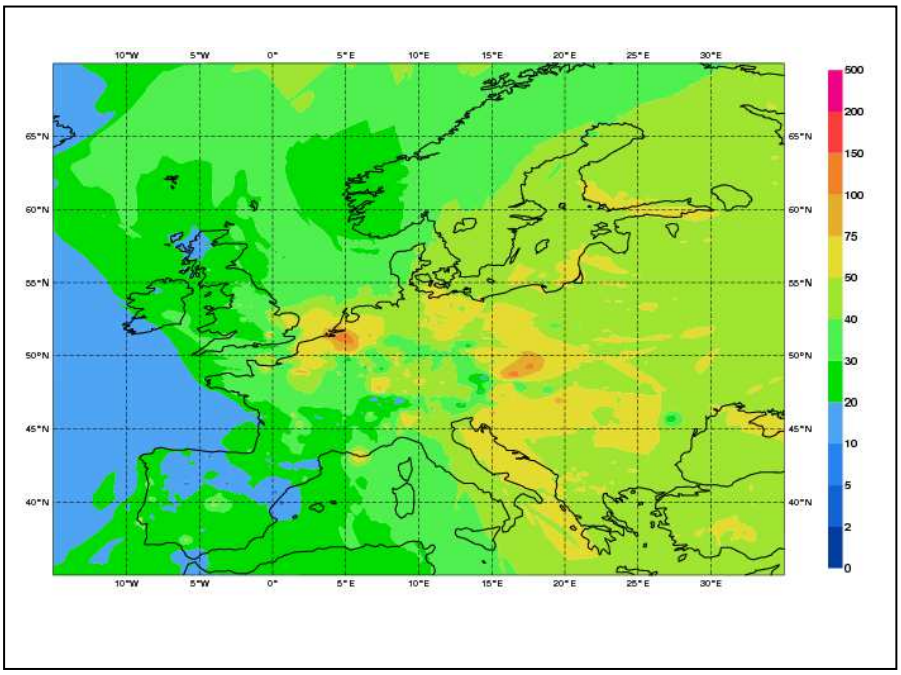
How to start with PM modelling for air quality assessment and planning relevant to the Air Quality Directive



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Laurence Rouïl, Bertrand Bessagnet (Eds.)

FAIRMODE
Forum for air quality modelling in Europe

European Topic Centre
on Air Pollution and
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Front page picture:
A snapshot of PM modelling result

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Executive summary

This document provides a first overview of questions and recommendations for the modelling of ambient particulate matter (PM_{10} and $PM_{2.5}$) concentrations in Europe. It is part of the activities of FAIRMODE (Forum for Air Quality Modelling in Europe; http://fairmode.ew.eea.europa.eu) working group 1 (WG1), which was since the end of 2013 led by the European Environment Agency (EEA). This review document has been developed to help support the needs of the EU Member States in addressing the European Air Quality Directive covering PM pollution (EC, 2008).

In general, the added-value of using models for air quality management can be summarized as follows:

- **Mapping air pollutant concentrations fields and air pollution patterns**: to get an overview of air pollutant concentrations over the whole targeted territory, even where no measurement data exists. Mapping is necessary for improving communication and raising awareness of the general public and competent authorities. It gives the most comprehensive representation of air pollution patterns.
- **Forecasting air pollution levels in a near or remote future**. Modelling is the only way to assess the potential evolution of concentrations in the future according to the variation of factors such as meteorology and emissions. Short term forecasting (commonly up to two days ahead) has the objective to inform the general public (e.g. sensitive population affected by asthma and other respiratory diseases) and authorities (e.g. who may also wish to implement short term action plans in case of pollution episodes). Long term scenario analysis aims at assessing the impact of emission control strategies. Impact assessment studies are necessary for decision making, and more generally urban planning.
- **Understanding the air pollution phenomenology**: analysing the model parameterisations, their sensitivity to changes and the model results in a large number of situations increases the capacity of the expert to interpret air pollution and to identify the most important drivers with regard to various meteorological situations. This is particularly relevant when episodes with persistent exeedances of regulatory threshold values occur: understanding the determining factors (local sources, long range transport, natural events, etc.) helps in targeting the most efficient emission control strategies.

In 2011, a first guide was published within the FAIRMODE WG1 initiative on “Modeling nitrogen dioxide for air quality assessment and planning relevant to the European Air quality Directive” (Denby et al., 2011a). This guide aimed at providing accessible guidance and good practices in the use of modeling tools to simulate nitrogen dioxide (NO_2) concentrations in urban areas.

It was not possible to provide such an operational document for PM issues: considering the high complexity that drives PM processes, in a first step, the scope of this document is to introduce a number of essential concepts regarding PM modeling and its operational use for policy purposes.

The aim of this document is as follows:

- To provide accessible information on the application of air quality models in regard to the European Air Quality Directive, with emphasis on the modelling of PM;
- To review questions and sources of information essential to consider before applying PM models
- To propose ‘good practices’ and keys for use of models for air quality assessment and planning applications, including needs and limitations.

The document is structured as an analysis of concrete issues with operational answers rather than a review of modeling tools.

- It provides an overview of the questions that can be considered according to the state of the art in the field of air quality modeling,
- As a consequence limitations for policy purposes will be identified and discussed
- It provides references and links to more detailed and relevant documents when required.

This document is intended to provide background information for authorities and technical organizations (such as environment agencies and local air quality monitoring networks) carrying out air quality assessment and planning activities in the framework of the European Air Quality Directive.

The document will not cover in detail the model contents, or scientific aspects of the methodologies, but it will review the questions that the policy user will have to consider before starting to define the best modeling approach considering his objectives. It will present data that is essential to gather, to efficiently run models, and concrete examples of successful applications.

Therefore, this document should be accessible to readers with limited experience in the area of air quality modelling, but with some experience in air quality monitoring or management. Its objective is to propose a methodology to assess “fitness for purpose” of modeling approaches to deal with given policy relevant questions over a given domain of interest.

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FAIRMODE
Forum for air quality modelling in Europe

How to start with PM modelling for assessment and planning relevant to the Air Quality Directives

1 Introduction

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1.1 Scope and aims

In 2011, a first guide was published within the FAIRMODE WG1 initiative on "Modeling nitrogen dioxide for air quality assessment and planning relevant to the European Air quality Directive" (Denby et al., 2011a). This guide aimed at providing accessible guidance and good practices in the use of modeling tools to simulate nitrogen dioxide (NO_2) concentrations in urban areas.

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1.2 Audience

This document is intended to provide background information for authorities and technical organizations (such as environment agencies and local air quality monitoring networks) carrying out air quality assessment and planning activities in the framework of the European Air Quality Directive.

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1.3 General overview on air quality modeling issues in the framework of the Air Quality Directive

Air pollution results from the presence of chemical compounds emitted into the air by anthropogenic activities and by natural sources (biogenic emissions from vegetation, soil erosion, sea salts, volcanic activity, and wild-land fires). Dynamic and chemical processes drive changes of the atmospheric composition, especially in the lowest layers of the atmosphere. Therefore, air pollutant concentrations and deposition affect air quality and can directly impact human health and ecosystems.

Models are just a mathematical representation of the real world. Air quality models aim at simulating the concentrations of air pollutants that humans can breathe and that can affect ecosystems and the environment. They should be suited to reproduce emissions, transport, chemistry and deposition of atmospheric chemical compounds. Models can be based on various mathematical approaches:

- Empirical, using suitable parameters from experimental studies, to fit and tune relevant equations describing air pollutant concentrations;
- Statistical, using extensive historical sets of observations to define relationships between concentrations and other relevant parameters;
- Deterministic, using numerical schemes to solve physical-chemical equations discretized on an appropriate grid.

To deal with air quality management issues, model results are supposed to complement measurement data from monitoring networks. Air quality legislation sets the main principles (number of stations and siting, standards in instrumentation, etc.) of the monitoring strategy that should be implemented in the MS depending on the concentration levels of the regulatory air pollutants. Particulate matter issues relate to PM_{10} and $PM_{2.5}$, (with aerodynamic diameter respectively lower than 10 and 2.5 $\mu g/m^3$), the latter being generally named as "fine particles". Monitoring strategy to be implemented in the MS, and limit values as well as quality objectives, are set in the Air Quality Directive (AQD; EC, 2008).

According to the AQD, MS have to divide their territory into a number of air quality (AQ) management zones. This is the basis of regulatory reporting. MS assess whether the air pollutant levels in their zones exceed or do not exceed the limit or target values (for an overview of what has been part of the official reporting for year 2010 by several MS see de

Smet et al., 2013). Monitoring networks provide routine observations at specific locations, using both automatic devices and manual measurements. This is the oldest and the most commonly used (and robust) way for air quality evaluation, but information remains restricted to the location of the observation sites (point measurements) and the time when the measurements are made.

The added-value of using models for AQ management can be summarized as follows:

- **Mapping air pollutant concentrations fields and air pollution patterns**: to get an overview of air pollutant concentrations over the whole targeted territory, even where no measurement data exists. Mapping is necessary for improving communication and raising awareness of the general public and competent authorities. It gives the most comprehensive representation of air pollution patterns.
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- **Understanding the air pollution phenomenology**: analysing the model parameterisations, their sensitivity to changes and the model results in a large number of situations increases the capacity of the expert to interpret air pollution and to identify the most important drivers with regard to various meteorological situations. This is particularly relevant when episodes with persistent exeedances of regulatory threshold values occur: understanding the determining factors (local sources, long range transport, natural events, etc.) helps in targeting the most efficient emission control strategies.

1.4 Fitness for purpose

AQ modelling, used in combination with observations from measurement networks, can be used as a management tool, provided that its performance in simulating air pollution variables is satisfactory, and its uncertainties are well understood and controlled. However, operational implementation of AQ models for decision making support is sometimes challenging because of the sensitivity of the results to input data and to model parameterisations that depend on the targeted spatio-temporal scale, meaning the size and level of detail of the region to be modelled as well as the time resolution required.

Spatio-temporal scale drives the model requirements together with the modelling objectives, and consequently, the choice of the kind of model to be used.

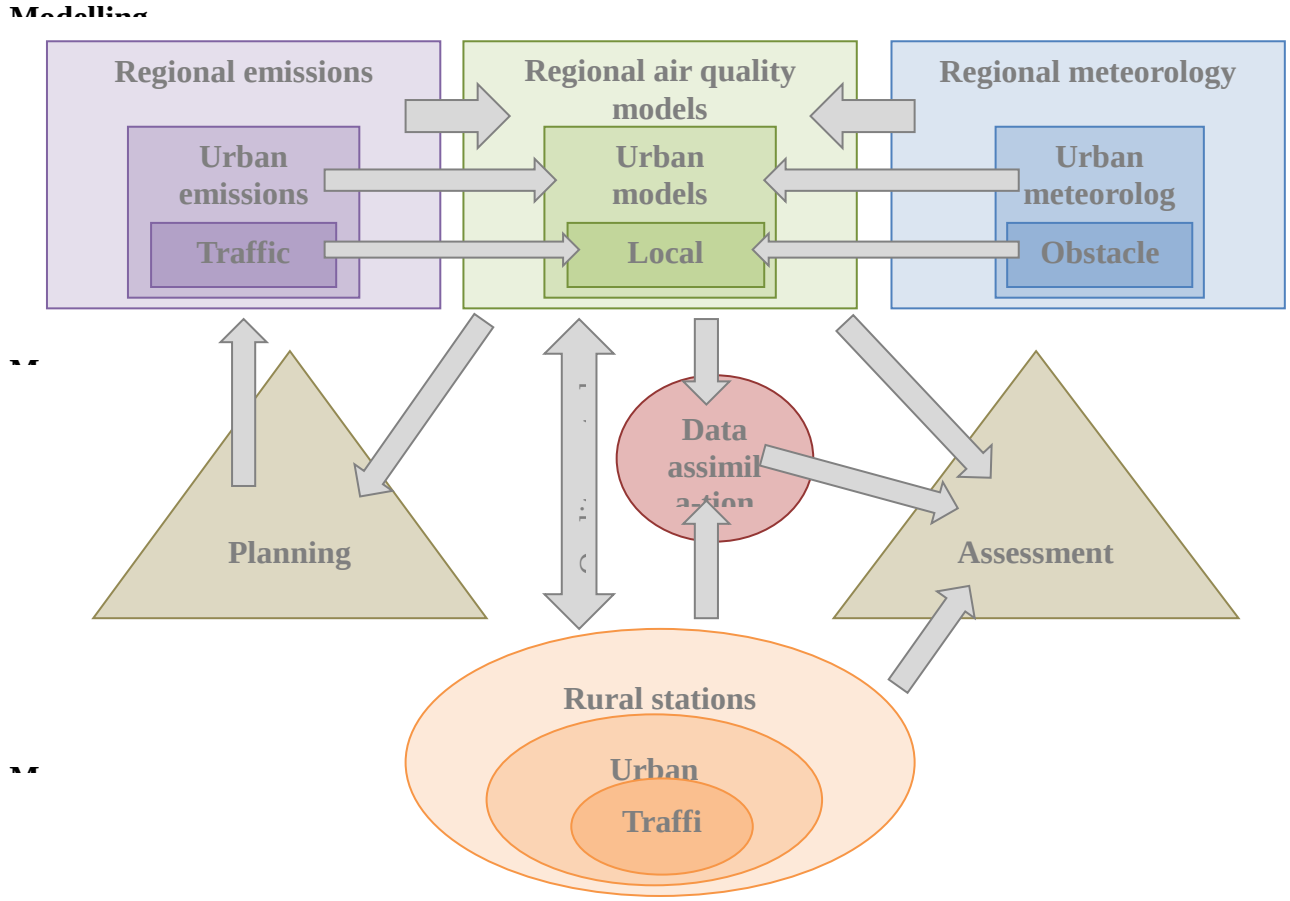

Figure 1: Schematic representation of air quality management and the connections with modelling and monitoring activities. Source: (Denby et al., 2011a)

The concept displayed in Figure 1, and taken from the "NO_2 guide on modelling nitrogen dioxide" (Denby et al. 2011a), separates various spatial scales of AQ modelling and highlights essential inputs and different kinds of applications that relate to them. Air quality is driven by physical-chemical phenomena which develop at various scales, depending on the considered sources, pollutants and their time of residence in the atmosphere, as well as meteorological parameters. Generally, model resolution is determined by the spatial scale, and the following notations are usually stated:

- *Local/hotspot scale* : Resolution: 1 - 100 m^2; domain size: < 1 km^2
- *Urban/agglomerate scale*: Resolution: 1 - 10 km^2; domain size: < 100 km^2
- *Regional scale*: Resolution: 10 - 50 km^2; domain size: national and continental.

Processes that drive pollutant formation and transport should be modelled with the correct spatial resolution for the targeted area. In the AQ modelling field, three kinds of processes are generally considered:

- *Dispersion processes*: transport and dispersion of pollutants driven by meteorological factors;
- *Chemical processes*: transformation and formation of chemical compounds due to pollutants' reactivity in the atmosphere;
- *Deposition processes* (especially for PM): deposition is a sink for air pollution;

Depending on the spatial scale those processes are more or less decisive (local dispersion of pollutant emitted by local sources versus long range transport of chemically reactive species for instance) and should be modelled with appropriate tools.

Moreover, consistency with the targeted scale is required for input data as well. It is important to get consistency between the spatial resolutions of model results and input data, which represent “driving forces”. Input data refers to:

- Emissions: anthropogenic and natural sources of air pollutants including resuspension due to traffic, soil erosion;
- Meteorological parameters: temperature, wind fields and turbulence, precipitation, relative humidity, atmospheric stability, and cloudiness are the main variables that influence atmospheric dispersion and chemistry;
- Boundary and initial conditions: initial state of the model and concentration levels outside the modelling domain (lateral and upper boundaries);
- Land cover: location and description of urban areas, largest roads, natural and agricultural areas, forests.

Beyond the spatio-temporal scale, suitability of air quality models refers also to the applications: assessment and mapping, forecasting and planning are the main ones in relation to requirements set by the AQD. Depending on the application, a type of model will be preferred to another or will be run in certain conditions (for instance coupled with data assimilation techniques for mapping concentration fields).

All these elements are addressed in this guide document.

1.5 Applications of PM modelling for the European Air Quality Directive

1.5.1 PM objectives in the air quality directive

In the European AQD, PM_{10} and $PM_{2.5}$ are considered with various quality objectives:

- For PM_{10}, legally binding limit values are set for both annual and daily averages: the yearly average shall not exceed **40 μg/m^3** and the number of days when the daily average exceeds **50 μg/m^3** must not be more than **35**. These limit values entered into force in January 2005, and the MS could ask to postpone compliance with these limit values until 201 (EC, 2014)1. But non attainment areas still exist in Europe in several countries.

- For $PM_{2.5}$ (see Annex XIV of the AQD: EC, 2008), a target value of **25 μg/m^3** is set for the yearly mean. This target value will, according to the AQD, become an annual limit value by the 1st of January 2015, and should be reduced to **20 μg/m^3** by the 1st of January 2020. Moreover, population exposure is evaluated: an Average Exposure Indicator expressed in μg/m^3 (AEI) shall be based upon measurements at urban background locations in zones and agglomerations and assessed as a three-calendar year running annual mean concentration (AEI for 2010 is based on 2008, 2009 and 2010 measurement). The AQD sets a target reduction for this indicator at the national level that should be respected in 2020. The target depends on the baseline AEI evaluated for 2010. In parallel, an exposure concentration obligation of **20 μg/m^3** is set for 2015.

As for other pollutants, in areas where PM limit values are exceeded and where objectives are not reached, the MS have to establish action plans to ensure compliance (article 23 of the AQD) and short term action plans when the risk of exceedance is high (article 24). Annex XV of the AQD describes the information required in the action plans: description of the episode, origin of the pollution, details of control measures and evaluation of their impact and efficiency to reduce pollution.

1.5.2 PM status in Europe

Thanks to the wealth of data gathered within the COST action 633 on "aerosol phenomenology" (COST, 2008), Putaud et al. (2010) reported a status of particulate matter mean values across Europe. This compilation aims at highlighting similarities and differences in aerosol characteristics, beyond considerations on PM mass concentrations, through a network of 40 sites selected for the study and distributed over 3 sub-regions: North-Western Europe, Southern Europe, Central Europe (Figure 2).

A large range of PM_{10} concentration levels (5–54 µg m^{-3} annual average) was observed through Europe. In each of the North-Western, Southern and Central areas, an increasing gradient in PM_{10} concentration is generally observed when moving from natural–rural background to kerbside sites. Exceptions were, however, observed in all three areas (e.g. lower or similar PM_{10} at urban or kerbside site levels compared to rural sites). As there is no reason why impacts of local to urban sources should be significantly smaller at these specific urban or kerbside sites, Putaud et al. (2010) assumed that these exceptions are due to local influence of regional PM_{10} background concentrations, and may occur everywhere. Annual and 24-h limit values are exceeded in all three areas of Europe, generally (but not solely) at urban and kerbside sites. Urban background PM_{10} annual mean and median values are significantly higher in southern Europe (median = 36 µg m^{-3}) compared to North-western and Central Europe (medians = 24 and 26 µg m^{-3}, respectively).

The range of the gradient in $PM_{2.5}$ concentrations observed across the network (3–35 µg m^{-3} annual average) is similar to that of PM_{10}. An increasing gradient in $PM_{2.5}$ is generally observed when moving from rural to urban background sites in North-western and Southern Europe. In Central Europe, $PM_{2.5}$ can be as large at rural sites as at urban background sites, and concentrations at kerbside sites do generally not appear to be particularly high compared to urban background locations. The 2010 EU target value for the annual $PM_{2.5}$ mean concentration (25 µg m^{-3}) was exceeded most often at urban background sites. The highest $PM_{2.5}$ concentrations at near-urban area, urban background, and kerbside sites are observed in Southern Europe.

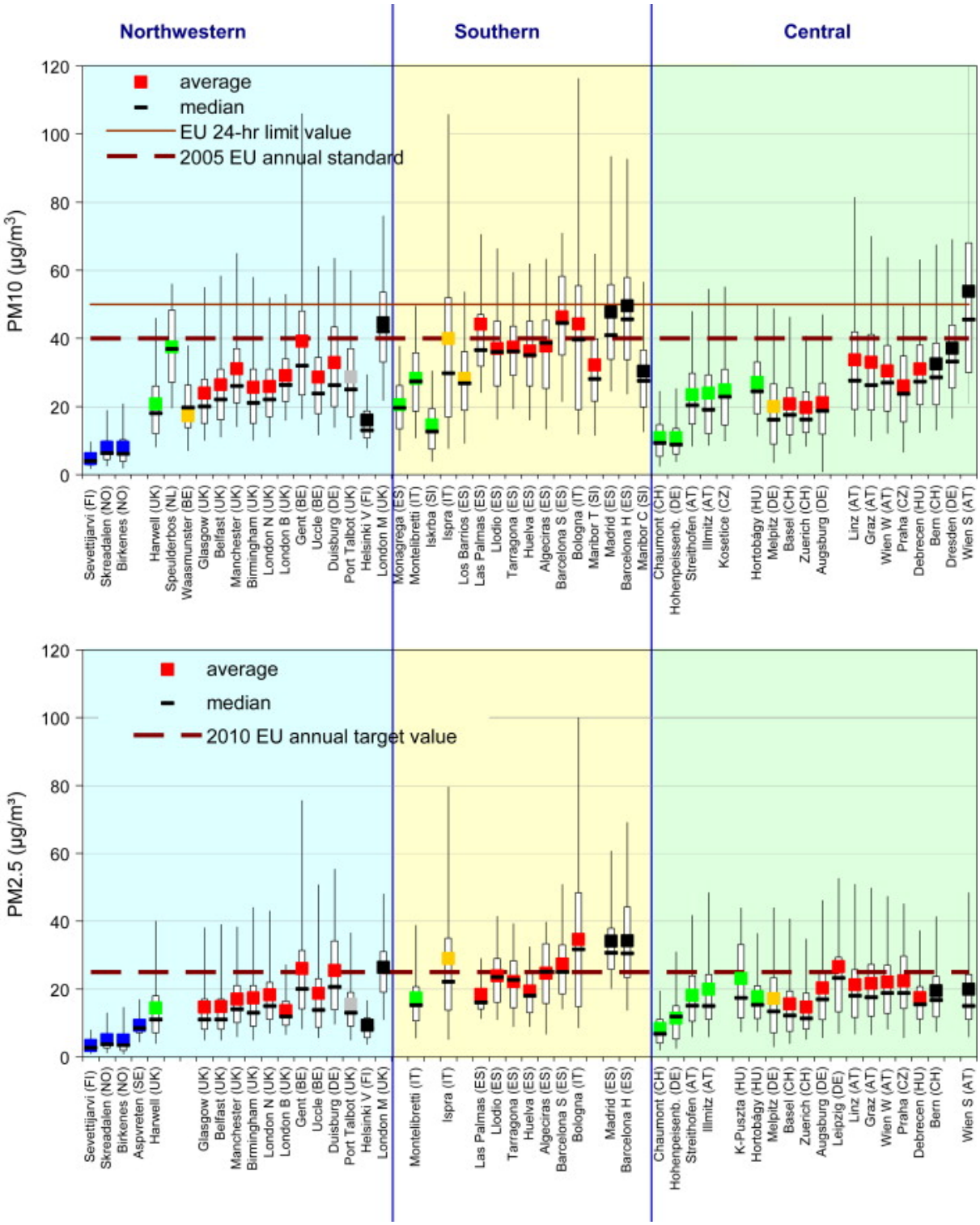

Figure 2: Annual averages of PM10 and PM2.5 mass concentrations, including the 5, 25, 50 (median), 75, and 95% percentiles of their 24-h integrated concentrations. Symbol colors indicate the type of site (blue: natural background, green: rural background, yellow: near-city, red: urban background, grey: industrial, black: kerbside) – Source: Putaud et al. (2010)

Air pollution due to high PM concentrations is considered to have the most adverse health impacts in Europe. Many MS still report PM_{10} exceedances, in particular of the daily limit value in many zones and agglomerations, i.e. they do not comply with the requirements of the AQD. The definition of efficient and relevant action plans to avoid such situations is often a challenge.

One reason is the complexity of chemical formation and transport processes leading to particulate matter pollution in the air.

Some other key observations related to PM are reported in the EEA's annual reports on air quality in Europe (EEA, 2013e):

- The small reductions observed in ambient particulate matter (PM) concentrations over the period 2001–2011 reflect the slowly declining emissions of primary PM and ammonia (NH_3) emissions.
- 33% of the EU urban population lives in areas where the EU air quality 24-hour limit value for PM (PM_{10}) was exceeded in 2011. For EEA-32[1] countries the estimate is 49 %.
- EU urban population exposure to PM_{10} levels exceeding the WHO air quality guidelines significantly higher; it comprises 88 % of the total urban population in 2011.

1.5.3 Applications of PM modeling

Because PM issues, in regard to both EU regulations and health, are far from being solved, modelling approaches provide an essential tool for addressing a number of questions raised by the AQD. The three key advantages of models are that they provide:

- the causal link between emissions, atmospheric processes and concentrations;
- the geographical distribution of concentrations;
- the ability to predict future air pollutant levels.

Therefore, the following applications of PM modelling according to the AQD can be considered:

- **Assessment and reporting[2]:**
  - Complementing fixed measurements in some areas and use for assessment where there is no exceedance.
  - Reporting (mapping) geographical distribution of PM levels and exceedances of limit values and long term objectives;
  - Reporting human exposure to exceedances: to assess and describe the geographical area and the number of inhabitants exposed.
- **Assessment of source contributions (source apportionment)** including long-range transport of PM
  - Understanding episodes genesis and characterizing the conditions (meteorology, emissions, boundary conditions) that favour their occurrence;
  - Contribution of road salting / sanding[3];
  - Contribution of natural PM sources (mainly desert dust, sea salt and wild-land fires)[4];
  - Contribution of "extra-national" (transboundary) PM pollution through long-range transport processes[5].
- **Assessment of the impact of long term action plans[6]**
  - Understanding of past trends of air pollution that allow the assessment of effective action plans in the future;

[1] 32 member countries of the EEA (EU-15 + EU-12 + EFTA-4 and Turkey)
[2] articles 6 and 7 of the AQD 2008/50/EC
[3] article 21 of the AQD 2008/50/EC
[4] article 20 of the AQD 2008/50/EC
[5] article 25 of the AQD 2008/50/EC
[6] articles 23 and 24 of the AQD 2008/50/EC

- Evaluation of the impact of emission control strategies (long term and short term action plans) on ambient PM concentrations.

- **Forecasting** :
  - to enhance public information and awareness[7];
  - to evaluate the relevance of short term action plans that can be decided few days in advance.

In the case of PM_{10}, an interesting feature has been established by scrutinizing the daily and annual mean concentrations measured in Europe: A strong correlation exists between the annual average concentration and the number of days when the 50 µg/m^3 daily limit value is exceeded. Figure 3 shows that given the current daily and annual mean limit values for PM_{10} in the AQD then the daily mean limit value is almost never exceeded if the annual mean limit value is not. This near linear relationship means that it will be sufficient to perform assessment of emission reduction strategies (using modelling) on annual mean PM_{10} concentrations to address the exceedances of daily and yearly limit values.

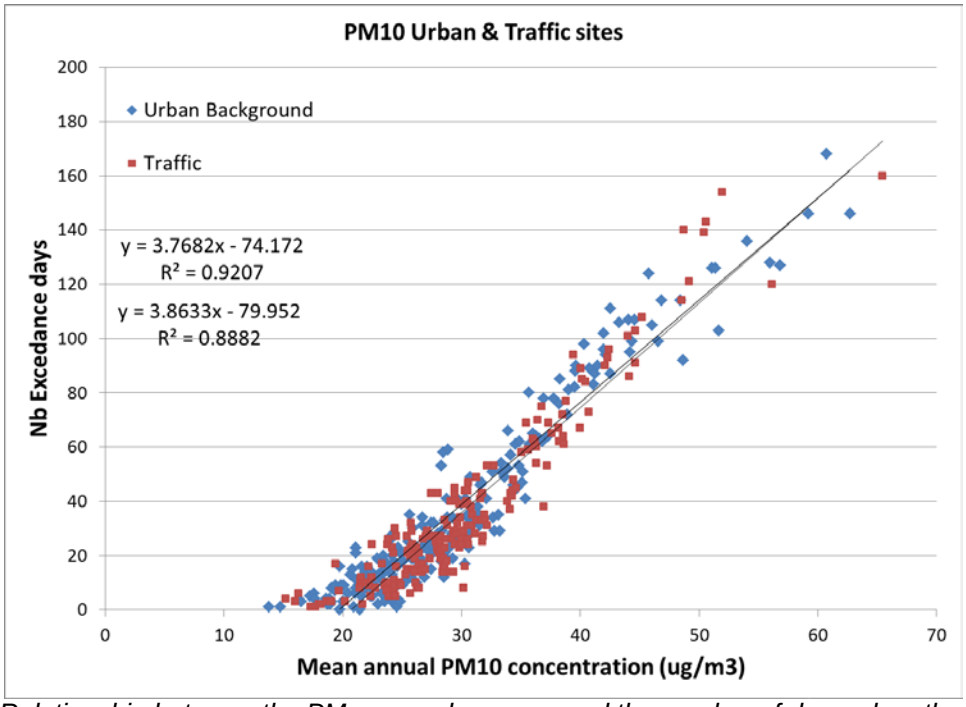

Figure 3: Relationship between the PM_{10} annual average and the number of days when the daily limit value is exceeded. The graph is established on the basis of urban and traffic sites recorded in the AIRBASE database (2009). Source: the EC4MACS project (http://www.ec4macs.eu/)

1.6 Structure of the report

This report is structured in separate parts that focus on the use of numerical models for calculating particulate matter (PM) concentrations in ambient air. It is intended to provide support to authorities and technical organizations which are in charge of air quality assessment or planning activities with special emphasis on PM. A large number of generic points, especially the description of the various AQ model categories, have been extensively covered in the previous guide dedicated to NO_2 modeling. Consequently references to that guide (Denby et al., 2011a) will be preferred to repetition and the present guide will rather provide insights on how to deal with specific PM issues.

[7] Article 26 of the AQD 2008/50/EC

The first part (chapters 2 to 4) is dedicated to the review of three major components of the particulate matter modeling approach:

- Emission data;
- Meteorological data;
- Quality control and evaluation.

The other chapters deal with questions that should be carefully considered by users who wish to run models at various scales and for different policy purposes in an appropriate way. They provide some views on how to interpret the results correctly.

Chapter 6 focuses on the analysis of PM modeling applications that can be applied in relation to requirements under the Air Quality Directives.

2 Main issues related to PM modelling

In this chapter a number of key questions that should be covered when the use of modelling is considered for **PM issues** is reviewed and discussed. It is out of the scope of this document to go deeply into scientific details. The objective is to provide the reader with an overview of the questions national and local experts and decision makers in the MS should be aware of when dealing with the operational use of models, for applications in air quality management, information to the general public, for forecasting, and for trend analyses and scenario studies. It is also out of the scope of the present document to go into detail of generic modelling principles that have already been detailed in (Denby et al., 2011a). In the present guide we focus on questions that are specific to PM. They are linked to the specific nature of this pollutant driven by specific physical-chemical processes.

The list of issues addressed below may not be exhaustive. However, the objective is to give an overview of the most relevant ones.

2.1 What should be modeled from aerosol typology and composition

There are various aerosol types depending on their sources and the geographical scale. It is common to make the distinction between **primary** and **secondary** particulate matter. The former one is directly emitted by sources while the second one is produced in the air via chemical processes (Figure 4). Therefore one could intuitively expect to find primary PM in the vicinity of sources, while secondary PM is more influenced by long range transport. However it is well-known that some primary particles, especially natural ones like desert dusts and sea salt are transported over very large distances.

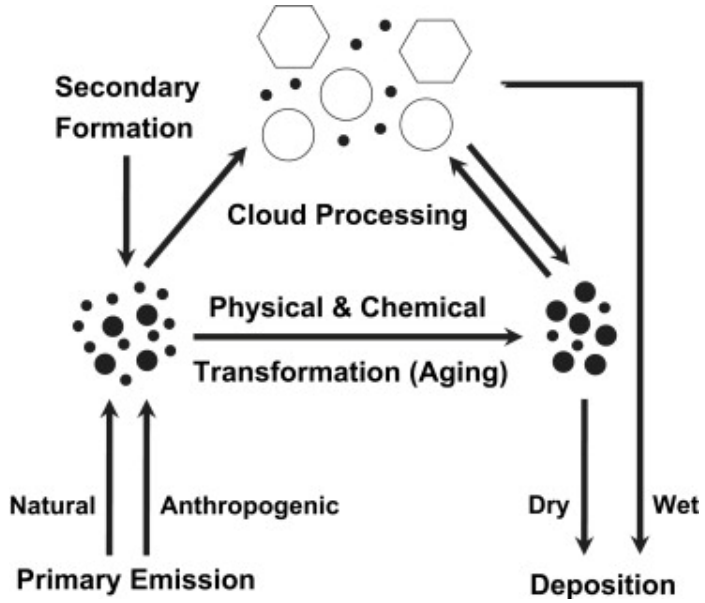

Figure 4: Primary emissions, secondary formation, and atmospheric processing of natural and anthropogenic aerosols (from Fuzzi et al., 2006 and Pöschl, 2005 in Monks et al., 2009).

Therefore, it **is necessary to consider the PM issue at both local and regional scales**. Some episodes are characterized by their regional typology, involving remote sources and a suite of chemical transformations. Others are driven by local sources, especially when stagnant meteorological conditions avoid dispersion and favour accumulation of those

primary particles, or when re-suspension processes inject deposited particles into the air again.

Currently mass is regulated in the EU legislation for PM_{10} and $PM_{2.5}$. There is a growing interest for very fine particles ($PM_{1.0}$) and even ultrafine particles ($PM_{0.1}$; UFP) because they might have particularly harmful effects on human health (see e.g. Viana et al., 2012). However, for UFP mass is no longer a relevant parameter to measure, and it should be replaced by the number of particles. So far particle numbers are regulated at the emission exhaust for vehicles (EC, 2007) only and not in the ambient air. Considering UFP changes significantly the modelling concept that has to be applied, and models to simulate concentration in number are still under development, **we recommend, for policy applications, to focus on the use of models to simulate PM mass concentration**.

2.1.1 Application guidance (1):

1. It is usual to distinguish the following categories to describe and model particulate compounds in PM_{10} and $PM_{2.5}$:
    - PPM : Primary particulate matter (carbonaceous species, other directly emitted mineral compounds)
    - Dust : Desert dust, soil erosion, road traffic particles
    - Sea salt
    - NH_4 : Ammonium
    - NO_3 : Nitrate
    - SO_4 : Sulphate
    - SOA: anthropogenic (ASOA) and biogenic (BSOA) secondary organic aerosols
2. Some specific components can also be simulated like persistent organic pollutants (POP), heavy metals, etc.
3. The aerosol size is closely linked to its composition. PM total mass is the indicator which is considered for regulatory purposes, with a distinction between PM_{10} and $PM_{2.5}$ (with reference to the diameter of the particle, measured in µm). Moreover, some compounds have characteristic sizes (Figure 5). Mineral dusts can be found in the largest bins, while the size of the major part of organic compounds ranges in the fine particulate matter range (lower than 2.5 µm)

Note that when going into details concerning the PPM, ASOA and BSOA composition, one can distinguish elemental *versus* organic carbon (EC and OC, respectively).

The particle size distribution is described in aerosol models. One can distinguish three major representations of the particle size distribution (PSD) in air quality models: continuous, sectional and modal. In the modal representation, the PSD is modelled by several log-normal distributions, also called modes (Devilliers et al., 2012). Usually, the modes are: the nuclei mode, the accumulation mode and the coarse mode. The accuracy of this approach is limited by the number of modes. In the sectional representation the particle size spectrum is divided into a finite number of sections (or bins), and the PSD is approximated by the integrated number, surface, mass or volume concentrations over each section, depending on the particle characteristics of interest.

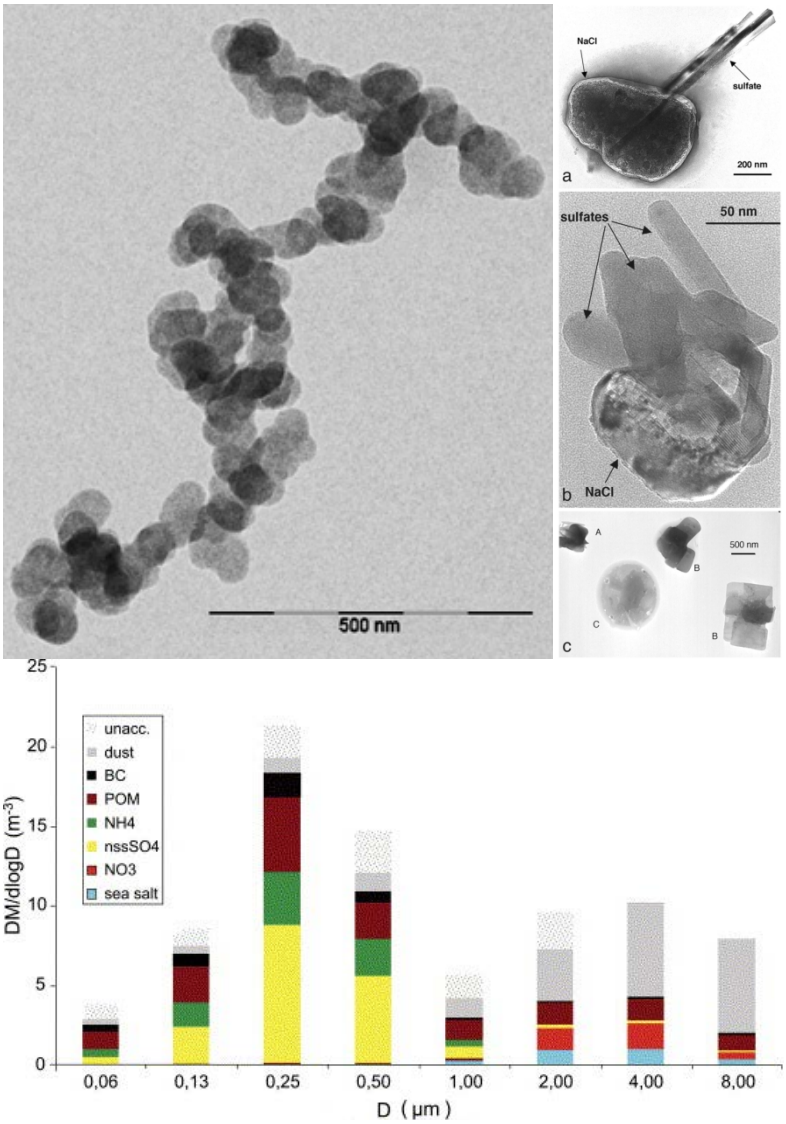

Figure 5: Size and structure of various PM compounds. Source : Cachier et al. (2005)

Modelling correctly ambient PM chemical compounds requires a good knowledge of the emissions and the chemical composition of primary PM and gaseous precursors. In particular secondary PM results from the presence of nitrogen compounds and volatile organic compounds (VOCs). Depending on emissions and meteorology the composition of particulate matter can significantly vary with the location and the period of the year.

An extensive study related to the aerosol composition throughout Europe has been carried out in 2008 in the framework of the COST action 633: “Aerosol Phenomenology - Physical and Chemical Characteristics of Particulate Matter at Rural, Urban and Kerbside sites” (COST, 2008). PM data ($PM_{2.5}$ and PM_{10} mass concentrations, chemical composition and size distribution) recorded at 40 monitoring sites were analysed.

Putaud et al. (2004) proposed an analysis of PM composition across Europe (Figure 6). In general, PM_{10} and $PM_{2.5}$ levels increase when moving from natural background to kerbside sites. It is however interesting to note that in Belgium, both PM_{10} and $PM_{2.5}$ concentrations are very similar at sites close to cities compared to urban background sites. PM_{10} and $PM_{2.5}$ mass concentrations can also be as high at rural areas or near city sites such as Illmitz (Austria) or Ispra (Italy), as at urban background sites such as Zurich and Basel (both Switzerland). These data were directly comparable since they were obtained with similar sampling and analytical methods run between 1998 and 2001. They highlighted that the PM

concentrations inside cities also depend on the background PM of the region where the cities are located. Further, the mass concentration of the coarse fraction (i.e. PM_{10} minus $PM_{2.5}$) is generally higher at urbanized sites compared to rural background. This shows that even coarse particles have predominantly anthropogenic sources[8]. Mineral dust represents generally the major single natural component of the coarse PM fraction. It is interesting to mention that the mineral dust concentrations observed in Barcelona (Spain), using methods similar to those used in Bern (Switzerland), are significant. Possible reasons are the aridness of the surroundings as well as frequent occurrence of Saharan dust transport towards Spain.

[8] As the aerosol coarse fraction mass and components are mostly obtained by difference (PM_{10}–$PM_{2.5}$), uncertainties in the coarse fraction concentrations are large.

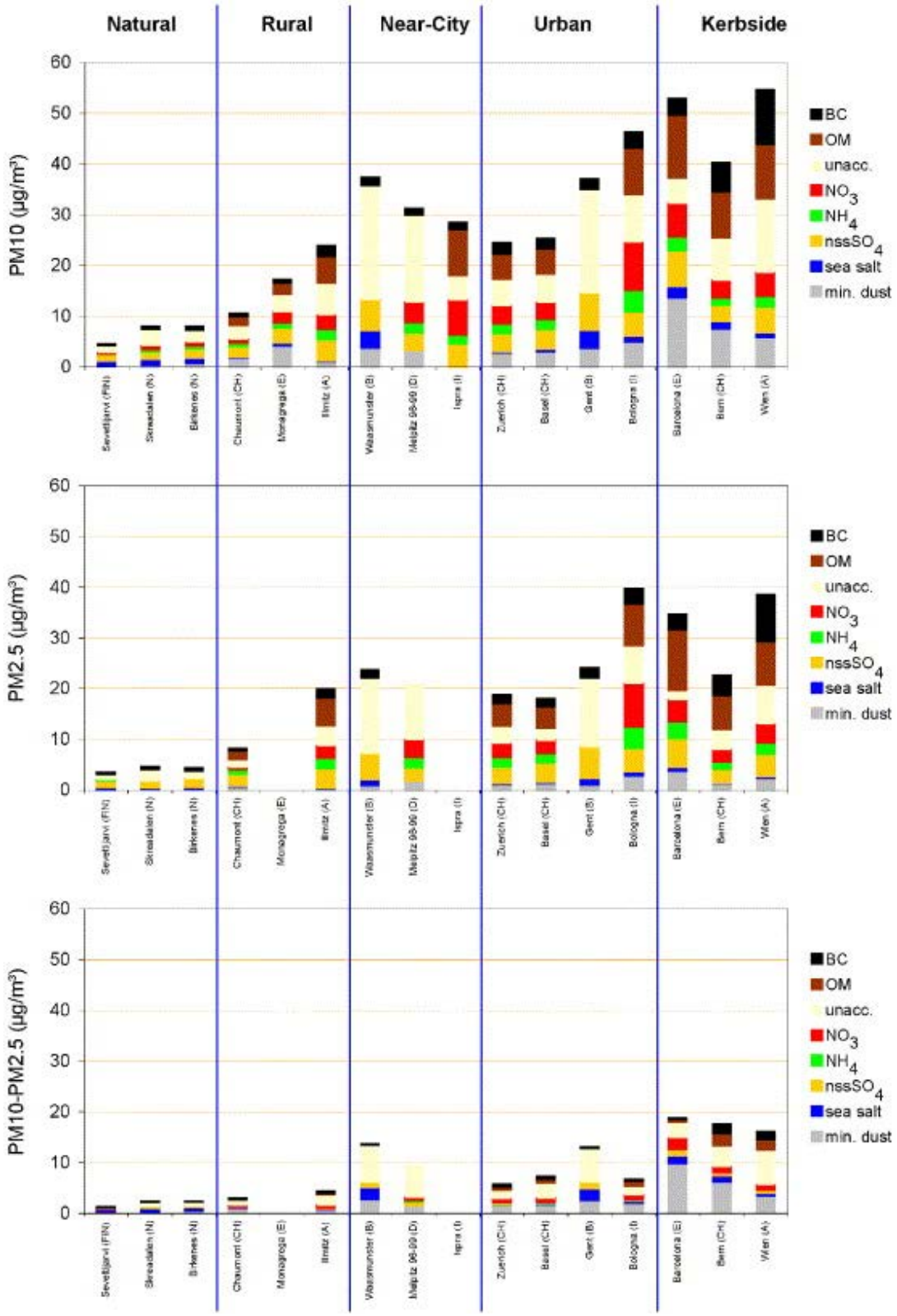

Figure 6: Absolute annual average concentrations of the main aerosol components in (a) PM10, (b) PM2.5 and (c) PM10–PM2.5. BC refers to black carbon, OM to Organic matter (Source : Putaud et al.,2004).

2.1.2 Application guidance (2)

1. Because of the importance of secondary formation processes, it is important to model correctly the aerosol composition (including secondary formation processes) in order to get reliable simulations of PM_{10} and $PM_{2.5}$ ambient concentrations.
2. At the regional scale, the secondary aerosol formation processes and transboundary transport are critical, and the model should be able to describe chemical processes. Chemistry-transport models are generally used.
3. At the local (typically street canyon) and urban scales, local and low height sources (traffic, residential heating, industrial diffusive sources) are the main influencing factors. Therefore accurate emission data should include a detailed description of primary components which are dispersed and transported after their emission. Dispersion models, whatever their complexity (see the FAIRMODE NO_2 guide for a detailed review, Denby et al., 2011a), can be applied provided relevant boundary conditions from extra-domain contributions are reliably assessed. Those conditions can be provided from regional models or from measurements.
4. At the local scale, only transport pathways that impact primary PM need to be considered. Secondary particles formation at the local scale is generally neglected.

2.1.3 Synthesis and recommendations for modelling

Depending on the geographical scale considered, two kinds of models can be used to simulate PM concentrations:

- **At the regional scale**, chemistry transport models (CTMs) should be used to take into account the large number of chemical reactions that are responsible for the formation and the transformation of PM compounds. Annex 1 gives an overview of the physical-chemical processes that should be considered. Some simplifications and assumptions have been introduced by the modellers (especially concerning the choice and reactions of precursors to build up modelled chemical species). Current models can be applied at the regional down to the urban scale with spatial resolution varying from 1 to 100 km^2. A number of CTMs are available and fit to simulate PM regional concentrations. A review of the available tools can be found in Annex 3.
- **At the local scale** (from the urban to the street levels), urban and street canyon models are often used. In such approaches, based on analytical equations, chemical transformation of particles is generally neglected, and those models are fitted to simulate the behaviour of primary compounds whatever their size class is (PM_{10} or $PM_{2.5}$): only transport and in some cases deposition processes are considered. A review of the available tools can be found in Annex 4.
- **Concerning models based on statistical or geostatistical methods**, they are generally dedicated to mapping and assessment. They are based on available observation data, which are processed together with auxiliary variables related to emissions, population density, land use, road patterns etc. However those approaches need large sets of observation data that are not always available. These kriging models are very well-developed for mapping NO_2 concentration fields. But the situation for PM is very different. Field campaigns based on the implementation of numerous and easy-to-use sampling systems (like passive tubes for NO_2) to grid the domain are much more difficult to set out for PMs, simply because of metrological limitations (it is still difficult to implement simple hand-held samplers for multiplication of field campaigns). Therefore such techniques are less developed for modelling PM concentration patterns than deterministic approaches.

2.2 Uncertainties in PM modelling

It is well known that PM concentrations at urban and local scales can be underestimated by regional chemistry transport models (see uncertainties below). The reasons for this can be summarized as follows:

- ***Uncertainties in emissions***: not all possible sources are considered, mainly wood burning, wild fires, diffusive emissions from industrial activity, and salting and sanding; uncertainties in the emission factors or activity data (particularly for natural emissions including sea salts, soil erosion, and wild fires). Significant uncertainties are also attached to the vertical distributions of a number of these sources.
- ***Uncertainties in some process parameterisations***: secondary organic aerosol production which involves a large number of VOCs and semi-VOCs is still studied in research projects and modelling schemes are not really mature. However, very recent research results are moving forward on this issue (Ahmadov, 2012; Zuend et al, 2012; Bergstrom et al., 2012). Operational use of SOA modules in chemistry transport models is still scarce. Another process which is not very well taken into account in current models is resuspension of particles from roads; however its contribution to local exceedances may be very high. Finally PM modelling is very sensitive to dynamical parameterisations that drive PM behaviour in the atmosphere. This is particularly true for the vertical diffusion coefficient (so called "K_z") and the deposition velocities. Values attributed to those parameters in the models can change significantly the simulated ambient concentration values, and high uncertainties remain, especially in urban areas.
- ***Numerical uncertainties*** and particularly the model resolution in both horizontal and vertical directions. The resolution of models has an impact on meteorology, emissions and on transformation processes.

Ranking or prioritizing sources of uncertainty is not easy, because it strongly depends on the characteristics of the modelling exercise. For instance, according to the type of episodes studied, the model sensitivity can be driven either by its ability to simulate correctly chemical compounds (so the driving factor is the chemical model parameterisation), or by dynamical processes (e.g. vertical mixing "K_z"). The location of the model receptor influences uncertainty analysis as well. However, it is generally agreed that uncertainties in emissions is the main driver, whatever the scale. Improving the modelled values of meteorological parameters (mixing height, temperature, wind speed...) over urban areas is also essential.

Very recently a wealth of new modelling results showed the improving performances of the PM modelling approaches, especially in chemistry transport models. Enhanced computing capacities allow modellers to deal with more and more complex processes and to refine spatial resolutions. However, PM model performances are closely linked to the quality of the emission inventory used. Therefore encouraging the improvement of PM and PM precursor emission inventories and ensuring consistency between city level and national emission inventories is essential. When mapping PM background concentrations, data assimilation approaches which allow correction of simulated concentration fields with measured PM observations where possible and relevant (according to the model resolution) can give improved results for use by decision makers and AQ managers. Relevant regional modelling results are already available on the European scale provided by the COPERNICUS[9] Atmosphere services developed through the MACC/MACC2 FP7 projects (http://www.Copernicus.eu/pages-principales/services/atmosphere-monitoring/ ; for more details, please see also section 5 of this report). Other approaches focus on the definition of appropriate corrections to be applied over urban areas using statistical analysis, proxies of

[9] Global monitoring for Environment and Security

sources (for example population density). The so-called “urban increment” elaborated within the EC4MACS[10] project is an example.

State of the art: Figure 7 sketches urban modelling issues. Processes within the urban sublayer up to the canopy layer have to be well simulated (i) to fit better with the spatial representativeness of measurements, and (ii) to better address the human exposure. Figure 8 based on recent CHIMERE model results (Bessagnet et al, 2012) illustrates the significant benefits of improving the horizontal spatial resolution of the model, and very reasonable performances in modelling PM. Those results have been confirmed recently by an intercomparison modelling exercise set-up under the aegis of the EMEP program (www.emep.int) for the Convention on Long Range Transboundary Air Pollution. To assess the impact of fine model resolution on air pollutant concentrations a sensitivity study has been coordinated by the TNO (NL) in 2012, involving four European chemistry-transport models (CTM), run over a whole year with various spatial resolutions ranging from 56 x 56 km^2 to 7 x 7 km^2.

The results (http://www.unece.org/index.php?id=30313) were assessed comparing model concentrations to observations for different site typologies available in the AIRBASE database. They clearly showed the added-value and the good performance of the current models to reproduce PM concentration fields in Europe.

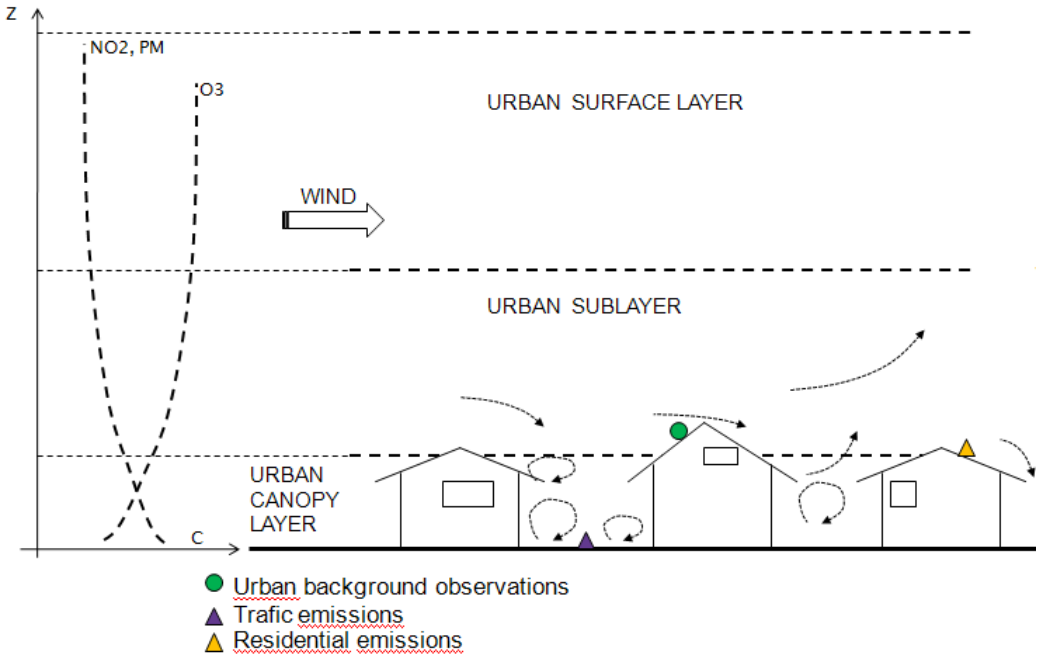

Figure 7: Urban modelling issues

Finally, the Copernicus-MACC2 project provides a lot of material to quantify the uncertainties of current regional models. Figure 9 and Figure 10 illustrate the statistical indicators daily updated and presented on the web site, to address model performances for simulating PM_{10} concentrations. It should be noted the good behaviour of the “ensemble” model which is a combination (the median) of results calculated by seven individual models..

[10] EC4MACS is a EU LIFE project : European Consortium for Modelling of Air Pollution and Climate Strategies (www.ec4macs.eu)

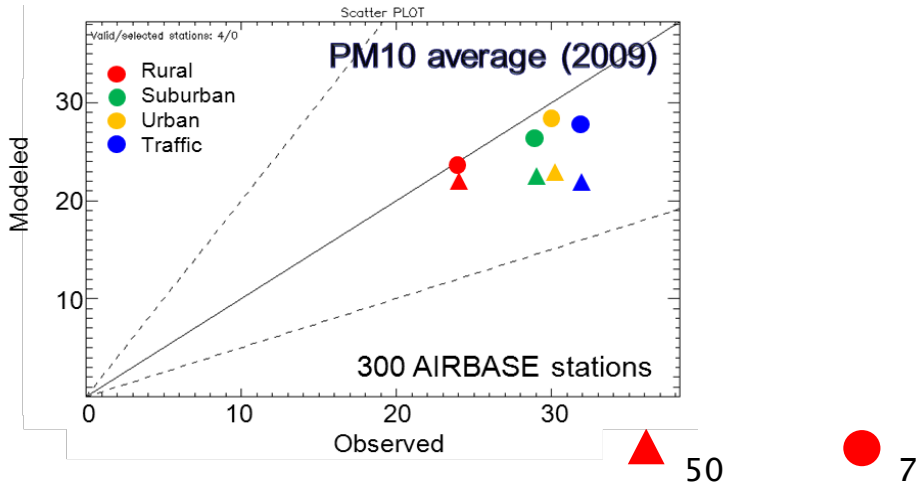

Figure 8: Observed versus modelled PM_{10} concentrations in 2009. Benefits of the refinement of the model spatial resolution. Source: CHIMERE results for the EC4MACS project.

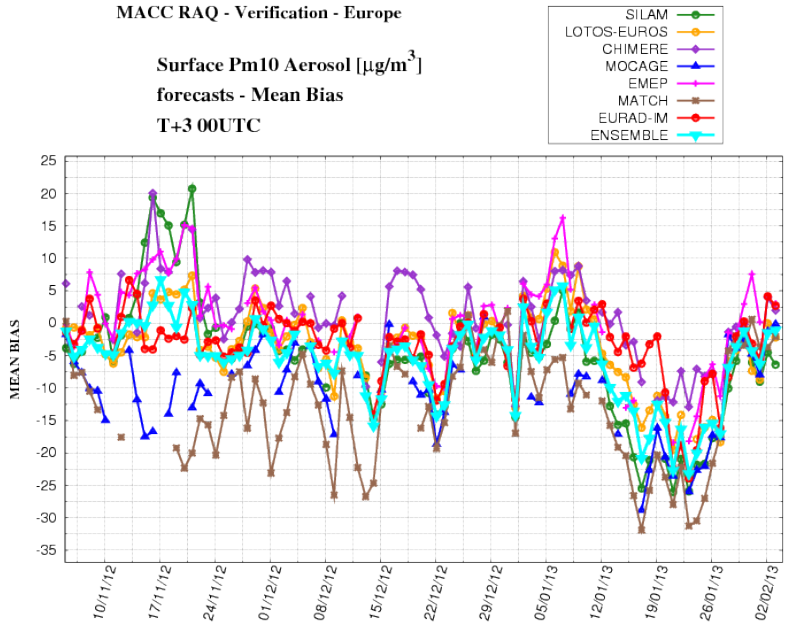

Figure 9: Evaluation process of the regional chemistry transport models (including the Ensemble) involved in the Copernicus/MACC services. The bias for PM_{10} daily mean concentrations is displayed for a 3 month period. Source : http://macc-raq.gmes-atmosphere.eu/

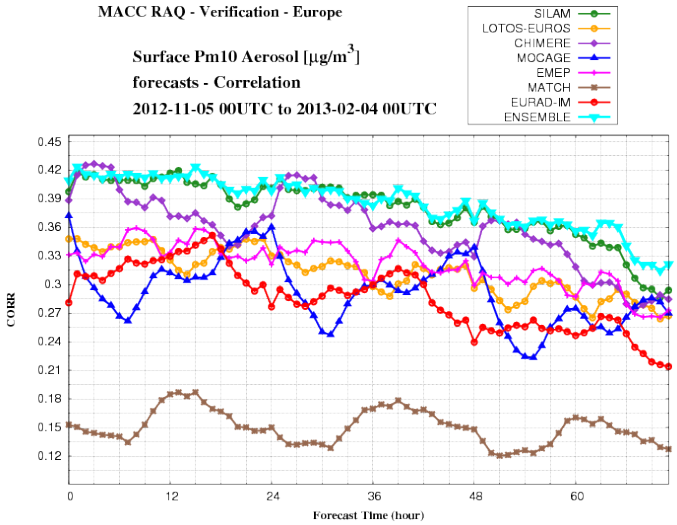

Figure 10: Evaluation process of the regional chemistry transport models (including the Ensemble) involved in the Copernicus/MACC services. The correlation for PM10 daily mean concentrations is displayed for a 3 month period. Source : http://macc-raq.gmes-atmosphere.eu/

2.3 Linking models of different scales

2.3.1 Main issue

The general scheme of scale bridging in model suites is depicted in Figure 11. Regional models provide boundary conditions to more local models (urban to street models) and are fed by global model boundary conditions. Air quality observations can be used to scale the model outputs for more accuracy. Air quality observations can be directly used in local models to get the background PM concentrations.

As shown in Figure 8, the difference between rural and urban PM_{10} concentrations is about 7 µg/m^3, this delta drops to only 2 µg/m^3 between urban and traffic sites (average values in 2009).

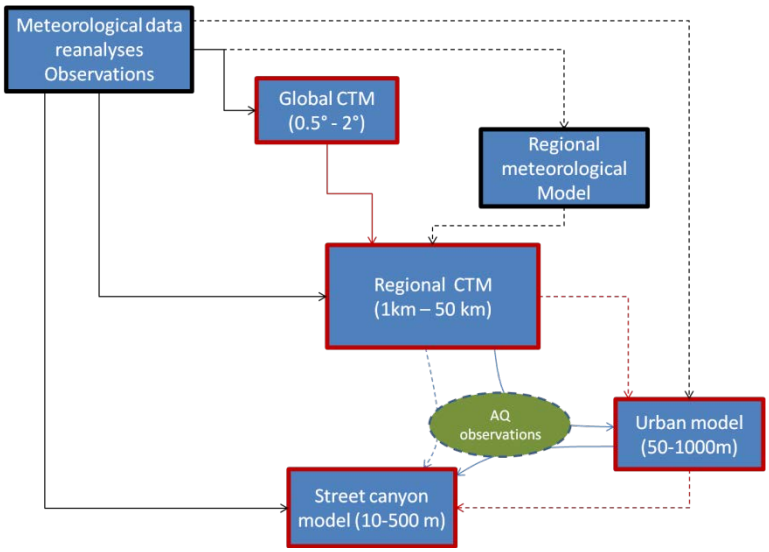

Figure 11: General flowchart of the model "cascade" from global to local scales for the inter-model fluxes of meteorological and chemistry data. Dash lines correspond to optional or possible paths. Observations can be assimilated in models or used to scale the model outputs.

2.3.2 Subgrid modelling : Incremental view of PM levels in urban environments

The "additive" approach (Figure 12) is a handy way for decision makers to describe and understand the origin of PM, but it can be sometimes misinterpreted (Lenschow et al., 2001).

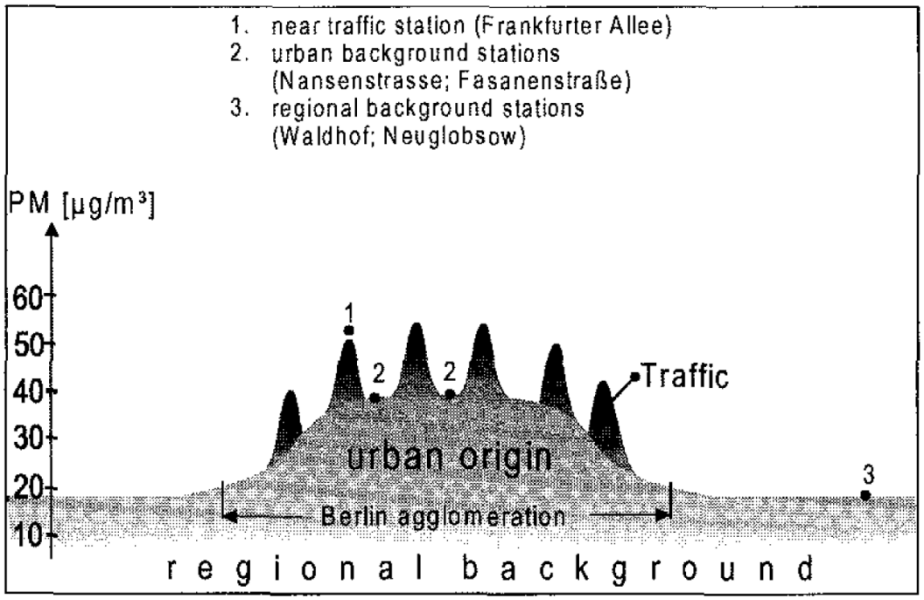

Figure 12: Schematic horizontal profile of the ambient PM10 concentration in Berlin according to Lenschow et al. (2001)

From a modelling point of view, the former City-Delta project brought together the 17 major European urban and regional scale atmospheric dispersion models (Thunis et al., 2007) and developed a generalized methodology to describe the increments in $PM_{2.5}$ concentrations in urban background air that originate - on top of the long range transport component - from local emission sources. These relationships associate the difference in the annual mean $PM_{2.5}$ concentrations between an urban area and the average concentrations calculated over the 50 x 50 km^2 grid cell surrounding the city with spatial variations in emission densities of low-level sources and city-specific meteorological and topographic factors. This methodology has been improved in the frame of ECMACS project (EU LIFE) to be used in the GAINS[11] air quality integrated assessment model (Amann et al., 2011).

Moussiopoulos et al. (2011) proposed a quite similar simple approach for accurately estimating an concentration increment on top of the regional background for urban areas in Europe. The method operates by establishing a functional relationship between the concentration increment and the local meteorological situation, the city characteristics, the urban emissions and background concentrations. This approach builds on earlier attempts to provide estimates of the urban increment for various pollutants, by improving key aspects concerning the treatment of the meteorological parameters. The method has been carried out for PM_{10} and NO_2 based on concentration measurements from 16 cities in order to fix the

[11] The Greenhouse gas – Air pollution Interactions and Synergies (GAINS) model has been developed as a tool to identify emission control strategies that achieve given targets on air quality and greenhouse gas emissions at least costs. GAINS considers measures for the full range of precursor emissions that cause negative effects on human health via the exposure of fine particles and ground-level ozone, damage to vegetation via excess deposition of acidifying and eutrophying compounds, as well as the six greenhouse gases considered in the Kyoto protocol. In addition, it also considers how specific mitigation measures simultaneously influence different pollutants. Thereby, GAINS allows for a comprehensive and combined analysis of air pollution and climate change mitigation strategies, which reveals important synergies and trade-offs between these policy areas.

functional relationship parameters using multiple regression. The functional relationship was then validated by estimating urban increments for the same pollutants in 7 additional urban conglomerations. The method can be potentially applied to a wider range of pollutants, including $PM_{2.5}$ and CO, depending on the availability of emission and regional background concentration data.

2.4 Sensitivity to boundary and initial conditions (Regional models)

2.4.1 Review of the literature

Regional chemistry transport models are usually initialized and driven at the boundaries (top and lateral) by global model outputs or measurements-derived profiles. Usually for PM, global model outputs are used in most regional models like CHIMERE (Vautard et al., 2005) and EMEP (Simpson et al., 2012).

Very few studies investigated the impact of the long range transport of PM to Europe. The hemispheric transport of PM was assessed by Brandt et al. (2012) with a chemistry transport model, for particles, the contributions from North America to Europe is around 0.9% (~0.05 $\mu g/m^3$). Liu et al. (2009) proposed a receptor modelling study with a global chemistry transport model in order to evaluate domestic versus background origins of $PM_{2.5}$ concentrations in several regions of the world (Table 1). Their results show that $PM_{2.5}$ from outside Europe contributed to 30 % of the background $PM_{2.5}$ concentrations in Europe. The main species contributing to the $PM_{2.5}$ is dust followed by sulphates. Organic matter and black carbon concentrations have according to Liu et al. (2009) mainly a domestic origin in Europe.

	Sources	Unit	Receptors									
			NA	SA	EU	FSU	AF	IN	EA	SE	AU	ME
Total		**$\mu g\ m^{-3}$**	**4.4**	**6.1**	**7.6**	**4.6**	**25.9**	**16.4**	**17.7**	**5.0**	**5.8**	**25.2**
	Sulfate	%	49	18	48	25	5	20	29	40	13	12
	BC	%	5	5	7	2	1	5	6	6	2	1
	OM	%	33	49	27	22	12	30	22	41	21	4
	Dust	%	14	28	18	50	82	45	43	13	64	83
Domestic		**$\mu g\ m^{-3}$**	**3.6**	**5.0**	**5.2**	**3.2**	**24.3**	**9.7**	**15.1**	**2.8**	**5.2**	**15.2**
	Sulfate	%	50	10	53	18	1	28	29	31	7	12
	BC	%	5	6	9	2	1	8	7	8	2	1
	OM	%	39	59	38	28	12	48	23	61	23	4
	Dust	%	5	25	0	51	85	16	41	0	68	83
Background		**$\mu g\ m^{-3}$**	**0.8**	**1.1**	**2.4**	**1.5**	**1.7**	**6.7**	**2.6**	**2.2**	**0.6**	**10.0**
	Sulfate	%	28	37	28	36	42	8	30	48	17	11
	BC	%	1	0	1	2	1	0	2	3	1	1
	OM	%	4	5	5	10	9	4	15	15	9	3
	Dust	%	52	42	58	46	41	87	51	30	27	84
	DMS	%	15	16	8	5	7	2	2	3	46	1

Table 1: Contributions to annual average area-weighted fine aerosol ($PM_{2.5}$) surface aerosol concentrations (SAC) (units: $\mu g/m^3$) over each receptor region. 'Total' indicates total fine aerosol ($PM_{2.5}$) concentrations including ammonium sulfate, black carbon (BC), organic mass (OM), and fine dust; 'Domestic' indicates aerosol concentrations resulting from local emissions; Background is the difference between 'Total' and 'Domestic' concentrations. The percent contribution from each aerosol species to each category (i.e., 'Total', 'Domestic', 'Background') is also quantified. Note: ''DMS'' represents sulfate aerosols derived from DMS (dimethyl sulphide), while ''Sulfate'' in the 'Background' category represents sulfate contributed from ROW (ROW= ships, airplanes, volcanoes, etc.). NA: North America, SA : South America, EU : Europe, FSU : Former Soviet Union, AF: Africa, IN: India, EA: East Asia, SE: Southeast Asia, AU: Australia, ME: Middle East. (Source: Liu et al., 2009)

Regarding the time frequency of PM boundary conditions to be used in models, some studies provide some guidance. In the frame of the AQMEII project, Schere et al. (2012) showed that for both O_3 and PM_{10}, using 3-hourly fields at the boundaries contributes to obtaining a slightly larger variability that is more in agreement with the observations for O_3 and NO_2. The time variability is impaired for PM_{10} showing that the predictability of dust events (intensity and occurrence) remains difficult, as demonstrated by Menut et al. (2009).

In Borge et al. (2010) simulations performed with the CMAQ model suggested that model performances were affected by spatial and seasonal factors, the results indicated that model-derived dynamic boundary conditions improved CMAQ predictions when compared to those based on static concentrations prescribed in the boundaries.

Concerning the initial boundary conditions, simulation results from Samaali et al. (2009) suggested the use of a spin-up period of longer than one week for a large (continental) domain and long-term simulation of $PM_{2.5}$ and O_3 rather than the 2–4 days commonly assumed in the literature.

2.4.2 Application guidance:

1. It is important to frame carefully the simulation domain and to account for the essential pollution sources. For instance, it is well known that the South-West of Russia is affected by frequent agricultural and biomass fires (Witham and Manning, 2007). If the regional domain encompasses this area and if the fire emissions are not accounted for, there will be a lack of PM in this region. A simulation targeting Western Europe will account for these phenomena through boundary conditions.
2. Usually global models account for biomass burning emissions; in that case it is suggested to limit the extension to the East in order to benefit from global model boundary conditions. This recommendation also applies to the issue of desert dust emissions in the southern boundaries.
3. For regional models, **dust and sulphate boundary conditions** appear to be very important compared to the other aerosol components. Consider material available from Copernicus/MACC services or other global models (GOCART http://acd-ext.gsfc.nasa.gov/People/Chin/gocartinfo.html - Chin et al, 2000) to pick up data for providing boundary conditions to regional models. Note that the time variability of dust outbreaks is difficult to capture by global models and this feature has a direct impact on the quality of the regional simulations.

3 Emission data

Specific and extensive review of the state of the art in the description of emission processes is needed when starting with PM modelling. Whatever the spatial scale considered, PM modelled concentrations are highly sensitive to emissions and their spatio-temporal variability. Therefore, emissions are still one of the main sources of uncertainty in PM modelling. Actual quantification of those uncertainties is still difficult, but a lot of progress has been made during the last five years, both on how to build up an emission inventory and to parameterize dynamic emission processes that are at least as important as PM chemistry to simulate PM mass concentrations.

From the modelling point of view, two classes of emissions can be distinguished: the usual "static" emission data that are usually found in yearly emission inventories as those officially reported within the Convention of Long Range Transboundary Air Pollution and under the EU's National Emission Ceiling Directive (NEC, 2001), and the "dynamic" emissions. The latter are characterized by large spatial variability as well as temporal variability (yearly, weekly and sometimes daily). This variability can be explained by the occurrence of pollution episodes that might not always be expected. For example:

- increasing use of wood for residential heating in very cold periods of the year;
- intensive use of fertilizers in spring in some parts of Europe leading to an increasing release of nitrogen compounds to the atmosphere;
- biogenic volatile organic compounds (VOC) mainly released by vegetation in summer periods;
- emissions due to sanding and salting in winter in northern European countries.

3.1 Sources of emission data: Bottom-up & top-down inventories

This section describes the usual inventories that are available with basic information on the methodologies (EMEP, TNO, national emission inventories).

3.1.1 The EMEP and the NECD emission inventories

The EMEP emission inventory is a major activity of the EMEP program (Co-operative program for monitoring and evaluation of the long-range transmission of air pollutants in Europe - www.emep.int) under the Convention on Long Range Transboundary Air Pollution. The Parties (countries) who ratified the Convention are asked to to report annually their emission totals of nitrogen oxides, sulphur dioxide, volatile organic compounds, ammonia, carbon monoxide, particulate matter (PM_{10}, $PM_{2.5}$, and Total Suspended Particles), heavy metals (9) and about 25 persistent organic pollutants (http://www.ceip.at/ceip/). Those are official emissions covering the Convention geographical domain (Europe from "the Atlantic to Oral mountains") available through regular processes. Emissions are described by activity sectors. The countries have been encouraged to submit "gridded emissions" better fitted for use by modelling teams. So far some countries who submit gridded emissions use a 50 km resolution. By 2015, the countries will be invited to report their emissions on a 0.1°*0.1° lat-long grid, according to the increased resolution of the EMEP simulation provided in support to the Convention work, which will also reach 0.1°*0.1°. Consolidation and QA/QC (including review, gap filling where no data are reported) of the official EMEP emission inventory is ensured by the Center on Emission Inventories and Projections hosted by UBA Vienna (http://www.ceip.at/ceip/).

The EU as a whole ratified the UNECE's Convention on LRTAP (UNECE, 1979) in 1982. Article 2 of the convention states that 'the Contracting Parties, taking due account of the facts and problems involved, are determined to protect man and his environment against air pollution and shall endeavour to limit and, as far as possible, gradually reduce and prevent air pollution including long-range transboundary air pollution'.

On 4 May 2012, the Executive Body for the LRTAP Convention adopted amendments to the Gothenburg Protocol (UNECE, 2013). The new text of the protocol includes national emission reduction commitments for the main air pollutants to be achieved in 2020 and beyond. Further, the revised protocol will include emission reduction commitments for fine PM. Black carbon (a short-lived climate forcer) is included as a component of PM. In 2013, parties were requested to report emissions data for NO_X, NMVOC, SO_X, NH_3, CO, HMs, POPs and PM, as well as associated activity data. Like last year, the EU also includes pollutants that can be reported additionally (As, Cr, Cu, Ni, Se, Zn, BaP, BbF, BkF, IP and TSPs; EEA, 2013a).

The NEC Directive requires all 28 EU Member States to report information annually concerning emissions for four important air pollutants: nitrogen oxides (NO_X), non-methane volatile organic compounds (NMVOC), sulphur dioxide (SO_2) and ammonia (NH_3) (EEA, 2013b).

3.1.2 The COPERNICUS MACC-TNO emission inventory

The COPERNICUS EU (www.Copernicus.eu) program aims at developing and maintaining operational services for monitoring the European environment. A set of services, developed under research FP7 projects called MACC and MACC-II (Monitoring Atmospheric composition and Climate) (http://atmosphere.Copernicus.eu/ *),* relate to regional (European) air quality. Those services provide air quality forecasts and analyses, past re-analyses and policy supporting tools to help in management of air pollution at the European scale. They are based on both modelling and measurements (in-situ and satellite).

The modelling activities are supported by access to a high resolution emission inventory developed under the projects to fulfil MACC modellers' needs. Air pollutant emissions in Europe (including PM) are described throughout Europe on a 6 to 7 km^2 resolution grid. The totals by countries are consistent with the official emissions reported under the EMEP process, but investigations have been carried out to improve spatial and temporal distributions of emissions by sector, as expected by the modellers. Annual emission data are available so far from 2003 to 2009.

3.1.3 The French inventory: an example of a bottom-up approach

The French "national spatialised inventory" for atmospheric pollutant emissions (INS) project was launched under the authority of the French ministry in charge of ecology in 2003. It aims at creating a full database of emission sources at high spatial and time resolutions (possibly 1*1 km^2 and 1 hour). The computing platform is fully operational from 2013 onwards. Through a dedicated web site, air pollutant maps will be available at various time and spatial scales. It will be an input of the PREV'AIR system (daily air quality forecast over Europe and France).

The methodology used in INS to evaluate emissions is, in general, bottom-up: emissions are evaluated from the product of unit activity data by an emission factor specific to this activity. For some activities, energy balances at national level can be made to correct the final emissions. INS platform accounts for 41 major pollutants and 549 speciations of these pollutants: acidification, eutrophication, photochemistry, greenhouse effect, heavy metals, persistent organic pollutants and particulate matter. Each INS emission is characterized by

an activity sector using three inputs keys: SNAP[12] activity, if necessary a rubric (additional information to snap activity) and an associated fuel (napfue). The time frequency ranges from hourly to annual bases.

The emissions sources are geographically distributed either as point sources (for some industrial sources), surface (residential, commercial and institutional, agricultural, diffuse industrial and road emission sources) or as linear (major roads, train, fluvial, aviation or maritime emission sources). In the case of ground sources, emissions are evaluated and distributed uniformly on low administrative level (municipality). Geographical information systems (GIS) are then used to spatially distribute and refine ground emissions: the INS uses the European database Corine Land Cover which describes the land use according to 44 classes (http://www.eea.europa.eu/data-and-maps/figures/corine-land-cover-types-2006). The Postgres /Postgis (database system) association is used to compute and distribute the emissions.

3.2 Pre-treatment of emissions

Models generally simulate hourly air pollutant concentration fields on a given modeling grid. Therefore they need to be fed by input data consistent with such constrains. Emission data issued from available emission inventories generally cannot be directly used by the models. A number of pre-processing operations are required to adapt the emission databases to the model format and to provide the appropriate input to models. Such operations are briefly described below with a focus on issues related to PM modeling.

3.2.1 Temporal disaggregation

We have noted in previous sections that emission inventories usually provide annual totals which generally need to be disaggregated on a relevant temporal scale for use in models (both deterministic or statistical ones). An hourly description of emissions might be required for running local and regional air quality models. This can be achieved in three steps by using (i) monthly profiles (or seasonal profiles), (ii) weekly and (iii) hourly profiles. The profiles are usually available by country, activity sectors and pollutants.

However, when local and national data and expertise are lacking, modellers can use for the diurnal profile a unique profile (Menut et al., 2012) for all compounds and countries (Figure 13). This might be inappropriate and further investigations on the temporal profiles may be necessary to improve model inputs.

[12] Selected Nomenclature for Air Pollution

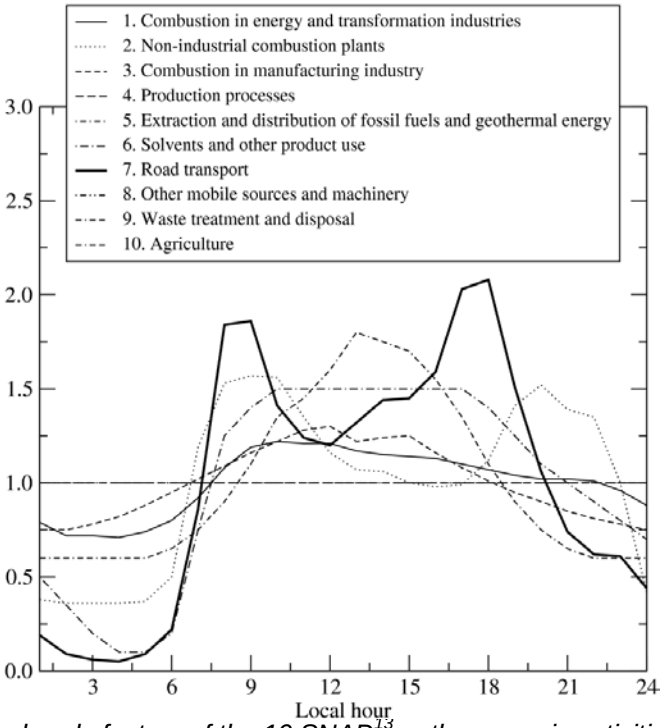

Figure 13: Emissions hourly factors of the 10 SNAP[13] anthropogenic activities sectors (Monday) traditionally used in chemistry transport models (from Menut et al., 2012)

For example, Menut et al. (2012) proposed a set of new profiles for the traffic emissions calculated from NO_2 traffic measurements. NO_2 is a good proxy to reallocate in time the road traffic gas and PM emissions. The emission factors are separately estimated for each weekday as presented in Figure 14. To distinguish between days is crucial since the traffic intensity is different on working days compared to weekends. For example, the morning traffic is very low for Saturdays and Sundays, compared to the other days of the week. Figure 15 presents some examples of these new hourly diurnal profiles of NO_2 concentrations for several cities in Europe. It is assumed that this variable is a good indicator of the road traffic activity in the cities. The difference between cities is large, highlighting the fact that traffic emissions do not have the same variability in time from one country to another. However, the profiles for two cities in the same country show roughly the same profile, confirming that the emission profile of mobile sources depends on the lifestyle and customs in a given country.

[13] SNAP : Selected Nomenclature for air pollution

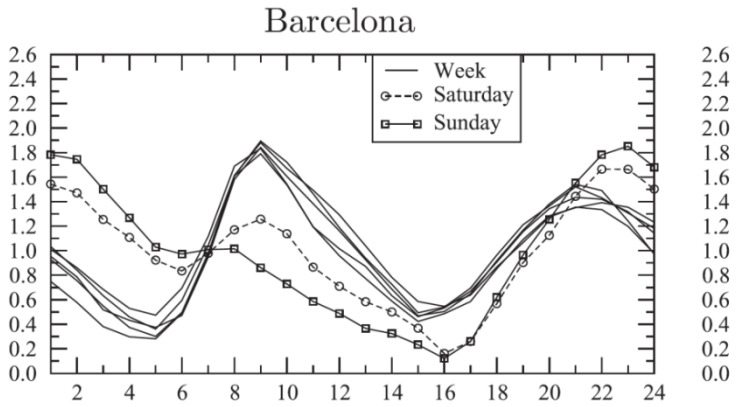

Figure 14: Hourly factors estimated in Barcelona for all weekdays and week-end – Source : Menut et al. (2012)

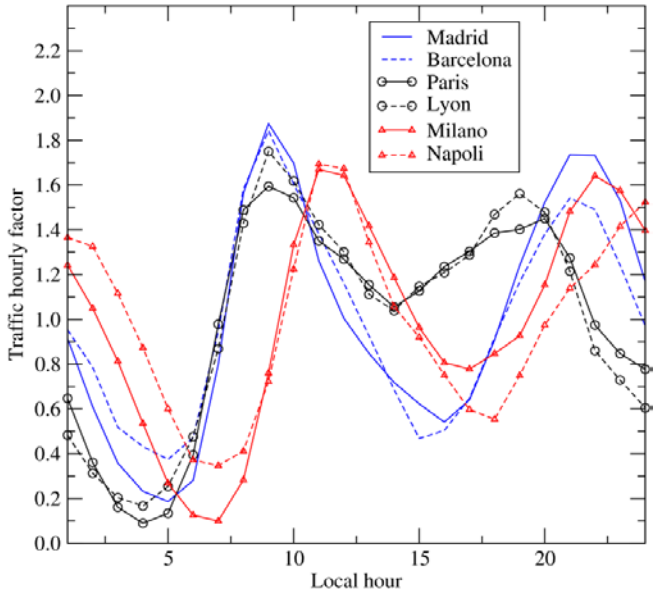

Figure 15: Hourly factors estimated with NO_2 measurements for different cities in various countries, for a given week-day (Monday here) – Source : Menut et al. (2012)

3.2.2 Spatial distribution of emissions

The emission inventories given in the example in section 3.1 have various spatial resolutions that may be inconsistent with the model resolution grid. Generally, available emissions are distributed over coarse grids, which have to be refined. Therefore the first step to build up emission inventories well-suited for modelling consists in re-gridding the anthropogenic emissions available at coarse resolution onto the high resolved target grid using adequate proxies. Databases of proxies are usually freely available on websites. Some commonly used examples are:

- USGS[14] : Land use data at 1 km^2 resolution (global)
- GLCF[15] : Land use data at 1 km^2 resolution (global)
- GLOBCOVER[16] : Land use data at 300 m^2 resolution (global)
- CORINE[17] : Land use data at 300 m^2 resolution (Europe)
- GRUMP[18] : Population data 1 km^2 resolution (global)
- EEA database[19] : Population data 100 m^2 resolution (Europe), Gallego (2010)

[14] http://landcover.usgs.gov/
[15] http://glcf.umiacs.umd.edu/
[16] http://postel.mediasfrance.org/en/PROJECTS/Preoperational-GMES/GLOBCOVER/
[17] http://www.eea.europa.eu/themes/landuse/interactive/clc-download
[18] http://sedac.ciesin.columbia.edu/gpw/

- EPER[20] : Large Source Point database (Europe)

For the land use data, usually ten to thirty (or more) categories can be identified and can be used to re-grid the emissions. A minimum of 1x1 km^2 resolution assures that the spatial scale of emissions is addressed with accuracy. As a first approach, an activity sector is associated to a land use category or a proxy (Table 2).

Emission sector or type	**Landuse category or Proxy**
Agriculture	"crops" and "grass land"
Road Traffic	Population and roads
Industry	"Artificial areas"
Residential heating	population
Forest fires	"forests" and "shrubs"
Industry	Large Source Points

Table 2: Landuse and associated proxy

3.3 Residential sector emissions (static and dynamic)

This sector is the most important sector for PM emissions in wintertime in suburban and rural areas. The high sensitivity of emission spatial patterns to account correctly for this contribution has been demonstrated in the EC4MACS project (www.ec4macs.eu), where a new spatial distribution of the SNAP2[21] emissions has been proposed from the EMEP reported data for the whole of Europe according to the population density distribution (http://www.ec4macs.eu/content/report/EC4MACS_Publications/MR_Final%20in%20pdf/Chimere_Methodologies_Final.pdf). $PM_{2.5}$ is mainly emitted by wood burning. As shown in Figure 16, a relationship can be drawn to reallocate $PM_{2.5}$ emissions not only in city centres but also over remote areas where wood burning is developed. Figure 17 represents the annual $PM_{2.5}$ SNAP2 emissions for 2009.

[19] http://dataservice.eea.europa.eu/

[20] http://www.eea.europa.eu/data-and-maps/data/member-states-reporting-art-7-under-the-european-pollutant-release-and-transfer-register-e-prtr-regulation-8

[21] According to the SNAP nomenclature for air pollutant emissions SNPA2 refers to "Non industrial combustion plants" www.eea.europa.eu/publications/EMEPCORINAIR/partb.pdf

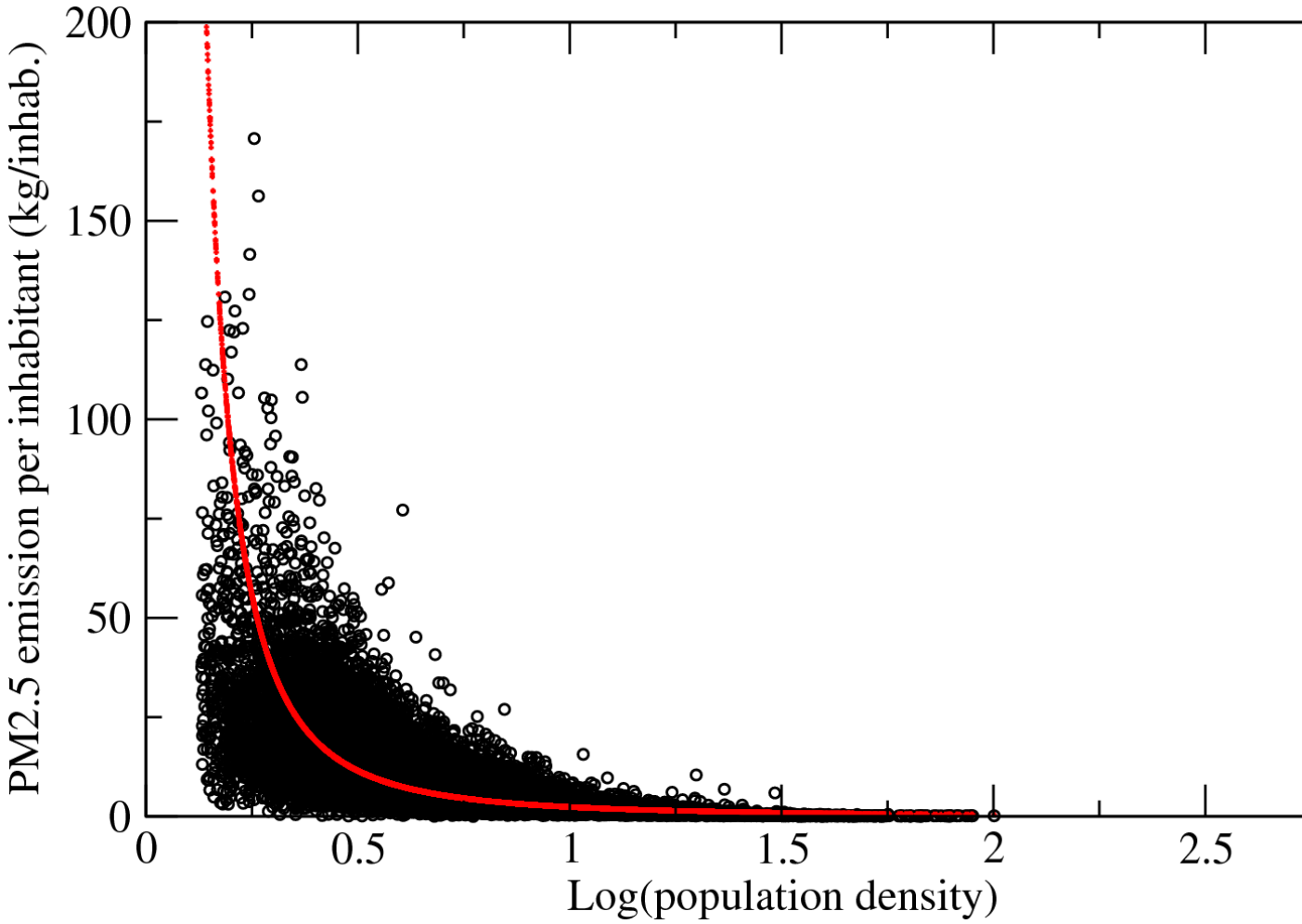

Figure 16: $PM_{2.5}$ emission per inhabitant (issued from the French bottom-up emission inventory) as a function of population density. Source : http://www.ec4macs.eu/content/report/EC4MACS_Publications/MR_Final%20in%20pdf/Chimere_Methodologies_Final.pdf

During cold spells, particularly wood burning is an important supplementary way of heating widely used in Europe. The activity is difficult to estimate, but it is possible to reallocate the emissions over cold spells with the “degree day” concept. This correction allows episode modelling and forecasting, or at least significant events can be considered when stagnant meteorological conditions avoid pollutant dispersion. The degree day is an indicator used as a proxy variable to express the daily energy demand for heating. The degree day for a day *j* is defined as:

D_j = max(0, 20-T_D) where T_D is the daily mean 2m air temperature. The daily modulation factor (F_j) is therefore defined as: $F_j = D'_j/\overline{D'}$. Where $D'_j = D_j + A.\overline{D}$ and $\overline{D'} = (1 + A).\overline{D}$ where *A* is a user defined modulation factor accounting for other kind of emissions (e.g. production of hot tap water). $\overline{D}$ is the yearly mean degree day. The factor F_j is applied to each hourly emission flux.

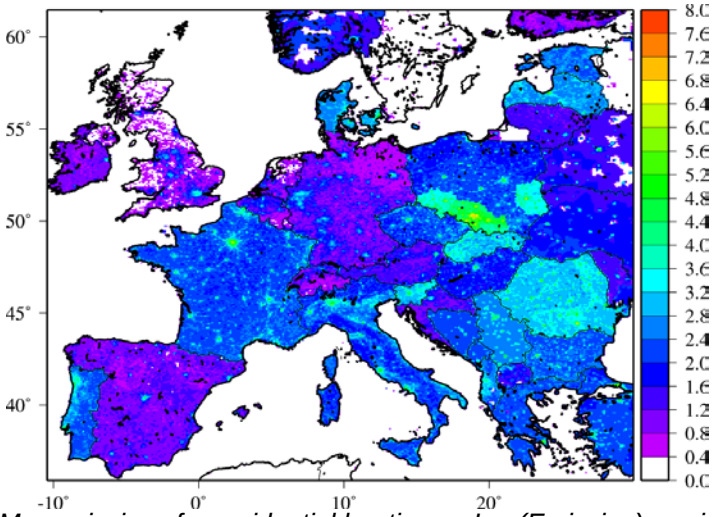

Figure 17: Annual $PM_{2.5}$ emissions for residential heating as Log(Emission) regridded with population data and rural/urban share ratio based on bottom-up approaches (source: INERIS for EC4MACS EU LIFE

3.4 Traffic emissions (direct and indirect)

3.4.1 Direct emissions

Because they represent a large part of anthropogenic emissions, road traffic emissions are carefully described in available emission inventories. Direct emissions (from vehicle exhaust) result from the product of emission factors (which depend on fuel used and vehicles categories according to EURO standards) and activity data (which depend on the number of vehicles for a given period on the target road segment). A detailed description of road traffic emission cannot be provided at the national scale. Only city-scale inventories can have a chance to describe with certain accuracy the fleet of vehicles and their number. They are based on models of traffic suited to describe the distribution of road traffic in the city regarding its road typology, the day and the time of the day etc.

Some cities have made large efforts to improve their inventory of road traffic direct emissions (Berlin, London, Paris, Vienna...) and several initiatives have been launched to support local and national authorities in improving their inventory methodologies (see for example EEA, 2013c; EEA 2013d) and the FAIRMODE (Forum for Air Quality Modelling in Europe) initiative itself (http://fairmode.ew.eea.europa.eu/emissions-projections-sg3). But improving direct traffic emissions both at local and regional/national scales remains an important challenge for PM modelling (Figure 18).

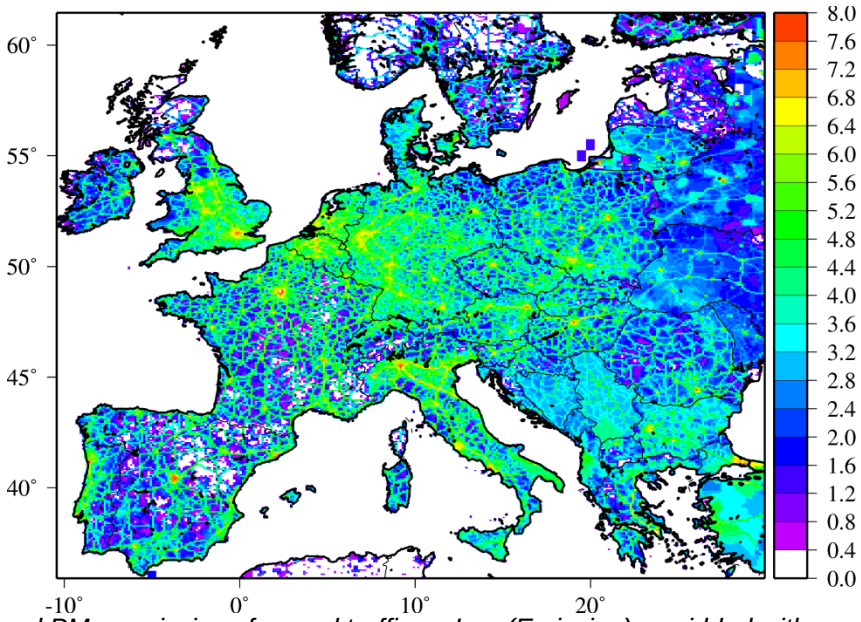

Figure 18: Annual $PM_{2.5}$ emissions for road traffic as Log (Emission) regridded with road data. Source INERIS for EC4MACS EU-LIFE and MACC EU FP7 with input data from TNO

3.4.2 Indirect traffic emissions: a major PM source

Road dusts can be defined as particulate matter (PM) generated from the road surface due to road surface wear. Tyre and brake wear may also be important "non-exhaust" emission sources. Particles generated by these frictional processes may be emitted directly to the air or deposit on the road surface and road shoulder and be re-suspended later due to vehicle induced turbulence and/or interaction between the tyres and the surface. Moreover, dust emitted in the vicinity of for example cities can be deposited on urban roads. Finally, the available mass for re-suspension may also be increased by the addition of sand or salt to the road surface, particularly important in countries where traction control methods such as traction sand and/or de-icing material are applied. This phenomenon results in enhanced production of road dust and salt that deposits on snow piles on the roadside during winter. When the snow melts in spring and the surface dries up, traffic induced turbulence causes the particles to be suspended into the air, and hence the PM_{10} (particulate matter with a diameter of 10 µm or less) concentrations increase significantly.

It has been estimated that at least 40% of total PM_{10} traffic emissions are due to road traffic re-suspension (Luhana et al., 2004). This ratio can reach up to 90% in springtime as reported by Folsberg et al. (2005) in Stockholm. So far, only few chemistry transport models or local Gaussian or street canyon models host re-suspension modules in an operational way. However the Swedish model SIMAIR has such capacities and is used in operational policy applications (Omstedt et al 2005)

As written in the Air Quality Directive "*Contributions to exceedances of particulate matter PM_{10} limit values attributable to winter-sanding or –salting of roads may also be subtracted when assessing compliance with air quality limit values provided that reasonable measures have been taken to lower concentrations*" (EC, 2008).

Therefore some European countries are interested in considering PM emissions from re-suspension, particularly in wintertime. Some modelling tools exist to assess the road traffic emissions by re-suspension. Currently, operational road dust emission models (EPA, 2006), the Tønnesen model (Tønnesen, 2000), the Omstedt model (Omstedt et al., 2005), the EMEP CORINAIR method (EMEP CORINAIR, 2013) are site-specific and highly empirical as they are based on local measurements. In particular, they include empirical constants (US

EPA, the Tønnesen model) or reference emission factors (the Omstedt model) applicable to only one or very few road environments.

Recently, Berger and Denby (2011) proposed a new and generalised road dust emission model. They based the emissions on road, tyre and brake wear rates and used the mass balance concept to describe the build-up of road dust on the road surface and road shoulder, depending on soil moisture. The model separates the emissions into a direct part and a re-suspension part, and treats the road surface and road shoulder as two different sources. The model was tested under idealized conditions as well as on two datasets in and just outside of Oslo in Norway during the studded tyre season. The model has been further developed by Denby et al, 2013 and applied over long past periods in Stockholm and Copenhagen.

In Spain, Pay et al. (2011) describe the inclusion of the re-suspension of particulate matter within the HERMES emission model (operationally implemented in the CALIOPE forecasting system) and the improvements obtained in the simulations of the PM_{10} mass over a domain covering Spain for a whole year of simulation (2004). The results indicate a remarkable improvement of the PM_{10} predictions, reducing the biases and errors by around 15-18% (2.6 $\mu g/m^3$ for the average bias in Spain). The emissions have strong local effects on the modelled particle concentration in or near the largest urban zones (up to 7 $\mu g/m^3$ as the annual average), albeit those positive effects are more limited in background areas, since the deposition mechanism was found to be a significant sink for these re-suspended particles in the chemistry transport model.

Even if these models offer the possibility to evaluate at local and urban scale the emission of road dust fraction of $PM_{2.5}$ concentrations, it is not yet possible to obtain the fraction of PM attributable to sanding and salting.

3.5 Wind blown dust emissions

Mineral dust is a natural compound in the atmosphere known to affect air quality in several regions of the world. An extensive overview of the dust contribution to PM concentrations in Europe is presented in (EEA, 2012).

Mineral dust emissions, transport, deposition and their impact on the radiative properties of the atmosphere have been particularly studied for the two main emitters in the world, Western Africa and China (Andreae and Crutzen, 1997; Sokolik et al., 2001). Worldwide measurement networks in Europe, North-America and elsewhere have been used, such as AERONET (Holben et al., 2001). Models such as the global model GOCART (Ginoux et al., 2004) and regional models such as, CHIMERE-DUST (Forêt et al., 2006;Menut et al., 2007) and DREAM (Nickovic et al., 2001; Perez et al., 2006, Park et al., 2007) or LOTOS-EUROS (Schaap et al., 2010) among others, integrate a specific module to account for dust emissions.

However, open scientific questions remain. The direct transport of mineral dust originated from arid areas toward large industrialized and urbanized areas has rarely been studied. Some studies report how the Taklamakan desert in north-western China directly impacts megacities (Sun et al., 2001; Zhang et al., 2003), and studies were also done for dust transport from Africa toward Europe (Ansmann et al., 2003; Perez et al., 2006). In Europe, most natural dust contributions to PM concentrations originate from the West African Sahara. However, the net contribution remains relatively low: it is rare to record a dust contribution of more than 20% of the total PM mass, as an averaged value over the year, even if sporadic episodes led to a massive contribution of dust during short periods (Simpson et al., 1999; Querol et al., 2004a, 2004b; Moreno et al., 2005; Escudero et al., 2007a). Figure 19 shows the main areas emitting dust from global MODIS observational data (Monks et al., 2009).

However, such episodes can contribute to exceedances of the daily limit value for PM_{10} as set in the EU's AQ Directive (EC, 2008). This issue is also discussed in the EEA Technical Report " Particulate matter from natural sources and related reporting under the EU Air Quality Directive in 2008 and 2009" (EEA, 2012).

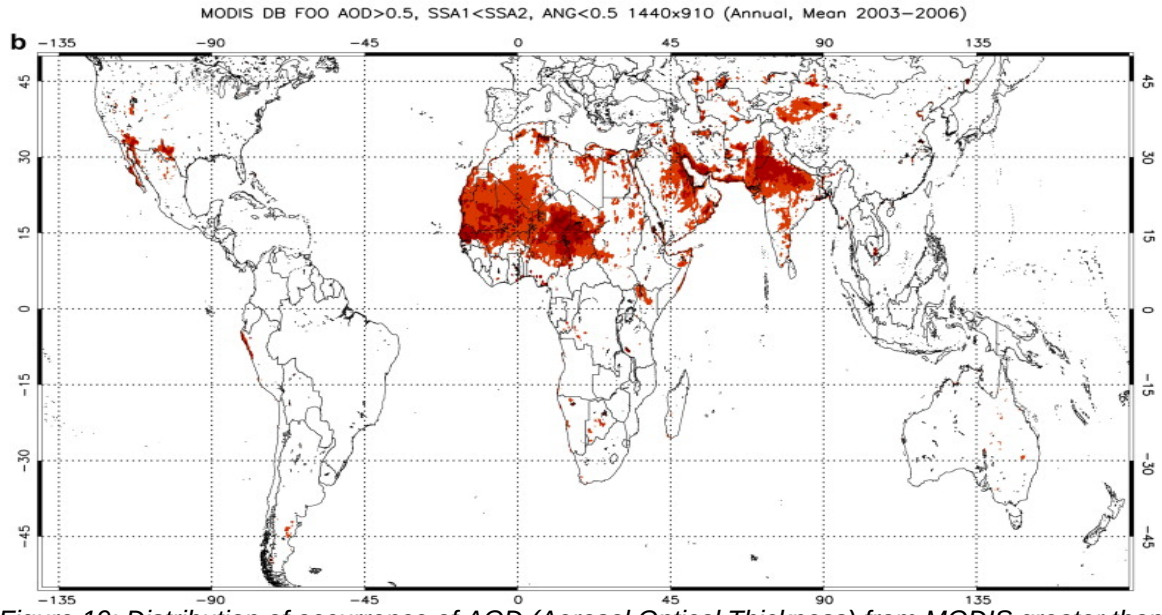

Figure 19: Distribution of occurrence of AOD (Aerosol Optical Thickness) from MODIS greater than 0.5: isocolor lines represent occurrences of 10, 25, 50 and 75% per year (source : Monks et al., 2009)

This natural contribution of mineral dust to European air quality has been particularly studied in countries frequently affected by Saharan dust outbreaks: Spain (Artinano et al., 2003; Alastuey et al., 2005; Escudero et al., 2007b; Querol et al., 2007), Portugal (Petzold et al., 2006), Italy (Kishcha et al., 2005), and more generally for the whole of Europe in studies for example Simpson et al. (1999), Querol et al. (2004b), and Viana et al. (2007). In the countries mentioned above, a large number of experimental field campaigns (Clairborn et al., 1998; Gomes et al., 2003), routine measurements of particle concentrations and speciation analyses (Putaud et al., 2004), were carried out to provide estimations of the relative contribution of natural dust particles to the total airborne PM budget in cities. According to Van Dingenen et al. (2004) the observed background annual average mass concentration of PM_{10} for the continental Europe is 7.0 ± 4.1 µg/m^3..

Issues and challenges

The Saharan dust outbreaks are sporadic and intense. Forecasting their emissions and transport aggregates a large number of uncertainties. Their predictability is very uncertain as estimated in Menut et al. (2009). To improve the modelling tools the first step will be to merge already existing large-scale modelling systems (such as hemispheric dust models) and regional CTMs. A second challenge is to avoid risks of false air quality alerts due to an overestimation of events producing and transporting natural dust. For a large part of CTMs currently applied for forecasts, boundary conditions are global monthly climatology from model outputs. This may be sufficient for some aerosols species, relatively constant in time and space. In the case of mineral dust, nor the mean nor the median values are able to accurately describe the temporal variability.

Wind-blown dust emissions inside Europe also remain poorly estimated. The strong dependency on meteorological variables like the wind speed and the soil moisture is a very

tricky issue. Dust emissions in Europe further depend on the evolution of natural vegetation and agricultural crops. Establishing a robust parameterization in CTMs remains a challenge.

3.6 Natural marine emissions

The ocean is a source of both primary aerosols, in the form of sea salt aerosols, and gas-phase species such as DMS (dimethyl sulphide) that can contribute to secondary aerosol production. The size distribution of aerosols though can be affected by pollution and weather patterns such as the stability of the mixing layer, precipitation and fog (Seguin et al., 2011). Sulfate aerosols over the oceans in the northern hemisphere originate from biogenic, anthropogenic, sea spray and lithogenic sources. DMS, a gas emitted as a result of the breakdown of the cell walls of biogenic matter in the ocean, can oxidize to form SO_2. SO_2 can oxidize further to form sulfuric acid, be taken up into existing aerosols or can be removed via dry deposition. Gaseous sulfuric acid will either add to the growth of existing aerosols or nucleate to form new aerosols which may be able to act as cloud condensation nuclei. Sea spray, containing sulfate, generally is found in larger aerosol diameters but can also act as cloud condensation nuclei in smaller aerosols (de Leeuw et al., 2011).

Aerosol sulfate can be introduced in the environment by anthropogenic sources, either directly or by the oxidation of SO_2. Distinguishing between anthropogenic and biogenic components in aerosols is important for determining anthropogenic impacts on aerosol sulfate burdens. As already mentioned, this issue is also discussed in an EEA Technical Report focussing on reporting natural contribution to PM limit value exceedances (EEA, 2012a). As chloride and sodium are the major components of the sea salt composition (Table 3) their direct emission in the atmosphere can affect the PM_{10} and $PM_{2.5}$ concentrations in coastal regions (Figure 20).

Element	Percent by weight
Cl	55.04
Na	30.61
SO4	7.68
Mg	3.69
Ca	1.16
K	1.1

Table 3: Composition of sea salt, based on the composition of sea water (Seinfeld and Pandis 1998), for components that contribute more than 1% to the sea salt mass

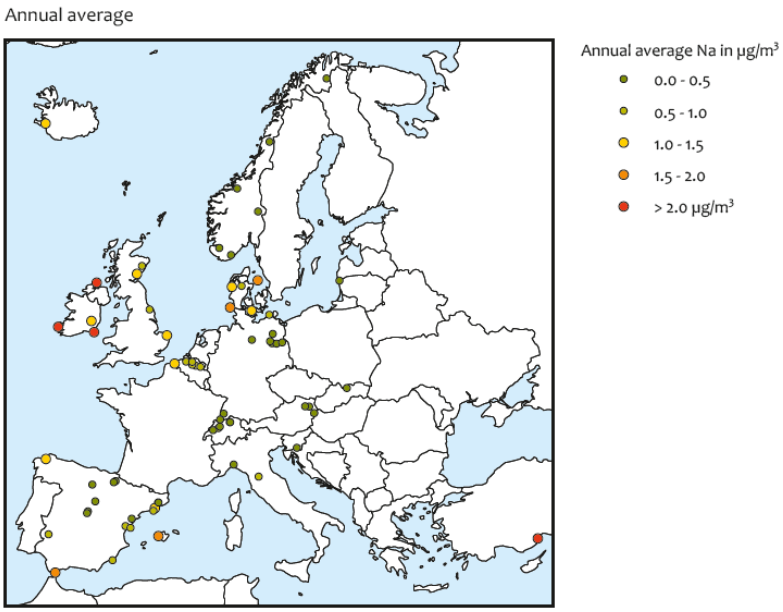

Figure 20: Sodium concentrations in Europe (source : Manders et al., 2009)

One difficulty arises for the implementation of sea salt emissions parameterization. There is an issue on horizontal scale and resolution. These parameterizations are very sensitive to wind speed and land cover, thus emissions sharply decrease until zero from the seaside to inland areas. This kind of gradient leads to large discrepancies between various model configurations. Manders et al (2010) recommend to use observations to help in deriving "*bias-corrected modelled distributions [which] serves a s a best estimate of the sea salt distribution across Europe*".

3.7 Ammonia emissions

According to official reporting, the agricultural sector represents 95% of anthropogenic ammonia emissions (e.g. EEA, 2013b). Ammonia is emitted from livestock housing and by volatilization in the atmosphere after manure and fertilizer applications. The natural fraction of ammonia emissions is difficult to estimate. It comes from the decomposition or breakdown of organic waste matter, animal waste, and the discharge of ammonia by biota. This fraction is most probably below 10% (Sapek, 2013).

Ammonia is an important PM precursor of ammonium nitrate in the air. In a "real world", ammonia emissions depend on the type of cattle, manures and fertilizers, spreading practices, meteorological and soil parameters (Sutton et al., 2011).

The usual time profiles for ammonia currently available are not accurate enough to catch the real instantaneous emission when a parameter (temperature, soil humidity and spreading practices) largely deviates from its average value. Moreover, if annual quantities are not available for the studied year, the closest documented year is chosen assuming that inter-annual variations of emissions are small. In order to better estimate large ammonia emissions during specific meteorological conditions, lately developed emission models allow to better account for ammonia emissions (Diaz Goebez et al., 2003) and the dynamical approach used in Skjoth et al. (2005) shows improved results in retrieving ammonia concentrations. Beuning et al. (2008) have developed ammonia emission models able to predict ammonia emissions peaks in Canada. In Europe, an inter-comparison exercise shows that ammonia emission models provided similar emission factors (Reidy, 2008). Therefore, a dynamical approach for the treatment of ammonia emissions needs to be

implemented in chemistry transport models to obtain better model predictions of high particulate matter episodes (Zhang et al., 2008).

At the French scale, Hamaoui et al. (2012) developed a dynamical emission module (Figure 21). This module accounts for soil moisture, temperature, wind speed and agricultural practices. The spatial distribution of ammonia emissions is rather different in spring. The ammonia emissions tend to be overestimated in Brittany compared to those computed in a "dynamical way" that attribute more emissions by crops.

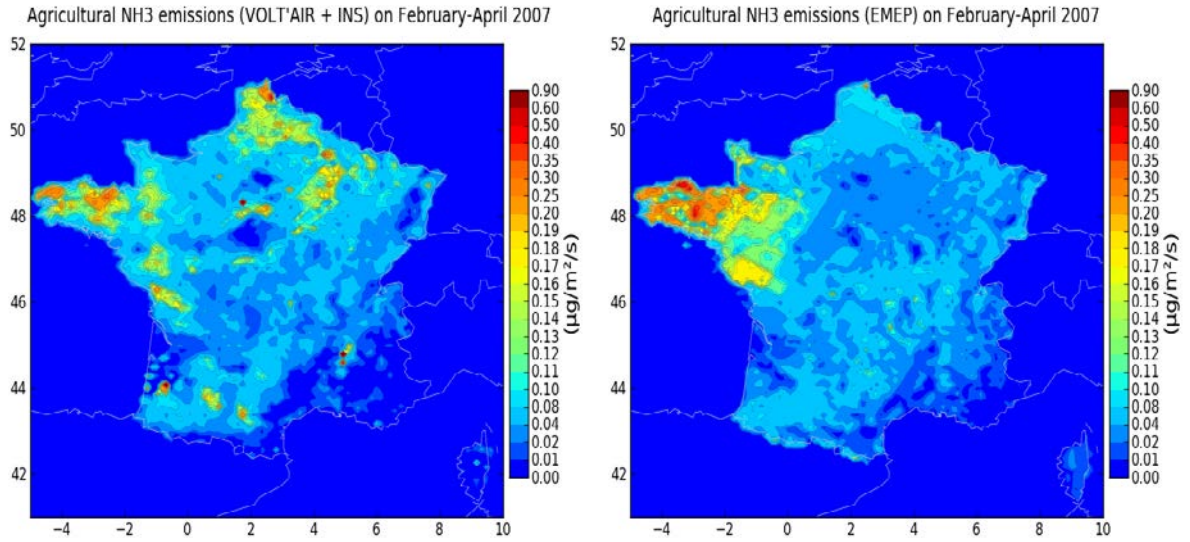

Figure 21: Differences in ammonia emissions between the EMEP emissions (right panel) and the VOLT'AIR model embedded in CHIMERE in spring 2007 over France.

However, ammonia in urban areas could be mainly emitted by other kinds of sources. As observed in Barcelona in summertime by Reche et al. (2012), values were significantly higher at urban background than at traffic sites, probably indicating the impact of emissions from biological sources, such as humans, sewage systems and garbage containers. Thus, the volatilization of NH_3 from the aerosol phase seems to be significant enough during summer to dominate over NH_3 emissions from traffic. Conversely, in winter levels were higher at traffic sites, suggesting a contribution from vehicle emissions. Indeed, NH_3 levels decreased by 55% with increasing distance (50 m) to the direct emissions from traffic. A significant correlation between NH_3 concentrations averaged for the different districts of the city and the number of waste containers per hour and m^2 was also obtained, highlighting the necessity for controlling and reducing the emissions from garbage collection systems.

3.8 Biogenic NO and VOC emissions

Biogenic emissions from vegetation and soils are a major source of PM precursors. Terrestrial ecosystems produce a diverse array of chemicals including many volatile and semi-volatile compounds that are emitted into the atmosphere. Some of these have an important role in atmospheric chemistry including reactive volatile organic compounds (VOC) for which terrestrial ecosystems are by far the biggest contributors to the global annual flux (Guenther et al., 2012). It is now widely recognized that these chemicals can influence atmospheric composition and quantitative estimates of their emissions into the atmosphere are needed for numerical assessments of past, present and future air quality and climate. A few biogenic compounds are now routinely included in air quality and earth system numerical models but the magnitude and variability of these emissions are not well known. Many other compounds are simply omitted from these models because they are thought to be unimportant or because their contribution is assumed to be accounted for by increasing the

emission of the compounds that are included in the models or because so little is known about the emission of these compounds.

Nitric oxide (NO) in the soil is produced by the microbial processes of nitrification and denitrification. NO emission originates from a natural pool of nitrogen and a fraction from fertilizer application. NO from soils can also be calculated in a dynamical way by models. An issue arises concerning the nitrous oxide (NO) emissions issued from the agricultural sector (SNAP10) that are reported in emission in emission inventories. As shown in Figure 22, NO emitted from soils is partially reported in Europe (EU 27) and unrealistic jumps are observed at the borders. Thus, there are two options to account for NO emissions in models, (1) either these unrealistic emissions are kept as input data and NO calculated by dynamic emission modules must not be used to avoid double counting, or (2) the official NO emissions are removed and NO calculated by emission modules must be used.

An example of a model to simulate biogenic emissions: the MEGAN system (Guenther et al., 2012)

The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1) is a modelling framework for estimating fluxes of 147 biogenic compounds between terrestrial ecosystems and the atmosphere using simple mechanistic algorithms to account for the major known processes controlling biogenic emissions. It is available as an offline code and has also been coupled into land surface models and atmospheric chemistry models. MEGAN2.1 is an update from the previous versions including MEGAN2.0 for isoprene emissions and MEGAN2.04, which estimates emissions of 138 compounds. Isoprene comprises about half of the estimated total global biogenic volatile organic compound (BVOC) emission of 1 Pg (1000 Tg or 1015 g). Another 10 compounds including methanol, ethanol, acetaldehyde, acetone, α-pinene, β-pinene, t−β-ocimene, limonene, ethene, and propene together contribute another 30% of the estimated emission. An additional 20 compounds (mostly terpenoids) are associated with another 17% of the total emission with the remaining 3% distributed among 125 compounds. Emissions of 41 monoterpenes and 32 sesquiterpenes together comprise about 15% and 3%, respectively, of the total global BVOC emission. Tropical trees cover about 18% of the global land surface and are estimated to be responsible for 60% of terpenoid emissions and 48% of other VOC emissions. Other trees cover about the same area but are estimated to contribute only about 10% of total emissions. The magnitude of the emissions estimated with MEGAN2.1 are within the range of estimates reported using other approaches and much of the differences between reported values can be attributed to landcover and meteorological driving variables. The offline version of MEGAN2.1 source code and driving variables is available from http://acd.ucar.edu/~guenther/MEGAN/MEGAN.htm and the version integrated into the Community Land Model version 4 (CLM4) can be downloaded from http://www.cesm.ucar.edu/.

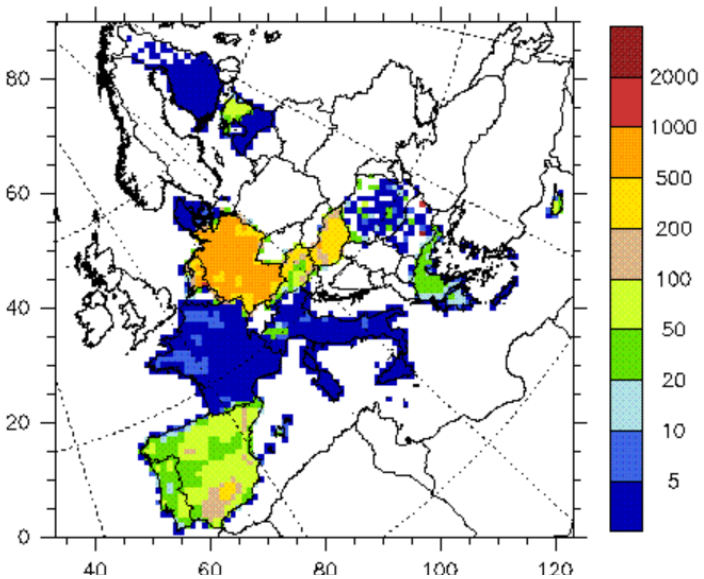

Figure 22: 2010 Emissions of NOx in Mg (emissions as used in EMEP models) for the agriculture activity sector in EU27. Several countries do not report NO emissions.

3.9 Fire emissions:

3.9.1 Background

The importance of fire activity on global atmospheric composition has been recognised since the late 1970s (Seiler and Crutzen, 1980). Its dramatic impact on air quality during extreme events became obvious during extended fires and smoke haze episodes in Indonesia in 1997, resulting in increased hospital admission and mortality (Heil and Goldammer, 2001). This particular event stressed the need for a better monitoring of exposure, implying improved quantification of fire emissions and of their impact on local and regional scales. Large efforts have been dedicated to the analysis of the chemical composition of fire plumes over recent decades, allowing the compilation of comprehensive reviews of emission factors for the observed species (Andreae and Merlet, 2001). These reviews, coupled to the availability of satellite observations of fire location since the mid1990s, have greatly facilitated the emergence of research on the large-scale variability and impacts of fires. Wildfire emissions are composed of large amounts of trace gases and aerosols. Among the trace gases emitted, the most abundant (90-95%) are carbon dioxide (CO_2) and carbon monoxide (CO), but also methane (CH_4), a series of volatile organic carbon compounds (VOCs) and inorganic species. In addition to primary species, emitted directly into the atmosphere, secondary species may be formed by chemical reactions within the plumes, in particular ozone.

To assess the impact of forest fires on air quality, Martins et al. (2012) applied the LOTOS-EUROS model and found an up to 20% contribution of fires to PM concentrations during specific periods in 2003-2005.

This issue is also discussed in a recent EEA Technical Report with focus on reporting natural contribution to PM_{10} limit value exceedances (EEA, 2012a).

3.9.2 Wild fire emission estimates

Ideally, the compilation of emission inventories requires knowledge of the quantity and type of vegetation burnt. Information on the fire type is also needed since plumes emitted by fires in difference combustion phases (flaming or smouldering) have very different compositions. However, except for the study of specific fires that were fully monitored (for example in the

case of prescribed fires) such information is missing and needs to be estimated. For this, scientists rely on a variety of information for instance:

- Observations of the fire characteristics: location, extent, energy
- Databases constructed from a set of measurements in fire plumes (in situ or laboratory experiments)
- Cartography of the vegetation, often based on satellite observations
- Modelling of the carbon cycle and vegetation dynamics

The classic formulation for calculating the emissions is based on the original work of Seiler and Crutzen (1980):

$$E_i = \sum_f A_f FL_f EF_{i,f}$$

where E_i is the emission (kg) for a given species i, A is the area burned (m^2), FL_f is the fuel load available for burning (kg dry matter per m^{-2}) for a fuel type *f* and $EF_{i,f}$ is the emission factor (g/kg dry matter). Inventories are generally constructed for large regions (for a specific country or globally) and fire and/or vegetation characteristics need to rely on approximations, leading to large uncertainties on the final estimates.

As also shown by Langmann et al. (2009), wild fires represent an important input of gaseous and aerosol compounds in the atmosphere. While domestic wood burning emissions can be included in common inventories (usually as residential sector contributions) vegetation fires emissions are difficult to estimate for past periods and a fortiori to predict in a forecast mode. Since vegetation fires strongly depend on meteorological factors and the biomass availability, they have a great inter-annual variability (Langenfelds et al., 2002). In Europe, the Mediterranean Basin and the Portugal (Miranda et al., 1994; Hodzic et al., 2007) are often affected by fires in summertime. During the 2003 fire outbreaks the modelled wildfire emissions caused an increase in average $PM_{2.5}$ ground concentrations from 20 to 200% (Hodzic et al., 2007) in some part of Europe.

3.9.3 Forecasting fires available in COPERNICUS/ MACC

Within the Copernicus/MACC-II project (EU FP7) Kaiser et al., (2012) developed a fire emissions model based on satellite observations (http://www.gmes-atmosphere.eu/about/project_structure/input_data/d_fire/).

The Global Fire Assimilation System (GFASv1.0) calculates biomass burning emissions by assimilating Fire Radiative Power (FRP) observations from the MODIS[22] instruments embedded on the Terra and Aqua NASA satellites. It corrects gaps in the observations, which are mostly due to cloud cover, and filters spurious FRP observations of volcanoes, gas flares and other industrial activity. The combustion rate is subsequently calculated with land cover-specific conversion factors. Emission factors for 40 gas-phase and aerosol trace species have been compiled from a literature survey. The corresponding daily emissions have been calculated on a global 0.5×0.5 grid from 2003 to the present. General consistency with the Global Fire Emission Database version 3.1 (GFED3.1) within its accuracy is achieved while maintaining the advantages of an FRP-based approach: GFASv1.0 makes use of the quantitative information on the combustion rate that is contained in the FRP observations, and it detects fires in real time at high spatial and temporal resolution.

GFASv1.0 indicates omission errors in GFED3.1 due to undetected small fires. It also exhibits slightly longer fire seasons in South America and North Africa and a slightly shorter

[22] MODIS (or Moderate Resolution Imaging Spectroradiometer) is a key instrument aboard the Terra (EOS AM) and Aqua (EOS PM) satellites http://modis.gsfc.nasa.gov/data/

fire season in Southeast Asia. GFASv1.0 has already been used for atmospheric reactive gas simulations in an independent study, which found good agreement with atmospheric observations. We have performed simulations of the atmospheric aerosol distribution with and without the assimilation of MODIS aerosol optical depth (AOD). They indicate that the emissions of particulate matter need to be boosted by a factor of 2–4 to reproduce the global distribution of organic matter and black carbon. This discrepancy is also evident in the comparison of previously published top-down and bottom-up estimates. For the time being, a global enhancement of the particulate matter emissions by a factor of 3.4 is recommended. Validation with independent AOD and PM_{10} observations recorded during the Russian fires in summer 2010 showed that the global Monitoring Atmospheric Composition and Change (MACC) aerosol model with GFASv1.0 aerosol emissions captured the smoke plume evolution well when organic matter and black carbon are enhanced by the recommended factor. In conjunction with the assimilation of MODIS AOD, the use of GFASv1.0 with enhanced emission factors quantitatively improves the forecast of the aerosol load near the surface sufficiently to allow air quality warnings with a lead time of up to four days.

3.10 Primary organic aerosol (POA) emissions

POA has been traditionally assumed to be non-volatile and nonreactive in atmospheric aerosol models. Even if these assumptions are used today by the great majority of Chemical Transport Models, and are integral parts of most mental models of the system, it has been clear for decades that neither they are not correct in many cases (Donahue et al., 2009). Most measurements of ambient organic PM concentrations were accompanied by serious negative (particle evaporation after collection on the filter) and/or positive artefacts (vapor adsorption on the filter), providing strong hints about the semi-volatile nature of organic aerosol (Turpin et al., 2000).

For more than twenty years, dilution samplers have been used to measure POA emission factors. Development of these samplers was motivated by the semi-volatile character of primary emissions. Although some primary emissions are clearly semi-volatile, models generally treat them as non volatile. The implicit assumption is that the partitioning measured using a dilution sampler is representative over the full range of atmospheric conditions simulated by the model, and that the semi-volatile primary mass is a small fraction of the total POA.

POA components that evaporate after dilution as they move away from their sources react in the gas phase, forming products with lower volatility that can condense back in the particulate phase (Robinson et al., 2007; Weitkamp et al., 2007). Both of these pathways oxidize primary emissions, forming OOA (oxidised organic aerosols). Even if some progress has been recently made to account for such processes in chemistry-transport models (see the VBS approach - volatilily basis set- in Bergstrom et al, 2012) they are not implemented yet in current PM models. This point will not be developed furthermore in the present document.

4 Meteorological data

4.1 Meteorology in air quality models

Predicted air pollutant concentrations are very sensitive to physical variables such as wind, temperature and specific humidity and diagnosed turbulent parameters (friction velocity, boundary layer height). A direct error concerning the meteorological fields used has a more or less direct and linear impact on concentrations (Menut, 2003; Minguzzi et al., 2005), whatever the model used. In addition, the selected horizontal resolution of the meteorological data has a large impact on results (Valari and Menut, 2008). In the 1990s, the global scale meteorological forecast system outputs, such as NCEP (National Centers for Environmental Prediction, Kalnay et al., 1996) and ECMWF (European Centre for Medium-Range Weather Forecasts, Bechtold et al., 2008) were used, mainly applying interpolations to re-grid the meso-scale data fields. In recent years, the forecast systems have evolved further and they now use meso-scale models, driven by global meteorological fields. These models are more adapted to fine resolutions and use more relevant land cover data and turbulence parameterizations.

4.2 Meteorological drivers – downscaling

In Europe, the American meso-scale meteorological models MM5 (Grell et al., 1994) and WRF (Skamarock et al., 2007) are the most widely used because they are easy to implement and to change. For forecasts, new systems are under development, which are based on the Integrated Forecast System (IFS) operated in ECMWF [23].

Figure 23 illustrates the impact of using a meso-scale model to refine the resolution of a meteorological re-analysis (Menut et al., 2012). The daily gridded observational dataset for precipitation, temperature and sea level pressure in Europe is called E-OBS[24], ERAi is the ERA-interim global ECMWF reanalysis, IPSLcm is the IPSL global coupled climate model (From Institut Pierre-Simon Laplace). The downscaling with a meso-scale model strongly affects the meteorological simulations. The Weather Research and Forecasting **(**WRF) model tends to increase the temperature cold bias (by -2K) and the precipitation positive bias by +1 mm/day. Over land, WRF tends to increases the wind speed by ±1 to 2 m/s, independently of the global model used.

[23] http://www.ecmwf.int/
[24] http://eca.knmi.nl/download/ensembles/download.php

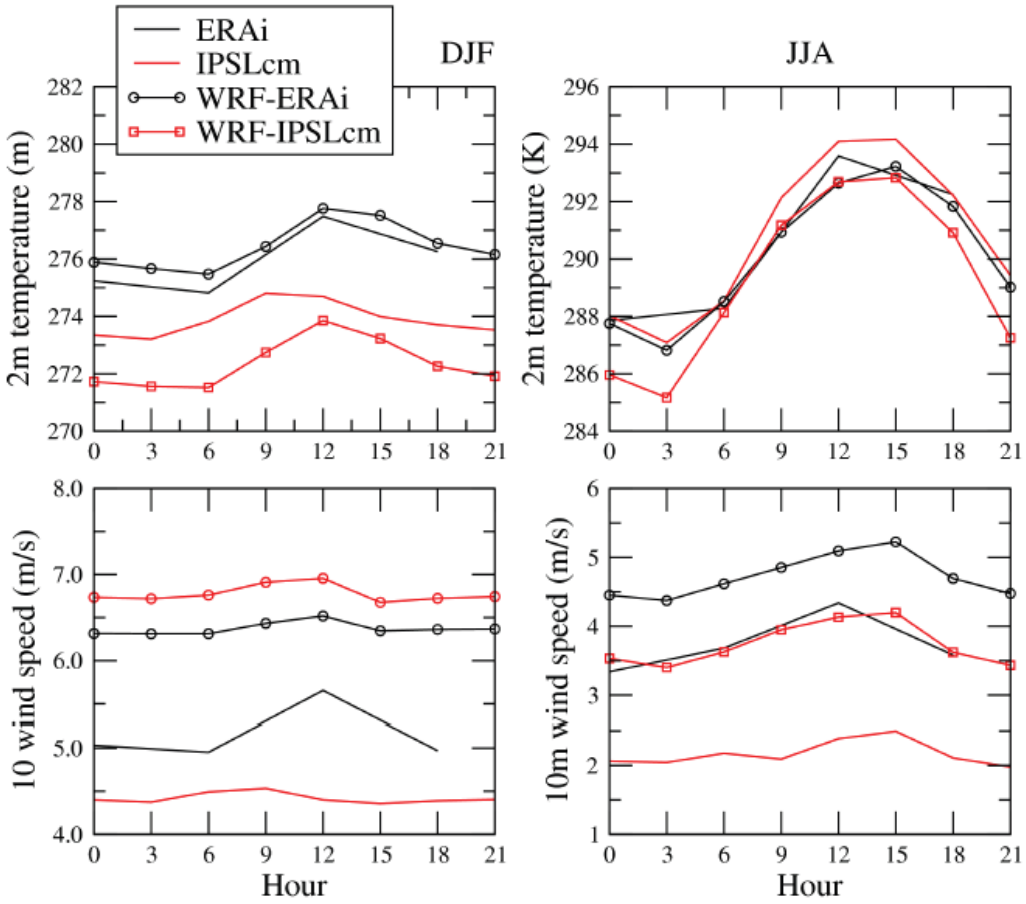

Figure 23: Diurnal cycles of 2m temperature (K) and 10m wind speed (m/s) for the periods DJA and JJA. Calculations are done over the western/central Europe modelled domain for the 1989-2005 period. E-OBS is the observational dataset, ERAi is the ERA-interim global ECMWF reanalysis, IPSLcm is the IPSL global coupled climate model, WRF is the mesoscale meterological model.

The temperature cold bias problem was addressed in Jimenez and Dudhia (2012). They proposed a correction, i.e. a parameterization that is based on the concept of a momentum sink term and makes use of the standard deviation of the subgrid-scale orography as well as the characteristics of the topographic field. Both the drag generated by the unresolved terrain and the possibility of an increase in the speed of the flow over mountains and hills. It could be shown that WRF presents a low wind speed bias, which is considered in the corrected scheme. The surface wind simulation over a complex-terrain region that is located in the northeast of the Iberian Peninsula has been improved with the inclusion of the new parameterization. In particular, the underestimation of the wind speed spatial variability resulting from the mentioned biases can be corrected. The importance of selecting appropriate grid points to compare with observations was also examined. The wind speed from the nearest grid point is not always the most appropriate one for this comparison, nearby ones can be more representative. The new scheme not only improves the climatological winds but also the intra-diurnal variations in the mountains, where the default WRF shows limitations in reproducing the observed wind behaviour.

4.3 Urban meteorology

In dispersion models at the urban scale, the lower part of the boundary layer is often represented by parameterizations derived from the theory of similarity of the surface layer[25]. The urban effects are then considered by changes in surface roughness and heat flux. Nevertheless, these formulations should only be used in the inertial sub-layer (ISL) which is well above the tops of buildings, i.e. in an average height of several tens meters. Indeed, in the sub-rough layer (RSL), i.e. the layer in the immediate vicinity of the urban canopy elements, the flow has a rather complex structure (Raupach, 1980) and the similarity theory cannot be applied.

This first layer of CTM used at urban scales may be under two times the average of the buildings height in big cities. It can be assumed that the corresponding Reynolds stress at this level has not yet reached its maximum value and the corresponding value of vertical diffusion coefficient, named Kz is overestimated even if we can also assume that Kz might be underestimated at the second level. A limit of the Kz methods that are using the theory of similarity is identified here.

To improve models at low levels over urban area, CTM modellers have to learn from what already exists in urban meteorology modelling: TEB (Town Balance Energy), SM2-U (Soil Model for Sub-Meso scale Urbanised version), MOSES (UM Surface Exchange scheme), BEP (Building Effect parameterization, AHF+R+A (Anthropogenic heat flux), MRF-Urb (Medium-Range Forecast Urban scheme), PALM-Urb (Parallelized Atmospheric Large-eddy Model). A summary of references to respective modules, modelling results, and discussions can be found in Mahura and Baklanov (2010).

Several studies deal with the so-called “urbanization” of meso-scale meteorological models (*e.g.*, Martilli et al. 2002; Dupont et al. 2004; Lee and Park 2008; Solazzo et al., 2010).Those models are based on a comprehensive set of equations linking the synoptic to the street scale circulation. Further, they include parameterisations for the turbulent exchange processes of momentum and mass. Solazzo et al. (2010) proposed a simple scheme for estimating spatially-averaged mean wind speed and the urban heat island over a selected neighbourhood area in Lisbon, Portugal. The results show the capability of the urban model to provide more accurate mean wind and temperature profiles. Moreover, the urban model has the advantage of being cost effective, as it requires small computational resources.

4.4 Expected impact of meteorological drivers on PM modelling

Various types of uncertainty studies on aerosol predictions have been performed:

- The impact of orography on meteorological parameters (Carvalho et al., 2006)
- The impact of horizontal resolution of meteorology on model calculations (Baertsch-Ritter et al., 2004; Menut et al., 2005)
- The impact of meteorological parameters on aerosols formation temperature, humidity and vertical mixing (Haywood and Ramaswamy, 1998; Penner et al., 1998; Easter and Peters, 1994)
- The impact of meteorology on gas/particle partitioning and the dry deposition of SOA (Bessagnet et al., 2010)
- The relationship between meteorological variables and PM concentrations (Mues et al., 2012)

[25]Similarity theory starts with the identification of the relevant physical parameters that characterize atmospheric processes; then dimensionless groups are formed from these parameters, and finally experimental data is used to find functional relations between dimensionless groups. Those functional relationships are used as simplified model parametrisations

- The impact of model MM5 configuration on aerosol predictions (Meij et al., 2009 ; Pernigotti et al., 2012)

The formation of ammonium nitrate (NH_4NO_3) is very sensitive to temperature while primary pollutants will be strongly dependent on stability conditions. As discussed in section 3.5, dust particles emissions will be very sensitive to wind speed and soil moisture. Soil moisture is a critical parameter very different from model to model and leads to large differences in emissions fluxes. The way to diagnose the vertical diffusion coefficient K_z strongly impacts the concentrations of primary pollutants. Vertical turbulent mixing takes place only in the boundary layer. Often, in the formulation of K_z, a minimal K_z is assumed, with a value about 0.01 m^2/s. The change of this value directly affects PM concentrations.

All the above-mentioned studies show a strong impact of the meteorology on modelled PM concentrations, whatever the kind of model used.

5 Quality control and evaluation

5.1 Model quality objectives in the AQD

The model quality objective described in the Air Quality Directive (AQD) along with the monitoring quality objectives, are given as a relative uncertainty (%). The AQD defines the modelling uncertainty as "*the maximum deviation of the measured and calculated concentration levels for 90 % of individual monitoring points, over the period considered, by the limit value (or target value in the case of ozone), without taking into account the timing of the events. The uncertainty for modelling shall be interpreted as being applicable in the region of the appropriate limit value (or target value in the case of ozone). The fixed measurements that have to be selected for comparison with modelling results shall be representative of the scale covered by the model*."

According to this definition, the AQD states that the uncertainty will be determined from the maximum of 90% of the available monitoring stations. Furthermore, it will be computed "without considering the timing of the event", which means that any temporal correspondence between modelled and observed values will be disregarded. The statistical indicator currently recommended for estimating modelling uncertainty is the Relative Directive Error (RDE) (Denby, 2010). It is mathematically defined at a single station as follows:

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

where OL_V is the observed concentration closest to the limit value concentration (LV) and M_{LV} is the correspondingly ranked modelled concentration. The maximum of this value found at 90% of the available stations is then the Maximum Relative Directive Error (MRDE). This formulation is similar to the one recommended (Stern and Flemming, 2004) called the Relative Percentile Error (RPE), which is defined at a single station as:

$$RPE = \frac{|O_P - M_P|}{P}$$

where O_P and M_P are the observed and modelled concentrations at the percentile (P), used to define the exceedance percentile.

5.2 Evaluation of PM models

In the literature, several intercomparison and evaluation exercises are reported for PM models : McKeen et al., 2007; Smyth et al., 2009; Stern et al., 2008; Hayami et al., 2008, Solazzo et al., 2012, Vautard et al, 2009 ; Vautard et al., 2007 ; Pernigotti, et al., 2013. Most of these model intercomparison exercises were performed at the regional scale with chemistry transport models.

In the most recent exercise, AQMEII (Solazzo et al., 2012), there was a clear tendency for models to underestimate PM_{10} concentrations in US and EU regions for background stations. Model results for $PM_{2.5}$ concentrations showed better performances but large uncertainty remains in the simulation of secondary organic aerosols schemes. Usually, the error statistics reported in Table 4 are classically used to assess the models. Discrepancies were attributed to meteorology and emissions.

Mean Bias	$(\bar{M} - \bar{O})$ with $\bar{M} = \frac{1}{N}\sum_{i=1}^{N} M_i$ and $\bar{O} = \frac{1}{N}\sum_{i=1}^{N} O_i$
Normalised Mean Bias	$NMB = (\bar{M} - \bar{O})/\bar{O}$
Root Mean Square Error	$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(M_i - O_i)^2}$
Correlation Coefficient	$R = \left(\sum_{i=1}^{N}(M_i - \bar{M})(O_i - \bar{O})\right) \Big/ \left(\sum_{i=1}^{N}(M_i - \bar{M})^2 \times \sum_{i=1}^{N}(O_i - O)^2\right)$
Centred Root Mean Square Error	$CRMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(M_i - \bar{M})(O_i - \bar{O})}$
Normalised Mean Standard Deviation	$NMSD = (\sigma_M - \sigma_O)/\sigma_O$ with $\sigma_X = \frac{1}{N}\sum_{i=1}^{N}(X_i - \bar{X})^2$
Mean Fractional Bias	$MFB = \frac{1}{N}\sum_{i=1}^{N}\frac{M_i - O_i}{(M_i + O_i)/2}$
Mean Fractional Error	$MFE = \frac{1}{N}\sum_{i=1}^{N}\frac{\lvert M_i - O_i\rvert}{(M_i + O_i)/2}$

Table 4: Usual error statistics to evaluate models (M and O refers respectively with Model and observations data, and N is the number of observations)

In the last decades, the so called Taylor diagrams have been used to summarize various statistical parameters as shown in Figure 24 in the AQMEII intercomparison exercise.

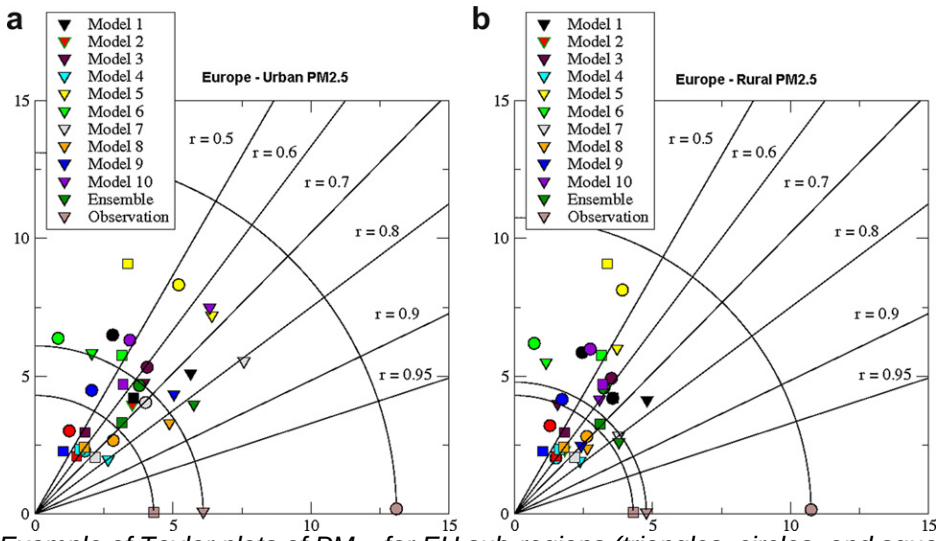

Figure 24: Example of Taylor plots of $PM_{2.5}$ for EU sub-regions (triangles, circles, and squares for sub-regions 1, 2, and 3, respectively) at a) urban and b) rural sites. Horizontal and vertical axes indicate the variance (μg/m3) and the arcs represent the curves of ideal variance for each sub-region (from Solazzo et al., 2012)

Since 2003, the EURODELTA exercises launched under the EMEP program, propose an on-going evaluation of model performances of chemistry transport models involved in the policy making process (Vautard et al., 2009). The current phase of EURODELTA provides an overview of PM model performances (with PM components) over four 1-month campaigns with common input datasets (emissions, meteorology and boundary conditions), model grids. The performance of the meteorology was also assessed. We can observe in Figure 25 the large variability of model results for PM_{10} even with common input data. This variability is mainly driven by the way natural emissions of PM (dust and sea salts) are handled in the models.

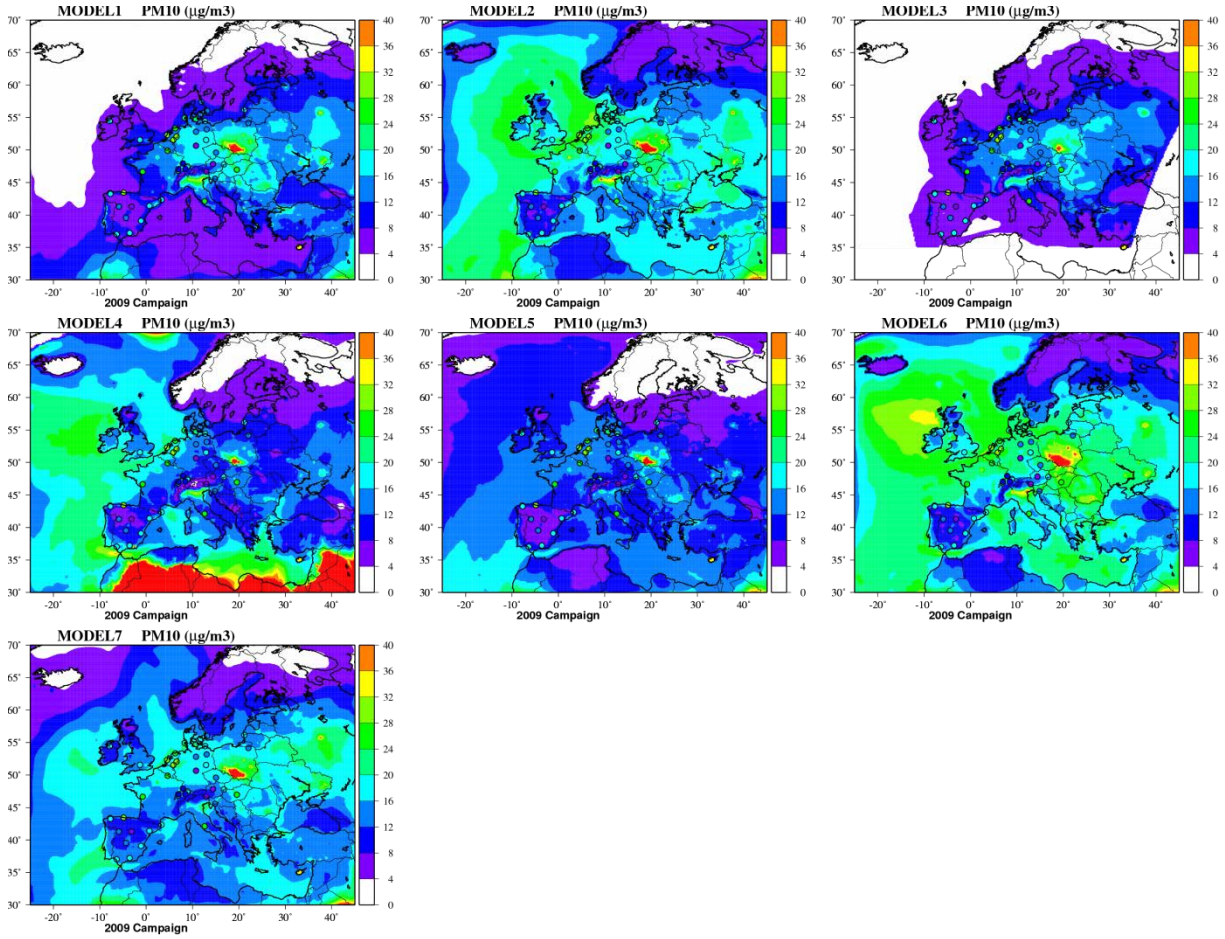

Figure 25: Mean PM_{10} model concentrations for seven models in the EURODELTA exercise for the 25/02 – 26/03/2009 periods (Observations are the coloured circles). In these results, MODEL1, 2, 4, 5, 6 uses the same input data.

In this exercise, the DELTA TOOL (Thunis et al., 2013, Thunis et al., 2012a, b; Pernigotti, et al., 2013) was used to assess the model performances with new criteria developed in the frame of the FAIRMODE Sub Group 4 ("model benchmarking"). One of the last improvements is the use of observation uncertainties as a new parameter in the criteria calculation for the evaluation.

5.3 The DELTA tool

DELTA is an Interactive Data Language (IDL) based model evaluation software using paired data of modelled and observed surface data (Thunis et al., 2012b: http://aqm.jrc.ec.europa.eu/DELTA/). It is designed for a rapid diagnostic of model performances in applications related to the implementation of the EU's AQD (EC, 2008). It therefore focuses on annual air quality assessments, dealing with three main pollutants (O_3, NO_2 and PM) for which EU Member States have the obligation of reporting. Planning activities (evaluation of emission scenarios) aimed at reducing air pollution will also be considered in the future[26]. The analysis module allows various statistical indicators to be generated, and their graphical visualisation in a variety of diagrams (bar plots, scatter plots, time series, Taylor, Target, Bugle, Soccer etc). The graphic user interface in DELTA facilitates the application of the tool even by less experienced modellers. DELTA offers two modes of use: exploration and benchmarking. In the exploration mode the user is free to select various time averaging, different pollutants, a selection of stations and the time period

[26] See FAIRMODE initiative: http://fairmode.ew.eea.europa.eu/.

on which model performances are evaluated. All these choices are performed through a user-friendly graphical interface, which allows switching straightforwardly from one diagram representation to another while keeping the same data selection. The tool offers the possibility of performing comparisons between model versions and/or between parameters and/or between monitoring stations (Figure 26). Various combinations of input selection options (parameters, stations, models etc.) could be used on the same diagram. Although the current version of DELTA does not make use of gridded model data, some conclusions on the spatial variability of the statistical indicators can be drawn from the comparison of station statistics grouped in categories (e.g. urban versus rural or belonging to different administrative region). The user can freely define the categories in which stations must be organized. In the benchmarking mode, DELTA produces performance reports in a standardized format structured around a set of selected statistical indicators. This should lead to a more harmonised way of looking at performances when this tool becomes more widely used. As the model uses pairs of model and observation, all model types can be assessed by this tool.

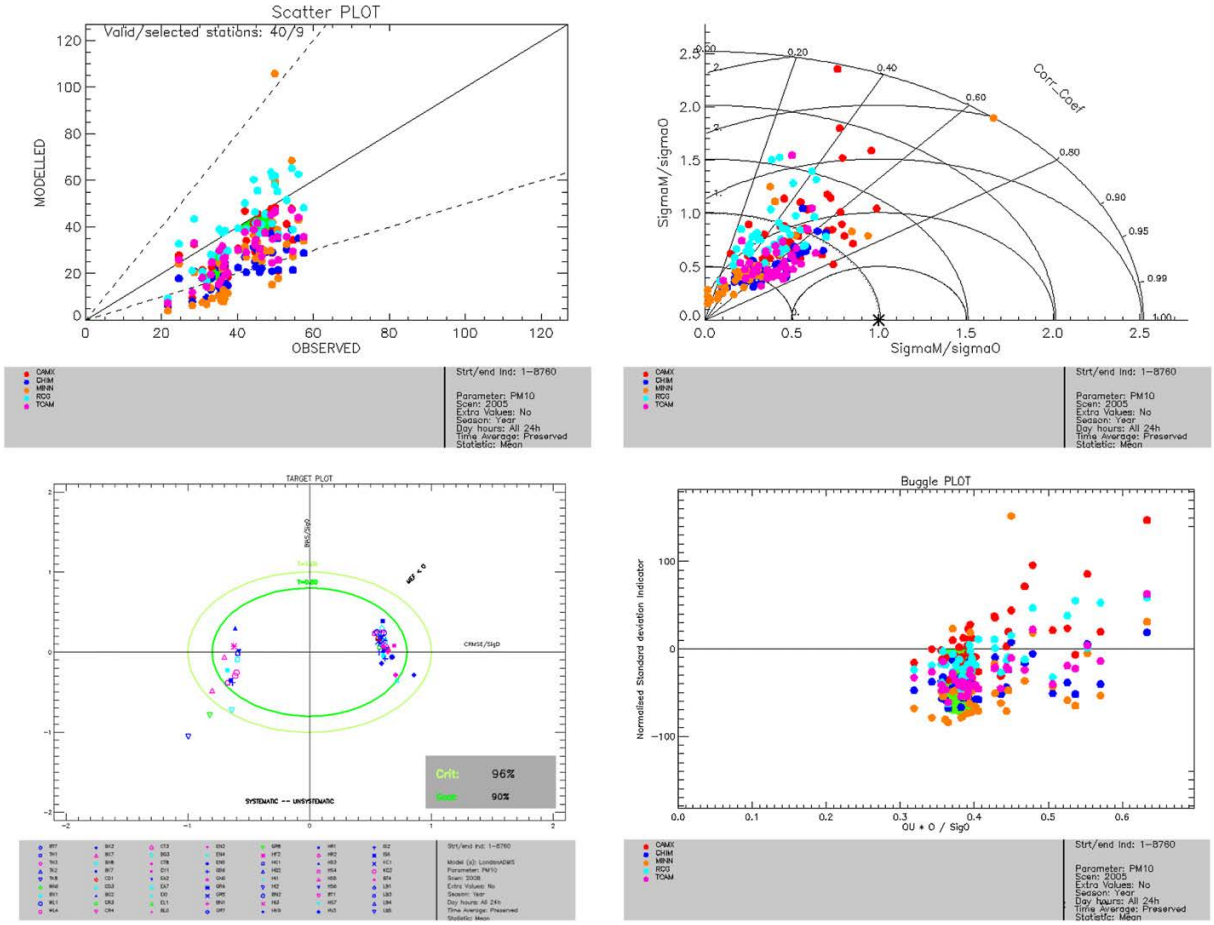

Figure 26: Examples of multi-dimensional diagrams for annual PM_{10} concentration obtained with DELTA by selecting 5 models results (MINNI, CHIMERE, TCAM, CAMX and RCG) and 61 monitoring sites in the Po Valley (Italy). Top left: scatter plot; top right: Taylor diagram; bottom left: Target diagram; bottom right: Bugle plot (from Thunis et al., 2012b).

6 PM modelling applications for the AQ Directive (AQD)

As previously mentioned, many zones and agglomerations in EU MS have been in exceedance of the limit values for PM_{10}. Pursuant to Article 22 of the AQD, MS had the possibility to notify the Commission when, in their opinion, the conditions were met in a given zone or agglomeration for being exempt from the limit values for PM_{10} (in force from the 1st January 2005). A time extension for compliance until the 1st June 2011 could be requested by MS provided they proved that:

- all appropriate measures were taken at national, regional and local levels to meet the deadline for the limit values, i.e. 1 January 2005, and that limit values had not been achieved because of the presence of one or more of the following elements: site-specific dispersion characteristics, adverse climatic conditions or transboundary contributions;
- Compliance with the limit values will be achieved at the expiry of the exemption period.

All the MS submitted a notification for exemption regarding the PM limit values to the European Commission (EC), and all, except the Netherlands, received some objections from the Commission:
(http://ec.europa.eu/environment/air/quality/legislation/time_extensions.htm).

Objections were related to insufficient information about:

- the measures taken at the initial attainment date,
- the measures to be taken for future compliance at the end of the time extension,
- the justification of the role played by the specific conditions for the exemption, i.e. site-specific dispersion characteristics, adverse climatic conditions or transboundary contributions.

In this legally binding process, negotiations between the concerned MS and the EC still continue. Modelling has been used by a large number of countries to apply for these time extensions, covering most of the relevant model applications for the AQD including:

- air quality and PM exceedance assessment,
- source apportionment studies, and
- planning and emission scenario analyses (see also de Smet et al., 2013).

These applications still remain a major activity for the problematic AQ management zones. Both national and local authorities in the countries still improve their action plans and the associated evaluation they provide to the EC. In this perspective modelling has played, and continues to play, a decisive role. This possible use of modelling, in policy application areas, was discussed in section 3, and practical recommendations are given below.

6.1 Modelling requirements for assessment

6.1.1 The AQ Directive requirements

According to the AQD, and whether they are engaged in the extension process discussed above or not, the EU MS have to report on the state of AQ in assessment zones that cover the country area, and they have to describe precisely the exceedances of limit and target values. The MS have to provide the EC with a good description of these situations, which includes at least a quantification of the area and population exposed. In the FAIRMODE

technical reference guide for AQ modelling (EEA, 2011) most of the aspects in regard to assessment and reporting of exceedances are presented and discussed.

In the current AQD (EC, 2008) ***modelling tools alone can only be used for reporting non-exceedance situations.*** This is when the concentrations are below the lower assessment thresholds, as defined in the AQD (EC, 2008, Appendix II). When concentrations are above the lower assessment threshold, but below the upper assessment threshold, then modelling alone is not sufficient and fixed measurements are also required (even if the number of measurement sites can be decreased where reliable modelling techniques are implemented). Above the upper threshold fixed measurements must be used as basis for the assessment and modelling may be used as supplementary material. Threshold values for PM are summarised below.

Lower assessment thresholds:

- For 24-hours average PM_{10} concentrations: 50% of the limit value (25 $\mu g/m^3$ must not be exceeded more than 35 times a year)
- For annual average PM_{10}: 50% of the limit value (20 $\mu g/m^3$)
- For annual $PM_{2.5}$ average: 50% of the limit value (12 $\mu g/m^3$)

Upper assessment thresholds:

- For 24-hours average PM_{10} concentrations: 70% of the limit value (35 $\mu g/m^3$ must not be exceeded more than 35 times a year)
- For annual average PM_{10}: 50% of the limit value (28 $\mu g/m^3$)
- For annual $PM_{2.5}$ average: 50% of the limit value (17 $\mu g/m^3$)

6.1.2 Use of models for assessment

When models are implemented for the assessment, a quality objective (regarding the results) is set to control uncertainty. For ***annual averages model uncertainty cannot exceed 50%***. Current models are more and more reliable. Several regional model intercomparisons and evaluation exercises (Cuvelier, 2007 ; Vautard, 2007) and more recent results from the new EURODELTA3 and COPERNICUS/MACC projects (see section 5) showed progress to reach this objective. Current regional models give satisfactory responses except in some areas with complex geographical terrain and the annual quality objective is generally fulfilled. At the smaller scale (city, street) model results are mainly driven by the emission inventory. Therefore where high resolution emissions inventories are available the objective is generally reached as well.

For ***hourly values and exceedances, no quality objective is set yet***. It is acknowledged that 50% can be challenging in many situations, especially because PM concentrations are highly sensitive to fluctuations in emissions (see chapter 4). Such difficulties can be overcome implementing data assimilation processes that help in correcting simulations with observations. Even simple procedures based on optimal interpolation and krigging give impressive and reliable results for assessment (see also de Smet et al., 2013).

The data assimilation processes for regional AQ assessment and their performances are extensively addressed within the Copernicus/MACC project that aims at developing the future Copernicus atmosphere services (http://atmosphere.Copernicus.eu/). In particular the yearly AQ assessment reports published by the project for the years 2007 to 2010 (2011 to be published by end of 2013) present the satisfactory performances of chemistry-transport models used together with data assimilation processes to map PM background concentrations in the European countries. The added-value of the Copernicus/MACC project is to bear the implementation of sophisticated and complex data assimilation chains that can

take advantage of various kinds of observation data (in-situ and satellite) to improve assessment[27] (e.g. Schneider et al., 2013)

For country-wide assessments, approaches based on kriging techniques and geostatistical analyses that have been largely evaluated by the EEA's European Topic Center on Air Pollution and Climate Change Mitigation (de Smet et al, 2009; de Smet et al, 2010); (Denby et al, 2011a); (Denby et al, 2011b) (Malherbe et al, 2009), are definitively efficient and relevant. Annex 4 gives an overview of the basic principles of such techniques and of the state of the art.

For determining limit value exceedances and their geographical distribution at both regional and urban scales, it is recommended to use deterministic models that will help to catch air pollution patterns. In that perspective, data assimilation or statistical approaches based on the analysis of historical datasets and systematic model biases are essential to correct the PM_{10} maps that have been derived from models. Note that to be consistent between the site representativeness area and the model resolution, traffic sites exceedances can only be considered by local and urban scale models. Ancillary data describing the station environment can be assimilated to improve locally the results (Denby et al, 2008). As a last point, the gradient between PM concentrations in regional background areas and at urban or traffic sites is generally much lower than for NO_2 concentrations. Therefore a good representation of background concentration levels is determinant whatever the scale, to provide good assessment maps.

Local situations where high daily values are recorded compared to background urban concentrations, are generally characterized by high contributions of re-suspension (the so-called "indirect traffic emissions") or construction/demolition work emissions (Amato, 2009), (Martuzevicius, 2011). Identification of areas where this kind of emissions can influence significantly PM levels is essential, and methodologies to assess emission levels continue to be developed (see for instance guidance provided by some US cities http://www.airquality.org/ceqa/index.shtml, http://www.dec.ny.gov/chemical/8912.html). Local measurements can help for the evaluation of the process.

For assessment and reporting PM values, recommendations are listed in section 6.1.3.

6.1.3 Application guidance for mapping and air quality assessment

1. Air AQ modelling requires a well-established and comprehensive emissions inventory, fitted to the geographical domain covered and the model (deterministic, geostatistical, or statistical) resolution.
2. The use of regional air quality model results combined with monitoring (with data assimilation or kriging approaches) is recommended to produce maps of background PM concentrations and exceedances. Kriging methods and/or optimal interpolation generally give very satisfactory results for a reduced computational cost.
3. A relevant set of monitoring stations must be used for data assimilation to correct model results and for mapping. This set should be representative of the targeted geographical scale, should provide significant historical data, and the associated metadata must be well-documented.
4. Improve assessment at the urban and local scales using appropriate models (see Annex 4) with accurate emission data; systematic biases of those models detected in step 1 can be corrected using statistical approaches[28].

[27] One should note that data results related to background concentrations are freely available from the service

[28] Generally those methods are based on an in-depth analysis of the historical behavior of the model to reproduce PM levels. Systematic biases when compared to the observations are established and corrected on the modeling results. Such approaches are successfully used in forecasting sciences.

5. Some indication of the uncertainty in the simulations (whatever the scale and the type of model) should be given. If necessary multiple runs reflecting the emission and model uncertainties may be made to indicate these uncertainties.

6.2 Modelling requirements for reporting exceedances of limit and target values

The mapping techniques described in the previous section apply for reporting exceedances of limit and target values. In the next sections, specific topics that relate to the occurrence of PM episodes when the PM limit values are likely to be exceeded are discussed.

6.2.1 Explaining exceedances due to natural events

According to the article 20 of the AQD Directive (EC, 2008), MS can transmit to the Commission the list of zones and agglomerations where some exceedances of the PM_{10} limit values are attributable to natural events. The most common situations relate to PM concentrations that can be influenced by dust events and sea-salt contributions in coastal areas. The EEA published in 2012 a Technical Report (EEA, 2012a) devoted to how to account for natural sources under the AQD. A number of data sources and methodological recommendations are reported in this document with an analysis of the practises in the concerned MS.

Mircea et al. (2008) and Mitsakou et al. (2008) presented two relevant examples where models were used to help identify and quantify the contribution of wind-blown dust from the Sahara. For dust emissions inside Europe, Vautard et al., (2005) showed that when applying a simplified approach towards wind-blown dust emissions within a chemistry-transport model the underestimation of modelled background PM concentrations compared to measurement results can partly be explained and corrected. The approach, statistically validated, uses meteorological parameters such as surface humidity and wind speed to give a more realistic picture of particulate matter levels in Europe.

By using a similar approach, Bessagnet et al. (2008) predicted a sudden and extensive dust event observed in Central Europe at the end of March 2007 (Birmili et al., 2008). PM peaks observed in Europe during this event could be attributed to long-range natural dust transport from the Ukrainian agricultural soils (Bessagnet et al, 2008) up to Western Europe (Figure 27).

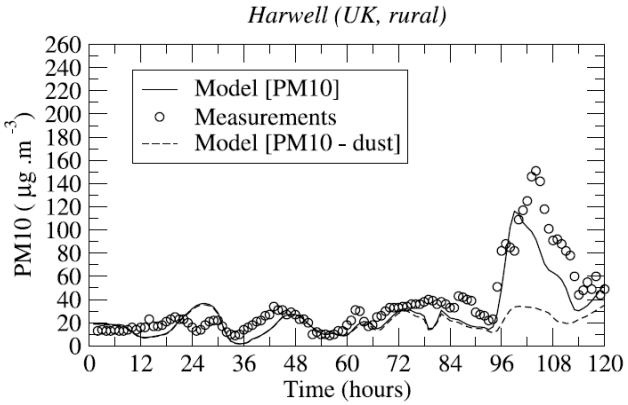

Figure 27: Contribution of dust in Harwell (rural station in UK) during the Ukraine dust studied in Bessagnet et al. (2008) and Birmili et al. (2008)

However, dust events and natural contributions to PM episodes remain difficult to predict and simulate because annual emission inventories cannot include them a priori. Observation and especially Earth observation are essential to monitor dust plumes and to adapt a posterior emission inventories to the episode period.

The Copernicus/MACC project proposes as well a service targeted to the evaluation and prediction of dust contribution thanks to the interpretation of satellite measurements (http://www.gmes-atmosphere.eu/d/services/gac/nrt/nrt_opticaldepth!36!Dust!Global!macc!od!enfo!nrt_opticaldepth!2013020400!!/). This is a valuable data source for boundary conditions that can improve regional or urban model results accounting for long range transport of desert dust.
Generally catching natural contribution in modelled PM concentrations requests use of observation data to correct emission inventories that cannot reproduce the spatio-temporal variability of such contributions. This step is clearly integrated in an application guidance given in section 6.2.2 below.

6.2.2 Application guidance to account for natural contribution in exceedances:

1. Assessing the natural contribution for reporting exceedance of the PM limit values requires regional modelling results (generally they result from long range transport), in combination with measurement data;
2. A systematic analysis of discrepancies between measurement data and modelling results should be conducted together with the analysis of meteorological data and backward trajectories to confirm the potential contribution of natural sources;
3. Satellite information might be used to support the evaluation, e.g. by checking daily the information available from the Copernicus/MACC services;
4. If a MS implements stations where mineral compounds are measured, they should be considered to quantify natural contribution to PM (rather PM_{10}) concentrations;
5. Correction of the model results can be performed using data assimilation or statistical techniques, using observed PM_{10} mass concentrations.

Within the continent, sea salt concentrations are usually below 0.5 µg/m^3. They can reach 2 to 3 µg/m^3 with highest values in wintertime. For cities located in coastal regions subtracting such values can have an impact on exceedances of the PM limit values set in the AQD (EC, 2008; EEA, 2012a). For the Netherlands, Manders et al. (2009) suggests that most of the exceedances of the daily PM limit value in Rotterdam would be below the limit value after subtracting sea salt contributions.

6.2.3 Reporting exceedance of PM limit values by winter sanding or salting of roads

According to the article 21 of the AQD (EC, 2008), MS can transmit to the Commission the list of zones and agglomerations where some exceedances of the limit values are attributable to sanding or salting road operations in winter.. Reporting on such situations requires an assessment of the associated emissions. This issue is discussed in the 'Emissions' chapter (see sections 3.4.2 and 3.5).

6.2.4 Explaining exceedances due to transboudary pollution

According to the article 25 of the AQD (EC, 2008), MS "*where any alert threshold, limit value or target value plus any relevant margin of tolerance or long-term objective is exceeded due to significant transboundary transport of air pollutants or their precursors, the Member States concerned shall cooperate* (…)".

Assessing transboundary contributions to PM exceedances that occur in European cities is a difficult task because they are not only driven by foreign emissions but also by atmospheric chemistry. A large part of PM issued from long range transport results from chemical transformation. Therefore, regional chemistry-transport models are the best fitted for the

purpose of transboundary fluxes analysis. A straightforward approach consists of running the models cancelling emissions from the target area country. It requires operational runs of CTM and can be computationally expensive. However, within the development of the Copernicus/MACC services, new products which assess foreign contributions during PM episodes situations will be developed and proposed to the MS by the end of 2014 (http://www.gmes-atmosphere.eu/services/aqac/policy_interface/source_allocation/?op=get).

6.3 Modelling requirements for source apportionment

One of the key issues for the decision maker is: **on which sectors should one decide stringent control measures to avoid exceedances and to reduce population exposure?** PM and their precursors' sources are multiple and their relative contribution in a given situation can vary with the period of the year and the location.

Aerosol particles are produced both by direct emissions of primary particulate matter as well as through the chemical transformation of several gas phase precursors, related to a considerable number of both anthropogenic and natural sources extending over regional scale areas (Seinfeld and Pandis, 2006). As a consequence, assessing the contribution of the different emission sources to the particulate matter concentration (the so-called source apportionment) is undoubtedly a crucial issue, needing robust and suitable approaches.

Source apportionment evaluations can be based on the analysis of observed data by means of several approaches such as: chemical balance modelling, statistical analysis (Larsen et al., 2012; Perrone et al., 2012) and tracer methods (Lukács et al., 2007; Querol et al., 2004) as well as by the application of modelling techniques.

Widely-applied modelling techniques are usually based on two possible approaches: Brute Force, or Direct Method and Zero-Out Modelling (Yarwood et al. 2004). Both techniques are, actually, more sensitivity analysis methods than source apportionment algorithms; however both of them can be applied to evaluate source contributions. Particularly, the Brute Force Zero-out method (Burr et al, 2011; Koo et al, 2009) estimates first-order sensitivity coefficients of the concentration variation as a response to small changes in emission values. Repeating such analysis to all the interested sources allows the relative weight of each emitting source to be quantified. The second and probably most applied approach is based on removing (i.e. setting to 0) the source of interest from the input set and then rerunning the model. The contribution of the source is then computed as a difference from the base case (i.e. with all sources).

Both methods are easy to implement but they feature two relevant limitations:

a) they are computationally expensive as they require a new simulation for each source to be evaluated;
b) in the case of non-linear systems (e.g. secondary pollutants) they do not conserve the mass, that is to say the sum of the computed impacts over all sources does not equal the total mass concentration.

To overcome these problems that can seriously undermine the effectiveness of the performed analysis, a different approach can be considered, usually referred as Reactive Tracers (or Tagged Species) method (Dunker et al., 2002; Zhang et al., 2005). The basic idea underlying this approach is the introduction of additional species (particularly, a set of species for each source to be apportioned) able to track the contribution of specific sources from the emission to final PM concentration, taking into account the same physical and chemical processes of the usual modelled variables. The approach relies on the information being available to split the original emission inventory. In general, a single tracer can track primary PM species whereas secondary particulate species require several tracers to track

the relationship between gaseous precursors and the resulting PM. A key point in this approach is that adopted algorithms for reactive tracers should ensure that mass is properly conserved but, at the same time, limiting as much as possible the additional request of computational resources.

In Annex 5 examples of source apportionment methodologies and results are given for Italy and the Lombardia region, and the UK.

6.4 Modelling requirements for planning and emission scenarios

6.4.1 General concepts

This is an obvious goal of modelling. Once the model is considered as reliable enough to reproduce current and past situations or has learnt enough from past situations (in case of statistical models) it can be applied to simulate future ones. Decision makers need to know the potential impact of the emission control strategies that they decide at the European or the local scales. Indeed, given information on emission factors and activities of various emission sectors it is possible to adapt and alter the total emissions through a range of assumed policy measures, that either affect the emission factors themselves (technological) or the activities (structural). A policy maker may come with a range of possible strategies that will need to be quantifiably assessed using air quality models. Or an AQ modeller may come with an emission requirement for which a policy maker will need to find a way of implementing it. In either case, the AQ model is applied, usually through multiple model runs, calculating the sensitivity of the modelled concentrations to a range of emission scenarios. How this is carried out depends on the requirements for any particular application.

Some applications are straight forward and related generally to primary PM compounds that are only transported through dispersion processes. They generally apply to local situations. In these cases dispersion models based on transport and diffusion equations can reproduce changes in concentrations due to changes in emissions. Street canyon models that target local exceedances, or Gaussian approaches adapted to the city scale or even Lagrangian or Eulerian models (see Annex 4) can be run with “appropriate” (in the sense that they are accurate enough) local emission inventories. Such inventories should discriminate properly the activity data from various sectors. Modelling uncertainty is directly correlated to emission uncertainties.

Other applications are more complex because the PM concentrations result from chemical processes that involve a large number of precursors and complex chemical relationships depending on meteorological parameters. PM pollution at regional scales or over large agglomerations relates to such phenomena and should be predicted with appropriate chemistry transport models (see Annex 3). As for ozone, those models should be able to simulate correctly the chemical regimes that stand in the targeted areas to ensure that relevant responses to emission reductions are calculated.

Modelling responses to emission reduction scenarios are strongly dependant on the meteorological fields that drive the simulation. Focussing on a given year can limit the assessment neclecting the effect of the meteorological variability. Therefore, it is recommended to simulate average concentrations over several years. The computational need can be high. If only one year is chosen for limiting costs, special care will be accorded to this choice: whether a “mean” year or an “extreme” one should be preferred will depend on the application. The following section 6.4.2 suggests some guidance.

6.4.2 Application guidance

1. Scenario calculations should be preferably carried out for a range of meteorological years, which also reflect the variability in PM background concentrations, rather than

just one single year. This will also provide information on the variability and uncertainty in the impact of the emission scenario.
2. If the geographical domain can be influenced by chemical processes it is essential to run chemistry transport models with appropriate chemical schemes.
3. In relation to the chemical schemes, special attention should be given to the emission inventory that feeds the model, to the description and quantification of the emission of precursor compounds and of their spatio-temporal variability.
4. Some indication of the uncertainty in the predictions should be given, e.g.. multiple runs reflecting the emission and model uncertainties may be made to indicate these uncertainties.

6.5 Modelling requirements for information and forecasting

6.5.1 General overview

Short term forecasting by applying models is a capacity that can be used by a Member State or local authorities to deal with their information duty. According to article 26 of the AQD (EC, 2008) and Annex XVI of the directive, they have to inform the general public about the occurrence of an air pollution episode and its evolution. Article 24 stipulates as well that short term action plans should be implemented if the risk that an episode develops is high. Forecasting capacities should be developed to facilitate access to such information and can help in raising public awareness. It should facilitate the management of an air pollution situation as well, anticipating future evolution. Forecasts are, by definition, delivered by modelling systems (deterministic or statistical) that should be sufficiently robust and reliable. Currently a number of PM forecasting systems are run throughout Europe at the national and urban scales.

Two types of forecast systems exist, both based on the use of meteorological data and chemistry models. They are built on “statistical” or “deterministic” methods. They can apply to both the local and regional scales. Essentially because of computer efficiency reasons and lack of maturity of deterministic models, efforts first focussed on the development and use of statistical models during the past decades (Karatzas et al, 2002; (Sun et al, 2008; Neto et al, 2009). Since they were easy to use and implement, simplified physics and chemistry equations systems were developed and tuned in order to estimate the probability of a pollution event. This is achieved by using only meteorological parameters such as mean wind speed, solar radiation and temperature (Hrust et al., 2009).

Using only a few equations or statistical relationships, statistical models allowed a fast computation (a major constraint for forecast systems). They were widely used for many years and showed satisfactory results, at least for ozone daily maxima (Zeldin and Thomas, 1975; Simpson and Layton, 1983; Robeson and Steyn, 1990). On the other hand they were rather crude and uncertain (no boundary conditions for example). are meanwhile used for air quality short term forecasts (Kolehmainen et al., 2001; Chaloulakou et al., 2003; Ibarra-Berastegi et al., 2008; Pfeiffer et al., 2009) and health impacts (Schlink et al., 2006). All these approaches are well-suited for urban issues because generally this kind of area is well-covered by measurement networks. These approaches are reviewed in a guidance which the US-EPA published in 2003 (http://www.epa.gov/airnow/aq_forecasting_guidance-1016.pdf) on forecasting approaches for predicting ozone and $PM_{2.5}$ concentrations.

Nowadays, deterministic models are available at both urban and regional scales for air quality forecasting applications (see e.g. the FUMAPEX project, Baklanov et al, 2007). At the urban scale, Gaussian, Lagrangian or even Eulerian approaches were developed. Considering PM concentrations, they generally focus on the transport of primary PM, what can be acceptable at the urban scale. For the regional scale chemical processes drive the formation of secondary aerosols that contribute significantly to PM mass concentrations.

Therefore it is no longer possible to ignore the complex chemistry that influences PM background concentrations and chemistry-transport models should be run.

Many air quality forecasting platforms can be found in Europe, some being national institutional systems (PREV'AIR, CALIOPE). The others are operated in research laboratories without operational constraints (7/24h). The PREV'AIR (www.prevair.org) system is a French national initiative (Honore et al., 2008; Rouïl et al., 2009) which is based on a partnership between French research institutes (CNRS and INERIS), and the national meteorological centre (Météo-France), acting under the aegis of the French ministry in charge of Ecology. PREV'AIR produces and provides forecast results every day of the year up to two days ahead, as an operational meteorological centre. The CALIOPE administrative structure (http://www.bsc.es/caliope) merges universities and governmental authorities: The Barcelona Supercomputing Center (BSC), the Spanish National Research Centre for Energy, Environment and Technology (CIEMAT), the Earth Sciences Institute "Jaume Almera" (IJA-CSIC) and the Mediterranean Center for Environmental Studies Foundation (CEAM) CALIOPE are on their way of becoming operational.

Table 5 below lists some of the forecasting systems currently running in Europe.

System (URL)	Country	METEO/CTM	References
CALIOPE (www.bsc.es/caliope)	Spain	WRF/CMAQ	Pay et al. (2012)
OPANA (artico.lma.fi.upm.es)	Spain	MM5/CMAQ	Cooter and Hutzell (2002)
(forecast.uoa.gr)	Greece	NCEP/SKIRON-CAMx	Kallos et al. (2007)
FARM (www.aria-net.eu/QualeAria)	Italy	RAMS/FARM	Zanini et al. (2005)
CETEMPS (pumpkin.aquila.infn.it/forechem/)	Italy	MM5/CHIMERE	Curci et al. (2008)
CHIMERE-DUST (www.lmd.polytechnique.fr/dust)	France	MM5/CHIMERE	Menut et al. (2009)
PREV'AIR (www.prevair.org)	France	MM5/CHIMERE-ARPEGE/MOGAG	Rouïl et al. (2009)
(http://cerea.enpc.fr/en/prevision.html)	France	WRF/Polair3D	Garaud et al (2010)
COSY (www.lmd.polytechnique.fr/cosy)	France	WRF/CHIMERE	Vautard et al. (2001)
PREVISAO-QAR (www.dao.ua.pt/gemac/previsaoqar)	Portugal	MM5/CHIMERE	Monteiro et al. (2005)
TRUMF (www.trumf.de)	Germany	COSMOS/RCG	Stern et al. (2003)
(www.lml.rivm.nl)	Netherlands	LOTOS-EUROS	Schaap et al. (2008)
THOR (thor.dmu.dk)	Denmark	ETA-MM5/DEHM	Frohn and Brandt (2006)
UK AQ forecast (http://uk-air.defra.gov.uk/forecasting/)	UK	WRF/CMAQ	http://uk-air.defra.gov.uk/library/
MACC II (http://www.gmes-atmosphere.eu/services/raq/raq_nrt/)	EU	IFS/CHIMERE-EMEP-EURAD-MATCH-LOTOS/EUROS-MOCAGE-SILAM	

Table 5: List of forecasting system in Europe

6.5.2 Forecasting products "PM oriented" in Copernicus/MACC

The Copernicus/MACC-II project has the overall functional objective of delivering reliable operational air quality forecasting products and information services at the European scale that support research, European environmental policy and the ongoing development of user-specific local services, like zooms over cities or parts of cities, smartphone alerts etc.. Copernicus/MACC-II promises to supply "aerosol" products (http://www.gmes-atmosphere.eu/services/aqac/) *inter alia*:

- Monitoring of the global distributions of greenhouse gases, reactive gases and aerosols through assimilations of satellite and in situ observations, using NRT, delayed-mode and reanalysis;
- Twice-daily forecasts of the global distributions of reactive gases and aerosols for several days ahead;
- Boundary values for regional modelling of troposphere and stratospheric chemistry, and local and urban modelling for air quality (gases and aerosols);
- Analyses and forecasts for the European domain based on an ensemble approach using multiple regional air-quality models;
- Air quality re-analyses for the European domain based on an ensemble approach using multiple regional air-quality models;
- Global fire analyses and estimates of emissions from fires for use in the global and European regional monitoring and forecasting systems;
- Surface fluxes of carbon dioxide, methane and aerosols produced using inverse methods;

In addition, Copernicus/MACC-II intends to respond promptly when needed to supply specific products related to major events involving atmospheric constituents such as volcanic ash and pollutants from major fires, especially in cases of particular importance to the European Union.

For air quality issues (forecasts, analyses and re-analyses of ozone, PM_{10}, $PM_{2.5}$ and nitrogen dioxide concentration fields), the MACC-II services will focus on the European scale with the provision of simulations with a 10 km^2 resolution in 2014 (the current resolution of the involved models ranges from 15 to 25 km^2). The ensemble approach is very promising with regard to reducing and controlling uncertainties and offers the most advanced air quality forecasts over Europe. With a 10 km^2 resolution, the MS which do not develop their own air quality forecasting system are encouraged to use those predictions to get an overview of expected air pollutant concentrations and to develop activities related to short term action plans or public awareness.

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Annex 1

Aerosol Physics and Chemistry modelling

In this chapter some basic concepts related to PM modelling will be detailed. In particular, the processes that should be parameterised in “up-to-date” models and how they are generally taken into account will be explained.

Aerosol microphysics

It is known that the mass concentration is governed by large particles, whereas the number concentration is governed by ultra fine particles. Current air quality models usually focus on the evolution of the mass concentration of particles, which is consistent with existing air quality regulations. As an example, the CHIMERE and Polair3d/Polyphemus chemistry transport models (CTM) (Bessagnet et al., 2004; Sartelet et al., 2007) only take into account particles with diameters respectively greater than 40 nm and 10 nm and solve the aerosol general dynamic equation for the mass concentration. Some air quality models simulate also the evolution of the number concentration; however, comparing their results with ambient measurements suggests that they are not as accurate for the number concentration as they are for the mass concentration (e.g., Zhang et al., 2006, 2010). The dynamics of particles is governed by several processes, which are condensation/evaporation, coagulation, nucleation and deposition. In this study, we focus on the condensation/evaporation process, which is usually considered to be the most challenging one due to the thermodynamics involved (gas/particle equilibrium, Kelvin effect) and its numerical issues.

There are three major representations of the particle size distribution (PSD) in air quality models: continuous, sectional and modal. The continuous distribution is the most accurate representation (Debry and Sportisse, 2007), but it can be unstable when too few dicretization points are used in three-dimensional (3D) applications. In the modal representation, the PSD is modelled by several lognormal distributions, also called modes. Usually, the modes are: the Aitken nuclei mode, the accumulation mode and the coarse mode (Binkowski and Shankar, 1995; Sartelet et al., 2006). The accuracy of this approach is limited by the number of modes. The modal aerosol model (MAM, Sartelet et al., 2006) is used and discussed in this study.

In the sectional representation, the particle size spectrum is divided into a finite number of sections (or bins), and the PSD is approximated by the integrated number, surface, mass or volume concentrations over each section, depending on the particle characteristics of interest (Gelbard et al., 1980; Jacobson and Turco, 1995; Seigneur, 1982; Seigneur et al., 1986; Zhang et al., 1999). The sections are internally mixed, i.e., particles in a section have the same chemical composition and the same representative diameter. The accuracy of a 2 sectional model depends strongly on the number of sections used to solve the aerosol dynamics.

Current sectional models can be divided in three approaches: (i) the lagrangian approach, (ii) the semi-lagrangian and (iii) the eulerian approach. In the lagrangian approach the particles are allowed to grow to their exact size and the particles are not redistributed over the fixed size grid. The semi-lagrangian approach was developed by (Jacobson, 1997a,b). Using this approach, the sections are fixed and the average diameter is allowed to vary within its size section, while in the eulerian approach, the mean diameters representative of each section are fixed. Although the semi-lagrangian approach has been shown to be very accurate

because it eliminates numerical diffusion (Zhang et al., 1999), it could lead to empty sections in some 0D case studies without constant emission fluxes.

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Annex 2

Natural Dust emissions: The process of saltation and sandblasting

Modelling dust entrainment into the atmosphere has been the subject of many studies since the early study of Bagnold (1941). Several detailed emission models have been proposed, for example by Marticorena and Bergametti, 1995; Alfaro and Gomes, 2001 and Nickovic et al., 2001. They are currently used in transport models for sensitivity studies, analyses or forecasts as in DREAM (Nickovic et al., 2001), CHIMERE-DUST (Forêt et al., 2006; Menut et al., 2005, 2007), CHIMERE (Vautard et al., 2005), LOTOS-EUROS (Schaap et al., 2010) among others. All these schemes have their origin in those developed by Marticorena and Bergametti (1995). The sandblasting flux *Fv* is expressed as the product of the saltation dust flux *Fh* and α, the sandblasting efficiency: $Fv = \alpha \times Fh$ with $\alpha = 5.10^{-5}$ m^{-1} an order of magnitude obtained from the measurements, over Northern Spain and Niger, of Gomes et al. (2003). The saltation flux is usually expressed as:

$$F_h = C \times u_{*s} \times (u_{*s}^2 - u_{*t}^2)$$

where u_{*s} denotes the saltation friction velocity, u_{*t} the threshold friction velocity and C is a coefficient that depends on several surface factors. The saltation friction velocity u_{*s} corresponds to that encountered on erodible parcels of the model grid cell, usually smoother than typical vegetated surfaces found in Europe. It is calculated by using the 10m wind field, a constant saltation roughness length of 5.10^{-4} m and the assumption of neutral stability as in most previous studies. In order to keep the formulation simple, the threshold friction velocity is assumed to depend only on gravimetric soil moisture and follows the formulation of Fecan et al. (1999).

Annex 3

Some examples of current chemistry transport models for PM modelling

Model	Source
CAMx	http://www.camx.com/home.aspx
CHIMERE	http://www.lmd.polytechnique.fr/chimere/
CMAQ	http://www.epa.gov/asmdnerl/CMAQ/index.html
DEHM	http://www.dmu.dk/en/air/models/
EMEP	http://www.emep.int/index_model.html
EURAD	http://www.eurad.uni-koeln.de/index_e.html
FARM	http://urbanemissions.info/daqi-farm.html
LOTOS-EUROS	http://www.lotos-euros.nl/
MATCH	http://www.smhi.se/en/Research/Research-departments/Air-quality/match-transport-and-chemistry-model-1.6831
MOCAGE	http://www.cnrm.meteo.fr/gmgec/spip.php?article87
NAME	http://www.metoffice.gov.uk/research/modelling-systems/dispersion-model
REM-Calgrid	http://www.geo.fu-berlin.de/met/ag/trumf/RCG/RCG_Internet_description.pdf
SILAM	http://silam.fmi.fi/

Annex 4

Some examples of current urban and street canyon, models for PM modelling

Model	Source
AIRQUIS-episode	http://www1.nilu.no/airquis/models_dispersion.htm
ADMS4	http://www.cerc.co.uk/environmental-software/ADMS-model.html
ADMS-Urban	http://www.cerc.co.uk/environmental-software/ADMS-Urban-model.html
AERMOD	http://www.epa.gov/scram001/dispersion_prefrec.htm
DEHM	http://www.dmu.dk/en/air/models/
GRAL(*)	http://pandora.meng.auth.gr/mds/showlong.php?id=133
IFDM (VITO)	http://pandora.meng.auth.gr/mds/showlong.php?id=50
OML	http://www.dmu.dk/en/air/models/oml/omlmodeldescription/
OSPM	http://www.dmu.dk/en/air/models/ospm/
PROKAS	http://www.lohmeyer.de/en/content/software-sales-distribution/product-overview/prokas
UDM-FMI	http://pandora.meng.auth.gr/mds/showlong.php?id=121

Gaussian models

In atmospheric dispersion modelling Gaussian type models are widely used in practical applications in many European cities in particular for regulatory purposes : yourAir system (London, Liverpool, Budapest, Vienna), Urbanair (Strasbourg), Sirane (Lyon, Granoble, Valence, Saint-Etienne).

The principle of Gaussian model is based on a Gaussian distribution of the plume in the vertical and horizontal directions under steady state conditions. The normal distribution of the plume is modified at greater distances due to the effects of turbulent reflection from the surface of the earth and at the boundary layer when the mixing height is low. The width of the plume is determined by σy and σz, which are defined either by stability classes (Pasquill, 1961; Gifford, 1976) or travel time from the source, or function of the Monin obukhov length.

Limitations

Concerning the urban scale modelling, systems should be able to allow for the various local scale effects, for instance, the influence of buildings and obstacles, downwash phenomena and plume rise, together with chemical transformation and deposition. Some advanced models can simulate some of the chemical transformations using post processing treatment of the chemistry. One limit of the Gaussian plume equation lies in the assumption that there is no interaction between plumes, which can become significant within urban environments. Regarding the particulate matter, most of the Gaussian models have no chemical process to assess the secondary organic aerosols, which can be important within the pollution plume (MEGAPOLI, 2011).

Gaussian models have been shown to consistently over predict concentrations in low wind conditions (Benson, 1984; Sokhi et al., 1998). Hybrid models, which use a combination of the Gaussian plume and puff models, include along wind dispersion of the pollutants in order to better estimate concentrations under low wind speed conditions (Sharan et al., 1996;

Thomson and Manning, 2001). A further limitation is a result of the simplified treatment of turbulence and meteorology so they are best suited to calculating hourly pollutant concentrations.

Since Gaussian plume equations assume a homogeneous wind field, it is not recommended that they are used for far field modelling as the meteorology is expected to change over such large distances. Caputo et al. (2003) observed that four Gaussian models calculated non-zero concentrations for the whole downwind domain and so suggested that they should be limited to distances a few tens of kilometres from the source.

The US-EPA today recommends the following computer packages for simulation of non-reactive chemicals:

US Gaussian models:

- AERMOD is a steady-state Gaussian plume model. It uses a single wind field to transport emitted species. The wind field is derived from surface,upper-air, and onsite meteorological observations. AERMOD also combines geophysical data such as terrain elevations and land use with the meteorological data to derive boundary layer parameters such as Monin-Obukhov length, mixing height, stability class, turbulence, etc., AERMOD is today replacing the ISC models for most regulatory applications in the US.

- CALINE4 is a steady-state Gaussian dispersion model designed to determine air pollution concentrations at receptor locations downwind of highways located in relatively uncomplicated terrain. CALINE4 is incorporated into the more refined CAL3QHC and CAL3QHCR models

- CTDMPLUS Complex Terrain Dispersion Model Plus Algorithms for Unstable Situations (CTDMPLUS) is a refined point source gaussian air quality model for use in all stability conditions for complex terrain. The model contains, in its entirety, the technology of CTDM for stable and neutral conditions. CTSCREEN is the screening version of CTDMPLUS.

- ASPEN The Assessment System for Population Exposure Nationwide (ASPEN) consists of a dispersion and a mapping module. The dispersion module is a Gaussian formulation based on ISCST3 for estimating ambient annual average concentrations at a set of fixed receptors within the vicinity of the emission source. The mapping module produces a concentration at each census tract. Input data needed are emissions data, meteorological data and census tract data. The Emissions Modelling System for Hazardous Pollutants (EMS-HAP) is needed to process the emission inputs into the ASPEN model or the ISC3 model. The ASPEN model was used in estimating annual ambient concentrations for air toxics pollutant in the National Air Toxics Assessment (NATA) Study.

- HOTMAC and RAPTAD - HOTMAC is a model for weather forecasting used in conjunction with RAPTAD which is a puff model for pollutant transport and dispersion. These models are used for complex terrain, coastal regions, urban areas, and around buildings where other models fail.

European Gaussian models

- ADMS-URBAN - A model for simulating dispersion on scales ranging from a street scale to city-wide or county-wide scale, handling most relevant emission sources such as traffic, industrial, commercial, and domestic sources. It is also used for air

quality management and assessments of current and future air quality vis-a-vis national and regional standards in Europe and elsewhere.

- CAR-FMI (Finland) - This model was developed by the Finnish Meteorological Institute (FMI) for evaluating atmospheric dispersion and chemical transformation of vehicular emissions of inert (CO, NOx) and reactive (NO, NO2, O3) gases from a road network of line sources on a local scale. It is a Gaussian line source model which includes an analytical solution for the chemical cycle NO-O3-NO2.

- CAR-International (The Netherlands) - Calculation of Air pollution from Road traffic(CAR-International) is an atmospheric dispersion model developed by the Netherlands Organisation for Applied Scientific Research. It is used for simulating the dispersion of vehicular emissions from roadway traffic.

- OPS-ST, The Operational Priority Substances short term model, OPS-ST (van Jaarsveld & Klimov, 2011) is derived from the OPS long-term model (van Jaarsveld 2004) and is used for modelling of dispersion and deposition of pollutants for the Netherlands on both local and national scale. The model can be described as a Lagrangian (trajectory) model which acts as a Gaussian plume model for local situations. Dry and wet deposition mechanisms are included as a function of particle size with some emphasis on the behaviour of large particles. Dry and wet deposition is taken into account as a function of particle size and the density of the particles.

- SIRANE is an operational urban dispersion model based on a simplified description of the urban geometry that adopts parametric relations for the pollutant transfer phenomena within and out of the urban canopy. The model contains three main mechanisms for transport in and out of a street: advection along the street axis, diffusion across the interface between the street and the overlying air flow and exchanges with other streets at street intersections. The dispersion of pollutants advected or diffused out of the streets is taken into account using a Gaussian plume model, with the standard deviations σ_y and σ_z parameterised by the similarity theory. (Soulhac, 2001-2011)

Street models (a review is given by Vardoulakis, 2003)

- OSPM is a practical street pollution model, developed by the Department of Environmental Science at Aarhus University (the former National Environmental Research Institute, Department of Atmospheric Environment).
- AEOLIUS is based on concepts and techniques previously used for the development of the Operational Street Pollution Model (Hertel and Berkowicz, 1989a), which was evolved from the CPBM. AEOLIUS and OSPM are semi-empirical models that calculate concentrations of exhaust gases on both sides of a canyon assuming three different contributions: (a) the contribution from the direct flow of pollutants from the source to the receptor, (b) the recirculation component due to the flow of pollutants around the vortex generated within the recirculation zone of the canyon, and (c) the urban background contribution. A Gaussian plume algorithm is used for the calculation of the direct contribution and a simple box model for deriving the recirculation component.

Other non Gaussian models

- CALPUFF is a multi-layer, multi-species non-steady-state puff dispersion model that simulates the effects of time- and space-varying meteorological conditions on

pollution transport, transformation and removal. CALPUFF can be applied on scales of tens to hundreds of kilometers. It includes algorithms for subgrid scale effects (such as terrain impingement), as well as, longer range effects (such as pollutant removal due to wet scavanging and dry deposition, chemical transformation, and visibility effects of particulate matter concentrations).

- MSS, Micro-SWIFT-SPRAY (Moussafir et al., 2004 and 2007), is a fast transport and dispersion modelling system. It is designed for local scale and takes into account buildings. MSS consists of SWIFT and SPRAY used in urban mode (Micro SWIFT, Micro Spray). Micro-SWIFT (Moussafir et al., 2004; Tinarelli et al., 2007) is an analytically modified mass consistent interpolator over complex terrain. Micro-SPRAY is an LPD (Lagrangian Particle Dispersion) model able to take into account the presence of obstacles. It directly derives from the SPRAY code (Anfossi et al., 1998).

Annex 5 : Examples of national experiences for source apportionment

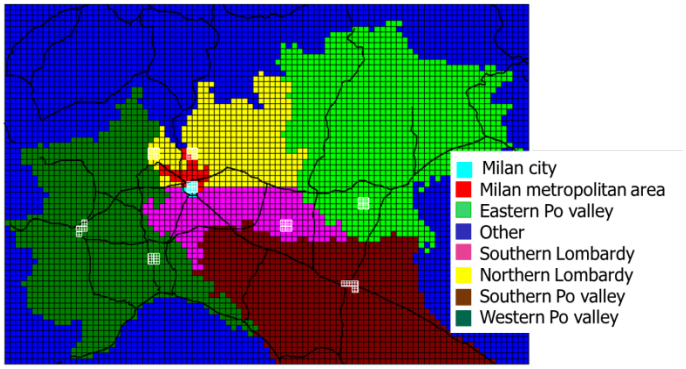
Application of Source Apportionment on the Milan area and the Lombardy region

An example of reactive tracers approach is the PM Source Apportionment Technology (PSAT, Yarwood et al., 2004; Wangstrom et al., 2008) implemented in the CAMx model (ENVIRON, 2008). PSAT algorithm is embedded into the CAMx code providing an effective method for modelling source apportionment when a large number of sources are used. It is also worth noting that this algorithm provides information on the current source contributions but not on the effect of any reduction or increase in a particular source.

PSAT uses reactive tracers to apportion both primary and secondary PM compounds among different source categories (e.g. transport, domestic heating, industrial processes) and source regions (e.g. city area, urban surroundings, regional basin, transboundary contribution). PSAT is designed to apportion the main primary and secondary PM compounds modelled in CAMx, such as: Sulphate, Nitrate, Ammonium, Secondary Organic Aerosol and a few primary classes including Elemental Carbon, Primary Organic Aerosol and Crustal material.

The first example illustrates a modelling analysis conducted in the framework of the POMI project (http://aqm.jrc.ec.europa.eu/pomi/) over a computational domain covering the whole Po Valley, but focused on the Lombardy region and, more specifically, the Milan area. Simulations have been performed over a 6x6 km^2 grid for the calendar year 2005 using CAMx (ver. 4.50) employing CB05 (Yarwood et al., 2005) gas phase mechanism and CF aerosol module (ENVIRON, 2008). CF module considers only two static size bins (Coarse and Fine), no aerosol dynamics and supposing that secondary aerosol is only in the fine fraction. Emission fields have been derived from local inventories for the four main analysed regions (Piemonte, Lombardia, Veneto and Emilia Romagna), the official Italian inventory for the remaining Italian regions included in the domain and the EMEP inventory outside Italian borders. Meteorological fields have been produced by MM5, driven by the NCEP (American National Centre for Environmental Prediction) 6 hourly Final Analyses (FNL) were used as initial and boundary conditions. Further details on the modelling set up are available in (Pirovano et al., 2009; 2010).

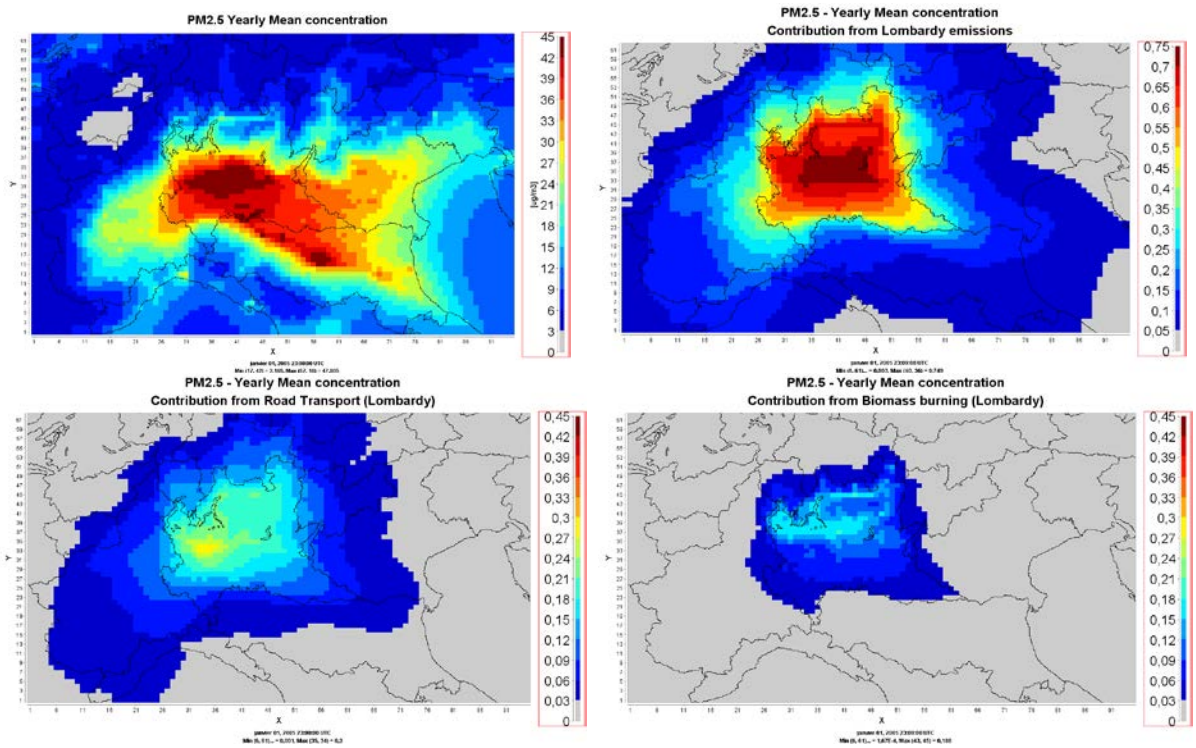
PSAT tool has been configured in order to track the contribution of 8 different geographical areas split among 6 emission groups. The first two areas cover the Milan administrative borders and the most urbanised surroundings, respectively. Together, they form the Milan metropolitan area. Lombardy region has been split in two areas: the northern part including the main cities and the southern part, mainly devoted to agriculture. The leftover part of the Po valley has been subdivided according to the regional boundaries into three main areas: eastern, western and southern Po valley, respectively. A last area has been designed to take into account the emissions of not Italian sources included in the domain.



Definition of the 8 source areas

Emission groups have been defined in order to track the anthropogenic activities mainly contributing to particulate matter, hence defining the following groups and their correspondence with the SNAP sectors: road transport (SNAP 7), biomass burning for domestic heating (SNAP 2), agriculture (SNAP 10), power plants (SNAP 1). The source apportionment analysis has been performed over the whole domain, whereas the analysis of the results have been focused on the Milan are and the Lombardy region. Model performance evaluation highlighted that bias in PM_{10} modelled concentrations was mainly related to PM coarse and to a fraction of organic matter. For these reasons, the source apportionment analysis has been focused on $PM_{2.5}$, not including secondary organic aerosol. As an example of PSAT application over a region, figure below shows the contribution of Lombardy emissions to the PM2.5 yearly mean concentrations.

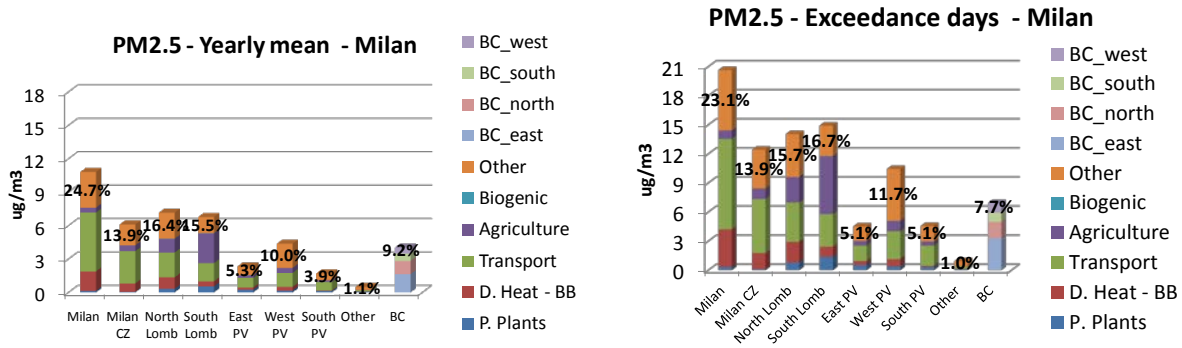
As shown in the map, Lombardy contribution extends over the whole Po valley, ranging between 10 and 70% (reaching 30% outside Lombardy). Likewise, the surrounding regions yield not less than 25% of total concentration inside Lombardy. The figure also compares the contribution of road transport and biomass burning in domestic heating. Road transport yields a contribution influencing $PM_{2.5}$ concentration also out of regional boundaries; conversely biomass burning influence is limited to Lombardy. This is due to the significant contribution of road transport to the secondary PM.



PM2.5 yearly mean concentration (top left); relative contribution of total (top right), road transport (bottom left) and biomass burning (bottom right) emissions from Lombardy to the total PM2.5 concentration.

PSAT tool optionally provides the results of the source apportionment analysis also to a set of user defined specific receptors. As an example Figure below, displays the contribution of each emission areas, stacked by emission categories. The picture shows that the total concentration at each receptor is due to the contribution yielded by several areas and categories, proving that is not possible to detect a single emitting source as the main responsible of particulate matter in Lombardy region.

The analysis also highlights that, as expected, the main contribution comes from the area to which the receptor belongs. However, it is worth noting that such contribution is generally lower than 40%, even in the most emitting area of Milan. Long range transport is accounted for the boundary conditions share, proving that particulate matter is mainly due to the Po valley sources. 0 figure shows also that road transport is the most relevant emitting sector in each area followed by either agriculture or domestic heating, according to the position of the receptor. Finally, the second diagram shows the differences in the total concentration as well as in the relative role played by each source during exceedance days.



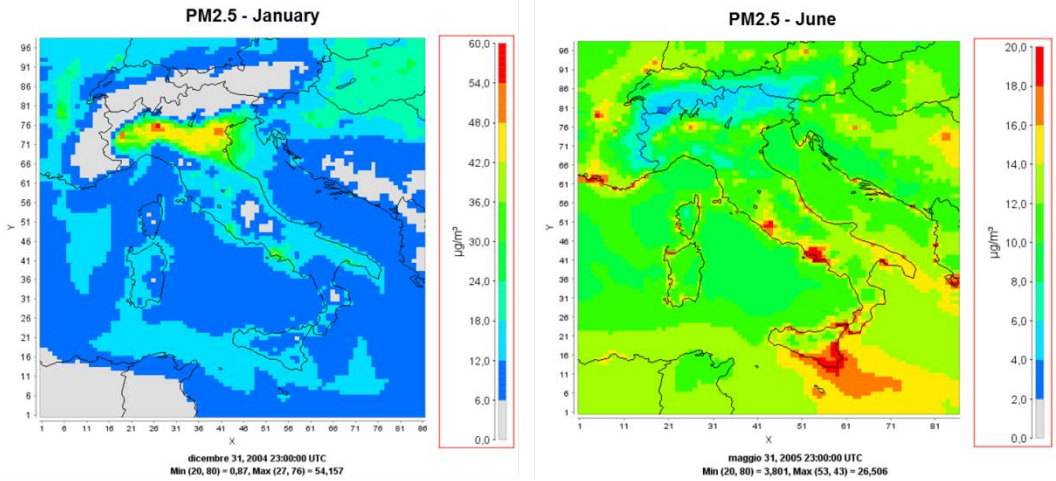
Contribution of the different emission areas, stacked by source categories, to the mean concentration of PM2.5 at Milan receptor. Labels show the total contribution of each emission area. The analysis refers to the whole year (left) as well as to the set of days where the PM10 computed concentration exceeded 50 $\mu g/m^3$

Application of Source Apportionment in Italy

A second example of PSAT application, concerning the whole Italy, derives from a project focused on road transport policies (Balzarini et al., 2011). Air quality simulation have been conducted by means of CAMx (ver. 4.50) employing the same configuration previously described. Meteorological fields have been reconstructed by means of the Weather Research and Forecast modelling system (WRF release 3.2.1; Skamarock et al., 2008) fed by ECMWF global analysis (http://www.ecmwf.int).

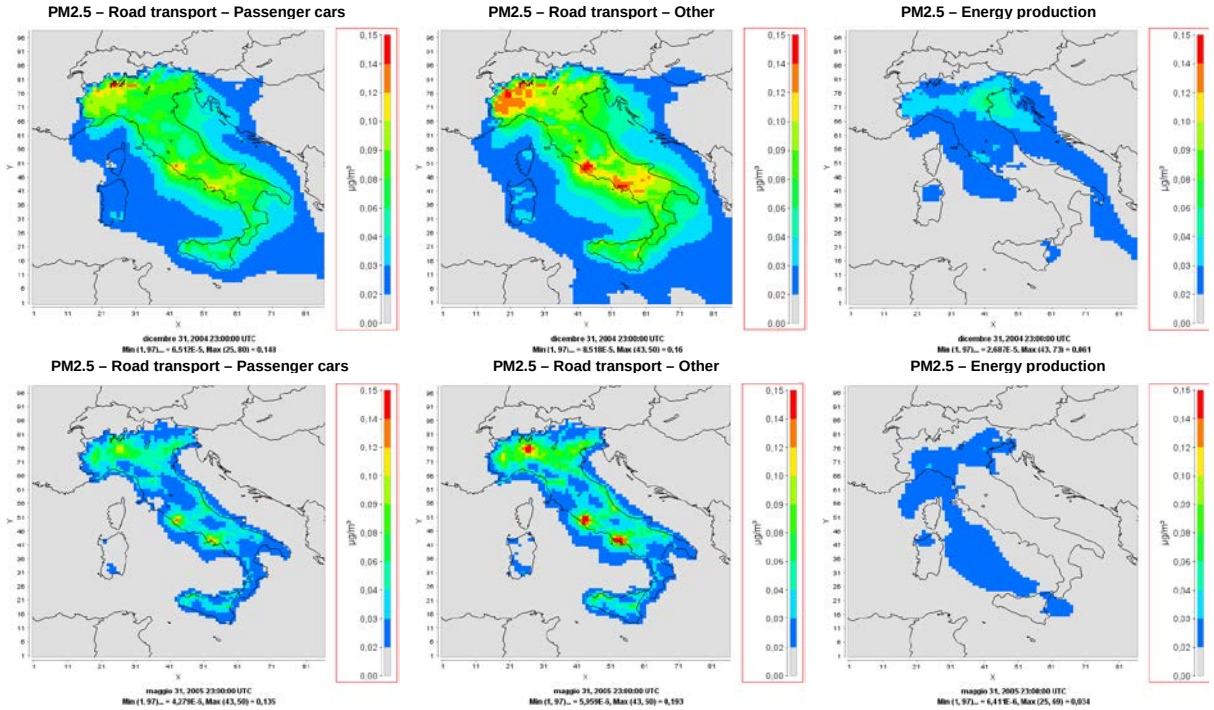
Anthropogenic gridded emissions have been compiled with the Sparse Matrix Operator Kernel Emission processor (SMOKE version 2.6, www.smoke.org). The Italian emission dataset has been derived from the Italian official inventory, while the EMEP database has provided emissions of the neighbouring countries. Natural emissions have been calculated with a sea salt processor based on Gong et al. (2002) and the MEGAN biogenic emission model (version 2.04; Guenther et al., 2006). Initial and boundary conditions have been obtained from a CHIMERE model run at European scale (Bessagnet et al., 2004). CAMx model has been applied for January and June 2005 over the Italian domain (1290x1470 km2) with a grid step of 15 km.

Figure below shows the monthly mean of modelled PM2.5 for both January and June 2005 in the Italian domain. During the winter, PM2.5 concentration ranges between 12 µg/m^3 to 60 µg/m^3. The highest values have been found in the Po Valley and near the major cities (Milan, Turin and Venice), caused by strong emissions and weak circulation conditions. Notably the area of Milan shows concentration values higher than 40 µg/m^3. In summer concentrations are lower and they range between 10 µg/m^3 to 18 µg/m^3. The highest values are located in the urban cities of Rome and Naples (~ 20 µg/m^3).



Simulation results of PM2.5 for January (left) and June (right) 2005 in the Italian domain

PSAT algorithm has been set up in order to account for the contribution to gas and PM concentrations of the following categories of the Italian emissions: passengers cars, other road transports and energy production. The highest contribution to the $PM_{2.5}$ concentration has been observed from road transport near the most polluted cities, such as Milan, Turin, Naples and Rome and along the main motorways (from 6% to 15% in January and from 3% to 15% in June).



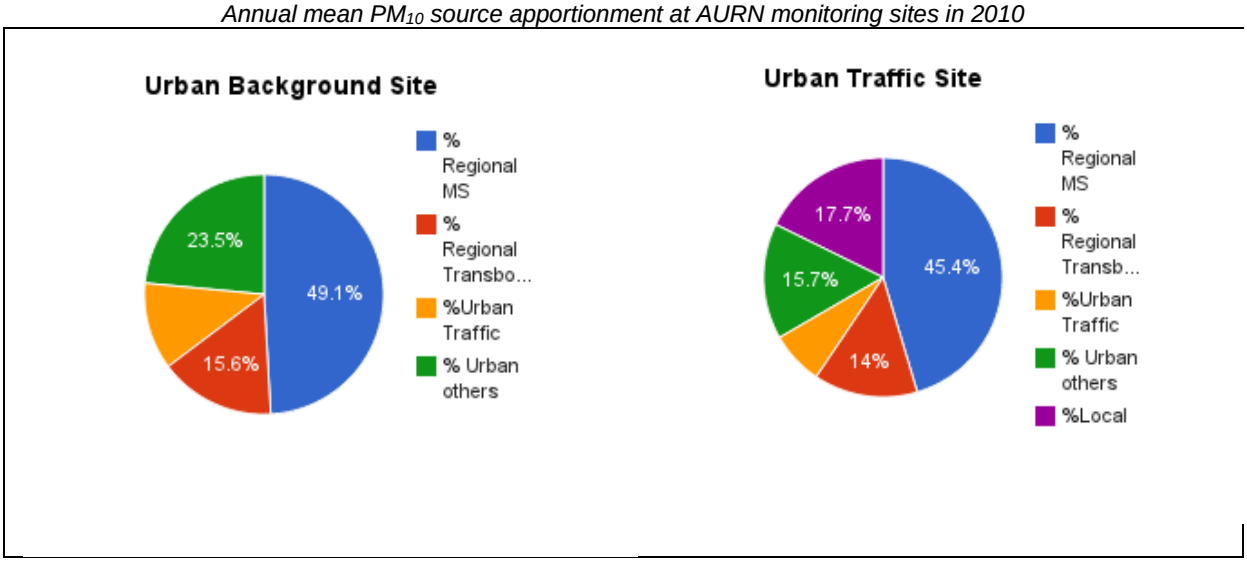
Contribution of passenger cars, other road transports and energy production to the total concentration of PM2.5 in January (top) and June 2005 (bottom). Data are represented as a fraction of the total concentration.

Source Apportionment in the UK

The UK makes extensive use of models to provide supplementary information as part of the annual assessment of compliance with limit and target values. The UK uses the Pollution Climate Mapping (PCM) model for this (Brookes at al 2012). This model is used to provide annual mean concentration estimates for both background and traffic (roadside) locations as required by Annex III of the AQD. Background concentrations are calculated for 1 km 1 km grid squares across the whole of the UK and roadside concentrations are calculated for approximately 9000 urban traffic locations. These traffic locations have been chosen so that the assessment is carried out at locations specified with in Annex III of the Directive.

The PCM model used for the UK compliance assessment reporting is an annual mean model and is routinely used to estimate source apportionment of annual mean PM concentrations. The average source apportionment for PM_{10} at urban background and traffic measurement stations in 2010 is illustrated in figure below.

Annual mean PM_{10} source apportionment at AURN monitoring sites in 2010

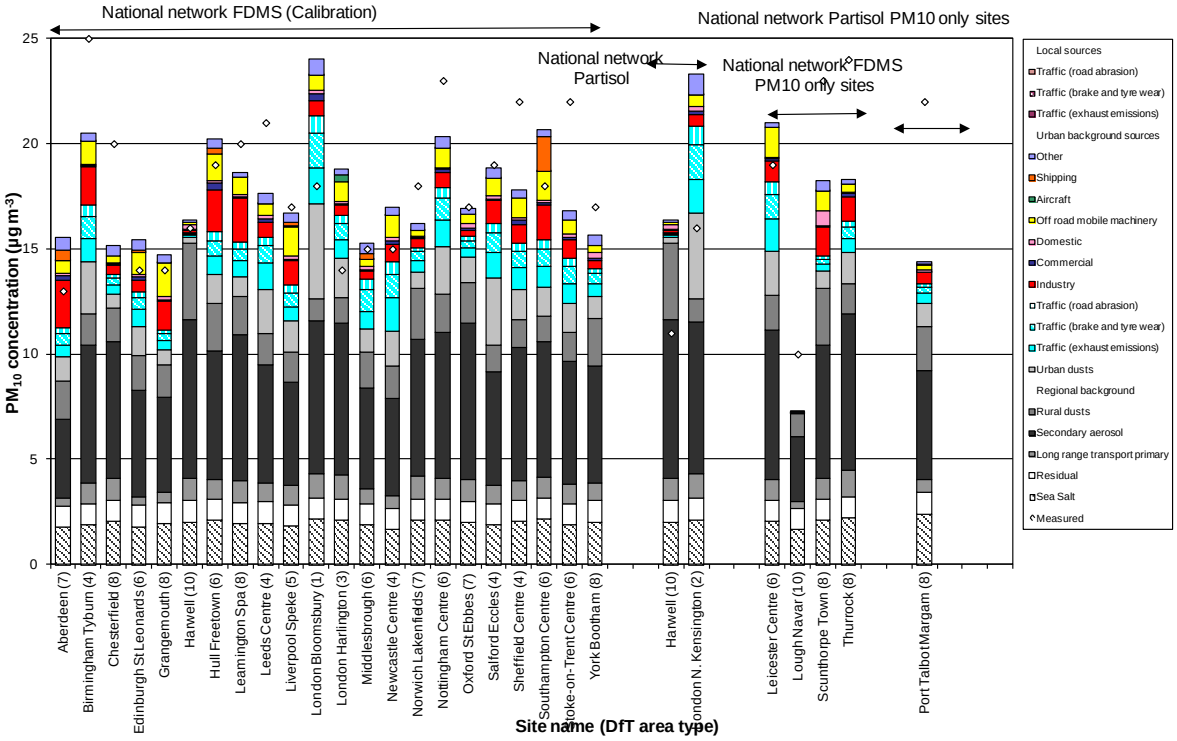


Road traffic (exhaust, brake and tyre wear and road abrasion) is the most important local source of primary PM along with contributions from off road machinery and industry. Ambient PM_{10} concentrations are dominated by the regional background, for which the most important components are secondary inorganic aerosol (SIA), secondary organic aerosol (SOA), regional primary PM and rural dusts (soil re-suspension). Note that in these plots the contributions from rural dusts, sea salt and the residual have been included in the regional

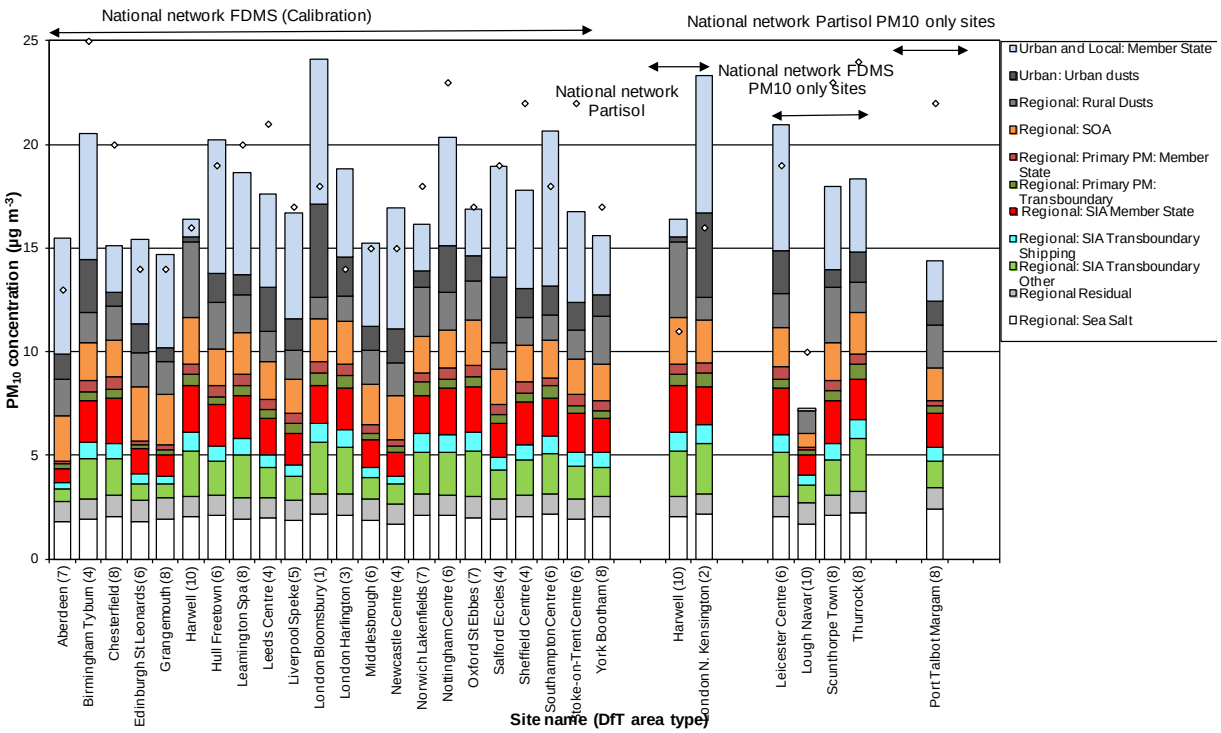
MS category. Only the contributions directly associated with emissions from sources included in emission inventories (SIA precursors and primary PM) have been included in the transboundary totals. Urban dusts have been included in the urban others totals. Details of the full set of contributions included for PM are provided in Figures below.

The contribution to the regional background PM_{10} from transboundary sources is important. More than half of the SIA is associated with emissions of precursor gases (NO_x, SO_2 and NH_3) from outside the UK. Source apportionment for SIA has been calculated using emission sensitivity coefficients derived from results from the EMEP model. About half of the regional primary PM is associated with emissions from outside the UK (transboundary). The regional background contribution (and thus transboundary) is also an important contributor to the total concentration at traffic locations. The contributions from brake and tyre wear are of similar magnitude to the contributions from exhaust emissions.

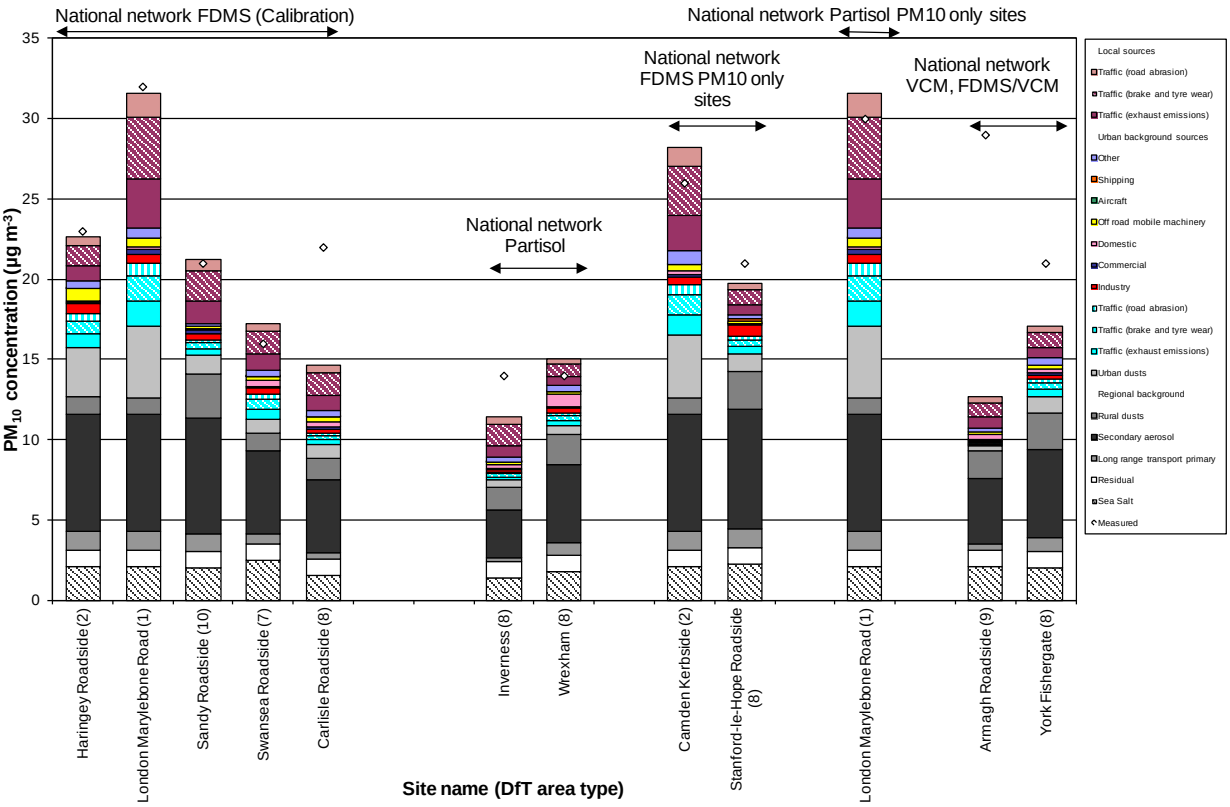
Annual mean PM_{10} source apportionment at background national network monitoring sites in 2010



Annual mean PM_{10} source apportionment at background national network monitoring sites in 2010: transboundary contributions highlighted



Annual mean PM_{10} source apportionment at national network roadside monitoring sites in 2010



The source apportionment analysis for PM_{10} includes the following contributions:

- Regional background

    - Regional: Sea Salt (natural)
    - Regional Residual (constant value, assumed natural)
    - Regional: SIA Transboundary Other (contribution to secondary inorganic aerosol from no-UK EU sources)
    - Regional: SIA Transboundary Shipping (contribution to secondary inorganic aerosol from shipping)
    - Regional: SIA Member State (contribution to secondary inorganic aerosol from UK sources)
    - Regional: Primary PM: Transboundary (contributions to regional primary PM from sources outside the UK)
    - Regional: Primary PM: Member State (contributions to regional primary PM from UK sources)
    - Regional: SOA (secondary organic aerosol)
    - Regional: Rural Dusts (wind blown dusts, depends on land use)
- Urban background sources
    - Urban dusts (vehicle and non-vehicle related)
    - Traffic (exhaust emissions, ubiquitous)
    - Traffic (brake and tyre wear, ubiquitous)
    - Traffic (road abrasion, ubiquitous)
    - Industry (point and area sources, important at some locations, present at all)
    - Commercial (non-domestic heating)
    - Domestic (domestic heating)
    - Off road mobile machinery (ubiquitous and quite large)
    - Aircraft
    - Shipping
    - Other (includes rail and accidental fires and bonfire night)
- Local Sources
    - Traffic (exhaust emissions)
    - Traffic (brake and tyre wear)

Traffic (road abrasion)