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Analysis and comparison of the national and regional emission inventories for the Greater Madrid Region through air quality simulations

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“Pour moi, ce n’est que l’atmosphère environnante
qui donne des sujets à leur juste valeur.”

Claude Monet (1840-1926)

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1. ABSTRACT

Emission inventories are extensive databases that intend to accurately describe the polluting activities that occur across a given geographic domain. These inventories are compiled basically to comply with reporting obligations and not for scientific use, so they rely on a series of hypotheses that might not be correct or reasonable. According to the **scale** of the domain that is being described, the availability of information will vary as well as the set of applied assumptions. These factors will have a direct influence in the quality, accuracy and representativeness of such an inventory. It could happen that inventories describing a small - scale domain stick to methodologies implemented by inventories describing larger scales in which the first domain is nested and still, exhibit considerable differences.

This study compared and contrasted **two** emission inventories describing the **Autonomous Community of Madrid** and neighbouring provinces under an air quality simulation approach. The chosen inventories were the National Emissions Inventory, compiled by the Spanish Ministry for the Environment for the whole territory of Spain and under a nation - wide perspective; and a Regional Emissions Inventory, ellaborated by the authorities of the Autonomous Community of Madrid only for the aforementioned region. The chosen air quality modelling system consisted in the Weather Research and Forecast model (*WRF*), the Sparse Matrix Operator Kernel Emissions (*SMOKE*) and the Community Multiscale Air Quality Model (*CMAQ*), coupled together. Predicted concentrations were then compared to observations from a 55 - monitoring stations network extended throughout the studied domain. Four air pollutants were chosen; nitrogen oxides (NO_x), sulphur dioxide (SO_2), and two sizes of particulate matter (PM_{10}, $PM_{2,5}$). The analysis was carried out attending to the *SNAP* nomenclature specified by the *EMEP/CORINAIR* methodology upon which both inventories are based. It was observed in every case that **road - traffic** was the main polluting activity.

Through the applications of statistical tools, the analysis of emissions at a cell level and cell - expansion procedures it was observed that the National Inventory shows better results for describing **on - road traffic** activities and **agriculture**, namely *SNAP07* and *SNAP10*. The accurate description of emissive activities, the good characterization of the vehicle fleet and the correct use of traffic emission factors are the main causes of such a good correlation. On the other hand, the Regional Inventory showed better descriptions for **non - industrial combustion** activities (*SNAP02*) and **industrial activities** (*SNAP03*). This inventory incorporated realistic emission factors, a reasonable fuel - mix apportionment and drew upon local information sources to describe these groups; the National Inventory to this respect relied on surrogation and national datasets which redounded in a poor reality representation. **Off - road transportation** (*SNAP08*) was similarly described by both inventories, while the rest of the *SNAP* activities showed a very marginal contribution to the overall emissions.

Further analysis revealed that **scales** can be **reconciled** through the assessment of the criteria used to describe the polluting activities. Finally a group of good - practices is presented, based on the findings of the present study. These practices strongly urge that emission inventories be compiled always with the underlying premise that the **best** approximation to reality should be attained over methodological rigour or readily available compilation guidelines.

2. INTRODUCTION

2.1. Theoretical background

The **atmosphere** is one of the five biogeochemical layers in which life develops. Its main component, **air**, is an important resource on which many living creatures rely. Its natural composition should not be altered; otherwise undesirable effects on life might take place. In general, the concept of **pollution** is that of degradation, of a loss of quality or a deviation from purity due to the introduction of external agents (Godish, 2004). Moreover, there is general consensus in affirming that in the absence of human activities at a given location, the environmental quality tends to be good enough or at least, the one determined by nature. Under this premise, the 2008/1/EC European Directive, related with the integrated pollution prevention and control defines the concept of pollution as *the direct or indirect introduction, as a result of human activity, of substances, vibrations, heat or noise into the air, water or land which may be harmful to human health or the quality of the environment, result in damage to material property, or impair or interfere with amenities and other legitimate uses of the environment* (Directive 2008/1/EC).

Clean air is considered to be a basic requirement of human health and well - being (World Health Organization, 2005). The former premises highlight the need for reaching a full understanding of the **air pollution** processes, as well as assuring that a clean air is being delivered. These needs have been recognised worldwide, and efforts have been carried out in the last decades to tackle air pollution by establishing stringent limits on emissions. It is therefore important to have enough quantitative information about such emissions, their sources and their temporal patterns in order to inform policy makers and citizens; to set rational and attainable objectives; to assess potential impacts on health and welfare; to evaluate environmental costs and benefits of certain policies; to monitor the state of the environment; to ensure liabilities are being properly assigned and so on (EMEP - CORINAIR, 2007). To attain all these, public administrations shall conduct programs for the research, testing and development of methods for sampling, measuring, monitoring, analysing and modelling air pollution processes (Clean Air Act, 2008).

Particularly, the quantification of **emissions** is quite an endeavour and sometimes it is not possible to measure them for all the existing sources directly. Not even for each one of the source types. In practice, emissions are estimated on the basis of measurements carried out at representative samples of the main sources and source types. They are further compiled into an **emission inventory** for a given domain. Emission inventories are databases that incorporate information on the location of emission sources, emission factors and activity rates (EMEP - CORINAIR, 2007).

Pulles (1999) accorded that the series of data present at an emission inventory must have four independent attributes or dimensions: (i) a chemical or physical identity of the emitted pollutant, (ii) the specific activity that causes the emission of such pollutant, (iii) the geographic location in which the emission is produced and (iv) a temporal profile. Emission inventories should have the following characteristics to be considered as best suited (Serveau et al., 2010):

- *Exhaustive*. Each and every single source must be taken into account for the total emission quan-

tification, trying to be as specific as possible.

- *Coherent*. The obtained series for a determined period of time must be homogeneous, implying consistent calculation methods and procedures.
- *Exact*. The estimations must be as exact as possible through the application of the most advanced knowledge to date.
- *Comparable*. Different inventories carried out according to the same methodology would be likely of being contrasted for better or worse results.
- *Transparent*. The series of methods and procedures used throughout the compilation process must be explicit so that sound verification processes might take place.

Regarding their construction, air pollution emission inventories can be compiled from various information sources, such as (i) estimates that apply emission factors and production inputs or outputs; (ii) real measurements that have been conducted under specific monitoring conditions and; (iii) compulsory data provided by each industry, coming from either self - monitoring practices or general estimates (Dos Santos and Moutinho, 2005).

Emission inventories describe the only direct link that exists between the atmosphere and human activities and thus, depict possible actions to be taken in order to mitigate high concentrations. They are spatially specific and the information is organised in gridded sets (Winiwarter et al., 2003). The construction of inventories might be done to cover individual urban areas, regional scales or even continents (Seika et al., 1996). Concretely Vautard et al. (2007) stated that the contribution of regional emissions to the European air quality will keep being dominant for the next decade, which highlights the need for carrying out representative emission inventories.

Since emission inventories are geographically specific, the scale upon which such inventories are compiled is an important issue to consider. Very often the chosen **scale** for the elaboration of the inventory is usually bounded by the political limits of a given geographic or administrative region. This premise underlies the fact that such region might be described by several inventories differently, usually depending on the scale upon which each one has been built. In the European Union, the compilation of emission inventories is fragmented over different national or regional agencies. The result is a series of various methods and standards for the categorisation of the sources and its emissions, even within single countries. Moreover, the emissions are collected using a local language, which complicates comparisons for a specific geographic domain for which two or more inventories exist (Maes et al., 2009).

This is completely true and it is closely related to the problem that originated this study in the firs place. Mesoscale air quality modelling needs boundary conditions that, most often, require a nested - domain approach. Therefore, the need for multiscale inventories and the problem of consistency is quasi - instrinsic to air quality modelling. Being this so, the importance of coordination between neighboring member states in drawing up and implementing air quality plans in short - term actions seems evident, especially for densely populated regions such as Europe (Directive 2008/50/EC). This scale factor is a key concept that must be taken into consideration by countries when complying with the Convention on Long Range Transboundary Air Pollution (*CLRTAP*), adopted in Geneva in 1979, as well as the different protocols established for specific pollutants; Helsinki (1985) and Oslo (1994) for sulphur; Sofia (1988) for nitrogen oxides; Geneva (1994) for volatile organic compounds; Århus (1998) for persistent organic pollutants and heavy metals (EMEP - CORINAIR, 2007; Borge et al., 2009).

To this respect, a difference should be noted regarding the specific objective that wants to be attained through the usage of emission inventories. Using inventory data for policy purposes might have assessment and control procedures as ultimate goal. A policy maker might be only interested in the fact that the inventory data intended for use are recognised as valid by the institutions involved in such policy making process. When using inventories for scientific research, inventory data must be evaluated as whether they are true or not as well as if they describe reality in a reliable way (Pulles, 1999).

Under both points of view, the **scale factor** is a very challenging issue. The difference in the development of scaled inventories lies in the level of detail of the input data, hypotheses, and analysed parameters. National inventories require a broader approach for emissions estimation, as they encompass sources with larger geographic scales, including air and marine transport and the national energy grid. An emission inventory at national level also depends upon many factors such as vehicle technology, socio - economic characteristics, transport policy, etc. This information is intrinsically included in each respective local inventory (Alonso et al., 2010).

Thus, it is almost imperative to carry out sound comparisons between inventories. Such comparisons might seek answers to two specific questions: (i) Is the contribution of specific sources to the total emissions for a specified area different? (ii) Is that result connected to the specific features of the area or to the way the inventory has been produced? However, in order to validly compare the emission data from several locations, a common basis for its categorisation is necessary (Seika et al., 1996). Also, special attention should be put to the size of populations and ecosystems exposed to air pollution and to the classification of the member states territory into zones reflecting the population density (Directive 2008/50/EC).

Usually, generic methodologies are implemented for describing a number of emitting activities, such as road transport, railways, industry, airports, shipping, and sources of domestic and commercial nature. These methodologies rely on the usage of readily available activity data, emission factors and first - hand assumptions. The fact of data being readily available is considered as the main restriction to develop consistent methodologies for the compilation of emission inventories; this fact also emphasises that the usual estimation procedures intend to obtain the same or similar data for all the administrative areas (Lindley et al., 2000).

The atmospheric emissions are usually estimated in terms of a statistical variable called **activity** (A) and specific **emission factors** (E_F) that describe the relationship between such activities and the emissions:

$$E = A \times E_F \tag{2.1}$$

This approach yields significant results if the emission factors are adequately chosen so that they describe accurately the emission behaviour under different conditions (Winiwarter et al., 2010). For this reason, the quantification of emissions is carried out according to their specific nature and type; scientists classify possible sources according to their behaviour at emission, namely point sources, area sources and line sources. Point sources are usually industrial facilities, power plants or generally, large emission outlets who have specific data, capacity, throughput and operating conditions. Area sources are a set of smaller sources that behave in a diffuse way and can thus be modelled as an emitting area. Finally, line sources are essentially emissive linear structures such as roadways, railways, inland navigation, shipping and aviation pathways, among others (EMEP - CORINAIR, 2007).

Two basic approaches for the compilation of emission inventories exist: the top - down and the bottom - up approaches. **Top - down** approaches have as starting point a given array of information that

corresponds to a bigger scale than the one intended to describe, thus needing to disaggregate it throughout the smaller scales. Their main advantages are that they rely on existing institutional data systems and that they can be calculated through the use of standardised processes (Alcorn and Lloyd, 2009). A top - down methodology is highly recommendable for identifying areas that are likely of being improved. Taking into account that the uncertainties associated with the emission inventories and the physical measurements is essential for carrying out a top - down approach. This methodology is indeed designed to diminish any possible uncertainties that might be related with the emission inventory and its comparisons (Funk et al., 2001).

Bottom - up methodologies rely on the use of more local inputs that better estimate the emission factors and activities, thus yielding more accurate levels on the local scale emission rates (Cook et al., 2006). Bottom - up inventories require a close cooperation between the specific emitting installations and the inventory compilers, as well as the use of customised installation templates or profiles. Bottom - up inventories are advantageous for defining boundaries, revealing data and emission factor gaps due to its higher resolution and detail degree (Alcorn and Lloyd, 2009). The main constraint that this approach has is the requirement of a large collection of datasets and a series of calculations, as well as a manual rollup or aggregation of results once level increases (Bellasio et al., 2007; Alcorn and Lloyd, 2009).

All the input data sets, such as activity data, emission factors and measurements have associated uncertainties. Therefore, the total emission inventory uncertainty is a function of the individual uncertainties associated to each one of the input parameters (Rypdal and Winiwarter, 2001). Potential errors might lie in the statistics used to describe the polluting activity, the emission factors per activity unit as well as the series of assumptions made to obtain such emission factors (Zachariadis and Samaras, 1997). Inventories described under a spatial resolution might be subject also to potential uncertainties due to the distribution of emissions along such a spatial dimension. Agreement exists in affirming that producing emission inventories without any statistical error is quite a difficult, not so say impossible task. When sources increase in number and variety, as well as the potential estimation methods used for their description, emission inventories become ultimately complex (Lindley et al., 2000).

Activity data are often subject to sizeable differences, basically because most of them have not been originally collected and prepared purposely as inputs for an emission inventory. Some of these data are not readily available and have been gathered through the use of different statistical procedures, regardless of the end use that they might have. Usually, activity data exhibit consistent yearly changes that need to be considered for inventory consistency purposes. It may be the case that compiling an emission inventory might need to consider site - specific activity data, either because they are the only data available or because such site is quite representative of the local emissions. In every case, the conciliation of the national - local issue is of extreme importance (Hiraishi et al., 1996).

Emission factors may carry higher uncertainties due to the lack of irrelevant measurements and a series of generalisations, as well as an insufficient understanding of the emission - generating process. On the contrary, activity data are usually statistical data, whose uncertainties are associated with the surveying process and the lack of investigations. In some situations, the uncertainties carried by emission factors can be estimated through the use of empirical data (Rypdal and Winiwarter, 2001). While evaluating the general quality of such data, Frey (2007) listed a series of questions that must be addressed by scientists and policy makers, always attaining to their potential variability and uncertainty. Some of the most important may be: *How well are emission factors estimated? What is the precision of such estimates? Do these estimates exhibit a systematic bias? Upon which criteria are these factors estimated? What are the key sources of such uncertainties? How are uncertainty levels reduced?*, among many others.

The definition of this variability is not straightforward. Frey (2007) defines uncertainty as the lack of knowledge whether a given emission value is indeed a true value. In the short term emissions are more variable than long term values, as well as they are uncertain. The following reasons are deemed of being substantial sources of variability in such levels:

- *Design*. Emitting sources do not have a generic design, rather a very particular one. Yet methodologies tend to override design issues that ultimately reflect themselves in differences in the emission factors.
- *Feedstock*. Certain types of emissions depend on the type and chemical composition of the feedstock that their processes incorporate. The most evident example is that of sulphur dioxide, whose emissions are a direct consequence of the sulphur content in coal and heavy fuel oils.
- *Background concentrations*. Environmental conditions such as humidity, pressure and temperature are deemed of influencing the emissions of some pollutants. For instance, volatile organic compounds tend to evaporate when temperatures increase.
- *Operational practises*. Some of the emitting sources exhibit differences between their operational practise levels which can be oriented to maximise efficiencies or production outputs, regardless of the emission rates. More intense operational practices lead to higher frequencies in maintenance levels.
- *Maintenance levels*. Not every emission source undergoes the same maintenance programmes. According to internal policies, these maintenance programmes might be more or less strict and would thus have a certain influence in the emission patterns.
- *Temporal patterns*. In some emission sources, specific temporal cycles and rates are usually evident. Emission characteristics tend to be transient rather than steady - state type.

On the other hand, a series of causes for uncertainty among emissions have been thoroughly observed and described (Frey, 2007):

- *Random sampling errors*. Usually of a statistical nature, they can be quantified on the grounds of confidence intervals and through investigating the standard error of the mean.
- *Measurement errors*. Every measurement process has associated a certain amount of error, related with the sampling and analysis processes. Such errors can be characterised as systematic and random, showing systematic errors a bias that impedes convergence. Random errors on the contrary, lead to imprecise measurements.
- *Non - representative data*. Assessing whether the compiled data are representative of a real situation or not, for a certain geographic area and a period of time, is a very complicated task. An additional difficulty arises when a dataset is effectively representative of an emission situation, yet it is not suitable for other circumstances, geographic areas or time periods. Using non - representative data leads to biases in the estimates.
- *Averaging time*. While some pollutants are measured on an hourly or daily basis, others are quantified by punctual stack tests which are characteristic of a few days. If the inventories are compiled under other temporal bases, errors associated with interpolation or extrapolation might become evident. One particular case of inconsistencies between averaging times might be the consideration of the driving cycle as a temporal base, given that driving cycles are highly variable regardless of speed averages being similar.

- *Omissions*. An omission is defined as the lack of substantial data in an emission inventory. A further statistical treatment occurs when some data are missing due to measurements losses or failures. Omissions related to under detections or non-detections by the measuring instruments could lead to missing or incorrect values, as well as the lack of speciation profiling for pollutants such as hydrocarbons or particles. According to the European legislation, there should be no need to gap - fill the reported inventory. Basically, three types of problems might be present that would eventually lead to a gap - filling; (i) when submissions from the whole national inventory are not provided for the immediate recent year or past years, (ii) when the emissions of some pollutants are missing for one or several time series and (ii) when sectorial emissions are not present and just national totals are provided (Directive 2008/50/EC).

- *Surrogate data*. A surrogate datum is defined as a situation in which reliable data are not available so a sensible judgement is made that leads to an analogy in which data are available. If the assumptions carried out to find the analogies are inappropriate, a sizeable amount of error might be introduced to the generated datasets.

Moroever, the amount of uncertainty also depends somehow on the aggregation level (Rypdal and Winiwarter, 2001). This uncertainty level can be widely avoided if the aggregation level increases; however, as scale is increased the analysis becomes less interesting (Rypdal, 2002). According to Winiwarter et al., (2009) the latter facts emphasise the need of assessing emission inventories constantly in terms of their quality to be reputed as reliable and accurate. A quality control system for an emission inventory is a series of technical activities that intend to measure and quantify as well as to assure the quality of the data that are being integrated into such an inventory (Hiraishi et al., 1996). Quality control procedures refer mainly to the comparison of independent and different datasets, in four basic modalities:

- Comparing emission data from past period observations.
- Comparing emission data from other regions that share meteorological and geographic similarities.
- Incorporating alternative emission data for the same domain and period of time.
- Contrasting inventories with on - site observations.

It is highly recommended to accomplish such **quality control** procedures using measured data, if possible. A well - sampled **observation** data set is fully independent and is therefore reliable for contrast issues. Two reasonable options to relate an emission inventory with a set of observation data are (i) the implementation of atmospheric models and further comparison between simulated results and (ii) the combination of ambient measurements and source apportionment techniques to estimate fluxes and inventory comparisons. Moreover, datasets need to be analysed attending to their particular temporal and spatial scales. Some observation sites are likely to be influenced by local scale emissions, highlighting the need of choosing an appropriate representation scale for the specific inventory or analysis. Furthermore, the comparison of measurements and inventories needs that inventories to be complete and to incorporate each and every one of the possible sources (Winiwarter et al., 2010).

Within the quality control procedures it is highly recommended, often as a good practice, that key source categories are identified and well characterised under a systematic and objective approach. A quantitative analysis focusing on these key categories and their changes in time might eventually conduct to significant improvements in the overall inventory system (Hiraishi et al., 1996). Key categories are defined by the European legislation as those responsible for 80% of the total national emissions (EMEP - CORINAIR, 2007). Additionally, it is important to consider categories that have undergone any technological or methodological changes.

Numerical **air quality models** have been built up to simulate many quantities that are observable in the atmospheric and chemical processes. Such variables have been described in terms of their behaviour in the atmosphere as a system, related either directly or indirectly to many actual physical processes. It is therefore reasonable to assume that if a model is simulating the atmospheric conditions of a given domain accurately, the inter - correlation among the simulated variables and the observations would be identical (Gilliam et al., 2006). Dispersion models and emission projection scenarios seem to be the unique reliable tools that are available to assess compliance of future legal emission limits. Air quality models may also be a valuable information link between emission inventories and measurements at air quality monitoring stations (Bellasio et al., 2007).

If any inconsistencies happen to occur, activity to undercover the sources of such discrepancies might take place so that better data are further delivered. Such discrepancies between measured and calculated pollutant concentrations might suggest that emission inventories could be improved. Yet, it is still unclear up to what extent this improvement must be carried out also on air quality models (Pulles, 1999).

Up to now, there is not a single model that is able to describe all the spatial and temporal scales in order to address air pollution as a whole. A series of computational requirements and restrictions have limited the resolution of different models, especially those that describe atmospheric processes on a global scale. Local and regional models, on the contrary, account poorly for global scale processes, usually those that occur outside the modelled domain but which have a direct influence on the studied region. This is particularly evident with long - range transport of air pollutants, such as intercontinental transport that affects background concentrations (Isaksen et al., 2009).

Global scale processes are usually of interest for describing long - term changes that occur from several decades to centuries, modelled over domains of several thousands of kilometres. On the other hand, urban scale models cover very local domains in which pollutants mix and interact on an almost immediate time scale. Modelling activities usually consider a hierarchy of different scales that are nested into each other. Such approach has as its final aim the communication and sharing of information between scales.

It is a common approach that nesting occurs only on a one - way direction, usually from the larger scale to the smaller one, and from the coarse model to the finer - scale model. The small - scale model should use output of the large model scale as boundary conditions; however, the large - scale model is also able to incorporate information from the finer scales in the form of distributions (Isaksen et al., 2009). An additional difficulty arises from this approach and it is the fact that the information is not always consistent between scales. There are sound efforts that aim to coordinate research among groups and models focusing on different scales, yet, there is still a long way to go until scale interactions are fully understood and correctly represented.

Under this perspective, **harmonisation** means that emissions calculated at a determined reference year, in both inventory and model analysis, must be consistent. The robustness of the inventory is strictly related with the consistency, within an acceptable uncertainty margin, between the inventory modelled data and real observations (ASTM D6589, 2010). In the case this does not occur, it is necessary to identify the causes of such departures between observations and modelled data. This evaluation process may include tracing the original sources of the emission factors used to construct the inventory through testing the sample set for appropriateness, reproducibility, statistical variance, etc.; the assessment of the robustness of the survey and compilation techniques might give strong hints about systematic differences (Lim et al., 1999).

In summary, to carry out a comprehensive analysis it is urgent to consider the model - emission inventory - measurements system as a whole. To this issue, several authors (Borrego, 2003; 2008; Chang and Hanna, 2004) define the total uncertainty (of both, the emission inventories and air quality models) as a contribution of three individual uncertainty sources: (i) the model uncertainty related with its theoretical formulation, the incorporation of comprehensive and realistic descriptions of the atmospheric and chemical phenomena, the numerical solutions and the chosen domains and structures; (ii) the input data uncertainty, associated to their representativeness, their validity and their quality; and (iii) the random variability. This category encompasses several stochastic and undetermined processes that affect invariably the quality of the measurements or input data.

The implementation of more advanced and realistic models or algorithms seems reasonable for minimising the first contribution, while incorporating accurate measurements, well placed monitoring locations and detailed inventories is recommended for improving the quality of input data. Conversely, the stochastic processes associated with the random variability are unavoidable and therefore render air quality modelling imperfect, or at least, subject to discussion (Borrego et al., 2008).

2.2. Current state of knowledge

Since the early 90's, initiatives in Europe regarding harmonisation in atmospheric modelling and decision making have been conducted. The two main problematic aspects were the great number of regulatory models that existed (and still exist) within Europe, as well as the fact that some models were not scientifically up - to - date. Even at such an early stage, there was a firm call for carrying out systematic comparisons of model predictions versus existing data sets from measurements or experiments leading to the filling of the existing knowledge gaps (Olesen, 2001).

One of the first comparisons carried out between emission inventories was carried out by Seika et al., (1996). The analysis of the methodologies on which the construction of the inventories relies has been discussed by the authors for the particular cases of New York, Los Angeles, Berlin and London, for both, local and national inventories. The cities of Los Angeles and New York in the United States were considered for the American case. The cities of Berlin and London, in Germany and the *UK* respectively, for the European case. No resolution has been chosen whatsoever, since no simulation was effectively performed over the selected cities. The studied pollutants were NO_x, VOC, CO and PM_{10} for each city and experimental measures have been carried out at a number of monitoring stations. Through comparing the compilation methodologies, the authors hunted for the potential discrepancy sources while suggesting measures for their consistence. The analysed data were consistently contrasted through their re - classification according to the *CORINAIR SNAP* nomenclature and evidence was found that inventories not only reflect true differences in emissions, but also distinctions which are related to the followed methodology for their construction.

With the rise of interest in the climate change phenomena, the development of strategies for better compilation of greenhouse gases emissions inventories were discussed in parallel. Jannsen et al., (1999) accomplished a comparison between the emissions obtained through an emission inventory and the predicted data from six atmospheric and two regional models for greenhouse effect gases. The chosen inventory database was *EDGAR* (Emission Database for Global Atmosphere Research). This database compiles emissions for several sectors organised on a $1^\circ \times 1^\circ$ grid resolution. The reported models were not actually used by the authors, though the quality of the yielded data was analysed. Four three - dimensional and one bi - dimensional Eulerian model were used for the global scale, as well as one three - dimensional Lagrangian model. Two three - dimensional Eulerian (*EUROS*) and one three - dimensional

Lagrangian (*COMET*) model were chosen for the regional scale. The datasets were compiled using both, bottom - up and top - down approaches; the chosen domain for the global scale was the world, while the north western region of Europe was used for the regional scale. It was demonstrated that the comparison between the inventory and the global simulations showed higher discrepancies, while the regional simulations yielded better data correlations. A similar discussion was extensively developed by Lim et al., (1999), in which a very first approach to emission inconsistencies is discussed.

In 2002, two important works were published in Germany. Mannschreck et al., (2002) carried out an extensive study of the emissions of the German city of Augsburg, with an approximate population of 255.000 inhabitants. The main goal of this study was to evaluate actual measures by comparing them with the values reported by emission inventories. The default level implied the use of mesoscale modelled data, while the superior level inventory used a high resolution road traffic model. The latter inventory also comprised a more detailed VOC profiling, according to their nature and emission source. For this specific work, VOC emissions were modelled on a national basis, whose starting points were the monthly consumption and production of solvent - containing products. The solvent content of these products was then spatially distributed on the population as to estimate an emission factor on a temporal base. The obtained data were validated with the observed emissions registered by two measuring stations located in the city.

Kühlwein et al., (2002) published a study which was a complimentary part of that developed by Mannschreck et al., (2002). This paper intended to evaluate the use of two different emission inventories with a different refinement degree by comparing modelled and measured data. In order to validate the estimated ratios, a Gaussian dispersion model and a chemical tracer through the chosen domain were implemented. The chosen domain comprised again, the German city of Augsburg. A series of mesoscale industrial inventories for the Bavarian region have been used, as well as mass balances for both, domestic and industrial emissions. For road traffic emissions, information on the road networks and traffic counts were used. For the emission of VOC, estimated consumptions of VOC - containing products and its further distribution on the population were applied. The uncertainty levels have been determined as statistic errors. For road traffic, deviations related with driving performances are expected, along with the uncertainties associated with emission values derived from dynamometer tests. A good refinement level in the inventory was obtained given the fact that the structure of the surrounding areas was considered, especially for road traffic emissions. It was therefore recommendable to use urban - scale emission data and detailed VOC profiles. The Gaussian dispersion model yielded reasonable results if compared with the transport of a chemical tracer, given the conditions of the city of Augsburg.

In the same year, Tsilingiridis et al., (2002) published a study whose main goal was to present a new emissions inventory with considerable detail for the Great Thessaloniki Area. Two different scales were used. For a local scale, an area of 5120 km^2 which corresponds to the Great Thessaloniki Area has been chosen, with a spatial resolution of 2×2 km^2. For the urban scale, a 3840 km^2 domain was evaluated using a 1×1 km^2 grid resolution. The authors strongly suggested the importance of having sufficient information regarding the small and medium sized point sources, such as dry cleaning facilities or gas stations, whose activities had a direct impact on the temporal and spatial distribution of the emissions. Except for the domestic, commercial and agricultural activities as well as industrial solvent use, every other activity has been modelled following a bottom - up approach. The emission factors have been taken from the *EMEP - CORINAIR* handbook. The used road traffic models were *COPERT II* and *ForeMove*. Since the obtained results supported the inventory data on actual measurements and simulations, a series of six emission abatement scenarios were chosen, simulated and analysed. Conclusions about the feasibility of these improvements were drawn.

The following year, Dommen et al., (2003) presented a study in which through the simulation of emission data obtained from two different inventories, the Italian and Swiss versions, several air quality parameters were calculated for the Lombardy region in northern Italy. A further validation of the estimates was done by comparing them with observed data provided by measuring stations. The chosen region comprised the Italian province of Lombardy, west Piedmont and the Swiss canton of Ticino. The chosen spatial resolution was $3\times3\ km^2$ with a 47×54 cell space discretisation. The road traffic emissions were simulated for three size - characteristic sites in the province; Milan for big urban traffic, Lecco for middle - size urban traffic and Valmadrera for small urban entourages. The SO_2, NO_x, CO and VOC emissions were followed through both, the Italian and Swiss inventory methodologies, under a bottom - up approach. The emissions of stationary sources were determined through local industrial inventories, while the biogenic emissions were estimated using a regional forest census. The Swiss inventory methodology is based in land use and disaggregated in a $1\times1\ km^2$ grid, to be regrouped into a $3\times3\ km^2$ grid resolution. The Eulerian model *LOOPS*, as well as the road traffic model *COPERT II* were used to process the inventories. In general, Dommen et al., (2003) discussed that there is sufficient error and uncertainty associated with the use of the same emission factors estimated through different methodologies. A clear relationship between the CO emissions and VOC with road traffic has been shown. Most of the stations that report high CO/NOx emissions are located in high - traffic zones and with propensity to vehicle congestion. Sizeable differences have been detected between the measured values for the region and the estimations obtained through the Swiss inventory.

A study of the air pollutants at the Maurienne Valley in France was completed using two different emission databases by Brulfert et al., (2005). The European *CORINAIR* and the Swiss *BUWAL - OFEPF* inventories were studied, both through simulations and comparison with the observed data. The chosen domain, the Maurienne Valley (130 km) is located in the French Alpine region that borders Italy. This zone is primarily affected by highway road traffic and by industrial activities related with metallurgy. A mesoscale approach was chosen and the emissions of SO_2, NO_x, CO, CH_4 and $NMVOC$ were followed. The atmospheric chemistry was modelled with *TAPOM*, whose boundary conditions were estimated with *CHIMERE*. The chosen simulation period comprised the dates between June 24^{th} to June 30^{th} and the dimensions of the domain were $74\times62\ km^2$.

Considerable deviations between inventories have been shown for several of the *SNAP* classes, whilst the uncertainty percentages inherent to the respective methodologies were unknown. Little differences were observed between inventories, due to the fact that the emission classes that compute for the grossest part of the total emissions were calculated using the same start data. Differences between inventories would have been evident in classes such as heating, but since the study was run during summer, the effect of these was slight (Brulfert et al., 2005).

Further attempts for an emission inventory analysis were carried out in South America by Zárate el al. (2007). Two versions of an emission inventory were generated for the city of Bogotá, Colombia. The first version was built with *CORINAIR* emission factors, while the second version was constructed with in - situ measurements and inverse modelling techniques. The chosen domain was the entire Cundinamarca province centred in the city of Bogotá, Colombia. A complete description of the physical and meteorological features of the city was made as well. The temporal resolution was one hour, within a 212×212 km grid with a 4 km resolution. The simulated date was March 6^{th} and 7^{th}, 2002 and the used models were *TAPOM* and *FVM* for this concrete study. The emission data were given by *DAMA* (the Colombian Environmental Monitoring Entity) in four categories: (i) production and services, (ii) air traffic, (iii) biogenic and (iv) on - road traffic. Emission estimates were made for NO_x, CO, $NMVOC$, CO_2, SO_2, CH_4 and PM_{10}. Although the *DAMA* organism has a specific classification for most of the point sources, the authors assigned classes to all of these according to their location and their weight in the total emissions.

Significant differences were observed between both inventories, basically due to the fact that the emission factors suggested by *CORINAIR* apply well to most of the European cases, but not to cities in developing countries. Interesting references to cities such as São Paulo and Mexico City were continuously made.

Mensink et al., (2009) studied scale interactions in a geophysical context, especially when cities exert a considerable impact on the regional air quality. Three scale levels were analysed and discussed. Local scale air quality was evaluated using a street canyon model. Then, changes in the air quality as a consequence of the distribution of open spaces at an urban scale were assessed. Finally, the dispersion of pollutants at a regional scale was observed for the Belgian province of Flanders. For the local scale analysis, the streets of the Ghentbrugge neighbourhood, located in Ghent (Belgium) were chosen. The urban scale was modelled using two feasible scenarios in the German region of the Ruhr, namely urban sprawl and satellite scenarios. The obtained simulations were then compared with the actual reference case. The Belgian region of Flanders was chosen as domain for the simulation of particle concentrations at a regional scale. The local scale analysis was carried out using a Gaussian dispersion model (*IFDM*) coupled with a street canyon model (*OSPM*). The necessary traffic data were obtained from a road traffic simulation model (*Paramics*). For the urban scale, the *AURORA* model was used, being fed with the appropriate geographical and meteorological data. The regional scale was simulated using the Eulerian air quality model (*EUROS*) for impact simulations. The results obtained for the local scale evaluation showed that around 40% of the measured $PM_{2,5}$ concentration came from the surrounding streets. At an urban scale, it has been shown that an urban sprawl scenario showed a marked change in comparison with the reference case. While the mean ozone exposure for both scenarios showed little change, adverse effects were evident when developing cities on a compact - polycentric scheme. According to the simulation carried out at a regional scale, around a 70% of the particles came from outside Flanders. Moreover, it was shown that the chemical composition of aerosols depended strongly on the season.

The study presented by Moussiopoulos et al., (2009) evaluated the impact of a series of reduction scenarios on the pollution of the Greek city of Thessaloniki. A set of measures from the Air Pollution Monitoring Network of Central Macedonia available at *AirBase* for NO_x, O_3, $NMVOC$ and PM_{10} was used. The authors clearly classified the monitoring stations whose data are to be used according to their entourage into urban, suburban and rural stations. The inventory was compiled for 2002 and the methodology used for this purpose was the one specified by *CORINAIR*. The inventory considered each one of the *SNAP* categories. A 1512 km^2 domain that encompassed the Great Thessaloniki Area, discretised into a 42×36 km grid with a 2×2 km^2 resolution was chosen and furtherly described through the use of the *OFIS* model. This work analysed the results attending to the air quality parameters that are recommended by the European Legislation. It strongly suggested the use of street canyon models for the internal urban traffic zones.

In general, Moussiopoulos et al., (2009) observed that the model values were in consonance with the actual measures. Further analysis lied on the fact of neglecting contributions from African dust transport, sea salt aerosols, etc. Particle emission factors were deemed uncertain due to its immature development stage. The authors quantified the uncertainty in the model estimations according to the European legislation, being 50% for PM_{10} and 30% for NO_2.

On a larger scale, the work of Alonso et al., (2010) was presented next year. The aim of this work was to build a regional urban vehicle emissions inventory for South America, taking into account the available inventories for major cities and a further atmospheric chemistry model. This inventory was then evaluated for two Brazilian cities through a simulation model and a series of observed measures. Three domains were evaluated in this study. An extensive area of South America comprised between the latitudes 14 $^\circ S$ and 32 $^\circ S$ and longitudes 37 $^\circ W$ and 55 $^\circ W$ with an 80 km grid was selected for the con-

tinental scale. Then, a region covering mainly the Southeastern and Central - Western regions of Brazil modelled under a 20 *km* grid was used for the regional scale. Finally, the cities of Minas Gerais and São Paulo were modelled for the evaluation of the local scale with a 5 *km* grid resolution. Several inventories were used as input data, at their respective scale but only focusing on road traffic emissions rather than in the whole set of anthropogenic emissions. For the regional scale inventory, the global databases *RETRO* and *EDGAR* were used. Basically for the local scale, the inventories of the Brazilian cities of São Paulo, Rio de Janeiro and Porto Alegre, were used; the inventories from the cities of Buenos Aires, Santiago and Bogota were also considered for the analysis. The construction of the regional mobile source inventory relies primarily on the extrapolation of local information to other Brazilian cities without inventories through a correlation with socio - economic indicators. Every inventory incorporated into this study was constructed through a bottom - up approach. One of the most important conclusions that were drawn from it was the fact that CO and NO_x fluxes correlate best with vehicle density. The elaboration of this new regional inventory affected notably the simulated CO and NO_x ratios and has yielded likewise, a better model performance according to the observations. The inclusion of this inventory in the global databases impacted greatly on the concentrations estimated formerly.

One of the most commendable efforts to harmonise atmospheric modelling is the Forum for Air Quality Modelling in Europe (*FAIRMODE*), under the coordination of the Joint Research Centre (*JRC*) of the European Commission. This forum was created in response to a series of requirements from the European Environment Agency (*EEA*), whose main scope was the execution of scientific research that would eventually lead to establish improved, reliable and validated tools upon which reasonable decisions could be based. Any modelling activity must necessarily comply with the Air Quality Directive requirements and must always provide causeways for joint interaction between member states.

The main objective of this forum is to establish a common infrastructure for reporting and storing air quality information, available to both, scientists and authorities. There is also a big interest in validating models and assuring the quality of modelled data as to identify limitations and detect error sources. To this effect, several intercomparison exercises at European or national level have been carried out. Under this framework is that the *EuroDelta* and *CityDelta* exercises happened to be, and where extensive research is being conducted as for now. Under the Clean Air for Europe (*CAFE*) programme scope, it is of priority to consider the urban scale as the domain of main interest, on which clearly human exposure occurs. Likewise, the *CityDelta* project is focused on a number of European cities that have been chosen to be representative of different air pollution patterns (Vautard et al., 2009).

The *CityDelta* project is an intercomparison study carried out to evaluate and assess the impact of emission reductions through the extensive usage of air quality models. The main question it intends to address is the extent to which regional model predictions differ from predictions obtained with finer resolution models (Cuvelier et al., 2007). A special interest is put into cities, in which most of the European population resides. Model simulations have been compared against observations as to understand model strengths and weaknesses; moreover, possible weak points in the emission inventories are identified. This work comprises studies carried out with six different models around four European cities: Berlin, Milan, Katowice, Paris and Prague. Two different resolutions were evaluated, a large - scale resolution (50 *km*) and a small - scale resolution (5 *km*) (Vautard et al., 2007).

Despite the important efforts dedicated to reduce the emission of air pollutants in Europe during the last decades, slight improvements have been experienced in many sectors; most of these sectors are indeed important such as industry or road transport (Vautard, et al., 2007). In the light of the above, the need for an urgent exchange of information between model developers, environmental managers, policy makers, inventory compilers and government institutions rises as necessary (De Leeuw et al., 1995).

2.3. Further developments and challenges

There is a general agreement that emission estimates are a crucial stage in the air quality modelling process. Yet, after nearly four decades the development of consistent and adequate emission estimates for its further application in air quality modelling is still a difficult chore. Moreover, there is a feeling that emission inventories are still subject of profound improvements in almost every stage of its compilation process (Wilkinson et al., 2005).

Intense activity has been carried out for combining the two main compilation approaches, top - down and bottom - up. Up to now, semi - bottom - up approaches, which are a combination of both methodologies but with emphasis on the latter, have been already tested in some works (Thao - Pham et al., 2008) . Alcorn and Lloyd (2009) agree that hybrid approaches might result in better - defined boundaries and higher levels of resolution and detail in data sets. They might also reveal more intuitively any data gaps. Yet, data conciliation is ultimately complex and time consuming.

There has been much discussion in past literature whether inventories should be compiled under a production - based scope rather than using a consumption - based frame; further developments might suggest that a correct approach would be considering an intermediate position between these two extremes (Peters, 2008). In the same line, a Swiss experience on compiling an emission inventory for ammonia (NH_3) hinted that conducting on - site surveys for stationary sources leads to results that are independent from the personal view of individual experts and thus, reflect more realistic conditions for such emissions. The development of regionally differentiated information, product of such surveys, allows better understanding levels of the abatement potentials for specific geographic regions (Reidy et al., 2008).

There is also an important call to improve the representation of local meteorological conditions as well as their quality, leading at the very end to an increase in the robustness of the model responses to emission controls (Thunis et al., 2007). To this issue, a study conducted by Vautard et al., (2007) showed that any significant improvements are limited when models do not generally use small - scale meteorology. Scale interaction, which is partly addressed by the present study, is also an issue that still needs to be fully described. Fine scale models generally encompass important urban scale effects, which are poorly represented by regional scale models (Thunis et al., 2007).

Since 2003, the United States Environmental Protection Agency (*USEPA*) has been developing a common framework to improve the quality of emission inventories. Such framework intends to prevent errors and bottlenecks during emission modelling through the development of a software infrastructure that would make the modelled emissions consistent across a series of projects. This issue highlights the importance of sharing emissions among institutions and parties, as well as the enhancement of transparency on such data. Further afield, the need for a reference document that includes best practice approaches for emission modelling is being addressed too. This includes also, the statistical methods that help identify differences between inventories and their compilation processes. Up to now, no method is able to fully describe all the differences and agreements that may show up between different inventories. A combination of methods is to be tailored according to the respective needs in any case (Winiwarter et al., 2003).

2.4. Legal background

2.4.1. International

In the international sphere, the Geneva Convention on Long - range Transboundary Air Pollution of 1979 (*CLRTAP*) is considered an essential framework for the control and reduction of pollution on a supra - national basis. This convention was signed by 34 governments and the European Union as the first international legally - binding instrument that dealt with air pollution under a broad regional scope. It came into force in 1983 and has been further extended by a number of protocols regarding specific pollutants; Helsinki (1985) and Oslo (1994) for sulphur; Sofia (1988) for nitrogen oxides; Geneva (1994) for volatile organic compounds; Århus (1998) for persistent organic pollutants and heavy metals (UNECE, 1979).

2.4.2. European Union

Airborne pollution has been addressed in the European Union through a series of laws and legal instruments. The Directive 1996/62/CE of the European Council of September 27^{th}, 1996, on ambient air quality assessment and management is the frame Directive from which many other directives and laws have originated. The general objective consecrated by this Directive was, among others, to define a common strategy in Europe to evaluate the ambient air quality of each of the member states through the use of common estimation methods and criteria (Directive 1996/62/EC).

To this effect, a series of daughter directives have been derived attending to different pollutants and the respective issues that arise from their emissions. The Directive 2008/50/EC of the European Parliament and of the Council of May 21^{st}, 2008 on ambient air quality and cleaner air for Europe is the legal background of the Clean Air for Europe (*CAFE*) program. This Directive emphasises the need of protecting human health and the environment as a whole, to mitigate emissions at the source, as well as identifying and implementing the most effective reduction strategies at local, national and community level (Directive 2008/50/EC).

2.4.3. Spain

The Royal Decree 1073/2002 of October 18^{th} is the transposition of Directive 1996/62/EC to the Spanish legal system, which along with Directive 1999/30/EC of the Council of April 22^{nd}, 1999 regulates the limit emission values for airborne sulphur dioxide, nitrogen dioxide, particles and lead (modified by Decision 2001/744/EC of the Comission).

Under Law 34/2007 on air quality and nature protection, the General State Administration is responsible for (i) defining any atmospheric polluting activity; (ii) compiling and updating periodically the national emission inventories; (iii) carrying out the evaluation, following and compilations of the technical information about pollution for complying with the obligations derived from international protocols and agreements; and (iv) integrating the information into the Spanish information system for pollution vigilance and control which would be further transmitted to the Autonomous Communities. These Communities will adopt control measurements and inspection procedures to guarantee the compliance with this Law and will eventually exert any sanctions as their ultimate prerogative (Royal Decree 1073/2002).

2.5. Scope and objectives

The concept of using air quality models whose final objective has been modifying or refining the input information for compiling emission inventories has been already reported (Vivanco and Andrade, 2006). These studies relied, however, on a series very general assumptions and simple compilation procedures while lacking a contrastable methodology.

As it has been already stated, evaluating an emission inventory in terms of its approximation to reality or its methodological goodness is a very difficult task. Up to now, the **scale issue** has been recognised as intrinsic to emission inventorying yet scientific activity intended to reconcile inventories has been rather shallow. It is thus that the present study appears as necessary and adequate, not only because it addresses the **scale issue** under an integral perspective, but also because it identifies the baseline causes of the inconsistencies between different - scale inventories.

For the specific purpose of this work, the **scale issue** and its associated problems will be tackled through the following sequence of activities:

- A detailed analysis of the general emission paterns across the Greater Madrid Region (*GMR*) for an array of selected pollutants.
- The build - up and run of an air quality model (*AQM*) with the same parameterisations and experimental setup, except for the emission inventories, to obtain a series of simulated ambient air concentrations produced by both databases.
- The statistical comparison of the simulated concentrations and the actual observations measured by a given monitoring network across the selected domain for the studied pollutants.
- The evaluation of the contribution of the emissions from neighbouring cells to those cells in which monitoring locations are emplaced, as to establish a spatial boundary for analysis.
- A detailed analysis of the emissions that occur at cells where stations are located, both in terms of the relative and absolute emissions for each pollutant.

Furthermore, this study pretends to establish a reasonable and easy - to - approach methodology that would eventually undercover mistakes and incorrect assumptions carried out during the inventory collection process and thus extrapolate findings to further compilation tasks. The ultimate objective of this work is to identify up to what extent do nested emission inventories interact while describing a certain geographic domain, which would eventually hint on strategies for reducing inconsistencies between them and producing **multi - scale inventories**.

3. METHODOLOGY

3.1. Air Quality Modelling System

The air quality modelling system (*AQM*) used is based on the Community Multiscale Air Quality (*CMAQ*) model (Byun and Ching, 1999; Byun and Schere, 2006). Emissions were processed by the Sparse Matrix Operator Kernel Emissions (*SMOKE*) modelling system (Institute for the Environment, 2009) as described in Borge et al., (2008). The meteorological fields needed to simulate air pollution processes have been generated with the Weather Research and Forecasting (*WRF*) modelling system (Skamarock and Klemp, 2008). . A schematic representation of the *AQM* is shown in figure 3.1.

The Community Multiscale Air Quality (*CMAQ*) model is a multi - pollutant, multiscale **air quality model** that incorporates a series of state - of - science techniques for the simulation of both, atmospheric and land processes of atmospheric pollutants and their precursors. It is also able to describe the processes that are involved in its transport, transformation and deposition, on both regional and urban scales. *CMAQ* is one of the most reliable tools for assessing air quality through the simulation of ambient air concentrations and deposition processes (wet or dry) of most pollutants. Its algorithms can handle all the major issues concerning photochemical oxidants, particulate matter, and acidic and nutrient deposition under a holistic approach.

The simulation system is also able to address tropospheric ozone, acid deposition, visibility and fine particles under a *one atmosphere* perspective, where the utmost complex interactions between regional and urban scales are confronted. It is thus quite versatile too for representing several spatial and temporal scales. *CMAQ* has been formulated to address regulatory assessments as well as scientific studies conducted by both, government and research institutions. *CMAQ* is also a valuable tool for simulating concentrations over a wide range of meteorological conditions and a variety of geographical areas (Byun and Ching, 1999).

CMAQ includes a variety of interface processors that deal with a certain number of input data for quantifying the emission and meteorological systems. It also incorporates a series of processors that calculate photolysis rates and that develop the initial and boundary conditions. One of the most interesting features it includes is an Advection and Diffusion module that addresses horizontal diffusion through an eddy diffusion approach. Through this module, the formation of aerosols and reactions between pollutants in aqueous phase is also described. A plume - in - grid module, which is also incorporated, includes a number of algorithms that deal with subgrid scale physical and chemical processes that have an impact in plumes released at given point sources. Also within the modelling system, a Particle Modelling and Visibility routine is included which is in fact one of the major advancements of *CMAQ*; it intends to address issues concerning fine and coarse particles, such as those related to sulphate, nitrate, ammonium, organics and aerosol water. This module might also be able to describe the deposition of semi - volatile organic compounds (*SVOC*) through the use of parametrisations (Byun and Ching, 1999; Eder and Yu, 2006).

Emissions have been modelled and processed with the Sparse Matrix Operator Kernel Emissions (*SMOKE*) Modelling System. It allows data processing through a high performance computing sparse matrix algorithms. This tool is quite useful in decision making regarding controls for regional and urban applications. It also includes a mechanism for preparing inputs for air quality research, having a prognostic function as well. *SMOKE* is able to deal with most of gaseous pollutants such as carbon monoxide (CO), nitrogen oxides (NO_x), volatile organic compounds (VOC), ammonia (NH_3), sulphur dioxide (SO_2), particulate matter (either $PM_{2,5}$ or PM_{10}) and a large number of toxic pollutants such as heavy metals, benzene or formaldehyde. An emissions processor such as *SMOKE* has as baseline function the transformation of resolution of emission inventory data to the input resolution of an air quality model. It transforms thus, the emission inventory through temporal allocation, chemical speciation and spatial allocation for the specific *AQM* requirements (Institute for the Environment, 2009).

The meteorological fields have been generated through the Weather Research and Forecasting (*WRF*) Model, also known as Advanced Research *WRF* (*ARW*). This model includes the latest developments for modelling under a fully comprehensible nonhydrostatic approach (Skamarock and Klemp, 2008). The *ARW* modelling system is a flexible, state - of - science simulation system that is suitable for a huge range of applications and to describe mostly any scale size and specifically tested and evaluated for air quality modelling applications in the Iberian Peninsula (Borge et al., 2008, Skamarock et al., 2008).

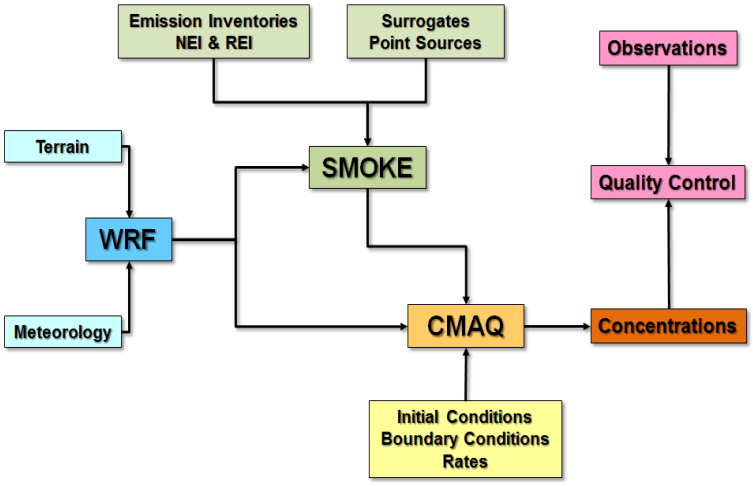

Fig. 3.1: Flux diagram of the air quality modelling system

3.2. Studied Domain

3.2.1. General Description

Greater Madrid Region

The Greater Madrid Region (*GMR*) is located in the centre of Spain; it is home to over 6 million people, of which approximately 4 million inhabitants are concentrated in the Madrid metropolitan area. This region concentrates around 14% of the total population of Spain over an 8.020 km^2 area, divided into 179 administrative municipalities (AECM, 2005; INE, 2009). According to the Statistical Atlas of

the Autonomous Community of Madrid, 51% of the regions land has an agricultural use, 11% urban use, and 3% industrial, the rest being forests and natural zones (AECM, 2005), .

The Greater Madrid Region (*GMR*) has basically a continentalised Mediterranean climate in which precipitations are scarce and irregular (between 300 y 800 *mm* yearly), with a strong thermal amplitude that shows cold winters (with temperatures as low as 6°C) and hot summers (temperatures above 22°C). It is located at an interior region away from the sea along a north - east south - west barrier, the Central System, which modifies the typical traits of the Mediterranean climate. This fact reduces precipitations and widens the thermal amplitude (AECM, 2005). The atmospheric dynamics is characterised by seasonal displacements of the Azorean anticyclone that if situated over the Iberian Peninsula, results in a dry season (summer). When this phenomenon moves southwards, it allows the entrance of western and south - western winds that provoke abundant precipitations during autumn and spring. During winter, these dynamics generate a less powerful thermal anticyclone (AECM, 2005).

The Greater Madrid Region is the most important economic hub in central Spain. During the last decade, economic activity in the Autonomous Community of Madrid (*CAM*) has grown above the Spanish mean growth rate, which is again, higher than the European mean. The Greater Madrid Region contributed to the National Spanish Economy with a 17,4% of the *GMP* in 2002. Most of the economic activity of Madrid is of industrial and service nature. Around 58% of the commercial activity in the region is concentrated in the Madrid municipality, which also encompasses 42% of the commercial facilities and 45% of the labour posts. The industrial activity in the *GMR* accounted for a 12% of the total Spanish business balance, rising up to 61.518 million € during 2007 (AECM, 2005; INE, 2009). Agricultural activities are not so important, whatsoever; productive agricultural land in the region barely surpasses 450 thousand hectares, roughly a 1% of the national total.

On the other hand, the Greater Madrid Region is an important communications and transport crossroads in the Iberian Peninsula. It had as to 2007, approximately 3.390 *km* of roads and highways and around 4.5 million vehicles, around 11% of the Spanish total. It is also one of the most important railroad traffic points in Spain, as well as a busy air transportation hub (INE, 2009).

3.2.2. Model Setup

Modelled Domain

The *GMR* was modelled as a grid of 48 $\times$ 48 km^2, with 2304 grid cells of a 4 *km* size each. This grid was centred aproximately between the 5,0 $^{\circ}W$ - 2,5$^{\circ}W$ longitudes and the 39,5 $^{\circ}N$ - 41,5$^{\circ}N$ latitudes. The studied region included the entire Autonomous Community of Madrid (*CAM*) as well as some portions of the provinces of Avila, Segovia, Valladolid, Guadalajara, Cuenca and Toledo. The geographic representation of the gridded domain is shown in figure 3.2.

The vertical structure for each of the models consists of 30 layers, out of which 9 are included within the first kilometre of height, the main objective being to describe as accurately as possible the phenomena that occur at the planetary boundary layer. The before mentioned geographic domain was studied from January 1^{st}, 2007 to December 31^{st}, 2007, with an hourly resolution for the 365 days of the year. This temporal span covered an 8.760 - hour period of time.

Model Configuration

In every case the model configuration was not changed as to attribute the observed differences *only* to the emission inventories. The meteorology was simulated with the *WRF* model whose inputs

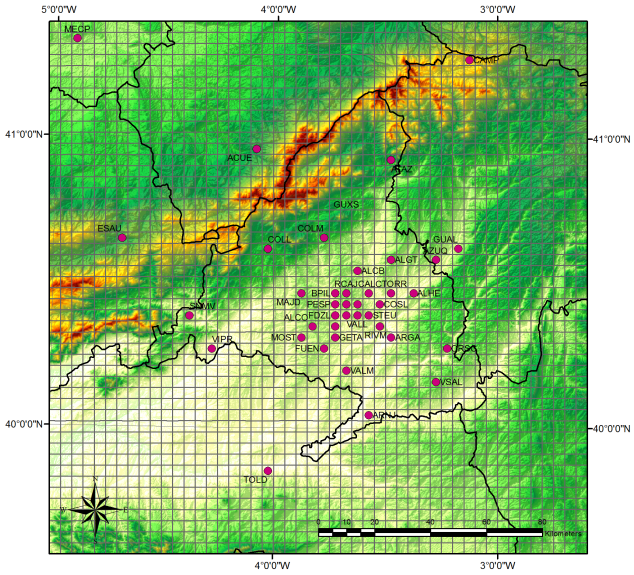

Fig. 3.2: Gridded domain over the Greater Madrid Region

(terrain elevation, landuse and land - water masks, soil humidity and temperatures) from the United States Geological Survey (*USGS*) and the European Centre for Medium - Range Weather Forecasts (*ECMWF*). The dynamic options and parameterisations for the meteorology simulations are the following (Borge et al., 2008):

- *Boundary layer.* Yonsei University (*YU*) Planetary Boundary Layer scheme.
- *Microphysics. WRF* Single - Moment 6 - class (*WSM6*) microphysics scheme.
- *Surface model.* Noah Land - Surface model.
- *Radiation.* Longwave: Eta Geophysical Fluid Dynamics Laboratory (*GFDL*) longwave radiation scheme. Shortwave: *MM5* shortwave radiation scheme.

The emissions have been prepared entirely according to what is stated in (Borge et al., 2008) . Further discussion on the preparation of the emission inventories will be carried out in the following sections. The chemistry and transport model related with *CMAQ* were introduced through the Meteorology - Chemistry Interface Processor (*MECIP*). The basic configuration of the chemistry and transport model is the following:

- *Advection.* Yamartino global mas - conserving scheme.
- *Vertical diffusion.* Asymmeteric Convective Model version 2 (*ACM2*).
- *Chemical mechanism. CB*-05 gas - phase mechanism.
- *Numerical integrator.* Euler Backward Iterative (*EBI*) solver.
- *Aerosols.* 4^{th} generation model *CMAQ* aerosol mechanism with extensions for sea salt emissions and thermodynamics.

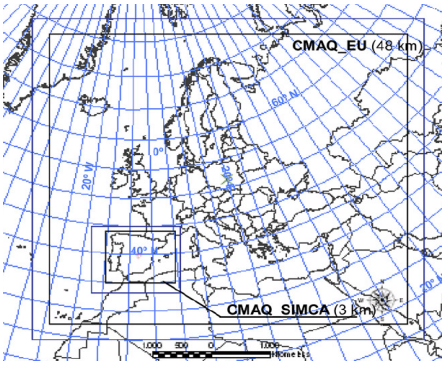

Fig. 3.3: Nested domains and projection for the Greater Madrid Region

3.2.3. Spatial analysis

In order to conduct a complete analysis of the entire domain, the punctual information obtained for the chosen *monitoring locations* was complemented with the spatial representation of the various datasets throughout the *GMR*. To this effect, a Geographic Information System (*GIS*) software (*ArcGIS* ®) was extensively used. The projection chosen for the spatial representation and the maps included in this work was the *Lambert conformal conic* projection (*LCC*) with parameters α =20°, β =60°, $\gamma = -3°$ (Annoni et al., 2003). The nested domains and the general projection can be seen in figure 3.3, and further described in (Borge et al., 2010).

3.3. Emission Inventories

3.3.1. National Emission Inventory

The National Emission Inventory is compiled every year by the Spanish Ministry for the Environment (Ministerio de Medio Ambiente, Medio Rural y Medio Marino - *MARM*) and was published in 2009 for the year of 2007. The National Emission Inventory follows a *SNAP* nomenclature and the general methodology specified by the *CORINAIR* framework (EMEP - CORINAIR, 2007). The maximum spatial resolution of the database is the fourth level (*NUTS 3*) of the *NUTS* territorial classification for statistical purposes, proposed by *EUROSTAT* which corresponds to the provincial level. The maximum temporal resolution is hourly. This inventory will be referred with the accronym *NAC* throughout the present work.

3.3.2. Regional Emission Inventory

The Regional Emission Inventory is compiled by the Environment, Housing and Territory Council of the Autonomous Community of Madrid (Consejería de Medio Ambiente, Vivienda y Ordenación del Territorio de la Comunidad de Madrid) and was published in 2009 for the year of 2007. This inventory is compiled on a yearly basis under the *SNAP* methodology and follows a series of recommendations from the European Environment Agency (*EEA*) and the International Panel for Climate Change (*IPCC*) (EMEP - CORINAIR, 2007). . The maximum temporal resolution of this database is hourly, yet also incorporating daily, monthy and yearly data. The spatial resolution upon which it has been constructed is based on a 1 $km \times 1$ km regular grid. This inventory will be referred with the accronym *CAM* throughout the present work.

3.4. Observational Datasets

The further comparison analysis was carried out over a network of 55 monitoring stations placed throughout the *GMR*. Of this observation network, 25 stations are located within the Madrid municipality, 24 more in the rest of the Autonomous Community of Madrid and 6 of them scattered in the adjacent provinces. According to their emplacement, stations are classified as *traffic*, *urban background*, *industrial* or *rural types*. Air quality concentration data for each of these stations were available on an hourly resolution for the studied pollutants. In table 3.1 the list of the 55 monitoring stations is presented with the measured pollutants at each precise location and their classification according to the Environment, Housing and Territory Council of the Autonomous Community of Madrid.[1] The network is also represented accordingly in figure 3.2.

It is important to note at this point that comparing simulated concentrations for a given cell and the actual observations measured by a monitoring station is not a straightforward procedure. Depending on the emplacement of the monitoring station, observations might be affected by phenomena that occur at a very local scale; this issue is specifically problematic for *traffic* stations, while *urban background* or *rural* stations seem to be representative of wider domains. Strictly speaking, comparisons should be carried out only with stations that prove to be representative enough of the cell on which they are emplaced. This fact in many cases, is not evident beforehand nor easy to be addressed. Further discussion on this issue is presented in section 4.7.

3.5. Selected Pollutants

Air quality models are able to simulate a wide array of substances across time and space, most of them also being measured by monitoring stations. However, the pollutants that were studied at the present work were chosen basically beacuse of their representativeness, their widespread nature and the health concerns they rise. The assessed pollutants were **nitrogen oxides** (NO_x), **particulate matter** (PM_{10}, $PM_{2,5}$) and **sulphur dioxide** (SO_2), since neither is sensitive to boundary conditions.

Nitrogen compounds play an important role in the formation of ground - level ozone and other photo - oxidants that are formed as a consequence of high concentrations, especially during summer smog periods. Such induced pollutants have adverse effects on vegetation and human health. Nitrogen compounds contribute also to the formation of secondary aerosols, one of the compounds present in fine particles (CFHA, 2005).

Particles come from very different sources, which are generally classified into three categories: anthropogenic sources of primary particles, anthropogenic sources of secondary particles and natural sources. In the anthropogenic sources of primary particles, mobile sources like road traffic - cars, trucks and vans - are widely found, although construction engines and machines are also important (CFHA, 2007).

Sulphur dioxide comes from both, natural and anthropogenic sources, being the latter by far the most important. Although acid rain has been traditionally the most studied issue concerning sulphur compounds emissions, it is now the formation of particulate matter that draws attention since it is an aerosol precursor that is likely to be transformed into sulphate aerosols (Vestreng et al., 2007).

[1] For further information on the monitoring networks of the Greater Madrid Region visit:
http://www.mambiente.munimadrid.es/opencms/opencms/calaire
http://gestiona.madrid.org/aireinternet/run/j/AvisosAccion.icm

Tab. 3.1: Stations of the Monitoring Network over the Greater Madrid Region

Station No.	Code	Station Name	Type	Pollutants
1	RECO	Recoletos	Traffic	NO_2, SO_2, PM_{10}
2	PCAR	Plaza del Carmen	Traffic	NO_2, SO_2, PM_{10}
3	PESP	Plaza de España	Traffic	NO_2, SO_2, PM_{10}
4	BPIL	Barrio del Pilar	Traffic	NO_2, SO_2, PM_{10}
5	PMAR	Plaza Dr. Marañón	Traffic	NO_2, SO_2, PM_{10}, $PM_{2,5}$
6	PSAL	Plaza Marqués de Salamanca	Traffic	NO_2, SO_2, PM_{10}
7	ESCA	Escuelas Aguirre	Traffic	NO_2, SO_2, PM_{10}
8	LUCT	Plaza Luca de Tena	Traffic	NO_2, SO_2, PM_{10}
9	4CAM	Cuatro Caminos	Traffic	NO_2, SO_2, PM_{10}
10	RCAJ	Ramón y Cajal	Traffic	NO_2, SO_2, PM_{10}
11	MBEC	Plaza Manuel Becerra	Traffic	NO_2, SO_2, PM_{10}
12	VALL	Puente de Vallecas	Traffic	NO_2, SO_2, PM_{10}
13	FDZL	Plaza Fernández Ladreda	Traffic	NO_2, SO_2, PM_{10}
14	PCST	Plaza de Castilla	Traffic	NO_2, SO_2, PM_{10}, $PM_{2,5}$
15	ASOR	Arturo Soria	Traffic	NO_2, SO_2, PM_{10}
16	GRIC	Gral. Ricardos - Farolillo	Traffic	NO_2, SO_2, PM_{10}
17	AEXT	Alto de Extremadura	Traffic	NO_2, SO_2, PM_{10}
18	MORZ	Moratalaz	Traffic	NO_2, SO_2, PM_{10}
19	ISPL	Isaac Peral	Traffic	NO_2, SO_2, PM_{10}
20	PONT	Paseo de Pontones	Traffic	NO_2, SO_2, PM_{10}
21	CALC	Final Calle Alcalá	Traffic	NO_2, SO_2, PM_{10}
22	CCAM	Casa de Campo	Urban Background	NO_2, SO_2, PM_{10}
23	STEU	Santa Eugenia	Traffic	NO_2, SO_2, PM_{10}
24	UEMB	Urbanización Embajada	Traffic	NO_2, SO_2, PM_{10}, $PM_{2,5}$
25	BARJ	Barajas Pueblo	Urban Background	NO_2, SO_2, PM_{10}
26	ALHE	Alcalá de Henares	Traffic	NO_2, SO_2, PM_{10}
27	ALCB	Alcobendas	Industrial	NO_2, SO_2, PM_{10}
28	ALCO	Alcorcón	Traffic	NO_2, SO_2, PM_{10}
29	ALGT	Algete	Urban Background	NO_2, SO_2, PM_{10}
30	ARNJ	Aranjuez	Urban Background	NO_2, PM_{10}
31	ARGA	Arganda del Rey	Industrial	NO_2, SO_2, PM_{10}
32	ATAZ	El Atazar	Rural	NO_2, SO_2, PM_{10}
33	COLM	Colmenar Viejo	Traffic	NO_2, PM_{10}
34	COLL	Collado Villalba	Traffic	NO_2, SO_2, PM_{10}, $PM_{2,5}$
35	COSL	Coslada	Traffic	NO_2, SO_2, PM_{10}
36	FUEN	Fuenlabrada	Industrial	NO_2, SO_2, PM_{10}
37	GETA	Getafe	Traffic	NO_2, SO_2, PM_{10}
38	GUXS	Guadalix de la Sierra	Rural	NO_2, SO_2, PM_{10}, $PM_{2,5}$
39	LEGA	Leganés	Traffic	NO_2, SO_2, PM_{10}
40	MAJD	Majadahonda	Urban Background	NO_2, SO_2, PM_{10}
41	MOST	Móstoles	Urban Background	NO_2, SO_2, PM_{10}
42	ORSC	Orusco de Tajuña	Rural	NO_2, SO_2, PM_{10}
43	RIVM	Rivas Vaciamadrid	Urban Background	NO_2, PM_{10}
44	SNMV	San Martín de Valdeiglesias	Rural	NO_2, PM_{10}, $PM_{2,5}$
45	TORR	Torrejón de Ardoz	Urban Background	NO_2, SO_2, PM_{10}
46	VALM	Valdemoro	Urban Background	NO_2, PM_{10}
47	VIPR	Villa del Prado	Rural	NO_2, SO_2, PM_{10}
48	VSAL	Villarejo de Salvanés	Rural	NO_2, PM_{10}
49	ACUE	Acueducto	Traffic	NO_2, SO_2, PM_{10}
50	AZUQ	Azuqueca de Henares	Urban Background	NO_2, SO_2, PM_{10}
51	CAMP	Campisábalos	Rural	NO_2, SO_2
52	ESAU	Estación de Autobuses	Traffic	NO_2, SO_2, PM_{10}
53	GUAL	Guadalajara	Urban Background	NO_2, SO_2, PM_{10}
54	MECP	Medina del Campo	Industrial	NO_2, SO_2, PM_{10}
55	TOLD	Toledo	Urban Background	NO_2, SO_2, PM_{10}

3.6. Statistic Analysis

After the air quality modelling (*AQM*) system has been run for both inventories, a sound comparison process between the output data from the simulations and the actual **measured** air pollutants concentrations was carried out. Given the fact that there are, for both simulations and measurements a set of 8760 data (which correspond to the total number of hours in a year) for the four pollutants, relying on **statistic indicators** seems more than reasonable.

The applied statistical indicators in this work seek to characterise quantitatively the performance of the model under different perspectives. According to Dufour (2006), the analysis would be carried out attending the following criteria:

- The ability to reproduce the temporal and geographical evolution of the prediction. Quantified through the *Pearson's correlation coefficient* (r).
- The characterisation of the average behaviour. Inspected through the *mean bias* (MB).
- The diagnosis of the general tendency for over or underprediction. Expressed globally by the *mean fractional bias* (MFB).

3.6.1. Pearson's correlation coefficient

$$r = \frac{\sum_{i=1}^{N} \left(M_i - \overline{M}\right)}{\left[\sqrt{\sum_{i=1}^{N} \left(M_i - \overline{M}\right)^2}\right] \cdot \left[\sqrt{\sum_{i=1}^{N} \left(O_i - \overline{O}\right)^2}\right]} \tag{3.1}$$

The *correlation coefficient* (r) is a widely used performance statistic that measures the degree to which two variables correlate linearly. Correlation coefficients of 1 indicate that a perfect linear relationship exists, while coefficients equalling zero, indicate that no such relationship between variables exists (USEPA, 2007). However, the use of the *correlation coefficient* is discouraged in some cases where outliers or extreme pairs are present, due to its sensitivity to such values (Wilmott, 1982).

3.6.2. Mean Bias

$$MB = \frac{1}{N} \cdot \sum_{i=1}^{N} \left(M_i - O_i\right) \tag{3.2}$$

The *mean bias* (MB) is a performance indicator that averages the difference between the observed and simulated concentrations over each pair in which the observed values are greater than zero. If the *mean bias* equals zero, this indicates that the model over predictions exactly cancel the model under predictions. The model bias has been defined in such a way that positive values indicate that the model prediction exceeds the observation, while negative values indicate likewise, underestimations (USEPA, 2007).

3.6.3. Mean Fractional Bias

$$MFB = \frac{2}{N} \cdot \sum_{i=1}^{N} \frac{M_i - O_i}{M_i + O_i} \tag{3.3}$$

Using a *mean fractional bias* (*MFB*) is recommended when the mean bias becomes large due to the misuse of a minimum threshold. For under and over - prediction, the fractional bias exhibits values of -67% and +67% respectively. This indicator is indeed reliable because it equally weights positive and negative bias estimates (USEPA, 2007). Its main disadvantage, however, is that the predicted concentration is found in both, the numerator and denominator. The *mean fractional bias* (*MFB*) can be especially useful for evaluating the performance of particle modelling, some of them with values close to zero and it ranges from -200% to +200% (Thunis et al., 2011).

3.7. Summary Diagrams

The use of summary diagrams is intended to have a better overview of the general behaviour of a simulation in the spirit of finding a compromise between the complexity of the evaluation and the need of providing simple and straightforward diagnosis tools for the user. In order to obtain these plots, the Joint Research Centre *DELTA* tool was used. This tool was conceived under the *FAIRMODE* [2] framework as a benchmarking tool for air quality models, while in this specific work it was applied to emission inventory benchmarking (Thunis et al., 2011). With this tool, two types of diagrams were drawn: *Taylor diagrams* and *target plots*.

3.7.1. Taylor diagrams

Taylor diagrams were first drawn by K.E. Taylor in 2001, as a mean of incorporating into a single plot a set of statistic indicators, summarising how well a series of model predictions matches actual observations. The statistic indicators that this plot incorporates are the *centred root - mean square difference*, their *correlation coefficient* and the amplitude of the variations in terms of the standard deviation. Patterns that agree well with observations will lie near the X - axis, exhibiting low *root - mean square errors* (Taylor 2001).

In summary, the *Taylor diagram* is able to characterise the relationship between an experimental field (mostly the data sets provided by a simulation or a model) and a reference field, based in actual observations. Interestingly, this diagram discards overall biases by subtraction of both data sets, thus characterising a centred error pattern. According to Thunis et al., (2011), data sets being located within an area limited by the curves r =0,4 and $r = 0{,}8$ and the curves $\sigma = 5$ and $\sigma = 10$ are representative of an acceptable model performance.

3.7.2. Target plots

The *target plot* is proposed as an alternative to the *Taylor diagram*, developed by Jolliff et al., (2009). It provides summarised information about the pattern statistics (*standard deviation* and *correlation coefficient*) incorporating also the *mean bias*. This diagram is a simple Cartesian coordinate system in which the unbiased *root mean square error* (*RMSE*) and the total *root mean square error* are the X and the Y axes respectively. The distance between the origin and the specific plotted point is equal to the total

[2] The *DELTA* tool is available from the JRC at: `http://aqm.jrc.it/DELTA/index.htm`

RMSE.

This diagram provides useful information about whether the standard deviation of the model predicted values is larger or smaller than the reference dataset simultaneously assessed on whether it has a positive or a negative bias. Moreover, normalising this diagram allows comparing performances among data sets that exhibit different units. However, it does not explicitly represent information about the correlation coefficient. It can be shown that values that appear on the diagram within the -1,0 $< X <$ 1,0 cannot be negatively correlated. Likewise, since the square of the normalised bias must always be positive, every point that shows an *RMSE* $<$ 1,0 is necessarily positively correlated too. Additionally, the outermost marker establishes that all points between it and the origin represent positively correlated datasets. If an r positive value is fixed between the origin and the outermost point, then the points between it and the origin are also greater than such fixed r value (Jolliff et al., 2009).

3.7.3. Selection criteria

In order to identify concrete issues of the inventories that should be improved, a *selection* process is carried out based on (i) the before mentioned statistic indicators and (ii) the results represented in the *Taylor diagrams* and *target plots*. The analysis will be focused on *monitoring locations* that either exhibit the *largest* or the *smallest departures* between emission inventories, *NAC* and *CAM*. The analysis of the locations that show the largest departures might reveal sensitive differences in compilation criteria between inventories; conversely, the analysis of locations where departures are almost nil could put into evidence valid considerations. As a general rule, stations correlated *better* but with slightly larger departures between inventories were preferred over stations with smaller departures but *lesser* correlations for analysis.

3.8. Emission analysis at cell - level

For the chosen stations, a detailed analysis of the emissions that occur at *cell - level* was completed, segregating such emissions according to the *SNAP* nomenclature (EMEP - CORINAIR, 2007). This evaluation was conducted in two main aspects for each cell, namely (i) **absolute emissions** for each pollutant and *SNAP* group, expressed in daily emitted metric tons ($ton \cdot day^{-1}$) (table 3.2); and (ii) the **relative emissions** of every *SNAP* group ($P_{SNAP_i,j}$) over the gross totals computed at a given cell for the studied pollutants, expressed as a percentage.

$$P_{SNAP_i,j} = \frac{100}{E_j} \cdot \sum_{k=1}^{n} E_{SNAP_i,j,k} \tag{3.4}$$

Where $E_{SNAP_i,j,k}$ are the emissions of pollutant j originated by activity k contained in $SNAP_i$ and E_j are the total produced emissions for pollutant j at a given cell, both in $ton \cdot day^{-1}$. Along with the cell - level analysis, additional information was frequently obtained through constant comparisons with the statistic indicators computed for every monitoring location.

An initial hypothesis would be that whenever the estimations obtained from the emissions inventory and the real emissions are similar, an air quality model will exhibit a better response while predicting pollutant concentrations. The spatial and temporal domains on which such emissions will have an influence on the ambient air quality at a given point are impossible to define beforehand. A feasible alternative

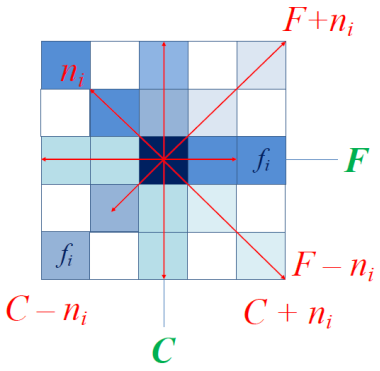

Fig. 3.4: Obtention of the influence zone for a given cell

would be an approach based on *wind frequencies* (N) and *intensities*. The main objective that was followed was to determine an *influence zone* of neighbouring cells around the cell where the monitoring station is found. This procedure studies an intermediate stage between considering *only* the emissions that occur at a cell level and *total* domain emissions, as in the study published by Vivanco and Andrade (2006).

This would produce an *expansion* effect that might contribute to the central emissions, weighed according to the *wind frequencies* and *speeds* (u) that occur at that precise location. In order to quantify the number of surrounding cells to be considered, one must have the *simulated* meteorological data for the studied location.The number of cells for every direction (n_i) and the weighing factors (f) are obtained according to the following equations, as a function of the *mean wind speed* ($\overline{u}$).

$$\overline{u} = \sum_{i=0}^{2\pi} u_i \cdot \frac{N_{i,j}}{N_i} \tag{3.5}$$

Tab. 3.2: Description of the *SNAP* categories

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

$$\boxed{n_i = \frac{\overline{u}}{d}} \tag{3.6}$$

$$\boxed{f_i = \frac{N_i}{n_i \cdot (N - c)}} \tag{3.7}$$

Where i is the *wind direction*, j is a given *wind speed* or *wind speed interval*, d is the cell size and c is the number of calms. Hence, the resulting *influence zone* can be determined from the coordinates of the cell where the monitoring location is being analysed (C, F) as suggested by figure 3.4. The general *SNAP* percentage ($P_{SNAP_i,j}$) for each neighbouring cell is obtained from equation 3.8. The overall *SNAP* percentage for the *expanded cell* ($P_{SNAP_i,exp}$) is calculated from equation 3.9.

$$P_{SNAP_i,j} = \frac{100 \cdot f_i}{E_j} \cdot \sum_{k=1}^{n} E_{SNAP_i,j,k} \tag{3.8}$$

$$\boxed{P_{SNAP_i,exp} = \sum_{P_{SNAP_i}=1}^{N} P_{SNAP_i,j}} \tag{3.9}$$

4. RESULTS AND DISCUSSION

4.1. Emission Analysis across the Greater Madrid Region

In the following section, an analysis in terms of the emissions that have been estimated for both inventories and for the selected pollutants. As it has been stated before, the analysis was carried out attending to the **absolute emissions** and the **relative emissions** that occur across the *GMR*.

4.1.1. Nitrogen oxides (NO_x) emissions across the Greater Madrid Region

Figure 4.1 shows the **absolute emissions** for NO_x within the *GMR*, for both inventories. Through inspection of the main *SNAP* categories that emit NO_x, it has been shown that *SNAP* categories *02* and *07* are responsible for most of the NO_x emissions within the *GMR*. The *CAM* shows very high emissions located throughout the cells along which the major highways lie, namely $A-2$, $A-3$ and $A-6$ and along the southeaster rim of the province of Madrid.

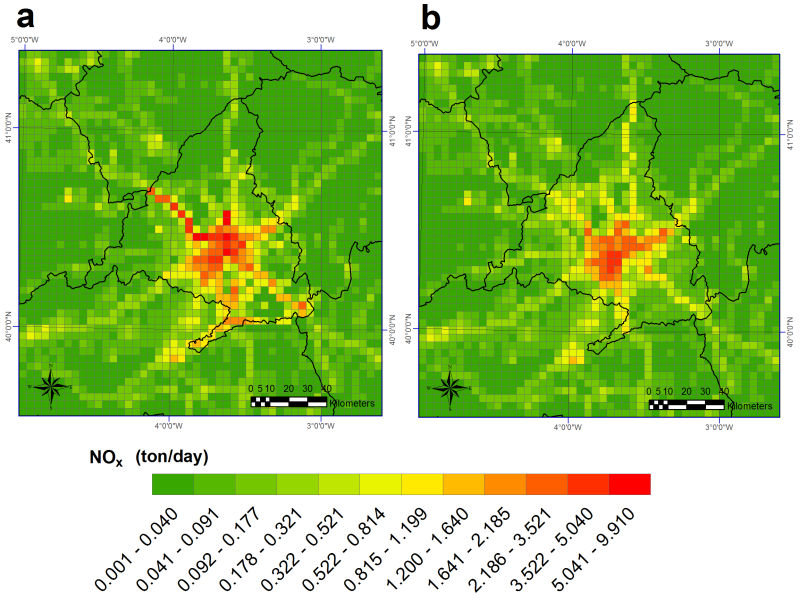

Fig. 4.1: Total NO_x emissions for the *GMR* according to a) *CAM* and b) *NAC* inventories

Figure 4.2 shows the *SNAP* percentage at cell - level for *SNAP02* and for both, *CAM* and *NAC* inventories. The *CAM* inventory clearly shows a marked contribution of domestic heating activities at areas that are reputed of having a less density of road networks, namely the southeastern part of the

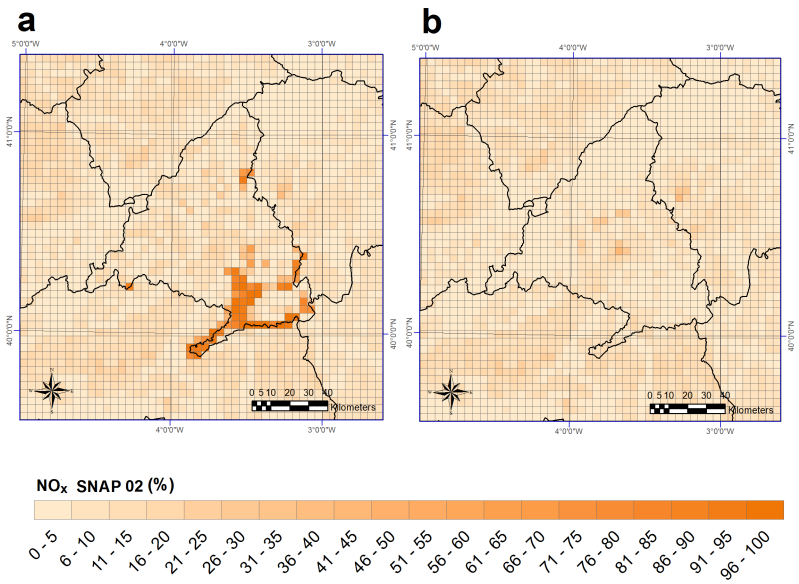

Fig. 4.2: NO_x SNAP02 percentage at cell level for a) *CAM* and b) *NAC* inventories

Madrid Autonomous community. This assumption seems reasonable since this area is of an emminently suburban nature (AECM, 2005). On the contrary, the *NAC* inventory considers quite a low contribution of *SNAP02* in the *GMR*. The difference between inventories is quite evident at the southern boundaries of the Autonomous Community of Madrid (*CAM*), where there is no continuity between the *SNAP02* percentages inside the *CAM* and the neighbouring province of Toledo.

Figure 4.3 shows the percentage at cell - level for *SNAP07*. Both inventories exhibit high percentages at cells that contain important regional roadways, as well as high emissions due to road - traffic at the urban nucleus of Madrid. Specifically, the *CAM* inventory assigned a very heavy contribution to highways and the city centre. The *NAC* inventory shows lower percentages for the city centre and cells where highways are located show moderate values. The lack of continuity between the *CAM* inventory and the *NAC* equivalent is also evident at the southeastern border, where the cells along the $A-3$ highway show higher percentages inside the *CAM* territory; once the boundary has been crossed, lower percentages are seen because the surrounding provices are described only by the *NAC* inventory. From figure 4.3 one can also see that the *NAC* inventory considers less *SNAP07* contributions for the city centre, which might hint that other activity groups should be present.

4.1.2. Particulate Matter (PM_{10}) emissions across the Greater Madrid Region

Figure 4.4 shows the **absolute emissions** for PM_{10} for the studied domain. It suggests again, that for the *CAM* inventory, road traffic at highways are overweighted, while exhibiting some emission hotspots along such roads. The *NAC* inventory reflects more emissions from off - road sources (cells not lying along major highways), showing also higher emissions for cells located at rural entourages. The *SNAP* activity groups that were evaluated as important **particulate matter** emission sources are *SNAP07*, *SNAP08* and *SNAP10* although other groups that may contribute are *SNAP02* and *03*.

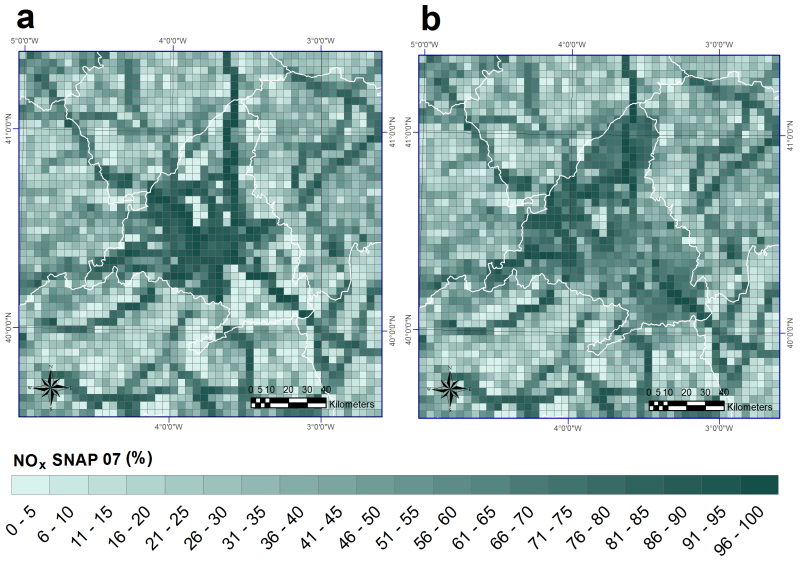

Fig. 4.3: NO_x SNAP07 percentage at cell level for a) *CAM* and b) *NAC* inventories

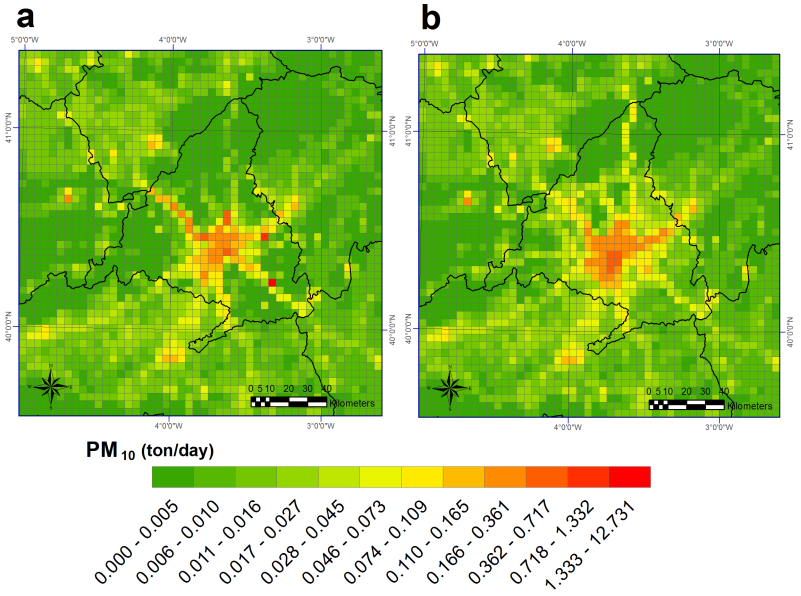

Fig. 4.4: Total PM_{10} emissions for the *GMR* according to a) *CAM* and b) *NAC* inventories

Figure 4.5 shows the *SNAP* percentage at cell - level for *SNAP07*. The *CAM* inventory shows high *SNAP07* percentages in the central area of the Autonomous Community of Madrid, where high traffic levels are usually registered. There is a noticeable variation in the cells along which highways are located once they cross the *CAM* boundaries, which of course reveals differences in compilation criteria. High contributions are concentrated in the southern area of the *CAM*, while just immediately in the neighbouring province of Toledo low contributions are observed except for the road cells. On the contrary, the *NAC* version considers a high contribution of *SNAP07* for areas that are emminently rural.

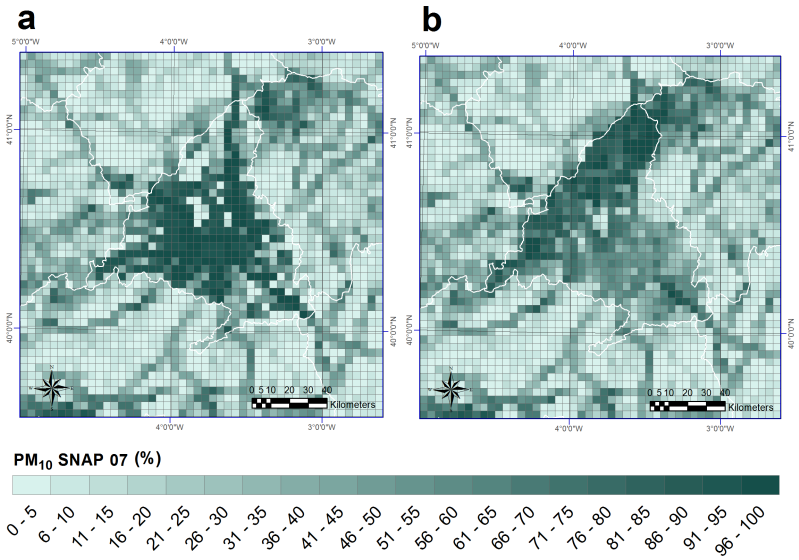

Fig. 4.5: PM_{10} SNAP07 percentage at cell level for a) *CAM* and b) *NAC* inventories

SNAP08 is also reputed for being a PM_{10} emitting activity group. Figure 4.6 shows the spatial representation of the *SNAP08* percentage at cell - level for the *GMR*. It is evident that there are noticeable differences between inventories. The *CAM* inventory considers high *SNAP08* contributions at specific rural entourages within the Autonomous Community of Madrid (*CAM*), where off - road transportation is very likely to be found.

This fact is also complemented by looking at the cells where some highways are located, namely $A-1$ in the North and $A-3$ in the south - east. Since PM_{10} emissions are closely related to activities that involve any type of combustion, it is evident that where highways lie *SNAP07* combustions would vastly predominate over *SNAP08*, which by definition must be *off - road*. This behaviour is neatly reproduced by the *CAM* inventory and not by its *NAC* counterpart.

Surprisingly, figure 4.6 considers *SNAP08* contributions within the territory of the Autonomous Community of Madrid (*CAM*) to be very low. This is clearly not reasonable since within the *CAM* there are indeed zones where road traffic does not contribute to particle emissions, basically rural emplacements already described in section 3.2.1 (AECM, 2005; ALMUDENA, 2011). Moreover, this fact does not give continuity to the emission pattern observed at the surrounding provinces, rendering the *CAM* as a diffuse center almost exempt from every *SNAP08* particle emission.

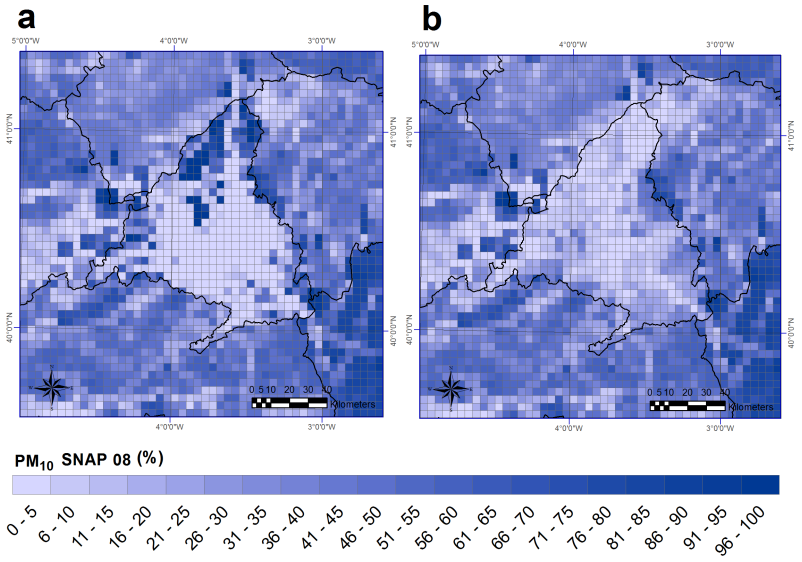

Fig. 4.6: PM_{10} SNAP08 percentage at cell level for a) CAM and b) NAC inventories

SNAP10 is an emitting activity group that would need to be analysed specifically for particles. The contribution of such a group at cell - level is depicted in figure 4.7 for the *GMR*. According to this figure the *CAM* inventory considers that, except for some cells, the contribution of *SNAP10* to the total PM_{10} emissions is very low. However, the fact that just a few cells have very high contribution percentages might suggest that the inventory did consider PM_{10} particles occuring at those specific locations. On the other hand, the *NAC* inventory considers a higher level of *SNAP10* contribution for locations outside the urban centres. Figure 4.7 shows that the *NAC* inventory is able to reproduce the silhouettes of some main highways, such as $A-2$ and $A-3$ where, certainly, PM_{10} emissions do not come from *SNAP10*.

4.1.3. Particulate Matter ($PM_{2,5}$) emissions across the Greater Madrid Region

Figure 4.8 is the spatial representation of the **absolute emissions** for $PM_{2,5}$ across the Greater Madrid Region (*GMR*). Although $PM_{2,5}$ is being emitted by several *SNAP* activities, special emphasis should be made on *SNAP02* and *SNAP08*. Even for this pollutant, strong contributions are put into evidence at most roads for the *CAM* inventory. The *NAC* inventory showed higher emissions for cells located at the urban centre of Madrid and at smaller cities throughout the adjacent provinces.

Since $PM_{2,5}$ are related to combustion processes, it is interesting to analyse the *SNAP02* category group for this pollutant. Figure 4.9 shows how *SNAP02* contributions are distributed along the studied domain, which are somewhat similar to those presented for NO_x in section 4.5.1. The *CAM* inventory shows a high contribution at the southeastern rim of the Autonomous Community of Madrid (*CAM*), which is of course reasonable since this area is not heavily influenced by on - road emissions. Almost at no other cell within the territory of the *CAM* does *SNAP02* contribute so greatly as in this area.

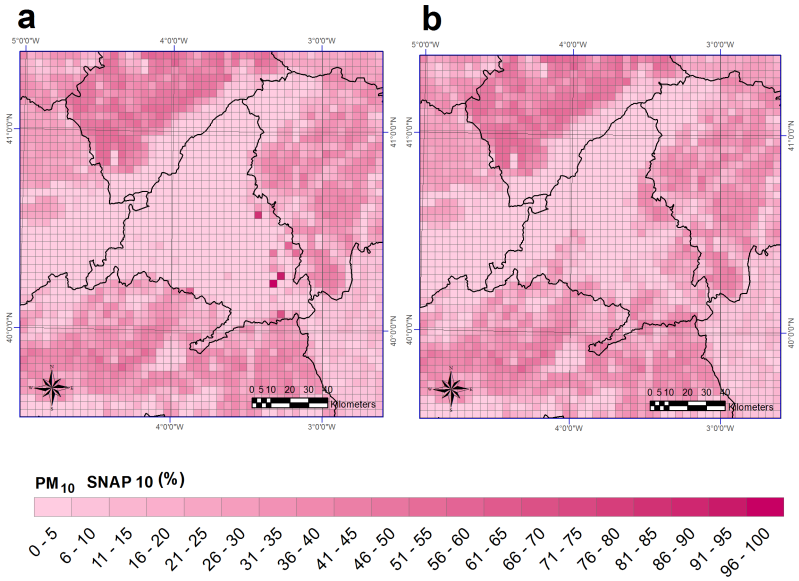

Fig. 4.7: PM_{10} SNAP10 percentage at cell level for a) *CAM* and b) *NAC* inventories

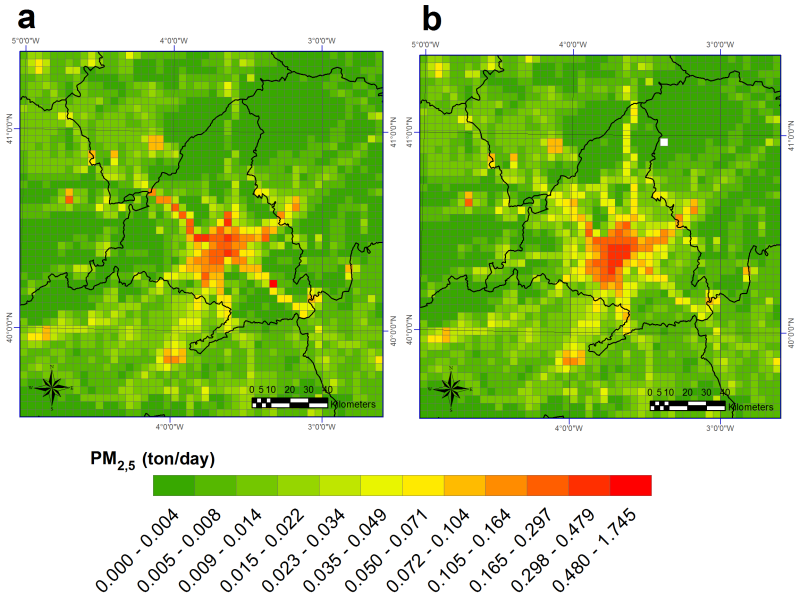

Fig. 4.8: Total $PM_{2,5}$ emissions for the *GMR* according to a) *CAM* and b) *NAC* inventories

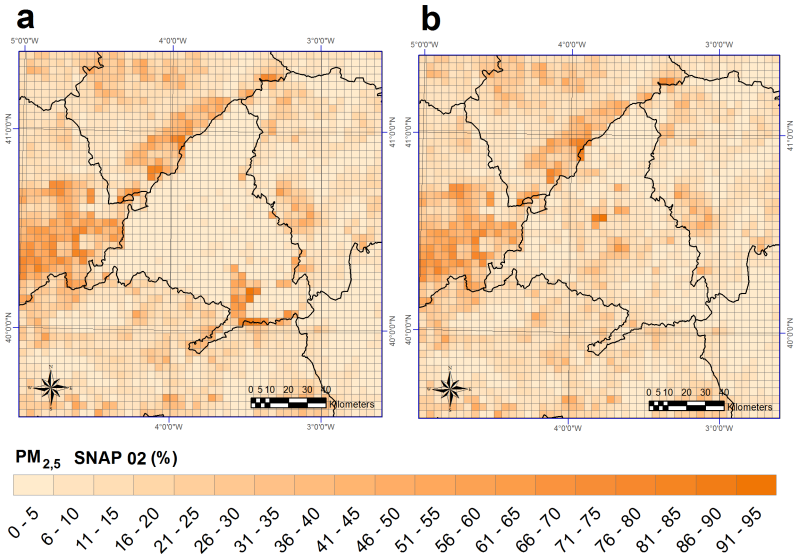

Fig. 4.9: $PM_{2,5}$ SNAP02 percentage at cell level for a) *CAM* and b) *NAC* inventories

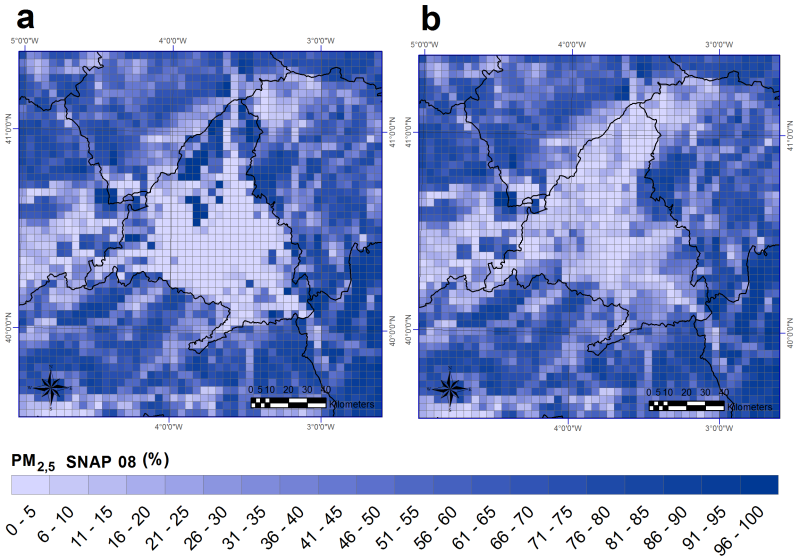

Fig. 4.10: $PM_{2,5}$ SNAP08 percentage at cell level for a) *CAM* and b) *NAC* inventories

On the contrary, the *NAC* inventory does not assign sizeable concentration spots of *SNAP02* influenced cells, which might suggest that this inventory lacks resolution to actually determine which zones are poorly influenced by traffic emissions. Another *SNAP* category that was analysed was *SNAP08*, especifically for the case of $PM_{2,5}$. Figure 4.10 represents the spatial distribution of the *SNAP08* percentage at cell level for the entire *GMR*. From this figure, it is evident that there are enough differences between inventories, as it is particullarly striking the fact that according to the *CAM* inventory almost no *SNAP08* is present within the territory of the Autonomous Community of Madrid.

On the other hand, this inventory is able to reflect spatially the cells along where the major highways lie, which the *NAC* inventory is unable to describe. The latter fact reflects a poor resolution on behalf of the *NAC* inventory, since cells where roads lie would be emminently influenced by *SNAP07* contributions; moreover, at rural regions the surrounding cells would be heavily influenced by *SNAP08* providing the effect of road representation neatly on the map.

4.1.4. *Sulphur dioxide (SO_2) emissions across the Greater Madrid Region*

Finally, figure 4.11 shows the sulphur dioxide **absolute emissions**. The *CAM* inventory shows great emissions on highways and roads, which is a direct consequence of the use of high - sulphur emission factors for *SNAP07* activities. The *NAC* inventory shows high SO_2 emissions at cells located in the very central part of the province of Madrid, more specifically at the urban nucleus.

The activity groups that contribute the most to SO_2 emissions are *SNAP02*, *SNAP03* and *SNAP07*. For *SNAP02*, the *CAM* inventory exhibits an analogous behaviour than the rest of the pollutants for *SNAP02* activities (sections 4.5.1, 4.5.7).

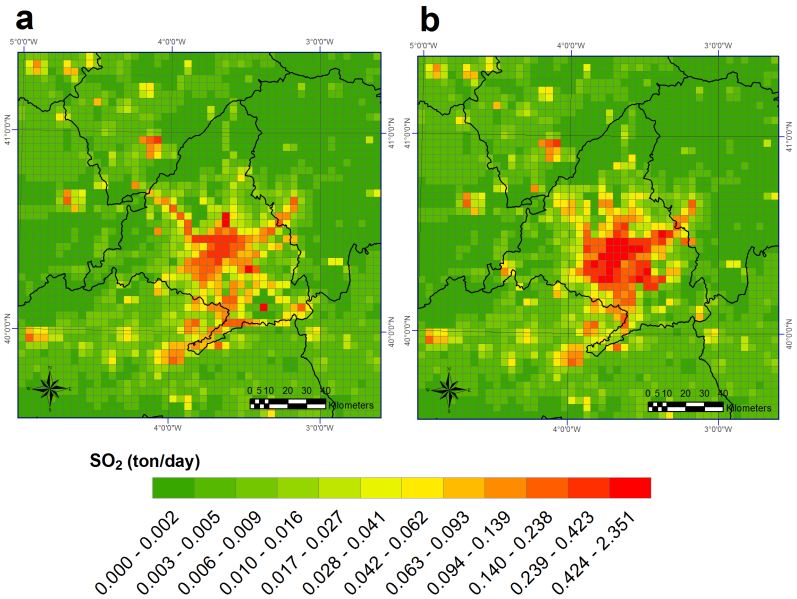

Fig. 4.11: Total SO_2 emissions for the *GMR* according to a) *CAM* and b) *NAC* inventories

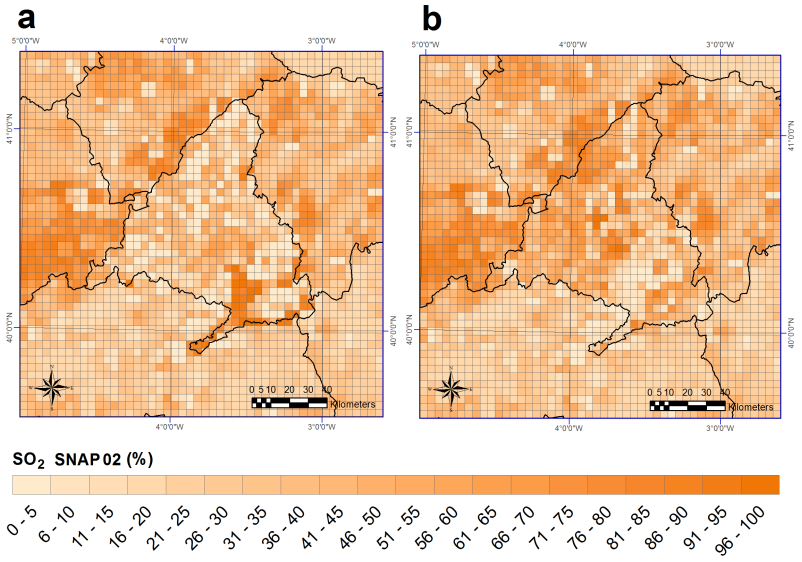

Fig. 4.12: SO_2 SNAP02 percentage at cell level for a) *CAM* and b) *NAC* inventories

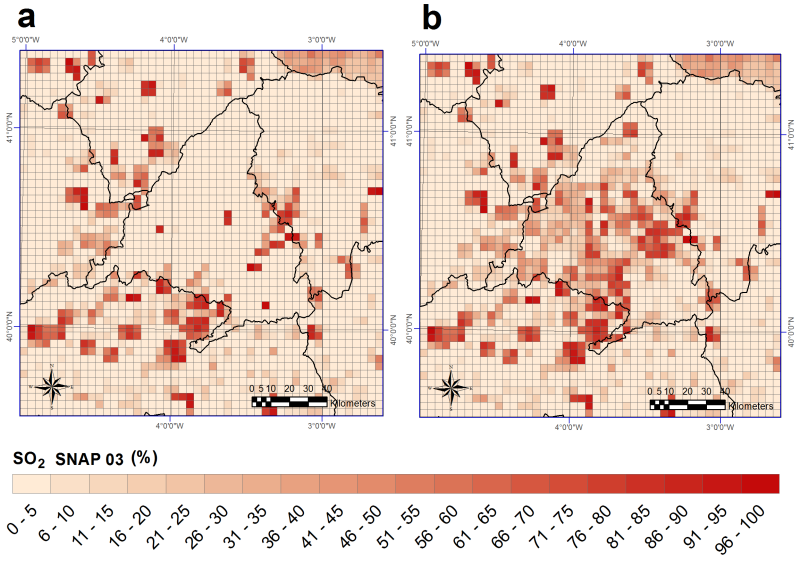

Fig. 4.13: SO_2 SNAP03 percentage at cell level for a) *CAM* and b) *NAC* inventories

This inventory exhibits great contributions of groups other than *SNAP07* at the southeastern corner of the Autonomous Community of Madrid, while contributions are poor at locations heavily influenced by on - road emissions. Conversely, the *NAC* inventory considers that *SNAP02* is responsible for sizeable SO_2 emissions throughout the *GMR*. Both facts are clearly presented in figure Figure 4.12.

Figure 4.13 shows the spatial representation of the *SNAP03* percentage at cell level for SO_2. Differences between both inventories are dramatic. While the *CAM* inventory identifies *SNAP03* contribution at very localised points, the *NAC* inventory considers a heavy contribution of *SNAP03* on SO_2 emissions over the entire Autonomous Community of Madrid (*CAM*). This very first assessment might not be completely right, since the *CAM* is not reputed for sheltering extensive industrial zones (AECM, 2005).

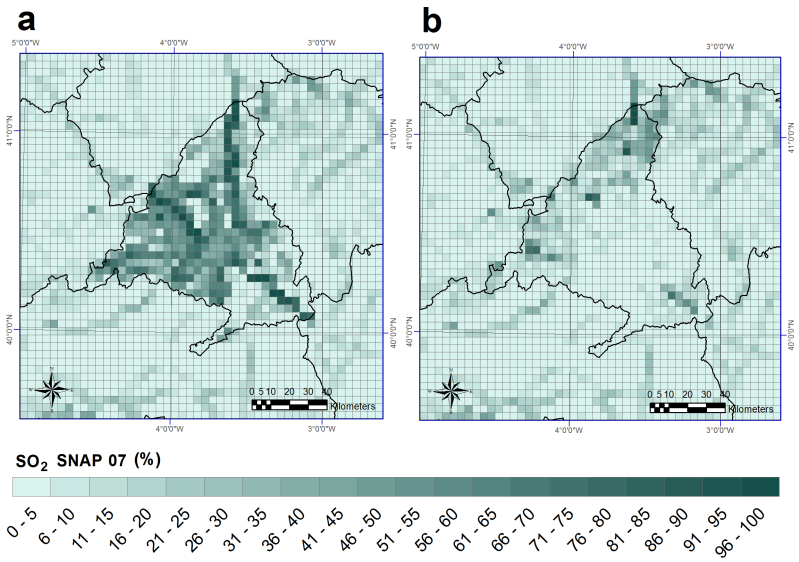

Fig. 4.14: SO_2 SNAP07 percentage at cell level for a) *CAM* and b) *NAC* inventories

Finally, the road - traffic contributions (*SNAP07*) are presented in figure 4.14. Very interesting differences can be easily spotted for each inventory; the *CAM* shows very high contributions of *SNAP07* throughout the territory of the Autonomous Community of Madrid (*CAM*). The road silhouettes are clearly presented and no continuity in the contribution patterns is seen at the neighbouring provinces whatsoever. However, the *NAC* inventory considers very low contributions for the territory of the *CAM*, and slightly higher for the northern rural regions. This behaviour clearly underlies different inventory compilation criteria for both versions.

4.2. Simulation Results

The simulation of the *GMR* domain was completed after a 17 - day period of time (420 hours), for an hourly resolution of the year 2007 and for **each** of the emission inventories, *NAC* and *CAM*.

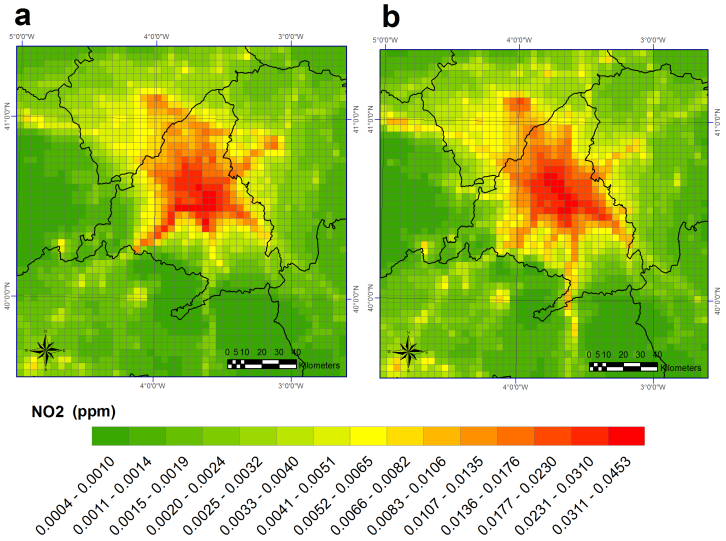

Fig. 4.15: Annual mean nitrogen dioxide (NO_2) concentration for a) *CAM* and b) *NAC* inventories

4.2.1. Simulation results for nitrogen dioxide - NO_2

As it is evident from figure 4.15, both emission inventories do exhibit some differences for this specific pollutant. The most noticeable one is the predominance of high concentration spots in the central *GMR* which corresponds to the urban nucleus of Madrid for the *NAC* than for the regional inventory. Since there are indeed differences for nitrogen dioxide, the sources of these differences were investigated.

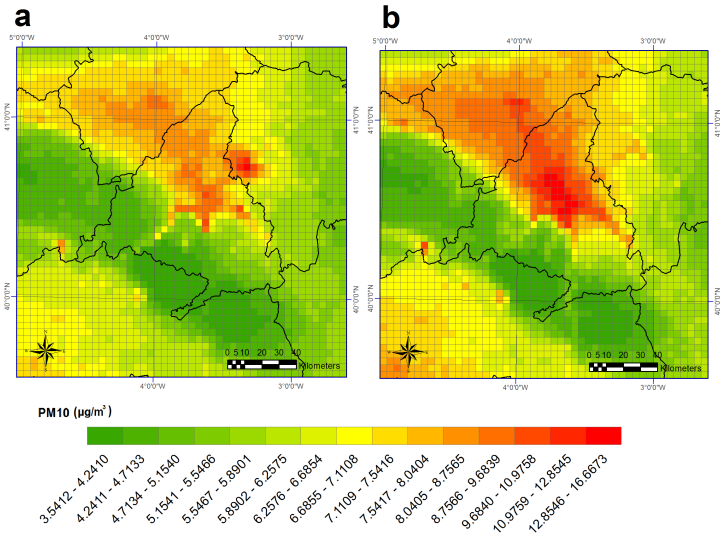

Fig. 4.16: Annual mean particulate matter (PM_{10}) concentration for a) *CAM* and b) *NAC* inventories

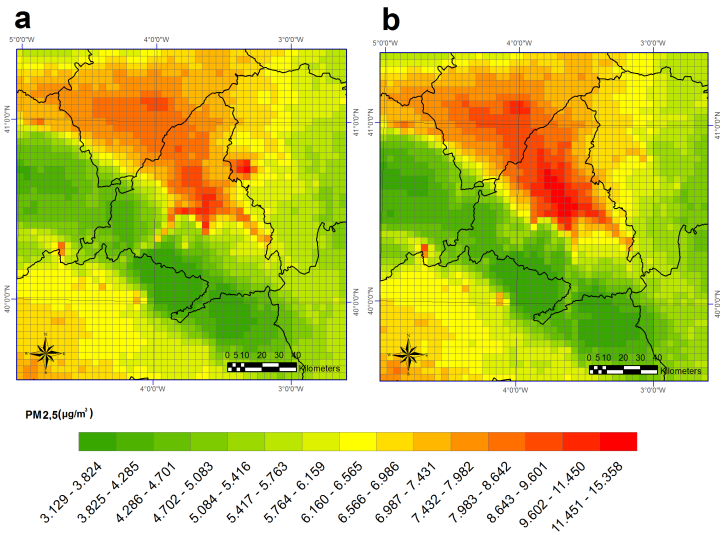

Fig. 4.17: Annual mean particulate matter ($PM_{2,5}$) concentration for a) *CAM* and b) *NAC* inventories

4.2.2. *Simulation results for particulate matter - PM_{10}, $PM_{2,5}$*

For the case of particulate matter (PM_{10}), there are also differences between inventories according to figure 4.16. Although the concentration *pattern* is very similar, there are higher concentrations in the central and north-western zones of the *GMR* for the *NAC* inventory, while the *CAM* inventory shows a concentration hotspot in the immediate north-eastern limit of the Madrid province with Guadalajara. As

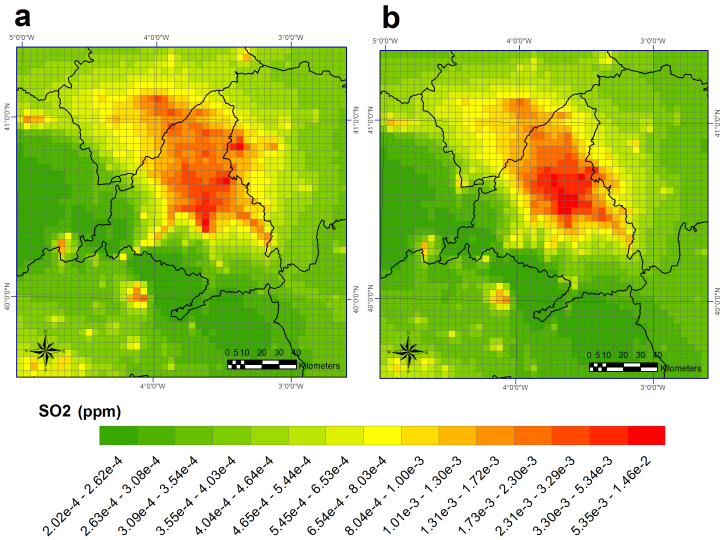

Fig. 4.18: Annual mean particulate matter (SO_2) concentration for a) *CAM* and b) *NAC* inventories

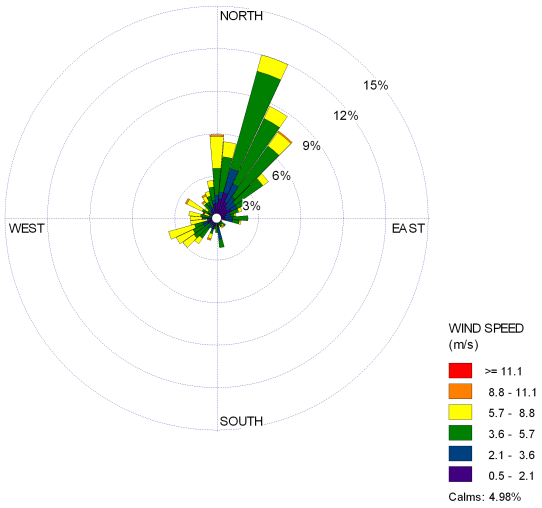

Fig. 4.19: Wind rose for simulated meteorological fields at cell $(29, 24)$

with nitrogen dioxide, the observed differences between inventories for PM_{10} motivated the evaluation of the underlying differences. Basically, an analogous behaviour to that of PM_{10} is observed for $PM_{2,5}$ too. A similar concentration *pattern* is observed at the central and north-western zones of the simulated domain for both inventories, being slightly higher for the *NAC* inventory than for the *CAM* equivalent. The study of the causes of the differences between inventories shown in figure 4.17 for $PM_{2,5}$ was also undertaken.

4.2.3. Simulation results for sulphur dioxide - SO_2

The differences shown in figure 4.18 suggested that further analysis on the different inventory compilation criteria for sulphur dioxide should be made. Although the concentration *patterns* are more or less analogous for both inventories, the regional *CAM* inventory accounts for some concentration hotspots that the *NAC* inventory does not exhibit.

4.3. Surroundings contribution and resolution analysis

In order to evaluate the contribution degree of the emissions of neighbouring cells for a given location, an analysis was carried out over the cell with (C, F) coordinates equal to $C = 29$ and $F = 24$ according to the general methodology outlined in section 3.8. In this cell three monitoring locations are emplaced: station 21 - Final Calle Alcalá (CALC), station 24 - Urbanización Embajada (UEMB), and station 25 - Barajas Pueblo (BARJ). Due to its location at the outskirts of the city and being the only cell with two *traffic* stations and one *urban background* site (table 3.1) this cell was considered as a convenient starting point for the evaluation of contributions. The *simulated* meteorological fields needed to compute these contributions were obtained directly from the *WRF* model and are represented as a wind rose in figure 4.19. It is important to notice that from figure 4.20, the *expanded cell* covers a considerable region of the *urban nucleus* of Madrid. This might strongly suggest that since most of the emissions at an urban nucleus tend to be of a similar nature, the analysis carried out over this cell would be representative of

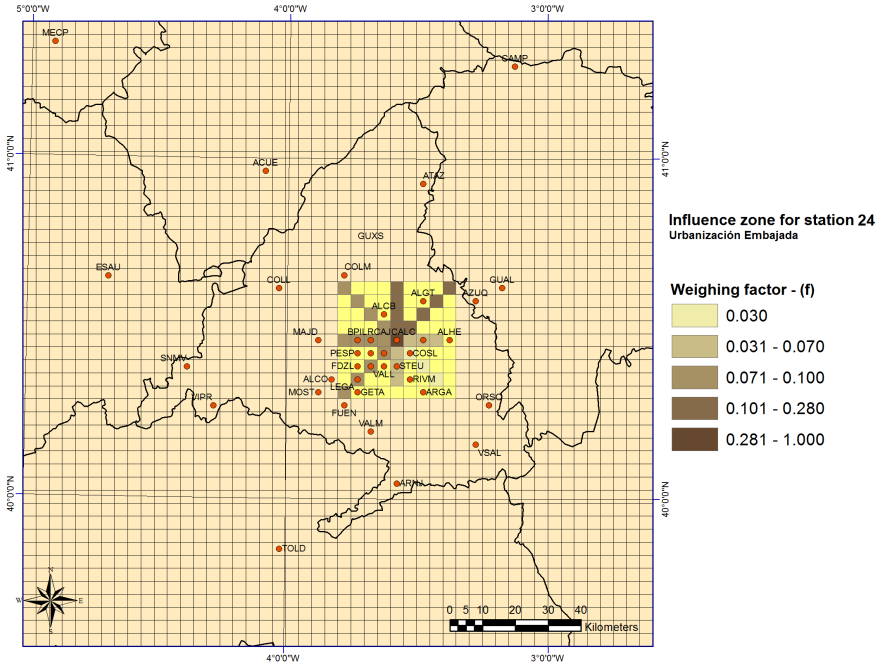

Fig. 4.20: Influence zone and weighing factors for cell (29, 24)

a vast extension of the *GMR*. Figure 4.20 also suggests that most of the contributions would eventually come from cells located in the north - east, north and south - west of the studied cell. For the general computation of the contributions, cell (29, 24) was weighed with a factor of $f = 1{,}0$, underlying the assumption that the emissions produced in this cell contribute 100% of the times to its pollutant concentrations.

The selected pollutants for this analysis were **nitrogen oxides** (NO_x) and **sulphur dioxide** (SO_2) which are not particullarly sensitive to boundary conditions. Moreover, since NO_x are produced mainly due to traffic activity, their **absolute** and **relative emissions** would undercover considerations made during the compilation of road traffic inventories. On the other hand, SO_2 is a very useful tracer for investigating activities related with combustion, namely *SNAP02* and *SNAP03*. Specifically for the latter activity group, the current analysis might reveal the influence of large point - sources on the studied cell.

The results of the analysis carried out over **nitrogen oxides** are presented in figure 4.21. Either for cell (29, 24) or the *expanded cell*, both inventories consider *SNAP07* as the most important emissive activity. Even further, the *CAM* inventory has assigned to this activity up to 90% of the NO_x emissions while the *NAC* inventory shows a more balanced contribution of *SNAP* groups other than *SNAP02* and *07*. Yet what seems striking in figure 4.21 is the fact that the total *expanded cell* shows an almost identical emission source pattern than the studied cell for *both* inventories.

This strongly suggests that the emissions occuring at neighbouring cells *do not* contribute greatly to the NO_x emissions for a given cell. This behaviour might find its primal cause in the fact that the NO_x emission sources at the neighbouring cells have been described under very similar conditions than those

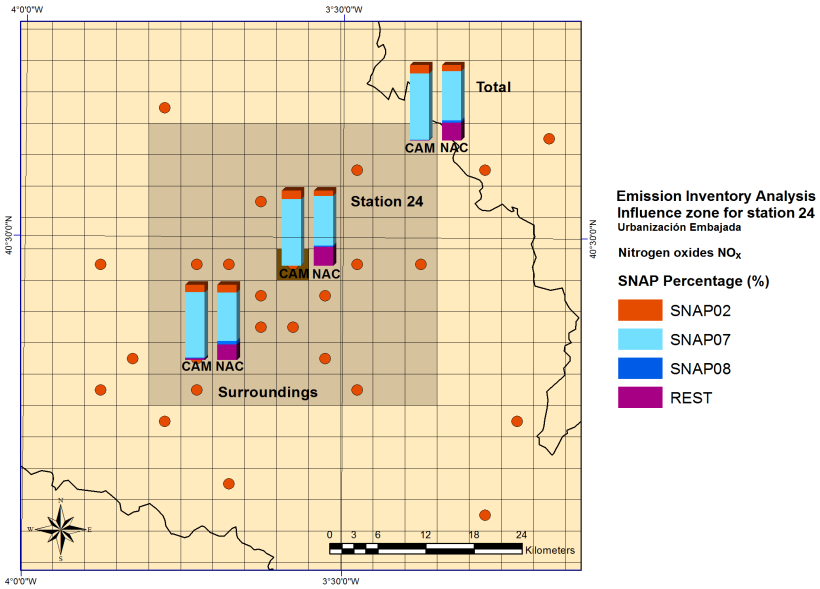

Fig. 4.21: Contribution analysis for nitrogen oxides (NO_x) around cell $(29, 24)$

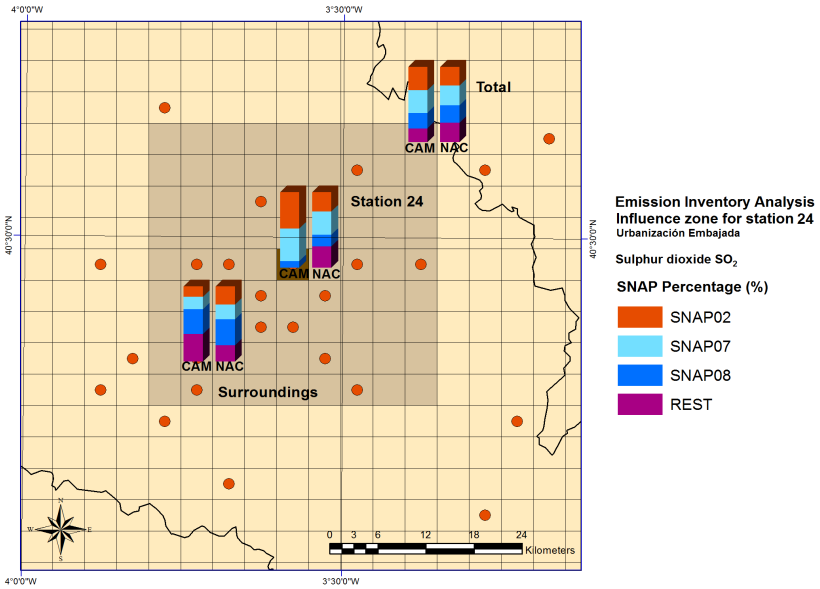

Fig. 4.22: Contribution analysis for sulphur dioxide (SO_2) around cell $(29, 24)$

applied to the studied cell. The latter might suggest that despite the fact of having a 4 *km* resolution, the *inventory compilation* resolution is much coarser for both, *NAC* and *CAM* inventories. In other words, both inventories have been compiled in such a way that it is useless to increase resolution since there is not an enough and reliable detail degree to describe it.

On the other hand, the results of the analysis on **sulphur dioxide** are shown in figure 4.22. SO_2 emissions clearly exhibit a different behaviour than the one described before for NO_x. While cell $(29,24)$ is clearly influenced by *SNAP02* for both inventories along with a sizeable *SNAP07* share, the *expanded cell* sees this influence buffered by the contribution of the *surroundings*. Interestingly, figure 4.22 shows that the emissions described by the *CAM* inventory do change after the *cell expansion* process; conversely, the *NAC* inventory shows a much smaller variation.

Hence, one must expect that since the *CAM* inventory is more sensitive to variations it should be too, more accurate for the chosen resolution for SO_2. Further afield, these changes suggest that the compilation structure for SO_2 is able to tell at a 4 *km* cell - level *where* are sulphur - related activities located. Unlike NO_x this analysis is easier for SO_2 due to the great contribution of point sources, rather than line sources. Yet, the degree of correctness of such considerations has not been evaluated so far.

4.4. Statistical Analysis

As to begin with the investigation, a group of stations was chosen for *each* pollutant from the information presented in the diagrams described in section 3.7. The selection criteria for these stations were emminently statistic and have already been explained in section 3.7.3. For the general purposes of this work, the term *well correlated station* means a location at which both *emission inventories* perform **very similarly** in addition to a fair statistical correlation degree. A *badly correlated station* is an emplacement at which the two *emission inventories* behave **differently**, regardless of its statistical correlation degree.

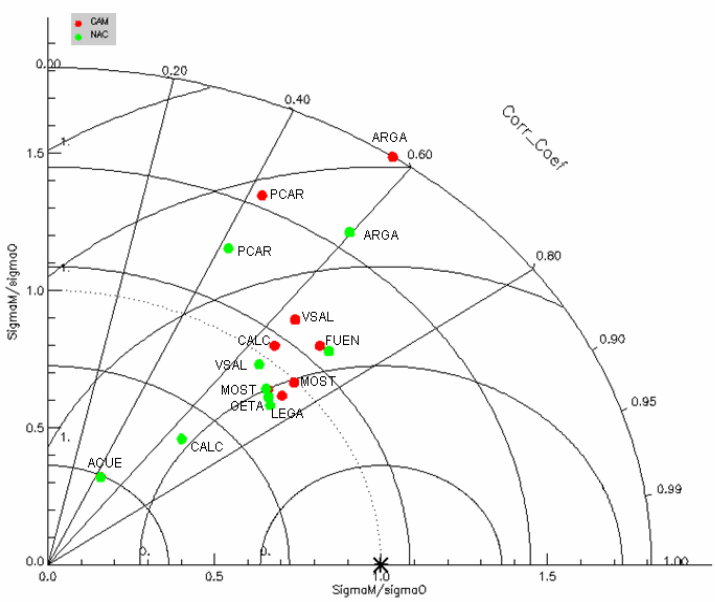

Fig. 4.23: Taylor diagram for chosen stations measuring NO_2

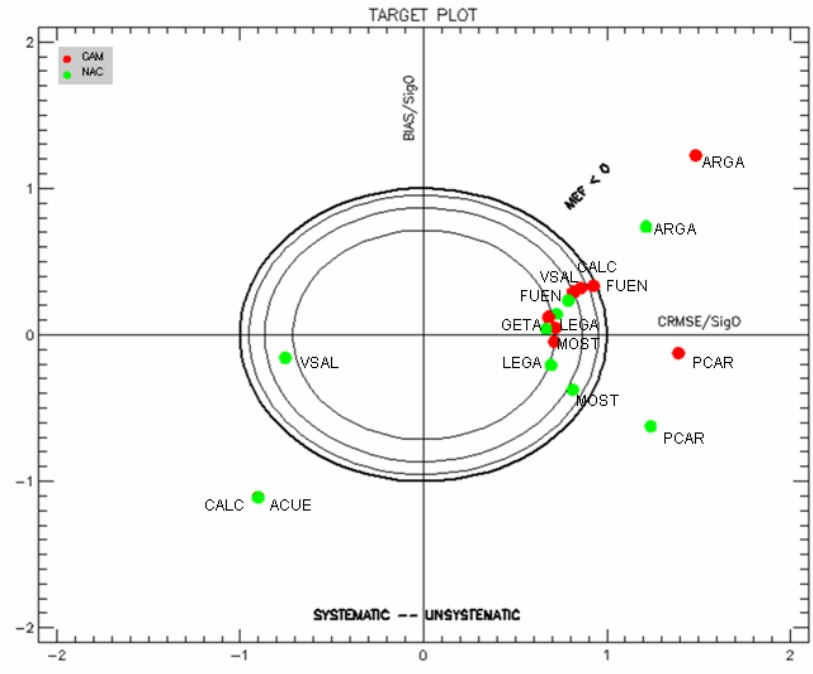

Fig. 4.24: Target plot for chosen stations measuring NO_2

4.4.1. Stations measuring nitrogen dioxide - NO_2

Although the reported pollutants for the emission inventories are total **nitrogen oxides** (NO_x), basically a sum of NO and NO_2, from this point further the assumption that *mostly all* the NO has been transformed into NO_2 will be considered valid in an effort to make comparisons easier. Such an assumption seems reasonable since **nitrogen dioxide** is a regulated pollutant, as well as one which rises concern according to what has been stated in section 3.5. Moreover, at urban entourages, NO rapidly oxidises to NO_2 (Fenger 2009; Monks et al., 2009). There is apparently no use in differentiating between NO and NO_2 at the present study, basically because the apportionment NO/NO_2 ratios were assumed and equally used at both experimental runs. The well correlated selected stations are presented in table 4.1, while the badly correlated stations chosen for this study are shown in table 4.2.

Tab. 4.1: Well correlated stations for nitrogen dioxide - NO_2

Station No.	Code	Station Name	Type
36	FUEN	Fuelabrada	Industrial
37	GETA	Getafe	Traffic
39	LEGA	Leganés	Traffic
41	MOST	Móstoles	Urban Background
53	ACUE	Acueducto	Traffic

Figure 4.23 is the *Taylor diagram* and figure 4.23 is the *target plot* for the before mentioned stations. As it is evident, most of the well correlated stations have *correlation coefficients* between $r = 0{,}6$ and $r = 0{,}8$, and lie along the $\sigma_M/\sigma_O = 1{,}0$ curve. Moreover, the red points of the *CAM* inventory are quite close to the green points of the *NAC* equivalent. Specifically, for station 53 (ACUE) both inventories correspond to be exactly the same, yet the *correlation coefficient* is somewhat low. Most of these stations lie inside the model efficiency score ($MEF > 0$) circle at the *target diagram*, which represent positively correlated model predictions and observations (Thunis et al., 2011). On the other hand, the badly correlated stations appear quite disperse in figure 4.23, exhibiting sizeable departures between the *CAM* and *NAC* points, being the largest deviation the one shown by station 21 (CALC). Figure 4.24 shows that most of the badly correlated stations lie outside the $MEF > 0$ circle, simultaneously exhibiting great deviations between inventories too.

Tab. 4.2: Badly correlated stations for nitrogen dioxide - NO_2

Station No.	Code	Station Name	Type
02	PCAR	Plaza del Carmen	Traffic
21	CALC	Final Calle Alcalá	Traffic
31	ARGA	Arganda del Rey	Industrial
48	VSAL	Villarejo de Salvanés	Rural

4.4.2. Stations measuring particulate matter - PM_{10}

Well correlated stations measuring PM_{10} are presented in table 4.3 as well as the badly correlated stations chosen which are shown in table 4.4, following the same criteria described above.

Tab. 4.3: Well correlated stations for particulate matter - PM_{10}

Station No.	Code	Station Name	Type
21	CALC	Final Calle Alcalá	Traffic
39	LEGA	Leganés	Traffic
44	SNMV	San Martín de Valdeiglesias	Rural

Tab. 4.4: Badly correlated stations for particulate matter - PM_{10}

Station No.	Code	Station Name	Type
26	ALHE	Alcalá de Henares	Traffic
28	ALCO	Alcorcón	Traffic

From figure 4.25 it is evident that the stations classified as badly correlated show wide departures between inventories, being station 26 (ALHE) the one with the largest deviation. Stations 21 (CALC) and 39 (LEGA) show the best *correlation coefficients*, never surpassing the r =0.6 line. Every station and inventory fell under the σ_M/σ_O =1,0 line which strongly suggests that observed PM_{10} concentrations are much higher than the actual modelled values.

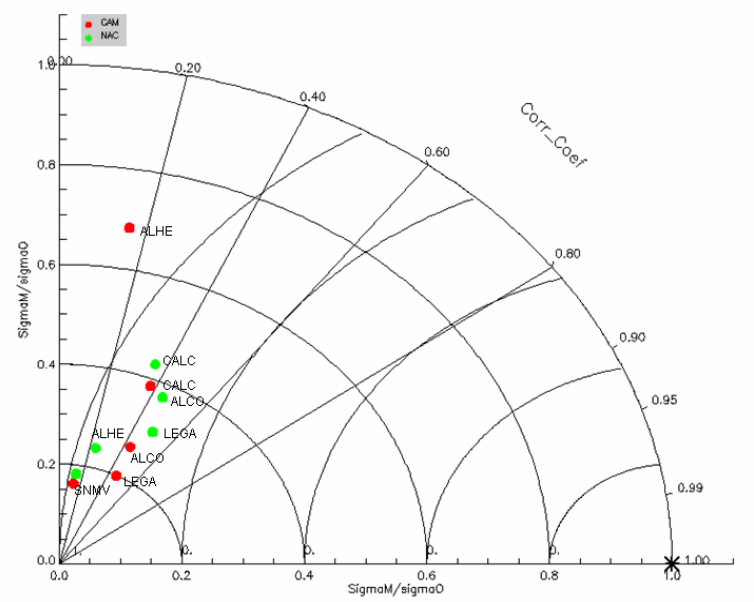

Fig. 4.25: Taylor diagram for chosen stations measuring PM_{10}

The *target plot*, (figure 4.26) clearly shows that there is indeed a *systematic error* in the general inventory compilation procedures for PM_{10}, since every station is located outside the $MEF > 0$ circle. Even the best correlated station of the entire monitoring network (station 21 - `CALC`) is barely underscored. This diagram highlighted the need to deeply study the general particle compilation criteria, as

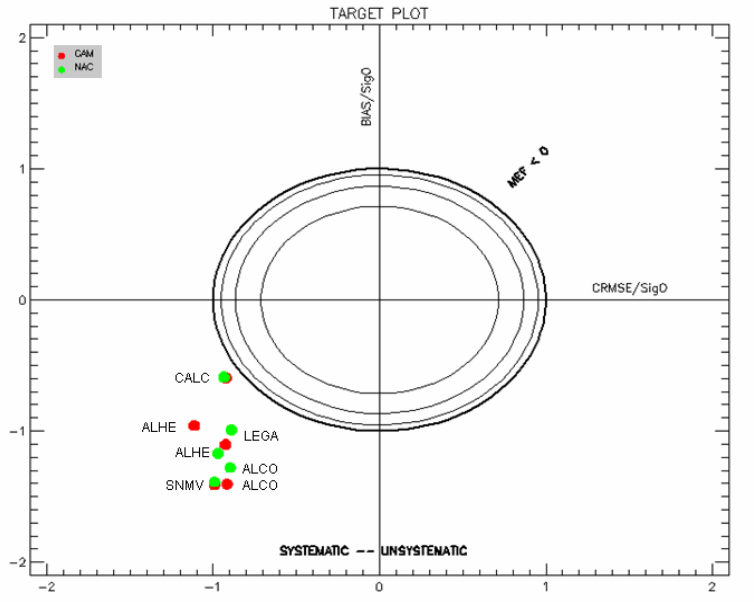

Fig. 4.26: Target plot for chosen stations measuring PM_{10}

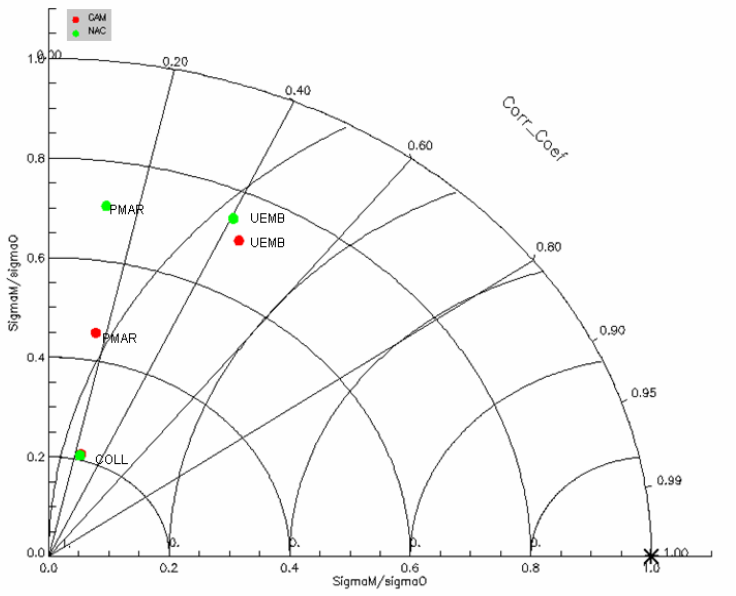

Fig. 4.27: Taylor diagram for chosen stations measuring $PM_{2,5}$

in the future sections of this work. The aforementioned premises might redound in the assumption that the systematic error present in every station of the modelled domain might lead to a considerable underestimation of the particle emissions; as it has been shown by the fact that σ_M/σ_O <1,0 for every case (figure 4.25), presumably because some emission sources are not being accounted.

4.4.3. *Stations measuring particulate matter - $PM_{2,5}$*

According to table 3.1, $PM_{2,5}$ is only measured by six locations in the entire monitoring network, therefore only three stations were chosen for analysis. For particulate matter ($PM_{2,5}$) the only badly correlated station is station 05 (PMAR). The rest, well correlated stations, are presented in table 4.5, following the criteria already applied for PM_{10} and NO_2. It is particullarly evident from figure 4.27 that station 34 hardly presents any departure between the *NAC* and the *CAM* inventories. However, the best correlated station for this pollutant is station 24 (UEMB), which shows a *correlation coefficient* of slightly above r =0,4 for both inventories.

Tab. 4.5: Well correlated stations for particulate matter - $PM_{2,5}$

Station No.	Code	Station Name	Type
24	UEMB	Urbanización Embajada	Traffic
34	COLL	Collado Villalba	Traffic

Moreover, according to figure 4.27, every station fell below the $\sigma_M/\sigma_O = 1{,}0$ which hints that the modelled values carry a systematic error due to the misconsideration of some particle emitting sources; this fact is further supported by figure 4.28, which presents every pointed outside the $MEF > 0$ circle. As

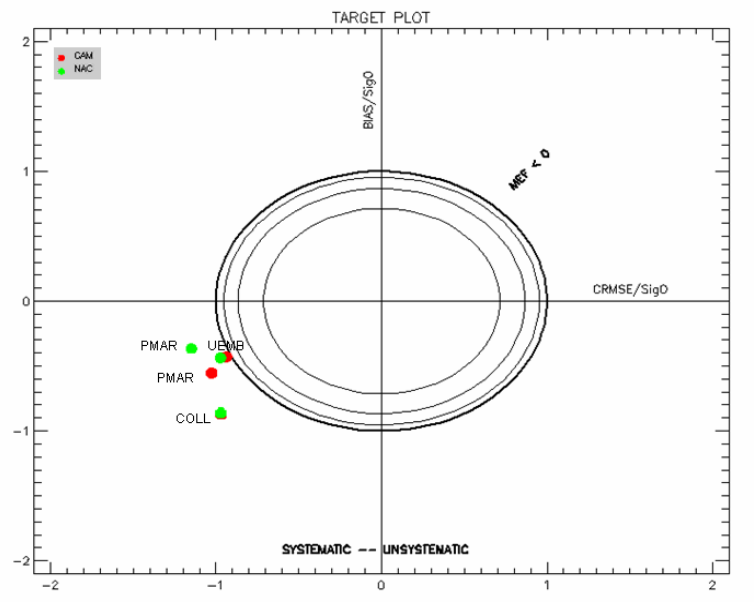

Fig. 4.28: Target plot for chosen stations measuring $PM_{2,5}$

this factor pointed out, it was necessary to carry on an extensive analysis on the causes of this behaviour and the particle compilation criteria for both inventories.

4.4.4. *Stations measuring sulphur dioxide - SO_2*

For sulphur dioxide, well correlated stations measuring SO_2 are listed in table 4.6 as well as the badly correlated stations which appear in table 4.7. For this specific pollutant, no *target plot* is presented due to the great dispersion for most stations provoked by great *mean bias* (*MB*) values.

Up to now, the tendency observed in figure 4.29 is way different to what has been already presented for other pollutants. Three out of four locations have $\sigma_M/\sigma_O \gg 1$, which indicates that there is either a big dispersion in modelled data (represented by a high σ_M) or a strong overestimation for this pollutant. The best correlated location corresponds to station 36 (FUEN), which shows a high *correlation coefficient* and a small departure between inventories. Station 53 (MECP) shows virtually no deviation between inventories, although the *correlation coefficient* is low. The latter is true because the *CAM* inventory does not consider any emission ocurring outside the Madrid province; for such cells, the emissions correspond to those described by the *NAC*. This fact thus, might be considered as a *good indicator* of the general *model skill*.

Tab. 4.6: Well correlated stations for sulphur dioxide - SO_2

Station No.	Code	Station Name	Type
36	FUEN	Fuenlabrada	Industrial
53	MECP	Medina del Campo	Industrial

Tab. 4.7: Badly correlated stations for sulphur dioxide - SO_2

Station No.	Code	Station Name	Type
04	BPIL	Barrio del Pilar	Traffic
31	ARGA	Arganda del Rey	Industrial

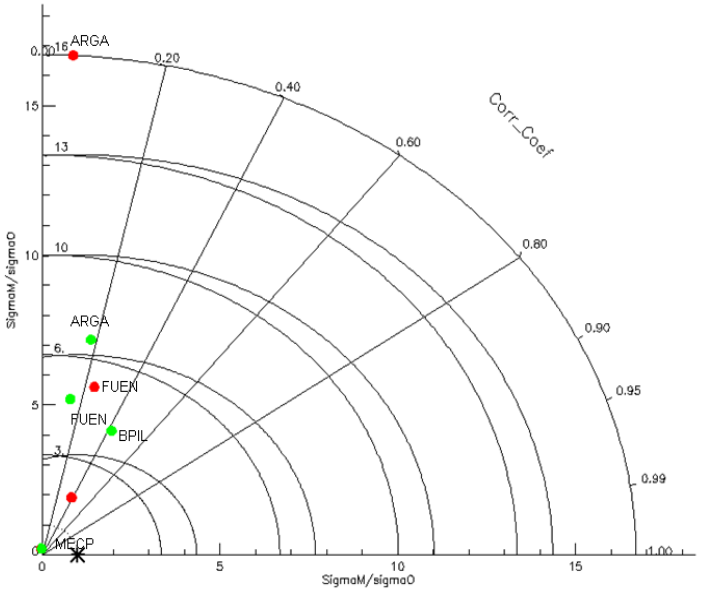

Fig. 4.29: Taylor diagram for chosen stations measuring SO_2

Conversely the worst correlated data pair is that of station 31 (ARGA), which shows the largest deviation between inventories along with poor *correlation coefficients*. Station 04 (BPIL) shows also sizeable departures between inventories, yet the *correlation coefficient* is better and the σ_M/σ_O smaller.

4.5. Emission analysis at cell - level

4.5.1. Detailed analysis for nitrogen oxides emissions - NO_x

Station 37 - Getafe (GETA) is labelled according to table 3.1 as a *traffic type* station and therefore suitable for measuring NO_2 coming emminently from traffic - related emissions. In line with what has been stated in section 4.4.1, the statistic indicators for this station are quite good so a deep analysis to undercover the causes of such a good correlation between observations and inventories. For the cell where this station is located - with coordinates $(26, 20)$ - the *SNAP07* category can be apportioned in the following activities (table 4.8).

Through inspection of figure 4.30, other stations that exhibit low *MB* values lie also within the southern region of Madrid, referred to as *urban south*. Such locations are station 36 - Fuenlabrada (FUEN) and station 39 - Leganés (LEGA).

Tab. 4.8: SNAP07 activities for station 37 (GETA). Emissions (E) in $[ton \cdot day^{-1}]$

Code	Description	$\%_{SNAP07,NAC}$	E_{NAC}	$\%_{SNAP07,CAM}$	E_{CAM}
070101	Passenger cars - highway driving	12%	0,33	13%	0,33
070103	Passenger cars - urban driving	33%	0,90	22%	0,56
070301	HDV - highway driving	7%	0,21	38%	0,98
070303	HDV - urban driving	25%	0,66	12%	0,32
070503	Motorcycles $> 50cm^3$ - urban dr.	–	–	11%	0,29
–	**Total**	100%	2,67	100%	2,53

Tab. 4.9: Well correlated stations at *urban south* region. Emissions (E) in $[ton \cdot day^{-1}]$

Station No.	Code	Station Name	$E_{NOx,NAC}$	$E_{NOx,CAM}$
36	FUEN	Fuenlabrada	1,64	1,82
37	GETA	Getafe	2,67	2,53
39	LEGA	Leganés	2,89	3,21

As it is evident from figure 4.30, station 37 (GETA) has very low MB values, namely MB =3,9 ppm for the *NAC* inventory and MB =1,8 ppm for the *CAM* version. The good correlation coefficients this station yields, as well as the *low* mean bias values (MB) might be attributed to a somehow similar characterisation of the *driving situations*, which according to table 4.8 are analogous and correspond with reality reasonably well (AECM, 2005; ALMUDENA, 2011).

Table 4.9 shows that both inventories compute very similar total emissions for *SNAP07*. The reason for this coincidence is the fact that both inventories consider a *similar* apportionment between driving situations and analogous contributions from passenger cars and HDV. In some cases, an excess in HDV might be compensated by the other inventory with an excess in motorcycles, to yield similar gross totals. A mean *driving situation apportionment* for the *urban south* region would be 15% for passenger cars under highway driving, 25% for passenger cars under urban driving, 23% of HDV under highway driving and 16% of HDV under urban driving.

Conversely, station 48 - Villarejo de Salvanés (VSAL) is classified as a *rural type* station (table 3.1), a good station in which the assumption of high *urban driving situations* might not be appropriate. The statistic indicators for this station are generally poor. The activity variables for the cell where this station is located $(35,16)$ are shown in (table 4.10). Although the emplacement for this station is considered as *rural*, it lies not far from the $A-3$ national highway. This fact interferes with the labelling of this station and clearly rules over the inventory compilation criteria that describe this location.

On the other hand, it is noticeable from table 4.10 that the *driving situations* are wrongly characterised. There is a high presence of vehicles under *urban driving situations* which might not agree with the station's label (*rural*). The difference between both *SNAP07* NO_x emissions is also striking, 0,74 $ton \cdot day^{-1}$ for *NAC* and 1,52 $ton \cdot day^{-1}$ for *CAM*, this is, sevenfold. Activity 070301 (HDV - highway driving) is responsible for this huge departure between *SNAP07* totals, while the rest of activities correspond fairly between inventories.

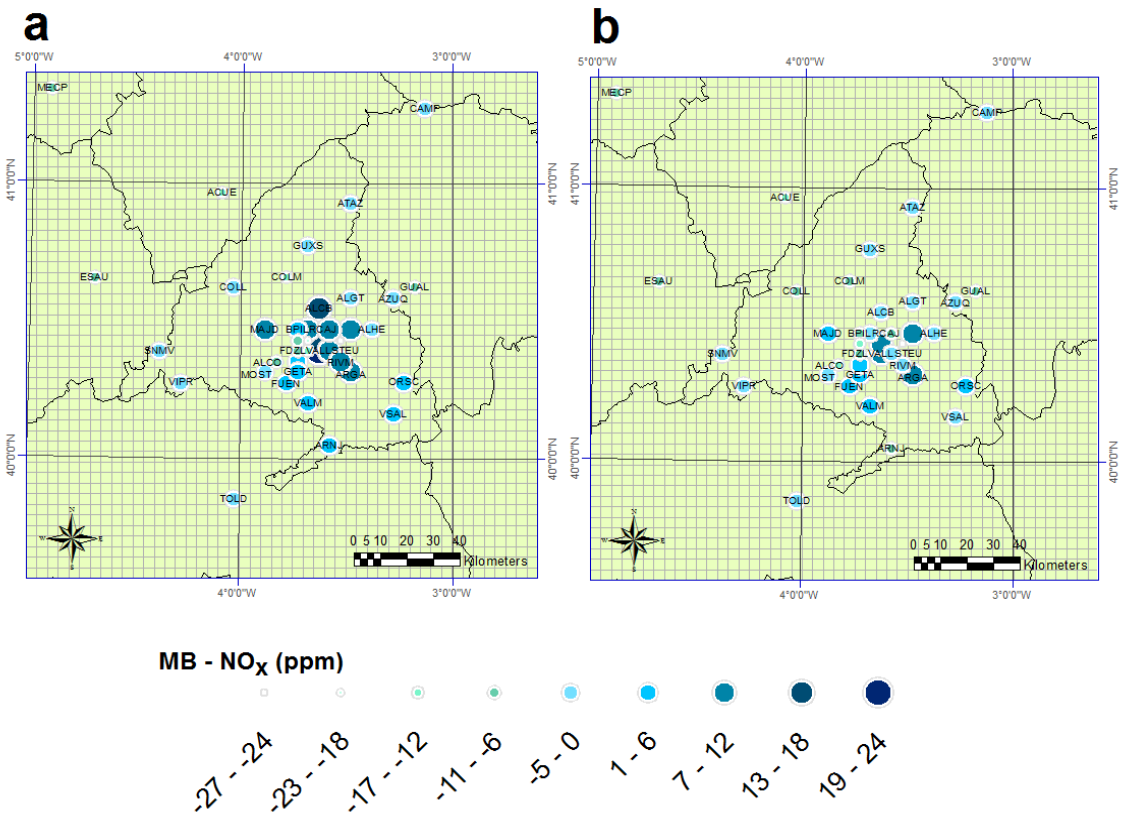

Fig. 4.30: Mean Bias (*MB*) values for NO_2 from a) *CAM* and b) *NAC* inventories

Tab. 4.10: SNAP07 activities for station 48 (VSAL). Emissions (E) in $[ton \cdot day^{-1}]$

Code	Description	$\%_{SNAP07,NAC}$	E_{NAC}	$\%_{SNAP07,CAM}$	E_{CAM}
070101	Passenger cars - highway driving	33%	0,24	23%	0,36
070103	Passenger cars - urban driving	14%	0,14	1%	0,02
070301	HDV - highway driving	20%	0,15	69%	1,06
070303	HDV - urban driving	10%	0,08	0%	0,01
–	**Total**	100%	0,74	100%	1,52

In order to have a broader scope of this situation, the same analysis was carried out over stations 31 - Arganda del Rey (ARGA), 43 - Rivas Vaciamadrid (RIVM), and Orusco de Tajuña (ORSC) which are located along the $A-3$ highway. Table 4.11 shows the emissions according to both inventories for activity 70303 (HDV - highway driving), for the stations placed along the $A-3$ highway. There are sizeable differences in both emissions according to table 4.11. In every case, the *CAM* inventory reports emissions far above the *NAC* equivalent. Further analysis on the cause of such noticeable departures is carried out in the forecoming section where methodological issues will be discussed.

Tab. 4.11: SNAP 070303 Emissions for $A-3$ stations. Emissions (E) in $[ton \cdot day^{-1}]$

Station No.	Code	Station Name	$E_{070303,NAC}$	$E_{070303,CAM}$
31	ARGA	Arganda del Rey	0,22	0,91
42	ORSC	Orusco de Tajuña	0,01	0,02
43	RIVM	Rivas Vaciamadrid	0,07	1,22
48	VSAL	Villarejo de Salvanés	0,15	1,06

4.5.2. Detailed analysis for NO_x emission inventorying

NO_x emissions originated from SNAP02 activities

Within this *SNAP* group, combustion devices with low thermal capacities are included, especially those destined for heat generation purposes, and to a lesser extent, heat and electricity (cogeneration) for individual use at residential and service buildings. Up to now, a detailed inventory of the combustion devices is not available. However, the information received from the major facilities that incorporate such devices (hospitals, commercial centres, office buildings, etc.) revealed that the nominal thermal power of these is almost completely below 50 MWh, for the concrete Spanish case.

It is openly recognised at the National Emissions Inventory Document (*NAC*) that in order to estimate the emissions of pollutants, it would be desirable to know, as detailed as possible, the characteristics of the installed devices, their operation cycles, emission control techniques and fuel consumptions. Since this information was non - existent in most cases, and due to the great variety of combustion devices existing at the residential, institutional and commercial sectors, the inventorying of this emissions was carried out basically through statistical procedures. The use of emission factors was applied at an area level.

The main activity variable that was used to estimate the emissions for this group was the fuel consumption for each device, expressed in Giga Joules (GJ) and quantified as a Low Heating Value (LHV). The fuel mix that this inventory has considered is quite diverse, expressed usually in mass or volume units, for either solid, liquid or gaseous fuels. The composition of such a fuel mix for this *SNAP* group includes coal, black lignite, petroleum coke, wood and woodchips, agricultural waste, fuel oil, gas oil, kerosene, natural gas, *LPG*, coke gas, biogas and manufactured gas. The information regarding the composition of each of the fuels came from several information sources. For instance, wood sulphur composition was obtained from the 1992 Perry's Chemical Engineering Handbook while its carbon content was estimated through the 1991 *OECD* document "Estimation of Greenhouse Gas Emissions and Sinks".

The energy balances were carried out according to a series of *EUROSTAT* and *OECD* documents, namely "Energy Balances Sheets" and "Energy Statistics of *OECD* Countries", which cover the temporal span between 1990 and 2006. Further information for 2007 was obtained through a series of questionnaires on coals, petroleum products and natural gas, reported by the Spanish Ministry of Industry, Tourism and Commerce (*MITYC*). Information on wood, vegetal coal and wood residues was obtained from a study published by the Institute for Energy Saving and Diversification (*IDAE*), published in 1995. Since this study includes information up to the year 1995, further data extrapolation and surrogation was carried out for year 2007. The emission factors for most fuels and devices were obtained from the

EMEP/CORINAIR guidebook (MARM, 2009).

The *CAM* inventory exhibits the same problem that has already been stated for the National version for this specific *SNAP* group. This means there is not a detailed inventory of the number and installation types of commercial, institutional and domestic boilers. This inventory applies exactly the same assumption that was made by the National version: all of the non industrial boilers have a nominal power of less than 50 MW. Moreover, due to the lack of information, the total number of installed boilers in the Autonomous Community of Madrid is still unknown. Most emissions related with this group come from fuel consumptions at boilers from heating systems, gas stoves and domestic hot water (*DMW*).

Since most of the information that is available regards fuel consumptions for global sectors such as agriculture, industry, transport, services, etc., the implemented method relies on the use of emission factors. The most relevant activity variables related to this sector are fuel consumptions and the proper assignation to the respective sectors accordingly. The considered information sources were the General Industry, Energy and Mines Direction of the Autonomous Community of Madrid (Dirección General de Industria, Energía y Minas de la Comunidad de Madrid); the 2007 National Emissions Inventory; and the 2007 Energetic Balance of the Community of Madrid (Balance Energético de la Comunidad de Madrid), among others (CAM, 2009).

NO_x emissions originated from SNAP07 activities

Vehicle Fleet Characterisation.

The "General Statistic Yearbook" published by the General Traffic Direction (*DGT*) of the Ministry of the Interior (*MI*) provides information about the current Spanish vehicle fleet, according to the following categories: (1) passenger cars, (2) buses, (3) trucks, (4) motorcycles, (5) mopeds, and (6) industrial tractors. For both inventories, the characterisation of the vehicle fleet was carried out attending to the disposed measures stated in Annex II of the Royal Decree 2822/1998 of September 23^{rd}, which approved the General Vehicle Regulations.

The characterisation of the vehicle fleet was accomplished through considering the registration years of vehicles as an indicator of their environmental characteristics. Thus, a relationship between the age of a given vehicle fleet and their emissions could be constructed through the use of the *COPERT* methodology (Ntziachristos and Samaras, 2000). Crossing the before mentioned information with data such as fuel type and fuel consumption gave a complete picture of the vehicle fleet.

Driving Situations Characterisation.

This characterisation of driving situations for both inventories was carried out according to the guidelines provided by *CORINAIR*, in which the problem of their description is solved through the definition of three different vehicle circulation conditions: highway, rural and urban (EMEP - CORINAIR, 2007). Yet, the National Inventory states that the lack of a strict European Norm is an impediment for a homogenous characterisation process between Member States. For the different existing vehicles, a representative circulation speed defines the driving situation according to table 4.12.

Fuel Consumptions.

The information regarding fuel consumptions has been obtained from a series of sources. The most important documents used where the *OECD* "Energy Statistics of *OECD* Countries" document, the *EUROSTAT* "Energy Balance Sheets" or the "Consumption Statistics of Petroleum Products" of the

Tab. 4.12: Driving situations according to *COPERT III*

Vehicle Type	Highway Driving	Rural Driving	Urban Driving
Passenger cars	105 km/h	65 km/h	25 km/h
LDV	100 km/h	65 km/h	25 km/h
HDV	95 km/h	60 km/h	25 km/h
Buses	95 km/h	60 km/h	25 km/h
Motorcycles	105 km/h	65 km/h	25 km/h
Mopeds	–	45 km/h	25 km/h

Ministry of Economy. The information from this documents has been extrapolated or surrogated when missing for a given year or specific data set, just as with other *SNAP* groups.

Travelled Distances.

The mean travelled distance assumed for the *NAC* inventory was 12 *km*, according to a series of estimations from a group of European countries described at the *COPERT* Methodology (Ntziachristos and Samaras, 2000). The consulted source for the estimation of travelled distances was the trip database kept by the General Highway Direction (*DGC*) of the Ministry of Development (*MF*). The information coverage was variable from year to year. For the period comprised between 1996 and 2007, data from the national highways, as well as the autonomic and regional are disaggregated in a province - resolution.

The *CAM* inventory calculation procedure involves a sum - product of the number of vehicles that circulate through a given road, the travelled distance on that road and an emission factor. The information sources that have been used are the Highway Maps of the Ministry of Development (*MF*) (2007) and data given by the General Highway Direction (*DGC*) for 2007. An analysis was carried out to each of the before mentioned sources, just to be found that some data from the General Highway Direction (*DGC*) are missing if compared to the Highway Maps of the Ministry of Development (*MF*). The overall vehicle classification presented by the General Traffic Direction (*DGT*) has been thoroughly listed for the National inventory (*NAC*) and since it does not correspond to that exposed by *COPERT III*, a series of assumptions were made to cluster vehicles within the latter criteria.

The general estimation of the travelled distances through each road section was made from the series published by the General Traffic Direction (*DGT*). The available data for each of the road sections in which the Community of Madrid has been divided were (i) length, (ii) mean daily intensity and (iii) HDV percentage. The circulating traffic on each of the road sections has been carried out through a distribution of the mean daily intensities obtained from the Ministry of Development (*MF*) or the General Highway Direction (*DGC*).

General Calculation Procedure.

The emissions of pollutants such as nitrogen oxides (NO_x) are, according to the *CORINAIR* methodology, a function of the motor operation schemes, cold start and hot start. The nature of the emissions according to each of the operation schemes is very different, which is actually reflected in the emission functions.

- *Hot start emissions*. Emissions have been calculated through the application of emissions factors to the travelled distances for each vehicle or also, through fuel consumptions previously estimated over such travelled distance and speed.

- *Cold start emissions*. Emissions produced under a cold start scheme are estimated as additional to those produced by the engine under a hot scheme. Although these emissions occur at every driving situation and for every vehicle category, the *CORINAIR* methodology has only emission factors for LDV available. Cold start emissions are a function of the ambient temperature too.

4.5.3. Discussion - NO_x emission inventories

NO_x emissions originated from SNAP02 activities

The activities that are related with the emission distortions are the use of boilers at commercial and institutional sectors (020103), as well as in residences (020302). Stationary engines in agriculture (020304) are also included in this category. The observed differences are a consequence of the used activity variables in both inventories. In the specific case of the *SNAP* 020304 group, the stationary engines have high emission factors for nitrogen oxides; in addition, the high activity variable assigned by the *CAM* inventory accounted for even higher NO_x emissions.

Small sources like domestic combustion sources are very numerous and they have significant impacts on emission inventories. These sources are usually not well characterised, yet they are expected to be different from large - scale combustion sources. For these sources, emissions do not only depend on the feedstock type but also on the way in which combustion is accomplished. Thus, the extrapolation of emission factors or emission rates within combustion types, sources or geographic locations should be extremely careful (Zhang and Morawska, 2002).

NO_x emissions originated from SNAP07 activities

Both emission inventories have determined the vehicle numbers based on statistical information provided by the General Traffic Direction (*DGT*), exhibiting slight differences. However, the interurban and highway driving situations considered by the *CAM* inventory correspond to the rural and interurban driving situations stated by the *NAC* counterpart. The driving speed considered by the *CAM* inventory is the one available for each driving situation, according to the available data and when such data are unknown, the default values proposed by the *NAC* inventory are incorporated; an exception occurs with the interurban driving situation speed which is kept at 100 km/h, while the *NAC* inventory's default speed is 105 km/h.

The *NAC* inventory obtained the trip data from the General Highway Direction (*DGC*), while the *CAM* version used the Highway Map of the Ministry of Development as data source. Although the methodology is analogous, the emission quantification was carried out by the *CAM* inventory for the interurban and rural driving situations through the use of a daily vehicle flow referred to a road length, while the *NAC* version incorporated trip information. It considers also the estimated trips for the 1990 - 2006 national emission inventories for the urban driving situation. For each driving situation, when the emission factor per travelled kilometre is not available from *COPERT*, the emissions should be estimated through fuel consumptions.

The trips calculated by the *NAC* inventory at interurban and rural driving situations incorporate annual trip data from the national, autonomic and local road networks provided by the *DGC*. Such data are detailed by province and vehicle class. Each of the calculated trips is distributed according to fuel type

and age through provincial vehicle fleets information; distributions according to each driving situation are also carried out. Trips made under an urban driving situation are estimated from fuel consumptions at these conditions, as well as through the use of emission factors.

The main problem for traffic related activity at the studied stations seems to be an excessive contribution of heavy duty vehicles in highway driving situation. In principle, activity ratios (traffic intensity) should be more accurately depicted by the *CAM* inventory, since it is based on link - specific traffic intensities. However, it seems that *COPERT* has not been run at road - level, being secondary emission factors used instead. This indicates that the emission factors derived for heavy duty vehicles are largely overestimated, thus pointing out the need of a purely bottom - up computation approach whenever detailed traffic data are available. Moreover, road traffic emission factors are based usually on measurements carried out using dynamometers under various driving cycles. Dynamometer tests help to determine the emission standards for regulation purposes, while they are not necessarily indicators of real driving conditions as well as the level of maintenance of the fleet of vehicles (Mellios et al., 2006).

Road traffic emission factors are usually obtained to represent long - term vehicle population averages for a certain vehicle class. Such emission factors are based on average and representative inputs. The activity data are usually transposed from larger geographic scales (such as national or regional) through the use of surrogates, usually population or census tracts. Additional difficulties seem to arise in the gathering and incorporation processes of consistent local traffic data for large domains, such as regional. However, it has been widely recognised that activities coming from larger geographic scales do not necessarily reflect local scale conditions. It is thus necessary to reconsider whether a top - down inventory, which is useful for natural scale applications, is still valid for its regional and local equivalents (Cook et al., 2006).

Usually emission factors that describe road traffic carry large uncertainty levels, basically due to external influences like driving behaviours, vehicle maintenance, the physical state of the roads, and so on. Traffic jams are usually not described by the emission factors either. Normally, through a series of emission campaigns, actual measured transport emissions are observed higher than the estimated or modelled emissions through emission factors (Mensink, 2000).

Another aspect that needs to be analysed is how traffic is being addressed. Traffic modelling tends to describe the fleet composition through its actual operation on a given road network by considering a certain number of vehicles driving through a specific road length. Emissions are thus modelled as a function of the attributes of the different vehicle types and a function of the driving condition too (Tønnesen, 2010).

Furthermore if the inventory would be compiled under a bottom - up scale, the on - site description of traffic is a complicated task, since traffic measurements are not usually available on a local scale to control or support any modelling effort. It is therefore imperative to carry out this traffic characterisation through the use of traffic models, which incorporate a series of assumptions (van de Kassteele, 2006; Tønnesen, 2010).

4.5.4. Detailed analysis for particulate matter emissions - PM_{10}

According to the selection procedure carried out in section 4.4.2 for PM_{10} stations, station 21 - Final Calle Alcalá (CALC) is considered a well - correlated measuring location. This station is labelled according to table 3.1 as *traffic type*, so it is an ideal site for evaluating particle emissions coming from road traffic sources. Table 4.13 shows a deeper insight into the traffic - related emissions that have been

compiled under both inventories. The *NAC* inventory generally overlooks highway driving situations for this location, while it concentrates basically in the urban modalities. Furthermore, a noticeable difference is the fact that the *NAC* inventory weighs importantly the contribution of LDV under urban driving situations, which the *CAM* inventory ignores completely. However, probably the most striking difference between the *NAC* and the *CAM* inventories is the fact that the latter does not consider sizeable emissions coming from *SNAP* categories 070700 and 070800, namely automobile tyre and brake wear and automobile road abrasion, reputed particle sources.

Tab. 4.13: SNAP07 activities for station 21 (CALC). Emissions (E) in $[ton \cdot day^{-1}]$

Code	Description	$\%_{SNAP07,NAC}$	E_{NAC}	$\%_{SNAP07,CAM}$	E_{CAM}
070101	Passenger cars - highway driving	–	–	36%	0,10
070103	Passenger cars - urban driving	40%	0,07	32%	0,09
070203	LDV - urban driving	13%	0,03	–	–
070301	HDV - highway driving	–	–	32%	0,09
070303	HDV - urban driving	12%	0,02	11%	0,03
070503	Motorcycles $> 50cm^3$ - urban dr.	–	–	14%	0,04
070700	Automobile tyre and brake wear	15%	0,02	–	–
070800	Automobile road abrasion	7%	0,01	–	–
–	**Total**	100%	0,17	100%	0,27

If assessing the gross totals, one can see that the *CAM* inventory computes 0,1 $ton \cdot day^{-1}$ above the *NAC* equivalent, even while overlooking important particle sources. The main contributing activity to this excess is a high presence of motorcycles $> 50cm^3$ under urban driving situations. According to figure 4.31, the MFB values suggest that the general tendency for underestimation of the *CAM* inventory is larger than the one shown for the *NAC* inventory, basically due to the misconsideration of some emissive sources. At this point it is worthy noticing that although this station was classified as *well - correlated* in section 4.4.2 still show poor statistic indicators.

Further analysis on this issue will be carried out on station 44 - San Martín de Valdeiglesias (SNMV) which according to section 4.4.2 is a classified as a well correlated station. This emplacement is ideal for analysing off - road contributions since it is labelled as *rural* (table 3.1).

Tab. 4.14: SNAP08 activities for station 44 (SNMV). Emissions (E) in $[ton \cdot day^{-1}]$

Code	Description	$\%_{SNAP08,NAC}$	E_{NAC}	$\%_{SNAP08,CAM}$	E_{CAM}
080600	Agriculture vehicles	14%	0,0004	–	–
080700	Forestry vehicles	–	–	7%	0,0002
080800	Industry vehicles	85%	0,0021	92%	0,0021
–	**Total**	100%	0,0025	100%	0,0022

For this *SNAP* group, the total emissions are analogous, being activity 080800 (Industry vehicles) the one that contributes the most for both inventories. Since both of them quantify the same emissions for this activity, it is clear that the criteria used to describe it are similar. The rest of the activites have a negligible contribution to the total emissions computed for this cell.

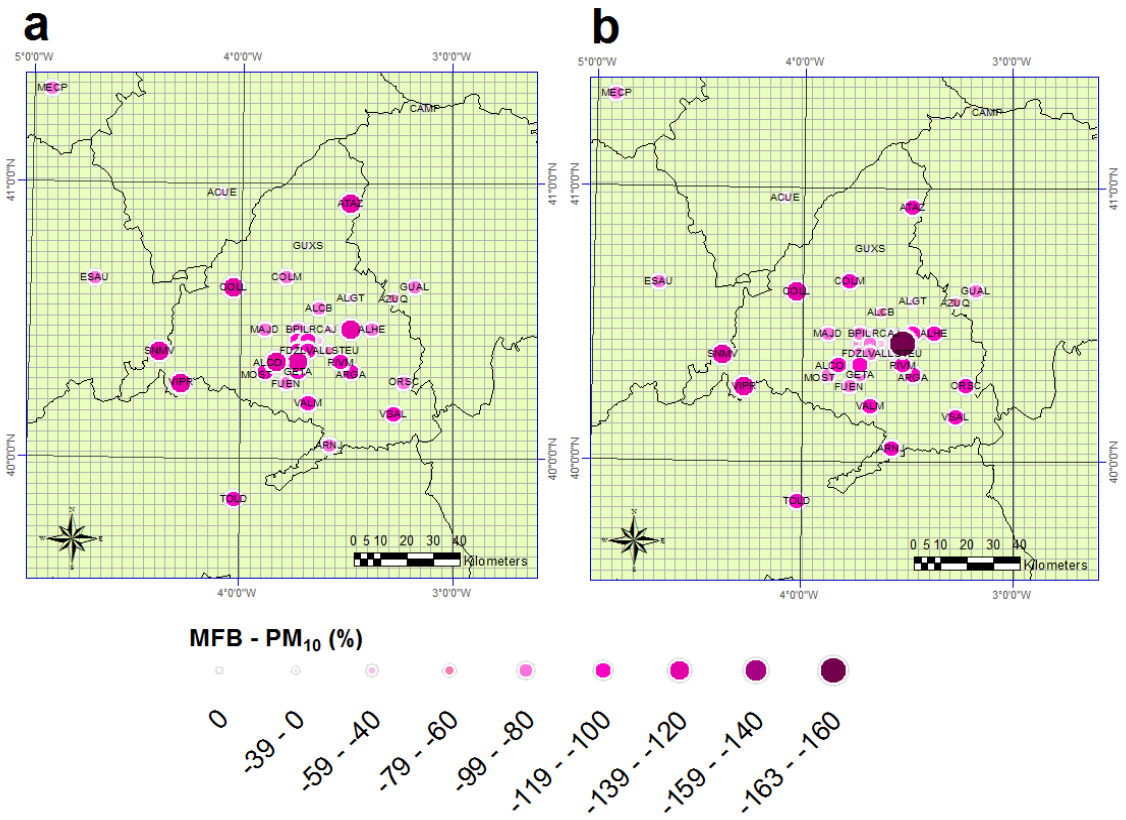

Fig. 4.31: Mean Fractional Bias (MFB) values for PM_{10} from a) *CAM* and b) *NAC* inventories

Again, a good station for assessing *SNAP10* contributions is station 44 - San Martín de Valdeiglesias (SNMV) (section 4.4.2). The considered activities for this cell are those listed below in table 4.15. The *NAC* inventory clearly consdiers more categories than the *CAM* counterpart, of which activity 100500 has an overwhelming contribution to the total emissions. If comparing the gross totals for both inventories, the emissions compiled by the *NAC* inventory are 60 times higher than the emissions considered by the *CAM* equivalent.

Tab. 4.15: SNAP10 activities for station 44 (SNMV). Emissions (E) in $[ton \cdot day^{-1}]$

Code	**Description**	$\%_{SNAP10,NAC}$	E_{NAC}	$\%_{SNAP10,CAM}$	E_{CAM}
100202	Arable land crops w/o fertilisers	7%	$1,2 \times 10^{-5}$	100%	$2,6 \times 10^{-6}$
100204	Market gardening w/o fertilisers	20%	$3,2 \times 10^{-5}$	–	–
100500	Manure management	72%	$1,2 \times 10^{-4}$	–	–
–	**Total**	100%	$1,6 \times 10^{-4}$	100%	$2,6 \times 10^{-6}$

Figure 4.31 shows that for station 44 (SNMV), the general understimation tendency is higher for the *CAM* inventory than for the *NAC* version. This basically suggests that the first inventory is counting much less emissions than the second one. However, since emissions for this *SNAP* group are not *considerable* in terms of absolute emissions, the differences show great disparities in compilation criteria.

4.5.5. Detailed analysis for PM_{10} emission inventorying

PM_{10} emissions originated from SNAP07 activities

The quantification of PM_{10} emissions according to both inventories has been carried out following the *COPERT* methodology, with the same underlying criteria than those stated in section 4.5.2. This is valid for categories related with vehicle flow and circulation (070100, 070200, 070300).

Furthermore, particle emissions are also considered through the estimation of brake and tyre wearing process. The algorithm involves the vehicle's number of axes, its loading level and its total capacity. The wearing phenomenon is also described by applying a correction factor related to the travelled distance and the circulation speed. Particle emissions are also accounted from the pavement abrasion phenomenon, which is estimated as a function of the travelled length and the vehicle class. This abrasion process is highly sensitive to the loading level of heavy - duty vehicles.

PM_{10} emissions originated from SNAP08 activities

For this *SNAP* group, the *NAC* inventory implemented emission factors based on the *CORINAIR* methodology and a the "Guidebook on the Estimation of the Emissions of Other Mobile Sources and Machinery" (Samaras and Zierock, 1994). In the case of particles, this inventory has assumed these mobile sources as diesel - powered, using specific emission factors quantified for the mean power of the vehicle fleet category. These information was spatially disaggregated with information coming from the Statistical Yearbook of the Ministry of Development (*MF*).

Interestingly, the *CAM* inventory states that the same methodology than the *NAC* variety has been applied to the quantification of particles. However, this inventory implemented a census of agriculture and forestry vehicles that has been used to characterise the fleet.

PM_{10} emissions originated from SNAP10 activities

In general, both inventories consider that particles are emitted as a consequence of the harvesting and ploughing operations on fields. The methodology used for the estimation of emissions from farming lands (*SNAP* 100102) is accomplished through the incorporation of emission factors available for arable soils; such emission factors are multiplied by the activity variable, a hectare in this case.

This procedure is carried out through the *RAINS* model, obtained from the *IIASA* document titled "Modelling particulate emissions in Europe, a framework to estimate reduction potential and control cost". The mean emission factor for arable soils, incorporated into this inventory, equals 0,1 $ton \cdot ha^{-1}$. The information regarding cultivated surfaces, disaggregated into crop categories has been taken from the "Agricultural and Food Statistics Yearbook", published by the Ministry of the Environment (*MARM*), which uses the European *NUTS-3* territorial classification. The emissions generated by livestock - related activities (100500) are originated basically trough feeding, manure and housing operations, as well as through the emission of hair, skin, pollen grains and insect parts. This emission group has been created exclusively by the inventory compilers since the *CORINAIR* methodology specifies that these activities would have been included under the 101001 and 101009 categories. It has been assumed that particles

emitted by livestock related activities are of both, PM_{10} and $PM_{2,5}$ type, and their emission factors per livestock unit were obtained from the *EMEP/CORINAIR* guidebook.

For the *CAM* inventory, the only activity on which specific differences with the *NAC* inventory were observed was that of cultures without fertilisers, more concretely regarding arable crop lands (100202). The most problematic pollutant is particulate matter, which is emitted during preparation for cultivation. The general methodology applied for this activity is the same applied by the *NAC* inventory which is based itself too on the *EMEP/CORINAIR* guidebook. For this purpose, the unfertilised cultivated surface had to be obtained from the Agricultural Atlas published by the Ministry of the Environment (*MARM*). The emission factors finally, came directly from the National inventory (*NAC*) on its former version.

4.5.6. Discussion - PM_{10} emission inventories

PM_{10} emissions originated from SNAP07 activities

While analysing particle emission factors, it has been studied that they change greatly according to season and location, as well as ambient concentrations. Ketzel et al., (2007) found that in places where conditions are strongly subject to seasonal variations (such as studded tyres, sand or salt usage) a time varying emission model for PM_{10} is necessary. For countries where conditions are milder, the use of a constant emission factor is reasonable. This premise might highlight the need of having specific emission factors that reflect the Spanish case in a more reliable way.

One problem that needs to be successfully addressed is the misconsideration of some important PM_{10} emission activities in the *CAM* inventory. Emissive activities such as the wear of brakes and clutches are usually within the coarse fraction (Garg et al., 2000; EMEP/CORINAIR, 2007) and are also a function of vehicle direct emissions (Wåhlin et al., 2006).

On the other hand, particles coming from road abrasion, tyre wear and road dust re - suspension are found in both, the fine and coarse fraction and they are not necessarily correlated with direct exhaust emissions. This source is also a function of a series of other variables and external factors such as tyre type, vehicle - induced turbulence, road conditions, weather and meteorology, use of studded/friction tyres, wetness of roads, temperature, salting and sanding, presence of side - strips, among others. The estimation of this factors is challenging and not at all straightforward (Gustafsson et al., 2005; Kupiainen et al., 2005; Johansson et al., 2006).

A study conducted by the United States Environmental Agency (*USEPA*) in 1999 applied to Europe, showed that road dust is by far the most importance source of particle pollution in the atmosphere. It was also manifested in this study that such emissions are unlikely to be confirmed by source apportionment analysis or by atmospheric observations. Most of road traffic emitted particles are due to brake and clutch abrasion, street surface abrasion, tyre abrasion and road dust suspension. This work also concluded that free flow traffic accounted for about 40% of total traffic emissions, being brake abrasion the most important particle emission cause (Winiwarter et al., 2010). This fact stresses out the need to importantly consider such emission sources at every inventory.

PM_{10} emissions originated from SNAP08 activities

The most relevant activity in the *SNAP* group is 080600, mobile machinery in agriculture, which comprises more than 50% of the emissions of this category for the entire *GMR*. The emissions reported by the *NAC* inventory are a 17% higher than those of the *CAM* inventory, basically because the former

includes a higher gas oil consumption. It is important to note that, according to section 4.5.5, both inventories incorporate the same methodology using the same assumptions as starting point. This is indeed true since according to table 4.14 emissions are almost equivalent.

The fact that the *CAM* inventory readily applies the methodology designed by the *MARM* for the *NAC* inventory might strongly hint that there is not enough information at local level to compile an inventory, at least for the Autonomous Community of Madrid (*CAM*).

Since there is a large number of off road engines and applications that effectively include it, it is difficult to quantify reasonable emission factors. Moreover, there are usually no registration databases for most off - road engines, thus making it difficult to estimate engine populations and age. Moreover, loading depends on the specific applications for each engine which vary greatly. The way in which inventories usually address this issue is through manufacturer sales surveys and surveys of users of these engines.

The quantification of emissions has been carried out for both inventories under a top - down approach that is based on fuel consumption. For this approach, emission factors are usually normalised to fuel consumption and engine activity is estimated through the total amount the engine consumes at a given operation scheme or cycle. Emission factors for engines from construction, agriculture, mining and forestry applications are obtained from engine manufactures which is not correct in some cases, because engines do not work at steady - state conditions (Kean et al., 2000).

PM_{10} emissions originated from SNAP10 activities

The causes for differences within this category are basically related with the considered activity variables and emission factors. Moreover, there are also divergences in criteria while accounting livestock units, especially regarding categories 100507 (laying hens) and 100508 (broilers).

Animal housing has been reputed of being the main particle source in the agricultural sector, having a strong influence in the local air quality. Within the animal housing activities, poultry facilities seem to be the most serious particle emitters and recognised as hazardous workplaces. Other agricultural operations such as ploughing, harrowing, disking and harvesting are deemed of being particle emitting processes. The extent to which particles are likely of being emitted by these operations is also related to the dryness of the cultivated soil (Winiwarter et al., 2010).

The determination of particle emissions for *SNAP10* typically includes emission factors referred to the number of farm animals. Yet, it has been observed that the determination of emissions improves when the emission factors are modeled trough production cycles of various types of livestock. Seedorf (2004) found out that relating the emission factors to the animal weight rather than the number of livestock heads is much more advantageous for scaling up and down with varying weights of animals of the same species. This approach might result in emission factors that represent median values per livestock class (Seedorf, 2004). This procedure may also include operations that contribute greatly to the emission inventories such as manure management.

In general, emissions related to land management need to be improved deeply, since the European methodology is reputed of carrying large uncertainties. It has also been found that there is a large departure between measured atmospheric PM_{10} levels and those modelled, meaning generally that emissions might be heavily underestimated (Erisman et al., 2008; Misselbrook et al., 2011).

4.5.7. Detailed analysis for particulate matter emissions - $PM_{2,5}$

To better analyse this *SNAP* group, analysis will be carried out on station 5 - Plaza Dr. Marañón (PMAR) which is classified as a badly correlated station according to what has been exposed in section 4.4.3. This station is located in the heart of the city of Madrid, at one of the most important crossroads of the urban network and thus classified as *traffic type* (table 3.1). The activities that have been accounted for the cell where this station is located are presented in table 4.16. The most striking difference that is extracted from table 4.16 is the fact that the emissions computed by the *NAC* inventory are fivefold the emissions considered by the *CAM* counterpart.

Tab. 4.16: SNAP02 activities for station 05 (PMAR). Emissions (E) in $[ton \cdot day^{-1}]$

Code	Description	$\%_{SNAP02,NAC}$	E_{NAC}	$\%_{SNAP02,CAM}$	E_{CAM}
020103	Combustion plants $< 50MWh$	15%	0,03	5%	0,001
020202	Combustion plants $< 50MWh$	85%	0,20	95%	0,01
–	**Total**	100%	0,23	100%	0,01

Category 020103 corresponds to commercial and institutional combustion equipments and category 020202 to residential boilers. Table 4.16 shows the percentages of emissions coming from each of the aforementioned combustion plants. Carrying out the same analysis over stations 24 - Urbanización Embajada (UEMB) and 34 - Collado Villalba (COLL), both inventories report *exactly* the same percentage for each of those two categories.

In order to analyse the issues mentioned above, station 34 - Collado Villalba (COLL) was chosen, which according to section 4.4.3 was classified as a well correlated station. This station lies outside the urban centre of Madrid and would be typically useful for analysing off - road emissions, although it has been labelled as *traffic type* according to table 3.1. Table 4.17 shows the emissions apportionment for $PM_{2,5}$ from *SNAP08* most emissive categories.

Tab. 4.17: SNAP08 activities for station 34 (COLL). Emissions (E) in $[ton \cdot day^{-1}]$

Code	Description	$\%_{SNAP08,NAC}$	E_{NAC}	$\%_{SNAP08,CAM}$	E_{CAM}
080203	Railways. Locomotives	5%	0,0004	90%	0,001
080800	Industry vehicles	91%	0,006	10%	0,0001
–	**Total**	100%	0,007	100%	0,001

From the detailed analysis of the emission inventory for *SNAP08*, there are sound differences between emission inventories. Considering the gross totals, the *NAC* inventory quantifies roughly seven times the total emissions considered by the *CAM* inventory. The first inventory gives a very high percentage to industry vehicles (080800), while the second assigns to railways (080203) a main contribution to the total emissions for *SNAP08*. From figure 4.32, station 34 (COLL) shows the soundest understimation tendency of the entire *GMR*.

This might strongly suggest the fact that several emission sources are not considered, basically because the *NAC* inventory computes bigger emissions than the *CAM* inventory and thus, underestimates

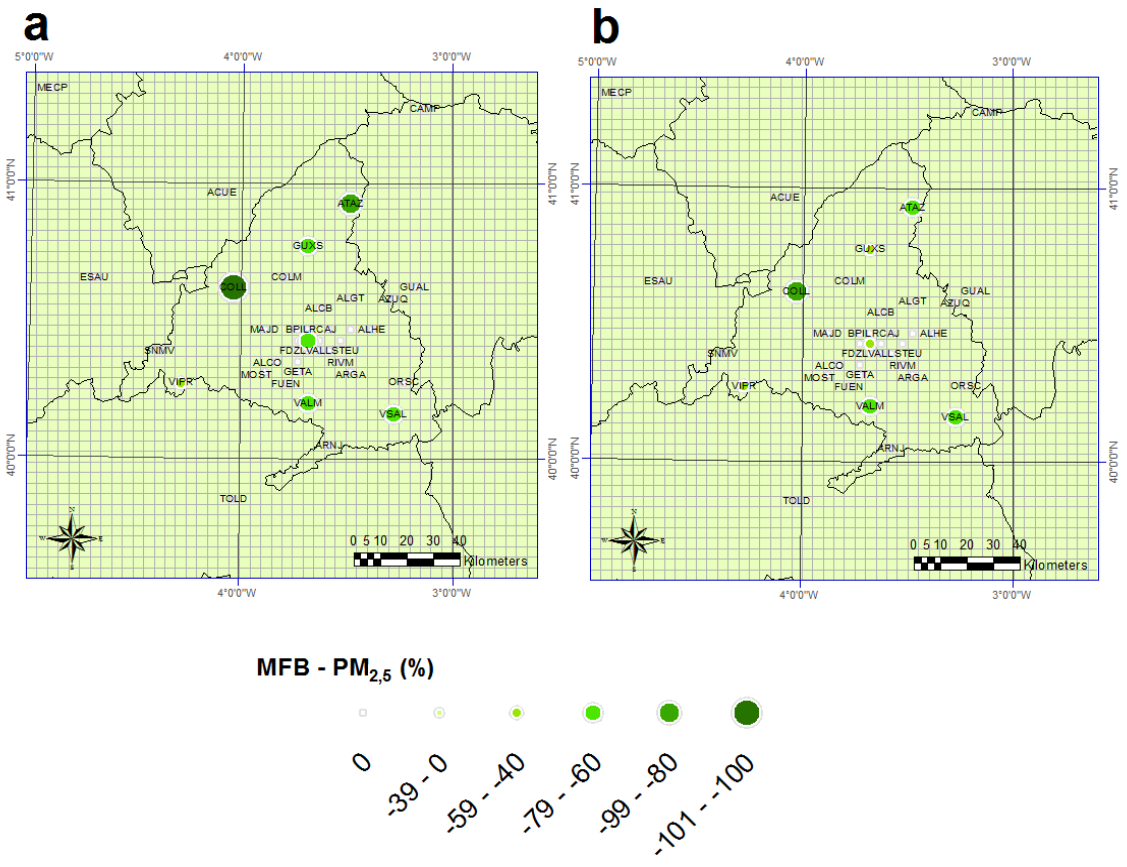

Fig. 4.32: Mean Fractional Bias (*MFB*) values for $PM_{2,5}$ from a) *CAM* and b) *NAC* inventories

less. What seems more dramatic is that, according to figure 4.32, *every* station in the *GMR* domain has a marked tendency towards *underestimation*; this fact might only be an indicator that the inventory compilation criteria are not at all efficient for $PM_{2,5}$.

4.5.8. Detailed analysis for $PM_{2,5}$ emission inventorying

$PM_{2,5}$ emissions originated from SNAP02 activities

The differences related with this category seem to originate at combustion plants from the residential sector (020202) and the stationary engines in agriculture activities (020304). The most viable interpretation for such differences follows the consideration of fuels and emission factors such as in section 4.5.2. In general, the *NAC* inventory is 727% higher than the *CAM* inventory at combustion plants at from residential sector activity (020202).

The $PM_{2,5}$ emissions are directly related with the consumption of wood, other forms of biomass and coals with high emission factors. The *NAC* inventory presents, thus, higher particle emissions through considering higher biomass and coal consumptions. For the stationary engines in agriculture (020304),

the *NAC* inventory is 99% lower than the *CAM* inventory estimate, again, due to the great discordance in the basic fuel mix criteria.

$PM_{2,5}$ emissions originated from SNAP08 activities

The main categories that have been assessed for this *SNAP* group are related with railroad transportation. The Spanish railways are administered entirely by *RENFE*, which provided the necessary information for the compilation of the inventory. The activity variable is the gas oil - diesel consumption in every case. The most important subcategory to consider in this case is *SNAP* 080203 which describes locomotives under transport situations, as well as the emissions generated from their boilers.

Since the activity variable that has been considered corresponds to the fuel consumption, generalised diesel propulsion has been assumed for the locomotive fleet. The consumption apportionment has been made considering line displacements (travellers or goods) through the incorporation of socioeconomic factors, provided by a relevant entity (*RENFE*). The territorial disaggregating process has been made according to the European *NUTS-3* territorial classification, using as information the publication titled "RENFE on a hundred maps", published by this entity. From this atlas, the longitudinal distribution index for each railroad has been deduced. This index has been used to further weigh train traffic intensity over the corresponding network portions, applied also to emissions.

For particles, the document titled "Development of a Database System for the Calculation of Indicators of Environmental Pressure Caused by Transport and Environment Database System (*TRENDS*). Detailed Report 3: Railway Module" has been adopted as a source for emission factors for diesel powered trains. In this case, the emission factor was referenced to an energy unit (Giga Joules), and thus has been transformed to mass units incorporating a Heating Value. Furthermore, it was assumed that emitted particles would be entirely of $PM_{2,5}$ type.

The main group that has been assessed on the *CAM* inventory on which apparent differences are shown is that of railroad transportation, specifically that of locomotives (080202). The general methodology for the quantification of emissions related to this group is the same than the one applied at the *NAC* inventory on its 2006 version, which is also based on the simplified *CORINAIR* methodology.

The activity variable used for the calculation of emissions is the gas oil consumption on railroad traffic within the Community of Madrid for the year 2007. The gas oil consumptions attributed to locomotives were obtained from the Railway Infrastructure Administration (Administración de Infraestructuras Ferroviarias - *ADIF*). The emission factors were obtained from the 2006 version of the *NAC* inventory.

4.5.9. Discussion - $PM_{2,5}$ emission inventories

$PM_{2,5}$ emissions originated from SNAP02 activities

Characterising domestic heating depends more on having an accurate description of the local environment rather than having realistic or representative emission factors. High variations have been observed caused basically by the lack of knowledge in the type of used fuels, the combustion equipments and the operation conditions (Winiwarter et al., 2010).

As it was already discussed in section 4.5.3, domestic combustion sources are very numerous and diffusely distributed. They are poorly characterised and expected to be different from large - scale combustion sources. For these sources, emissions do not only depend on the feedstock type but also on the way in which combustion is accomplished. Emission factors for these activities must be carefully chosen

and extrapolated (Zhang and Morawska, 2002). Studies conducted by Sandradewi et al., (2008) and Glasius et al., (2006) strongly suggested that the impact of domestic heating in the air quality conditions might be as high as road traffic concerning $PM_{2,5}$; domestic heating is also sensitive to seasonal variations (having high emission rates in winter) and rural entourages, where wood combustion is still an extended energy source.

$PM_{2,5}$ emissions originated from SNAP08 activities

One of the most important issues that appears when examining combustion emissions from trains is the fact that they cannot be considered as a point source or a line source. Recent studies agree that trains are not to be considered exclusively for a given cell, but rather as a fast moving point source (Burchill et al., 2011). Emission factors for particulate matter are very different for railroad diesel engines. For this specific activity group, the analysis carried out in section 4.5.6 is valid too.

4.5.10. Detailed analysis for sulphur dioxide emissions - SO_2

The chosen station for carrying out the analysis is station 4 - Barrio del Pilar (BPIL), which is classified as a badly correlated station according to section 4.4.4. Labelled as *traffic type* (table 3.1), this station is located at an emminently residential area, so it is a good spot for evaluating the effect of *SNAP02* (domestic heating) contributions on total SO_2 emissions. The analysis of the activities that occur at this location are described in table 4.18. The gross emissions reported for this *SNAP* group at this cell reveal that the national inventory has computed almost a double amount of SO_2. Moreover, the percentages of each activity over the gross totals for the *CAM* do not differ at all from those presented in section 4.5.7, yet those reported by the *NAC* inventory have changed.

Tab. 4.18: SNAP02 activities for station 04 (BPIL). Emissions (E) in $[ton \cdot day^{-1}]$

Code	**Description**	$\%_{SNAP02,NAC}$	E_{NAC}	$\%_{SNAP02,CAM}$	E_{CAM}
020103	Combustion plants $< 50MWh$	51%	0,187	5%	0,008
020202	Combustion plants $< 50MWh$	49%	0,178	95%	0,160
–	**Total**	100%	0,365	100%	0,168

The station upon which the activity analysis was carried out was station 31 - Arganda del Rey (ARGA). This station is classified as *industrial type* and therefore ideal to assess the degree on which industrial activity (*SNAP03*) actually contributes to the emissions. This station was observed to be badly correlated for SO_2 according to what has been shown in section 4.4.4. The details about the activities that occur at cell - level are listed in table 4.19.

Perhaps the most evident difference between inventories is the amount of activities that each one of them considers. While the *NAC* inventory considers a balanced contribution of different industrial activities, the *CAM* inventory considers lime processes (030312) as virtually the only contributor to the emissions. The gross totals suggest that the compilation criteria for the same location are completely different; the *CAM* inventory has computed 9 times the emissions accounted by the *NAC* inventory. For analysing road traffic SO_2 emissions more deeply, station 36 - Fuenlabrada (FUEN) was chosen. This station has shown a good correlation degree according to the statistical analysis carried out and is labelled as *industrial type* (table 3.1). The activities that occur at this cell are listed in table 4.20 for *SNAP07*.

Tab. 4.19: SNAP03 activities for station 31 (ARGA). Emissions (E) in $[ton \cdot day^{-1}]$

Code	Description	$\%_{SNAP03,NAC}$	E_{NAC}	$\%_{SNAP03,CAM}$	E_{CAM}
030103	Combustion plants $< 50MWh$	41%	0,110	1%	0,003
030104	Gas turbines	11%	0,029	–	–
030105	Stationary engines	6%	0,015	–	–
030204	Plaster furnaces	6%	0,015	–	–
030312	Lime	34%	0,089	99%	2,460
–	**Total**	100%	0,264	100%	2,463

Tab. 4.20: SNAP07 activities for station 36 (FUEN). Emissions (E) in $[ton \cdot day^{-1}]$

Code	Description	$\%_{SNAP07,NAC}$	E_{NAC}	$\%_{SNAP07,CAM}$	E_{CAM}
070101	Passenger cars - highway driving	15%	0,001	9%	0,005
070103	Passenger cars - urban driving	40%	0,036	32%	0,020
070203	LDV - urban driving	15%	0,001	7%	0,004
070301	HDV - highway driving	10%	0,0009	18%	0,010
070303	HDV - urban driving	15%	0,001	11%	0,007
070503	Motorcycles $> 50cm^3$ - urban dr.	–	–	18%	0,01
–	**Total**	100%	0,009	100%	0,056

Table 4.20 shows a good concordance with figure 4.14, in which the emissions reported by the *CAM* inventory are much greater than the emissions reported by the *NAC* counterpart. For the specific case of station 36 (FUEN), the emissions reported by the *CAM* inventory are sixfold the emissions reported by the *NAC* version, which further stresses the need to hunt for the sources of these departures. Further analysis regarding these differences will be addressed in the following sections.

4.5.11. Detailed analysis for SO_2 emission inventorying

SO_2 emissions originated from SNAP02 activities

The description of *SNAP02* has already been discussed at sections 4.5.1, 4.5.7. However, since the determination of sulphur - related emissions follows a mass balance based upon fuel consumptions, a fuel - analysis allows for discrepancies among inventories. The distribution assigned by the *NAC* inventory has been described in the sections mentioned above. On its behalf, the fuel distribution for the *CAM* inventory includes the following assumptions:

1. *Petrol* and *gasoline* consumptions (95, 97 and 98) have been assigned entirely to the transport sector.
2. *Fuel oil* consumptions were assigned entirely to the industrial sector.
3. *LPG* consumption was apportioned between the industrial, transport, domestic and agricultural sectors.
4. *Gas oil type B* was completely attributed to the agricultural sector.

5. *Gas oil type A* and *kerosene* were entirely assigned to the transport sector.

6. *Gas oil type C* was apportioned between the industrial, domestic and service sectors until reaching the total consumption values.

SO_2 emissions originated from SNAP03 activities

There are deep differences throughout the compilation process for this *SNAP* group for the *NAC* inventory, which basically rely on the detail degree of the specific information that was used for obtaining the activity variables. In mostly every case, this activity variable was related with socioeconomic data that were provided by different industrial sectors. The basic socioeconomic datum has been the fuel consumption (in Giga Joules), which is particularly difficult to obtain basically due to discrepancies between the different statistical sources, which can be highly magnified when data for the specific facilities are known. Such "plant - level" information has been obtained in most cases through an individualised questionnaire when facilities were located within a big point source. Industrial facilities that incorporate processes such as steel, car assembly, pulp and paper and aluminium were thoroughly censed and further spatially disaggregated through the *NUTS* territorial classification.

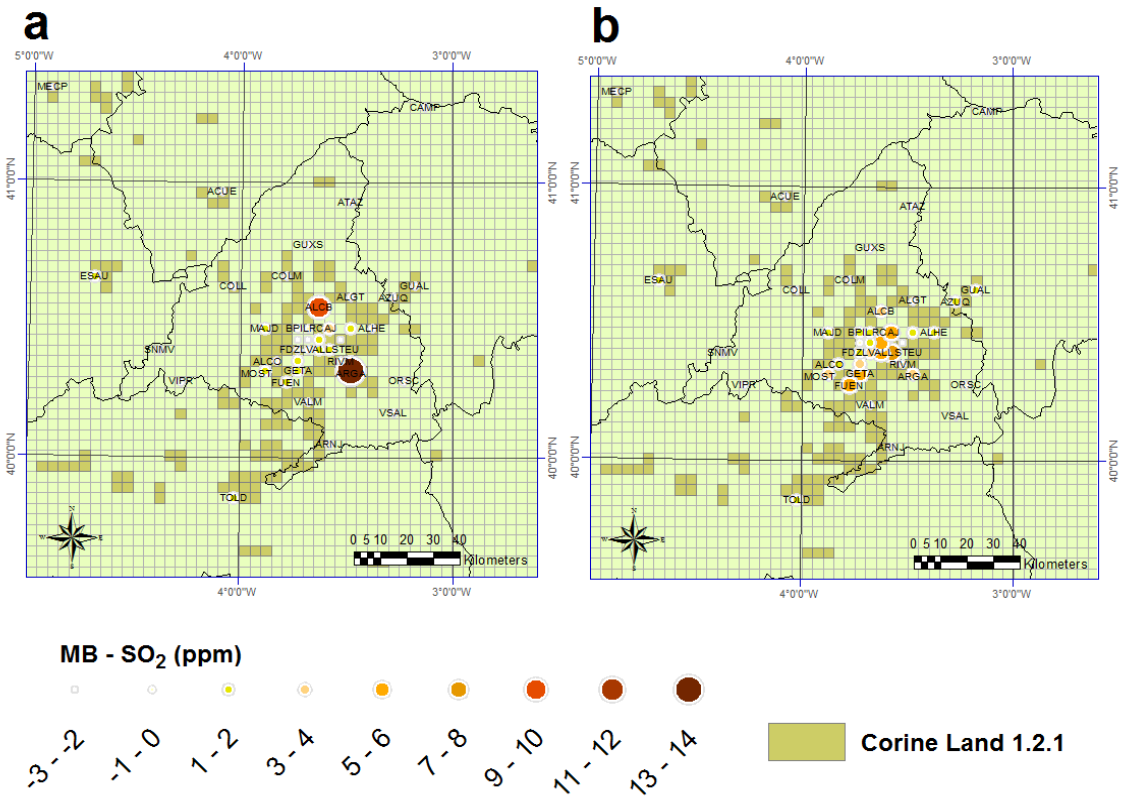

Fig. 4.33: Mean Bias (*MB*) values for SO_2 from a) *CAM* and b) *NAC* inventories and *CLC* 1.2.1

For the iron and steel - related activities, information provided by the Union of Steel Companies (Unión de Empresas Siderúrgicas *UNESID*) has been applied, regarding fuel consumptions and product sales. The non - metallic mineral sector, which involves brick and tile manufacturing as well as ceramic materials, has disaggregated the information on a nation - wide basis through provincial consumptions of such products. The non - ferrous metal sector has used provincial outputs, while the pulp and paper sector has applied production shares for every province. The rest of the sectors, due to the absence of a better information, have been described using information from the document titled "The National Income of Spain and its Provincial Apportionment", published by the *BBVA* Foundation; this involved a sector distribution according to the gross added value for each product and industrial sector.

Combustion in Boilers, Gas Turbines and Stationary Engines.

The *NAC* inventory considers within this group, three activities to be described: combustion plants < 50 *MW* (030101), gas turbines (030102) and stationary engines (030103). These categories are commonly described as part of a broader group called non - specific combustion, which is defined as any type of combustion that is not specific of any industrial activity. For these specific activities, there is great difficulty in obtaining individualised data, since there are many operating facilities and in most cases, a direct knowledge of the concrete characteristics of these processes is difficult (used fuels, ignition conditions, incorporated technologies, etc.).

In order to solve this issue, great efforts have been invested in a crossing procedure between fuel consumptions and different *SNAP* categories, as already described in section 4.5.1. This has been accomplished using a series of fuel consumption balances from the *EUROSTAT* Energy Balance Sheets as well as the *OECD* document "Energy Statistics of *OECD* Countries". The National inventory has considered a wide variety of fuels for these activities, such as coal, black lignite, petroleum coke, wood and woodchips, agricultural waste, fuel oil, gas oil, kerosene, natural gas, *LPG*, coke gas, biogas and manufactured gas.

The characterisation of this activity group is a bit different for the *CAM* inventory. There is great difficulty to actually describe this group due to the great variety of combustion activities and the availability of individual data for each one of the facilities installed within the Greater Madrid Region (*GMR*). However, a reasonable estimation was made through the Energetic Balance of the Community of Madrid, which through consumptions surpluses, distributed such amounts associated to combustion plants. Even so, the information concerning individual industries came partially from a questionnaire applied to each installation under the National Atmospheric Emissions Register (Registro Nacional de Emisiones a la Atmósfera - *RENADE*) and the Pollution Release and Transfer Register (*PRTR*) frameworks. This survey concluded that for the studied region (*GMR*), there are no devices with a thermal power superior to 50 *MW*.

In order to determine the emissions coming from gas turbines (030104) and stationary motors (030105) respectively, the information obtained by the *RENADE* survey was useful to describe cogeneration facilities within the Community of Madrid. As it has been already stated, the consulted information sources were the General Industry, Energy and Mines Direction of the Autonomous Community of Madrid (Dirección General de Industria, Energía y Minas de la Comunidad de Madrid); the 2007 National Emissions Inventory; the 2007 Energetic Balance of the Community of Madrid (Balance Energético de la Comunidad de Madrid); the General Environmental Direction of the Autonomous Community of Madrid (Dirección General de Medio Ambiente de la Comunidad de Madrid), through the *RENADE* and *PRTR* surveys; and individual and specific questionnaires applied to certain facilities within the Community.

Processes without Contact.

This group of *SNAP* activities involves industrial processes where combustion is carried out at a furnace yet neither the flames nor the flue gases have a direct contact with the treated materials. Within this group, and for the specific case of the *GMR*, the only accounted activity was plaster furnaces (030204).

The basic sources of information for this category were a series of documents published by the Spanish Ministry of Industry, Tourism and Commerce (*MITYC*) titled "Plaster, Lime and Natural Cement Industries" where information related to the energetic inputs in plaster production is available. Moreover, this information was complemented with data provided by the Technical and Corporate Plaster Association (Asociación Técnica y Empresarial del Yeso - *ATEDY*), which facilitated fuel consumptions and outputs. For this *SNAP* activity, the emission factors that were applied were those reported by the *EMEP/CORINAIR* guidebook. For the specific case of *LPG*, generic emission factors have been used since the *CORINAIR* handbook does not include them.

On the other hand, the *CAM* inventory only considered the emissions attributed to plaster furnaces (030201), since it is the only type of activity under this *SNAP* subcategory that actually exists within the Greater Madrid Region (*GMR*). The activity data have been provided to the inventory compilers directly by the plaster factories of the Community through an extensive information request procedure. On the contrary, the emission factors that have been calculated for the plaster furnaces were those reported by the *NAC* on its 2006 version. Surprisingly, it was natural gas the only fuel assumed to be actively used in such processes.

Processes with Contact.

These processes have as main characteristic the direct contact between the combustion flames or the flue gases and the treated materials at the hearth of the furnace. The activities that are accounted according to the *NAC* inventory under this category for the specific case of the *GMR* are the following: steel and iron reheating furnaces (030302); grey iron foundries (030303); secondary aluminium production (030310); lime (030312); asphalt concrete plants (030312); bricks and tiles (030319); ceramic materials (030320); an paper - mill industries (030321).

As it has been already stated, for the steel and iron reheating furnaces category (030302), the used information was treated at a point level (mostly integral, individual steel plants). The information source was the Union of Steel Companies (Unión de Empresas Siderúrgicas *UNESID*). The *EMEP/CORINAIR* handbook was the preferred source of emission factors .

For the secondary aluminium production (030310), the information that was incorporated into the 2007 inventory came originally from the Spanish Association of Aluminium Refiners (Asociación Española de Refinadores de Aluminio - *ASERAL*), as well as from the Spanish Ministry of Industry, Tourism and Commerce (*MITYC*). Most emission factors were considered as generic, since they were absent from the *EMEP/CORINAIR* guidebook except those attributed to fuel oil consumptions.

The lime industrial activity (030312) has been described through the incorporation of information from the National Lime and Derivates Manufacturing Association (Asociación Nacional de Fabricantes de Cales y Derivados de España - *ANCADE*). The information has been treated at an area - level and the emission factors were taken from the *EMEP/CORINAIR* guidebook, where available.

The emissions from the bricks and tiles (030319) industry were characterised through the inclusion of information provided by the Spanish Association of Clay Bricks and Tiles (Asociación Española de Fabricantes de Ladrillos y Tejas de Arcilla Cocida - *HISPALYT*). It has been stated that this specific activity is highly atomised throughout the Spanish territory, making it difficult to obtain enough data with a certain degree of reliability. Likewise, the information concerning the ceramic materials industry (030320) has been facilitated by the Spanish Association of Ceramic Tile Manufacturers (Asociacin Española de Fabricantes de Azulejos, Pavimentos y Baldosas Cerámicas - *ASCER*). The emission factors were obtained entirely from the *EMEP/CORINAIR* guidebook.

Finally, for paper - mill industries (030321) the information was provided by the Association of Paper Manufacturers (*ASPAPEL*) through a "Statistical Report", while the socioeconomic datum that has been incorporated was that of fuel consumptions in energy units (Giga Joules). As with the before mentioned categories, the emission factors were taken from the *EMEP/CORINAIR* guidebook.

The *CAM* inventory on its behalf characterises this subgroup based upon other assumptions. Concerning the grey iron foundries (030303), there have been identified only two facilities within the Greater Madrid Region (*GMR*). To describe these facilities, emission factors from the *NAC* inventory on its 2006 version were obtained, except for particles, for which emission factors proposed by *CEPMEIP* (Coordinated European Programme on Particulate Matter Emission Inventories) were implemented.

In the case of secondary aluminium production (030310), information provided directly by the manufacturers was included into the inventory and used for activity description, yet the emission factors that were included were those obtained from the *NAC* inventory on its 2006 version. Exactly the same happened concerning lime production processes (030312): private information was provided and the emission factors from the *NAC* inventory were drawn.

For bricks and tiles manufacturing (030319), the information describing the activity variables was obtained in the first place from companies and facilities within the territory, under the frameworks of the *RENADE* and *PRTR* surveys; emphasis was made on outputs and fuel consumptions. Emission factors were drawn from the *NAC* inventory of 2006. The production of ceramic materials (030320) that occurs in the *GMR* is basically porcelain stoneware and tiling; hence, outputs are expressed as produced square meters of tiling. Again, the variable activity was properly described through the information obtained from the *PRTR* surveys and handed to the General Environmental Direction of the Community. Finally, for paper - mill drying processes (030321), the activity variables were elucidated through fuel consumptions reported to the Community of Madrid. Only one paper mill has already completed the *RENADE* survey, stating fuel oil and biomass consumption as fuels.

SO_2 emissions originated from SNAP07 activities

The general procedure for estimating road traffic emissions (*SNAP07*) has been detailed in section 4.5.2. The emission computations of sulphur dioxide (SO_2) depend basically on the composition and the amount of fuel used in every trip, as well as on the circulation conditions of every vehicle. According to the *COPERT* methodology, it is considered that the total fraction of sulphur is being emitted to the atmosphere in the form of SO_2, assumption which is not always true. SO_2 emissions are quantified thus, through a mass balance between sulphur and oxygen, incorporating also an additional term influenced by the travelled distance for each vehicle class.

4.5.12. Discussion - SO_2 emission inventories

SO_2 emissions originated from SNAP02 activities

For SO_2 emissions, the discussion regarding fuel considerations has been already developed in sections 4.5.1, 4.5.7. Reddy and Venkataraman (2002) have reported that the use of average fuel - based emission factors for sulphur dioxide may be somewhat misleading, since relevant information about production and pollution control technologies might be omitted. In the absence of technology related information, assumptions have to be made either to enhance or to worsen emission factors through the use of arbitrary values.

SO_2 emissions originated from SNAP03 activities

The most significant activities within this group are the use of industrial combustion boilers, stationary engines and gas turbines (030301) and combustion processes related with cement (030311) and lime (030312). The first group is indeed complex due to the high number of sectors that incorporate such processes (iron and steel, paper and pulp industries, non - metallic minerals, etc.). The emission factors depend on the fuels and the installation, therefore the emissions would also depend on the correct adjudication of such variables. The *CAM* inventory assigned consumptions within this sector for natural gas, gas oil, fuel oil, *LPG* and coal; the *NAC* inventory developed higher fuel consumptions considering petroleum coke, wood, agricultural refuse, gas oil, fuel oil, natural gas, *LPG*, biogas and refinery gas, as well as differentiating sectors and activities. According to Vestreng et al., (2007), the key sources for SO_2 are the sectors "combustion in energy and transformation industries", "non-industrial combustion plants", "combustion in manufacturing industries and production processes".

The *NAC* inventory handles emissions at a point - source level to a certain extent. To disaggregate such information, production percentages, consumptions, sales rates and economic indicators are applied. In the case of the cement and lime industries, the emission inventories exhibit differences in the estimation of the activity variable rather than in the final emission values. As with the former category, the differences between inventories are clearly due to the use of sulphur - containing fuels, as well as their consumption rates.

Since the *NAC* has the *provincial* level as the maximum data resolution, the emission processing has been carried out through a *surrogation*, as described in Borge et al., (2008) applied over the *industrial and commercial* land uses specified by the *CORINE* land cover database. The *CORINE* land cover (*CLC*) are data derived from satellite images for the period of the 1990's and 2000 ($\pm$ one year) that provide information about land cover changes for a substantial part of Europe. Availability of these data can contribute to new approaches for the assessment of the European landscape, for instance in the context of environmental and economic accounting, diversity, modelling of its properties, etc. These possibilities are given by the fact that land cover reflects the biophysical state of the real landscape (Feranec et al., 2010).

The information obtained from *EMEP/CORINAIR* as well as its methodology usually involves the disaggregation of emissions through the use of surrogates in order to obtain a spatially resolved emission inventory, especially if these emissions are to be used as input for air quality models situated in Europe. A spatial surrogate or substitute variable is a number between zero and one that specifies the fraction of the emissions that occur in a particular location or grid cell of the model domain. Normally, any type of geographic feature is used to weigh emissions into grid cells more specifically than a uniform distribution (Maes et al., 2009). The before mentioned fact is evident also in figure 4.33, in which the *MB* values are presented for every monitoring location in the *GMR*. The cells coloured in crimson represent cells where the *CLC* 1.2.1 (*industrial and commercial*) category is present. This figure strongly

suggests that the sound overestimations carried out by the *NAC* inventory are caused by the surrogation process through the usage of *CLC* criteria; overestimations being represented as high - positive *MB* values (USEPA, 2007). On the other hand, the pattern of *CLC* 1.2.1 land use cells shown in figure 4.33 shows great similarities with the high *SNAP03* contribution cells at the map represented in figure 4.13 for the *NAC* inventory.

Maes et al., (2009) conclusively observed that data uncertainty decreased when using coarser spatial grids and that disaggregation through surrogates works relatively fine for resolutions higher than 5 *km*. Hence, the use of surrogate disaggregation is not recommendable for finer resolutions, encouraging the use of local or regional emission inventories instead, when available. Typically, SO_2 is not a good pollutant to be surrogated since it has been observed that few large point sources dominate the spatial emission pattern for such pollutant.

In general, there is common agreement that the accuracy of emission determination increases with the level of reporting and resources available for in - country quality control (Vestreng et al., 2007). The latter fact emphasises the need of conducting surveys and questionnaires such as *PRTR* or *RENADE* which have been applied to industrial facilities at the *CAM* inventory. To this respect, the use of plant specific consumptions and regional averages for general sources tend to result in better SO_2 estimations than an overall emission factor for a given category (Reddy and Venkataraman, 2002).

SO_2 emissions originated from SNAP07 activities

The differences in emissions are noticeable between inventories. The *NAC* inventory yielded a SO_2 271 *ton* emission for 2007, while the *CAM* inventory gave a 2.876 *ton* emission. According to the latter, the incorporated emission factors were those of the *NAC* inventory for 2004. Being this so, the sulphur content in fuels would be much higher in 2004 than in 2007, basically because the legal limit imposed in 2005 was 50 *ppm* for gas oil; in 2004, the sulphur content for this fuel rounded 350 *ppm*.

The case of the *CAM* inventory is a typical situation in which the obtained predicted concentrations are directly proportional to the emission factors used. This fact stresses dramatically the need of carrying out and assessment of the emission factors achieved either by direct comparison of the real measured emissions and the calculated data from the models or by selecting the emission factors that yield reasonable concentrations, once used in the dispersion models. Additionally, emission factors should reflect any changes and must be constantly updated to keep up with any modifications (Marmur and Mamane, 2003). Further afield, the consideration of a high SO_2 emissive *SNAP07* category is in clear contradiction with the current efforts in the European Union to restrict the emissions of this pollutant (Vestreng et al., 2007).

4.6. General Summary

In the sections already described, a detailed analysis of the emission inventories has been carried out. In this section, a general summary will be conducted, so that through generalisation, differences between inventories would be more easily identified and improvement proposals further formulated.

4.6.1. Data and information scale

The scale of the information and data upon which an emission inventory is compiled is of paramount importance, since it will determine its resolution and reach. Moreover, it is closely related to *top - down* approaches, where emissions are likely to be surrogated. This issue is particullarly evident for the *NAC*

inventory, which has implemented a series of statistics and databanks reported mostly for *Spain*; these data are likely to be organised in provincial schemes, without any finer resolution. On its behalf, the *CAM* inventory is also affected by this issue since some of the inventory compilation criteria it has incorporated derive from those formulated by the *NAC* inventory in the firstplace. The information *scale* factor is typically important for identifying *point sources*, such as activities classified under *SNAP03* or *SNAP01*. For the above mentioned cases, every situation in which local information has competed against surrogates, better results were obtained for *bottom - up* approaches.

In line with what has been affirmed in section 4.6.1, surrogation is one of the main sources of discrepancies between inventories and of deviations from real concentrations. Surrogation is a common practice while compiling emission inventories under *top - down* approaches, and relies in fragmenting a large domain into smaller and almost equal sub - domains. An underlying assumption associated to this process is that none of these smaller domains has a significant attribute that differentiates them from the rest. This extrapolation did not seem to work adequately for SO_2 under *SNAP03* category for the *NAC* inventory, and although it had a somewhat solid theoretical background (the *CORINE* land use criteria), it led to greatly weigh industrial activity at the *GMR*. As a consequence, some cells accounted emissions coming from activities that do not *exist* in reality.

Another example of a surrogation error is the way in which *COPERT III* was run at the *CAM* inventory for characterising *SNAP07*. For this inventory, *COPERT III* was not run at road - level (for every street), yet it was run at a general level for the entire Autonomous Community of Madrid (*CAM*) and then surrogated according to the type of road and vehicle fluxes. This fact is the cause of the very high contributions registered for highway driving patterns for almost every pollutant. The same issue is the origin of the huge weight motorcycles have at the *CAM* inventory.

One issue that is worth a special mention is the *fuel apportionment criteria* incorporated into the inventories. Virtually each one of the pollutants studied in this work is related to *combustion* activities, which consume fuel. Since it is ultimately complex to have a precise census of the exact fuel types used in every combustion installation within a given domain, a series of assumptions should be made almost always to distribute fuels among sectors or activities. The *NAC* inventory has considered a constant fuel - mix, which represents Spain's fuel consumptions on a general basis. Moreover, the composition of this fuel - mix has been characterised by gathering data from different information sources, most of them prepared and reported for international clusters such as *EUROSTAT* or *OECD*. Keeping this heterogeneous fuel composition (which includes fuels such as agricultural waste or fuel oil) for *every* activity might lead to unrealistic descriptions, some of them as absurd as affirming that *domestic boilers are fueled with petroleum coke*. To this respect, the *CAM* inventory has made a much more reasonable set of assumptions by apportioning some fuels completely to certain sectors (section 4.5.11). Although these criteria might be exclusive, they at least assure that no inappropriate fuel assignations are made.

4.6.2. Quality and validity of emission factors

It has been observed that most of the emission factors drawn by *both* inventories to describe different activities come from the *EMEP/CORINAIR* handbook. Other emission factors, such as those used to compute sulphur dioxide (SO_2) emissions have been obtained through mass balances and taking emission limits as restrictions. To this respect, the *CAM* inventory has overlooked the past application of a composition specification for sulphur containing fuels for vehicles, hence resulting in high SO_2 emissions throughout the domain for *SNAP07*.

4.6.3. Omissions

There is general consensus in saying that the more extensive and detailed an emission inventory is, the better correlation with reality it will attain. Although omitting a certain number of emissive activities is not abnormal during the compilation process, there are some activities that should not be excluded if the inventory wants to reach a certain degree of accuracy. To this respect, the *CAM* inventory has omitted brake and tyre wear and road abrasion as potential PM_{10} emissive activities, which is conceptually wrong considering what has been discussed in section 4.5.6. An analogous situation has been observed for PM_{10} computed for *SNAP10*. The *CAM* greatly misses activities that are reputed for being highly emissive, such as manure management. Both, simplicity and easiness in computation procedures might have been reason enough for omitting such activities.

4.6.4. Information formats

Compiling emission inventories is an information - intensive activity. Therefore, the available information needed to compile them must be easy to process and as straightforward as possible. Most of the information regarding *SNAP08* and *SNAP10* according to the *NAC* inventory has been obtained through the processing of a series of atlases and maps that have been published by different government institutions. For example, to characterise $PM_{2,5}$ emissions for *SNAP08*, the *NAC* inventory incorporated information from an atlas provided by *RENFE*; PM_{10} for *SNAP10* were estimated through information from the Agricultural Atlas published by the Ministry of the Environment (*MARM*).

4.7. Other uncertainties

The scheme upon which this methodology is based departs from the basic assumption that *observations* are *true values*, that have been determined *impartially* and under the *best* experimental conditions. Although *observations* are indeed and independent data set useful for comparisons, the *quality* of such data has been subject of extensive studies. The most challenging issue pertaining *observations* is the fact that monitoring stations are highly dominated by the *local* scale.

While comparisons between observations and model output are useful for carrying out an assessment of model performance, they exhibit sizeable limitations. Model metrics and statistic indicators can only be quantified for grid cells in which monitoring stations are available. This fact means that such metrics might only reflect performance in areas that actually have monitoring clusters, which are usually urban areas and regions where problems occur. Hanna (2007) defines the *nonrepresentativeness* of the observations as one of the most concerning obstacles while trying to assess the general skill of an air quality model. This *nonrepresentativeness* means that the observation is not entirely representative of the area where the instrument is located; even further, the observation point is not representative of the grid - volume average that is actually being simulated (Hanna, 2007; Swall and Folley, 2009; Denby et al., 2010).

It is therefore unrealistic to assume that measured values at a monitoring location are indeed representative of the cell average. This means that even if the model has a perfect performance with almost no observational error, it is impossible to actually obtain paired modelled and observed values on a one - to - one line (Swall and Foley, 2009).

Moreover, the air transport and transformation processes that occur at local scale are somewhat difficult to describe. Pollution levels are sensitively affected in streets with a disturbed or obstructed atmospheric flow, sometimes by more than one order of magnitude compared to free spaces. If buildings

are considered within the parametrisation of roughness, significant errors might be present while describing the source - receptor relationships (Moussiopoulos, 1999).

Because of the complex interactions between natural wind and the number of obstacles present in cities, differences within concentrations over relatively short distances might be observed. Usually monitoring stations are located close to the emission sources, where large concentration gradients occur; this fact usually accounts for stations not being representative.

Artificial obstacles such as buildings that impede free wind flow patterns are also responsible for most of the stagnant conditions within urban areas. The well known street canyon condition is usually found in densely built entourages. At the bottom of this street canyon, vehicles emit in the form of buoyant plumes, dispersing the emitted gases afterwards because of motion. Local pollutant concentrations and wind patterns are influenced substantially by such buoyancy effects caused by the high temperature of the car exhaust gases, solar radiation, soil and window reflection, among other factors (Berkowicz et al., 1996; Moussiopoulos, 1999).

The European legislation specifies that stations should be located in critical points, where high concentrations happen to be and where sizeable population groups are exposed. They should also avoid sampling microenvironments in their immediate emplacement zone. Likewise, traffic monitoring sites should seek to be representative of a surrounding area of around 200 m^2 (Vardoulakis et al., 2005).

Finally, the mathematical modelling is also a limitation that up to now still exerts a certain influence. It is therefore necessary to implement more physically realistic and computationally efficient algorithms. The monitoring instruments that actually carry out the measurements can also be improved, increasing accuracy. Yet, the stochastic fluctuations that are a natural characteristic of the atmosphere are impossible to be eliminated (Vardoulakis et al., 2002).

5. GENERAL DIAGNOSES ON THE EMISSION INVENTORIES

5.1. Emission inventories strengths and weaknessses

5.1.1. National Inventory

Strengths

The strongest point that the NAC inventory exhibited is a *more accurate* description of *SNAP07*. The computed emissions for this *SNAP* group yielded better results for road - traffic, simultaneously considering more emissive activities, such as wear and abrasion processes for particles. These facts make the NAC inventory more realistic and accurate. A favourable asset this inventory included is the fact that the road - traffic model, *COPERT III* was run for every road existing in the *GMR*. The used emission factors were also up - to - date with the legal limits and emissions, hence reflecting a much more realistic situation.

The quantification of *SNAP10* was better described by this inventory too, since it incorporated information gathered by the Ministry of Agriculture which is accurate despite being available for a national scale. This inventory considers more activities for this specific group which led to *less* underestimations, specifically for particles. *SNAP08* was also efficiently described, especially for particles although the information sources were not at all direct and needed further processing.

Weaknesses

The NAC inventory showed sound weaknesses at the rest of the *SNAP* categories. The consideration of a *generic* fuel - mix for Spain, applied exactly in the same way to every combustion activity regardless of its specific features or the conditions under which it is conducted. This resulted in unaccurate descriptions of the emissions that actually occur at a given cell, which sometimes did not correspond to real emission patterns. Another strong weakness this inventory exhibited is the *scale* upon which the information has been compiled, which is national. The maximum resolution this information may have is the *provincial* level, which to describe the *GMR* needs to be subrogated somehow. Specifically for SO_2, this subrogation was carried out through landuse criteria which resulted in describing activities that do not actually occur. Some information sources that have been used are published as atlases and maps, which needed a further dissemination process that might have been subject of inaccuracies and extra assumptions.

5.1.2. Regional Inventory

Strengths

It is precisely at those *SNAP* groups where the NAC missed to accomplish an accurate description that the CAM inventory appeared as strong. The fuel apportionment assumptions that have been made allowed this inventory to better quantify emissions coming from *SNAP02* and *SNAP03*. Moreover for this last *SNAP* group, the consulted information sources are strictly local and concise, usually in the

form of surveys and questionnaires. For some activities *better* emission factors than the ones available from the *CORINAIR* methodology have been incorporated; this is an asset for the *CAM* inventory, which suggests that a certain degree of *quality control* of the implemented emission factors for some categories. The *SNAP08* group for particle emissions is described exaxctly the same as in the *NAC* version by this inventory.

Weaknesses

The biggest failure of this inventory was its poor description of *SNAP07*, by far the activity group that contributes the most to air pollution throughout the *GMR*. The cause of this irregularity was the fact that *COPERT III* has not been run for every road within the studied domain, rather subrogated. This yielded excessive contributions from highway driving situations and motorcycles, which are somehow an exaggeration of the real emission pattern. Another weakness was the misconsideration of sulphur content limits for the calculation of SO_2 emission factors. This yielded extremely high *SNAP07* contributions for SO_2, which were further magnified by the excessive weight that traffic had at the *GMR* for this inventory. The misconsideration of a series of *SNAP10* activities was also a source of sizeable underestimations for this inventory, especially for particulate matter. This fact was the cause of an almost two - magnitude order departure between inventories for this pollutant. Although both inventories incorproated emission factors from the *CORINAIR* methodology, these were more suitable for the *NAC* inventory than for the *CAM* basically because the *CORINAIR* methodology has been designed for national reporting obligations.

6. HARMONISATION REMARKS AND PROPOSALS

6.1. Harmonisation strategies

At this point, a series of strategies are presented to improve the harmonisation degree between the *CAM* and the *NAC* inventories. These strategies could be further extended and applied to several emission inventories that describe the same given domain or emission inventories that describe domains nested within another.

6.1.1. Scale priority

The establishment of a scale priority is a fundamental departure point for choosing the information upon which the emission inventories will be compiled. For describing a given domain, information that is available for the scale of this domain should be chosen over information available for larger or smaller scales. If information from larger scales is used, data could be lost or overlooked during disaggregation procedures; when using smaller scales, the risk of rolling - up datasets that have been obtained under different methodologies might lead to inconsistent descriptions and coverages.

If possible and according to the before mentioned premises, the data selection process should also take into consideration the origin of such information. The selection preference should favour datasets coming in the following order:

1. *Local information.* For point - sources, information based on environmental reports for factories and production facilities should be preferred over all others, provided that such data comply with the rest of the factors mentioned below.

2. *Specialised information.* Information coming from databases that monitor activity variables and emission factors or that convey statistical information should be chosen for non - point - sources or for sources that do not have installation - level data. A beforehand assumption would be that the level of expertise applied to the gathering process of these data should conduct to better quality emission estimations.

3. *Methodologies.* It is at this level that the implementation of general methodologies such as *EMEP* seems adequate. Although these methodologies are very extensive and cover almost every emissive activity desirable of being described, they depart from a general pespective. This perspective is the one that allows an easy - application to almost every European case; however, it is still unclear how much this methodology reflects the concrete cases for every European country or even more, for the local cases within any Member State.

4. *Tailored information.* When data gaps occur or when some information is deemed of not being reliable, some experts tailor information up to the needs of the specific activity to be described. This information has been created exclusively to cover the needs of punctual information lacks and even if complying with the statistical and methodological rigour, its use should be limited.

5. *Generic information.* The concept of generic information is that of any dataset that has been reported without the express intention to be used as part of an emission inventory. If this type of information is being included, it should undergo enough consistency tests.

6.1.2. Subrogation limit

It might happen that while compiling emission inventories, information from a scale bigger than that of the studied domain is only available. Although subrogation is a smart procedure to transfer these large - scale to smaller scales, there exists a limit scale under which the subrogated information is no longer reasonable. This subrogation limits can be determined or assessed following a similar methodology than the one described in section 4.3. The information obtained from such analysis would help to identify at what *scale* are these boundaries located and whether the cells defined by such boundaries are sensitive to resolution changes.

6.1.3. Reliability and accuracy of the information

Given the fact that the emission inventory compiling process needs to be fed with a great deal of data, the quality of this information is crucial to guarantee the representativeness of such an inventory. It is therefore of paramount importance to assure that the information comes from organisations or databases that have consistent and verifiable methodologies for data collection and processing. When possible, the collection of these data should be carried out under normalised conditions. Information must be transparent, accesible and constantly quality - checked. Information must also be actual and valid, preferring newer datasets over older versions.

It is certainly a good practice to compare between similar sets of statistical data reported by one or more institutions for complying with several national and international requirements as to ensure consistency amongst different emission inventories. This might improve the data quality that flows through a series of national statistical offices, ministries and institutes that collect data for reporting obligations. This strategy stresses out the fact that there should be enough discussion regarding *which* variables (emission factors, activities, etc.) should be included in the inventory; incorporating information that has not been checked, accuracy tested or contrasted might produce unrealistic descriptions.

6.1.4. Uniformity and consistency of the information

In line with what has been stated before, there should be a certain degree of similarity among information coming from different data sources. It should be noted that this issue does not encourage to prefer well - established methodologies over punctual information; it is an urge to assure that most of the datasets included in the inventory are *comparable* and *consistent*. Certainly, the usage of a fully - detailed methodological framework for compiling inventories is downright clever to hunt for inconsistencies, gaps, false assumptions, miscalculations and so on.

6.1.5. Information formats

Since emission inventories require considerable loads of data, the need of having easy - processable information is paramount. This means that information should be provided in straightforward formats, which would need a minimal amount of processing time to be successfully included into the

inventory. Information presented in maps, atlases, plots and charts may have enough uncertainties associated especially if the methodology used to produce them is not clearly specified. Data obtained from such information sources might be affected by interpolation, krigging, poor graphic resolution, bad interpretations, etcetera. Ideally data should be presented as *raw* figures; this would dramatically decrease the uncertainty degree the emission inventory attains during its rolling - up process.

6.1.6. Expertise and common sense

Usually when compiling an emission inventory, the compiler comes across a dilemma between describing a given activity with the *best* methodological resources yet reproducing reality inaccurately or even worse, not reproducing it. If this is the case, *common sense* should always prevail. It is useless to consume time and efforts describing activities and behaviours that do not really occur, even if the underlying information has solid methodological backgrounds. The before mentioned premise is a consequence of the very first aim of an emission inventory which is to accurately reflect polluting activities within a given domain. Moreover it highlights the dual nature of inventories, being both scientific and political instruments.

6.1.7. Transparency and peer review

This issue is a call for emission inventories and methodologies to be open to reviewing and crosschecking activities. It is precisely through the conduction of studies and works such as this one that inventories can be evaluated in terms of their goodness and their reliability in the description of reality. To this respect, methodologies should be open, explicit, transparent and detailed. An emission inventory should be subject of constant revisions and a sound crosschecking should be among the required reporting obligations. It is also an urge for discussion and debate, aiming to reach a consensus that would further redound in emission inventories of a superior quality.

7. CONCLUSIONS

The comparison between national and regional scale emission inventories for the Greater Madrid Region (*GMR*) was carried out aiming to understand and clearly identify the reasons for the lack of consistency and to have a better insight of the relevant questions that arise while looking for a common inventorying framework. It has been made evident that both inventories have as starting point the *EMEP/CORINAIR* methodology, yet differ substantially.

According to the general performance of the Air Quality Model, the estimates produced by the Regional Inventory (*CAM*) are better for almost every *SNAP* group except for *road traffic* (*SNAP07*) and *agriculture* (*SNAP10*). At these weak points, the National Inventory (*NAC*) is more accurate. Since *road - traffic* is the *main* polluting activity that occurs in the *GMR*, it is impossible to override completely the National Inventory.

This study has shown that the sources of disagreement between inventories, manifested by the deviations between simulated concentrations and actual observations are due to a series of issues. The **data** and **information scale** upon which each of the inventories has been compiled is different; the National version incorporating data from national and supra - national entities (national ministries, international agencies, etc.), the Regional inventory using information coming from national and regional authorities. In general, it was observed that *local* information sources provide better results. The use of this information redounded in the consideration of different fuel - mixes for both inventories, hence reporting higher or lower emissions for different pollutants incorporating the same *EMEP/CORINAIR* emission factors.

The **scale factor** was further manifested in subrogation practices and their implications on the reproduction of reality for certain zones of the *GMR*. Often it is not possible to use the same references, but the understanding of the methodological differences may facilitate the development of specific procedures to harmonise basic databases, a fundamental requirement to achieve consistency across scales. Other issues such as **omissions** and complicated information **formats** seemed to play a key role in lessening the quality of both inventories.

It is evident that a given **reconciliation** point should be reached between both inventories, combining the best of both worlds. It would be highly recommendable that the Regional Inventory reproduce the methodology followed by the National version to describe road traffic, as it would certainly achieve better results. The bottom-up approach is preferred when there is enough information to support a very detailed emission estimation, but a top-down approach in combination with an updated high - resolution landuse/population cover may provide a more accurate picture of general emission distribution patterns. If basic reference statistics are properly **harmonised**, both approaches should lead to quite similar results, being the differences due to the use of more specific information available only at finer scales.

One of the main conclusions that can be drawn from the present study is that sound research efforts are still to be made, aimed basically at developing more consistent methodologies for the description of urban emission inventories. A general methodology on how to address these issues has been presented

in this study. The outputs from studies like these will also help in the future research needed to provide some guidance on the requirements and methodological framework for emission projections and scenario definition procedures, needed to apply regulations.

Still a considerable amount of effort should be made to reconcile scales and resolutions for emission inventories. Up to now, this task is still largely conditioned by the fact that emission inventories are not intended for scientific use, rather to comply with emission reporting and vigilance obligations. However, the fact of having a **real** and **accurate** emission inventory would result in *better*, *cheaper* and *more assertive* monitoring and control policies whose final aim is assure a good environmental quality for life and health.

About the author.

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8. NOMENCLATURE

- A ⟶ Activity variable.
- C ⟶ X - cell coordinate. Spanish *columna*.
- c ⟶ Number of calms.
- $CRMSE$ ⟶ Centred root mean square error.
- d ⟶ Mean cell size.
- E ⟶ Absolute emission.
- E_F ⟶ Emission factor.
- E_j ⟶ Absolute emission at cell j.
- $E_{SNAP_i,j,k}$ ⟶ Absolute emission of *SNAP* group i at cell j and for activity k.
- F ⟶ Y - cell coordinate. Spanish *fila*.
- f_i ⟶ Weighing factor for neighbouring cells along i direction.
- $\overline{M}$ ⟶ Mean modelled concentration.
- MB ⟶ Mean bias.
- ME ⟶ Mean error.
- MEF ⟶ Model efficiency.
- MFB ⟶ Mean fractional bias.
- M_i ⟶ Modeled concentration for hour i.
- N ⟶ Total number of hours or frequencies.
- N_i ⟶ Frequency for wind direction i.
- n_i ⟶ Number of considered cells along wind direction i.
- $N_{i,j}$ ⟶ Frequency for wind direction i and wind speed j.
- $\overline{O}$ ⟶ Mean observed concentration.
- O_i ⟶ Observed concentration for hour i.
- $P_{SNAP_i,j}$ ⟶ Relative emission of *SNAP* group i at cell j.
- r ⟶ Pearson's correlation coefficient.
- $RMSE$ ⟶ Root mean square error.

- α ⟶ Lambert conformal conic projection standard parallel 1.
- β ⟶ Lambert conformal conic projection standard parallel 2.
- γ ⟶ Lambert conformal conic projection central meridian.
- σ ⟶ Mean standard deviation.
- σ_M ⟶ Mean standard deviation of the modelled data.
- σ_O ⟶ Mean standard deviation of the observations.

- $\%_{SNAP_i,j}$ ⟶ Activity percentage over the given *SNAP* category.

- *AQM* ⟶ Air Quality Modelling system.
- *CAFE* ⟶ Clean Air for Europe Programme.
- *CAM* ⟶ Autonomous Community of Madrid.
- *CAM* ⟶ Inventory of the Autonomous Community of Madrid.
- *CMAQ* ⟶ Community Multiscale Air Quality Model.
- *CLC* ⟶ *CORINE* land cover.
- *COPERT* ⟶ Computer programme to calculate emissions from road traffic.
- *CORINAIR* ⟶ Coordination of Information on the Environment - Air.
- *CORINE* ⟶ Coordination of Information on the Environment.
- `CorrCoef` ⟶ Pearson's correlation coefficient.
- *EEA* ⟶ European Environment Agency.
- *EMEP* ⟶ European Monitoring and Evaluation Program.
- *FAIRMODE* ⟶ Forum for Air Quality Modelling in Europe.
- *GIS* ⟶ Geographic Information System.
- *GMR* ⟶ Greater Madrid Region.
- HDV ⟶ Heavy duty vehicles.
- *IIASA* ⟶ International Institute for Applied Systems Analysis.
- LDV ⟶ Light duty vehicles.
- *LPG* ⟶ Liquefied Petroleum Gas.
- *MARM* ⟶ Spanish Ministry for the Environment.
- *MF* ⟶ Spanish Ministry of Development.
- *NAC* ⟶ National Emissions Inventory.
- *NMVOC* ⟶ Non - Methane Volatile Organic Compounds.
- *NUTS* ⟶ Common Nomenclature for Statistic Territorial Units.
- *OECD* ⟶ Organisation for Economic Cooperation and Devlopment.
- PM_x ⟶ Particulate Matter.
- *PRTR* ⟶ Pollutants Release and Transfer Register.
- *RAINS* ⟶ Regional Air Pollution Information and Simulation.
- *RENADE* ⟶ Atmospheric Emissions National Register.
- `SigmaM` ⟶ Mean standard deviation of the modelled data.
- `SigmaO` ⟶ Mean standard deviation of the observations.
- *SMOKE* ⟶ Sparse Matrix Operator Kernel Emissions.
- *SNAP* ⟶ Selected Nomenclature for Air Pollution.
- *USEPA* ⟶ United States Environmental Protection Agency.
- *VOC* ⟶ Volatile Organic Compounds.
- *WRF* ⟶ Weather Research and Forecasting.

- CH_4 ⟶ Methane.
- CO ⟶ Carbon monoxide.
- CO_2 ⟶ Carbon dioxide.
- NH_3 ⟶ Ammonia.
- NO ⟶ Nitric oxide.
- NO_2 ⟶ Nitrogen dioxide.
- NO_x ⟶ Nitrogen oxides.
- $PM_{2,5}$ ⟶ Particulate matter with $\emptyset \leq 2{,}5\ \mu m$.
- PM_{10} ⟶ Particulate matter with $\emptyset \leq 10\ \mu m$.
- PM_x ⟶ Particulate matter.
- SO_2 ⟶ Sulphur dioxide.

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BIBLIOGRAPHY

[1] Alcorn, J., Lloyd, S. 2009. From the Trenches: Top - down and Bottom - up *GHG* Inventory Approaches. In: *FES* - East Conference, Bethesda, MD, USA. June 17th, Available online at: `http://www.ndcee.ctc.com/technologies/ESOH/Alcorn_FES-East_GHG_Trends_Final.pdf`. Last visit: April 28^{th}, 2011.

[2] Alonso, M.F., Longo, K.M., Freitas, S.R., Mello da Fonseca, R., Marécal, V., Pirre, M., Gallardo Klenner, L. 2010. An urban emissions inventory for South America and its application in numerical modelling of atmospheric chemical composition at local and regional scales. Atmospheric Environment 44. 5072 - 5083.

[3] Annoni, A., Bernard, L., Lillethun, A., Ihde, J., Gallego, J., Rives, M., Sommer, E., Poelman, H. Condé, S., Greaves, M., Uotila, P., Teixeira - Pinto, J., Agria - Torres, J., Lampinen, R., Kayadjanian, M., Schmidt - Seiwert, V., Meiner, A., Luzet, C., Wirthmann, A. 2003. Short Proceedings of the 1^{st} European Workshop on Reference Grids. Joint Research Centre (*JRC*). Ispra, Italy. Available online at: `http://eusoils.jrc.ec.europa.eu/Projects/Alpsis/Docs/ref_grid_sh_proc_draft.pdf`. Last visit: June 18^{th}, 2011.

[4] ASTM D6589. 2010. Norm - Standard Guide for Statistical Evaluation of Atmospheric Dispersion Model Performance.

[5] Atlas Estadístico de la Comunidad de Madrid (*AECM*). 2005. Consejería de Economía e Innovación Tecnológica. Comunidad de Madrid. Madrid, Spain.

[6] Banco de datos municipal y zonal ALMUDENA. 2001. Instituto de Estadística. Dirección General de Economía, Estadística e Innovación Tecnológica. Consejería de Economía y Hacienda. Comunidad de Madrid. Madrid, Spain. Available online at: `http://www.madrid.org/desvan/almudena/almudena.html`. Last visit: June 28^{th}, 2011.

[7] Bellasio, R., Bianconi, R., Corda, G., Cucca, P. 2007. Emission inventory for the road transport sector in Sardinia (Italy). Atmospheric Environment 41, 677 - 691.

[8] Berkowicz, R., Palmgren, F., Hertel, O., Vignati, E. 1996. Using measurements of air pollution in streets for evaluation of urban air quality - meteorological analysis and model calculations. The science of the total Environment 189/190. 259 - 265.

[9] Borge, R., López, J., Lumbreras, J., Narros, A., Rodríguez, M.E. 2010. Influence of boundary conditions on *CMAQ* simulations over the Iberian Peninsula. Atmospheric Environment 44. 2681 - 2695.

[10] Borge, R., Lumbreras, J., De La Paz, D., Rodríguez, M.E. 2009. Air quality modeling: bridging national and continental scales. In: The 18^{th} World *IMACS/MODSIM* Congress, Cairns, Australia. July 13^{th} - 17^{th}, 2009. Available online at: `http://www.mssanz.org.au/modsim09/F10/borge.pdf`. Last visit: March 20^{th}, 2011.

[11] Borge, R., Lumbreras, J., Rodríguez, M.E. 2008. 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 and Software 23. 1026 - 1044.

[12] Borrego, C., Monteiro, A., Ferreira, J., Miranda, A.I., Costa, A.M., Carvalho, A.C., Lopes, M. 2008. Procedures for estimation of modeling uncertainty in air quality assessment, Environment International, 34, 613 - 620.

[13] Borrego, C., Schatzmann, M., Galmarini, S. 2003. Quality assurance of air pollution models. In: Moussiopoulos N (ed.), *SATURN* - Studying air pollution in urban areas - *EUROTRAC-2* subproject final report,

Springer Verlag, Heidelberg, Germany, chapter 7, 155-183. Available online at: http://aix.meng.auth.gr/saturn/finalreport/l-ch7.pdf. Last Visit: March 28th, 2011.

[14] Brulfert, G., Chollet, J.P., Jouve, B., Villard, H. 2005. Atmospheric emission inventory of the Maurienne valley of an atmospheric numerical model. Science of the Total Environment 349. 232 - 248.

[15] Burchill, M.J., Gramotnev, D.K., Gramotnev, G., Davison, B.M., Flegg, M.B. 2011. Monitoring and analysis of combustion aerosol emissions from fast moving diesel trains. Science of the Total Environment 409. 985 - 993.

[16] 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.

[17] 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, *USEPA* National Exposure Research Laboratory, Research Triangle Park, NC. Available online at: http://www.epa.gov/AMD/CMAQ/ch01.pdf. Lastvisit:May10th,2011.

[18] CFHA 2007. Les poussières fines en Suisse. Rapport de la Commission fédérale de l'hygiène de l'air (*CFHA*). Office fédéral de l'environnement, des forêts et du paysage (*OFEFP*, Ed.). Cahier de l'environnement 384. Bern, Switzerland. p.142.

[19] CFHA, 2005. Les polluants atmosphériques azotés en Suisse. Rapport de la Commission fédérale de l'hygiène de l'air (*CFHA*). Office fédéral de l'environnement, des forêts et du paysage (*OFEFP*, Ed.), Cahier de l'environnement 384. Bern, Switzerland. p.174.

[20] Chang J.C., Hanna S.R. 2004. Air quality model performance evaluation, Meteorol. Atmos. Phys., 87, 167-196.

[21] Comunidad Autónoma de Madrid. 2009. Actualización del inventario de emisiones de contaminantes a la atmósfera en la Comunidad de Madrid. Consejería de Medio Ambiente, Vivienda y Ordenación del Territorio. Exp: 34 - D/06. Madrid, Spain.

[22] Cook, R., Touma, J.S., Beidler, A., Strum, M. 2006. Preparing highway emissions inventories for urban scale modeling: A case study in Philadelphia. Transportation Research Part D. 396 - 407.

[23] Council Directive (*EC*) 2008/1/EC of January 15th, 2008 concerning the Integrated Pollution Prevention and Control. L 24/8. Official Journal of the European Union. 29.1.2008. Available online at: http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:024:0008:0029:en:PDF. Last visit: April 12th, 2011.

[24] Council Directive (*EC*) 2008/50/EC of May 21st, 2008 on ambient air quality and cleaner air for Europe. L 152/2. Official Journal of the European Union. 11.6.2008. Available online at: http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:152:0001:0044:EN:PDF. Last visit: April 14th, 2011.

[25] Council Directive (*EC*) 1996/62/EC of September 27th, 1996 on ambient air quality assessment and management. L 296. Official Journal of the European Union. 21.11.1996. Available online at: http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31996L0062:EN:HTML. Last visit: April 15th, 2011.

[26] Cuvelier, C., Thunis, P., Vautard, R., Amann, M., Bessagnet, B., Bedogni, M., Berkowicz, R., Brandt, J., Brocheton, F., Builtjes, P., Carnavale, C., Coppalle, A., Denby, B., Douros, J., Graf, A., Hellmuth, O., Hodzic, A., Honoré, C., Jonson, J., Kerschbaumer, A., de Leeuw, F., Minguzzi, E., Moussiopoulos, N., Pertot, C., Peuch, V.H., Pirovano, G., Rouil, L., Sauter, F., Schaap, M., Stern, R., Tarrason, L., Vignati, E., Volta, M., White, L., Wind, P., Zuber, A. 2007. *CityDelta*: A model intercomparison study to explore the impact of emission reductions in European cities in 2010. Atmospheric Environment 41. 189 - 207.

[27] De Leeuw, F., Berge, E., Grønskei, K., Tombrou, M. 1995. Review on requirements for models and model application, Report of the European Topic Centre on Air Quality to the European Environmental Agency. Copenhagen, Denmark. Available online at: http://www.eea.europa.eu/publications/topic_report_1996_18/at.../file. Last Visit: May 3rd, 2011.

[28] Denby, B., Georgieva, E., Larssen, S., Guerreiro, C., Li, L., Douros, J., Moussiopoulos, N., Fragkou, L., Gauss, M., Olesen, H., Miranda, A.I., Dilara, P., Thunis, P., Lappi, S., Rouïl, L., Lükewille, A., Querol, X., Martin, F., Schaap, M., van den Hout, D., Kobe, A., Silibello, C., Vincent, K., Stedman, J., Goncalves, M., Pirovano, G., Volta, L., van Pul, A., González - Ortiz, A., Roberts, A., Oettl, D., Jericevic, A. 2010. Guidance on the use of models for the European Air Quality Directive. A working document of the Forum for Air Quality Modelling in Europe - *FAIRMODE*. Guidance document for modelling *ETC/ACC* deliverable. October 11^{th}, 2010.

[29] Dommen, J., Prévôt, A.S.H., Baertsch - Ritter, G., Maffeis, G., Longoni, M.G., Grüebler, F.C., Thielmann, A. 2003. High - resolution emission inventory of the Lombardy region: development and comparison with measurements. Atmospheric Environment 37. 4149 - 4161.

[30] Dos Santos - Lucon, O., Moutinho - Dos Santos, E. 2005. The *HORUS* model - inventory of atmospheric pollutant emissions from industrial combustion in São Paulo, Brazil. Environmental Impact Assessment Review. 25, 197 - 214.

[31] Dufour, A. 2006. Simulation et Prévision de la Qualité de l'Air aux Echelles Continentale et Régionale. Doctoral Thesis. University of Toulouse III - Paul Sabatier. Toulouse, France. Available online at: `http://www.cnrm.meteo.fr/spip.php?article500&lang=fr`. Last visit: May 25^{th}, 2011.

[32] Eder, B., Yu, S. 2006. A performance evaluation of the 2004 release of Models-3 *CMAQ*. Atmospheric Environment 40. 4811 - 4824.

[33] Erisman, J.W., Bleeker, A., Hensen, A., Vermeulen, A. 2008. Agricultural air quality in Europe and the future perspectives. Atmospheric Environment 42. 3209 - 3217.

[34] European Monitoring and Evaluation Program - *CORINAIR*. European Environment Agency. 2007. *EMEP/EEA* Inventory Guidebook - 2007. Technical report 16/2007. Available online at: `http://www.eea.europa.eu/publications/EMEPCORINAIR5/Introduction_Aint_2007.pdf`. Last visit: May 4^{th}, 2011.

[35] Fenger, J. 2009. Air pollution in the last 50 years - from local to global. Atmospheric Environment 43. 13 - 22.

[36] Feranec, J., Jaffrain, G., Soukup, T., Hazeu, G. 2010. Determining changes and flows in European landscapes 1990 - 2000 using *CORINE* land cover data. Applied Geography 30. 19 - 35.

[37] Frey, C.H. 2007. Quantification of Uncertainty in Emission Factors and Inventories. 16^{th} Annual International Emission Inventory Conference. Emission Inventories: Integration, Analysis, and Communications. United States Environmental Protection Agency (*USEPA*). Raleigh, NC. May 14 - 17.

[38] Funk, T.H., Chinkin, L.R., Roberts, P.T., Saeger, M., Mulligan, S., Páramo - Figueroa V.H., Yarbrough, J. 2001. Compilation and evaluation of a Paso del Norte emission inventory. The Science of the Total Environment 276. 135 - 151.

[39] Garg, B.D., Cadle, S.H., Mulawa, P.A., Groblicki, P.J. 2000. Brake wear particulate matter emissions. Environmental Science and Technology 34, 4463 - 4469.

[40] Gilliam, R.C., Hogrefe, C., Rao, S.T. 2006. New methods for evaluating meteorological models used in air quality applications. Atmospheric Environment 40. 5073 - 5086.

[41] Glasius, M., Ketzel, M., Wåhlin, P., Jehnsen B., Monster, J., Berkowicz, R., Palmgren, F. 2006. Impact of wood combustion on particle levels in a residential area in Denmark. Atmospheric Environment 40, 7115 - 7124.

[42] Godish, T. 2004. *Air Quality*. 4 ed. Lewis Publishers. Boca Raton FL. United States. Pp. 26 - 27.

[43] Gustafsson, M., Blomqvist, G., Dahl, A., Gudmundsson, A., Ljungman, A., Lindbom, J., Rudell, B., Swietlicki, E. 2005. Inhalable particles from the interaction between tyres, road pavement and friction materials. VTI Publication No. 521. Available online at: `http://www.vti.es/EPiBrowser/Publikationer/English/R521.pdf` Last visit: May 10^{th}, 2011.

[44] Hanna, S.R. 2007. Chapter 4: A review of uncertainty and sensitivity analyses of atmospheric transport and dispersion models. Developments in Environmental Science. Vol. 6. Editors: C. Borrego and E. Renner. Elsevier Ltd. 331 - 352.

[45] Hiraishi, T., Nyenzi, B., Carruthers, I., Gillet, M., Kruger, D., Lopez, C., Meyer, L., Miguez, D., Minxing, W., Nazarov, I., Odingo, R., Pachauri, R., Penman, A., Stone, J. 1996. Revised 1996 *IPCC* Guidelines for National Greenhouse Gas Inventories. Intergovernmental Panel on Climate Change. Available online: `http://www.ipcc-nggip.iges.or.jp/public/gp/english/`. Last Visit: May 5^{th}, 2011.

[46] Institute for the Environment. 2009. *SMOKE* v2.6 User's Manual. University of North Carolina. Chapel Hill, NC. Available online at: `http://www.smoke-model.org/version2.7/html/ch01.html`. Last visit: May 10^{th}, 2011.

[47] Instituto Nacional de Estadstica (*INE*). 2009. Anuario Estadstico de España. Madrid. Spain. Available online at: `http://www.ine.es/prodyser/pubweb/anuarios_mnu.htm`. Last Visit: May 10^{th}, 2011.

[48] Isaksen, I.S.A., Granier, C., Myhre, G., Bernsten, T.K., Dalsøren, S.B., Gauss, M., Klimont, Z., Benestad, R., Bousquet, P., Collins, W., Cox, T., Eyring, V., Fowler, D., Fuzzi, S., Jöckel, P., Laj, P., Lohmann, U., Maione, M., Monks, P.S., Prévôt, A.S.H., Raes, F., Richter, A., Rognerud, B., Schulz, M., Shindell, D., Stevenson, D.S., Storelvmo, T., Wang, W.C., van Weele, M., Wild, M., Wuebbles, D. 2009. Atmospheric composition change: Climate - Chemistry interactions. Atmospheric Environment 43. 5138 - 5192.

[49] Janhäll, S., Jonsson, Å, Molnár, P., Svensson, E.A., Hallquist, M. 2004. Size resolved traffic emission factors of submicrometer particles. Atmospheric Environment 38. 4331-4340.

[50] Janssen, L.H.J.M., Olivier, J.G.J., van Amstel, A.R. 1999. Comparison of CH_4 emission inventory data and emission estimates from atmospheric transport models and concentration measurements. Environmental Science and Policy. 295 - 314.

[51] Johansson, C., Norman, M., Gindhagen, L. 2006. Spatial and temporal variations of particle mass (PM_{10}) and particle number in urban air - implications for health impact assessment. Environmental Monitoring and Assessment 127. 477 - 487.

[52] Jolliff, J.K. Kindle, J.C. Shulman, I, Penta, B., Friedrichs, M.A.M, Helber, R. Arnone, R.A. 2009. Summary diagrams for coupled hydrodynamic-ecosystem model skill assessment. Journal of Marine Systems 76. 64 - 82.

[53] Kean, A.J., Sawyer, R.F., Harley, R.A. 2000. A fuel - based assessment of off-road diesel engine emissions. Journal of Air and Waste Management Association 50. 1929 - 1939.

[54] Ketzel, M., Omstedt, G., Johansson, C., Dring, I., Pohjola, M., Oettl, D., Wåhlin, P., Lohmeyer, A., Haakana, M., Berkowicz, R. 2007. Estimation and Validation of $PM_{2,5}/PM_{10}$ exhaust and non - exhaust emission factors for practical street pollution modelling. Amospheric Environment 41. 9370 - 9385.

[55] Kühlwein, J., Wickert, B., Trukenmüller, A., Theloke, J., Friedrich, R. 2002. Emission - modelling in high spatial and temporal resolution and calculation of pollutant concentrations for comparisons with measured concentrations. Atmospheric Environment 36. Supplement No. 1. S7 - S18.

[56] Kupiainen, K.K., Tervahattu, H., Räisänen, M., Mäkelä, T., Aurela, M., Hillamo, R. 2005. Size and composition of airborne particles from pavement wear, tires and tractor sanding. Environmental Science and Technology. 39, 699 - 706.

[57] Lim, B., Boileau, P., Bonduki, Y., van Amstel, A.R., Janssen, L.H.J.M., Olivier, J.G.J., Kroeze, C. 1999. Improving the quality of national greenhouse gas inventories. Environmental Science and Policy 2. 335 - 346.

[58] Lindley, S.J., Conlan, D.E., Raper, D.W., Watson, A.F.R. 2000. Uncertainties in the compilation of spatially resolved emission inventories - evidence from a comparative study. Atmospheric Environment 34. 375 - 388.

[59] Maes, J., Vliegen, J.,Van de Vel, K., Janssen, S., Deutsch, F., De Ridder, K., Mensink, C. 2009. Spatial surrogates for the disaggregation of *CORINAIR* emission inventories. Atmospheric Environment 43. 1246 - 1254.

[60] Mannschreck, K., Klemp, D., Kley D., Friedrich, R., Kühlwein, J., Wickert, B., Matuska, P., Habram, M., Slemr, F. 2002. Evaluation of an emission inventory by comparisons of modelled and measured emission ratios of individual HCs, CO and NO_x. Atmospheric Environment 36 Supplement No. 1. S81 - S91.

[61] Marmur, A., Mamane, Y. 2003. Comparison and evaluation of several mobile-source and line-source models in Israel. Transportation Research Part D 8. 249 - 265

[62] Mellios, G., van Aalst, R. Samaras, Z. 2006. Validation of road traffic urban emission inventories by means of concentration data measured at air quality monitoring stations in Europe. Atmospheric Environment 40. 7362 - 7377.

[63] Mensink, C., De Ridder, K., Deutsch, F., Lefebre, F., van de Vel, K. 2009. Examples of scale interactions in local, urban and regional air quality modelling. Atmospheric Research 89. 351 - 357.

[64] Mensink, C. 2000. Validation of urban emission inventories. Environmental Monitoring and Assessment 65. 31 - 39.

[65] Ministerio de Medio Ambiente y Medio Rural y Marino. 2009. Inventario Nacional de Emisiones a la Atmósfera 1990 - 2007. Dirección General de Calidad y Evaluación Ambiental. Secretaría de Estado de Cambio Climático. Madrid, Spain.

[66] Misselbrook, T.H., Cape. J.N., Cárdenas, L.M., Chadwick, D.R., Dragosits, U., Hobbs, P.J., Nemitz, E., Reis, S., Skiba, U., Sutton, M.A. 2011. Key unknowns in estimating atmospheric emissions from *UK* land management. Atmospheric Environment 45. 1067 - 1074.

[67] Monks, P.S., Granier, C., Fuzzi, S., Stohl, A., Williams, M.L., Akimoto, H., Amann, M., Baklanov, A., Baltensperger, U., Bey, I., Blake, N., Blake, R.S., Carslaw, K., Cooper, O.R., Dentener, F., Fowler, D., Fragkou, E., Frost, G.J., Generoso, S., Ginoux, P., Grewe, V., Guenther, A., Hansson, H.C., Henne, S., Hjorth, J., Hofzumahaus, A., Huntrieser, H., Isaksen, I.S.A., Jenkin, M.E., Kaiser, J., Kanakidou, M., Klimont, Z., Kulmala, M., Laj, P., Lawrence, M.G., Lee, J.D., Liousse, C., Maione, M., McFiggans, G., Metzger, A., Mieville, A., Moussiopoulos, N., Orlando, J.J., O'Dowd, C.D., Palmer, P.I., Parrish, D.D., Petzold, A., Platt, U., Pöschl, U., Prévôt, A.S.H., Reeves, C.E., Reimann, S., Rudich, Y., Sellegri, K., Steinbrecher, R., Simpson, D., ten Brink, H., Theloke, J., van der Werf, G.R., Vautard, R., Vestreng, V., Vlachokostas, Ch., von Glasgow, R. 2009. Atmospheric composition change - global and regional air quality. Atmospheric Environment 43. 5268 - 5350.

[68] Moussiopoulos, N., Vlachokostas, G., Tsilingiridis, G., Douros, I., Hourdakis, E., Naneris, C., Sidiropoulos, C. 2009. Air quality status in Greater Thessaloniki Area and the emission reductions needed for attaining the EU quality legislation. The Science of the Total Environment 407. 1268 - 1285.

[69] Moussiopoulos, N. 1999. Urban Air Pollution: Current Knowledge and Future Research. Proceedings of *EUROTRAC* Symposium '99. Editors: P.M. Borrell and P. Borrell. Transactions on Ecology and the Environment. Vol. 28. *WIT* Press. 679 - 688.

[70] Ntziachristos, L., Samaras, Z. 2000. *COPERT III* Computer programme to calculate emissions from road transport. European Environment Agency. Copenhagen, Denmark. Available online at: `http://www.eea.europa.eu/publications/Technical_report_No_50`. Last visit: April 10^{th}, 2011.

[71] Olesen, H.R. 2001. Ten years of Harmonisation activities: Past, present and future. In: 7^{th} International conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes. Belgirate. May 2001. Available online at: `http://www.harmo.org/Docs/TenYears.pdf`. Last Visit: April 15^{th}, 2011.

[72] Peters, G. P. 2008. From production-based to consumption - based national emission inventories. Ecological Economics 65. 13 - 23.

[73] Pulles, T. 1999. Can and should European inventories be improved? Proceedings of *EUROTRAC* Symposium '99. Editors: P.M. Borrell and P. Borrell. Transactions on Ecology and the Environment. Vol. 28. *WIT* Press. 9 - 15.

[74] Reddy, M.S., Venkataraman, C. 2002. Inventory of aerosol and sulphur dioxide emissions from India: I - Fossil fuel combustion. Atmospheric Environment 36. 677 - 697.

[75] Reidy, B., Rhim, B., Menzi, H. 2008. A new Swiss inventory of ammonia emissions from agriculture based on a survey on farm and manure management and farm - specific model calculations. Atmospheric Environment 42. 3266 - 3276.

[76] Rypdal, K. 2002. Uncertainties in the Norwegian emission inventories of acidifying pollutants and volatile organic compounds. Environmental Science and Policy 5. 233 - 246.

[77] Rypdal, K., Winiwarter, W. 2001. Uncertainties in greenhouse gas emission inventories - evaluation, comparability and implications. Environmental Science and Policy 4. 107 - 116.

[78] Samaras, Z., Zierock, K.H. 1994. Guidebook on the estimation of the emissions of other mobile sources and machinery. Subparts 'Off-road vehicles and machines', 'railways' and 'inland waterways'. European Environment Agency. Copenhagen, Denmark.

[79] Sandradewi J., Prévôt, A.S.H., Weingartner E., Schmidhauser, R., Gysel, M., Baltensprenger, U. 2008. A study of wood burning and traffic aerosols in an Alpine valley using a multi - wavelength aethalometer. Atmospheric Environment 42, 101 - 112.

[80] Seedorf, J. 2004. An emission inventory of livestock - related bioaerosols for Lower Saxony, Germany. Atmospheric Environment 38. 6565 - 6581.

[81] Seika, M., Metz, N., Harrison, R.M. 1996. Characteristics of urban and state emission inventories - a comparison of examples from Europe and the United States. The Science of the Total Environment 189/190. 221 - 234.

[82] Serveau, L. 2010. Inventaire des Emissions de Polluants Atmospheriques en France. Series Sectorielles et Analyses Entendues. Centre Interprofessionel Technique d'Etudes de la Pollution Atmosphérique (*CITEPA*). Paris, France. Available online at : `http://www.citepa.org/publications/Inventaires.htm`. Last visit: April 28^{th}, 2011.

[83] Skamarock, W.C., Klemp, J.B. 2008. A time-split nonhydrostatic atmospheric model. Journal of Computational Physics 227. 3465 - 3485.

[84] Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Duda, M.G., Huang, X.Y., Wang, W., Powers, J.G. 2008. A Description of the Advanced Research *WRF* Version 3. *NCAR* Technical Note. National Center for Atmospheric Research. Boulder, Colorado, USA. Avaible online at: `http://www.mmm.ucar.edu/wrf/users/docs/arw_v3.pdf`. Last visit: May 10^{th}, 2011.

[85] Spain. Royal Decree 1073/2002, of October 18^{th} on the evaluation and air quality management related to sulphur dioxide, nitrogen dioxide, nitrogen oxides, particles, lead, benzene and carbon monoxide. BOE No. 260. October 30^{th}, 2002. Available online at: `http://www.boe.es/boe/dias/2002/10/30/pdfs/A38020-38033.pdf`. Last visit: April 10^{th}, 2011.

[86] Swall, J.L., Foley, K.M. 2009. The impact of spatial correlation and incommensurability on model evaluation. Atmospheric Environment 43. 1204 - 1217.

[87] Taylor, K. E. 2001. Summarizing multiple aspects of model performance in a single diagram, J. Geophys. Res. 106 (D7). 7183 - 7192.

[88] Thao - Pham, T. B., Manomaiphiboon, K., Vongmahadlek, C. 2008. Development of an inventory and temporal allocation profiles of emissions from power plants and industrial facilities in Thailand. Science of the Total Environment 397. 103 - 118.

[89] Thunis, P., Rouïl, L., Cuvelier, C., Stern, R., Kerschbaumer, A., Bessagnet, B., Schaap, M., Builtjes, P., Tarrason, L., Douros, J., Moussiopoulos, N., Pirovano, G., Bedogni, M. 2007. Analysis of model responses to emission-reduction scenarios within the *CityDelta* project. Atmospheric Environment 41. 208 - 220.

[90] Thunis, P., Georgieva, E., Galmarini, S. 2011. A procedure for air quality models benchmarking. Joint Research Centre (*JRC*). Ispra, Italy.

[91] Tønnesen, D. 2010. Urban Air Pollution 2000-2015. Results from monitoring and modeling in Oslo and Trondheim. Report OR 51/2010. NILU Norsk institut for luftforskning - Norwegian Institute for Air Research. Kjeller, Norway. Available online at: `http://www.nilu.no/index.cfm?ac=publications&folder_id=4309`. Last visit: May 25^{th}, 2011.

[92] Tsilingiridis, G., Zachariadis, T., Samaras, Z. 2002. Spatial and temporal characteristics of air pollutant emissions in Thessaloniki, Greece: investigation of emission abatement measures. The Science of the Total Environment 300. 99 - 113.

[93] United Nations Commission for Europe (*UNECE*). 1979. Convention on Long - Range Transboundary Air Pollution. Geneva, Switzerland. Available online: `http://www.unece.org/env/lrtap/full%20text/1979.CLRTAP.e.pdf`. Last Visit: April 20^{th}, 2011.

[94] United States of America. Clean Air Act. 2008. Chapter 85 - Air Pollution Prevention and Control. Available online at: `http://www.epa.gov/air/caa/`. Last visit: April 20^{th}, 2011.

[95] United States Environmental Protection Agency (*USEPA*). 2007. Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze. Office of Air Quality Planning and Standards. Air Quality Analysis Division. Air Quality Modeling Group. Research Triangle Park, North Carolina. United States.

[96] van de Kassteele, J. 2006. Statistical air quality mapping. Doctoral Thesis. Wageningen University. Wageningen, Netherlands. Available online at: `http://edepot.wur.nl/121778`. Last visit: May 25^{th}, 2011.

[97] Vardoulakis, S., González - Flesca, N., Fisher, B.E.A., Pericleous, K. 2005. Spatial variability of air pollution in the vicinity of a permanent monitoring station in central Paris. Atmospheric Environment 39. 2725 - 2736.

[98] Vardoulakis, S., Fisher, B.E.A., González - Flesca, N., Pericleous, K. 2002. Model sensitivity and uncertainty analysis using roadside air quality measurements. Atmospheric Environment 36. 2121 - 2134.

[99] Vautard, R., Schaap, M., Bergström, R., Bessagnet, B., Brandt, J., Builtjes, P.J.H., Christensen, J.H., Cuvelier, C., Foltescu, V., Graff, A., Kerschbaumer, M., Krol, M., Roberts, P., Rouïl, L., Stern, R., Tarrason, L., Thunis, P., Vignati, E., Wind, P. 2009. Skill and uncertainty of a regional air quality model ensemble. Atmospheric Environment 43. 4822 - 4832.

[100] Vautard, R., Builtjes, P.H.J., Thunis, P., Cuvelier, C., Bedogni, M., Bessagnet, B., Honoré, C., Moussiopoulos, N., Pirovano, G., Schaap, M., Stern, R., Tarrason, L., Wind, P. 2007. Evaluation and intercomparison of Ozone and PM_{10} simulations by several chemistry transport models over four European cities within the *CityDelta* project. Atmospheric Environment 41. 173 - 188.

[101] Vestreng, V., Myhre, G., Fagerli, H., Reis, S., Tarrason, L. 2007. Twenty-five years of continuous sulphur dioxide emission reduction in Europe. Atmospheric. Chemistry and Physics 7, 3663 - 3681.

[102] Vivanco, M.G., Andrade, M.F. 2006. Validation of the emission inventory in the São Paulo Metropolitan Area of Brazil, based on ambient concentrations ratios of CO, $NMOG$ and NO_x and on a photochemical model. Atmospheric Environment 40. 1189 - 1198.

[103] Wåhlin, P., Berkowicz, R., Palmgren, F. 2006. Characterisation of traffic - generated particulate matter in Copenhagen. Atmospheric Environment 40, 2151 - 2159.

[104] Wilkinson, J.G., Loomis, C., Souten, D., Emery, C. 2005. Emissions Modelling in Action: Suggestions to Improve the Process of Developing Emissions Estimates for Use in Air Quality Modelling Studies.14^{th} International Emission Inventory Conference - Transforming Emission Inventories - Meeting Future Challenges Today. Las Vegas, Nevada, April 11 - 14.

[105] Willmott, C.J. 1982. Some comments on the evaluation of model performance. Bulletin of the American Meteorological Society 63.1309 - 13.

[106] Winiwarter, W., Kuhlbusch, T.A.J., Viana, M. Hitzenberger, R. 2010. Quality considerations of European *PM* emission inventories. Atmospheric Environment 43. 3819 - 3828.

[107] Winiwarter, W., Dore, C., Hayman G., Vlachogiannis, D., Gounaris, N., Bartzis, J., Ekstrand, S., Tamponi, M., Maffeis, G. 2003. Methods for comparing gridded inventories of atmospheric emissions - application for Milan province, Italy and the Greater Athens Area, Greece. The Science of the Total Environment 303. 231 - 243.

[108] World Health Organization. 2005. *WHO* Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulphur dioxide. Global update. Summary of risk assessment. Geneva, Switzerland. Available online at: `http://www.who.int/phe/health_topics/outdoorair_aqg/en/index.html`. Last visit: May 4^{th}, 2011.

[109] Zachariadis, T., Samaras, Z. 1997. Comparative assessment of European tools to estimate traffic emissions. International. Journal of Vehicle Design (Special Issue) 18 (3/4), 312 - 325.

[110] Zárate, E., Belalcázar, L.C., Chappier, A., Manzi, V., van der Bergh, H. 2007. Air quality modelling over Bogotá, Colombia: Combined techniques to estimate and evaluate emission inventories. Atmospheric Environment 41. 6302 - 6318.

[111] Zhang, J., Morawska, L. 2002. Combustion sources of particles: 2. Emission factors and measurement methods. Chemosphere 49. 1059 - 1074.