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Analysis of Contributions to NO_2 Ambient Air Quality Levels in Madrid City (Spain) through Modeling. Implications for the Development of Policies and Air Quality Monitoring

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As environmental standards become more stringent (e.g. European Directive 2008/50/EC), more reliable and sophisticated modeling tools are needed to simulate measures and plans that may effectively tackle air quality exceedances, common in large cities across Europe, particularly for NO_2. Modeling air quality in urban areas is rather complex since observed concentration values are a consequence of the interaction of multiple sources and processes that involve a wide range of spatial and temporal scales. Besides a consistent and robust multi-scale modeling system, comprehensive and flexible emission inventories are needed. This paper discusses the application of the WRF-SMOKE-CMAQ system to the Madrid city (Spain) to assess the contribution of the main emitting sectors in the region. A detailed emission inventory was compiled for this purpose. This inventory relies on bottom-up methods for the most important sources. It is coupled with the regional traffic model and it makes use of an extensive database of industrial, commercial and residential combustion plants. Less relevant sources are downscaled from national or regional inventories. This paper reports the methodology and main results of the source apportionment study performed to understand the origin of pollution (main sectors and geographical areas) and define clear targets for the abatement strategy. Finally the structure of the air quality monitoring is analyzed and discussed to identify options to improve the monitoring strategy not only in the Madrid city but the whole metropolitan area.

Keywords: Air Quality Modeling; Source Apportionment; NO_2; CMAQ; Urban Air Quality; Madrid

Introduction

Despite recent efforts made in Europe, a large percentage of the European population is currently exposed to air pollutant concentrations above the European Union and World Health Organization reference levels (EEA, 2013). This is mainly relevant to urban areas, where the majority of the European population lives, leading to adverse effects on public health. Even from the regulatory point of view, there are still pending issues in Europe. For instance, air concentrations of NO_2 lags clearly behind the decreasing trend of NO_X emissions (Guerreiro, C. et al., 2010). As a consequence, the main cities in Europe are finding serious difficulties to comply with the air quality standards defined in the EU Directive 2008/50/EC for NO_2. Non-attendant areas and urban agglomerations are legally bound to develop and apply air quality plans (AQPs) to meet legal standards. Such plans and strategies are based on abatement measures that usually entail significant social, economic and political costs. Therefore, it is important to guarantee that any measure is actually aimed at the sectors causing air quality problems so they can effectively improve air quality levels.

The inherent complexity of urban environments requires simulation tools to assess air quality levels to be able to support the analysis and evaluation of a variety of policies and emission abatement measures previous to their implementation (Denby, B. et al., 2011).

This evaluation must begin with a source apportionment analysis that can provide essential information regarding the basic emission abatement strategy, the maximum feasible air quality improvement related to the main emitting sectors as well as the external constrains. This paper reports on the source apportionment carried out for the Madrid city (Spain) for the development of an AQP to comply with NO_2 standards in the near future (Borge, R. et al., 2014). Despite satisfying a legal requirement, this analysis is also useful to assess the layout of the air quality monitoring stations and to identify options to improve the monitoring strategy in the region.

Case Study

Madrid is the capital and largest city in Spain, located in the centre of the Iberian Peninsula with a total population of 5 million people in its metropolitan area. Madrid City Council and surrounding municipalities are connected through a dense road network and constitute a continuous urban area with a large density of air quality monitoring stations (**Figure 1**).

Despite the experienced population and traffic increase, air quality levels have improved in the city over the last decade. However, some pollutants like nitrogen dioxide (NO_2) still exceed the limit values (LV) according to the European legislation. The NO_2 annual average recorded in most of the city's traffic air quality monitoring stations is above the LV (40 µg/m^3), as it can be seen in the records of the monitoring station

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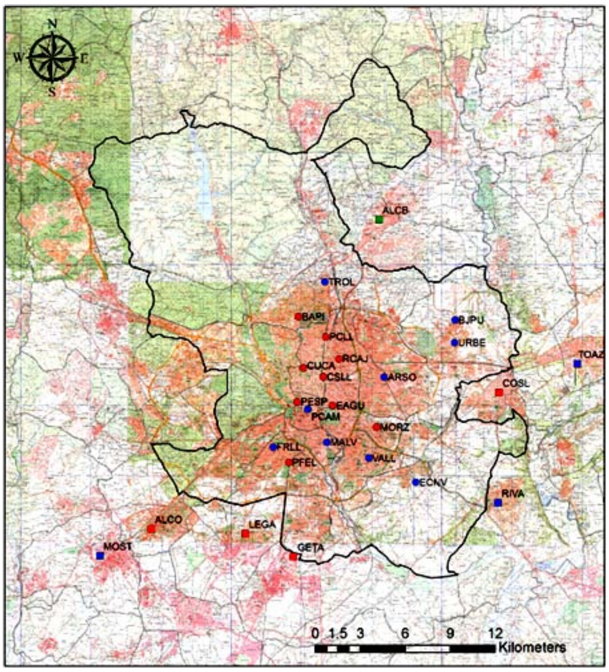

Figure 1.
Madrid city metropolitan area and air quality monitoring stations. Air quality monitoring stations of the Madrid City council and Madrid Greater Area networks are represented by circles and squares respectively. Traffic stations are represented in red, urban background in blue and industrial stations in green.

(**Figure 2**). This phenomenon is basically attributed to heavy traffic levels and to a strong dieselization of the fleet in recent years (Kassomenos, P. et al., 2006).

Methodology

Modeling System and Domains

The mesoscale modeling system is based on the Weather Research and Forecasting (WRF) (Skamarock & Klemp, 2008), the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system (Institute for the Environment, 2009), and the Community Multiscale Air Quality (CMAQ) (Byun and Ching, 1999; Byun and Schere, 2006). Details about specific configuration and adaptation to the Spanish conditions can be found respectively in Borge et al. (2008a, 2008b, 2010).

Four nested domains were used in order to capture international, national, regional and local contributions to NO_2 to ambient concentration in Madrid with a maximum resolution of 1 km^2 (**Table 1**).

Special care was taken to keep the consistency among the emission inventories used across the scales. This is a crucial issue for the source apportionment exercise since it includes an analysis of emission contributions from different geographic areas. Further information about emission compilation and harmonization can be found in Borge, R. et al. (2014). The emission inventory for the innermost domain (**Figure 1**) is based on a combination of top-down and bottom-up methods specific for each of the main emission sources in the city. The most relevant feature is the integration of a the City Concil's mesoscale road traffic model within the emission model so very detailed, 1-h, 1-km^2 resolved emissions can be computed for any given traffic situation (Borge, R. et al., 2012a). Total emissions at SNAP group level in the innermost modeling domain (**Figure 1**) are given in **Table 2**.

Table 1.
Domains for the mesoscale modelling system.

Domain	Geographic scope	X-Y dimensions (km)	Horizontal resolution (km)
D1	Europe	6144 × 5376	48
D2	Iberian Peninsula	1200 × 960	16
D3	Greater Madrid Region	192 × 192	4
D4	Madrid Metropolitan Area	40 × 44	1

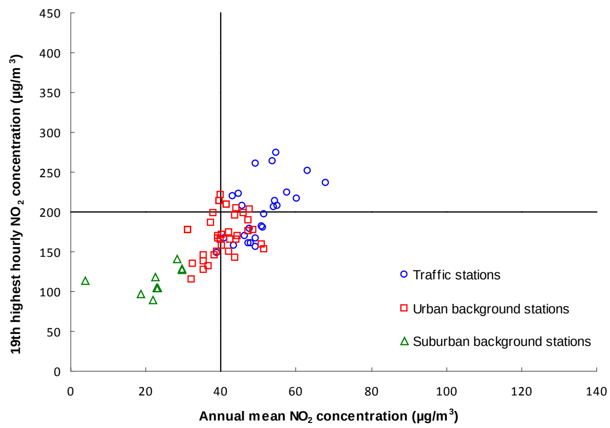

Figure 2.
Observed NO_2 values (corresponding to the annual and hourly NO_2 limit values defined in the European AQ Directive) in the Madrid air quality monitoring network for the years 2010-2012.

Source Apportionment Methodology

A zero-out methodology was followed in this application. The contribution of a particular emission source or region can be estimated through the brute force method (BFM), sometimes referred to as single-perturbation method (Samaali, M. et al., 2011 and references within). This method relies on the analysis of the change in the pollutant concentration that would occur if a given emitting source is removed from the simulation (usually referred to as zero-out sensitivity runs). This approach has been used in the past to isolate the response of complex, nonlinear systems to one particular sector in source apportionment and sensitivity analysis (Cohan, D.S. et al., 2005). This method has inherent limitations in accurately describing sensitivities but it may be useful to approximate the effect of potential emission reductions in a particular source or origin area as pointed out before in several studies (Koo, B. et al., 2009; Carmichael, G. et al., 2010; Leung et al, 2007). This analysis was originally performed separately for those locations inside and outside the Madrid municipality, since this is a legal requirement. For this study, both analyses have been merged so a more scientifically-sound and coherent view can be given. Reductions of 100% (zero-out) were simulated for the most relevant anthropogenic emissions:

- Residential, commercial and institutional combustion (RCI)

Table 2.
Emissions in the Madrid metropolitan area (metric tons per year).

SNAP group*	CO	NH_3	NO_X	PM_{10}	$PM_{2.5}$	SO_2	VOC
01	225	0	243	50	29	1128	1
02	10004	0	3680	520	410	2731	1104
03	2238	0	10689	265	210	2494	1217
04	1083	130	108	51	32	70	3782
05	0	15	0	0	0	0	2056
06	0	212	0	0	0	0	48828
07	22070	250	27961	1506	1205	157	4365
08	2711	0	4171	360	360	287	769
09	441	2036	1769	26	26	6	5267
10	357	1543	56	90	13	0	17
11	32	605	125	0	0	0	4682
latoT	**39161**	**4791**	**48802**	**2868**	**2285**	**6873**	**72088**

*SNAP groups: 01—Combustion in energy and transformation industries; 02—Non-industrial combustion plants; 03—Combustion in manufacturing industry; 04—Production processes; 05—Extraction and distribution of fossil fuels; 06—Solvent and other product use; 07—Road transport; 08—Other mobile sources and machinery; 09—Waste treatment and disposal; 10—Agriculture; 11—Other sources and sinks (Nature).

(SNAP 02);
- Industry (SNAP 03 & 04);
- Road traffic (SNAP 07);
- Other mobile sources (planes, trains, machinery) (SNAP 08);
- Sources outside the modeling domain (rest of the country and international contributions).

The total impact and therefore, the maximum theoretical benefits that can be harvested by implementing abatement options in these sectors were derived from the comparison of the assessment of the individual runs with the base case (considering all emissions). All the emission processing and scenarios for the sensitivity runs were done through the SMOKE system. Further details on this procedure can be found in R. Borge et al., (2012b).

Results and Discussion

Individual results for the model grid cells where air quality monitoring traffic stations are located are shown in **Figure 3**. As expected, according to their classification, NO_2 concentration levels at all these locations are clearly dominated by contributions form road traffic, with shares ranging form 59.8% to 75.3% and an average value of 69.4%. It is worth noting, however that this contribution seems to be smaller for the stations located at the right-hand side of the graph. Those stations (COSL, ALCO, LEGA, GETA) correspond to monitoring stations of the Greater Madrid Region, that they are located in the outskirts of the urban area. In contrast, the influence of sources outside the Madrid Region (rest of the country) is larger, which is totally reasonable. While national contributions inside the Madrid municipality are around 10% of total NO_2 concentration levels, they reach a 15% as an average in the monitoring stations closer to the modeling boundary. A similar trend is seen for international contributions, although the differences are smaller (3.4% Vs 5.6%). In all cases local emissions (those originated inside the region) are responsible for the majority of the observed concentrations (around 85% as an average). The second source in importance is the RCI sector, responsible for 4.5% of NO_2 ambient concentration. This ratio exhibits a larger variability, with larger values in the city center (RCAJ, CUCA) except for some locations where other sectors (mainly SNAP 09) have a bigger influence locally due to the proximity to waste management plants. The contributions from other mobile sources are similar in all the traffic stations (around 4.2%). This is probably due to the difficulty to allocate emissions from gardening and cleaning machinery as well as construction and industrial vehicles, that are spatially distributed all over the city depending on the population and road network density. Finally, the results point out that the influence of industry in the air quality of the city center is negligible; (less than 2%). This makes sense, since the industrial activity in Madrid (very moderate) is located in the surroundings of the city. This can be clearly spotted in the results corresponding to the stations of the Greater Madrid Region, mainly Coslada (COSL) where 9.4% of NO_2 is produced by industry. It should be noted that the SNAP 01 sector is totally unimportant in Madrid since there are no power plants in the region.

Results form non-traffic stations are shown in **Figure 4**. Despite most of these stations are considered urban background locations, the structure of NO_2 contributions is rather similar to that shown in **Figure 3**. Contributions of road traffic emissions to air quality background levels roughly represent 2/3 of total pollution. As expected, industry has a bigger influence in these environments not directly exposed to traffic emissions. This is particularly true for those stations of the Greater Madrid Region (again those in the right-hand side in **Figure 4**) where industrial activity clearly constitutes the second most important pollution source (16.3% as an average). Although Alcobendas (ALCB) is the only station label as industrial, it does not present a specially high contribution from this sector (14.7%), lower that other stations such as Móstoles (MOST) and Torrejón (TOAZ) (15.1% and 21.2% respectively). The contribution of other sectors not explicitly considered in the analysis represents around 5.5% of total NO_2 values as an average, although it presents quite some variability. Barajas Pueblo presents an unusually high share for this sector (16.0%). This may be due to the influence of emissions generated in the Madrid/Barajas international airport neither included in the SNAP 08 nor the SNAP 07 sector.

Conclusion

A comprehensive source apportionment study was performed in the Madrid metropolitan area through the application of a multi-scale, multi-pollutant air quality modeling system (WRF-SMOKE-CMAQ). The study was based in the single-perturbation method, i.e. removing emissions from each of the main sectors one at a time and comparing the results with those of the baseline scenario (complete emission inventory).

The results from a series of locations throughout the inner-most modeling domain indicate the different relative weight of the contributions of the sectors analyzed. Despite these differences, it can be seen that road traffic is the main contributor to

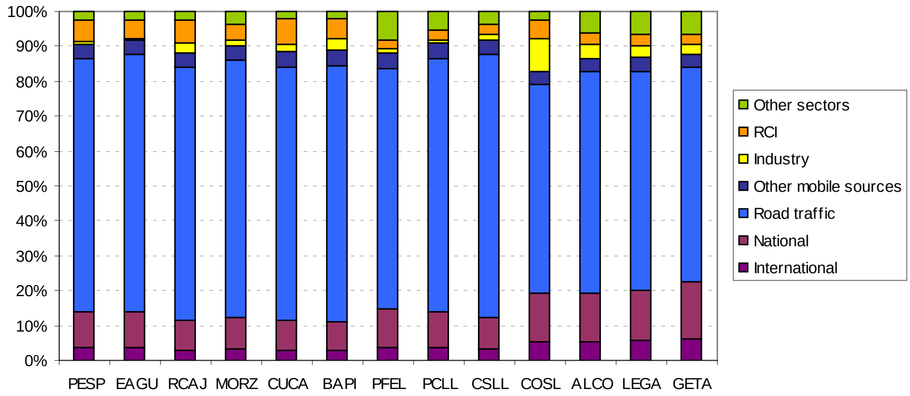

Figure 3.
Results of the NO_2 source apportionment analysis performed at the location of traffic air quality monitoring stations.

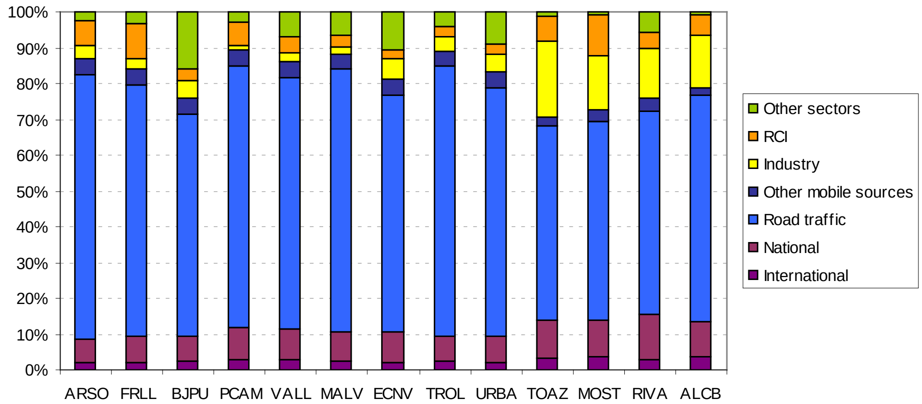

Figure 4.
Results of the NO_2 source apportionment analysis performed at the location of (mostly) urban background monitoring stations.

NO_2 levels all over the region, specially in the city center with contributions up to 75% of total concentration (86% if non-local sources, those outside the Greater Madrid Region are not considered). Contributions form other countries are relatively unimportant (around 1% in most cases). Similarly, pollution from outside the region has a small incidence (approximately 3% - 4%). National influences (sources in Spain but outside the Greater Madrid Region) are more important, mainly in the city outskirts, but it represents less than 10% of total NO_2 as an average) This means that NO_2 levels observed in Madrid are basically due to local sources, so air quality issues can be tackled by local AQP. In particular, these plans should be clearly directed to reduce emissions in the road traffic sector, responsible of 69.4% and 66.4% of nitrogen dioxide ambient concentration observed in traffic and urban background locations respectively (as an average). These figures indicate that abatement emissions in the road traffic sector may be particularly effective. As shown in **Table 2**, the relative contribution of the SNAP 07 sector is 57.3% in the geographical domain analyzed (D4).

While their contribution in terms of emissions is well below 60%, their share in resulting NO_2 ambient concentration levels is close to 70%. This may be related to two factors: 1) emissions form road traffic are released at ground level (unlike those of the RCI or industrial sectors) and 2) their NO_2/NO_X emission factor is higher due to a strong fleet dieselization phenomenon (R. Borge et al., 2012).

This conclusion is clearly reflected in the Madrid AQP for the horizon 2011-2015 (Madrid City Council, 2012). Consistently with the results of the source apportionment exercise performed, most of the measures in the plan (up to 70) were targeted to the road traffic sector. This abatement strategy includes a Low Emission Zone (LEZ), reduction of road capacity and pedestrianized areas in the city center, renovation of city bus fleet to incorporate clean technologies (electric, hybrid natural gas-fuelled buses), etc. According to the results shown in this AQP would achieve a 40% reduction of NO_X emissions from the road traffic sector in the modeling domain. As a result of all these measures, a global decrease of 31% in NO_X emissions is expected in the year 2014 within D4 (respective to the emissions of 2007 used as reference scenario for the source apportionment analysis presented here). Additional model runs (R. Borge et al., (2014) indicate that annual NO_2 levels may be reduced by 34% as an average; approximately 15 $\mu g/m^3$ in the city centre, also with an important impact in the metropolitan area (-7 $\mu g/m^3$ as an average in the modeling domain). 1-hour concentration peaks may also decline by 40% approximately in most of the city (**Figure 5**).

The second most important contributor, far from road traffic, is the RCI sector, responsible for approximately 5% of ob-

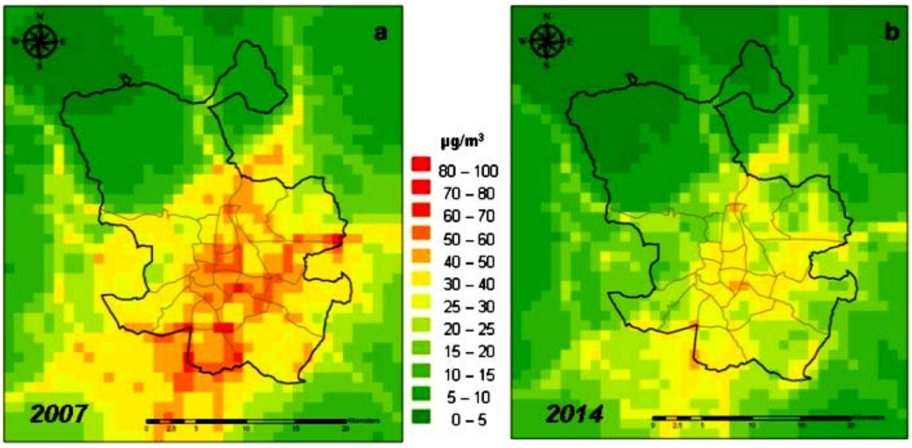

Figure 5.
Expected effect of the Madrid AQP in NO_2 concentration. Annual average for year 2007, considered for the source apportionment study (a) and 2014, temporal horizon of the AQP (b).

served NO_2 in Madrid. A similar influence can be attributed to mobile sources other than road traffic. The contribution of industry is low in general although it may be important in the surroundings of the city. According to the source apportionment study performed, the industrial sources in the region are responsible of 5% and 16% of NO_2 levels in the traffic and urban background stations of the Greater Madrid Region, and therefore may constitute a sensible target for additional measures, especially in the east area of the Madrid metropolitan area.

As for the air quality monitoring strategy, the results form this study indicate that the relative amount of traffic stations, although high, may be adequate since traffic is mainly responsible for air quality problems in the region. However, further analysis should be done to understand to what extend this stations may be providing redundant results (the source apportionment results are very similar in all the stations) and therefore the air quality monitoring network may be simplified and reduced, considering the minimum requirements established in the Directive 2008/50/EC. On the other hand, the potential need for some industrial stations in the eastern part of the Madrid metropolitan area may be considered.

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REFERENCES

Borge, R., Alexandrov, V., del Vas, J.J., Lumbreras, J., & Rodríguez, E. (2008a). A comprehensive sensitivity analysis of the WRF model for air quality applications over the Iberian Peninsula. *Atmospheric Environment, 42,* 8560-8574. http://dx.doi.org/10.1016/j.atmosenv.2008.08.032

Borge, R., de Miguel, I., de la Paz, D., Lumbreras, J., Perez, J., & Rodríguez, M. E. (2012a). Comparison of road traffic emission models in Madrid (Spain). *Atmospheric Environment, 62,* 461-471. http://dx.doi.org/10.1016/j.atmosenv.2012.08.073

Borge, R., López, J., Lumbreras, J., Narros, A., & Rodríguez, E. (2010). Influence of boundary conditions on CMAQ simulations over the Iberian Peninsula. *Atmospheric Environment, 44,* 2681-2695. http://dx.doi.org/10.1016/j.atmosenv.2012.08.073

Borge, R., Lumbreras, J., & Rodríguez, M. E. (2008b). Development of a high-resolution emission inventory for Spain using the SMOKE modelling system: A case study for the years 2000 and 2010. *Environmental Modelling & Software, 23,* 1026-1044. http://dx.doi.org/10.1016/j.envsoft.2007.11.002

Borge, R., Lumbreras, J., Pérez, J., de la Paz, D., Vedrenne, M., de Andrés, J. M., & Rodríguez, M. E. (2014). Emission inventories and modeling requirements for the development of air quality plans. Application to Madrid (Spain). *Science of the Total Environment, 466-467,* 809-819.

Borge, R., Lumbreras, J., Pérez, J., de la Paz, D., Vedrenne, M., & Rodríguez, M. E. (2012b). Emission inventories and modeling activities for the development of air quality plans in Madrid (Spain). *2012 USEPA International Emission Inventory Conference "Emission Inventories—Meeting the Challenges Posed by Emerging Global, National, Regional and Local Air Quality Issues",* Tampa, 13-16 August 2012.

Byun, D. W., & Ching, J. K. S. (1999). Science algorithms of the EPA models-3 community multi-scale air quality (CMAQ) modeling system. EPA/600/R-99/030, US EPA National Exposure Research Laboratory, North Carolina.

Byun, D. W., & Schere, K. L. (2006). Review of the governing equations, computational algorithms, and other components of the models-3 community multiscale air quality (CMAQ) modeling system. *Applied Mechanics Reviews, 59,* 51-77. http://dx.doi.org/10.1115/1.2128636

Carmichael, G., & Wild, O. (2010) Global and regional modelling. In: Hemispheric Transport of Air Pollution. *Task Force on Hemispheric Transport of Air Pollution.*

Cohan, D. S., Hakami, A., Hu, Y., & Russell, A. G. (2005). Nonlinear response of ozone to emissions: Source apportionment and sensitivity analysis. *Environmental Science & Technology, 39,* 6739-6748. http://dx.doi.org/10.1021/es048664m

Denby, B., Douros, I., & Fragkou, L. (2011). Modelling of nitrogen dioxide (NO_2) for air quality assessment and planning relevant to the european air quality directive. *Forum for Air Quality Modelling in Europe (FAIRMODE) Guidance Document.*

European Environment Agency (EEA) (2013). Air quality in Europe—2013 report. EEA Report No 9/2013.

Guerreiro, C., Horálek, J., de Leeuw, F., Hak, C., Nagl, C., Kurfürst, P., & Ostatnicka, J. (2010). Status and trends of NO_2 ambient concentrations in Europe. ETC/ACC Technical Paper 19/2010.

Institute for the Environment. SMOKE v2.7 User's Manual (2009). University of North Carolina, Chapel Hill.

Kassomenos, P., Karakitsios, S., & Papaloukas, C. (2006). Estimation of daily traffic emissions in a South-European urban agglomeration during a workday. Evaluation of several "what if" scenarios. *Science of the Total Environment, 2-3,* 480-490. http://dx.doi.org/10.1016/j.scitotenv.2006.08.018

Koo, B., Wilson, G. M., Morris, R. E., Dunker, A. M., & Yarwood, G. (2009). Comparison of source apportionment and sensitivity analysis in a particulate matter air quality model. *Environmental Science & Technology, 43,* 6669-6675. http://dx.doi.org/10.1021/es9008129

Leung, F. Y. T., Logan, J. A., Park, R., Hyer, E., Kasischke, E., Streets, D., & Yurganov, L. (2007). Impacts of enhanced biomass burning in the boreal forests in 1998 on tropospheric chemistry and the sensitivity of model results to the injection height of emissions. *Journal of Geophysical Research, 112,* D10313. http://dx.doi.org/10.1029/2006JD008132

Madrid City Council (2012). Madrid's air quality plan 2011-2015. Approved by the Governing Board of Madrid City Council on April 26, 2012. General Directorate of Sustainability, Government Division of Environment, Safety and Mobility, Madrid City Council.

Samaali, M., Bouchet, V. S., Moran, M. D., & Sassi, M. (2011). Application of a tagged-species method to source apportionment of primary PM2.5 components in a regional air quality model. *Atmospheric Environment, 45,* 3835-3847. http://dx.doi.org/10.1016/j.atmosenv.2011.04.007

Skamarock, W. C., & Klemp, J. B. (2008). A time-split nonhydrostatic atmospheric model. *Journal of Computational Physics, 227,* 3465-3485. http://dx.doi.org/10.1016/j.jcp.2007.01.037