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Science of the Total Environment 407 (2009) 3269-3281

Contents lists available at ScienceDirect



Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv



Emissions variation in urban areas resulting from the introduction of natural gas vehicles: Application to Barcelona and Madrid Greater Areas (Spain)

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ARTICLE INFO

Article history: Received 10 September 2008 Received in revised form 11 December 2008 Accepted 19 January 2009 Available online 5 March 2009

Keywords: Emissions Alternative fuels Urban pollution Natural gas vehicles Air quality management

ABSTRACT

On-road traffic is the major contributor to pollutant emissions in urban areas. Nowadays different emission abatement strategies are being tested in order to improve urban air quality (e.g. the European Commission currently promotes the use of natural gas as an alternative fuel). Several feasible scenarios regarding the introduction of natural gas vehicles (NGV) are studied in the two main cities of Spain (Barcelona and Madrid) by using the HERMES emission model. The most suitable emission factors to NGV are selected among those available in the literature. The account of emissions in the base case scenario estimated for a typical summertime polluted day of the year 2004 reflects that in Barcelona 86% of primary pollutants come from on-road traffic compared to 93% in Madrid, because of the heavier industrial activity in the former. The introduction of NGV in urban zones would have a positive effect on emissions, whose extent largely depends on the substituted fleets and the conurbation characteristics. Maximum reductions in NO₂ emissions in Madrid are attributed to the substitution of 10% of the oldest diesel and petrol cars, while in Barcelona the change of 50% of the oldest commercial light vehicles becomes more effective. PM_{2.5} and SO₂ emissions can be significatively reduced with the introduction of NGV instead of the oldest commercial light vehicles. The substitution of conventional fuels by natural gas must reach around 4% to achieve significative reductions in traffic emissions (larger than 5%). This work focuses on air quality issues, therefore GHG emissions are not included, nevertheless this kind of associated impact has to be considered by the decision makers. Assessing the efficacy of environmental improvement strategies entails a realistic design of emission scenarios and their evaluation. The detailed emission account provides a fundamental basis for the air quality modelling and its comparison among scenarios.

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1. Introduction

Atmospheric pollution is the environmental factor with the largest impact on human health in Europe and is responsible for the largest number of diseases related to the environment (EEA, 2005a). Tropospheric ozone and particulate matter affect the human health (EC, 2005; WHO, 2004). Specifically fine particles (diameter less than 2.5 µm) are associated with increased mortality (EEA, 2005b; Pope and Dockery, 2006).

The largest contributions to the emissions of atmospheric pollutants in urban areas, where 80% dwellers in Europe live, come from the transportation sector, especially from on-road transport (Chin, 1996; Cirillo et al., 1996; Palmgren et al., 1996, 1999; Costa and Baldasano, 1996; Oduyemi and Dadvison, 1998; Colvile et al., 2001; Crabbe et al., 1999; Ghose et al., 2004). Technological improvements and emissions control legislation led to unitary emissions reduction by

vehicle; nevertheless the increase of the vehicles number makes these efforts insufficient to accomplish the more and more restrictive air quality standards. Moreover in some cases the new technologies increase specific pollutant emissions; e.g. oxidation catalyst systems or particulate filters lead to an increase in NO₂ emissions (Carslaw et al., 2007).

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

For all these reasons it is fundamental to evaluate the most suitable strategies for reducing the contribution of traffic emissions to air pollution in urban areas. Currently these strategies are mainly addressed to: (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.

The strategies to reduce the km travelled include public transport improvement, both from the infrastructural and service point of view; parking places restriction; roads construction to allow the easy flow of high occupation vehicles (taxis, buses or private cars with a high occupation rate); or the enhancement of mopeds and cycle roads. Also the introduction of taxes to use urban infrastructures contributes to a

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^{0048-9697/\$ –} see front matter @ 2009 Elsevier B.V. All rights reserved. doi:10.1016/j.scitotenv.2009.01.039

reduction in the congestion of urban zones (EC, 2001b). Parking taxes are the simplest example, but some big cities are testing more elaborated formulas, like London, which has introduced an urban toll with electronic car identification and an electronic system for taxes collection (Beevers and Carslaw, 2005).

The use of alternative fuels and the introduction of new technologies provide a way for reducing the unitary emissions by vehicle. The EU White Paper on transport policy (EC, 2001b) points out that the urban transport is a suitable market for alternative energies. Several European cities have taken this path: Paris, Florence, Stockholm, Luxembourg, Barcelona or 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 of energy supply (EC, 2000) lays down the foundations to energy diversification. The European Commission (EC, 2001a, 2007) proposes the introduction of up to 20% of alternative energies in transport at 2020. It anticipates the use of biofuels in the short term, NG in the medium-term and fuel cells or hydrogen internal combustion engines in the long term.

This work assesses the impacts on emissions of the introduction of NG as an alternative fuel in Barcelona and Madrid urban areas, which are the most populated cities of Spain and are commonly affected by air pollution episodes. Under this perspective only primary pollutant emissions are estimated, including carbon monoxide (CO), nitrogen oxides (NO_x), sulphur dioxide (SO₂), non-methane volatile organic compounds (NMVOCs) and PM₁₀ PM_{2.5} emissions are set separately, due to its effects related to human health.

2. Methods

The study areas are centred in the two largest and most populated cities of Spain, where problems related to poor air quality and traffic derived emissions are of special concern. These are: (1) the Spanish administrative capital, Madrid, which accounts for 2.840 million inhabitants; and is located in the Central Iberian Peninsula, where a domain is defined covering 373 km²; and (2) the second largest conurbation of Spain, Barcelona, located in the northern Mediterranean coast, where a 130 km² area is defined including 1.770 million inhabitants (Fig. 1).

The account of emissions was gathered on 18 June, 2004. The day is selected on the basis of two criteria: (1) a poor air quality episode, set as the worst case scenario to analyse the effects of emissions change in air quality by means of a subsequent analysis; and (2) a usual traffic circulation pattern (working days), in order to obtain representative results.

The high resolution (1 km², 1 h) HERMES emissions model, specific for the Iberian Peninsula (Baldasano et al., 2008), was used to evaluate the change in traffic emissions for each scenario. This model focuses on the estimation of gas and particulate matter pollutants, including the ozone precursors. It considers biogenic and anthropogenic emission sources, taking into account on-road, ship and planes traffic, airports and ports, power generation, industrial sectors, domestic and commercial sectors. It follows the methodologies and criteria of previous emission models developed for the Eastern Iberian Peninsula: EMICAT2000 (Parra et al., 2004, 2006) and EMIVAL2000 (Arévalo et al., 2004). The reference year chosen is 2004, since it is the most recent year in which all the required data for the development of HERMES are available.

The on-road transport 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, 2006). The vehicle fleet is classified by fuel type, weight, age and cubic capacity. The speed dependant emission factors are specific for each vehicle category. Hot exhaust, cold exhaust and evaporative emissions are considered, and also particulate matter produced by brakes abrasion, tire wear and pavement erosion.

The vehicle fleet is defined for Spain, and specifically for Barcelona and Madrid areas (Fig. 2) for the year 2004 (data provided by the national traffic management organisation of Spain (Dirección General de Tráfico)), and distributed in 72 categories. The model includes the definition of the road network, divided in stretches (inside the 1 km² cells), with specific temporary disaggregating profiles (distinguishing day-type: weekday-holiday, and month), average speed circulation, annual average daily 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, 50% of the national urban traffic volume is included, covering the whole road network for Barcelona and Madrid Greater Areas.

Mobility data (Table 1) were introduced in the urban areas to distinguish groups of specific vehicles by activity (i.e. taxis from diesel cars, or urban buses from heavy duty vehicles categories). According to the data five zones are defined in Barcelona (Fig. 1, right pannels), where the distribution of vehicles by activity was differentiated. The downtown area (zone 1. Main roads) is characterized by a high percentage of taxis (18.0% of the total volume of vehicles) and a lower proportion of heavy duty vehicles (14.8%). The ring-roads (zone3. Ronda de Dalt and zone 4. Ronda Litoral) and the industrial area of Zona Franca (zone 5) have a 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 leads to the definition of four zones (Fig. 1, left panels), consisting of approximately concentric areas from the downtown. The taxis volume in the downtown is around 20.3%, being just 6.0% on the outskirts. The ratio of heavy duty vehicles varies from 5.8% (zone 2) to 8.9% (zone 4). The mobility patterns and the distribution of vehicles by activity differ between Madrid and Barcelona, partially due to the historically larger industrial activity in Barcelona.

3. Implementation of the traffic emissions module

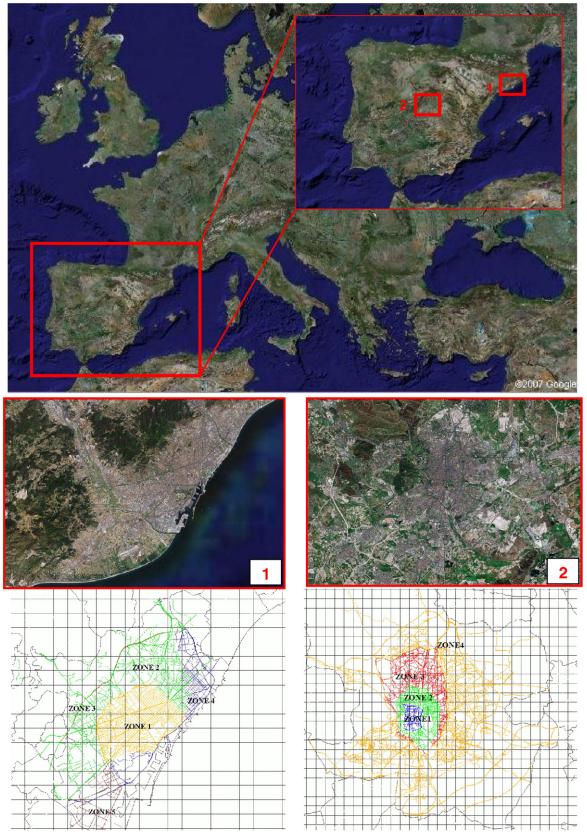
In order to evaluate the change in emissions due to the introduction of natural gas vehicles (NGV) there are two requirements: (1) estimation of the speed-dependant emission factors for different categories of NGV (cars, light duty vehicles-LDV-, and heavy duty vehicles-HDV-); (2) definition of the vehicle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

3.1. 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, 2007)). 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 was carried out. To achieve the objectives of this work: (1) speed-dependant emission factors are required; and (2) emission factors for European standards are preferred due to the location of the study areas (Spain).

Currently, the pollutant emissions for different NGV technologies 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; Nylund and Erkkila, 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 use of natural gas concentrates on urban areas. Specifically, it is used as a fuel



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in the urban bus 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 2). There is an acceptable agreement in VOCs and NO_x 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

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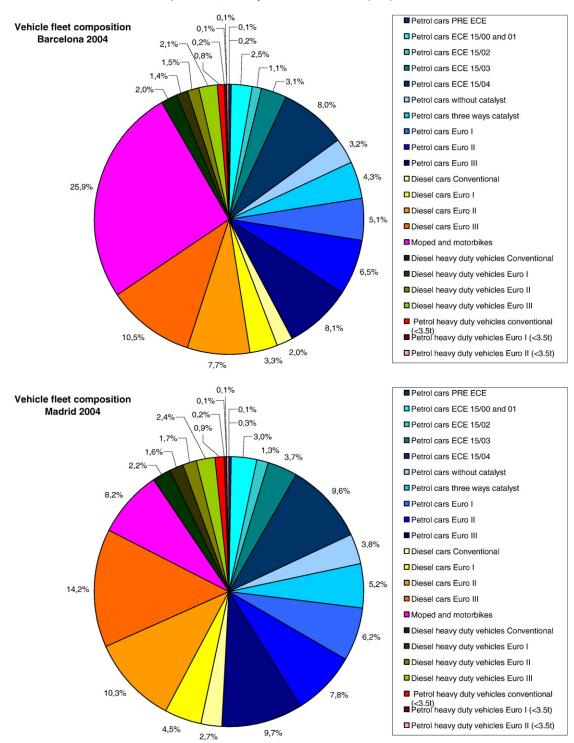


Fig. 2. Vehicle fleet composition in 2004 in Barcelona (up) and Madrid (down): different categories of petrol cars (blue), diesel cars (orange), moped and motorbikes (pink), categories of diesel HDV (green) and petrol HDV (red).

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 the average emission factor in some cases the standard deviation values, i.e. the estimations for Enhanced Environmentally Friendly Heavy Duty Vehicles (EEV HDV) indicate an emission factor of 1.54 g CO km⁻¹, and the standard deviation is 1.81 (Table 2).

The available emission rates of NG light duty vehicles are sparse (Table 2). The ascribed datasets were originated by the European Natural Gas Vehicles Association (ENGVA) and obtained by testing

specific vehicles (i.e.: Euro IV Ford Transit vans). These emission factors are not comprehensive enough to extract conclusions. Although the data dispersion is lower than in the HDV case, this fact is attributed to the common origin of most 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 values for these estimates are 0.25 and 0.02, respectively). The peer reviewed values are consistent, 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

Table 1 Vehicles distribution in each circulation zone in Barcelona (up) and Madrid (down), differentiating taxis from private cars and buses (urban and intercities) 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	67.70%		20.30%	3.80%	8.20%
1st ring road					
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	81.40%		9.70%	2.30%	6.60%
the M-30	00.000/		6.000/	0.50%	0.000
Zone 4. Outside the M-30	82.60%		6.00%	2.50%	8.90%

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

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 with the parameters of this work. However the peer reviewed NGV emission factors reported by Ristovski et al. (2004) agree with the results in this study, obtaining large variations in emission levels for different vehicles. The technology and condition of the vehicles are parameters largely affecting the final emissions; nevertheless the compressed natural gas engines

Table 2

Emission factors for compressed NG (CNG) HDV and LDV.

present lower emissions of toxic compounds than petrol or diesel equivalent engines (Ristovski et al., 2004).

The emission correction factors relate the emissions of the alternative fuel vehicle, like NGV, to an existing technology or fuel, like diesel or petrol vehicles (Table 3). With the exception of the ENGVA data, the peer-reviewed sources do not provide correction factors for all pollutants in this study (NO_x, NMVOCs, CO, SO₂ and PM₁₀) for HDV, LDV and cars, although they are an accurate reference. The correction factors for NG LDV with respect to petrol LDV agree for NMVOCs and NO_x, being on average 12% 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, although for CO (average correction factor 165%, standard deviation: 170%) and NO_x (average correction factor: 38%, standard deviation: 18%) the reliability of the correction factors is lower. They provide larger VOCs emissions for the HD-NGV than those of the diesel vehicles, mainly due to the CH₄ contribution, being the reported NMVOCs lower in all cases. The PM emissions for a HD-NGV are on average 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

Emission factors for CNG Hea	avy duty vehicles (g k	m^{-1})				
Emission standard	СО	VOCs	NMVOCs	NO _x	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) ⁽
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)
Average EII (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) ^(a)
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 $(g \text{ km}^{-1})$	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	010 1	2.5	0.005	Samaras and Zierock
Average (g km $^{-1}$)	1.54	1.25	0.03	4.78	0.01	Sumarus und Zieroek
Standard deviation	1.81	0.70	0.01	3.41	0.01	
Emission factors for NG light	duty vehicles (g km ⁻	¹)				
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	0.00	0.02		ENGVA (2006) ^{(b),(c)}
Euro IV	0.37	0.06		0.02		ENGVA (2006) ^{(b),(c)}
Unknown	0.14	0.00	0.02	0.02		Kremer (1999)
Average (g km $^{-1}$)	0.14	0.08	0.02	0.08		Memer (1555)
Standard deviation	0.47	0.08	0.02	0.03		
	0.25	0.02	0.02	0.05		

Average values and standard deviation of data obtained from different sources.

^(a) More than 200 tests carried out within 34 individual buses (included diesel buses present at the study, but not presented 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.

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Table 3

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Emission correction factors for NG-LDV respect to petrol LDV and diesel LDV and correction factors for NG-DV with respect to diesel vehicles.

NGV category	Reference vehicle	Correction	Correction factor (CF) ^(a)					
		$CF = EF_{NG}/EF_{reference-vehicle}$						
		CO	VOCs	NMVOCs	NO _x	PM		
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 (<7.5 t)	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) SO₂ emission factor is considered as zero, because of the low sulphur content (less than 10 ppb) of NG.

^(b) TWC: three ways catalyst.

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 the reduction factors provided by ENGVA (2006) shown in Table 3 are used. They represent the emissions reduction of each pollutant for a NGV referred to a diesel

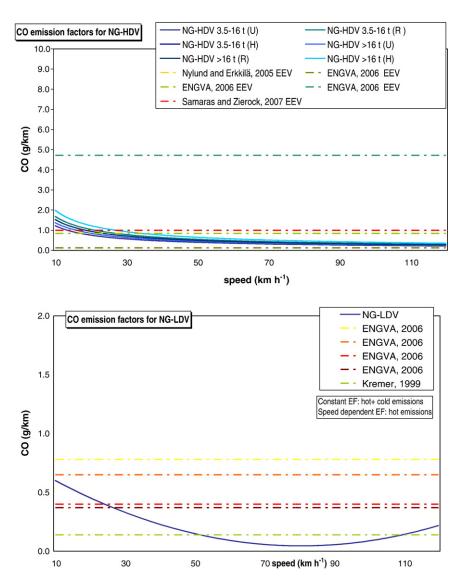


Fig. 3. CO emission factors estimated for natural gas heavy duty vehicles NG-HDV (up) and light duty vehicles NG-LDV (down). The dotted lines present the peer-reviewed constant emission factors and the solid lines those estimated by the chosen correction factors (ENGVA, 2006) applied to the correspondent diesel emission factors (EEA-EMEP CORINAIR, 2000).

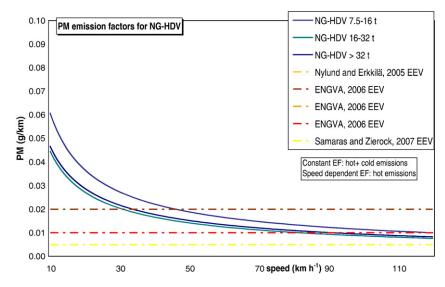


Fig. 4. PM emission factors estimated for natural gas heavy duty vehicles NG-HDV. The dotted lines present the peer-reviewed constant emission factors and the solid lines those estimated by the chosen correction factors (ENGVA, 2006) applied to the correspondent diesel emission factors (EEA-EMEP CORINAIR, 2000).

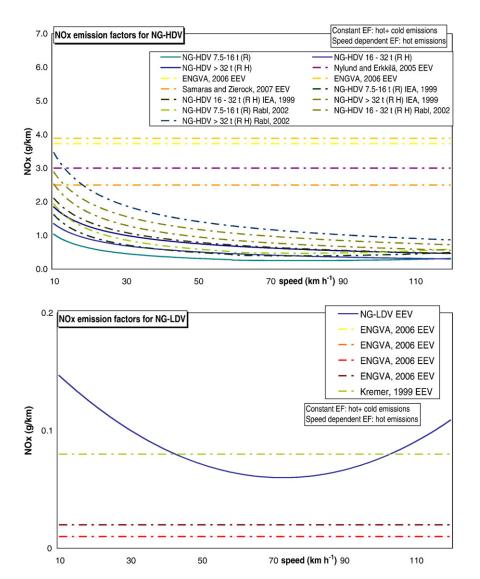


Fig. 5. NO_x emission factors estimated for natural gas heavy duty vehicles NG-HDV (up) and light duty vehicles NG-LDV (down). The dotted lines present the peer-reviewed constant emission factors and the estimations done with the correction factors from IEA (1999) and Rabl (2002). The solid lines present the emission factors estimated by the chosen correction factors (ENGVA, 2006) applied to the correspondent diesel emission factors (EEA-EMEP CORINAIR, 2000).

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 CH₄ (95%) (Rabl, 2002) it would be possible to estimate CH₄ emissions. Nevertheless this work focuses on air quality issues, therefore the analysis of the GHG emissions variation is not carried out.

Nowadays, the speed-dependant correlation for the NGV emission factors is not available. Usually the emission correction factors proposed by various 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) 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 to the peer-reviewed factors where possible. These present the same order of magnitude, especially for CO (Fig. 3) and PM (Fig. 4). Nevertheless the speed dependency shows that at low speed the consideration of 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; albeit the U shaped speed dependency for LDV is not captured by the constant emission factors (Fig. 5).

3.2. Definition of scenarios

The base case scenario (EB) 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 18, June, 2004. To implement the different scenarios the vehicle fleet composition in both 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 in the urban zones is modified to introduce those scenarios of adding natural gas vehicles categories (Tables 4 and 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 in Barcelona and Madrid.

Changing the 50% of the taxis (E2) 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 the vehicle fleet are estimated in the industrial and portuary 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 weighing more than 3.5 t have been changed. Expected changes in emissions in this scenario will be much larger in Barcelona than in Madrid, due to the vehicle fleet composition and the higher weight of light commercial vehicles in the former.

The substitution of 10% of private cars (E5) 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

Table 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 3.5–7.5 t	0.04%	0.06%	0.05%	0.06%	0.06%
	Heavy duty diesel vehicle 7.5–16 t	0.50%	0.80%	0.67%	0.73%	0.80%
	Heavy duty diesel vehicle 16–32 t	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 7.5–16 t	0.33%	0.53%	0.49%	0.44%	0.53%
	Heavy duty diesel vehicle 16–32 t	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 3.5–7.5 t	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 7.5–16 t	0.76%	0.48%	0.75%	0.70%	0.99%
	Heavy duty diesel vehicle 16–32 t	0.20%	0.13%	0.20%	0.18%	0.26%
	Heavy duty diesel vehicle >32 t	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,	Petrol and diesel cars	10.97%	12.90%	9.73%	9.51%	8.29%
commercial vehicles, heavy duty freight transport vehicles	Diesel and petrol HDV <3.5 t	11.12%	6.95%	10.98%	10.19%	14.41%
	Diesel and petrol HDV 3.5-7.5 t	0.25%	0.20%	0.27%	0.25%	0.34%
	Diesel HDV 7.5-16 t	1.59%	1.81%	1.97%	1.81%	2.32%
	Diesel HDV 16-32 t	0.44%	0.51%	0.55%	0.51%	0.65%
	Diesel HDV >32 t	0.06%	0.04%	0.06%	0.06%	0.08%
	Total	24.45%	22.40%	23.57%	22.31%	26.09%

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Table 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 3.5–7.5 t	0.10%	0.07%	0.06%	0.07%
	Heavy duty diesel vehicle 7.5-16 t	1.17%	0.75%	0.71%	0.77%
	Heavy duty diesel vehicle 16–32 t	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 7.5–16 t	1.02%	0.65%	0.62%	0.67%
	Heavy duty diesel vehicle 16–32 t	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 3.5–7.5 t	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 7.5–16 t	0.26%	0.19%	0.21%	0.29%
	Heavy duty diesel vehicle 16–32 t	0.07%	0.05%	0.06%	0.08%
	Heavy duty diesel vehicle >32 t	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,	Petrol and diesel cars	16.31%	14.83%	12.25%	10.51%
commercial vehicles, heavy duty freight transport vehicles	Diesel and petrol HDV <3.5 t	3.74%	2.63%	3.01%	4.06%
	Diesel and petrol HDV 3.5–7.5 t	0.29%	0.20%	0.21%	0.27%
	Diesel HDV 7.5-16 t	2.45%	1.58%	1.54%	1.73%
	Diesel HDV 16-32 t	0.34%	0.22%	0.22%	0.25%
	Diesel HDV >32 t	0.02%	0.02%	0.02%	0.02%
	Total	23.15%	19.47%	17.25%	16.84%

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 6) to 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 assimilable 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 substitution of 6.0%–12.9% of petrol and diesel by natural gas, which is near to the 10% proposed to 2020. The fuel change in the combined scenario is around 20% (10%) in Barcelona (Madrid). These values are larger than the most optimistic ratio proposed by the EU. Its analysis would provide a representation of the aggregated effect of all scenarios.

4. Emissions analysis

The vehicle 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., 2003). This is reflected in the emissions account for the base case scenario: 86% of the total mass emissions (NO_x, NMVOCs, CO, SO₂ and PM₁₀) in Barcelona come from on-road traffic, compared to 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 (81% in Barcelona, 94% in Madrid), CO (98% in Barcelona, 100% in Madrid) and NMVOCs emissions (77% in Barcelona and 79% in Madrid) Fig. 6. The industrial component importance in Barcelona is also reflected in the SO₂ emissions (53%). In Madrid the traffic and energy consumption of domestic and commercial sectors substitute the industry as main SO₂ emitter (contributing a 53% and 42%, respectively). Concerning PM₁₀, the contribution of the industrial sector is also important in Barcelona

(35%), while in Madrid it is almost negligible (2%). Then the measures for the control of on-road traffic emissions are expected to be more effective in reducing emissions in Madrid than in Barcelona.

The particulate matter emissions from on-road traffic are mainly included in the $PM_{2.5}$ fraction in the HERMES estimations. 2.0 t d⁻¹ of $PM_{2.5}$ in Barcelona and 4.2 t d⁻¹ in Madrid are attributed to on-road traffic, accounting for more than 85% of the PM_{10} in both cities (2.2 t d⁻¹ and 4.8 t d⁻¹, respectively). All activity sectors emit 2.6 t d⁻¹ of $PM_{2.5}$ in Barcelona and 4.6 t d⁻¹ in Madrid. $PM_{2.5}$ disaggregation is estimated according to the chemical mechanism Carbon Bond IV (Gery et al., 1989). The 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 estimations of the HERMES traffic module for PM coarse (PM with aerodynamic diameter ranging between 2.5 and 10 μ m) 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 traffic flows and the typology of vehicles do not vary between scenarios.

Table 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	1376		3607		
fleet composition					
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.

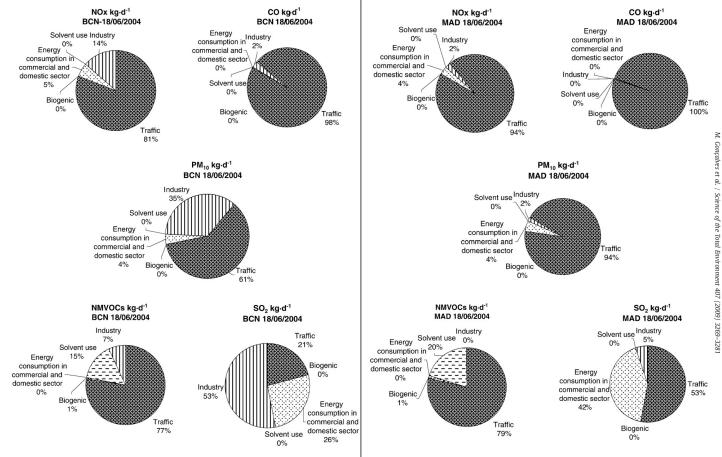


Fig. 6. Source apportionment for Barcelona greater area emissions (left) and for Madrid greater area emissions (right) the 18, June, 2004.

Fig. 7 depicts the weighted change in traffic emissions for each scenario, where the sum of all scenarios (E7) involves the largest relative variation in all emission compounds. 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 100% of the urban buses (E1), 50% of the intercity buses (E3) and 100% of HDV (E6) are negligible in terms of emissions change (Tables 7 and 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 SO₂ 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 SO₂ 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). Substituting the freight transport vehicles in Barcelona (E6) reduces SO₂ 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, Fig. 2). Regarding E3, changes are more significant in Madrid, due to the larger number of intercity buses, making the SO₂ emissions from traffic reduce up to 4.0%, meanwhile in Barcelona the traffic emissions do not decrease over 2.2% for any pollutant.

Table 7

Emissions reduction from on-road traffic (kg d^{-1}) for each scenario in Barcelona.

Emissions from road traffic in Barcelona									
Pollutant (kg d^{-1})	NO _x	NMVOCs	CO	SO ₂	PM10	Total			
Base case scenario (EB)	23,949	72,740	116,162	736	7356	223,244			
%	NO _x	NMVOCs	CO	SO_2	PM_{10}				
E1-EB	- 3.6%	-0.2%	-0.2%	-4.2%	- 3.1%				
E2-EB	-2.8%	-0.1%	-0.2%	-6.2%	-4.2%				
E3-EB	-2.0%	-0.1%	-0.1%	-2.2%	- 1.8%				
E4-EB	- 15.1%	-1.0%	- 8.3%	- 15.1%	-24.5%				
E5-EB	- 7.8%	-3.3%	- 12.1%	-3.9%	-4.6%				
E6-EB	-3.4%	-0.2%	-0.2%	-4.1%	-2.8%				
E7-EB	- 34.7%	-5.0%	-21.0%	- 35.6%	-41.0%				

Further definition of the scenarios on Section 3.2.

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 7 and E5 in Madrid, changing 10% of private cars (reduction in NO_x traffic emissions of 10.9%) Table 8. The vehicle fleet composition is the key factor for these differences (Fig. 2). Madrid vehicle fleet is characterized by a high ratio

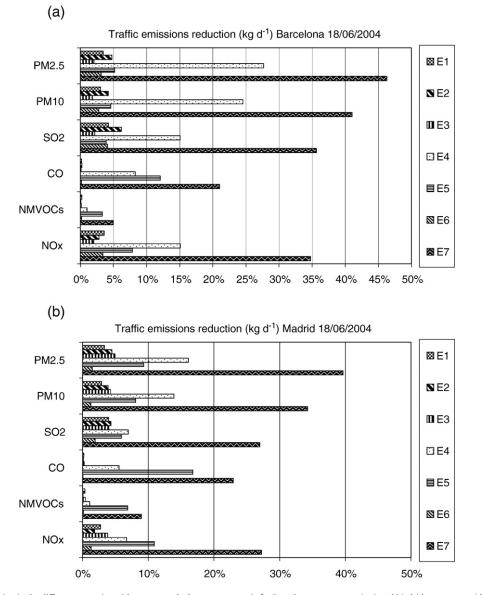


Fig. 7. Traffic emissions variation in the different scenarios with respect to the base case scenario for Barcelona greater area (up) and Madrid greater area (down). Scenarios EB, E1–E7 detailed on Section 3.2.

Table 8

Emissions reduction from road traffic (kg d^{-1}) for each scenario in Madrid.

Emissions from road traffic in Madrid									
Pollutant (kg d^{-1})	NO _x	NMVOCs	CO	SO ₂	PM_{10}	Total			
Base case scenario (EB)	66,700	95,767	297,574	1832	18,238	485,432			
%	NO_x	NMVOCs	CO	SO ₂	PM_{10}				
E1-EB	-2.7%	-0.4%	- 0.2%	- 3.9%	-2.9%				
E2-EB	-1.8%	-0.1%	-0.1%	-4.3%	- 3.9%				
E3-EB	- 3.8%	-0.4%	-0.2%	-4.0%	-4.3%				
E4-EB	-6.7%	-1.1%	- 5.5%	-6.9%	- 13.9%				
E5-EB	-10.9%	-6.9%	- 16.8%	-5.9%	-8.1%				
E6-EB	- 1.3%	-0.1%	-0.1%	- 1.9%	- 1.3%				
E7-EB	- 27.3%	-8.9%	-22.9%	-27.0%	- 34.3%				

Further definition of the scenarios on Section 3.2.

of private cars and taxis; meanwhile Barcelona shows an important fraction of commercial light vehicles. Commercial vehicles are petrol or diesel LDV (<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 (Fig. 7)).

 SO_2 and $PM_{2.5}$ reductions are mainly noticeable when transforming to NG of 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%. On the other hand, due to the large contribution of industrial emissions in Barcelona area, if all emission sectors are considered, the reduction in SO_2 emissions accounts just for 3.1%. $PM_{2.5}$ from on-road traffic is reduced by 27.7% in Barcelona and by 16.1% in Madrid.

The substitution of 50% of taxis by NG cars (E2) involves a change in the 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 the variation of emissions (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, affecting clearly the emissions differences between scenarios.

5. 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, European large cities are already trying out different strategies, among others the introduction of alternative fuels like natural gas.

Several scenarios of natural gas vehicles introduction 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 buses and coaches, 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 SO₂ and PM emissions, especially when substituting old commercial LDV (<7.5 t). The decrease estimated in PM is mainly caused by the reduction in the emissions of the fine fraction (lower than 2.5 μ m), 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.

This work also shows that 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 high resolution HERMES emission model reveals as a powerful tool to assess scenarios of emission variation 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 are needed in the future to increase the size of 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 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.

Acknowledgments

The authors gratefully acknowledge E. López for the implementation of HERMES. The information for implementing industrial emissions was provided by the Environmental Department of the Catalonia Government (Spain). Also we acknowledge the collaboration of Natural Gas Corporation and the help of the European Natural Gas Vehicles Association (ENGVA) in assessing the emission factors for natural gas vehicles. This work was funded by the projects CICYT CGL2006-08903 and CICYT CGL2006-11879 of the Spanish Ministry of Education and Science and CALIOPE project 441/2006/3-12.1 of the Spanish Ministry of the Environment. Images presented in Fig. 1 were obtained from Google Maps, © 2007 – Images DigitalGlobe, Terrametrics, NASA.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.scitotenv.2009.01.039.

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