concentration by 3.1% and 6.0% (Table 4-6, Table 4-7).

The NO_x emissions increase has different effects in Barcelona and Madrid NO_2 and O_3 concentrations. The NO_2 maximum hourly concentration locally increases over some urban roads of Barcelona (Figure 4.6), nevertheless these increases are compensated by reductions in other areas, leading to an overall reduction in 24-hr average concentration over the Barcelona greater area around 0.5% (Table 4-6).

The O_3 levels increase; being the maximum concentrations around 4.0 $\mu g\ m^{-3}$ larger than in the base case at some points of the urban area (Figure 4.6), and the 8-hr average concentration over the Barcelona greater area 0.2% higher (Table 4-6). In Madrid area increases around 0.5 to 1.0 $\mu g\ m^{-3}$ in maximum hourly NO_2 are observed (Figure 4.7), moreover the 24-hr average concentration in the Madrid greater area is 0.3%.

Nevertheless the O_3 concentrations slightly decrease in the urban area, the 8-hr average concentration in the Madrid greater area is 0.03% lower than in the base case, and specifically downtown it is around 0.1% lower (Table 4-7). The NO_x-VOCs ratio in the urban areas conditions the photochemical regime, which defines the response of O_3 and NO_2 concentrations to changes in emissions. As observed for the previous scenarios the response in both cities clearly differs, being Barcelona a typically VOCs controlled area where reductions on NO_x concentrations lead to increases in O_3.

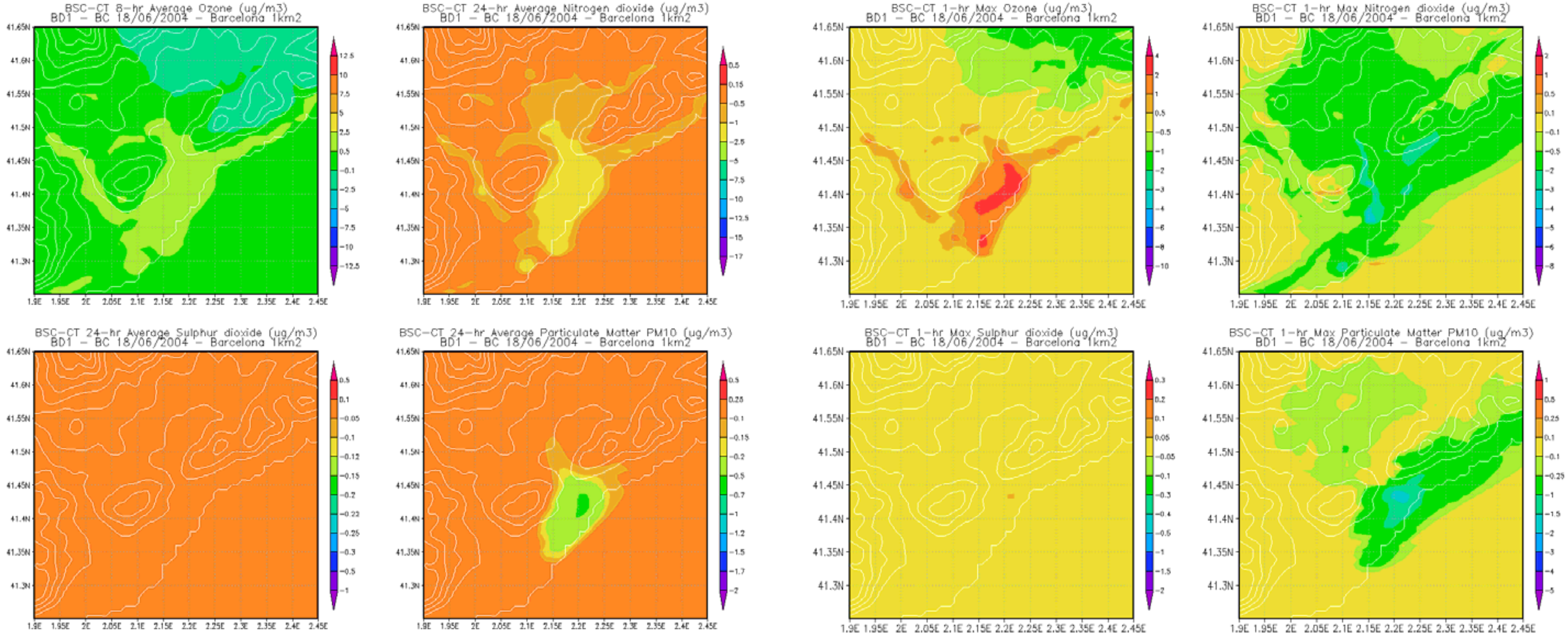


Figure 4.4. 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentration and 1hr-maximum O_3, NO_2, SO_2 and PM_{10} concentration variation between the base case and the B1 scenario in Barcelona area. 18, June, 2004.

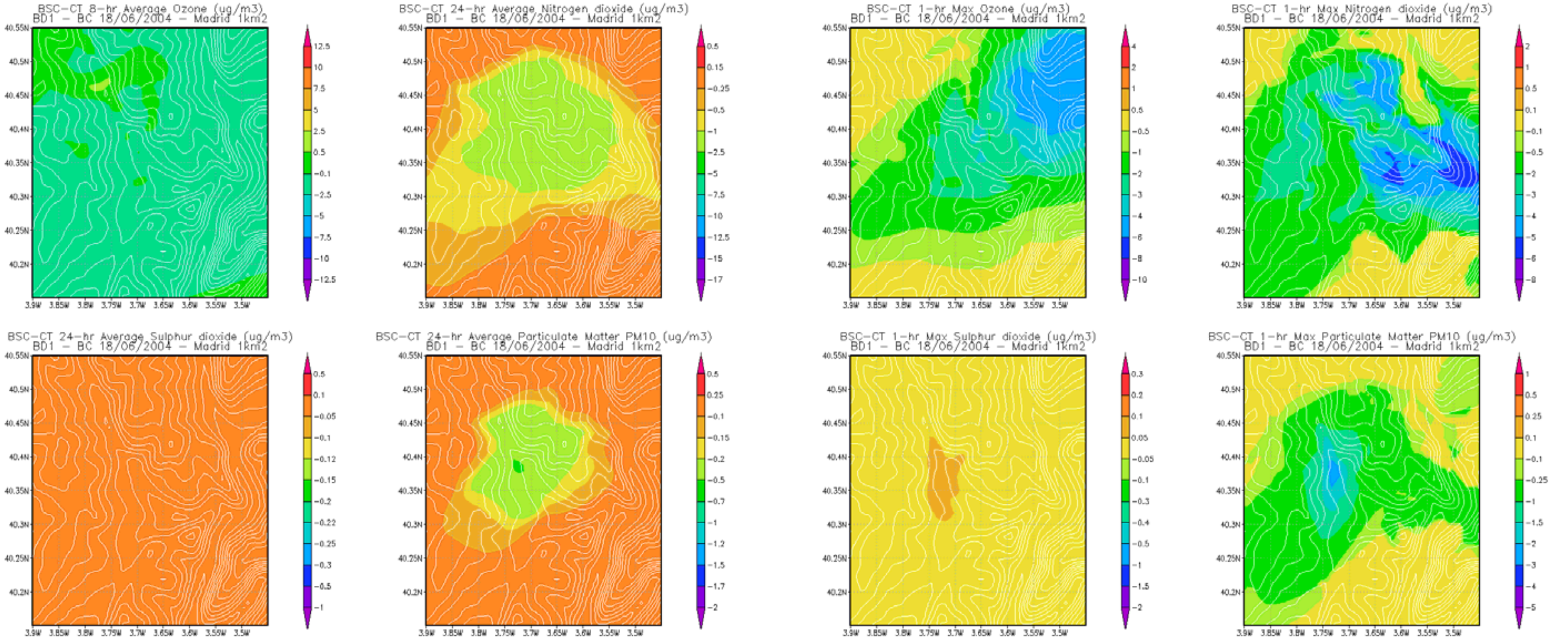
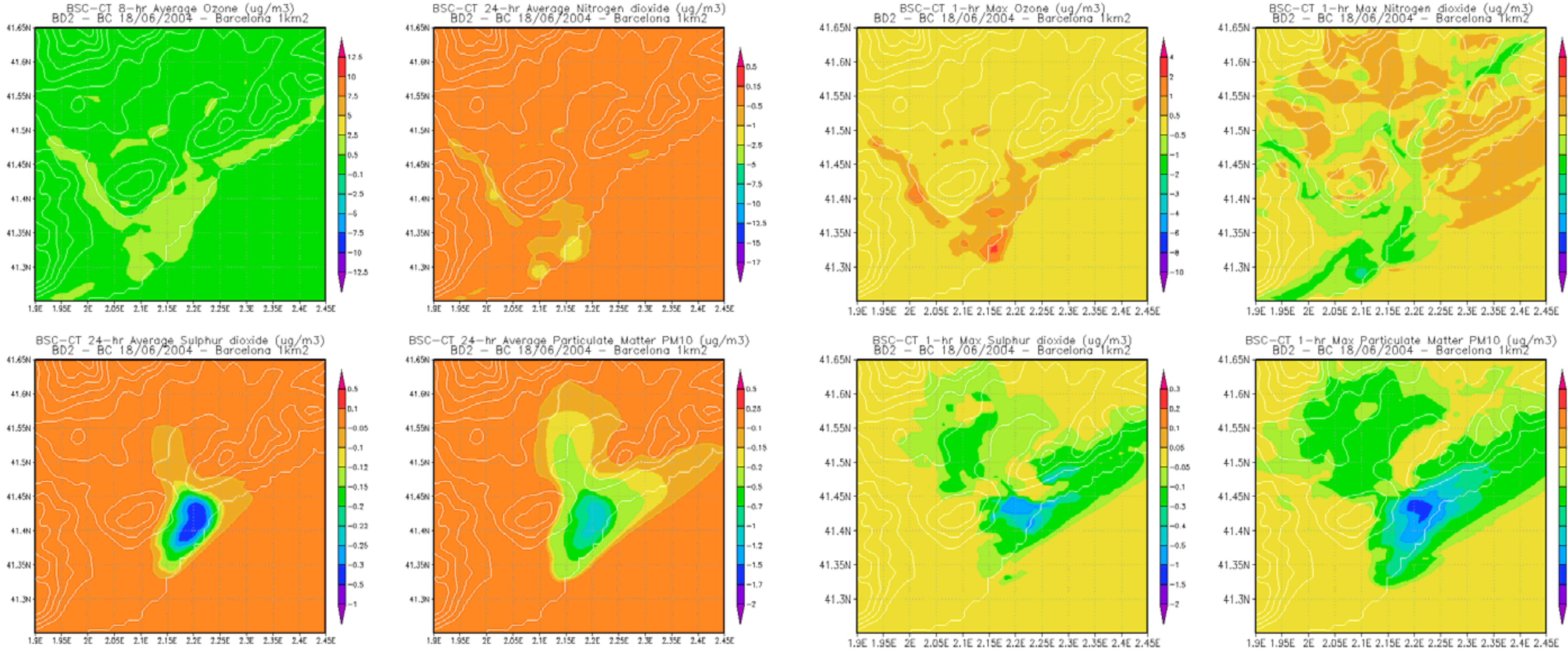


Figure 4.5. 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentration and 1hr-maximum O_3, NO_2, SO_2 and PM_{10} concentration variation between the base case and the B1 scenario in Madrid area. 18, June, 2004.



BSC-CT 8-hr Average Ozone (ug/m3)
BD2 – BC 18/06/2004 – Barcelona 1km2

BSC-CT 24-hr Average Nitrogen dioxide (ug/m3)
BD2 – BC 18/06/2004 – Barcelona 1km2

BSC-CT 1-hr Max Ozone (ug/m3)
BD2 – BC 18/06/2004 – Barcelona 1km2

BSC-CT 1-hr Max Nitrogen dioxide (ug/m3)
BD2 – BC 18/06/2004 – Barcelona 1km2

BSC-CT 24-hr Average Sulphur dioxide (ug/m3)
BD2 – BC 18/06/2004 – Barcelona 1km2

BSC-CT 24-hr Average Particulate Matter PM10 (ug/m3)
BD2 – BC 18/06/2004 – Barcelona 1km2

BSC-CT 1-hr Max Sulphur dioxide (ug/m3)
BD2 – BC 18/06/2004 – Barcelona 1km2

BSC-CT 1-hr Max Particulate Matter PM10 (ug/m3)
BD2 – BC 18/06/2004 – Barcelona 1km2

Figure 4.6. 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentration and 1hr-maximum O_3, NO_2, SO_2 and PM_{10} concentration variation between the base case and the B2 scenario in Barcelona area. 18, June, 2004.

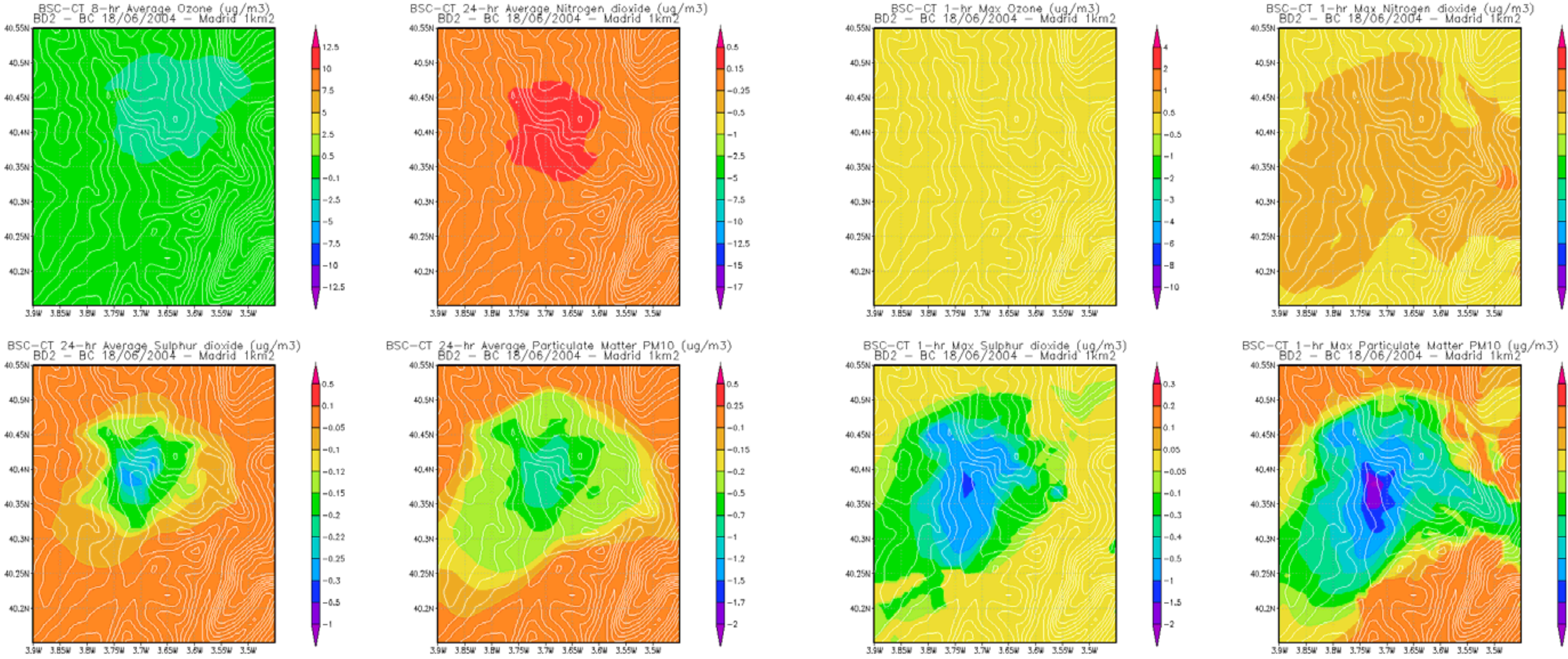


Figure 4.7. 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentration and 1hr-maximum O_3, NO_2, SO_2 and PM_{10} concentration variation between the base case and the B2 scenario in Madrid area. 18, June, 2004.

Table 4-6. 8-hr average O_3, 24-hr average NO_2, SO_2, PM_{10} and $PM_{2.5}$ concentration in the Barcelona Greater Area (BGA) and Barcelona downtown (B-D) in the base case (BC), B1 and B2 scenarios during the 18 June, 2004.

18 June, 2004	8-hr ave. O_3		24-hr ave. NO_2		24-hr ave. SO_2		24-hr ave. PM_{10}		24-hr ave. $PM_{2.5}$	
Conc. (µg m^3)	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D
BC	107.7	64.7	23.0	77.6	8.4	20.4	15.9	30.4	12.3	26.8
B1	108.0	66.7	22.7	75.7	8.4	20.5	15.8	30.0	12.2	26.4
B2	107.9	65.4	22.9	77.2	8.4	20.1	15.8	29.6	12.2	25.9
	8-hr ave. O_3		24-hr ave. NO_2		24-hr ave. SO_2		24-hr ave. PM_{10}		24-hr ave. $PM_{2.5}$	
Diff. (µg m^3)	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D
BC-B1	-0.2	-2.0	0.3	1.8	-0.001	-0.05	0.04	0.37	0.04	0.37
BC-B2	-0.2	-0.7	0.1	0.3	0.02	0.3	0.08	0.84	0.08	0.84
% diff.	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D
BC-B1	-0.2%	-3.2%	1.5%	2.4%	-0.01%	-0.25%	0.23%	1.21%	0.29%	1.37%
BC-B2	-0.2%	-1.2%	0.5%	0.4%	0.3%	1.5%	0.49%	2.76%	0.63%	3.14%

Table 4-7. 8-hr average O_3, 24-hr average NO_2, SO_2, PM_{10} and $PM_{2.5}$ concentration in the Madrid Greater Area (MGA) and Madrid downtown (M-D) in the base case (BC), B1 and B2 scenarios during the 18 June, 2004.

18 June, 2004	8-hr ave. O_3		24-hr ave. NO_2		24-hr ave. SO_2		24-hr ave. PM_{10}		24-hr ave. $PM_{2.5}$	
Conc. (µg m^3)	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D
BC	127.0	121.7	17.3	34.7	2.7	2.4	10.2	12.6	6.7	9.1
B1	126.5	121.4	16.7	32.9	2.7	2.5	10.2	12.3	6.6	8.8
B2	126.9	121.6	17.4	34.8	2.6	2.3	10.1	12.0	6.5	8.5
	8-hr ave. O_3		24-hr ave. NO_2		24-hr ave. SO_2		24-hr ave. PM_{10}		24-hr ave. $PM_{2.5}$	
Diff. (µg m^3)	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D
BC-B1	0.5	0.3	0.6	1.8	-0.003	-0.01	0.08	0.28	0.08	0.28
BC-B2	0.0	0.1	-0.1	-0.2	0.05	0.2	0.18	0.54	0.18	0.54
% diff.	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D
BC-B1	0.4%	0.2%	3.7%	5.2%	-0.11%	-0.5%	0.76%	2.26%	1.16%	3.14%
BC-B2	0.03%	0.1%	-0.3%	-0.5%	1.8%	6.9%	1.74%	4.31%	2.65%	5.98%

4.4 Conclusions

This work evaluates the effects in urban air quality of biodiesel use. Two feasible scenarios of B20 (20% biodiesel – 80% diesel fossil blend) are designed for Barcelona and Madrid urban fleets. The WRF-ARW/HERMES/CMAQ modelling system provides their effect on fuel consumption, emissions and air quality for the selected case of study, the 17-18 June, 2004.

The B1 scenario considers the substitution of 10% of the oldest petrol and diesel cars by Euro III cars fuelled by B20.

The changes in emissions and air quality observed are due to the fuel substitution, around 0.9% and 1.2% of pure biodiesel introduced in Barcelona and Madrid, respectively. But also to the vehicle fleet renewal considered, which slightly reduces the total fuel consumption from the base case (from 0.5% to 0.7% lower in the urban areas), and compensates the NO_x emissions increase due to the biodiesel use. The NO_x, NMVOCs, CO and PM emissions decrease. The Barcelona NO_x emissions decrease by 5.5% (1.2 t d^{-1}). Being the 24-hr average NO_2 concentration over the greater area by 1.5% (from 23.0 to 22.7 µg m^{-3}) lower than in the base case. The VOCs sensitive regime of this area induces an increase of O_3 levels, being the 8-hr average concentration 0.2% higher than in the base case (from 107.7 to 108.0 µg m^{-3}). The final concentration being lower than the European target for human health protection of 120 µg m^{-3}. This scenario in Madrid has a deeper impact because the vehicle fleet is mainly composed by cars. The NO_x emissions decrease in the urban area by 8.1% (5.4 t d^{-1} lower), reducing in this case both the 24-hr average NO_2 but also the 8-hr average O_3 concentrations in the Madrid greater area (by 3.7% and 0.4% respectively). In spite of the reduction the final O_3 concentration is 126.5 µg m^{-3}, which is still larger than the European target for the 8-hr average concentration.

The $PM_{2.5}$ emissions decrease by 3.7% (0.06 t d^{-1}) in Barcelona and by 5.0% in Madrid (0.2 t d^{-1}), reducing the 24-hr average concentration of this pollutant around 0.3% and 1.2% in Barcelona and Madrid greater areas. The largest reductions concentrate in downtown, where the cars circulation is enhanced face to outskirts. The Barcelona $PM_{2.5}$ levels are higher than in Madrid, which has to be attributed not only to on-road traffic emissions, but also to power generation plants and other industrial sources located in the surroundings. This is the main cause of the slight effect that on-road traffic related PM emissions reductions have in final levels of this pollutant in the area.

The SO_2 emissions increase due to the sulphur content in the diesel fossil introduced in the new cars, by 0.8% in the Barcelona area (0.01 t d^{-1}) and 0.9% in Madrid area (0.02 t d^{-1}). Nevertheless its impact on average SO_2 concentrations in the conurbations is not appreciable: 0.01% increase in Barcelona and 0.11% in Madrid, being the final levels 8.4 µg m^{-3}, and 2.7 µg m^{-3}, respectively

The B2 scenario considers the effect of fuelling with B20 all the diesel vehicles in Barcelona and Madrid urban areas, without vehicles typology changes. The fuel substitution it involves is comparable to the 2020 objective set by the European Union (5.75%).

The NMVOCs, CO, SO_2 and PM emissions decrease in the urban areas, while a slight increase in NO_x emissions is observed. The impact of the 1.0% (0.2 t d^{-1}) NO_x emissions increase in Barcelona is moduled by tropospheric chemistry and does not affect the NO_2 levels but in small areas over the conurbation. Overall, the 24-hr average NO_2 concentration over the greater area decreases by 0.5%. Again the O_3 concentration increases by 0.2% (from 107.7 to 107.9 µg m^{-3}), which still does not represent a risk for human health taking into account the European Directives.

The on-road traffic of Madrid emits 0.7% (0.5 t d^{-1}) more NO_x than in the base case, which increase average NO_2 levels by 0.3% (from 17.3 to 17.4 µg m^{-3}). The O_3 response to this scenario is a slight reduction (by 0.03%), which does not represent an appreciable change in final levels (from 127.0 to 126.9 µg m^{-3}).

The $PM_{2.5}$ emissions reductions by 8.4% (0.15 t d^{-1}) in Barcelona and by 12.0% (0.5 t d^{-1}) in Madrid have a direct effect in final levels of this pollutant in the conurbations, being the 24-hr average $PM_{2.5}$ concentration reduced by 3.1% and 6.0% in Barcelona and Madrid downtown, respectively.

The SO_2 from on-road traffic decreases by 14.0% and 13.8% in Barcelona and Madrid, respectively. Their effect in Barcelona (0.3% reduction in 24-hr average concentration) is lower than in Madrid (1.8% reduction in 24-hr average concentration), due to the presence of other sources different from traffic contributing to this pollutant emissions.

This test shows that it is fundamental to define in detail the new emissions scenarios, taking into account not only the new vehicles or alternative fuels that are going to be tested but also the existent characteristics of the vehicles fleets and typology of the study areas. The high resolution modelling permits also to provide detailed effects such as differential variation in O_3 response to the NO_x abatement strategies over the same urban area.

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5.1 Introduction

The introduction of new technology vehicles, such as hybrid vehicles (Demridöven and Dutch, 2004), is one of the available strategies for the short term reduction of on-road traffic emissions in urban areas. The quantitative evaluation of its effects in air quality is fundamental to help decision makers. Air quality modelling is the most suitable tool to obtain geographically distributed information and to define air pollutants variation in advance, including secondary pollutants such as tropospheric O_3 (Sillman et al., 1998; Jiménez and Baldasano, 2004; Ponche and Vinuesa, 2005).

The most common hybrid vehicles are those combining an electric and an internal combustion engine. They can be categorized with respect to the level of electric power integration in the powertrain system and the engine-electric motor coupling strategy, being the parallel and the series configurations the most common. The fuel economy is based on braking energy recovering and the electric motor use to provide additional power in specific situations. Previous studies show that fuel economy peaks under urban driving conditions, where reductions from 40% to 60% respect to conventional technologies are observed (Christidis et al., 2005; Fontaras et al., 2008). This kind of technology could contribute to lower emissions and less petroleum use at small or negative social cost (Demirdöven and Deutch., 2004).

Nowadays the hybrid technology is available in cars, light duty vehicles and heavy duty vehicles, but actually there are mainly cars in the European markets (Badin et al., 2000). In Europe the hybrid fleet still represents a small percentage of the total number of vehicles. Some cities use hybrid urban buses (like London) or introduce measures to promote the use of this kind of vehicles (fiscal promotion, taxes exemption, etc.), and some projections indicate that the introduction of hybrid vehicles in the European market in a 7-10 years term could be important (Rijkeboer et al., 2004), suggesting that hybrids would reach a 6% of market share in 2010 and slightly above 12% by 2020 (Christidis et al., 2005).

In Spain, several policies are being implemented to promote their use: measures related to their purchase, i.e. some communities subsidize up to a 30% of the prize difference between a hybrid and a conventional vehicle; or their use, i.e. the Madrid Council plans to introduce this kind of vehicles in the public fleets, to homologate them to be used as taxis, etc (Madrid Council, 2006). Indirectly some measures like the European directives in emissions limits for vehicles (the Euro 5 limit COM(2005)683 in 2008/9), the planned objectives to the CO_2 emissions by vehicles agreed by the EU and the vehicles manufacturers (from 140 g km^{-1} in 2008 to 120 g km^{-1} in 2010), or the local initiatives to improve air quality as those being developed to Barcelona (Generalitat de Catalunya, 2006), affect the hybrid vehicles introduction.

This work defines two hybrids introduction scenarios in Barcelona and Madrid urban areas, the largest conurbations of Spain. The first scenario considers a low penetration and introduces a 10% of gasoline-electric hybrid cars instead of the oldest diesel and petrol private cars, the second scenario is more optimistic, considering the introduction of a 30% of gasoline-electric hybrids instead of the oldest diesel and petrol

cars and taxis of the urban areas. The WRF-ARW/HERMES/CMAQ modelling system permits to assess the effects on air quality (O_3, SO_2, NO_2 and PM concentrations) with high resolution (1 km^2, 1hr), during a typical photochemical pollution episode of 2004.

5.2 Methods

A deep discussion on the selection of the 17-18 June, 2004 as case of study can be found on *section 2.2.4* of *Chapter 2* and *Annex 1.* The modelling system and domains configuration are described in detail in *sections 2.2.1* and *2.2.3* of *Chapter 2.* The on-road traffic emissions module of HERMES is described in *section 3.2.1* of *Chapter 3*. Therefore this section will focus on the scenarios definition for hybrid vehicles introduction.

5.2.1 Scenarios definition

The planned scenarios include: (H1) **the introduction of 10% of gasoline-electric hybrid cars instead of the oldest petrol and diesel private cars in Madrid and Barcelona** and (H2) **the introduction of 30% of gasoline-electric hybrid cars instead of the oldest petrol and diesel private cars and taxis**. The necessary changes in the HERMES emissions model to introduce the hybrids scenarios are: (1) the modification of the vehicle fleet composition of the urban areas of interest; and (2) the introduction of speed dependent emission factors for the new categories of vehicles: hybrid cars.

5.2.1.1 Introduction of hybrid cars in the vehicles fleet of Barcelona and Madrid urban areas

The vehicle fleet composition for Barcelona and Madrid urban areas is modified in the new scenarios, by adding the hybrid cars category and removing in the same proportion the oldest petrol and diesel cars. The percentage of hybrid cars in the new fleets is estimated in base of the number of vehicles in the 2004 urban fleets, 925 839 in Barcelona and 1 678 942 in Madrid, being 66% and 82% cars, respectively.

The H1 scenario introduces 5.7% of hybrid vehicles in Barcelona and removes the petrol cars previous to 1979 and 23% of those accomplishing the ECE-15/03 (from 1980 to 1984). Moreover the conventional diesel cars are eliminated, those previous to 1996. In Madrid the scenario H1 introduces 7% of hybrid vehicles, instead of all the petrol cars older than 1979, 25% of petrol ECE-15/03 and the diesel conventional cars. The scenario H2 includes 19.7% of hybrid vehicles in Barcelona and 24.7% in Madrid, removing all the petrol cars previous to 1990, including on average 41% of petrol cars with conventional injection system without catalyst (from 1990 to 1992). All diesel conventional cars are changed, including taxis.

The prediction of future trends in hybrids market for Spain is beyond the scope of this work. Nevertheless a rough estimation of the implications of the proposed scenarios in a 5 years period permits us to assure their feasibility. Assuming that the introduction of new vehicles is constant and equal to this registered for 2004 (29% in Barcelona and 27% in Madrid). The H1 renewal implies that by 3.9% and 5.2% of new vehicles in a 5 years period may be hybrid cars in Barcelona and Madrid, respectively. The H2 introduction would involve that 13.5% and 18.2% of new vehicles may be hybrids in Barcelona and Madrid, respectively. The scenario H1 is of the same order of magnitude that some estimations for 2010 (Christidis et al., 2005), being the H2 scenario in the order of magnitude of medium term scenarios.

5.2.1.2 Emission factors for petrol hybrid cars

The EEA-EMEP CORINAIR methodology (Samaras and Zierock, 2007) provides speed dependent emission factors for CO, VOC and NO_x. It also permits to estimate the fuel consumption. The HERMES emissions model estimates additionally CO_2, SO_2, CH_4, NMVOCs, N_2O, NH_3 and particulate emissions (PST, PM_{10} and $PM_{2.5}$). The CO_2 and SO_2 emissions are estimated as a function of the fuel consumption, considering the content of carbon and sulphur in the gasoline. To estimate the particulate matter by combustion, the N_2O and the NH_3 emissions a relation between the fuel consumption of a hybrid car and a petrol Euro III car is estimated and the emission factors are considered to maintain this rate. The coarser PM produced by brakes abrasion, tire wear and pavement erosion are equal to those of petrol cars. The methane emissions are calculated from the total VOCs emissions, considering that the ratio CH_4/VOCs depends on the type of fuel, taking as a reference this of petrol Euro IV vehicles. The cold emissions for all these pollutants and the VOC evaporative emissions are estimated as analogous to petrol Euro III cars (the most recent category implemented in HERMES).

5.3 Results and discussion

5.3.1 Changes on pollutants emissions and fuel consumption

The emissions outcome for the base case scenario is deeply discussed in *Chapter 4*, *section 4.3.2.* Here just a summary to facilitate the reader's comprehension is provided.

The on-road traffic sector is the main contributor to NO_x (57%), NMVOCs (43%) and CO (87%) emissions in the NEIP domain, being also the second source of primary particulate matter (27% of PM_{10} and 32% of $PM_{2.5}$), only surpassed by the combustion in manufacturing industry processes. A 21% of these emissions from traffic are specifically associated to the Barcelona urban area (corresponding to a defined domain over the greater area of 130 km^2, which .accounts for 1.8 million inhabitants, see *Figure 3.6* on *Chapter 3* for details).

The on-road transport is also the main emitter of NO_x (74%) and CO (95%) in the CIP domain. The particulate matter origin can be attributed to the industrial combustion (63% of PM_{10} and 54% of $PM_{2.5}$), but the on-road transport involves the emission of 42% of the total emitted $PM_{2.5}$. Moreover, the metropolitan area of Madrid is larger than Barcelona, and the traffic in the conurbation accounts for 27% of total pollutants emissions (taking into account a 373 km^2 domain defined over the urban area, which accounts for 2.8 million inhabitants). The largest amount of emitted NMVOCs is attributed in the region to the biogenic emissions (54%).

The contribution of on road traffic to greenhouse gases emissions is also important, being in the NEIP domain responsible for 31% of equivalent CO_2 emitted (81985 t d^{-1}), and in CIP for 49% (58866 t d^{-1}). The global warming potential of CH_4 was considered as 23 and of N_2O of 296 referred to CO_2 (Houghton et al., 2001) to make these estimations.

The main benefit of introducing hybrid vehicles concerns fossil fuel savings, which in the NEIP domain are estimated in 0.4% and 1.6% respectively for the H1 and H2 proposed scenarios (Table 5-1). The impact of the same assumptions in the CIP domain is larger, reducing fossil fuel consumption in 1.4% and 5.1% (Table 5-2). This is due to the higher number of vehicles in Madrid face to Barcelona, together with the particular fleet composition (82% of the vehicle fleet of Madrid are cars). The direct effect is the reduction of NO_x, NMVOCs, CO, SO_2 and PM emissions from on-road traffic. The H1 scenario saves 1.8 t of NO_x d^{-1} in the NEIP and 7.8 t of NO_x d^{-1} in the CIP, and the H2, 7.3 t d^{-1} and 33.0 t d^{-1}, respectively (Table 5-1; Table 5-2). The largest changes occur in CO emissions, which are reduced by the H1 scenario in 13.2 t d^{-1} in the NEIP and 50.7 t d^{-1} in Madrid. If the H2 scenario is tested on-road traffic emits 36.2 t d^{-1} of CO in the NEIP and 126.0 t d^{-1} in the CIP less than in the base case.

Table 5-1. On road traffic emissions and fuel consumption (t d^{-1}) for the Northeastern Iberian Peninsula domain during 18 June, 2004. Comparison of the base case scenario, the introduction of a 10% of hybrid cars (H1 scenario) and of a 30% of hybrid cars (H2 scenario).

On-road traffic emissions and fuel consumption (t d^{-1})18/06/2004							
	NO_x	NMVOCs	CO	SO_2	PM_{10}	$PM_{2.5}$	FuelCons.
	NEIP	NEIP	NEIP	NEIP	NEIP	NEIP	NEIP
BC	153.9	361.9	514.6	3.9	11.3	9.7	7764.0
H1	152.1	359.5	500.9	3.9	11.2	9.6	7729.1
H2	146.6	352.2	477.0	3.8	11.1	9.5	7638.7
Differences	NO_x	NMVOCs	CO	SO_2	PM_{10}	$PM_{2.5}$	FuelCons.
(t d^{-1})	NEIP	NEIP	NEIP	NEIP	NEIP	NEIP	NEIP
BC-H1	1.8	2.4	13.7	0.02	0.10	0.10	34.9
BC-H2	7.3	9.7	37.7	0.05	0.19	0.19	125.3
%	NEIP	NEIP	NEIP	NEIP	NEIP	NEIP	NEIP
BC-H1	1.1%	0.7%	2.7%	0.5%	0.9%	1.0%	0.4%
BC-H2	4.8%	2.7%	7.3%	1.3%	1.7%	2.0%	1.6%

Table 5-2. On road traffic emissions and fuel consumption (t d^{-1}) for the Central Iberian Peninsula domain during 18 June, 2004. Comparison of the base case scenario, the introduction of a 10% of hybrid cars (H1 scenario) and of a 30% of hybrid cars (H2 scenario).

On-road traffic emissions and fuel consumption (t d^{-1})18/06/2004							
	NO_x	NMVOCs	CO	SO_2	PM_{10}	$PM_{2.5}$	FuelCons.
	CIP	CIP	CIP	CIP	CIP	CIP	CIP
BC	173.6	324.2	574.5	4.4	12.2	10.5	8724.6
H1	165.8	317.9	522.9	4.3	11.8	10.1	8600.4
H2	140.6	300.1	446.4	4.2	11.5	9.8	8281.3
Differences	NO_x	NMVOCs	CO	SO_2	PM_{10}	$PM_{2.5}$	FuelCons.
(t d^{-1})	CIP	CIP	CIP	CIP	CIP	CIP	CIP
BC-H1	7.8	6.3	51.7	0.07	0.4	0.4	124.2
BC-H2	33.0	24.1	128.1	0.18	0.7	0.7	443.3
%	CIP	CIP	CIP	CIP	CIP	CIP	CIP
BC-H1	4.5%	2.0%	9.0%	1.6%	3.3%	3.9%	1.4%
BC-H2	19.0%	7.4%	22.3%	4.1%	5.6%	6.5%	5.1%

The outcome performed for the tested scenarios indicates that the fossil fuel savings induce reductions on equivalent CO_2 of 111.8 t d^{-1} in the H1 and 402.7 t d^{-1} in the H2 in the Northeastern Iberian Peninsula, which constitute a 0.4% and 1.6% reduction of the GHG originated by on-road traffic in the region. In the Central Iberian Peninsula the equivalent CO_2 is reduced in 395.1 t d^{-1} (H1) and 1412.0 (t d^{-1}), emitting the on-road traffic sector 1.4% and 4.9% less GHG than in the base case.

5.3.2 Air quality in the urban areas.

A summary of the model evaluation can be found on *section 4.3.3* of *Chapter 4*. The air quality conditions in the study areas for the base case are described on *section 4.3.4* of *Chapter 4*, here only those aspects relevant to clarify the results discussion are shown.

The maximum O_3 levels occur in downwind areas from the Barcelona and the Madrid conurbations (Figure 4.2), the main NO_x emitters in the studied regions. Values above the population information threshold of 120 µg m^{-3} for the 8-hr average concentration (Dir 2002/3/CE) occur in the northern area of the NEIP and in the southern area from Madrid city in the CIP. In addition, the Barcelona and Madrid urban areas suffer the highest concentrations of NO_2 and PM_{10}.

Quantifying the change in pollutants concentration over the whole greater areas permits to assess the overall effects of the proposed scenarios. Defining two smaller areas (4 km^2) over downtown provides a more detailed description of their effects (see *Figure 3.9* on *Chapter 3* for details).The introduction of hybrid vehicles pretends to have benefits in urban air quality, where major problems are related with NO_2 and PM_{10} concentrations (*Figure 4.2* on *Chapter 4*), especially in Barcelona area. In Madrid the photochemical regime also involves high O_3 levels in the conurbation (above the 120 µg m^{-3} threshold for the 8-hr average concentration). The differences in emissions origins, photochemical regime (NO_x-VOCs ratio) and atmospheric transport behaviour in both regions are reflected in the final air quality levels assessed (*Figure 4.2* in *Chapter 4*, Table 5-4). These factors also condition the different response to analogous on-road transport management strategies.

The introduction of 10% of hybrid cars instead of the oldest private cars, H1 scenario, is positive in terms of NO_2, SO_2 and PM (both PM_{10} and $PM_{2.5}$) in the conurbations (Figure 5.1). It reduces the NO_2 24 hr average concentration in the Barcelona greater area (see *Figure 3.9* on *Chapter 3* for details) in 1.8% and the $PM_{2.5}$ in 0.3% (Table 5-3). The effects assessed over the smaller downtown area (see *Figure 3.9* on *Chapter 3* for details) follow the same trends; being the reductions on NO_2 and $PM_{2.5}$ concentrations by 3.1% and 1.3%, respectively. The finest fraction of particulates is the most affected, because it is originated mainly by fuel combustion processes or chemical production. The reduction on fuel consumption causes maximum hourly $PM_{2.5}$ concentrations 3.3% lower in downtown (from 46.2 µg m^{-3} to 45.1 µg m^{-3}; Table 5-5).

The impact of this scenario in Madrid is larger than in Barcelona, being the NO_2 and $PM_{2.5}$ levels by 5.3% and 2.1% lower than in the base case (24 hr-average concentrations for the whole greater area). These are coherent with the previously shown changes on emissions. When the changes in the vehicle fleet are more pronounced, the effects both in emissions and air quality are deeper. The H2 scenario reduces NO_2 and $PM_{2.5}$ peaks in Barcelona downtown by 12.6% and 5.3%, respectively, while in Madrid downtown they decrease 29.4% and 13.6% (Table 5-5).

The emissions abatement in the urban areas reduces the formation of NO_2 and secondary particulates in the urban plumes. These pollutants levels decrease for the Barcelona and Madrid downwind areas in both scenarios (Figure 5.1 and Figure 5.2), being the effects more pronounced in the H2 scenario in the Madrid region.

The low SO_2 concentration in the Barcelona greater area remain almost unaffected when introducing the hybrid cars scenarios, indicating its industrial origin (Table 5-3). The maximum hourly concentration in the downtown is reduced 0.03% in the H1 scenario and 0.1% in the H2 (Table 5-5). The impact is larger in Madrid, where a 2.7% reduction on hourly maximum level is estimated downtown in the H1 scenario and a 6.9% in the H2 (Table 5-5). The maximum concentrations in the base case are much lower than in Barcelona (7.0 µg m^{-3} face to 41.1 µg m^{-3}; Table 5-5); where abatement strategies may be addressed to the emissions of the combustion and manufacturing industry sector.

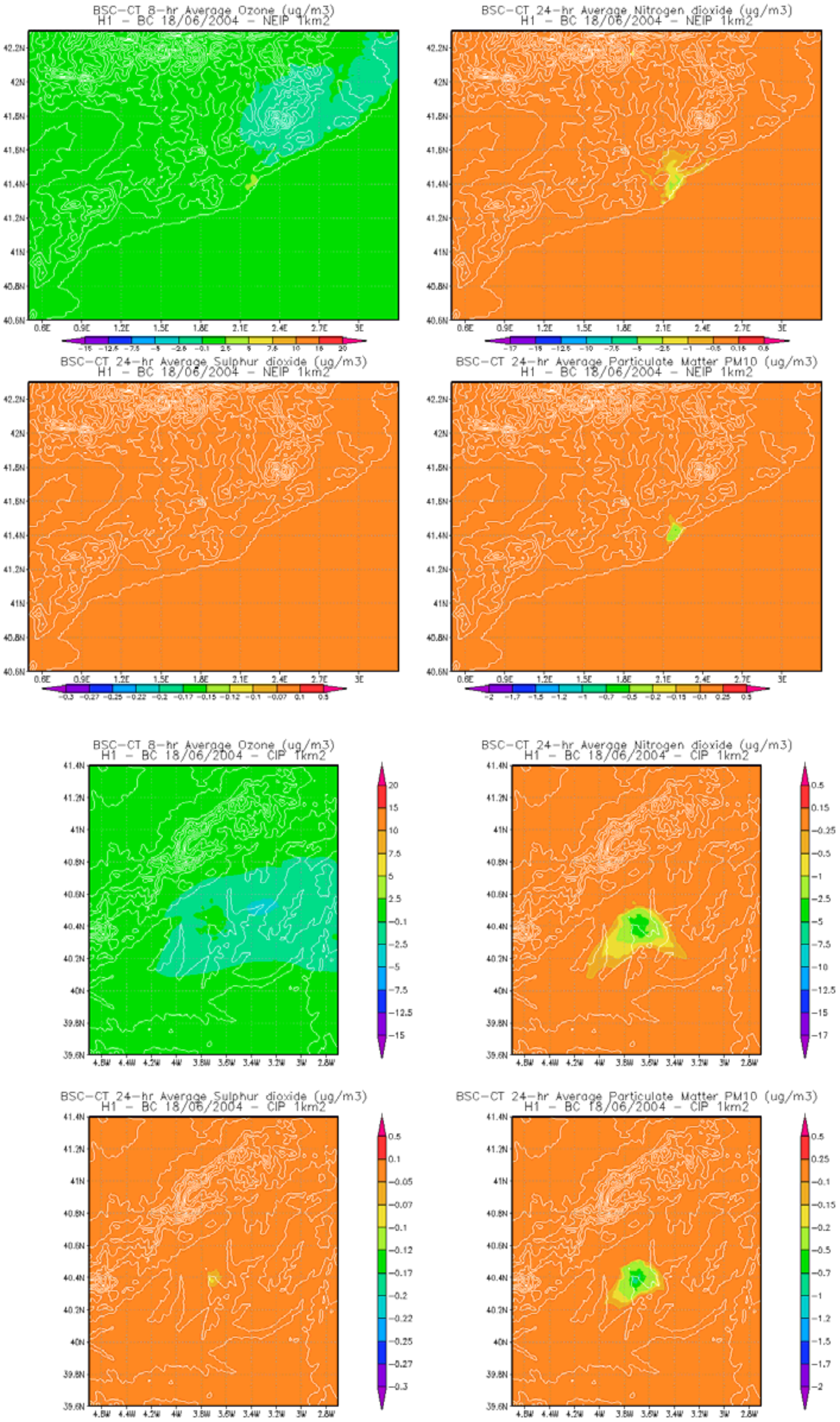

Figure 5.1. Difference in 8-hr average O_3 and 24-hr average NO_2, SO_2 and PM_{10} concentrations between the H1 and the BC scenarios in the NEIP (up) and the CIP domain (down) during 18 June 2004

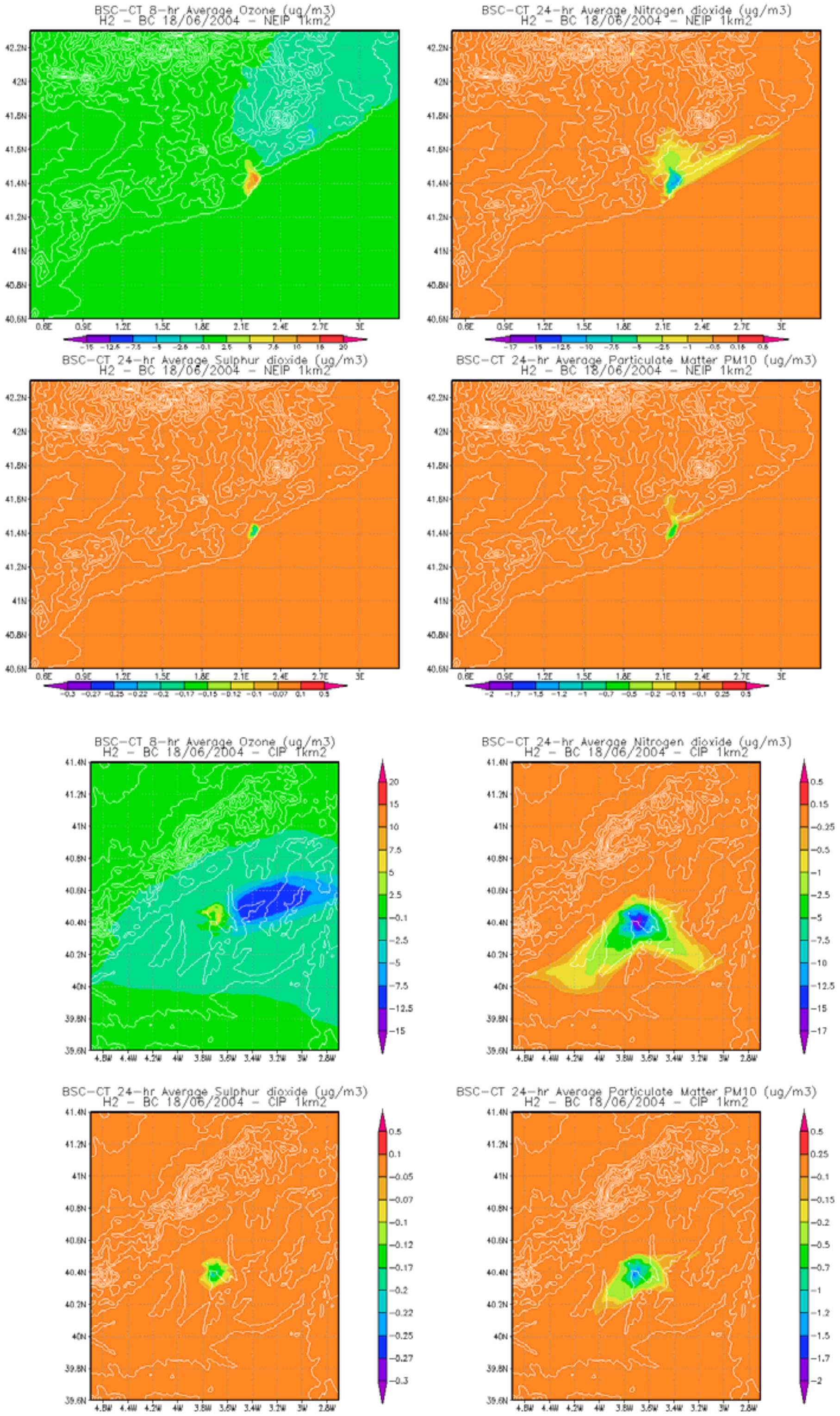

Figure 5.2. Difference in 8-hr average O_3 and 24-hr average NO_2, SO_2 and PM_{10} concentrations between the H2 and the BC scenarios in the NEIP (up) and the CIP domain (down) during 18 June 2004.

Table 5-3. 8-hr average O$_3$, 24-hr average NO$_2$, SO$_2$, PM$_{10}$ and PM$_{2.5}$ concentrations in the Base Case (BC), the H1 scenario (introduction of 10% of hybrid cars) and the H2 scenario (introduction of 30% of hybrid cars) in the Barcelona greater area (BGA) and the Barcelona Downtown (B-D) area. Differences in average concentration between the BC and H1 and H2 scenarios.

18 June, 2004	8-hr ave. O$_3$		24-hr ave. NO$_2$		24-hr ave. SO$_2$		24-hr ave. PM$_{10}$		24-hr ave. PM$_{2.5}$	
Conc. (µg m^3)	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D
BC	107.7	64.7	23.0	77.6	8.4	20.4	15.9	30.4	12.3	26.8
H1	108.1	67.4	22.6	75.2	8.4	20.4	15.8	30.1	12.3	26.4
H2	108.5	73.6	21.7	67.8	8.4	20.3	15.8	29.9	12.3	26.3
	8-hr ave. O$_3$		24-hr ave. NO$_2$		24-hr ave. SO$_2$		24-hr ave. PM$_{10}$		24-hr ave. PM$_{2.5}$	
Diff. (µg m^3)	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D
BC-H1	-0.3	-2.7	0.4	2.4	0.005	0.1	0.03	0.4	0.03	0.4
BC-H2	-0.8	-9.0	1.3	9.8	0.01	0.2	0.03	0.5	0.03	0.5
	8-hr ave. O$_3$		24-hr ave. NO$_2$		24-hr ave. SO$_2$		24-hr ave. PM$_{10}$		24-hr ave. PM$_{2.5}$	
% diff.	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D	BGA	B-D
BC-H1	-0.3%	-4.1%	1.8%	3.1%	0.1%	0.3%	0.2%	1.2%	0.3%	1.3%
BC-H2	-0.7%	-13.8%	5.7%	12.6%	0.2%	0.8%	0.2%	1.7%	0.3%	2.0%

Table 5-4. 8-hr average O$_3$, 24-hr average NO$_2$, SO$_2$, PM$_{10}$ and PM$_{2.5}$ concentrations in the Base Case (BC), the H1 scenario (introduction of 10% of hybrid cars) and the H2 scenario (introduction of 30% of hybrid cars) in the Madrid greater area (MGA) and the Madrid Downtown (B-D) area. Differences in average concentration between the BC and H1 and H2 scenarios.

18 June, 2004	8-hr ave. O$_3$		24-hr ave. NO$_2$		24-hr ave. SO$_2$		24-hr ave. PM$_{10}$		24-hr ave. PM$_{2.5}$	
Conc. (µg m^3)	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D
BC	127.0	121.7	17.3	34.7	2.7	2.4	10.2	12.6	6.7	9.1
H1	126.6	121.8	16.4	32.1	2.7	2.4	10.1	12.1	6.6	8.6
H2	125.5	123.0	13.2	22.4	2.6	2.3	10.0	11.7	6.5	8.2
	8-hr ave. O$_3$		24-hr ave. NO$_2$		24-hr ave. SO$_2$		24-hr ave. PM$_{10}$		24-hr ave. PM$_{2.5}$	
Diff. (µg m^3)	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D
BC-H1	0.4	-0.2	0.9	2.6	0.01	0.05	0.1	0.5	0.1	0.5
BC-H2	1.5	-1.4	4.1	12.2	0.04	0.1	0.2	0.8	0.2	0.8
	8-hr ave. O$_3$		24-hr ave. NO$_2$		24-hr ave. SO$_2$		24-hr ave. PM$_{10}$		24-hr ave. PM$_{2.5}$	
% diff.	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D	MGA	M-D
BC-H1	0.3%	-0.1%	5.3%	7.5%	0.5%	2.0%	1.4%	3.8%	2.1%	5.2%
BC-H2	1.2%	-1.1%	23.8%	35.3%	1.3%	5.0%	2.4%	6.6%	3.6%	9.2%

Table 5-5. Hourly maximum concentration of O_3, NO_2, SO_2, PM_{10} and $PM_{2.5}$ for the base case (BC) scenario in the Barcelona Downtown area (B-D) and the Madrid Downtown (M-D) area during 18 June, 2004.

B-D	O_3	NO_2	SO_2	PM_{10}	$PM_{2.5}$	M-D	O_3	NO_2	SO_2	PM_{10}	$PM_{2.5}$
BC	79.6	116.9	41.1	49.9	46.2	BC	150.0	96.4	7.0	30.6	27.1
H1	82.3	114.0	41.1	48.7	45.1	H1	148.7	91.7	6.8	28.5	25.0
H2	88.3	102.2	41.1	47.4	43.8	H2	147.0	68.1	6.5	26.9	23.4
Difference	O_3	NO_2	SO_2	PM10	$PM_{2.5}$	Difference	O_3	NO_2	SO_2	PM_{10}	$PM_{2.5}$
BC-H1	-2.7	2.9	0.01	1.1	1.1	BC-H1	1.3	4.6	0.19	2.1	2.1
BC-H2	-8.7	14.7	0.04	2.5	2.5	BC-H2	3.0	28.3	0.48	3.7	3.7
% diff.	O_3	NO_2	SO_2	PM10	$PM_{2.5}$	% diff.	O_3	NO_2	SO_2	PM_{10}	$PM_{2.5}$
BC-H1	-3.4%	2.5%	0.03%	2.3%	2.5%	BC-H1	0.9%	4.8%	2.7%	6.9%	7.8%
BC-H2	-11.0%	12.6%	0.1%	5.0%	5.3%	BC-H2	2.0%	29.4%	6.9%	12.0%	13.6%

The NO_x emissions locally act as an O_3 sink. Therefore the introduction of hybrid cars with the consequent NO_x emissions reductions, may increase local O_3 concentrations. This is the overall effect in Barcelona, where both scenarios propitiate an increase in O_3 levels (Figure 5.3 and Figure 5.4). The 8-hr average concentration over the whole metropolitan area increases 0.3% in the H1 and 0.7% in the H2, but the effect could be locally more important, being 4.1% (13.8%) higher the O_3 levels in the Barcelona downtown area in the H1 (H2) scenario (Table 5-3). Nevertheless the highest concentration achieved is 108.5 $\mu g\ m^{-3}$, which is below the EU target for human health protection (Dir 2002/3/CE). In Madrid, higher O_3 levels occur in the metropolitan area (Figure 4.2; Table 5-4), with 8-hr average concentrations of 127.0 $\mu g\ m^{-3}$ in the base case. The introduction of hybrid cars produces a different effect on O_3 concentration depending on the analyzed period. When the O_3 production does not exist or remains low, the effect of reducing NO_x emissions is the local increase of O_3 levels respect to the base case (both when introducing 10% or 30% of hybrid cars). The NO_x role as an O_3 sink is mitigated (Figure 5.3 and Figure 5.4). In the maximum O_3 production period (from 11.00 UTC to 15.00 UTC) the urban O_3 levels are reduced because of the lower amount of NO_x available to react. This behaviour affects the O_3 peaks downtown, being 0.9% lower in the H1 scenario than in the base case and 2.0% lower in the H2 scenario (Table 5-5). Due to the NO_x emissions reductions in the conurbations both scenarios decrease downwind O_3 concentrations (Figure 5.1 and Figure 5.2)

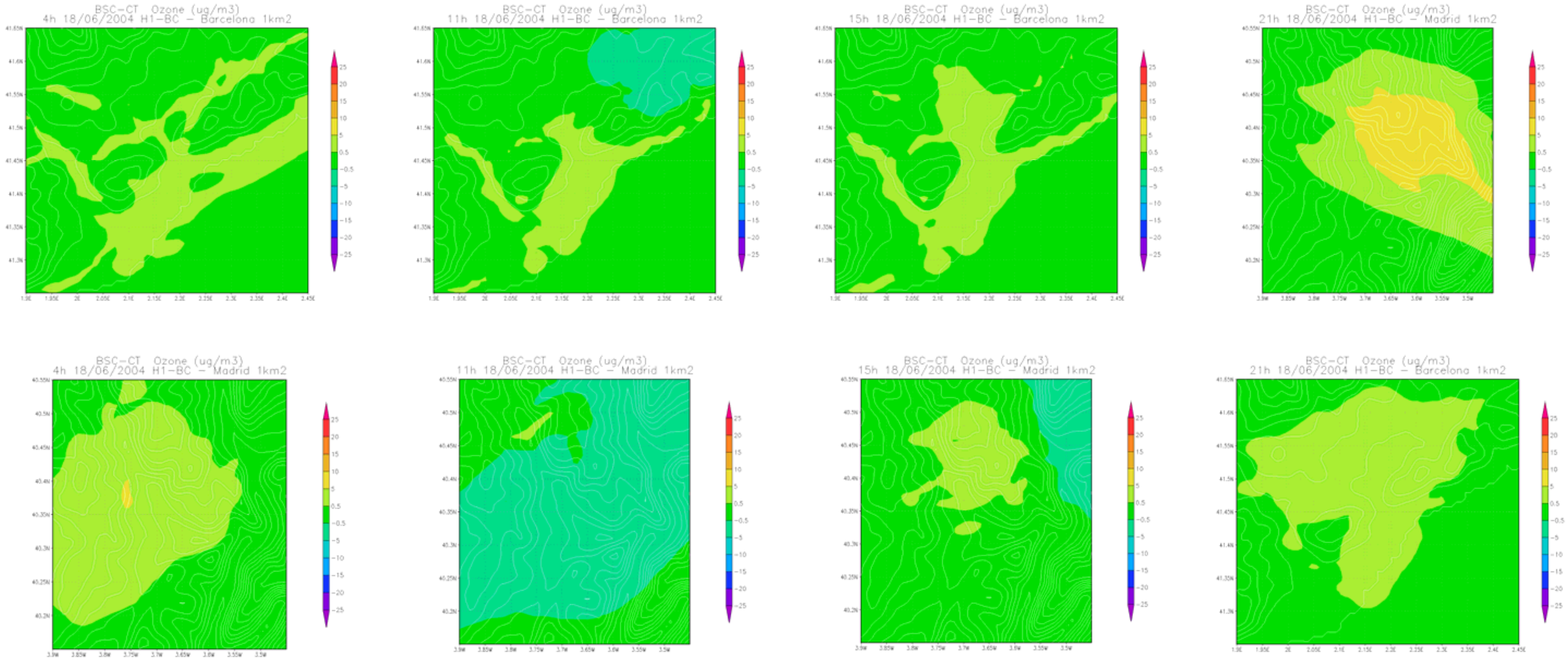

Figure 5.3. Differences in O_3 hourly average concentration between the H1 and the BC scenario at 04.00 UTC, 11.00 UTC, 15.00 UTC and 21.00 UTC of 18 June, for the Barcelona area (up) and the Madrid area (down)

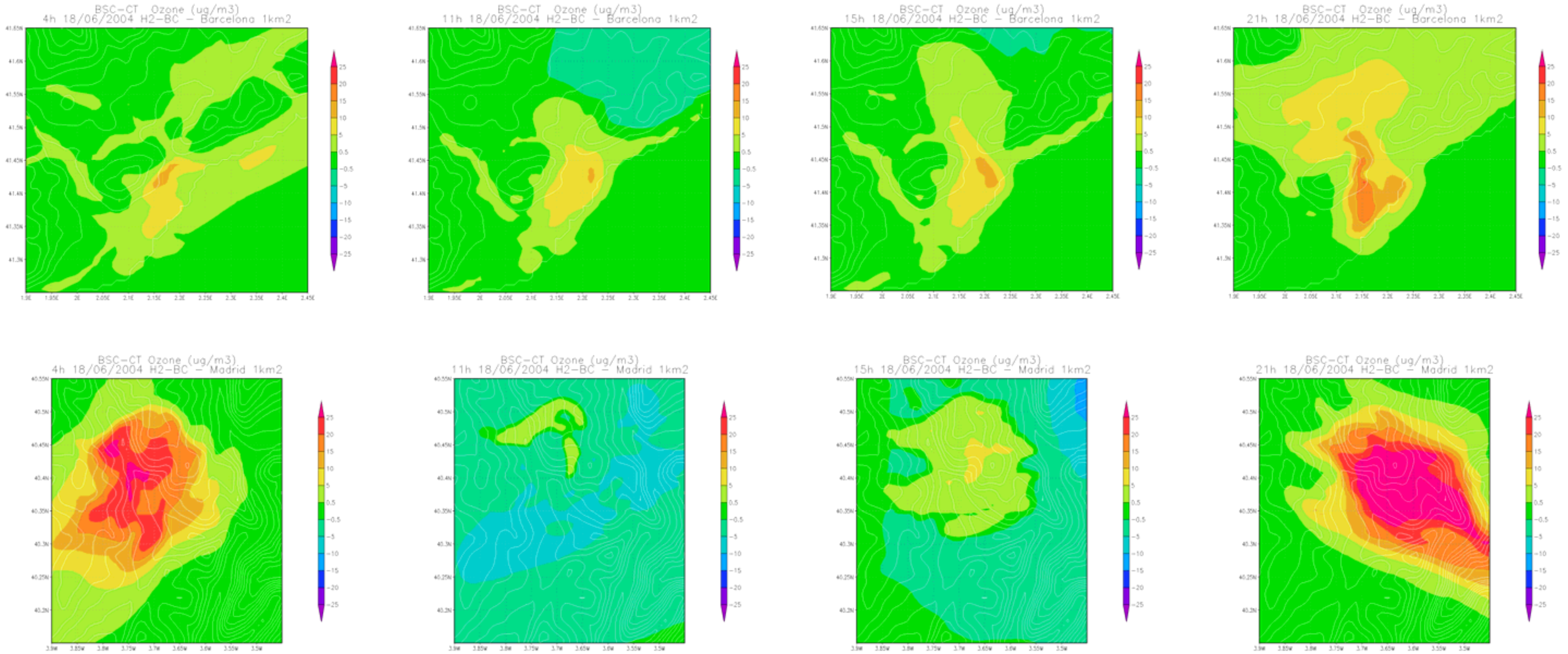

Figure 5.4. Differences in O_3 hourly average concentration between the H2 and the BC scenario at 04.00 UTC, 11.00 UTC, 15.00 UTC and 21.00 UTC of 18 June, for the Barcelona area (up) and the Madrid area (down)

5.4 Conclusions

The improvement of urban air quality in the southern European region is a major concern. Strategies leading to the on-road traffic emissions abatement are being tested, intending to reduce NO_2 and PM levels, but also O_3 concentrations. This work assesses the effects on air quality, fossil fuel consumption and GHG emissions of the introduction of hybrid cars in the Barcelona and Madrid urban areas, the largest conurbations of Spain. The WRF-ARW/HERMES/CMAQ model permits to design different emissions scenarios and to evaluate its environmental benefits taking into account the specific characteristics of the studied areas.

The introduction of 10% of gasoline-electric hybrid cars instead of the oldest petrol and diesel cars (H1) and the substitution of 30% of the oldest petrol and diesel cars, including taxis, by gasoline-electric hybrid cars (H2) are tested for Barcelona and Madrid. The different responses to these changes are mainly due to: the particular vehicle fleet composition, the different contribution of economical or activity sectors to total emissions, the topography, meteorological conditions and the atmospheric transport, being Barcelona a typically coastal city and Madrid a continental one, and finally the different chemical regime existing in both of them (different NO_x-VOCs ratio that directly affects O_3 production response to emissions abatement strategies). All these particularities are taken into consideration in the atmospheric model used.

The overall effect in both cities involves the reduction on NO_2 and $PM_{2.5}$ levels. Which is more pronounced in the H2 scenario, the daily NO_2 and $PM_{2.5}$ concentrations are on average 5.7% and 0.2% lower in Barcelona and 23.8% and 3.6% lower in Madrid, respectively. The largest contribution of on-road transport to total emissions and the fact that the vehicle fleet is mainly constituted by cars (82%) is on the origin of the higher impact of this scenario in Madrid.

The combustion in manufacturing industries is the largest contributor to SO_2 emissions in the Northeastern Iberian Peninsula domain, which makes the on-road traffic management not being an effective strategy to reduce SO_2 levels in the region. The introduction of 30% of petrol hybrid cars, which reduces the SO_2 emissions in 0.05 t d^{-1}, involves the daily average concentrations of this pollutant decreasing just 0.1% in the metropolitan area of Barcelona. The effects of the same scenario in Madrid are notably higher, with SO_2 daily concentration reductions of 1.3%.

The O_3 concentrations in the conurbations locally increase, because of the limitation of the O_3 titration by fresh NO_x emissions. Nevertheless, the photochemical regime involves this increase being more important in Barcelona, while in Madrid the O_3 levels during the central hours of the day are reduced. In fact, the maximum concentrations downtown are lower in the H1 and H2 scenarios than in the base case. The introduction of hybrid cars in the urban areas (both H1 and H2) has positive effects in downwind areas, decreasing specially the NO_2, $PM_{2.5}$ and O_3 levels.

The implications of the on-road traffic sector not only concern local or regional air quality levels, but also greenhouse gases emissions. The equivalent CO_2 emitted by on-road traffic is reduced in 0.4% and 1.6% in the Northeastern Iberian Peninsula when introducing H1 and H2, respectively. In the Central Iberian Peninsula the equivalent CO_2 from on-road traffic decreases 1.4% (H1) and 4.9% (H2), reflecting the larger fossil fuel savings in this region. This work is centred in the vehicle use phase, the benefits of this technology in terms of GHG emissions may be confirmed over the whole life cycle of the vehicles.

The WRF-ARW/HERMES/CMAQ is a powerful tool to help decision makers. Thanks to the high resolution used and the intensive characterization of the studied areas (detailed vehicle fleet compositions, topography and emissions sources) permits to design realistic scenarios and detect subtle differences in air quality parameters.

6 Air quality models sensitivity to speed representation. Evaluation of a 80 km h^{-1} speed restriction in the Barcelona metropolitan area

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6.1 Introduction

The strategies addressed to reduce on-road traffic emissions in urban areas may have two orientations. They intend to either reduce the number of vehicles circulating on conurbations, in particular by urban tolls or parking taxes, or to mitigate the unitary emissions by vehicle, either using alternative fuels and new technology vehicles (hybrids, fuel cells, natural gas, biofuels, etc.) or changing the vehicle circulation speed.

The reduction of the number of vehicles circulating and therefore fossil fuel consumption decreases emissions and greenhouse gases outcome related to on-road traffic sector. The introduction of alternative fuels and new technologies normally involves the reduction of specific pollutants emissions. However, it may have secondary impacts such as unexpected changes in emissions composition (Richter and Williams, 1998; Fenger, 1999). Moreover the continuous growth of the vehicles use in the next 25 years would cancel the effects of technological improvements; therefore an exclusively technological answer to environmental problems related to transport could not be expected (Cannibal and Lemon, 2000). The combination of different strategies may be applied in the future.

A complementary way of reducing traffic emissions consists in changing the speed circulation patterns. The speed dependency of emissions varies as a function of the pollutant, depending on the vehicle age, weight and cubic capacity of the engine. Therefore a unique optimal speed circulation for atmospheric pollutants for the whole range of vehicles in an urban vehicles fleet does not exist (Keller et al., 2008). Nevertheless, it is a widely adopted traffic management strategy, because its benefits concern not only pollutants emissions, but also reduces congestion, noise and traffic accidents.

The design of strategies to reduce tropospheric O_3 is affected by the non-linearity of the reactive transport of pollutants and the uncertainty that the kinetics of O_3 represent in atmospheric chemistry (Sillman et al., 1998; Jiménez and Baldasano, 2004). Therefore the evaluation of air quality management strategies requires the use of air quality models to perform quantitative impact studies (Ponche and Vinuesa, 2005). The third generation Eulerian grid models represent nowadays the state of the art in air quality modelling. The emissions inventories are found one of the main causes of uncertainties in this kind of models predictions (Russell and Dennis, 2000). In particular, on-road traffic modules need more emissions measurements from vehicle types and pollution control technologies to parameterize emissions (Carslaw and Beevers, 2005). Currently, the most used on-road traffic emission models, such as MOBILE or COPERT, apply average speeds to estimate the emission factors for vehicles. An accurate prediction of circulation speed is a key issue to get better traffic emissions models (Smit et al., 2008).

This work pursues two objectives: (1) to assess the **performance of the** WRF-ARW/HERMES/CMAQ air quality model when **changing the constant speed** by road stretch initially considered in HERMES by **hourly speed cycles** obtained from experimental campaigns for several roads in the Barcelona Metropolitan area; and

(2) to analyze the **effects of introducing a speed limit of 80 km h^{-1} in the road network of Barcelona Metropolitan area** on air quality, which is planned by the regional administration to ameliorate the air quality conditions.

6.2 Methods

A deep discussion on the selection of the 17-18 June, 2004 episode can be found on *section 2.2.4* of *Chapter 2* and *Annex 1.* The modelling system and domains configuration are described in detail in *sections 2.2.1* and *2.2.3* of *Chapter 2* and the on-road traffic emissions module of HERMES in *section 3.2.1* of *Chapter 3*. Therefore this section will focus on the scenarios definition for hybrid vehicles introduction.

6.2.1 HERMES modifications and the 80 km h-1 scenario definition

This section describes the three different emissions cases analyzed in this work: (1) the base case of HERMES, which considers constant speed by road stretch (CS); (2) the HERMES modified case, which introduces variable speed cycles for the Barcelona area (VS); and (3) the scenario considering the introduction of a 80 km h^{-1} speed limit in the Barcelona area (V80).

6.2.1.1 From constant to variable speed in HERMES on-road traffic module

During the last decades the information included and processed in the emissions models became more and more complex, intending to be closer to reality. In this sense from the constant average speed by road type (highway, road and street) considered by on-road traffic emission models in the nineties (Costa and Baldasano, 1996), there was an evolution to a specific speed definition by road stretch (Parra et al., 2006). The next step in speed representation might be oriented towards the inclusion of variable speeds, based on measurement campaigns, as proposed in this work.

Initially a constant average speed by road stretch (obtained from the TeleAtlas cartography for 2004) was considered in the HERMES traffic module. The real circulation conditions, such as traffic congestion, involve variations of this ideally represented speed.

Hourly averaged speeds obtained from measurement campaigns (RACC, 2008) are implemented in the main roads of the Barcelona Metropolitan area. The campaign covers an area of 30 km of radius from the conurbation of Barcelona and provides information about traffic circulation speed in the working days from 15 May to 28 July, 2006 and from 20 September to 17 October, 2006, in selected roadways (the B-10, Ronda Litoral, B-20 (C-32), Ronda de Dalt - Túnel de Vallvidriera (C-16), C-58, C-31 (N and S), C-32, B-23 and A 2), which involve 360 road stretches of those defined in HERMES (in pale blue and red dots in Figure 6.1). These include both roads and highways.

The constant speed circulation was changed in these locations by a daily speed cycle based on the real data with hourly discrete values (i.e. Figure 6.2 depicts the daily speed cycle for the C-31 highway stretches as defined in HERMES modified). The traffic volume (number of vehicles by km in a daily basis) affected is 32% estimated for the Barcelona area (Figure 6.1, in yellow).

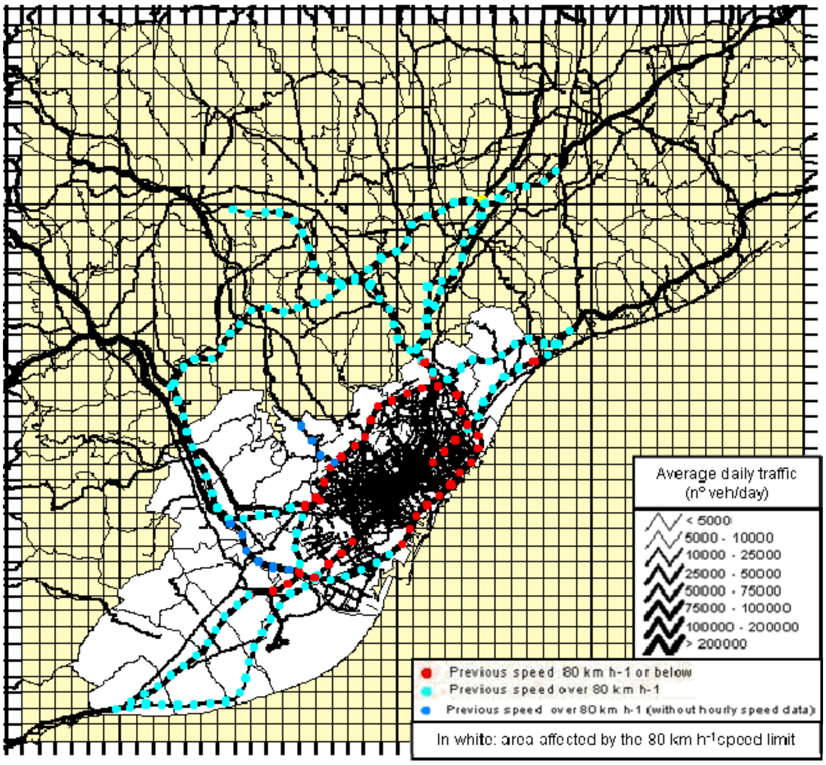


Figure 6.1. Road network description implemented in HERMES, zoom over the Barcelona area, the thickness of the road stretches indicates the average daily traffic (AVT) circulating by them. The pale blue and red dots denote those locations where hourly speed data were available. The speed limit measure was introduced only in the Barcelona Metropolitan area (in white). The red dots denote those locations affected by the speed limit that were already circulating at or below 80 km h^{-1}, and therefore remain unaffected by the limitation. The blue dots inside the Barcelona Metropolitan area were changed to 80 km h^{-1} in the V80 scenario. The area of the north-eastern Iberian Peninsula selected for the emissions variation analysis is depicted in yellow. It covers 2112 km^2, with 1457 km^2 over land surface)

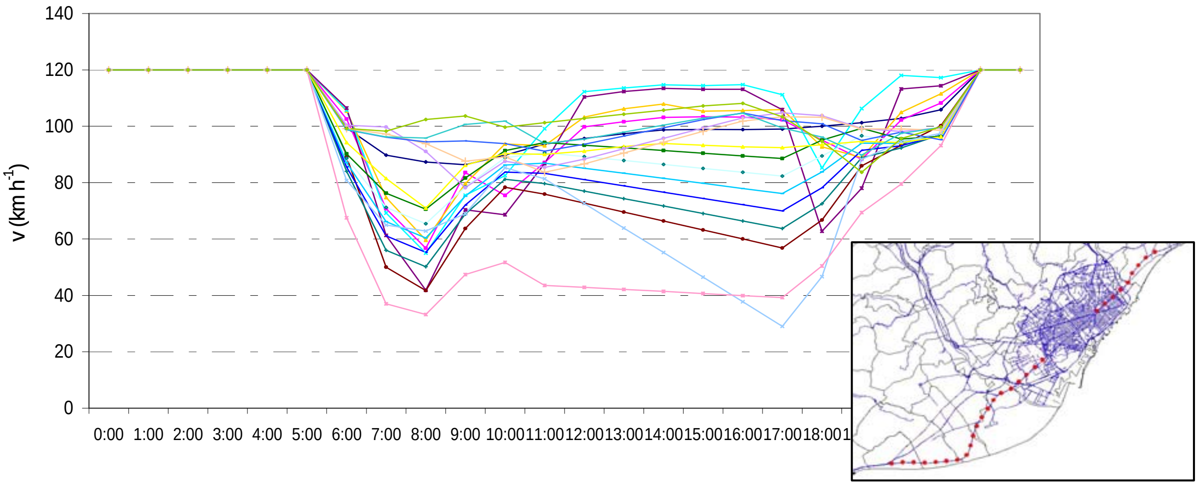


Figure 6.2. Speed dependency considered in HERMES modified for the depicted highway in the Barcelona area (red dots). The variable speed data are available from 6:00 to 23:00 hr, the 24:00 to 5:00 hr period is covered with the previous constant speed provided by the TeleAtlas cartography. In this case the constant speed previously considered in HERMES was 120 km h^{-1}

6.2.1.2 80 km h^{-1} speed limit scenario (V80)

The model is used to assess the pollutants emissions variation and the air quality changes when introducing a traffic management scheme based on regulated speed. The plan of the regional government imposes a maximum speed of 80 km h^{-1} for motorways, dual carriage-ways and main roads of the Barcelona Metropolitan area. This scenario is introduced in the traffic emissions module by changing the speed circulation in the affected road stretches. The new speed limit is introduced only in those roads in which had a previous average speed higher than 80 km h^{-1} (in blue dots in Figure 6.1), taking into account real driving conditions and congestion patterns. It is assumed that congestion would not affect these 80-limited roads, because they were not affected before and the road capacities may increase by decreasing the circulation speed. Therefore constant speed of 80 km h^{-1} is considered for the whole road links affected.

6.3 Results and discussion

6.3.1 Traffic emissions analysis. Base case with constant speed (CS), base case with variable speed (VS) and 80 km h^{-1} speed limit scenario (V80)

The Northeastern Iberian Peninsula domain covers 83398 km^2 and is located in the Mediterranean littoral. The major sources of pollutant emissions are the urban areas of Barcelona, accounting for 3.1 million inhabitants, and Tarragona, which is located in a densely industrialized area, and the road network connecting the Iberian

Peninsula with France, all of them located along the coastal axis. The HERMES – CS model estimates 158.1 t d^{-1} of NO_x emitted for 18 June. Fifty per cent is produced by on-road traffic, which constitutes the main source of primary pollutants in the region, contributing also with 48% of the 759.6 t d^{-1} of NMVOCs emitted in the domain. In this particular case 33% comes from biogenic sources. The power generation and the industrial sectors are the main emitters of SO_2 and primary particles, accounting for 93% and 73% of the mass emissions (162.6 t d^{-1}, 47.4 t d^{-1}), respectively.

The Barcelona urban traffic (Figure 6.1) is responsible for 47% to 58% of on-road traffic emissions depending on the pollutant. The variable speed introduction in the model (HERMES-VS) produces a reduction of 4.2 t d^{-1} of NO_x, 0.1 t d^{-1} of SO_2 and 0.3 t d^{-1} of primary particulate matter (PM_{10}) emissions, while the NMVOCs emissions increase in 0.5 t d^{-1}. The largest changes occur in CO emissions estimates, considering constant speeds by stretch the total amount of CO was 610 t d^{-1}; the HERMES-VS provides an estimation of 595 t d^{-1} emitted for the whole north-eastern Iberian Peninsula domain (see domains definition in *section 2.2.3* of *Chapter 2*)

Focusing the analysis on the Barcelona area (Figure 6.1) the changes in the traffic emissions (Figure 6.3) by the introduction of the variable speed involve reductions of 5.7% of NO_x, 5.1% of CO, 4.8% of SO_2 and 5.1% of PM_{10} emissions. The NMVOCs estimated for the area increase by 0.3%. These changes would have effects in pollutants concentrations predicted in the area and hence in the performance of the model estimates.

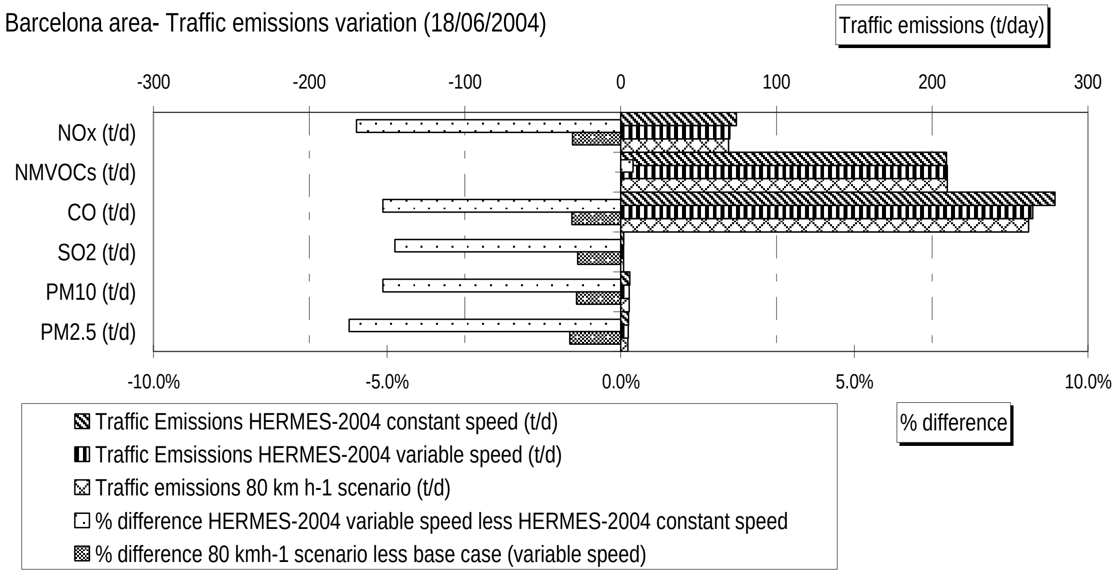


Figure 6.3. Traffic emissions estimations provided by HERMES in the base case (constant speed), in the modified base case (variable speed) and in the scenario that considers the 80 km h^{-1} speed limit in the Barcelona area. The emissions account is provided for the Barcelona area (Figure 6.1) for the 18 June, 2004.

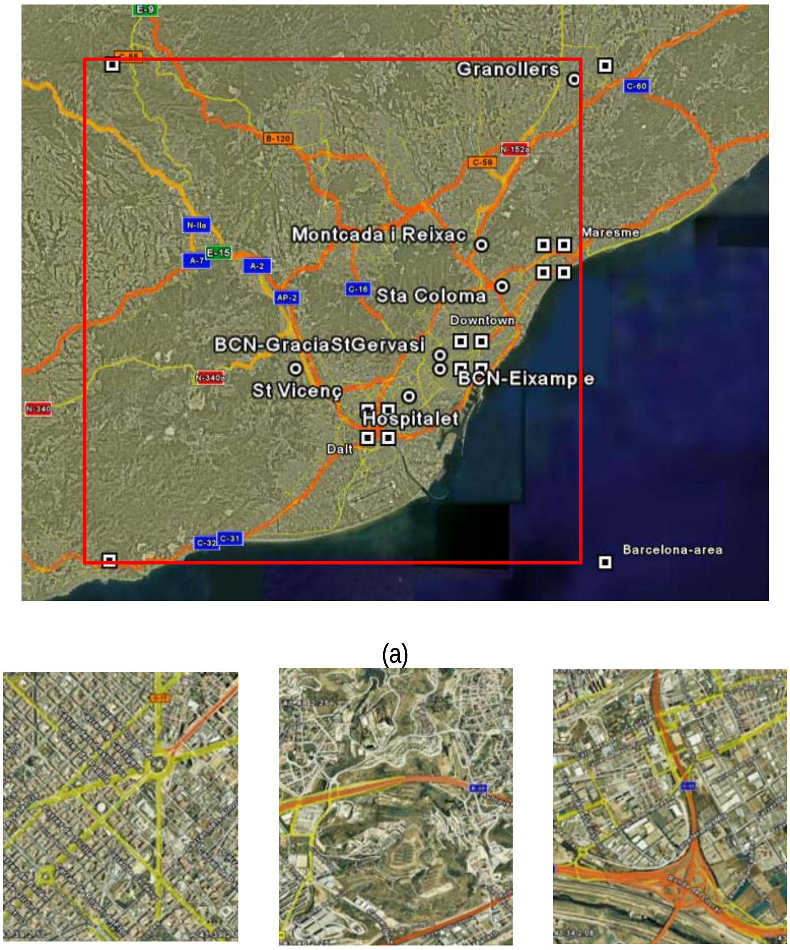
The strategy of introducing an 80 km h^{-1} speed limit for the Barcelona Metropolitan area road network (V80 scenario) reduces emitted NO_x and CO from traffic in 1.0%, and PM_{10} and SO_2 by 0.9% within the Barcelona area, while the NMVOCs emissions increase in 0.01% respect to the variable speed base case (Figure 6.3).

These variations are four times lower than those due to the change from constant to variable speed, because several limited roads were already circulating at speeds lower than 80 km h^{-1}, especially during the daytime periods in which congestion is larger: morning and afternoon traffic peaks (Figure 6.2). Keller et al. (2008) obtain equivalent emissions trends when introducing the 80 km h^{-1} speed limit in Swiss highways, previously considered as circulating at 120 km h^{-1}; NO_x emissions decrease during summertime by 4.3% and NMVOCs emissions remain almost constant. The vehicle fleet composition and the extent of the management measure differ from our study and they do not consider congestion effects, which explain the differences observed in the emissions variations ratio.

6.3.2 Air quality model evaluation. Changes in model performance when including HERMES-VS

Air quality surface station hourly data (provided by the Environmental Department of the Catalonia Government, Spain) over the domain of study are used to evaluate the performance of WRF-ARW/HERMES/CMAQ predicting ground-level O_3, NO_2, SO_2 and PM_{10} during the episode of 17-18 June, 2004. The European Directives 1999/30/EC, 2002/3/EC and 2008/50/EC assume an uncertainty of 50% for O_3, and between 50% and 60% for NO_2 and SO_2 hourly concentrations estimations, for the air quality objective for modelling assessment methods. This uncertainty is defined as the maximum error of the measured and calculated concentration levels. The statistical values obtained as a result of the evaluation considering the estimated emissions of HERMES with constant speed representation by road stretch meet the uncertainty objectives set by the European Directives.

Additionally the modified modelling system, when introducing variable speed cycles in the Barcelona Metropolitan area, is used to assess the air quality levels in the region. These predictions were validated against the same air quality stations data. The results for several stations selected over the Barcelona area (Figure 6.4-a, Table 6-1)) indicate that the model underestimates O_3 concentrations, both using HERMES-CS and HERMES-VS emissions as inputs. The introduction of variable speeds based on experimental data involves light improvements in O_3 predictions, reducing in some cases the statistical parameters, i.e. in L'Hospitalet or Montcada air quality stations from 1 to 2% reductions on MNBE, MNGE and UPA are observed.



(a)

(c) (d) (e)

Figure 6.4. a) Selected air quality stations in the Barcelona area to perform the model assessment with HERMES and HERMES-modified. Inside the red square: Barcelona area (40x40 km^2, 41°15'0"N -41°36'36"N; 1°49'48"E - 2°18'36"E) c) Barcelona downtown (2x2 km^2, 41°23'24"N - 41°24'36 "N; 2°10'12"E - 2°11'24"E), d) Barcelona Maresme (2x2 km^2,,41°27'36"N - 41°28'48"N; 2°15'0"E - 2°16'12.00"E), e) Barcelona Dalt (2x2 km^2, 41°20'24"N - 41°21'36"N; 2° 4'48"E - 2° 6'0.00"E) domains selected for the evaluation of the management strategy effects on air quality

The underpredictions of NO_2 concentrations occurring in the Barcelona area (i.e. BCN-Eixample, L'Hospitalet or St Vicenç) are not improved with the methodological change in HERMES. The NO_x emissions from traffic considering the real time speed cycles are lower than those previously estimated by using the HERMES data with constant speed. The underpredictions may be caused by traffic related emissions, either due to uncertainties in emissions factors or to an underestimation of the real traffic volumes. Nevertheless, the origin of the discrepancies between modelled and measured NO_2 levels could not only be attributed to the emissions estimates, but also to the chemistry representation in the model or the meteorological predictions, which particularly showed problems in representing wind fields during calm situations such as those investigated in this study (Jiménez et al., 2008).

The model tends to underestimate both SO_2 and PM_{10} average and peak concentrations in the Barcelona area. When introducing the HERMES-VS data the PM_{10} predictions remain almost constant, improving at some points (i.e. 1% reduction in the UPA for the BCN-Eixample PM_{10} estimates) but worsen in others (i.e. 1% increase of the MNGE in the L'Hospitalet station for PM_{10} predictions). One of the reasons for the PM_{10} underpredictions in the area can be found in the emissions model itself, which does not take into account natural sources of primary particulates such as erosive or saltation processes or marine aerosols which may be important for the area of study (Vautard et al., 2005a). Moreover the inaccuracies in representing accumulation and transport patterns during this low gradient pressure situation could be other source of these underpredictions. The SO_2 emissions contribution of the on-road traffic sector in the Northeastern Iberian Peninsula domain accounts for 2% of the daily mass emissions. The improvement of the traffic representation in the Barcelona area (which accounts for 49% of SO_2 traffic related emissions in that region) does not involve appreciable changes in model performance, indicated by constant statistical parameters for all stations.

Table 6-1 Model evaluation statistical parameters for the HERMES (CS- constant speed) and the HERMES-modified (VS – variable speed) simulations for several AQS (Figure 6.4-a) in the Barcelona area. Green colour reflects an improvement in and red colour a worsening.

BCN-Eixample AQS	MNBE (%)		MNGE (%)		UPA (%)	
	CS	VS	CS	VS	CS	VS
O_3	-11%	-11%	11%	11%	-19%	-18%
NO_2	-42%	-42%	44%	44%	-29%	-29%
PM_{10}	-35%	-35%	40%	40%	-2%	-1%
SO_2	-37%	-37%	60%	60%	-47%	-47%
L'Hospitalet AQS	MNBE (%)		MNGE (%)		UPA (%)	
	CS	VS	CS	VS	CS	VS
O_3	-14%	-13%	18%	17%	-29%	-28%
NO_2	-21%	-23%	35%	36%	-15%	-15%
PM_{10}	-37%	-38%	47%	48%	-53%	-54%
Sta Coloma AQS	MNBE (%)		MNGE (%)		UPA (%)	
	CS	VS	CS	VS	CS	VS
O_3	-48%	-48%	48%	48%	-52%	-52%
NO_2	49%	49%	62%	62%	3%	3%
SO_2	-26%	-26%	39%	39%	14%	14%
BCN-Gracia St Gervasi AQS	MNBE (%)		MNGE (%)		UPA (%)	
	CS	VS	CS	VS	CS	VS
NO_2	-54%	-55%	54%	55%	-46%	-46%
PM_{10}	-7%	-7%	35%	35%	8%	9%
SO_2	-48%	-48%	52%	52%	-8%	-8%
St Vicenç AQS	MNBE (%)		MNGE (%)		UPA (%)	
	CS	VS	CS	VS	CS	VS
O_3	-8%	-8%	18%	17%	-13%	-13%
NO_2	-12%	-12%	72%	72%	1%	1%
PM_{10}	-23%	-23%	63%	63%	-66%	-66%
Granollers AQS	MNBE (%)		MNGE (%)		UPA (%)	
	CS	VS	CS	VS	CS	VS
O_3	-9%	-8%	21%	20%	0%	2%
NO_2	6%	3%	57%	57%	-42%	-42%
SO_2	-75%	-75%	75%	75%	-84%	-84%
Montcada AQS	MNBE (%)		MNGE (%)		UPA (%)	
	CS	VS	CS	VS	CS	VS
O_3	-25%	-23%	27%	26%	-32%	-31%
NO_2	1%	-3%	32%	33%	-11%	-11%

6.3.3 Air quality variation when introducing the limit in speed circulation to 80 km h^{-1}

The photochemical simulation results for the **Northeastern Iberian Peninsula** during the 17-18 June, 2004 show that the maximum O_3 concentrations occur in downwind areas from Barcelona city after the maximum photochemical activity hours (Figure 6.5–a,b show the simulation results for the 18 June). During the day the increase of the solar radiation and the temperature (reaching 30-35ºC) promote the high levels of O_3, exceeding in some cases the population information threshold (180 μg m^{-3}) and the 8-hr average objective to human health protection (120 μg m^{-3}). The stagnant conditions involve very similar atmospheric circulation patterns during the 17-18 June, controlled by the sea breezes regime. The main difference concerns the photochemical pollution accumulation that takes place over the Mediterranean Sea, a characteristic process in this region during summertime (Baldasano et al., 1994; Jiménez et al., 2006). The major air quality problems in the **Barcelona urban area** are related with NO_2 and PM_{10} levels. The hourly maximum NO_2 concentrations exceed 200 μg m^{-3} and the PM_{10} overpasses the 50 μg m^{-3} (Figure 6.6).

When introducing the 80 km h^{-1} limitation, the 24-hr average NO_2 concentration over the whole **Barcelona area** (Figure 6.4-a) on the selected days decreases by 0.7% and 0.8% (Table 6-2). The largest reductions are observed in those areas that are directly affected by the speed limitation. In the Maresme and Dalt areas, which located in the northern and southern ways out of Barcelona conurbation respectively (Figure 6.4-c,d) the reduction of the 24-hr average NO_2 concentration amounts up to 5.7%. Both areas include roads limited to 80 km h^{-1}. The effects in downtown (Figure 6.4-b), where most city dwellers may be affected are less, the average NO_2 reduction being 0.1% and 0.3%. The benefits are also reflected in the urban plume, specifically the hourly maximum NO_2 concentration reduces up to -2.5 μg m^{-3} in the downwind region (Figure 6.7).

The NO_x emissions reductions in a VOCs limited area cause an 8-hr average O_3 concentrations increase of 0.1% over the **Barcelona area** (Table 6-2). In detail the largest changes in O_3 concentration (up to 3.5 μg m^{-3} on average; Figure 6.8) are observed in the areas over the speed-limited roads. Specifically the Maresme area reflects deeply the management measure effect; increases in O_3 average concentration amount up to 2.5%, due to the dominant northwesterlies controlling the pollutants plume displacement. The effect decreases with distance to the conurbation. The 8-hr average O_3 concentrations downwind are between 0.75 and 1.25 μg m^{-3} higher than those in the base case (Figure 6.7), which is two orders of magnitude lower than the average value in these areas, ranging from 100 to 120 μg m^{-3}.

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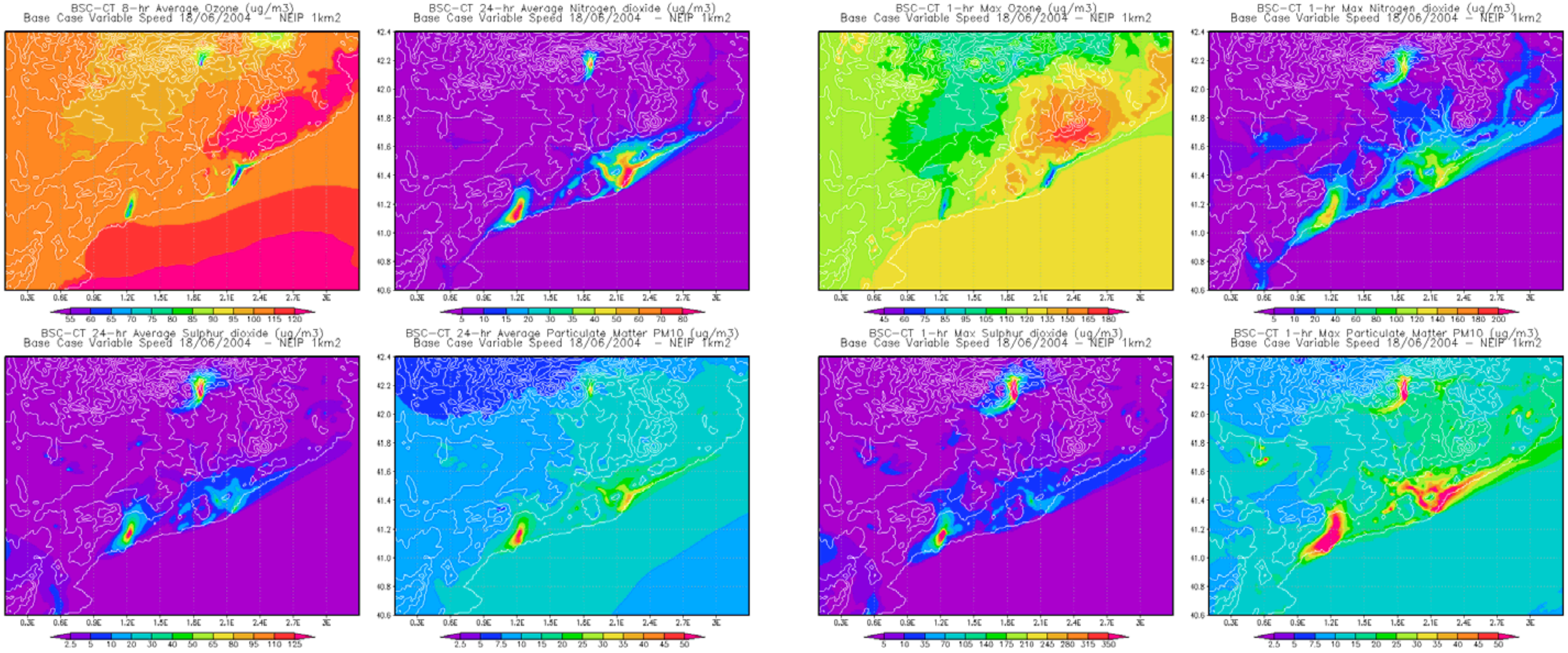
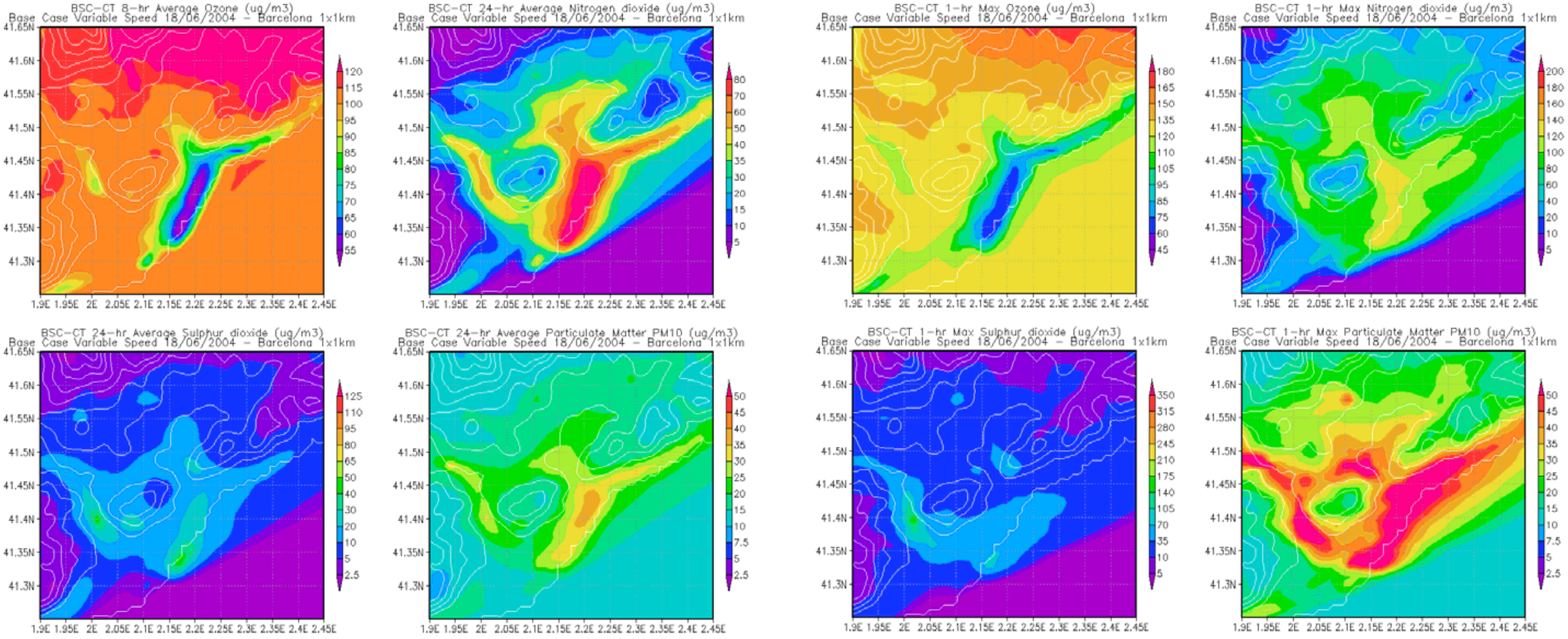


(a) (b)

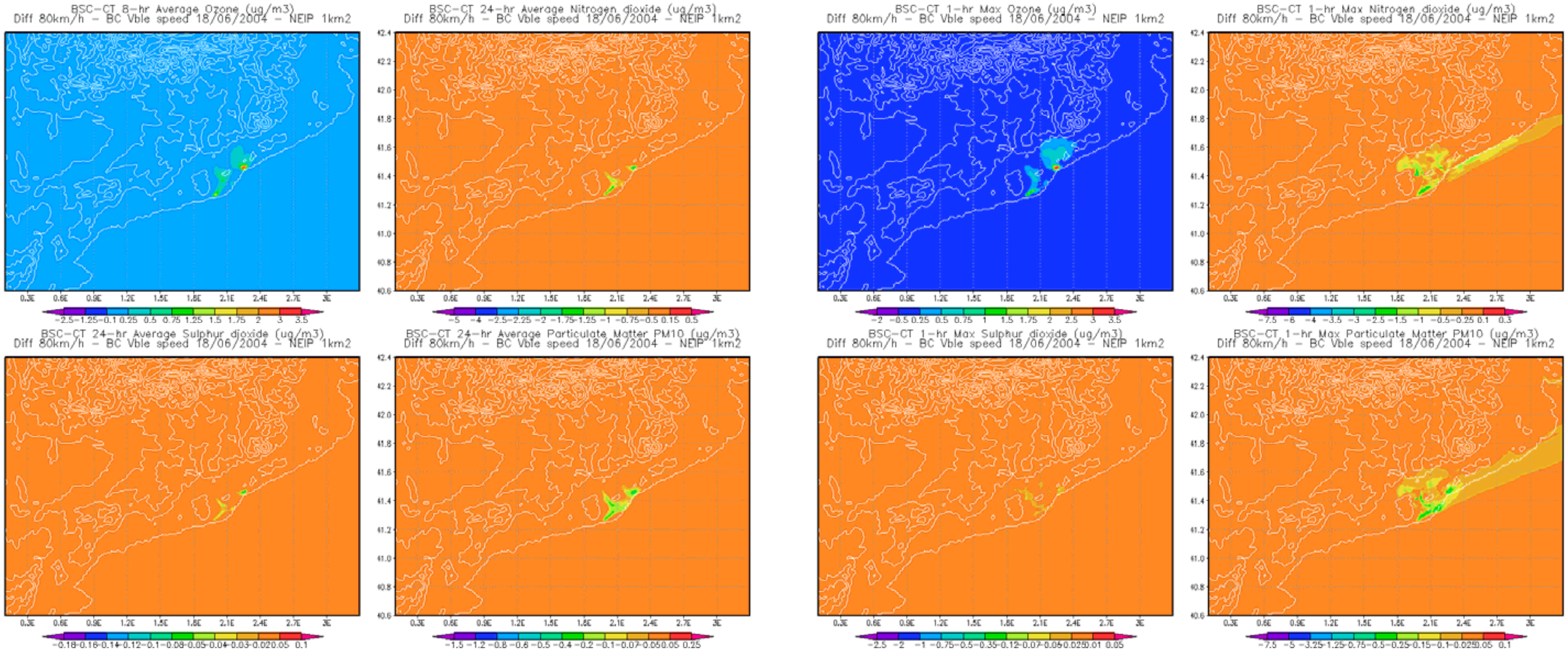
Figure 6.5. (a) 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentrations (µg m^{-3}) and (b) maximum hourly concentrations of O_3, NO_2, SO_2 and PM_{10} (µg m^{-3}) in the north eastern Iberian Peninsula domain in the base case estimated with HERMES-VS (variable speed). 18 June, 2004.



(a)

(b)

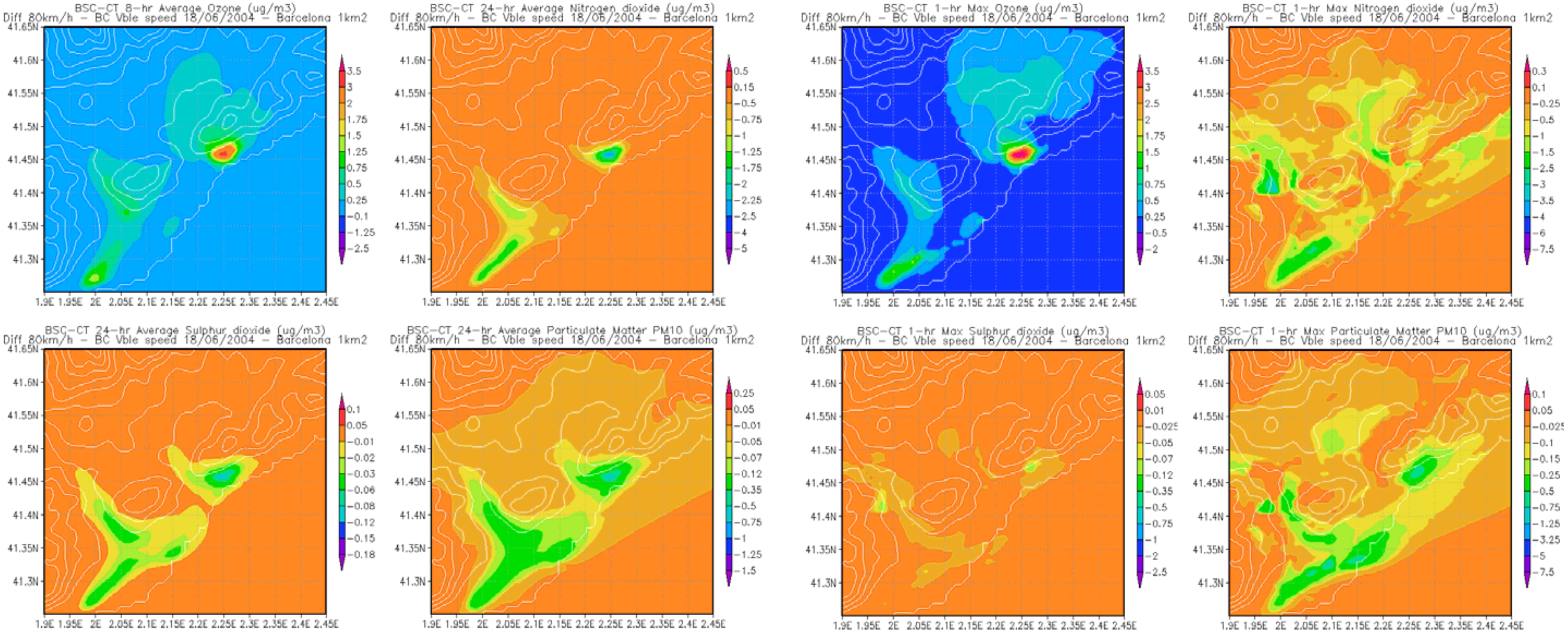
Figure 6.6. (a) 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentrations (µg m^{-3}) and (b) maximum hourly concentrations of O_3, NO_2, SO_2 and PM_{10} (µg m^{-3}) in the Barcelona area in the base case estimated with HERMES-VS (variable speed). 18 June, 2004.



(a)

(b)

Figure 6.7. Differences in (a) 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentrations (µg m^{-3}) and (b) maximum hourly concentrations of O_3, NO_2, SO_2 and PM_{10} (µg m^{-3}) in the Northeastern Iberian Peninsula when introducing the speed limit of 80 km h^{-1}.18 June, 2004..



(a)

(b)

Figure 6.8. Differences in (a) 8-hr average O_3, 24-hr average NO_2, SO_2 and PM_{10} concentrations (µg m^{-3}) and (b) maximum hourly concentrations of O_3, NO_2, SO_2 and PM_{10} (µg m^{-3}) in the Barcelona area when introducing the speed limit of 80 km h^{-1}. 18 June, 2004

The reduction on primary pollutants emissions affects the SO_2 and PM_{10} local levels over Barcelona area (Figure 6.7). The maximum hourly concentrations of SO_2 and PM_{10} are up to 0.12 µg m^{-3} and 0.75 µg m^{-3} lower than the base case values, respectively. PM_{10} is also slightly reduced in the urban plume (-0.05 µg m^{-3}, Figure 6.8). In the urban area the $PM_{2.5}$ fraction dominates the particulate matter concentration, ranging the reductions from -0.3% to -4.2% in mass (Table 6-2). The coarse fraction of particulate matter (PM_{10}-$PM_{2.5}$) is hardly affected by the introduced measure, indicating that the main reductions of PM are due to the reduction on exhaust combustion particulates and decrease of precursors' emissions.

The SO_2 levels in the urban area are relatively low compared to the legal thresholds. The maximum 24-hr average concentration occurs in the downtown area during 18 June, and reaches 20.4 µg m^{-3} (Table 6-2), which is far from the legislation threshold (125 µg m^{-3}). The speed limit restriction decreases the average concentration downtown by 0.1%. The largest reductions occur in the Dalt area (-5.3%).

The effects on studied air quality parameters concentrate over the locations of the 80 km h^{-1} limited speed roads, the largest changes being those estimated for the Dalt and the Maresme areas. The average downtown levels vary up to ±0.3%, reflecting a lighter effect in those areas not directly affected by the measure (Table 6-2).

Table 6-2 Pollutant concentrations (µg m^{-3}) in selected domains of the Barcelona area for the 17-18 June in the base case scenario (VS) and the scenario introducing speed circulation limits of 80 km h^{-1} (V80) in selected domains of the Barcelona area: BCN – Barcelona area; BCN DT – Barcelona Downtown; BCN M – Barcelona Maresme; BCN D – Barcelona Dalt (Figure 6.4).

VS: HERMES-VS: variable circulation speed based on measurement campaign.
V80: Scenario including speed limitations to 80 km h^{-1} in the Barcelona Metropolitan Area road network.
17 J: 17 June, 2004; 18 J: 18 June, 2004.

Conc (µg m^{-3})		24 hr average NO$_2$		24-hr average SO$_2$		8-hr average O$_3$		24 hr average PM$_{10}$		24 hr average PM$_{25}$	
		VS	V80	VS	V80	VS	V80	VS	V80	VS	V80
BCN	17 J	22.8	22.7	8.7	8.7	105.6	105.8	15.0	15.0	15.0	14.9
	18 J	22.9	22.7	8.4	8.4	108.1	108.3	15.9	15.8	15.8	15.8
BCN DT	17 J	74.6	74.5	20.0	20.0	55.6	55.6	29.8	29.8	29.8	29.8
	18 J	77.0	76.7	20.4	20.4	67.2	67.3	30.4	30.4	30.3	30.2
BCN M	17 J	40.5	39.5	9.6	9.5	74.2	76.0	19.6	19.4	19.5	19.3
	18 J	49.6	48.4	11.2	11.2	79.6	81.6	25.2	24.9	25.1	24.8
BCN D	17 J	34.9	34.1	11.8	11.8	93.5	93.7	16.8	16.7	16.7	16.5
	18 J	35.1	33.1	11.5	10.9	102.3	102.6	17.9	17.4	17.8	17.3
Difference between the V80 scenario and the base case (HERMES-VS)											
Conc (µg m^{-3})		24 hr average NO$_2$		24-hr average SO$_2$		8-hr average O$_3$		24-hr average PM$_{10}$		24-hr average PM$_{25}$	
		V80-VS	%	V80-VS	%	V80-VS	%	V80-VS	%	V80-VS	%
BCN	17 J	-0.2	-0.7%	-0.01	-0.1%	0.1	0.1%	-0.03	-0.2%	-0.03	-0.2%
	18 J	-0.2	-0.8%	-0.01	-0.1%	0.2	0.1%	-0.03	-0.2%	-0.03	-0.2%
BCN DT	17 J	-0.1	-0.1%	-0.01	-0.03%	0.05	0.1%	-0.04	-0.1%	-0.04	-0.1%
	18 J	-0.2	-0.3%	-0.01	-0.1%	0.1	0.2%	-0.1	-0.2%	-0.1	-0.2%
BCN M	17 J	-1.0	-2.5%	-0.04	-0.4%	1.8	2.4%	-0.2	-1.1%	-0.2	-1.1%
	18 J	-1.2	-2.3%	-0.05	-0.4%	2.0	2.5%	-0.3	-1.0%	-0.3	-1.0%
BCN D	17 J	-0.7	-2.1%	-0.02	-0.2%	0.2	0.2%	-0.1	-0.7%	-0.1	-0.7%
	18 J	-2.0	-5.7%	-0.61	-5.3%	0.3	0.2%	-0.5	-3.0%	-0.5	-3.0%

6.4 Conclusions

In developed countries different strategies are being tested and put into practice in order to abate the negative environmental effects of on-road traffic, without decreasing population mobility. State-of-the-art air quality models and computer resources allow to assess the effects of hypothetical mitigation measures in advance.

The average speed dependency of current on-road traffic emissions models predictions, such as HERMES, based on COPERT III methodology, demands for more precise and realistic representation of vehicles circulation speed. The traffic emissions account based on constant speeds does not take into account the characteristics of the road networks or the speed variability due to specific driving behaviour. Variable speed based on measurement campaigns is supposed to reduce uncertainties in emissions and air quality estimations. The HERMES emissions model was modified by introducing variable speed data, which caused reductions of the NO_x, CO, SO_2 and PM_{10} traffic emissions estimates for the Barcelona area by 5.6%, 5.1%, 4.8% and 5.1% respectively.

The variable speed introduction in the WRF-ARW/HERMES/CMAQ model locally improved the O_3 predictions (around 1% reduction on the statistical parameters estimated), even though the results differed for the NO_2, SO_2 and PM_{10} concentrations, depending on the air quality station considered. Further work must be oriented to extend these measurement campaigns in time, in order to represent the circulation patterns that could occur for at least an annual cycle. The speed representation by vehicle type might be also introduced.

The modelling system was applied to assess the effects on air quality of the 80 km h^{-1} speed restriction planned for the Barcelona Metropolitan area. The analysis of real circulation patterns shows that the traffic on some of the affected roads was still circulating at speeds around 80 km h^{-1} or lower, mitigating the effects of the management strategy. The changes in emissions estimates of the limit speed introduction are lower than those expected. The NO_x, SO_2, PM_{10}, NMVOCs and CO traffic emissions over the Barcelona area are 3.2% lower on average when considering variable speed than those estimated with the constant speed by road stretch representation, while the introduction of the speed limitation to 80 km h^{-1} causes a further reduction of only 0.6%. This may be due to both the effect of traffic congestion and the relatively small affected area, exclusively the Barcelona Metropolitan area. Consequently the effects on air quality predicted by the model focus on the Barcelona area, mainly over the affected roads, where the reductions on NO_2, SO_2 and PM_{10} levels reach up to 5.7%, 5.3% and 3.0% reductions on 24-hr average concentration, respectively. The NO_x emissions decrease in a VOCs limited area, such as Barcelona, produces local increases of O_3 concentrations, especially in the urban plume over the roads affected by the speed limit (up to 2.4%); nevertheless the O_3 concentrations in downwind areas remain practically constant. The most positive effects of the management measure is observed for $PM_{2.5}$, the most dangerous fraction for human health.

This work highlights the need for more detailed emissions inventories, specifically concerning on-road traffic, in order to improve the reliability of air quality modelling as a management tool and help decision makers. It also shows the importance of introducing more realistic parameters into the emissions models and the changes that they involve in the air quality model predictions.

7 Conclusions

7.1 General conclusions

This PhD thesis has evaluated different strategies for on-road traffic emissions abatement in terms of emissions, fuel consumption and air quality, by means of the WRF-ARW/HERMES/CMAQ modelling system. It has focused on the largest urban areas of Spain: Barcelona and Madrid, during a representative photochemical pollution episode of 2004, which fits in a usual traffic circulation pattern (correspondent to working days): the 17-18, June, 2004.

The main goals proposed for this work (see *section 1.6, Chapter 1*) were achieved. The WRF-ARW/HERMES/CMAQ model, representative of the state-of-the-art in air quality modelling, proved to be a versatile tool to assess subtle changes in urban air quality and designing detailed scenarios. It permitted to assess the effects of different urban management strategies, particularly those related to the abatement of emissions from on road traffic. It provided not only the changes in urban air quality but also those produced in downwind regions affected by the pollution plume. The highly detailed emissions inventory and the high resolution applied (1 km^2, 1hr) are the main features that allow this kind of developments. Besides, the availability of the MareNostrum Supercomputer, holded by the BSC-CNS, reduced the computational times considerably.

The on-road traffic management strategies intend to decrease emission levels by varying the circulation speed, reducing the number of vehicles or introducing alternative fuels or new technology vehicles. All these changes can be defined in detail in the HERMES emissions model, which particularly in the urban areas of Barcelona and Madrid takes into account the specific vehicle fleet composition and has a highly detailed definition of the road network and the traffic volumes.

The model proved also to be capable of defining the pollutants transport patterns for a coastal and a continental environment, the Northeastern Iberian Peninsula and Central Iberian Peninsula, which house Barcelona and Madrid urban areas, respectively. In the case of study, a low pressure gradient situation over the Iberian Peninsula, the mesoscalar flows are dominant. The model reproduces the sea-breezes regime behaviour in the coastal domain and the katabatic and anabatic winds control in the mountain areas. The contribution of atmospheric processes to gaseous pollutants concentration was quantified by means of the Integrated Process Analysis tool available in the CMAQ model, and a deep analysis of the atmospheric behaviour under the PBL was carried out; concluding that the transport by horizontal and vertical advection set the locations of the maximum pollutants concentration. The photochemical pollution formation occurs at high levels in the coastal domain (the Northeastern Iberian Peninsula), due to the recirculation patterns caused by the sea-breezes and the very complex topography. On the other hand the continental domain (the Central Iberian Peninsula) has a simpler behaviour. The pollutants dynamics are controlled by thermal phenomena, originated by surface heating by solar radiation. The Madrid pollutants plume affects the south-eastern and the eastern region due to the advected flows, being the photochemical pollution produced in the entire tropospheric column. Both locations show different dynamics of pollutants dispersion.

A revision of the most feasible strategies to reduce on-road traffic contribution to urban air pollution in the short term was carried out. It led to the definition of several scenarios including alternative fuels use, such as (1) **natural gas**, and (2) **biodiesel**; and new technology vehicles introduction, such as (3) **hybrid vehicles**. Also (4) the introduction of a **speed circulation limit** was evaluated in terms of air quality.

The proposed scenarios agree with the European Union criteria, which promote natural gas and biofuels use not only to ameliorate urban air quality conditions, but also to reduce GHG emissions and to increase the security of the energy supply. The use of hydrogen as a fuel, also promoted by the EU, is still in a development stage; therefore it was not considered as a short term option. On the other hand the hybrid vehicles constitute the first step in the path to the electric vehicles and are already being introduced in the EU markets. Finally, speed limits are currently usual in different European cities, because of being short term strategies that allow reducing pollutants emissions, but also noise and traffic accidents. Specifically the Catalonia Government has introduced a 80 km h^{-1} speed limit in the Barcelona Metropolitan area, whose effects were characterized by means of the WRF-ARW/HERMES/CMAQ model.

The strategies for the abatement of emissions show different responses in both studied areas. These are mainly due to: the specific vehicle fleet composition, the different contribution of economical or activity sectors to total emissions, the topography, meteorological conditions and the atmospheric transport, and the different chemical regime (different NO_x-VOCs ratio that directly affects O_3 production response to emissions abatement strategies). All these particularities are taken into consideration.

The HERMES emissions model is continuously improved, not only in the framework of this work, but also due to the developments that the Earth Sciences Department of the BSC-CNS carries out. To perform this study the most suitable emission factors and vehicle categories for the new vehicles tested were added. Vehicles included in the same category (weight, age, cubic capacity) were differentiated by their activity (public, private or freight transport), in base of mobility data. Also variable speed cycles from measurement campaigns in the Barcelona area were included instead of the constant speed by road stretch previously considered. Besides, during the execution of this work, emissions from ports and airports, ships and planes traffic were added to the model. These are included in the simulations of Chapters 4 to 6.

The model evaluation for the base cases was carried out separately for each version of the WRF-ARW/HERMES/CMAQ modelling system; the results are shown in the correspondent chapter. They agree with the European and US-EPA recommendations for air quality modelling to be used with regulatory purposes. Each chapter presents its own conclusions, here a summary of those related with different management strategies is provided together with a comparison among their effects.

7.2 Individual scenarios and comparison of the strategies addressed to on-road traffic emissions abatement

On-road transport is responsible for the largest amount of emitted NO_x (57% in the Northeastern Iberian Peninsula –NEIP- and 74% in the Central Iberian Peninsula –CIP-). It largely contributes to $PM_{2.5}$ emissions (32% in the NEIP and 42% in the CIP), together with power generation and industrial combustion; and NMVOCs (43% in the NEIP and 44% in the CIP), together with biogenic sources. The traffic contribution to emissions concentrates in urban areas: Barcelona and Madrid account for 21% and 46% of the total emissions of this sector in the Northeastern and Central Iberian Peninsula, respectively.

The analysis performed for the selected case of study, the 17-18 June 2004, in the Barcelona and Madrid urban areas, showed that the air quality improvement strategies should be addressed mainly to reduce NO_2 and PM ground levels. The on-road transport emissions are on the origin of the high levels of NO_2 (24-hr average concentrations above 80 µg m^{-3} in Barcelona and above 60 µg m^{-3} in Madrid) and PM_{10} (24-hr average concentrations above 45 µg m^{-3} and 35 µg m^{-3} in Barcelona and Madrid, respectively).

On-road traffic emissions also contribute to the O_3 formation in the urban plumes, resulting in high O_3 levels in downwind areas from the conurbations. Exceedances of the O_3 European air quality targets (180 µg m^{-3} for the hourly concentration and 120 µg m^{-3} for the 8-hr average concentration) are observed in these regions, and also in Madrid city during the morning (17-18 June, 2004).

Despite the SO_2 concentrations are higher in the downtown areas than outskirts, they do not involve problems related to human health. They remain in both cities below the WHO recommendations and the EU thresholds (350 µg m^{-3} for the hourly concentration). The SO_2 levels are higher in Barcelona than in Madrid, which is mainly attributed to the industrial activity of the region.

The strategies for on-road traffic emissions abatement provide a way for reducing its contribution to urban air pollution. This work evaluated several scenarios for Barcelona and Madrid, defined in under a feasible perspective and taking into account specific vehicle fleets:

(1) seven scenarios of natural gas vehicles introduction;

(2) two scenarios of biodiesel use;

(3) two scenarios of hybrid cars introduction;

(4) one scenario of an 80 km h^{-1} speed limit introduction.

These scenarios were evaluated for the Northeastern Iberian Peninsula and Central Iberian Peninsula domains, but smaller areas over the Barcelona and Madrid conurbations were defined.

The **effects in emissions** were assessed in the areas specifically affected by the scenarios:

(1) A 130 km^2 area over **Barcelona** and a 373 km^2 area over **Madrid**; which cover the urban roadways affected by the urban fleet change (natural gas, biodiesel and hybrids scenarios).

(2) The emissions budget for the 80 km h^{-1} speed limit scenario was assessed over a 2112 km^2 square covering all the **Barcelona** Metropolitan area and all the roadways where hourly speed data were introduced.

The **effects in air quality** were assessed for:

(1) The **Barcelona** (BCN) and **Madrid** (MAD) **areas** (40x40 km^2 squares centred in the conurbations),

(2) And **smaller domains** (2x2 km^2) **over zones directly affected by the scenarios** (e.g. the Barcelona-Dalt domain used in the speed limit scenario covers several roadways affected by the 80 km h^{-1} speed limit) or **over areas with high population density** (all scenarios were assessed in zones defined over downtown).

7.2.1 Natural Gas Vehicles Introduction

The use of natural gas as a fuel substituting fossil diesel or gasoline involves reductions on emissions of NO_x, NMVOCs, CO and PM, while the CH_4 emissions increase. These changes widely vary depending on the vehicle technology and the substituted fuel.

The tested scenarios included the substitution by natural gas vehicles (NGV) of: (NG1) **all the urban bus fleet**; (NG2) **50% of the oldest taxis**; (NG3) **50% of the intercity buses**; (NG4) **50% of light commercial vehicles**; (NG5) **10% of the oldest private cars**; (NG6) **all the heavy duty freight transport vehicles**; and (NG7) **combination of all the rest**. The oldest categories of petrol and diesel vehicles in the Barcelona and Madrid urban fleets were removed. The NGVs introduced mainly agree with the EEV (Enhanced Environmental Vehicle) standards.

All the tested scenarios had a positive effect on emissions from on-road traffic. The combined scenario, when up to 26% of the vehicle fleet circulating in Barcelona and up to 23% in Madrid was transformed, involved the largest reductions in ozone precursors and the rest of primary pollutants emissions (38.4 t d^{-1} in Barcelona and 98.8 t d^{-1} in Madrid). NGVs are useful to reduce SO_2 and PM emissions, especially when substituting commercial light duty vehicles.

Concerning urban air quality levels, the NGVs introduction involves a decrease in the NO_2, SO_2 and PM_{10} and an increase of O_3 concentrations. The substitution of the 100% of the urban buses, of the 50% of the intercity buses and of the 100% of HDV are negligible in terms of emissions change. None of them involves a fleet change larger than 1.5% neither in Barcelona nor in Madrid. Even considering the introduction of NGV accomplishing the EEV standards, the conventional fuel substitution has to reach certain critical values (around

4%) for being effective in the reduction of emissions. Their effects on O_3, NO_2, SO_2 and PM_{10} concentrations are below 3% in the urban areas (BCN, MAD).

The urban fleet composition of Madrid makes specifically effective the 10% of private cars substitution, which reduces NO_2 concentration up to 8.2% (MAD). The 50% of light commercial vehicles change involves reductions specifically in SO_2 and $PM_{2.5}$. Particularly in Barcelona urban area, the 24-hr average concentrations decrease by 0.6% and 4.9%, respectively. The combined scenario involves the largest air quality levels change in the urban areas, specifically in Madrid, where a reduction on NO_2 of 20.6% and PM_{10} of 14.9% is achieved. This scenario also involves the largest increase in O_3 levels, a 2.5%, which leads 8-hr average concentration in Madrid to be 88.1 $\mu g\ m^{-3}$ (below the EU target of 120 $\mu g\ m^{-3}$ for human health protection)

Collateral impacts of using NGV such as the construction of facilities for gaseous fuel supply and the possible increase in the GHG outcome by the methane emissions increase have to be considered by the policy makers.

7.2.2 Use of biodiesel as an alternative fuel

Two feasible scenarios of B20 (20% biodiesel – 80% diesel fossil blend) use are designed for Barcelona and Madrid urban fleets.

The B1 scenario considers the substitution of 10% of the oldest petrol and diesel cars by Euro III cars fuelled by B20.

The changes in emissions and air quality observed are due to the fuel substitution, around 0.9% and 1.2% of pure biodiesel introduced in Barcelona and Madrid, respectively. But also to the vehicle fleet renewal considered, which slightly reduces the total fuel consumption from the base case (from 0.5% to 0.7% lower in the urban areas), and compensates the NO_x emissions increase due to the biodiesel use. The NO_x, NMVOCs, CO and PM emissions decrease. Their effect is noticed in NO_2 concentrations, which are reduced by 1.5% and 3.7% in Barcelona (BCN) and Madrid (MAD) areas. The VOCs sensitive regime of Barcelona induces an increase of O_3 levels, around 0.2%. In Madrid the reduction of 0.4% in O_3 levels is not enough to accomplish the European targets (120 $\mu g\ m^{-3}$)

The B1 scenario effect in $PM_{2.5}$ is positive both in Barcelona (BCN) and Madrid (MAD), being the reductions lower in the former (0.3% lower face to 1.2%) due to the vehicle fleet composition (less cars proportion than in Madrid) and the other emission sectors that contribute to the PM outcome.

The SO_2 emissions increase due to the sulphur content in the diesel fossil introduced in the new cars, nevertheless its impact on average SO_2 concentrations in the conurbations is not appreciable: 0.01% increase in Barcelona (BCN) and 0.11% in Madrid (MAD) and remaining the final concentrations at low levels.

The B2 scenario considers the effect of fuelling with B20 all the diesel vehicles in Barcelona and Madrid urban areas, without changing the vehicles typology. The fuel substitution estimated is comparable to the 2020 objective set by the European Union (10%).

The NMVOCs, CO, SO_2 and PM emissions decrease in the urban areas, while a slight increase in NO_x emissions is observed. The NO_x emissions increase in Barcelona is moduled by tropospheric chemistry and does not affect the NO_2 levels but in small areas over the conurbation. Moreover, a slight increase in O_3 levels occurs. The on-road traffic of Madrid emits more NO_x than in the base case, which increase average NO_2 levels by 0.3%. The O_3 response to this scenario is a slight reduction (by 0.03%, in MAD domain), which does not represent an appreciable change in final levels.

The deepest impact of this scenario concerns $PM_{2.5}$ concentration, which is reduced by 3.1% and 6.0% in Barcelona and Madrid downtown, respectively. The SO_2 levels are slightly reduced, in Barcelona (0.3% reduction - BCN) less than in Madrid (1.8% - MAD), due to the presence of other sources different from traffic contributing to this pollutant emissions.

7.2.3 Hybrid cars introduction

The introduction of 10% of gasoline-electric hybrid cars instead of the oldest petrol and diesel cars (H1) and the substitution of 30% of the oldest petrol and diesel cars, including taxis, by gasoline-electric hybrid cars (H2) are tested for Barcelona and Madrid.

The overall effect in both cities involves the reduction on NO_2 and $PM_{2.5}$ levels, which is larger in the H2 scenario. The daily NO_2 and $PM_{2.5}$ concentrations are on average 5.7% and 0.3% lower in Barcelona (BCN) and 23.8% and 3.6% lower in Madrid (MAD), respectively. In addition it reduces the SO_2 emissions in 0.05 t d^{-1}, which causes the daily average concentrations of this pollutant decreasing just 0.2% in the area of Barcelona (BCN). The effects of the same scenario in Madrid (MAD) are notably higher, with SO_2 daily concentration reductions of 1.3%. The O_3 concentrations in the conurbations locally increase, because of the limitation of the O_3 titration by fresh NO_x emissions. Nevertheless, the photochemical regime involves this increase being more important in Barcelona, while in Madrid the O_3 levels during the central hours of the day are reduced. In fact, the maximum concentrations downtown are lower in the H1 and H2 scenarios than in the base case. The introduction of hybrid cars in the urban areas (both H1 and H2) has positive effects in downwind areas, decreasing specially the NO_2, $PM_{2.5}$ and O_3 levels.

The implications of the on-road traffic sector not only concern local or regional air quality levels, but also greenhouse gases emissions. The equivalent CO_2 emitted by on-road traffic is reduced in 0.4% and 1.6% in the Northeastern Iberian Peninsula when introducing H1 and H2, respectively. In the Central Iberian Peninsula the equivalent CO_2 from on-road traffic decreases 1.4% (H1) and 4.9% (H2), reflecting the larger fossil fuel savings in this region. This work is centred in the vehicle use phase, the benefits of this technology in terms of GHG emissions may be confirmed over the whole life cycle of the vehicles.

7.2.4 Speed variation. 80 km h^{-1} speed restriction in the Barcelona area

The introduction of an 80 km h^{-1} speed limit in the Barcelona area planned by the Catalonia Government was assessed in terms of air quality (V80).

The speed dependency of current on-road traffic emissions models predictions, such as HERMES, demands for more precise and realistic representation of vehicles circulation speed. The traffic emissions account based on constant average speeds do not take into account the characteristics of the road networks speed variability due to specific driving behaviour. Variable speed based on measurement campaigns is supposed to reduce uncertainties in emissions and air quality estimations. The HERMES emissions model was modified by introducing variable speed data. This change locally improved the O_3 predictions (around 1% reduction on the statistical parameters estimated), even though the results differed for the NO_2, SO_2 and PM_{10} concentrations, depending on the air quality station considered.

The analysis of measured circulation patterns shows that the traffic on some of the affected roads was still circulating at speeds around 80 km h^{-1} or lower, mitigating the effects of the 80 km h^{-1} limit management strategy.

The changes in emissions estimates of the limit speed introduction are lower than those expected. The NO_x, SO_2, PM_{10}, NMVOCs and CO traffic emissions over the Barcelona area are 3.2% lower on average when considering variable speed than those estimated with the constant speed by road stretch representation, while the introduction of the speed limitation to 80 km h^{-1} causes a further reduction of only 0.6%. This may be due to both the effect of traffic saturation and the relatively small affected area, exclusively the Barcelona Metropolitan area.

Consequently the effects on air quality predicted by the model focus on the Barcelona area, mainly over the affected roads, where the reductions on NO_2, SO_2 and PM_{10} levels reach up to 5.7%, 5.3% and 3.0% reductions on 24-hr average concentration, respectively. The NO_x emissions decrease in a VOCs limited area, such as Barcelona, produces local increases of O_3 concentrations, especially in the urban plume over the roads affected by the speed limit (up to 2.4%); nevertheless the O_3 concentrations in downwind areas remain practically constant. The most positive effects of the management measure is observed for $PM_{2.5}$, the most dangerous fraction for human health.

The hourly speed data included to assess this scenario do not take into account possible changes in congestion when introducing the 80 km h^{-1} speed limit. An increase in circulation speed during the traffic peaks due to the reduction of congestion could provide further improvements in air quality levels (due to the U shaped functions of main emission factors). A conservative approach was taken into account in the implementation of this scenario and the effect of congestion reduction was not introduced in the model intending not to manipulate the available speed data. Measurement campaigns during the speed limit operation would be necessary to improve this aspect.

7.2.5 Comparison of the strategies for on-road traffic emissions abatement and concluding remarks

All the strategies evaluated for on-road traffic emissions abatement produced reductions on primary pollutants emissions (NO_x, SO_2, NMVOCs and PM). Table 7-1 shows the changes in traffic emissions for comparable scenarios: the substitution of 10% of the oldest private cars by natural gas cars (NG5), B20 Euro III fuelled cars (B1) or hybrid cars (H1) in Barcelona (BCN) and Madrid (MAD). The effects of introducing the 80 km h^{-1} speed limit (V80) is also included for the Barcelona area. These are comparable in absolute terms (not in relative values due to the updates in the HERMES model).

The largest reductions on NO_x and $PM_{2.5}$ emissions are attributed to hybrid electric cars and natural gas vehicles use. The use of 10% of natural gas cars in Madrid instead of the oldest petrol and diesel cars would reduce the NO_x emissions in 7.3 t d^{-1}, if the substitution was by petrol hybrid electric cars the reduction would be of 7.6 t d^{-1}. These scenarios would save 0.4 t d^{-1} of $PM_{2.5}$ in Madrid. The vehicle fleet composition of Barcelona (lower percentage of private cars) reduces the impact of these scenarios (the NG5 scenario emits 1.9 t d^{-1} of NO_x and 0.1 t d^{-1} of $PM_{2.5}$ less than the base case). The NO_x emissions of biodiesel fuelled vehicles are slightly higher than those of the correspondent diesel fossil vehicles; nevertheless the fleet renewal compensates this fact in the shown scenario (Table 7-1).

The main advantage of B20 fuelled cars use is the reduction of $PM_{2.5}$ and SO_2 emissions when compared to diesel fossil vehicles (note that the B1 scenario accounts for more SO_2 emissions than the base case, this is due to the substitution of petrol cars by B20 fuelled cars, which are feed in 80% with diesel fossil).

Table 7-1. On road traffic emissions change attributed to selected scenarios. BC: base case, NG5: introduction of 10% of natural gas cars; B1: introduction of 10% of Euro III B20 fuelled cars; and H1 introduction of 10% of petrol hybrid electric cars, instead of the oldest diesel and petrol cars in Barcelona (BCN) and Madrid (MAD).V80: introduction of an 80 km h^{-1} speed limit in the Barcelona Metropolitan area.

	On road traffic emissions change t d^{-1} (%)							
	NOx		NMVOCs		$PM_{2.5}$		SO_2	
	BCN	MAD	BCN	MAD	BCN	MAD	BCN	MAD
BC-NG5	1.9 (7.8%)	7.3 (10.9%)	2.4 (3.3%)	6.6 (6.9%)	0.10 (5.2%)	0.39 (9.3%)	0.03 (3.9%)	0.1 (5.9%)
BC-B1	1.2 (5.5%)	5.4 (8.1%)	2.5 (2.8%)	6.5 (4.8%)	0.06 (3.7%)	0.21 (5.0%)	-0.01 (-0.8%)	-0.02 (-0.9%)
BC-H1	1.7 (7.9%)	7.6 (11.4%)	2.3 (2.6%)	6.2 (4.6%)	0.09 (5.3%)	0.40 (9.5%)	0.02 (2.6%)	0.07 (3.8%)
BC-V80 *	0.7 (5.7%)		-0.03 (-0.01%)		0.05 (1.1%)		0.02 (0.9%)	

(*) The selected area to estimate the emissions is not the same as in the other cases and the speed representation in the base case also differs, therefore relative values (%) are not comparable – see Figure 3.6 on Chapter 3 and Figure 6.1 on Chapter 6, for details.

They have positive effects on NMVOCs emissions (emitting 6.5 t d^{-1} less than in the base case in Madrid and 2.5 t d^{-1} in Barcelona), which also considerably decrease in the natural gas and hybrid vehicles scenarios (6.6 and 6.2 t d^{-1} saved in Madrid; and 2.4 and 2.3 t d^{-1} in Barcelona, respectively). Nevertheless, in the case of natural gas use the increase of CH_4 emissions is an associated impact to consider. The introduction of an 80 km h^{-1} speed limit promotes the reduction of NO_x and $PM_{2.5}$ emissions in 0.7 t d^{-1} and 0.05 t d^{-1}, respectively. The emission factors dependence of the circulation speed varies as a function of the specific type of vehicle, but also on the pollutant considered, not existing an optimal circulation speed in common for all of them, being the NMVOCs emissions practically invariable. The emissions model improvement with the inclusion of hourly speeds for the Barcelona area showed a decrease on the expected emissions change, nevertheless this scenario is in the same order of magnitude for $PM_{2.5}$ and SO_2 emissions reductions than the changes produced by the fleet renewal and introduction of B20 in 10% of private cars in Barcelona.

Considering the same vehicle fleet change for different technologies or alternative fuels can provide a measure of the effectiveness of each of them in abating urban air pollution. As aforementioned during this discussion the comparative results for this study have to be analyzed carefully, because during the elaboration of this work the emissions inventory evolved and the versions used to different scenarios analysis were not identical. Assuming that ports and airports emissions were not included in the natural gas vehicles scenarios, a comparative summary for the 10% of the oldest petrol and diesel cars substitution in Barcelona and Madrid areas is shown in Table 7-2 (corresponding to the traffic emissions changes detailed in Table 7-1).

The natural gas cars introduction could provide a feasible way for reducing PM_{10} and SO_2 levels in the urban areas (up to 1.4% and 0.7% reductions estimated for Madrid levels), moreover the hybrid cars are particularly useful to reduce PM_{10} concentrations in Madrid area (1.4% reductions).

The largest reductions on NO_2 levels occur in Madrid when introducing hybrid cars (5.3%), although natural gas cars use could provide almost the same effects (5.2%). The particularities of the vehicle fleet composition and contribution of other emission sources involves that the same scenario in Barcelona only reduces the NO_2 levels by 1.8%.

The O_3 concentration behaviour also depends on the study area, it is characterized by a reduction in daytime Madrid levels, but an increase in Barcelona, due to the lower presence of fresh NO_x that acts as a local O_3 sink and the different chemical sensitivity of the study areas, which influences the O_3 response. The variations in the urban areas are below 1.0% for all scenarios (Table 7-2), and the effect downwind proved to be positive in terms of O_3 concentrations.

Finally, the introduction of a 80 km h^{-1} speed limit is positive in the Barcelona urban area, but the overall effect is lower than the 10% cars substitution aforementioned. Although comparable reductions in daily average concentrations are observed for PM_{10}, being in average 0.2% lower than in the base case. The speed limit effects are deeper in these areas specifically affected by the management measure.

Table 7-2. Average concentration variation in the Barcelona and Madrid urban areas (40x40 km^2 domains defined; see *Chapter 3* for details) attributed to selected scenarios. BC: base case, NG5: introduction of 10% of natural gas cars; B1: introduction of 10% of Euro III B20 fuelled cars; and H1 introduction of 10% of petrol hybrid electric cars, instead of the oldest diesel and petrol cars in Barcelona (BCN) and Madrid (MAD).V80: introduction of an 80 km h^{-1} speed limit in the Barcelona Metropolitan area.

		8-h ave. O_3			24-h ave. NO_2		
		BC (µg m^{-3})	Diff (µg m^{-3})	(%)	BC (µg m^{-3})	Diff (µg m^{-3})	(%)
BCN	NG5	102.5	-0.14	-0.14%	25.1	0.26	1.05%
	BD1	107.7	-0.25	-0.23%	23.0	0.34	1.47%
	H1	107.7	-0.34	-0.32%	23.0	0.42	1.81%
	V80	107.6	-0.16	-0.15%	22.9	0.19	0.82%
		24-h ave. PM_{10}			24-h ave. SO_2		
		BC (µg m^{-3})	Diff (µg m^{-3})	(%)	BC (µg m^{-3})	Diff (µg m^{-3})	(%)
BCN	NG5	15.3	0.05	0.33%	8.17	0.008	0.09%
	BD1	15.9	0.04	0.23%	8.39	-0.001	-0.01%
	H1	15.9	0.03	0.20%	8.39	0.005	0.06%
	V80	15.9	0.03	0.21%	8.39	0.006	0.07%
		8-h ave. O_3			24-h ave. NO_2		
		BC (µg m^{-3})	Diff (µg m^{-3})	(%)	BC (µg m^{-3})	Diff (µg m^{-3})	(%)
MAD	NG5	126.8	0.45	0.35%	17.0	0.88	5.19%
	BD1	127.0	0.52	0.41%	17.3	0.64	3.68%
	H1	127.0	0.41	0.32%	17.3	0.91	5.26%
		24-h ave. PM_{10}			24-h ave. SO_2		
		BC (µg m^{-3})	Diff (µg m^{-3})	(%)	BC (µg m^{-3})	Diff (µg m^{-3})	(%)
MAD	NG5	10.1	0.14	1.39%	2.85	0.02	0.75%
	BD1	10.2	0.08	0.76%	2.67	-0.003	-0.11%
	H1	10.2	0.14	1.40%	2.67	0.01	0.52%

The changes in concentration are geographically distributed in a heterogeneous way. The **selection of the adequate areas for the analysis** of the air quality changes is a **fundamental step** in this kind of studies, and it has to be carefully considered. Along this work several small areas (4 km^2) were defined at some specific points of the conurbations, to provide air quality improvements in city centres or in those areas mostly affected by the measures. This analysis was possible thanks to the high resolution employed and provided interesting conclusions.

Table 7-3. Average concentration variation in the Barcelona and Madrid downtown areas (2x2 km^2 domains defined, see *Chapter 3* for details) attributed to selected scenarios. BC: base case, NG5: introduction of 10% of natural gas cars; B1: introduction of 10% of Euro III B20 fuelled cars; and H1 introduction of 10% of petrol hybrid electric cars, instead of the oldest diesel and petrol cars in Barcelona downtown (BCN-DT) and Madrid downtown (MAD-DT).V80: introduction of an 80 km h^{-1} speed limit in the Barcelona Metropolitan area.

		8-h ave. O_3			24-h ave. NO_2		
		BC (µg m^{-3})	Diff (µg m^{-3})	(%)	BC (µg m^{-3})	Diff (µg m^{-3})	(%)
BCN – DT	NG5	70.7	-1.8	-2.53%	75.4	1.9	2.49%
	BD1	64.7	-2.0	-3.15%	77.6	1.8	2.36%
	H1	64.7	-2.7	-4.13%	77.6	2.4	3.05%
	V80	67.2	-0.1	-0.18%	77.0	0.2	0.31%

		24-h ave. PM_{10}			24-h ave. SO_2		
		BC (µg m^{-3})	Diff (µg m^{-3})	(%)	BC (µg m^{-3})	Diff (µg m^{-3})	(%)
BCN – DT	NG5	28.6	0.5	1.79%	18.7	0.08	0.41%
	BD1	30.4	0.4	1.21%	20.4	-0.05	-0.25%
	H1	30.4	0.4	1.17%	20.4	0.05	0.26%
	V80	30.4	0.1	0.21%	20.4	0.01	0.06%

		8-h ave. O_3			24-h ave. NO_2		
		BC (µg m^{-3})	Diff (µg m^{-3})	(%)	BC (µg m^{-3})	Diff (µg m^{-3})	(%)
MAD – DT	NG5	121.6	-0.5	-0.39%	34.3	1.4	4.04%
	BD1	121.7	0.3	0.21%	34.7	1.8	5.24%
	H1	121.7	-0.2	-0.14%	34.7	2.6	7.51%

		24-h ave. PM_{10}			24-h ave. SO_2		
		BC (µg m^{-3})	Diff (µg m^{-3})	(%)	BC (µg m^{-3})	Diff (µg m^{-3})	(%)
MAD – DT	NG5	12.6	0.7	5.56%	2.8	0.1	2.63%
	BD1	12.6	0.3	2.26%	2.4	0.0	-0.47%
	H1	12.6	0.5	3.78%	2.4	0.0	1.95%

Analyzing the results in Table 7-2 we can conclude that none of the tested scenarios (10% of private cars change or 80 kmh^{-1} speed limit) reduces the concentration pollutants above 6% in the greater areas (40x40 km^2 defined domains). To obtain larger effects in average air quality levels over urban areas, deeper changes in the vehicle fleets or traffic volumes have to be considered.

Nevertheless, the changes in specific areas can be noticeable even with a relatively low vehicle fleet change, for example those achieved in the downtown of Barcelona and Madrid (4 km^2 defined domains) for the

10% of private cars substitution are shown in Table 7-3, together with the effect of the 80 km h^{-1} speed limit in Barcelona. The NO_2 and PM_{10} levels change in the downtown areas reflects noticeable improvements by the use of those strategies for the abatement of on-road transport emissions. For example the use of hybrid cars reduces by 3.5% the NO_2 average concentration in Barcelona downtown and by 7.5% in Madrid downtown. And the maximum reductions in PM_{10} levels in both city centres are achieved with the natural gas cars use, which provide 1.8% and 5.6% lower concentrations (in Barcelona and Madrid, respectively). In addition, when considering the 80 km h^{-1} speed limit introduction in the Barcelona Metropolitan area, the air quality levels downtown slightly change, but as shown in previous sections of this document (see *Chapter 6* for details), its effects are noticeable in areas closer to the affected roads.

All the proposed scenarios were designed under a feasible perspective, considering current plans and policies or market trends as a reference. These considerations lead to the definition of different vehicle fleet changes and different percentage of fuel substitutions. Their effects in urban air quality were positive in general terms, but the extent of these changes limited. A brief comparison of the effects for all scenarios can be found below.

Madrid proved to be a more favourable scenario to introduce strategies for on-road traffic emissions abatement than Barcelona, where other emission sources must be controlled in addition, especially when referring to the PM_{10} and SO_2 levels. All tested scenarios for the Barcelona area show reductions on PM_{10} levels lower than 4% and on SO_2 lower than 1%. The largest variation occurs when changing 50% of the oldest commercial vehicles by natural gas vehicles (0.65% lower SO_2 concentration and 3.99% lower PM_{10}).

The NO_2 levels are more sensitive to changes in traffic emissions, i.e. they decrease by 5.7% when introducing 30% of hybrid electric cars in the Barcelona area. The effect of NO_x emissions abatement in Barcelona is negative in terms of local O_3 concentration; nevertheless these increases in average concentrations are always below 1% (occurring a 0.7% increase when changing 30% of the oldest cars by hybrid electric cars). Except for O_3, the levels of pollutants are higher in Barcelona than in Madrid, so that it becomes essential to introduce air quality management strategies in this area. These control strategies should be addressed not only to on-road transport, but also to power generation and industrial sources.

The response to the tested scenarios of Madrid air quality levels is much deeper, especially when referring to NO_2 concentrations. The 30% of the oldest petrol and diesel cars substitution by hybrid cars could reduce by 23.8% the NO_2 levels in the urban area, and changing 10% of the private cars by natural gas cars provides 8.2% reductions.

The abatement of SO_2 and PM_{10} levels could be addressed both by introducing natural gas vehicles as commercial vehicles (substituting 50% of the oldest commercial vehicles by NGV reduced the SO_2 concentrations by 2.3% and the PM_{10} by 6.2%) or by using B20 blends in the whole diesel fleet (which provides SO_2 levels 1.8% and PM_{10} levels 7.6% lower than the base case scenario).

The O_3 response to emissions change depends on the NO_x-VOCs ratio reduction. O_3 levels could increase when introducing natural gas vehicles in the conurbation, but the estimated change is lower than 1%. When introducing hybrid cars slight reductions on local O_3 concentrations are observed (up to 1.2% when changing 30% of cars), these reductions occur mainly during the maximum photochemical activity hours (midday).

The response to the scenarios introduction could be locally more important. The deepest changes occur in these areas specifically affected by the strategy, i.e. in case of introducing a 80 km h^{-1} speed limit in Barcelona, the effect in downtown air quality levels is below 1.0%, but in the areas over the affected roadways the NO_2 concentration could be reduced by 5.7%, the SO_2 by 5.3% and $PM_{2.5}$ by 3.0% (see Barcelona-Dalt domain definition on Chapter 6 for details).

In conclusion:

- ✓ This PhD thesis proves that there is not a unique solution that permits to achieve acceptable air quality parameters in the urban areas in the short term. Combinations of different measures, both technical and of urban planning have to be taken in consideration.
- ✓ The selection of the areas for the assessment of the strategies is a fundamental step of their analysis. The heterogeneous geographical distribution of their effects in air quality could lead to distorted interpretations. There is not a stablished methodology for this selection, but considering those areas inside the cities with the highest population density, the poorest air quality conditions or those mostly affected by the measures may be right choices.
- ✓ The alternative fuels use provides a feasible way to reduce on-road traffic emissions, but normally involves associated impacts. In the case of natural gas the increase of CH_4 emissions and in the case of biodiesel the increase of NO_x emissions are those directly related with air quality, but other impacts such as new infrastructures construction in case of gaseous fuels supply or land use changes in case of biofuels have to be considered. Even the hydrogen used as a fuel is not exempt of this kind of associated impacts (e.g. emissions and waste in the power plants or hydrogen production sites and those derived from the fuel transport).
- ✓ The strategies to reduce fuel consumption do not risk of increasing specific pollutants emissions, among them the hybrid vehicles promotion, the urban fleet renewal and the circulation speed limits are recommendable. Nevertheless the air quality changes that can be achieved with this kind of strategies in the short term are limited and none of them involves zero emissions.
- ✓ A revision of the current urban mobility parameters have to be addressed by the decision makers: the reduction of the km travelled and/or the number of vehicles circulating could be the key for reducing pollutant emissions without associated impacts.

- ✓ Air quality modelling proves to be the most suitable tool to assess the effects of these strategies in advance. The eulerian three-dimensional mesoscalar models applied with high spatial resolution are the preferable option for this kind of developments. It is essential to obtain the information geographically distributed with the adequate resolution to distinguish areas inside a conurbation: downtown, outskirts, etc. Moreover the mesoscalar models provide the effect of the urban pollution plume in downwind areas, which it is not possible with other high resolution models such as street canyon.

- ✓ The interpretation of results has to consider the hot spots inside the conurbation: densely populated areas and areas directly affected by the measure, but also the picture in a regional scale, because the effects of urban management strategies could spread hundreds of km away.

- ✓ The emissions characterization is a key factor to provide the effect of activity changes in air quality. The HERMES model has a highly detailed description of the road network, traffic volumes and vehicle fleet composition of both Barcelona and Madrid, which constitute fundamental data for the scenarios design and assessment. It has also the capacity of distinguishing vehicles by their activity (private, public or freight transport) which permits to design scenarios in a realistic way. Moreover this kind of models can be continuously improved. The increase in the capacity for information storage and processing provides a way for representations closer to reality, such as the introduction of hourly speed data in some roadways of the Barcelona area.

- ✓ The introduction of new scenarios involves the addition of new categories of vehicles: alternative fuels or new technology vehicles, which have associated new emission factors. These are nowadays an important source of uncertainty, as shown along this dissertation. There is a need for more experimental data concerning emissions for alternative fuels and new technology vehicles, although the important efforts done in this field during the last decade. I.e. the EMEP-CORINAIR methodology published in the year 2007 provides emission factors for biodiesel use or hybrid vehicles, which were not included in the previous version of the year 2003.

- ✓ Each strategy must be assessed for the specific area of application, not being possible the extrapolation of results. Factors such as the vehicle fleet composition, the road network characteristics, the emission sources in the region, the atmospheric circulation patterns, the meteorological conditions or even the chemical sensitivity regime affect the response of the management strategy applied.

- ✓ In Barcelona, the SO_2 and PM_{10} levels improvement should be addressed not only under an on-road traffic management perspective, but also considering industrial sources and power generation plants located in the surrounding areas. The strategies addressed to on-road traffic emissions abatement could be useful to reduce NO_x levels, nevertheless an increase in local O_3 levels have to be expected. On the contrary the O_3 concentrations in those regions mostly affected by the Barcelona urban plume could be reduced by these strategies.

- ✓ The urban pollution levels in Madrid are highly dependant on on-road traffic emissions, therefore the strategies addressed to on-road transport emissions abatement would be effective, not only to ameliorate the urban air quality, but also that registered in downwind areas. The O_3 levels could be reduced in the conurbation by the NO_x emissions abatement, specifically during the maximum photochemical activity areas, which is especially interesting due to the high O_3 levels registered in the conurbation.
- ✓ The quantitative assessment of different strategies for on-road traffic emissions abatement is possible thanks to the use of atmospheric modelling and provides a helpful and objective management tool for decision makers.

7.3 Future works and development

The WRF-ARW/HERMES/CMAQ modelling system proves to be a suitable tool for the management and assessment of urban air quality especially when applied with high spatial and temporal resolution. The high resolution HERMES emission model permits to assess emission variation scenarios based in real changes of application and it provides the base to design scenarios by changing the vehicle fleet composition in urban areas of the Iberian Peninsula.

The available information about emission factors for new technology vehicles or alternative fuels is sparse. Efforts may be done in a future in order to enlarge current databases and provide speed dependant emission factors. These would necessarily have to reflect the changes in vehicles fleet composition that are already taking place, especially in urban areas. The use of correction factors, such as those used in this work for estimating the natural gas or biodiesel emissions are an alternative, but they do not constitute the ideal solution. There is an important gap in the currently available literature, which needs for more experimental tests concerning these new developed vehicles, in order to provide accurate emission factors for different types of vehicles (ideally divided by weight, age and cubic capacity of the engine) and in different driving conditions (urban, road and highway). Speciation factors, for NO_x, VOCs and PM emissions will be also required. They constitute an important issue to accurately assess final concentrations of pollutants.

On the other hand, this work highlights the need for more detailed emissions inventories, specifically concerning on-road traffic, in order to improve the reliability of air quality modelling as a management tool and help decision makers. It shows the importance of introducing more realistic parameters into the emissions models, such as the variable speed by road stretch taken from measurement campaigns. This kind of information may be included in the model in an hourly basis for an annual cycle, or at least for one type-day (working day-holiday) by month.

The $PM_{2.5}$ of anthropogenic origin is directly related with combustion processes. On road traffic is an important source of organic carbon and elemental carbon emissions. This work frequently assessed the PM_{10} fraction of particulate matter, because currently most of the air quality stations provide hourly concentrations of total suspended particulate matter and PM_{10}. The model evaluation in the base case is fundamental to provide information about the accuracy of the results. The availability of $PM_{2.5}$ concentrations and PM fractions divided by composition (sulphate, nitrate, ammonia, elemental carbon and organic carbon) could be useful to evaluate the base case concentrations predictions. Nowadays, efforts are being done in the $PM_{2.5}$ characterization.

This work focuses on an episodic event corresponding to a western recirculation in the synoptic scale. It is related with high pollutants levels in the study areas and representative of the most frequent situations of photochemical pollution. This permitted us to depict the effects of strategies for on-road traffic emissions abatement in the worst case scenario. Nevertheless, the work could be extended to define their consequences in an annual cycle by the selection of a representative episode for each synoptic situation. The application of the Integrated Process Rate analysis for each of these situations may provide a quantitative description of the atmospheric chemistry and dynamics and could be helpful to detect if the model is behaving well.

Future scenarios can be planned and assessed, such as the hydrogen fuel cells vehicles introduction, or a combination of different measures, which is probably a more realistic scenario than setting individual impacts.

This work focuses on the use phase of vehicles which is directly related with urban air quality issues. The urban traffic sector is known to have also implications on climate change and transboundary air pollution. The greenhouse gases outcome was carried out when possible; nevertheless to effectively asses the climate implications of the new technologies and alternative fuels introduction in urban areas the whole life cycle may be considered. The integrated assessment models may be also useful to provide the associated costs or human health effects of this kind of initiatives, but their results have to be considered carefully and to be subject to a deep revision of each step to reduce the related uncertainties.

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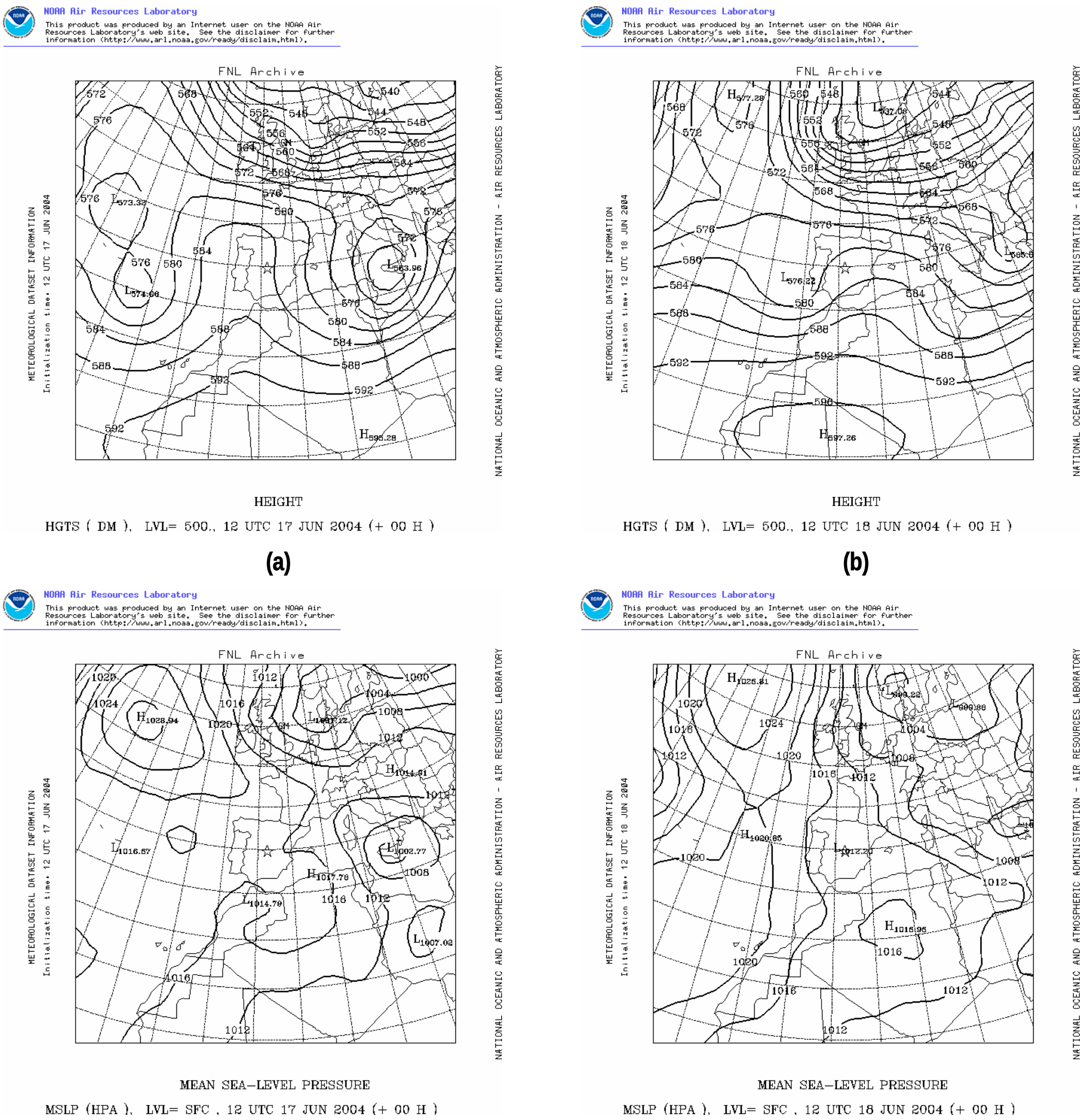
A. Annex 1. Air quality and meteorological conditions in the North-eastern and Central Iberian Peninsula during the 17-18 June, 2004.

Index of figures

This study focuses on the largest urban areas of Spain: Barcelona and Madrid (see location in Figure A.1). These are frequently affected by photochemical pollution episodes during summertime. To perform the simulations a critical episode of photochemical pollution was selected according to air quality data monitored in the study areas. The second imposed criteria was the selection of working days in order to avoid distortions of the traffic circulation patterns, necessary to provide representative results in the traffic management strategies analysis.

Figure A.1 Location of Barcelona (1) and Madrid (2) greater areas.

The chosen episode of 17-18 June, 2004 corresponds to a typical summertime low-pressure gradient with very high levels of photochemical pollutants (especially O_3 and PM_{10}) over the entire Iberian Peninsula. These days were characterized by a weak synoptic forcing, dominated by a western recirculation (Figure A.2), so that mesoscale phenomena, induced by the particular topography of the regions, may be expected to be dominant. A high sea-level pressure and negligible surface pressure gradients over the domain characterize this episode, with low northwesterlies aloft. The large solar radiation and the low cloudiness (Figure A.3) promote the development of mesoscale phenomena like mountain winds, sea and land breezes in coastal areas, and the development of the Iberian thermal low in the centre of the Iberian Peninsula.



(a)

(b)

(c)

(d)

Figure A.2 Meteorological analysis of the synoptic situation of 17-18 June, 2004. a) 500 hPa geopotential height at 12.00 UTC 17 June. b) 500 hPa geopotential height at 12.00 UTC 18 June; c) mean sea level pressure at 12.00 UTC 17 June; d) mean sea-level pressure at 12.00 UTC 18 June.

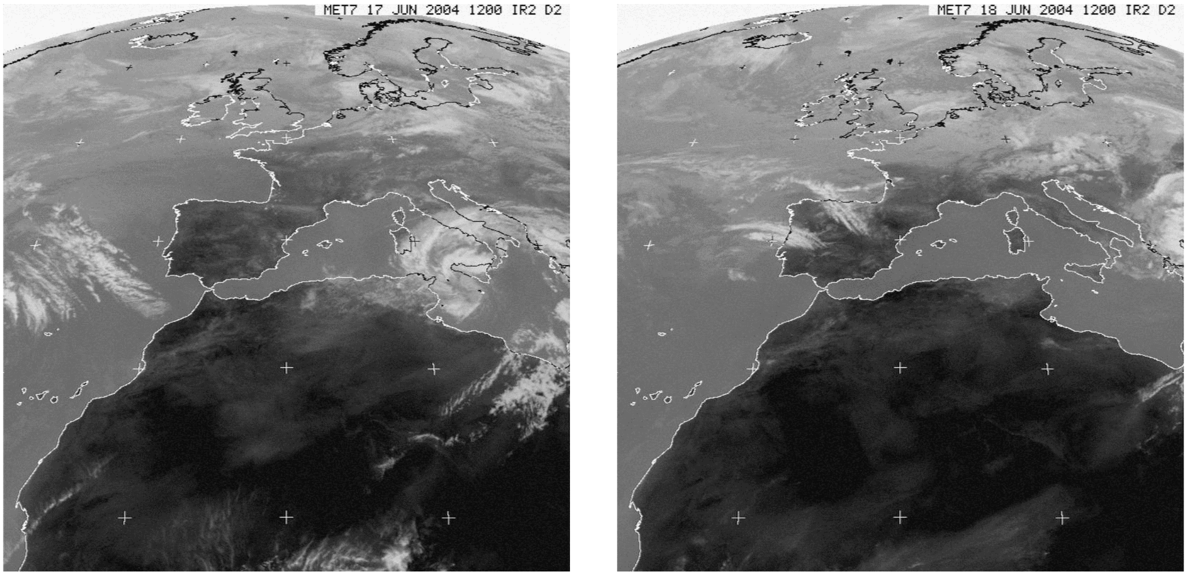


Figure A.3. Meteosat IR image for the 17 June (a) and the 18 June (b) 2004.

The WRF-ARW meteorological model provides the necessary inputs for the air quality simulations in the study areas. The surface flows in both domains are characterized by thermal origin circulations. In the NEIP the daily and nocturnal patterns differ. Being during the night dominant the valley and katabatic winds, with light land breezes in the coastal area (Figure A.4). They produce NE and SE drainages by the main valleys.

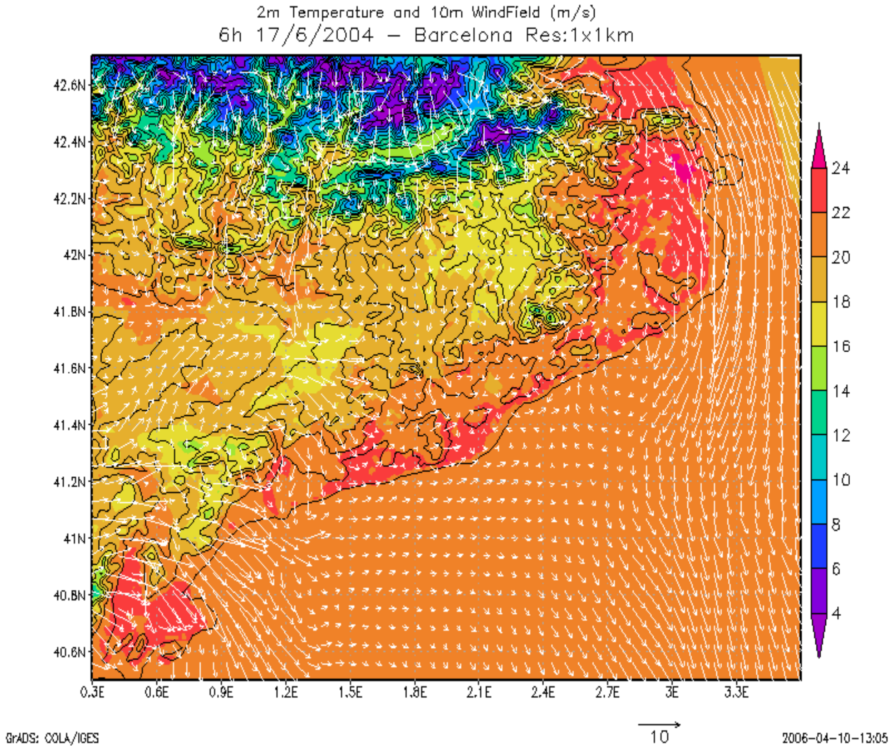


Figure A.4. Nocturnal wind flows and surface temperature (6.00UTC-17 June, 2004) in the NEIP domain.

During daytime the sea-breezes development controls coastal surface flows, inland the anabatic and valley winds develop. Along the day the sea-breezes penetrate inland, channelled by the river valleys, until 18.00-19.00 UTC when the wind starts shifting to turn into the night-time regime. The daytime surface temperature overpasses 28ºC (Figure A.5)

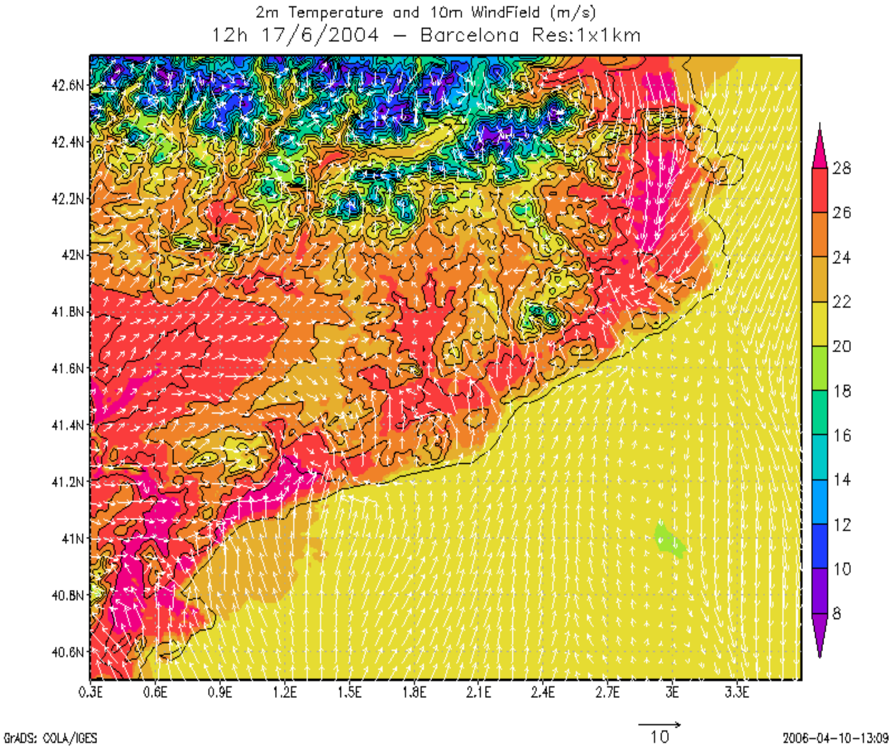


Figure A.5. Diurnal wind flows and surface temperature (12.00UTC-17 June, 2004) in the NEIP domain.

The CIP domain is characterized by thermal winds under the influence of the synoptic flow. In this episode, during night-time the katabatic winds and valley drainages dominate (Figure A.6), while during daytime the anabatic winds enhanced by the valley channelled winds up to the mountains promote flows with a NW or NE component depending on the day (17-18 June) . Central hours of the day are characterized by high temperatures and weak winds (Figure A.7).

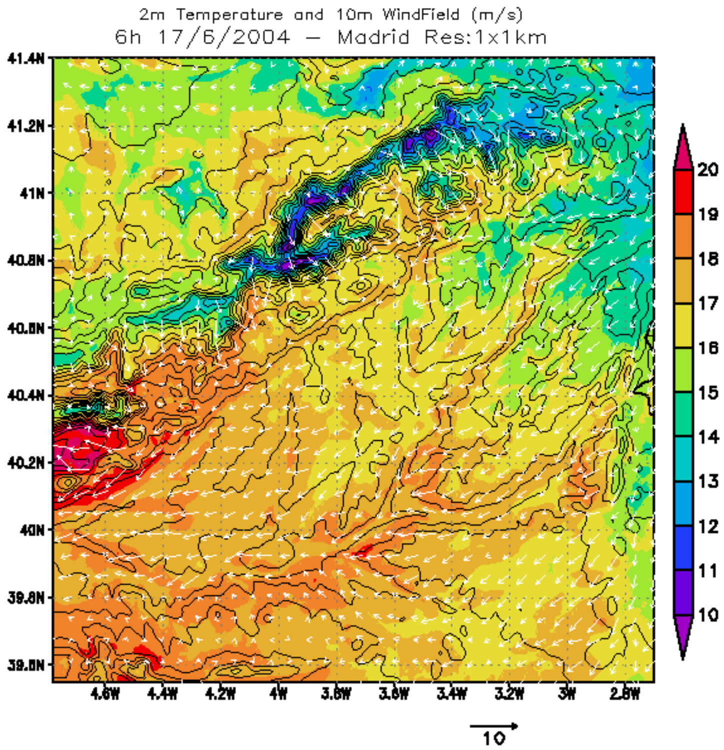


Figure A.6. Nocturnal wind flows and surface temperature (6.00UTC-17 June, 2004) in the CIP domain.

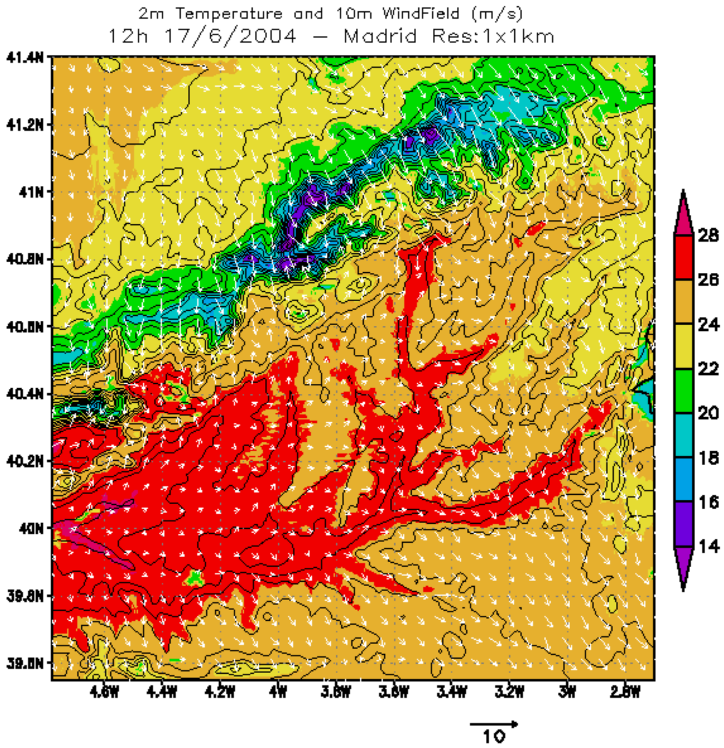


Figure A.7. Diurnal wind flows and surface temperature (12.00UTC-17 June, 2004) in the CIP domain.

These meteorological conditions favour the photochemical pollutants formation and their accumulation, being normally associated with poor air quality conditions in the studied regions, as deeply discussed along the document. The observed air quality levels in the North-eastern and central Iberian Peninsula, where Barcelona and Madrid are located, are provided by the *"Xarxa de Vigilància i Previsió de la Contaminació Atmosférica"* –XVPCA- and the "*Red de Calidad del Aire de la Comunidad de Madrid*". In both areas exceedances of the EU population information threshold for the hourly O_3 concentration (180 µg m^{-3}) are registered -Figure A.8 – The NO_2 concentrations are on average lower in the Central Iberian Peninsula for the selected episode, while the north-eastern region register hourly concentrations over 200 µg m^{-3} -Figure A.9- in several air quality stations. The hourly SO_2 concentrations remain below the limit value defined by the European Union (350 µg m^{-3}), nevertheless the levels are higher in the North-eastern Iberian Peninsula domain than in the Central Iberian Peninsula. On the contrary particulate matter –PM_{10}- concentrations are higher in this region compared to the North-eastern Iberian Peninsula. In both cases the hourly observed concentrations are frequently over the 50 µg m^{-3} limit for the 24-hr average concentration.

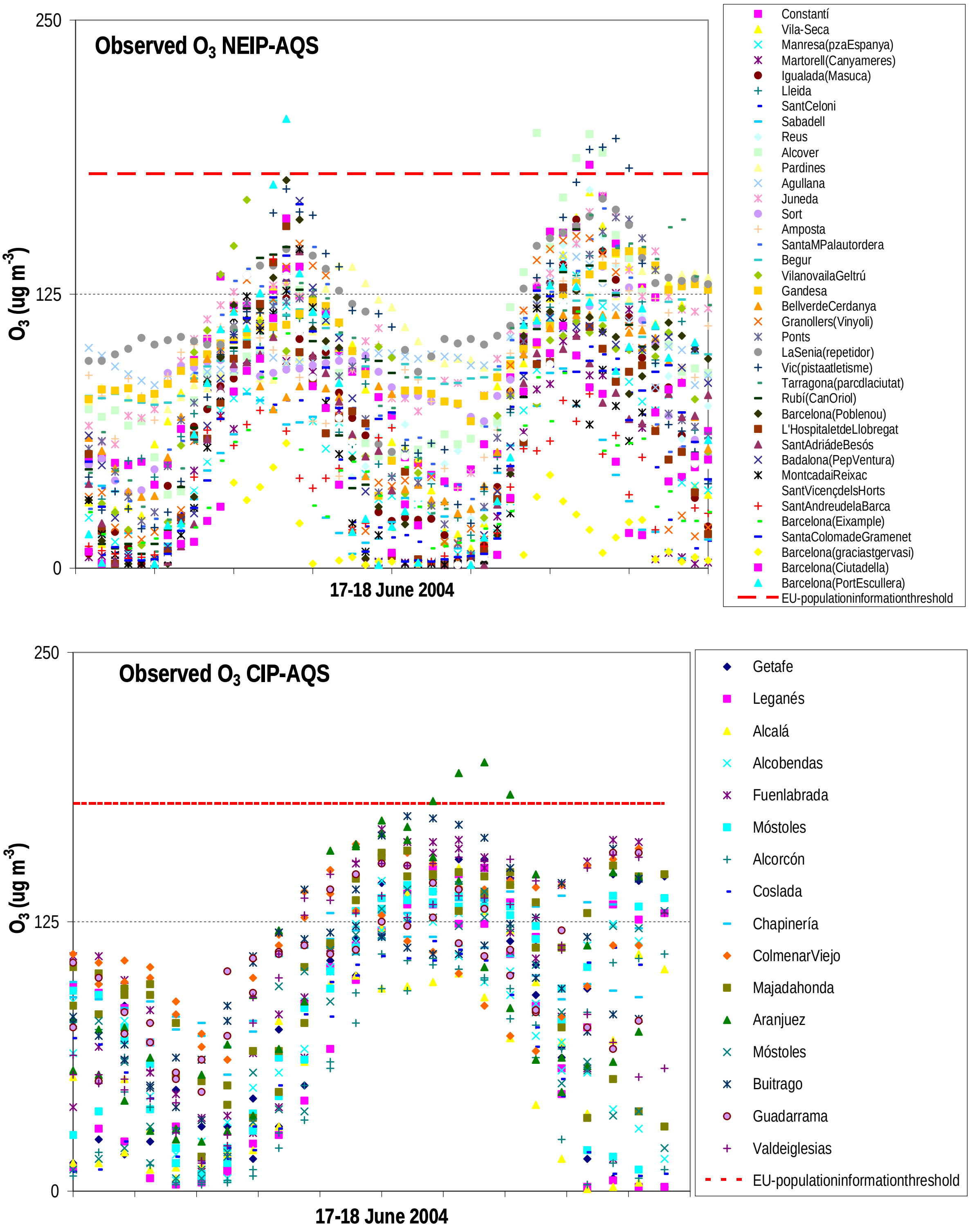
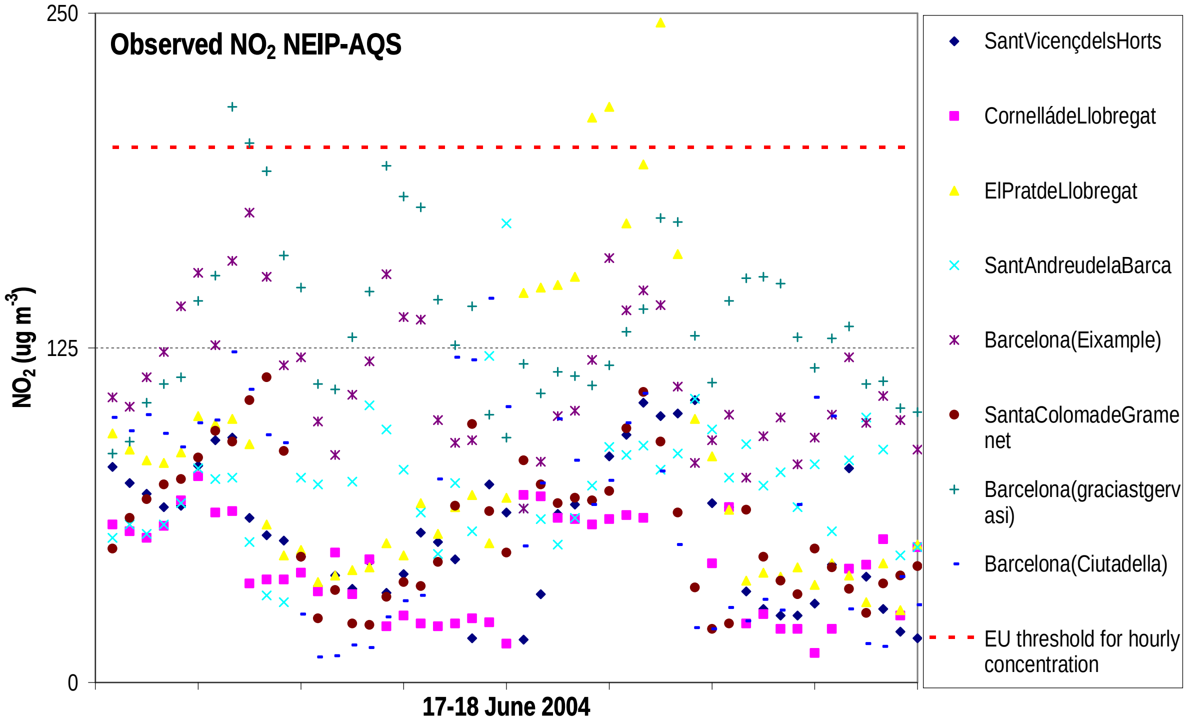


Figure A.8. Observed hourly O3 concentration (μg m-3) in the NEIP -up- and CIP –down- air quality stations for the 17-18 June, 2004.



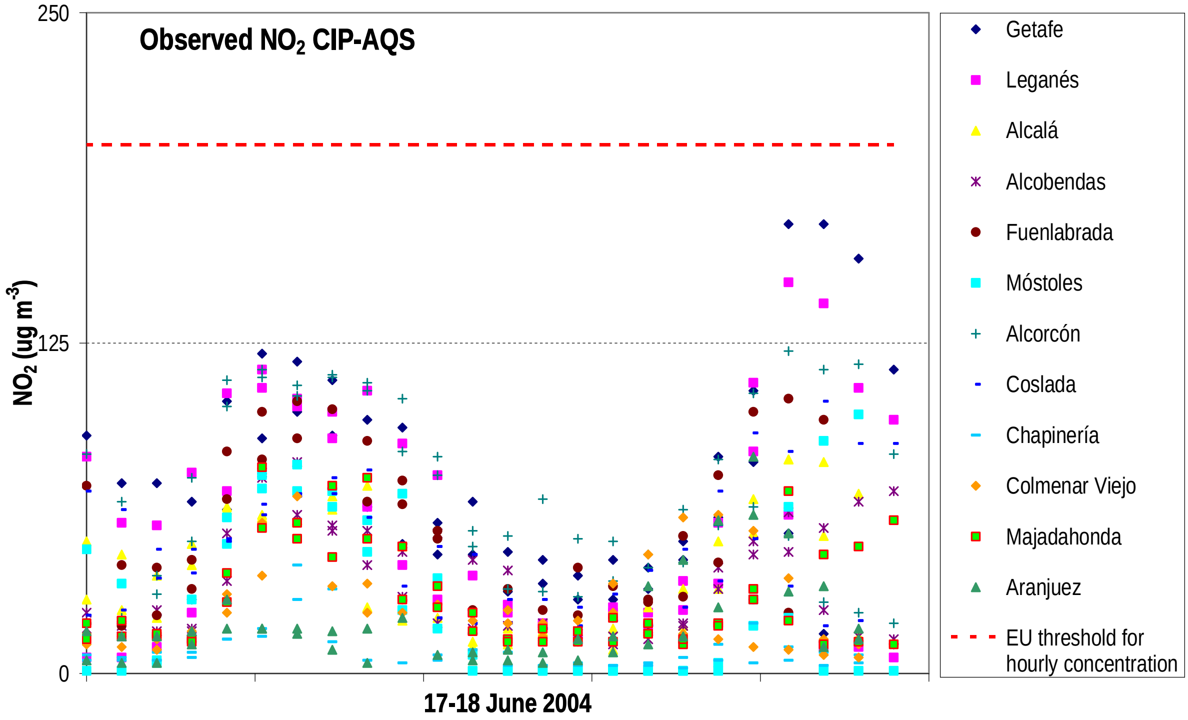
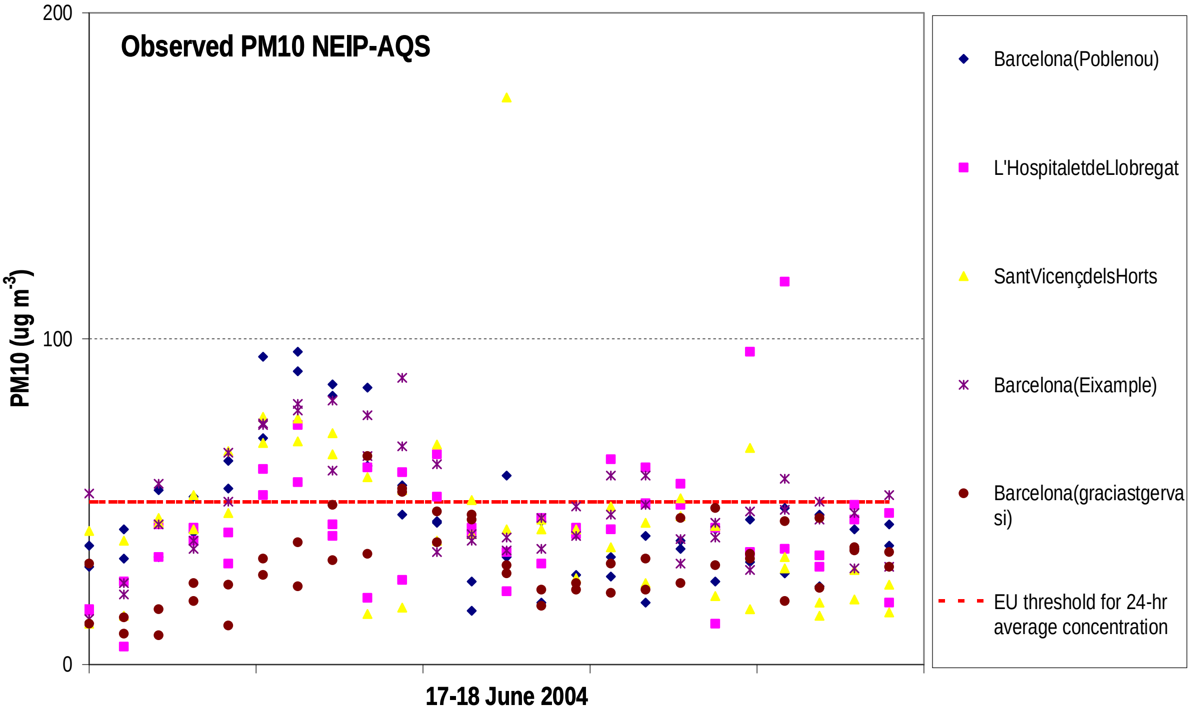


Figure A.9 Observed hourly NO$_2$ concentration (μg m^{-3}) in the NEIP –up- and CIP –down - air quality stations for the 17-18 June, 2004**.**



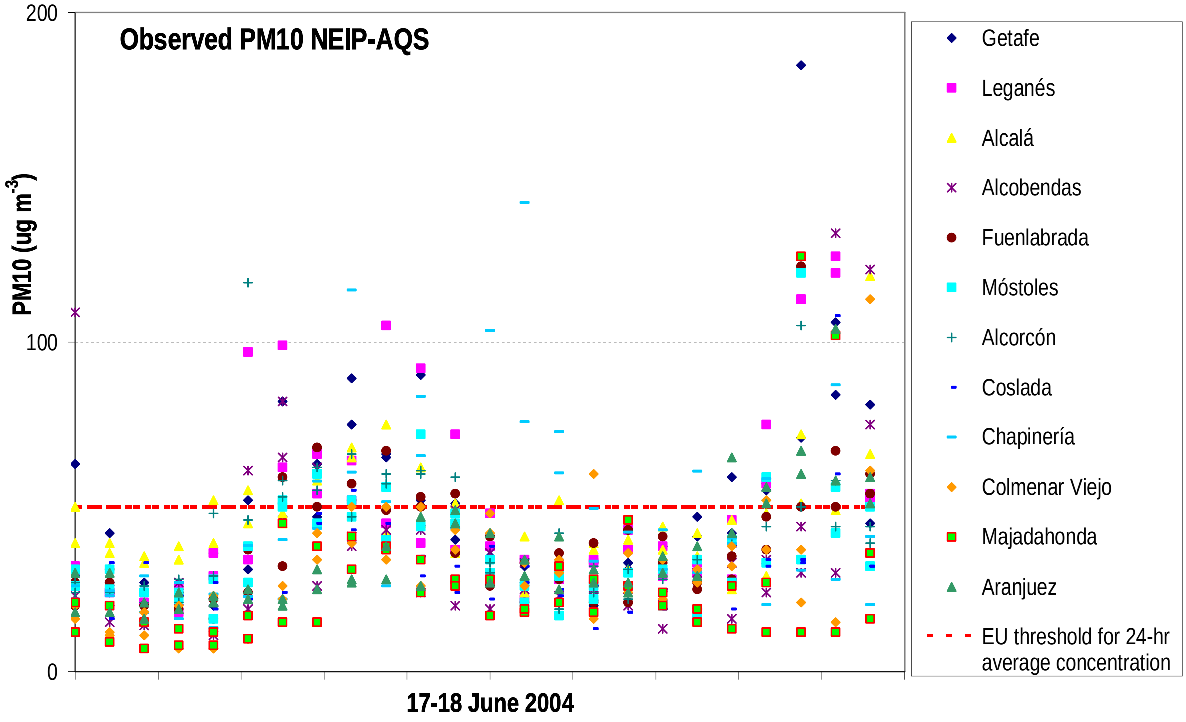


Figure A.10. Observed hourly PM_{10} concentration (µg m^{-3}) in the NEIP –up- and CIP –down- air quality stations for the 17-18 June, 2004.

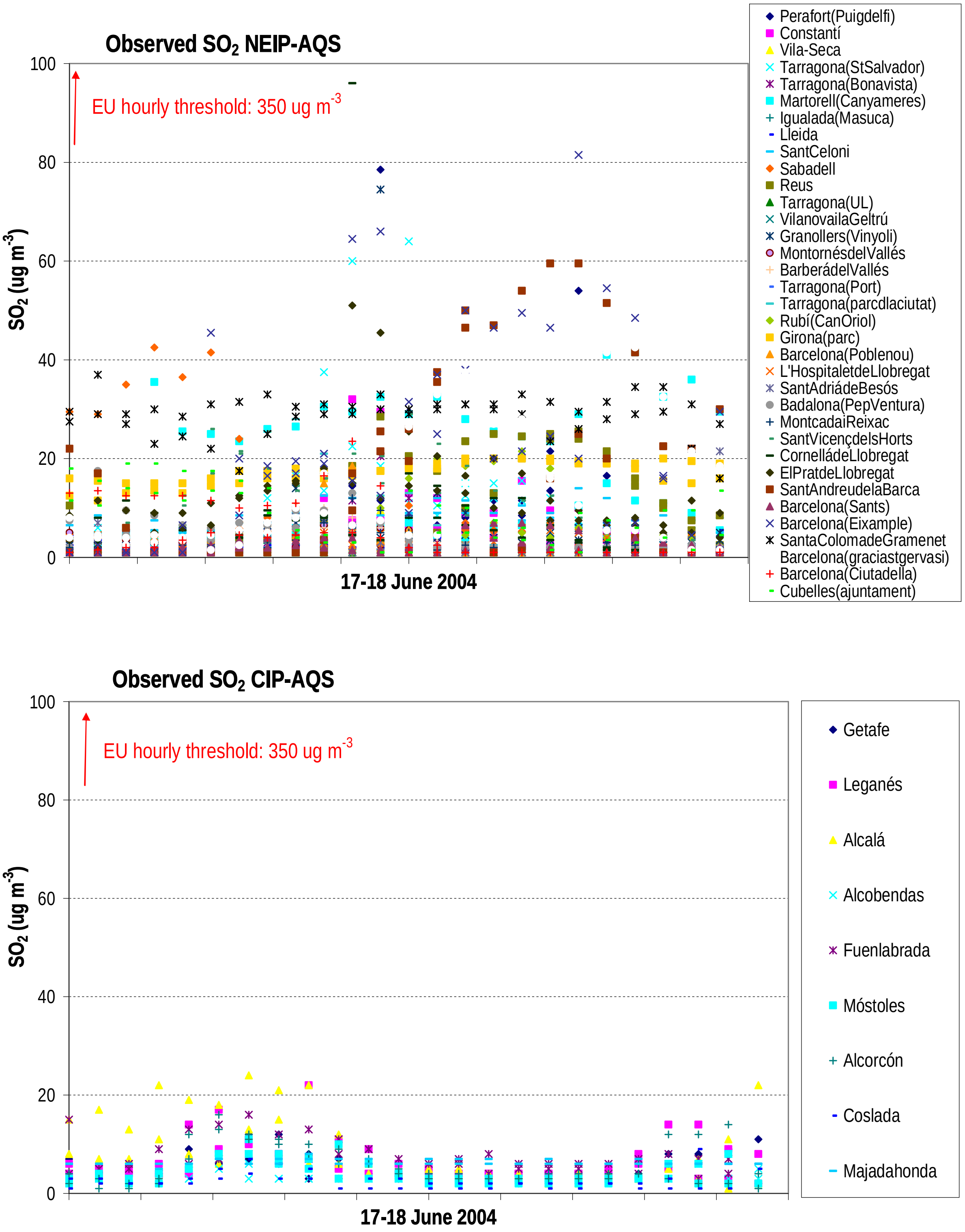


Figure A.11 Observed hourly SO_2 concentration (µg m^{-3}) in the NEIP –up- and CIP –down- air quality stations for the 17-18 June, 2004.