

# Using a Coupled Air Quality Modeling System for the Development of an Air Quality Plan in Madrid (Spain): Source Apportionment and Analysis Evaluation of Mitigation Measures

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## Abstract

In this contribution, we use a coupled air quality modelling system (AQM) as a tool to design and develop an air quality plan in Madrid. AQM has allowed us to obtain a preliminary evaluation of the effect of mitigation measures over regional and local air quality levels. To achieve these goals, we have prepared a sophisticated AQM, coupling the meteorological model WRF, the emission model AEMM, and the photochemical model CMAQ. AQM was evaluated using the whole modelling vear 2010 working with high horizontal resolution, 3 km for the region of Madrid and 1km for urban metropolitan area of Madrid. Two different analyses have been realized: a source apportionment exercise following a zero-out methodology to obtain the contribution to the air quality levels of the different emission sector; and an evaluation of the main mitigation measures considered in the air quality plan using sensitivity analysis. The air quality plan was focused on the improvement of NO<sub>2</sub> levels and AQM analyzed the effect of the mitigation measures during ten episodes of 2011 where NO<sub>2</sub> or O<sub>3</sub> levels were the highest of the year; so we analyzed the effect of the mitigation plan in worst conditions. Results provided by the AQM system show that it accomplishes the European Directive modelling uncertainty requirements and the mean absolute gross error for 1-h maximum daily NO<sub>2</sub> is 31% over locations with higher levels of this atmospheric pollutant; the road traffic is the main contributor to the air quality levels providing a 81% for NO<sub>2</sub>, 67% for CO and 46% for PM<sub>10</sub>; measures defined in the plan achieve to reduce up to 11  $\mu$ gm<sup>-3</sup> NO<sub>2</sub> levels offering highest reductions over urban areas with traffic influence.

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## **Keywords**

Environmental Assessment, Air Quality Modelling, CMAQ, Emissions, Madrid, Air Quality Plan, Mitigation Measures

## **1. Introduction**

The largest amount of gases and aerosols emitted into the atmosphere are generated in cities with poor land extension and large population (about 50% population in 0.1% land area). These emissions influence weather and climate [1] and health. Recently, pollution has been included as one of the cancer-causing agents by the World Health Organization [2]. Even pollutant concentrations remain high, particularly in urban areas, air emissions have been reduced significantly in recent years [3]. Road traffic emissions associated with combustion and road dust resuspension processes are the main causes of pollution in urban areas and conurbations [4]-[7].

In these areas, there are high levels of nitrogen dioxide (NO<sub>2</sub>) and particulate matter (PM<sub>10</sub>) by comparison with the air quality standards (European Directive EC/2008/50). In Spain, annual average values of NO<sub>2</sub> and PM<sub>10</sub> are elevated in many urban air quality measurement stations with traffic influence [8]. Whereas, high ozone levels are measured in rural or suburban areas located downwind of urban or industrial locations and where local ozone precursors are lacking [9] [10]. Scientific studies has demonstrated that exposure to a high levels of NO<sub>2</sub>, O<sub>3</sub> or PM<sub>10</sub> can increase respiratory problems as inflammation, can lead to asthmatic responses in sensitive people or even cause premature death [11]-[15].

In order to improve air quality levels in urban areas, in the last years they have been developed international and national action plans [16]-[18]. Policies over traffic sector to improve air quality in urban areas have followed different strategies associated: to decrease variables associated with traffic which directly affect the amount of pollutant emissions (velocity or intensity vehicles flow); and to change Vehicles Park distribution, to introduce new technologies or alternative fuels [19] [20].

In the same way, Madrid has developed an ambitious action plan to improve the air quality in the last years. Previously to the development of the air quality plan evaluated in this paper, the Regional Government of Madrid developed the Air Quality and Climate Change Strategy 2006-2012 (Plan Azul). This plan established an amount of 111 measures with a degree of compliance of 87%. Furthermore, the Regional Government of Madrid updates periodically its emission inventory (14 versions in the last 24 years), and considers air quality modelling to evaluate mitigation measures prior to adopting them.

In this sense, air quality modelling has become a useful tool for administrations since it provides them a method to deal with human resources, production, emergency proceedings or to improve existing air quality plans and test abatement strategies. In the last years, local administrations have used models to prepare air quality plans in urban areas as the Plan Azul case. Models are able to provide the difference of pollutants concentration and a quantitative assessment of the effect of policies and mitigation plans [21]-[27].

This work aims to investigate the effect on air quality concentrations of measures proposed by the air quality plan called Air Quality and Climate Change Strategy of the Regional Government of Madrid 2013-2020 (Plan Azul +). Specifically, we will analyze one scenario with all measures proposed applied over emissions inventory, affecting traffic, residential and industrial sectors (section 2.3). The study includes a numerical deterministic evaluation that shows the accuracy of the air quality modelling outputs; and a source apportionment analysis to know the contribution to air quality levels of each emission sector.

We have used WRF-ARW/AEMM/CMAQ (Section 2.2) modelling system to evaluate the impact of each emission scenario by sensitive analysis (comparison between scenarios). To develop this air quality modelling system, we have followed the recommendations proposed by [28] on the Guide on the use of models for the European Air Quality Directive.

Description of the modelling system used, is presented in Section 2, as well as the area characteristic, data used, episode selection and mitigation measures proposed. A detailed analysis of the results obtained is presented in Section 3, and finally, some conclusions are reported in Section 4.

## 2. Methodology

A short summary of the modelling system, the area of study, the data used for the emission estimation, the pe-

riod analyzed, the action plans considered and their corresponding scenarios is included below.

#### 2.1. Area Characteristic, Data Used and Episode Selection

The area of study has been Madrid in the centre of the Iberian Peninsula over the Central Plateau. The Community of Madrid is surrounded by the autonomous communities of Castile and León and Castile-La Mancha and covers the 1.6% percent of the territory of Spain. Madrid and its metropolitan area is the third-largest in the European Union and due to its economic activity, high standard of living, and market size, is considered one of the major financial centre of Southern Europe. Madrid is served by highly developed communication infrastructures and one of the regions best connected by roads and railways in Europe.

The population of Madrid metropolitan area reached in 2012 a population of 6.5 million (around 14% of Spain). The Community of Madrid is composed on 179 municipalities, being Alcalá de Henares, Alcobendas, Alcorcón, Fuenlabrada, Getafe, Leganés, Madrid, Móstoles, Parla and Torrejón de Ardoz the most populated. Madrid is the capital and largest city in Spain.

Madrid presents a varied topography combining mountain peaks rising above 2000 m, holm oak dehesas and low lying plains, being 650 meters the average altitude. Peñalara is the highest mountain in Madrid, reaching 2428 m.a.s.l., located in the Guadarrama mountain range in the west region of the Community.

Since a climate point of view Madrid has a temperate Continental Mediterranean climate with cold winters with temperatures below 0°C habitually. During summer temperatures rises above 30°C and frequently reach 40°C in July. Yearly average precipitation levels are below 500 mm, distributed throughout the year and with maximums in autumn and spring. Hottest and driest regions are reproduced in the flatter areas on the south of the region, whereas coldest and wettest areas are located in the mountain ranges. In the urban areas of Madrid the climate is modified by the heat island effect, increasing mainly nocturnal temperatures.

Anthropogenic contribution dominates pollutant air emissions in Madrid. Transport emissions (road and non-road traffic) from the metropolitan area of Madrid are the main CO,  $NO_x$  and particulate matter emission sector, representing between a 53 and an 86% of the total emissions. Airport represents a important contribution to the emissions of the whole Community of Madrid. On other hand, industrial emissions dominate  $SO_x$  and NMVOCs (non-methane volatile organic compounds) emissions.

In Figure 1, we show models domains used for simulations (Section 2.2) that represents the Community of Madrid.

Regarding the air quality levels, ozone and nitrogen dioxide limit values fixed by the European Air Quality Directive EC/2008/50 has been exceeded during the last years. In 2010, the  $O_3$  threshold information value was exceeded in 30 occasions in the air quality stations handled by the Regional Government of Madrid. NO<sub>2</sub>



Figure 1. Models domains for simulations (left panel). Zoom domain of Community of Madrid and the Urban Metropolitan area of Madrid.

maximum 1-hour limit value was exceeded in 31 occasions but not exceeding the tolerance fixed by the Directive (18 occasions permitted per year and station). In the recent years  $SO_2$  and  $PM_{10}$  levels have showed a decrease, whereas  $O_3$  has showed a trend to rise. The rest of pollutants remain constants with exceedances of the  $NO_2$  annual limit value.

To realize this study we have chosen 2010 as modelling year. The whole calendar year has been considered to analyze the source apportionment of every emission sector, and 10 meteorological episodes of 48 hours in 2010 have been considered to evaluate each mitigation measure. We have selected meteorological episodes with the highest  $NO_2$  and  $O_3$  concentrations measured, evaluating mitigation scenarios in the worst case since an air quality point of view. 5 meteorological episodes correspond on highest  $O_3$  concentration and 5 on highest  $NO_2$  concentration.

We have characterized episodes using air quality measurements from the Air Quality Network that belongs to the Environment and Territorial Planning Agency of the Regional Government of Madrid. In Table 1, we show the date of every episode selected,  $NO_2$  and  $O_3$  maximum 1-h per episode and annual average of these statistics.

**Table 1.**  $NO_2$  and  $O_3$  daily maximum 1-h values measured in the air quality stations of the Community of Madrid during meteorological episodes selected and annual average (U correspond to urban station; S, suburban; R, rural; T, traffic; I, industrial; and F, background).

	Period and NO <sub>2</sub> daily maximum 1-h ( $\mu$ gm <sup>-3</sup> )					Period and $O_3$ daily maximum 1-h ( $\mu$ gm <sup>-3</sup> )						
Air Quality Station	17/03	20/10	28/10	04/11	28/12	Annual average	24/06	06/07	08/06	11/08	20/08	Annual average
Alcalá de Henares (UT)	68	129	119	97	161	68	151	164	100	194	159	93
Alcobendas (UI)	131	174	134	156	172	67	148	181	70	163	153	82
Alcorcón (UF)	141	186	179	188	135	80	135	154	72	141	139	86
Algete (SF)	68	69	79	53	50	34	166	186	105	175	168	102
Aranjuez (UF)	50	113	127	104	74	55	109	103	81	130	144	81
Arganda del Rey (UI)	82	75	79	50	63	45	85	133	81	178	119	78
Colmenar Viejo (UT)	129	155	168	118	131	82	135	139	91	98	150	85
Collado Villalba (UT)	209	233	153	163	149	75	155	174	95	165	174	90
Coslada (UT)	112	84	217	202	188	101	130	139	69	177	150	78
El Atazar (RF)	4	13	100	6	49	11	171	171	106	134	153	104
Fuenlabrada (UI)	168		154	175	114	82	122	143	67	138	121	79
Getafe (UT)	96	200	219	234	126	82	130	131	79	124	127	76
Guadalix de la Sierra (RF)	43	45	63	31	58	26	85	183	80	148	170	92
Leganés (UT)	158	133	147	149	157	93	118	123	46	145	134	75
Majadahonda (SF)	145	144	121	155	144	67	153	181	95	139	152	95
Móstoles (UF)	149	126	129	139	135	71	73	114	87	104		77
Orusco de Tajuña (RF)	4	31	25	29	23	13	148	138	116	219	144	102
Rivas Vaciamadrid (SF)	102	148	159	130	121	79	124	134	89	160	137	84
San Martín de Valdeiglesias (UF)	48	34	42	32	54	20	118	137	108	143	150	89
Torrejón de Ardoz (UF)	82	150	172	123	88	58	113	97	83	138	137	76
Valdemoro (SF)	98	84	89	77	87	56	124	125	67	153	133	84
Villa de Prado (RF)	36	32	64	11	56	18	130	119	96	89	126	85
Villarejo de Salvanés (RF)	42	95	125	120	80	49	128	121	95	185	125	90

## 2.2. Modeling Approach and Emissions Inventory Used

The design, implementation and configuration of the air quality modelling system have been made by researchers with an extensive experience as modellers [29] [30]. The air quality modelling system has been set with the optimum parameterizations to reduce the uncertainty of the models [31]-[33]. The authors have applied this kind of models as forecast tool as assessment tool of mitigation plans [26] working in collaboration with different regional and local administrations (Environmental and Water Agency of Andalusia Government, Environment and Territorial Planning Agency of Regional Government of Madrid and Territory and sustainability Agency of Catalan Government).

Three models compose the air quality modelling system: a meteorological model, an emission model and a photochemical model. The recommendations and requirements indicated in the Guide on the use of models for the European Air Quality Directive [28] have been used for the models configuration, and also to choose the optimum kind of models used to evaluate the air quality plans. This coupled air quality modelling system has been applied and tested successfully in urban, industrial and mine areas. Urban areas as Madrid, Barcelona, Seville (Spain) or Nice (France); industrial areas as Ponferrada or Tarragona (Spain); and mine areas as Calama (Chile). The air quality modelling system showed has been evaluated using Maximum Relative Directive Error [28] referred in the European Directive EC/2008/50. Results obtained from this evaluation accomplish the model uncertainty limits according to the Directive for the pollutants O<sub>3</sub>, NO<sub>2</sub>, PM<sub>10</sub>, SO<sub>2</sub> and CO, having used measurements from more than 120 stations (urban, suburban and rural locations) during a period of four years. In section 3.1 we show the evaluation of the air quality modelling system developed in the region of Madrid.

The following paragraphs outline the main features of the three models which compose the modelling system.

#### 2.2.1. Meteorological Model

The mesoscale meteorological model used is Weather Research and Forecasting-Advanced Research (WRF-ARW) version 3.3 [34]. WRF model was configured with four nested domains with 27 (first domain), 9 (second domain), 3 (third domain) and 1 km (fourth domain) of horizontal resolution (Figure 1). First domain, called d01, covers the southwest of Europe and the north of Africa with  $108 \times 97$  grid cells. Second domain (d02), covers the whole of the Iberian Peninsula with  $142 \times 118$  cells. And the inner domains cover the Community of Madrid (d03 with  $52 \times 55$  cells) and the city of Madrid and its metropolitan area (d04 with  $61 \times 43$  cells). The vertical resolution includes 32 levels, 22 below 1500 meters, with the first level at approximately 15 meters and the domain top at about 100 hPa. The vertical structure covers the whole troposphere and a resolution decreasing slowly with height in order to allow low-level flow details to be captured. The higher resolution close to the surface is a common practice in air quality studies in order to better represent the physical-chemical processes within the Atmospheric Boundary Layer [35]-[38]. Initial and boundary conditions for domain d01 were supplied by the National Center for Environmental Prediction and National Center for Atmospheric Research (NCEP/NCAR) Climate Forecast System Reanalysis with 0.5° of spatial resolution and 6 hours of temporal sampling. We use a WRF physical configuration used in preliminary studies [26] that provides good results for air quality applications in the Iberian Peninsula [39]. Two-way nesting is used as relationship between domains for the three external domains (D01, D02 and D03) and one-way nesting for D04 due to computational issues.

#### 2.2.2. Emission Model

Air Emission Model of Meteosim, AEMM [26] [40] is a numerical, deterministic, Eulerian, local-scale model developed by Meteosim S.L. It allows obtaining the intensity of emissions in different areas, either anthropogenic (traffic, industry, residential, etc.) or natural (emissions caused by vegetation or erosion dust) for the area of interest. AEMM has been applied to the area of Madrid. AEMM considers elevated sources with his 8 levels vertical distribution. Monthly, weekly and vertical profiles are taken from the Unified EMEP model, and they are applied to determine the value of an emission for each month and day of the year, and vertical level. Two different methodologies are used to obtain emissions in each domain. By one hand, we use top-down methodology to calculate emissions for d02 domain using the European annual inventory EMEP/MSC (EMEP Chemical Transport Model www.emep.int), and our disaggregation is based on land used CLC2006 (Corine Land Class 2006) with 250 meters of resolution, coupled with different statistical functions depending on socio-economic variables [41]. On the other hand, we use the emission inventory that belongs to the Regional Government of Madrid with  $1 \times 1 \text{ km}^2$  of horizontal resolution, to adapt emissions for d03 and d04 domains. Madrid emissions

inventory version 2010 includes emissions classified by Selected Nomenclature for Air Pollution (SNAP) sectors (**Table 2**). Additionally, we use bottom-up methodology to calculate natural emissions for d02, d03 and d04 domains. As natural emissions we consider those caused by vegetation [42] or erosion dust [43] using parameterizations, land uses and meteorological outputs from WRF. These emissions are adapted and speciated by AEMM model to the requirements of the chemical module of CMAQ.

AEMM model also includes an emission projections module called AEMM-EP. This module estimates future emissions in the Community of Madrid. AEMM-EP does not realize a specific forecast, according with considerations of the EMEP/EEA emission inventory guidebook 2013 [44]. Projections are a tool to assess what might happen it we take no action, what might be achieved with actions we are committed to and what else could be done (EMEP/EEA 2013). Projections importance lies in considering different developments in the economy, technologies or policies for a sustainable development. In this way, projections are a tool to know what happens to the amount of atmospheric emissions without considering mitigation measure, with measures already taken, and considering further actions.

#### 2.2.3. Photochemical Model

The U.S. Environmental Protection Agency models-3/CMAQ model is the one used to simulate the physical and chemical processes into the atmosphere [45]. CMAQ is an open-source photochemical model which is updated periodically by the research community. In this contribution we use CMAQv4.7.1, considering CB-5 chemical mechanism and associated EBI solver [46] and AERO5 aerosol module [47]. Regarding atmospheric chemistry, CB5 considers 155 chemical reactions that involve NOx, non-methanic volatile organic compounds (NMVOCs) or ozone (O<sub>3</sub>). Additional details regarding the latest release of CMAQ can be found on the Community Modelling and Analysis System (CMAS) Center (www.cmascenter.org). CMAQ model uses the same configuration as the WRF simulation. Initial and boundary conditions for d02 domain are provided by the results of simulation of d01 domain. And the same relationship is followed between d02 and d03, d03 and d04. Meteorology-Chemistry Interface Processor (MCIP) version 3.6 is used to prepare WRF output to CMAQ model. And AEMM model prepares emissions as AERO5 and CB5 modules require.

The whole year 2010 has been modelled with simulations of 48 hours of duration for every day of the year. In order to minimize the effects of the initial conditions, the first 24 hours of each simulation have been discarded as they have been considered as spin-up time.

The air quality modelling simulations have run in Meteosim's computing cluster, which has 27 nodes and more than 212 cores.

CNAD Sector	Emissions from Madrid Emission Inventory 2010 (tonnes per year)							
SNAP Sector —		NH <sub>3</sub>	NO <sub>x</sub>	$PM_{10}$	PM <sub>2.5</sub>	SO <sub>x</sub>	VOCs	
S1: Combustion in energy and transformation industries	369	0	213	37	37	0	1	
S2: Non-industrial combustion plants	4,349	0	4,517	140	130	1,146	453	
S3: Combustion in manufacturing industry	3,586	0	7,213	228	125	2,034	382	
S4: Production processes	7,895	0	160	336	168	103	664	
S5: Extraction and distribution of fossil fuels and geothermal energy	0	0	0	1	0	0	2,070	
S6: Solvent and other product use	0	16	0	0	0	0	47,824	
S7: Road transport (urban roads, non-urban roads and motorways)	51,974	688	40,956	2,675	2,190	41	5,237	
S8: Other mobile sources and machinery (railways, inland shipping, air transport)	5,464	0	6,486	487	487	428	672	
S9: Waste treatment and disposal	256	1,204	608	12	12	504	9	
S10: Agriculture	690	2,795	116	1,279	215	8	1,795	

<b>Table</b>	2. SNAP	sectors	considered	into the	he N	Madrid	Emission	Inventory	/ and t	their p	ollutant	emissi	ons
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#### 2.3. Modeling Scenarios

In the following lines, we explain the modelling scenarios defined and the methodology used to evaluate them.

#### 2.3.1. Source Apportionment

The first analysis realized is a source apportionment exercise. The aim of this analysis is to obtain the contribution to the air quality levels of the different emission sector. To accomplish with this goal a zero-out methodology was followed, also know as the brute force method or as single-perturbation method [48] [49]. The application of this methodology consists on the comparison between the results of the air quality modelling system executed considering all emission sectors regarding the results obtained by the same system turning off one source of emissions. Turning off a specific sector is equivalent to reduce a 100% (zero-out) its emission value. This approach lets to isolate the response in nonlinear systems. In our case, we have realized nine modelling different scenarios turning off sectors. We have turned off snap sectors and to simplify we have considered S3, S4 and S6 as an only one sector (called S346). Additionally, we have turned off natural emissions included in the modelling system.

#### 2.3.2. Mitigation Measures Effect

The second analysis focus on the evaluation of mitigation measures over the air quality levels. We take into account mitigation measures considered in the Air Quality and Climate Change Strategy of the Regional Government of Madrid 2013-2020 (Plan Azul +). More information about Plan Azul + can be found at the official environmental webpage of the Community of Madrid. In **Table 3**, we show mitigation measures considered and their effect over atmospheric emissions for the Community of Madrid as a whole. Mitigation measures defined in the Plan are focused on the reduction of NO<sub>2</sub> levels primordially. For this reason we focus our attention on the effect of the Plan over NO<sub>2</sub> and O<sub>3</sub>.

Previously to analyze the combined effect of all mitigation measures considered in **Table 3**, individualized analysis was realized for different strategic measures. Considering the results obtained, some measures were accepted or modified or denied. Measures finally planned and accepted were those that good results were found in terms of reduction of air quality levels.

Sector	Mitigation measure	SO <sub>x</sub> (%)	NO <sub>x</sub> (%)	CO (%)	NMVOCs (%)	PM <sub>10</sub> (%)
S2	<ul> <li>*NO<sub>2</sub> emissions reduction from the Cogeneration Plant Barajas</li> <li>*Incorporation of environmental criteria in administrative authorizations regarding air pollution from industries</li> <li>*Use of clean fuels in the residential sector</li> <li>*Improving energy efficiency in the residential sector</li> <li>*Environmental adaptation of livestock farms</li> </ul>	-2.78	-8.04	-1.35	-0.67	-1.79
S346	*Register RIECOV and new authorizations in accordance with the Spanish lay 34/2007				-6.07	
S5	<sup>*</sup> Integration of the Phase II agreement as EESS Madrid to advance and improve the regulatory obligations in the matter				-3.66	
S7	<ul> <li>*Renewal of the fleet autotaxi fuels and clean technologies</li> <li>*Public-private partnership to promote the use of gas vehicles collaboration</li> <li>*Implementation and consolidation of charging infrastructure and encouraging the use of electric vehicles</li> <li>*Renewal of institutional fleet under environmental criteria</li> <li>*Urban and inter-city buses cleaner</li> <li>*Renewal the vehicles park with more efficient models</li> <li>*Low emission zones and residential areas of priority</li> <li>*Circulation efficient vehicles by BUS HOV lanes</li> <li>*To promote gas fuel for duty vehicles in the corridor Madrid— Castile La Mancha—Valencia</li> </ul>	-5.93	-7.15	-2.19	-2.76	-8.30
<b>S</b> 8	<sup>*</sup> Implementation of the AENA agreement in Barajas. <sup>*</sup> Environmental adaptation of livestock farms	-11.22	-9.12	-14.04	-21.57	-0.32

Table 3. Mitigation measures classified by SNAP sectors and their emission reduction estimation in comparison with the base case scenario.

As [28] recommends sensitivity analysis has been made in order to evaluate the results obtained by the Air Quality Modelling system considering Plan Azul + emissions. The basis of a sensitivity analysis is to compare the results obtained in the real scenario versus the results obtained modifying the emissions. These emission variations result from the implementation of mitigation measures. The reduction of pollutant concentrations can directly be determinate using this approach.

## 3. Results and Discussion

In the following subsections we present a evaluation of the air quality modelling system, the source apportionment analysis realized, the effect of mitigation measures defined in the Plan Azul + over air quality levels, and the emission projections for 2020.

#### **3.1. Air Quality Modeling Evaluation**

Two evaluations have been realized to evaluate the accuracy of the air quality modelling system designed and developed. By one hand, we have used the uncertainty definition for modelling of the European Directive EC/2008/50, and on the other hand, we have realized a numerical deterministic evaluation. Twice evaluations have been developed for the whole 2010 year.

As European Directive suggests, models must be verified and validated before they can be used for air quality assessment or management [28]. The quality objectives for a model are given as a percentage uncertainty. The definition of the uncertainty of the models is ambiguous in the Directive. Since values may be calculated, a mathematical formula would have made the meaning much clearer, as such, the term "model uncertainty" remains open to interpretation. Despite this, [28] suggests that it should be called the Relative Directive Error (RDE) and defines it mathematically at a single station as follows:

$$RDE = \frac{|O_{LV} - M_{LV}|}{LV}$$
(2)

where  $O_{LV}$  is the closest observed concentration to the limit value (LV) or the target value for ozone and  $M_{LV}$  is the correspondingly ranked modelled concentration. The maximum of this value found at 90% of the available stations is then the Maximum Relative Directive Error (MRDE). MRDE values and Directive recommendations are showed on Table 4. Results indicate that model uncertainty requirement is achieved for all pollutants and so, the air quality modelling system presented in this paper can be used for the aims the Directive considers.

Statistical metrics for photochemical model performance assessment are calculated for surface ozone and nitrogen dioxide concentrations at 23 measurement stations (Table 1). We consider  $NO_2$  and  $O_3$  because mitigation measures are focused on the reduction of these atmospheric pollutants. The two multi-site metrics used are the mean normalized bias error (MNBE) and the mean normalized gross error (MNGE). The U.S. Environmental Protection Agency [50] developed a guideline indicating that it is inappropriate to establish a rigid criterion for model acceptance or rejection. However, building on past air quality modelling applications [51] common values ranges have been established [29]. The accepted criteria are MNBE,  $\pm 5$  to  $\pm 15\%$ ; and MNGE,  $\pm 30$  to +35%. For the entire period studied (2010), the results in Table 5 show the statistics metrics of daily maximum 1-h and 8-h values for O<sub>3</sub> and maximum 1-h and daily values for NO<sub>2</sub>.

year.				
Pollutant	Description	MRDE (d03)	MRDE (d04)	Recommendation
NO <sub>2</sub>	Hourly limit value	35%	39%	<50%
$NO_2$	Annual limit value	34%	27%	<30%
$\mathbf{PM}_{10}$	Annual limit value	45%	46%	<50%
O <sub>3</sub>	Target value	13%	15%	<50%
СО	Limit Value	13%	14%	<50%
$SO_2$	Hourly limit value	11%	10%	<50%
SO <sub>2</sub>	Daily limit value	8%	8%	<50%

Table 4. MRDE values calculated using the air quality modelling system predictions taking into account the whole 2010

Table 5. Wittbe and Wittbe statistical values corresponding to 1002 and 03 concentrations for the domains dos and dot.										
		D	omain d03		Domain d04					
Statistical	NO <sub>2</sub>		C	<b>)</b> <sub>3</sub>	$NO_2$		O <sub>3</sub>			
	Maximum 1-h	Daily	Maximum 1-h	Maximum 8-h	Maximum 1-h	Daily	Maximum 1-h	Maximum 8-h		
MNBE (%)	9	-1	9	14	16	4	15	22		
MNGE (%)	41	28	24	29	38	21	29	36		

**Table 5.** MNBE and MNGE statistical values corresponding to NO<sub>2</sub> and O<sub>3</sub> concentrations for the domains d03 and d04.

Results indicate that the model shows a clear tendency to overestimate ground level ozone and NO<sub>2</sub> concentration, being MNBE positive in the major part of the cases. Ozone prediction shows a better accuracy than NO<sub>2</sub> forecast. NO<sub>2</sub> worst values are obtained for measurement stations located in rural areas (Algete, Orusco de Tajuña or Villa de Prado), whilst the best results are obtained in urban stations like Alcorcón, Leganés or San Martín de Valdeiglesias. The opposite result is obtained for the ozone evaluation: best results in rural areas (El Atazar, Orusco de Tajuña or Villarejo de Salvanés) and worst results in urban stations (Coslada, Arganda del Rey or Móstoles). These results show that the model predicts better  $NO_2$  and  $O_3$  in locations where measured levels of each one of these pollutants are higher. Analyzing the daily profile of ozone, we have observed a typical overestimation during the night. This fact can be associated to the model does not represent nocturnal physicochemical processes accurately enough [52] or night-time emissions profile. To solve this problem often evaluation statistics are calculated using only the hourly observation-predictions pairs for which the observed concentration is greater than a specific value [29]. We have used 60  $\mu$ gm<sup>-3</sup> as cut-off value [53]-[55] and when we apply this restriction, reductions of 9% (maximum 8-h) and 13% (maximum 1-h) have been obtained. In the same way for NO<sub>2</sub> concentrations we have eliminated very low concentrations, and a cut-off of 25  $\mu$ gm<sup>-3</sup> has been defined. The application of this restriction improves forecast between a 12% - 15%. The correlation coefficient evaluated using maximum 1-h value is 0.7 for ozone concentration (d03 and d04) and 0.8 and 0.9 for NO<sub>2</sub> concentration (d03 and d04 respectively).

## 3.2. Source Apportionment Analysis

The emission inventory values showed on **Table 2** provides that traffic sector (S7) is the main responsible to the emissions of the whole region of Madrid for CO (59% of contribution),  $NO_x$  (68%),  $PM_{10}$  (47%) and  $PM_{2.5}$  (60%), whilst S346 is the main for SO<sub>2</sub> (73%) and NMVOCs (89%). As we have commented previously we have followed a zero-out methodology to realize the source apportionment analysis for the air quality levels using CMAQ photochemical model.

In **Figure 2**, we show the contribution of the different snap sectors and natural contribution (calculated using AEMM model) to the levels of NO<sub>2</sub>, O<sub>3</sub>, CO, SO<sub>2</sub>, PM<sub>10</sub> and PM<sub>2.5</sub> using different statistical daily values.

As we could expect traffic sector is the main responsible to NO<sub>2</sub> levels with contributions between 73% - 89%. Second most important contribution corresponds to other mobile sources, airport mainly, with up to a 12% in some municipalities. For this pollutant agriculture is a relevant sector in municipalities away the urban metropolitan area of Madrid. In the case of ozone, again traffic sector is the main contributor with a percentage between 57% - 77%. Other mobile sources and non-industrial combustion plants are the second and the third contributor sector, respectively, with values between 7% - 19% and 7% - 12%. CO results are very similar than those obtained for O<sub>3</sub> with a most relevant contributor is traffic sector in some municipalities (Getafe and Leganés) more industrialized. PM<sub>10</sub> and PM<sub>2.5</sub> main contributor is traffic sector (33% - 59%). In comparison with NO<sub>2</sub> or O<sub>3</sub> the percentage is lower and the relevancy of the other sectors is higher. Agriculture affects an 11% - 36%, being most important for PM<sub>10</sub> than PM<sub>2.5</sub>; and S346 provides a percentage of 8% - 21% to the particulate matter levels. Finally, the distribution of SO<sub>2</sub> contributors is different, being S2 (Alcorcón 61% and Móstoles 55%), S346 (Alcalá de Henares 39%) or S8 (Alcobendas 43% and Coslada 48%) the main contributors to the air quality levels.

Results achieved are according with the same obtained for [27]. The urban metropolitan area of Madrid is strongly dominated by local sources, mainly traffic. In this area natural emissions are not important, and only provide a remarkable contribution in areas far away of Madrid (up to 5% for  $PM_{10}$  and  $O_3$ ).



Figure 2. Contribution of the emission sectors (snap and natural) to the air quality levels for different municipalities in the region of Madrid.

## 3.3. Effect of Mitigation Measures over Air Quality Levels

As we can comment previously a sensitivity analysis has been made considering all mitigation measures of **Table 3** and comparing with the results obtained in the base case. Real emissions (industry, traffic, natural, etc.) from the emission inventory are considered in the base scenario. In order to analyze the effect of the mitigation plan, the comparison has been made in some daily statistical values; focus our attention on NO<sub>2</sub> and O<sub>3</sub>.

Geographically results are shown in the air quality zones of the region of Madrid

(<u>http://gestiona.madrid.org/azul\_internet</u>) or municipalities, depending if results are provided by d03 or d04 domain. In Figure 3 and Figure 4, the difference and the relative difference obtained in any cell which is contained in the air quality zone for NO<sub>2</sub> and O<sub>3</sub>. As modelling periods have been selected using NO<sub>2</sub> and O<sub>3</sub> highest levels



Figure 3. Difference (left) and relative difference (right) of daily maximum 1-h of  $NO_2$  (up) and  $O_3$  (bottom) between Plan Azul + scenario and base case scenario over the whole region of Madrid.

criterion, for this pollutants the results corresponds to the five periods defined in **Table 1** (the effect of the mitigation plan is analyzed during episodes while  $NO_2/O_3$  levels are higher than the average annual value). In the rest of cases (PM<sub>10</sub>, PM<sub>2.5</sub>, CO and SO<sub>2</sub>), the results correspond to the average of ten periods defined in **Table 1**.

The effect of the mitigation plan directly results in a reduction of the levels of primary pollutants such as NO<sub>2</sub>. The highest nitrogen dioxide reductions are reached in Madrid city centre and around the big neighbour towns. The application of Plan Azul + mitigation plan reduces about 15% of nitrogen dioxide values in Madrid air quality zone and Corredor del Henares air quality zone; 9% in Cuenca del Alberche air quality zone; 8% in Urbana Noroeste air quality zone; 7% in Urbana Sur air quality zone; and 3% in Cuenca del Tajuña air quality zone.

The comparison of the effect over NO<sub>2</sub> hourly maximum values between base case scenario and Plan Azul + mitigation plan is showed in **Table 6**. We show mean and maximum difference corresponding to the average and the maximum of grid cell values for each air quality zone. NO<sub>2</sub> hourly maximum values are reduced up to  $11 \,\mu gm^{-3}$  in Madrid air quality zone, and up to  $9 \,\mu gm^{-3}$  in Corredor del Henares air quality zone.

The effect of mitigation plans over ozone does not produce a direct reduction of this pollutant. The effect of the mitigation plans depends on the kind of area (urban, suburban or rural), on the effect over volatile organic compounds emissions of every measure, and on the weekend effect [56]-[58]. There may be a reduction of  $NO_x$  and NMCOVs, but this reduction may not be sufficient to reduce ozone or other factors could involve the elimination of the potential ozone depletion. In this sense, the influence of the actions that lead to the reduction of



Figure 4. Difference (left) and relative difference (right) of daily maximum 1-h of NO<sub>2</sub> (up) and O<sub>3</sub> (bottom) between Plan Azul + scenario and base case scenario over the urban metropolitan area of Madrid.

Air Quality Zone	NO <sub>2</sub> Max. 1h ( $\mu$ gm <sup>-3</sup> )	Mean Difference (µgm <sup>-3</sup> )	Maximum Difference (µgm <sup>-3</sup> )
Madrid	76	-2.53	-11.30
Corredor del Henares	58	-1.24	-8.97
Urbana Sur	51	-0.92	-3.68
Urbana Noroeste	42	-1.37	-3.24
Sierra Norte	21	-1.03	-2.47
Cuenca del Alberche	30	-1.63	-2.75
Cuenca del Tajuña	24	-0.28	-0.69

Table 6. Effect of mitigation plans over NO2 1-h Maximum values in the Air Quality Zones.

pollutants should be considered in a potential increase in tropospheric ozone concentrations in the study area. For these reasons, when Plan Azul + have been developed, testing has been realized to obtain reductions of NO<sub>2</sub> without high increases of O<sub>3</sub>, or increasing ozone only in those locations where ozone levels are lower. In this way, the application of Plan Azul + increases about 2% of ozone values in the region of Madrid. The highest increase of ozone levels is around the city centre, where there is the highest reduction of pollutants as NO<sub>2</sub>. O<sub>3</sub> hourly maximum values increase up to  $6 \,\mu \text{gm}^{-3}$  in Madrid air quality zone, and up to  $4 \,\mu \text{gm}^{-3}$  in Corredor del Henares air quality zone. Table 7 is showed the effect of Plan Azul + over every air quality zone. We show

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Air Quality Zone	O <sub>3</sub> Max. 1h (µgm <sup>-3</sup> )	Mean Difference (µgm <sup>-3</sup> )	Maximum Difference (µgm <sup>-3</sup> )
Madrid	84	1.14	3.85
Corredor del Henares	89	0.27	3.28
Urbana Sur	90	0.19	1.23
Urbana Noroeste	91	0.30	1.79
Sierra Norte	95	0.06	0.27
Cuenca del Alberche	89	0.20	0.70
Cuenca del Tajuña	94	0.02	0.09

**Table 7.** Effect of mitigation plans over  $O_3$  1-h Maximum values in the Air Ouality Zones.

mean and maximum difference corresponding to the average and the maximum of grid cell values for each air quality zone.

For the rest of pollutants the effect of the Plan is not so remarkable, with global reductions of CO,  $PM_{10}$ ,  $PM_{2.5}$  and  $SO_2$  lower than 5%. Anyway, we have identified that Plan Azul + have a local effect over these pollutants in specific locations as, for example, near the International Airport of Madrid, increasing the effect up to a 30%.

Using the modelling year 2010, we estimate that the application of the Plan could reduce the number of exceedances of the hourly limit value of  $NO_2$  in a 20%, and the exceedances of  $PM_{10}$  in a 5%. Not changes in the number of ozone exceedances have been estimated.

## 4. Conclusions

A coupled air quality modelling system has been used for the design and preliminary evaluation of an air quality plan over a region with exceedances and high levels of atmospheric pollutants. The numerical modelling system accomplishes with the European Directive requirements and its accuracy is good enough as to use for evaluate air quality plans and mitigation measures. Results of evaluation also show that the system provides high accuracy over locations with higher levels of NO<sub>2</sub> and O<sub>3</sub>.

Results obtained show that the main sector contributor to the emissions and air quality levels over Madrid is the road traffic, followed for other mobile sources and non-industrial combustion plants as second and third contributors respectively. Moreover, air quality levels are determined basically for local contributions in Madrid and its urban metropolitan area. In this way, mitigation measures designed and evaluated have been focused on this sector.

We have observed that the Plan designed is optimum to reduce NO<sub>2</sub> levels, reducing up to  $11 \ \mu gm^{-3}$  the concentration over the city of Madrid. Highest reductions of this pollutant are located over urban areas with traffic influence, coinciding with regions where NO<sub>2</sub> levels traditionally are higher. The air quality plan has the effect with opposite sign and provides slight increases of ozone concentration (1% - 2%) in areas with typically ozone levels which are low. We expect that the application of this Plan will reduce the number of exceedances of the NO<sub>2</sub> limit value and not affects considerably to the number of exceedances for ozone. Mitigation measures defined in the plan do not affect remarkably to the levels of CO, SO<sub>2</sub> or particulate matter.

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