Guide on modelling Nitrogen Dioxide (NO_2) for air quality assessment and planning relevant to the European Air Quality Directive

Result of activities in the FAIRMODE Working Group 1

Version 4.6

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Preface to version 4.6 of this document

This is the third draft of the model application document for NO_2 modelling in regard to the European Air Quality Directive (2008/50/EC). This document is complementary to the previously developed and published model application document 'The application of models under the European Union's Air Quality Directive: A technical reference guide' (EEA, 2011) in that it concentrates on the actual modelling methods and the scientific aspects of these, rather than on the interpretation and general use of models for AQ Directive applications.

This document is the result of ETC/ACM activities for EEA within FAIRMODE Working Group 1 and was initiated in response to the model activities of Member States around the 'notification of postponement' for NO_2 and benzene*. Various elements of this document, e.g. emission inventories and quality assurance, are being actively addressed in other FAIRMODE activities in order to further develop modelling guidance.

This version (v4.6), builds on the previously openly available version (v3.3) and is the result of several iterations with the modelling and model user communities in the EEA member countries. A large portion of this document is the result of contributions from this community. Though the major part of the text and editing has been the responsibility of ETC/ACM, more than 30 people have contributed to the document in various ways, ranging from significant textual contributions, to corrections, to general discussions on modelling as well as to the contribution of examples. The document has been presented at a number of meetings in the years 2010 and 2011 and this has helped stimulate the participation of the community.

Though the document is intended to present 'good practise' in the modelling of NO_2 it is by no means 'universally accepted good practise'. What is clear from the range of inputs and comments provided from the community is that there is a healthy diversity of views on what constitutes 'good practise' and what is 'fit for purpose', two concepts that are regularly referred to when applying models. What is often considered scientific 'good practise' may not be as 'fit for purpose' in applications for the AQ Directive. This may lie in the fact that the AQ Directive is not requesting scientifically well founded results but is requesting, to a large degree, tools for policy implementation and compliance checking. The intention of this document is to address mainly the scientific basis but always keeping in mind the AQ Directive as the intended application.

One other aspect in this regard, and in regard to the contributions from the community, is that it is difficult to provide firm recommendations on modelling that are consensual, i.e. that the large majority agree to. Within this document then we are not providing precise recommendations, as these need more discussion within the FAIRMODE community. Instead we provide *application guidance*, which is a non exhaustive list of indicative recommendations. These will need to be further discussed by the FAIRMODE community.

One of the most widely received comments from the community is that diversity and flexibility should be encouraged and that modellers and model users should not be restrained to adhere to fixed recipes or methodologies. FAIRMODE agrees with these views but it also considers that there is a need for common reference points when models are being applied to a common application, i.e. the European Air Quality Directive.

Contribution table version 3.3

Following the 3rd FAIRMODE plenary session (May 2010) a number of people agreed to and, as a result, contributed to the development of this model application document. The contributors are listed in the following table with a short indication of where their contributions were provided.

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Panagiota Dilara	panagiota.dilara@jrc.ec.europa.eu	JRC ISPRA	Chapter 4, Emissions. Some corrections and inclusions.
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Addo van Pul Joost Wesseling	addo.van.pul@rivm.nl Joost.Wesseling@rivm.nl	RIVM	Contributions on NO2 chemistry modelling. Chapter 3.3. Variety of comments on NL perspectives.
John Stedman	John.Stedman@aeat.co.uk	AEA	Comments and additions mostly concerning NO2/NOx emission ratio.
Helge Rørdam Olesen	hro@dmu.dk	NERI	Comments in chapters 2 and 3. Detailed description of OSPM chemistry.
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David Carruthers Martin Seaton	David.Carruthers@cerc.co.uk martin.seaton@cerc.co.uk	CERC	Comments and additions to Sections 2.1, 2.2, 2.3, 2.4, 2.5, 3.1, 3.2, 4 and Appendix 1
Linton Corbet	linton.corbet@chmi.cz	CHMI	Contributions and corrections to Sectors 3.4
Lia Fragkou John Douros	lia@aix.meng.auth.gr jdouros@aix.meng.auth.gr	AUTH	Significant contributions to Chapters 2 and 4 and Source apportionment Chapter 6.3
Niemi Jarkko	Jarkko.Niemi@hsy.fi	HSY	Recommended inclusion of AERMOD and other EPA models
Rafael Borge Julio Lumbreras	rborge@etsii.upm.es jlumbreras@etsii.upm.es	UPM	Comments on Chapter 2 and significant content in Chapter 4
Philip Thunis	philippe.thunis@jrc.ec.europa.eu	JRC ISPRA	Chapter 7.2 Validation of models

Contribution table version 4.6

The previous version (v3.3) was made available for comment and contribution after the 4'th FAIRMODE plenary in June 2011. The contributors to the new version of this document (v4.6) are listed in the following table with a short indication of where their contributions were provided.

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Jose Maria Baldasano	jose.baldasano@bsc.es	Barcelona Supercomputing Centre, Spain	Inclusion of HERMES emission model for Spain
Michael Schatzmann	schatzmann@zmaw.de	Meteorological Institute, University of Hamburg, Germany	Critical comments on the validity of models in general and on the representativeness of input data
Fernando Martin Llorente	fernando.martin@ciemat.es	Research Center for Energy, Environment and Technology (CIEMAT), Spain	A number of comments and improvements concerning CFD models. Various text corrections
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Ingo Düring	ingo.duering@lohmeyer.de	Lohmeyer GmbH, Germany	Addition of limited mixing NO2 scheme, addition of models PROKAS.
Pete Roberts	pete.roberts@concawe.org	CONCAWE	Various comments and corrections throughout the document
Elke Trimpeneers	trimpeneers@irceline.be	IRCELINE, Belgium	Information on RIO-corine downscaling methodology applied in Belgium
David Carruthers	David.Carruthers@cerc.co.uk	CERC, UK	Various comments on text
Anke Lükewille	Anke.Luekewille@eea.europa.eu	EEA, Denmark	Various comments in text. Update of UK request for postponement
Stijn Janssen	stijn.janssen@vito.be	VITO, Belgium	Reference to AURORA-FIDM coupling.
Stefano	Stefano.galmarini@jrc.,ec.europa	JRC ISPRA	Additions and corrections to

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Ingrid Sundvor	is@nilu.no	NILU, Norway	Review of chapter 3 Chemistry
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Cristina Guerreiro	cbg@nilu.no	NILU, Norway	Review of Chapter 6, Modelling applications for the Directive
Núria Castell Balaguer	ncb@nilu.no	NILU, Norway	Review of Chapter 4, Emissions. General textual changes.
Bruce Rolstad Denby	bde@nilu.no	NILU, Norway	Reorganized document. Section 6 (Application to the directive) moved to section 2.

Contents

Summary

The European Air Quality Directive (2008/50/EC) sets limit values for the concentration of Nitrogen Dioxide (NO_2). It is a requirement that all air quality zones and agglomerations assess the levels of NO_2 and report these to the European Commission. If NO_2 concentrations are found to exceed these limit values then the Member States are obliged to take appropriate steps to reduce these concentrations. In order to do this effectively, air quality management plans must be developed and possible measures assessed. Air quality modelling then becomes an essential tool for local and national authorities to manage their air quality.

This document provides initial guidance on the use of models for air quality assessment and planning, with particular regard to modelling NO_2 concentrations. The document is intended as guidance for authorities and modellers to help stimulate 'good practise' in air quality modelling and provide an overview and description of methods and tools available for air quality modelling. Since NO_2 is generally a local and urban scale problem, this document concentrates on applications within cities.

In regard to the limit values set for NO_2, of which one is for hourly mean percentile concentrations and the other for annual mean concentrations, it is important to note that the annual mean limit value of 40 $\mu g/m^3$ is the most stringent of these two. It is very rare that the hourly mean percentile limit value is exceeded when the annual mean is not. This has important consequences for modelling since predicting the 19'th highest hourly mean concentration is far more uncertain than predicting the annual mean concentration. Several simpler model types may be quite adequate for annual mean concentrations, but inadequate for hourly mean percentiles.

Nitrogen oxides (NO_X) are emitted in all combustion processes and the major source of these in urban areas is traffic. NO_X is emitted as both NO and NO_2. Unlike some other regulated pollutants NO_2 and NO are highly reactive gases on short time scales (minutes) through the oxidation of NO with ozone and the photo-dissociation of NO_2 to NO. As such chemistry plays an important role in determining concentrations on all scales, be they local hotspots, urban or regional scales. For this reason special consideration must be given to the description of chemistry in the models used to assess NO_2.

One of the reasons for the development of this document has been the efforts by Member States to provide well founded air quality plans in regard to the 'notification of postponement', whereby Member States can delay the achievement of the limit values if concrete and realisable plans can be shown to be in progress that will ensure compliance by the revised deadline. For this reason it is important that the results of NO_2 modelling not only provide suitable assessment of the current concentration levels but that they can also provide realistic concentrations in the future, given changes in emissions, background concentrations and meteorological conditions. It is thus not enough that an air quality model provides verified assessments but that the sensitivity of the model to these changes must also be properly assessed and verified.

The document describes and provides some basic guidance on a number of elements of air quality modelling. These include dispersion, chemistry, emissions, meteorology and quality control. The modelling described here is almost exclusively source oriented modelling. This means that the air quality models discussed are process based mathematical models that take emissions and calculate concentrations from these. Empirical or statistically based models are not discussed to any great extent (with the exception of empirically based chemistry schemes) because process based models are the most useful for planning applications.

Though this document provides basic guidance, and hopefully understanding, of air quality modelling it cannot replace expert knowledge in the application of the models. Indeed, the level of complexity of any air quality or meteorological model warrants a corresponding level of expertise in its application and interpretation.

1 Introduction

This document provides an overview and recommendations for the modelling of ambient NO_2 concentrations in urban areas. 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). It has been developed to help support the needs of the Member States in addressing the European Air Quality Directive (2008/50/EC), termed 'AQ Directive' in this document.

1.1 Scope and aims

This document has the following aims:

- To provide accessible guidance on the application of air quality models in regard to the European Air Quality Directive, with emphasis on the modelling of nitrogen dioxide (NO_2);
- To encourage 'good practise' with the use of models for air quality assessment and planning applications.

The document cannot cover all details in regard to modelling but will cover the following aspects:

- Will provide an overview suitable for understanding the needs and limitations when applying models, with emphasis on the modelling of NO_2;
- Will provide suitable background information for authorities to make informed choices when requesting modelling to be carried out;
- Will provide references and links to more detailed and relevant documents when required.

There are certain areas that this document does not cover, but have been described in the previously published 'Technical reference guide' for modelling (EEA, 2011). These include:

- Methods for combining models and monitoring;
- Non-process based modelling such as statistical interpolation of monitoring data.

1.2 Audience

This document is intended to provide background information for authorities carrying out air quality assessment and planning activities, but is particularly oriented towards applications relevant to the European Air Quality Directive. The document is not intended to provide detailed modelling methodologies and as such it 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.

1.3 What is an air quality model?

A model, in its most general sense, is a representation of the real world. This is true for conceptual models, for physical models or for complex mathematical models. We use models to understand the world and to represent it in an understandable way. Air quality models are no different in this sense. We use air quality models to represent processes that result in varying concentrations of varying pollutants, often with the major aim of predicting/prognosing air quality given a certain real world situation. In this sense the model produces a quantifiable result, one that can also be measured and compared. An air quality model can be of various forms. It may be conceptual (e.g. we expect high pollution levels when the wind speed is low), it may be empirical (e.g. the concentrations we measure show a statistically significant dependence on the volume of traffic), or they may be process oriented (e.g. we can mathematically represent in some way the process of dispersion). In this document we concentrate largely on the process oriented models, sometimes called 'source models', that are capable of converting emissions (or proxies for emissions) to ambient air concentrations.

In essence all models are parameterisations, i.e. a number of parameters (information) are used to calculate a result. The concept 'parameterisation' is often used to refer to models where the processes are not well

represented but where at least the relevant parameters that influence the result are included. Parameterised models, or parts of models, are often combined with mathematical models describing physical processes. Most air quality models are some combination of both of these.

It is desirable to have as many physical processes as possible represented by the model, but this can quickly lead to a level of complexity that not only makes calculation using numerical methods impossible but also puts high demands on the quantity and quality of information needed to apply the models. Many air quality models are thus a balance between speed of calculation, available information and understanding/representation of the processes. For operational applications the balance tends more to favour speed and less available information, whilst for research applications the balance tends to favour process understanding and detailed information. This is reflected in the concept of 'fitness for purpose'.

It is useful, as with many process models, to separate air quality models into different elements. Of course this will vary, dependent on the model used. For statistical empirical models there may be no need to split the model, but for complex process based models this will be the case. The major elements of an air quality model are:

- Emissions (major source of most pollutants);
- Wind fields and turbulence (providing meteorological information for the dispersion model);
- Dispersion processes (transport and dispersion of the pollutants);
- Chemical processes (chemical transformation of the pollutants);
- Removal processes (wet and dry deposition);
- Boundary and initial conditions (initial state of the model and conditions at the model boundaries).

All these elements are found in this model application document with particular emphasis on the first four aspects.

1.4 Fitness for purpose

When applying any modelling system it is important to keep in mind the suitability of the model for the specific information required and the environment in which it is to be applied. In other words, its 'fitness for purpose'. In regard to NO_2 this can take on many forms. For instance, some simplified chemistry schemes are very suitable in cold climates but are not accurate in warmer sunnier regions. There are also a variety of dispersion model types and some of these may be very suitable for determining annual mean concentrations but will not be suitable for calculating hourly means or percentiles.

The different AQ Directive applications will place demands on the suitability of a model. For assessment applications the aim of a model is to reproduce the existing air quality situation as well as possible, but for planning purposes the air quality model is required to predict the changes in air quality. This later application requires that the 'dynamic response' (the ability of the model to correctly represent changes in concentrations as a result of changes in emissions or meteorology) is well represented by the model, whilst the former makes no requirement on this.

1.5 Air quality modelling and management

Air quality modelling is an integral part of management. In Figure 1 the connections between modelling, management and monitoring are schematically represented. Whilst air quality assessment has traditionally been carried out using monitoring, it is not possible to carry out planning activities without the use of some form of air quality model. For assessment purposes the combination of models and monitoring, through data assimilation methods, should provide the most comprehensive information for understanding and assessing the current air quality situation.

It is common and useful to separate the physical scales of the modelling. Throughout this document we refer to the following spatial scales and typical model resolution:

Local/hotspot scale	(Resolution: 1 - 100 m; extent: 1 – 3 km)
Urban/agglomerate scale	(Resolution: 1 - 10 km; extent: 20 - 200 km)

Regional scale (Resolution: 10 - 50 km; extent: 200 km - continental)

There is a variety of models currently in use in Europe and the development of models has been carried out, to a large extent, independently by different countries and institutes within countries. Some of these models have been harmonized nationally but there are currently no models in Europe that can be considered to be 'standard'. An overview of models currently in use in Europe can be found through two repositories. These are:

- COST 728/732 model inventory hosted by the University of Hamburg (http://www.mi.uni-hamburg.de/Model-Inventory.6295.0.html?&no_cache=1)
- Model Documentation System (MDS) established by the European Environment Agency (http://air-climate.eionet.europa.eu/databases/MDS/index_html).

The first of these is more technically oriented towards model developers whilst the second is oriented towards model users, providing more general information.

In addition it is important to note that the US EPA provides a range of 'preferred/recommended' and 'other' models through their 'Support Center for Regulatory Atmospheric Modeling (SCRAM)' web site (www.epa.gov/ttn/scram/). At this site a number of often used models are available along with extensive documentation and data sets used for validation. Within Europe no such preferred or recommended model(s) exists.

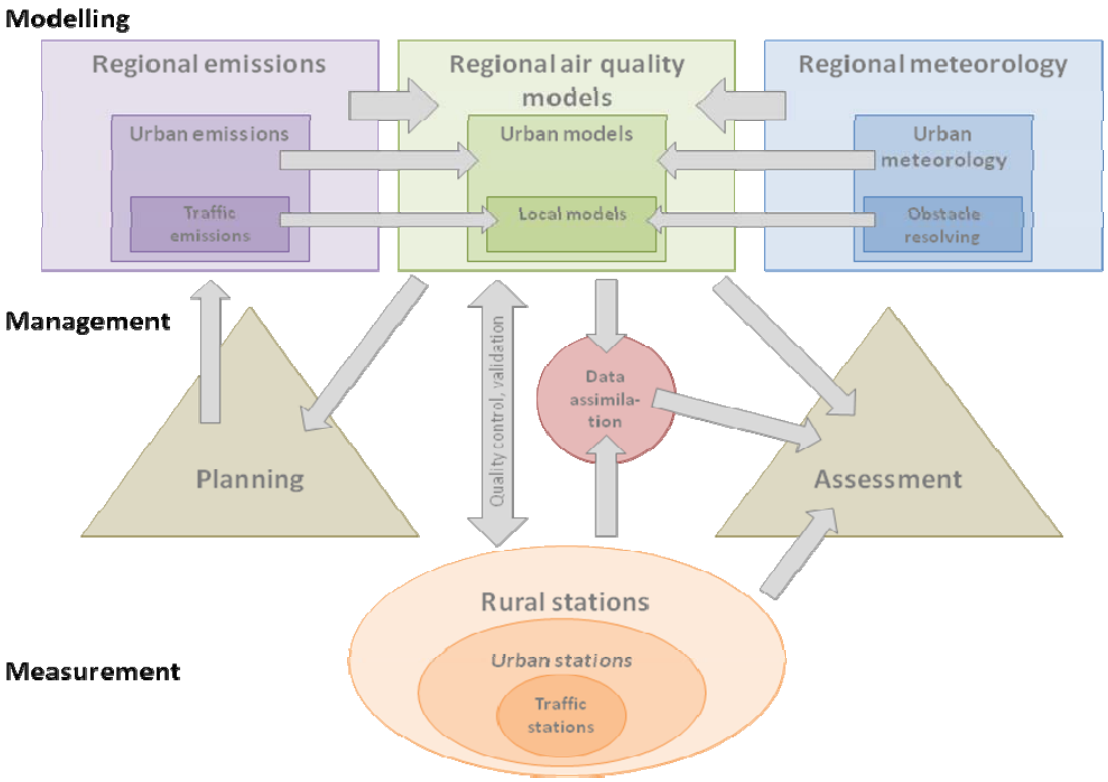

Figure 1: Schematic representation of air quality management and the connections with modelling and monitoring activities.

1.6 Applications of modelling for the European Air Quality Directive

This document will deal chiefly with applications of models in regard to the European Air Quality Directive (EC 2008). These applications include:

1. Assessment and reporting of the exceedance of limit values
2. Assessment of source contributions (source apportionment)

3. Long term air quality planning and management
4. Short term mitigation and forecasting of air quality

In regard to exceedances there are currently two limit values applicable for NO_2. These are the annual mean limit value of 40 μg/m^3 and the 19'th highest hourly mean concentration of NO_2 in a calendar year (99.78 percentile) that should not exceed 200 μg/m^3. In general it is the annual mean limit value that is the most limiting, i.e. there are rarely exceedances of the hourly mean limit value if there are not exceedances of the annual mean limit value. This is illustrated in Figure 2 where annual mean concentrations are plotted against the 19'th highest hourly mean concentrations of NO_2, taken from all of Europe (AirBase, 2011) for the years 2006 – 2008. From a total of 5623 data points only on 9 occasions is the 99.78'th percentile limit value exceeded when the annual mean limit value is not. Accurately predicting, using models, the 19'th highest hourly mean concentration is a difficult task for any model and so most of this document will concentrate on modelling of the annual mean concentration of NO_2. However, for short term mitigation applications it is necessary to provide predictions of hourly mean concentrations.

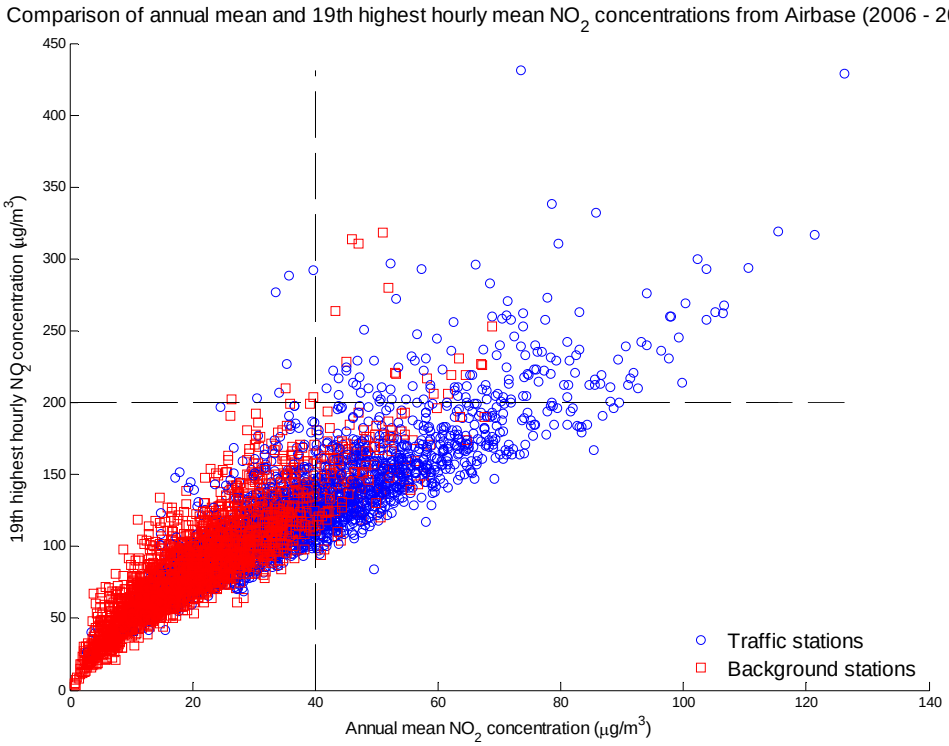

Figure 2. Relationship between the annual mean concentration of NO_2 and the 99.78'th percentile (19'th highest hourly mean concentration). Data taken from AirBase (2006-2008). In total 1987 traffic station data points and 3636 background station data points are shown. Only on 9 occasions is the 99.78'th percentile limit value exceeded when the annual mean limit value is not. Dashed lines indicate the respective limit values.

1.7 Overview of the document

The document is separated into a number of chapters. It starts by addressing Modelling applications for the AQ Directive (Chapter 2). These include the following:

- Notification of postponement
- Assessment
- Source apportionment
- Planning and emission scenarios

It then goes on to describe the major components of air quality modelling (Chapters 3 – 6). These include:

- Dispersion and transport
- Chemistry
- Emissions
- Meteorology

Within these chapters a number of model types and formulations are described. Where appropriate these are presented in the following three subjects:

  - *Description:* Overview of the model or method and its main application area
  - *Examples:* Mostly tabulated examples with short descriptions and references
  - *Application guidance:* Indicative guidance when applying the models or methods

The *Application guidance* is intended as a forerunner to actual recommendations on each of the topics. At this stage they are presented as indicative guidance on particular issues, rather than fixed recommendations, and these will be further discussed within FAIRMODE by the community.

The document concludes on one of the most important aspects of modelling, Quality control and evaluation (Chapter 7).

2 Modelling applications for the AQ Directive

2.1 Modelling requirements for planning and notification of postponement

The European Commission requires detailed information on the current and future air quality management plans of a Member State if a 'notification of postponement of the attainment deadline' (COM2008-403) is to be delivered for NO_2 (or Benzene). Information concerning this notification can be found on http://ec.europa.eu/environment/air/quality/legislation/time_extensions.htm. This information is summarised in a number of forms (SEC2008 2132) but should chiefly be provided through accompanying documentation. This accompanying documentation will to a large extend be based on modelling activities, since the impact of emission reductions are difficult, if not impossible, to assess without the application of models of some form.

A separate guidance document 'Commission Staff Working Paper concerning guidance on preparing a notification of a postponement of the deadline for attaining the limit values for NO2 under Directive 2008/50/EC on ambient air quality and cleaner air for Europe' is available and provides more detailed information on reporting requirements (http://ec.europa.eu/environment/air/quality/legislation/pdf/sec_2011_300.pdf).

In regard to modelling of NO_2, the notification will need to include information relating to:

1. Air quality assessment: A recent assessment (reference year) of the observed/modelled air quality including the level of exceedance of the limit value (Form 2).
2. Source apportionment: An assessment (reference year) of the cause (source apportionment) of the observed/modelled concentrations and exceedances (Form 3a and 4a).
3. Air quality 2010: An assessment for the original deadline year (2010) of the expected/assessed air quality and its causes (Form 4a). This includes an assessment of the impact of measures (Form 7 Annex A) and why already implemented plans have not been successful in achieving the limit values (Form 3b).
4. Predicted air quality 2015: A description of the air quality plan (Form 5a, 7, 7 Annex B), the impact of the emission measures (Form 5b) and an assessment of the predicted air quality without these measures (Form 4b) for the extended deadline year (2015).

In addition to these air quality assessments some other relevant sets of information need to be addressed. These include:

1. A justification for the non-implementation of suggested/recommended measures (Form 6).

2. A demonstration that other EC Directives, particularly those relating to emissions, have been fulfilled (Forms 8, 9).

Given the above the most important modelling aspects are:

1. Air quality assessment
2. Source apportionment
3. Planning and emission scenarios

Examples

All notifications and decisions for NO_2 extensions are publicly available through the Commission website at http://circa.europa.eu/Public/irc/env/ambient/library?l=/notifications_extensions&vm=detailed&sb=Title. The following two examples of requests for postponement are provided for illustration.

The Netherlands

The Netherlands has been granted a derogation period (postponement) by the European Commission for NO_2 until the end of 2014. To monitor the development of the air quality, a combined monitoring and modelling system has been developed and is presently being implemented. The system has several dispersion models (among which SRM-1 and SRM-2) and contains the best estimates for the development of emission factors and background concentrations in the near future. It uses a large amount of information on ten thousands of streets (all streets in those parts of the Netherlands where air quality may be an issue), to calculate the expected PM_{10} and NO_2 concentrations between the present day and 2015. Every year an assessment is made to determine if additional measures are needed, by the government or by local authorities, to comply with the limit values at the end of the derogation period (e.g. Beijk et al., 2010).

http://circa.europa.eu/Public/irc/env/ambient/library?l=/application_extensions/nl/notification/notification_official&vm=detailed&sb=Title

United Kingdom

The United Kingdom (UK) was granted a postponement of the deadline for attaining the annual limit value for NO_2 until 1 January 2015. In the UK, the NO_2 monitoring assessment is delivered through the Automatic Urban and Rural Monitoring Network (AURN). Models are used to supplement the annual monitoring assessments. The GIS-based Pollution Climate Mapping (PCM) air dispersion model is used to estimate annual mean NO_X concentrations at background and roadside locations. Nitrogen oxide emission estimates from the UK National Atmospheric Emission Inventory (NAEI) are a model input. For the notification, the NAEI was also the basis for NO_x and NO_2 emission projections. The base year 2008 was used for the modelled AQ assessment and for calculating the baseline NO_2 concentration projections for the new attainment year 2015. The projection provides the information required to develop the air quality plans.

http://circa.europa.eu/Public/irc/env/ambient/library?l=/notifications_extensions/uk/notification_22092011/official_notification/110921_technical/_EN_1.0_&a=d

2.2 Modelling requirements for assessment and reporting exceedances

In chapter 6 of the Technical reference guide for modelling (EEA, 2011) most of the aspects in regard to assessment and reporting of exceedances are presented and discussed. We provide here a summary of the information in the AQ Directive that is relevant for NO_2 modelling.

2.2.1 Assessment

In the current AQ Directive (EC, 2008) modelling alone can only be used for reporting clear non-exceedance. This is when the concentrations are below the lower assessment threshold, as defined in the AQ Directive (Appendix II, 2008/50/EC) for any particular pollutant. 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. Above the upper threshold fixed measurements must be used as basis for the assessment and modelling may be used as supplementary material. These threshold values are summarised below.

Lower assessment threshold

For NO_2 the lower assessment threshold value is given by:

- Hourly means: 50% of the limit value (i.e. 100 µg/m^3, not to be exceeded more than 18 times in any calendar year)
- Annual mean: 65 % of limit value (i.e. 26 µg/m^3)

Upper assessment threshold

For NO_2 the upper assessment threshold value is given by:

- Hourly means: 70% of the limit value (i.e. 140 µg/m^3, not to be exceeded more than 18 times in any calendar year)
- Annual mean: 80 % of limit value (i.e. 32 µg/m^3)

2.2.2 Quality objectives for NO_2 modelling in the AQ Directive

Associated with the modelling of NO_2 is also a quality objective (Appendix I, 2008/50/EC). This quality objective is defined for both hourly and annual NO_2 mean concentrations and is defined to be:

- Hourly: an *uncertainty* of 50%;
- Annual: an *uncertainty* of 30%.

Interpretation of the AQ Directive term '*uncertainty*' for modelling is provided in Section 7.1 of this document and more extensively in Section 3.6 of the Technical Reference guide for modelling (EEA, 2011).

In general, the quality objective of 30% for annual mean concentrations should be obtainable by models when they are applied below the upper threshold limit. However, as is pointed out in the discussion in the Technical reference guide for modelling, it is strongly dependent on interpretation of the AQ Directive text and the actual concentration levels of pollutants.

The hourly mean quality objective of 50% for NO_2 may be achievable for models below the lower threshold limit but may be very difficult to achieve in more polluted environments, especially when levels are strongly episodic.

2.2.3 Reporting of exceedances

The method for reporting air quality assessment to the European Commission (in regard to modelling) has been presented and discussed in the Technical Reference guide for modelling (EEA, 2011), Section 4.1. The reader is referred to that document for more information.

2.3 Source apportionment

The identification of pollutant sources and the accurate quantification of their individual contributions to the ambient pollutant levels, here revered to as source apportionment, is an integral part of any successful Air Quality Management Plan (AQMP). Source apportionment of NO_2 is therefore particularly valuable, especially in areas where routine observations reveal high concentrations that are close to or exceeding the EU limit value.

Source apportionment methodologies for NO_2, as for other pollutants e.g. particulate matter or volatile organic compounds, rely both on modelling as well as on monitoring data. When applying any source apportionment methodology as part of an AQMP, it is important to ensure that the selected approach will enable the researcher/authority to:

- confirm the principal source to which exceedances of NO_2 limit values can be attributed. In the case of NO_2 the responsible sector is mainly road traffic, although in many areas industrial plants and/or power generation are also relevant sources.
- quantify what proportion of the exceedances of NO_2 is due to background emissions or local emissions from busy roads in the area of interest, in order to decide on whether local traffic management measures or national measures would be the most suitable approach for reducing emissions and achieving EU air quality objectives.

- determine the extent to which different vehicle types and driving conditions contribute to NO_2 traffic emissions, in order to examine the efficiency of relevant traffic management scenarios.

Most source apportionment studies in European urban areas indicate that road traffic is the most significant contributor to the total NO_2 concentrations and that the most important sources are heavy duty vehicles and stationary vehicles in queues.

2.3.1 NO_2 source apportionment using monitoring data

There are a number of methods for carrying out source apportionment using monitoring alone, but these are more limited for the case of NO_2. This usually involves the use of station pairs or triplets, allowing the different scales (regional, urban and local) to be assessed by subtracting the 'background' contributions. In addition, methods using wind roses may also be applied for defining directions of sources and in particular for determining if traffic stations are measuring the up or downwind concentrations from a particular road.

For a complete assessment of the source contributions in an urban area more information is required than is available through monitoring alone, especially in the case of a reactive pollutant such as NO_2. For instance, it is not only emissions of NO_x and primary NO_2 that will affect the NO_2 concentrations but also the availability of ozone for oxidation. As such, emissions from a neighbouring country or region that affect ozone, rather than NO_x, can also affect NO_2 concentrations in a transboundary fashion. The same can be true on the local and urban scale. The contribution to NO_2 concentrations from a road located in the centre of a large city could be significantly less than one with the same NO_x emissions on the upwind outskirts of the city since it is there that ozone is most readily available. As such source apportionment must be carried out in a holistic fashion and on all scales if the sources are to be correctly apportioned.

Due to these non-linear chemical reactions affecting NO_2, source apportionment for NO_2 is not as physically meaningful or understandable as it may be for the more non-reactive NO_X. Therefore it is recommended that a source apportionment for NO_X be calculated along with an indicative source apportionment for NO_2. They will often be quite similar but a source apportionment for NO_2 may highlight particular vehicle types for which primary NO_2 emission is important.

2.3.2 NO_2 source apportionment using air quality modelling

The most common and comprehensive approach to source apportionment is based on the direct numerical simulation using air quality models. A sensitivity method can then be used, where a model base case simulation is set up and then repeated with perturbed emissions from a particular source, e.g. industry or diesel traffic. The difference in the concentration output between the base case and the sensitivity case accounts then for the effect of changing emissions from that source on air pollutant concentrations at all receptor sites. This sensitivity approach using dispersion models for NO_2 source apportionment enables the apportionment of the contribution of each identified source to the overall NO_2 ambient levels at any given point in space and time.

Because NO_2 is a reactive species and its formation from NO will also be dependent on the levels of ozone available, the total NO_2 concentrations will not simply be the sum of the individual emissions. When carrying out source apportionment studies for NO_2 it is very useful to also apportion NO_X, as this is a much more conserved and linear quantity. Doing so will provide more information on the contribution of the individual NO_X emissions sources.

The starting point of such a system is the emissions inventory. Information concerning the emission rates of NO, NO_2, VOC, and even CO when complete chemical schemes are used, are required on the local and urban scale. Further to the local emissions it is also necessary to have good source of information in regard to the boundary conditions of the urban region or, in cases where the modelling system covers larger regions, also the emission inventories from neighbouring regions or countries. This is one advantage of nested modelling systems that provide not just information concerning the local source contributions but also regional and transboundary contributions.

In regard to transboundary transport it is useful to review the EMEP Source Receptor matrices (www.emep.int/SR_data/index_sr.html) to assess the impact of emissions from other countries. Unfortunately NO_x and NO_2 are not included in these matrices and so for NO_2 they are not very useful. However, the impact of

NO_x and VOC emissions on ozone (SOMO35 and AOT40) are a part of these matrices and can be indicative of the impact of these emissions on at least ozone concentrations.

The disadvantages of this methodology relates to the computer resources required, as well as to the need for a thorough emissions inventory, without which such modelling cannot be usefully applied. A lot of effort and time for developing and evaluating the model may also limit the applicability of using complex air quality models. Such an evaluation must also include a validation of the 'dynamic sensitivity' of the model, i.e. if the model response to changes in emissions is appropriate or not (see Section 7.3).

2.3.3 NO_2 source apportionment using receptor modelling

An alternative to using direct monitoring data or complex dispersion models is the application of receptor models, which statistically associate measurements of air quality at selected receptor sites with potential sources. Typical methods include Chemical Mass Balance (CMB) analysis, statistical algorithms such as regression, Principal Component Analysis (PCA) and Positive Matrix Factorization (PMF). Such methods are regularly applied to the source apportionment of particulate matter or VOC's, using the chemical analysis of the particles or compounds to provide the information necessary for separating the sources. These methods provide the source contributions at the measurement site but do not directly provide emission factors for the sources. Dispersion models are required for this. It is also important to note that receptor modelling usually requires intensive monitoring campaigns that are usually limited in time and spatial coverage.

In the case of NO_2 the chemical composition of NO_2 is the same no matter which source is emitting. As such the use of receptor modelling for NO_2 source apportionment will require other information, e.g. other pollutants that are emitted in different proportions from the different sources. In a study by Thornhill et al. (2010), the U.S. EPA PMF 3.0 receptor model has been applied to resolve gasoline engine exhaust from diesel exhaust and to calculate NO_X emission factors associated with each fuel. To do this a large number of exhaust related compounds were measured, including CO, benzene, toluene, alkylated aromatics, formaldehyde, acetaldehyde, acetone, ammonia, particle number, $PM_{2.5}$, and black carbon (BC). As a result, the emissions of NO_X related to diesel and gasoline vehicles, under a variety of driving conditions could be assessed. Bruno et al. (2001) also used measurements of a range of pollutants (CO, NO_X and benzene toluene m+p-Xylene (BTX)) and applied PCA analysis to these to identify two major contributing sources. Coupling this information to known traffic volumes and known dispersion conditions allows the derivation of 'real world' emission factors for the various vehicle categories under differing driving conditions.

SO_2 has also been used in a number of studies to differentiate between industrial and traffic contributions, based on the ration of SO_2 to NO_X. Such studies have been undertaken by Yuval et al. (2007) and Nirel and Dayan (2001) using differing statistical methods which also included the use of meteorological data and the known temporal variation of the emission sources.

Application guidance

1. Direct assessment of monitoring data at local (traffic/industry), urban and regional sites combined with meteorological data can provide preliminary information concerning the sources of NO_2. However, this information is limited to the individual site and does not provide direct information on emissions.
2. Receptor modelling can be used to help establish 'real world' source contributions and relative emissions for various vehicle categories and other sources. Dispersion models are required to infer emission factors from source contributions. Such methods can be considered if emission factors are considered to be sufficiently uncertain.
3. Air quality modelling is the only method that can provide 'urban wide' source apportionment assessment.
4. Source apportionment should be calculated for both total NO_X and for NO_2 in all instances if possible.
5. Air quality modelling requires a well established and comprehensive emissions inventory.

2.4 Planning and emission scenarios

The main advantage of air quality modelling is its usefulness as a planning tool. 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 policy implementations, that either effect the emission factors themselves (technological) or the activities (structural). As such a policy maker may come with a range of possible strategies that will need to be quantifiably assessed using air quality models, or an air quality modeller will come with an emission requirement that a policy maker must find a way of implementing. In either case, the air quality 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, e.g. reducing the emission of NO_X on a particular road by a particular amount will reduce the contribution from that particular road to the total NO_X concentration proportionally. However, NO_2 is non-linearly related to NO_X (see Figure 4) and the reduction in the ambient NO_2 concentration will depend on other factors as well, including any changes in primary NO_2 emissions, the amount of available ozone, changes in other pollutant emissions such as VOC, the level of background NO_2 and the distance from the source. So, though the primary driver behind NO_2 concentrations in the urban environment is no doubt NO_X emissions it is not the sole process.

One aspect that is often not discussed or presented when implementing emission scenarios is the uncertainty and variability in the calculations. Variability in the calculated concentrations for future emission scenarios will occur chiefly as a result of the inter-annual variability of meteorology. This will affect aspects such as dispersion conditions, atmospheric transport and production of ozone, as well as having an impact on the energy production (heating/coolling). The major source of uncertainty in scenario calculations includes both the uncertainty in the model and uncertainty in the current and future emission scenarios. These uncertainties and variability should be included in any scenario calculation if air quality models are to be useful applied for decision making purposes.

Application guidance

1. Some indication of the uncertainty in the predictions should be given. If necessary multiple runs reflecting the emission and model uncertainties may be made to indicate these uncertainties
2. Scenario calculations should be carried out for a range of meteorological years, which also reflect the variability in ozone 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.
3. In regard to NO_2 it is important that the chemical scheme, if parameterised, can reflect the changes in emissions, background concentrations and meteorology, e.g. it should be able to represent changes in NO_2/NO_X emission ratios and should reflect the impact of changes in ozone levels.

3 Dispersion and transport

Dispersion modelling, combined with chemistry, provides the physical link between emissions and concentrations. In many urban environments the major NO_2 pollutant source is traffic. If concentrations are to be correctly modelled near traffic sources then specialised dispersion models are required for open road and street canyon environments. NO_2 is not, however, just a near source pollutant problem. Large areas can be highly polluted and this requires both urban and regional scale models to effectively describe the total pollutant problem.

Dispersion models can generally be categorised by their type (e.g. Gaussian, Lagrangian, Eulerian) or by their application (e.g. street canyon, urban models). In general a dispersion model will consist of a meteorological part, a pollutant dispersion part and a chemical part. Sometimes these parts are inseparable, e.g. a particular Gaussian dispersion model will have been developed using a particular meteorological pre-processor, or they may be interchangeable, e.g. Lagrangian particle models may use almost any wind field. Different models use different combinations of the meteorological, dispersion and chemical parts and as such it is not always straight forward to categorise an individual model. Table 1 lists the different major model types, indicating their suitability (fitness for purpose) for the different applications. In the following chapter we look at the major model types and their related applications as follows:

1. Gaussian open road line source models
2. Street canyon models
3. Lagrangian particle models (various applications)
4. Urban scale models (Gaussian/Lagrangian and Eulerian)
5. Regional scale models (mainly Eulerian)

Table 1. Fitness for purpose matrix for dispersion models. Shown are the four major model types (columns) and applications (rows). Fitness for purpose is indicated by colour and appropriate comments. Green = 'fit for purpose'; Orange = 'conditionally applicable'; Purple = 'not fit for purpose'.

Model types and applications	Gaussian models	Lagrangian particle models	Obstacle resolving Eulerian models (CFD)	Terrain resolving Eulerian models
Open roads			No obstacles, computationally expensive	Unresolved
Street canyon	In combination with parameterised wind field model	In combination with parameterised wind field model	Computationally expensive	Unresolved
Urban scale	Requires homogenous meteorology	Computationally expensive	Not computationally feasible	
Regional scale	Requires homogenous meteorology	Computationally expensive	Not computationally feasible	

3.1 Gaussian open road line source modelling

Description

Most open road line source (ORLS) models are based on the Gaussian slender (thin) plume approximation that is also extensively used for calculating concentrations from industrial stack emissions. ORLS models are intended for use where there are few, or no, obstacles and to calculate concentrations within a few hundred meters of the roads themselves. These are so called 'steady state' models, which means that they represent the concentration distribution over a period of time that is:

- longer than the travel time of the pollutant
- suitably long to average out the variability arising from turbulent fluctuations
- long enough for the emission source to be considered constant in time

As such they can be used to calculate mean concentrations on time scales of roughly an hour and upwards relatively close to source. Most models use turbulence parameterisations that are valid for one hour.

The major components and characteristics of Gaussian ORLS models are:

1. A meteorological pre-processor that takes observed or modelled meteorology and adapts it to the dispersion model. Most importantly turbulent intensity and wind speed at emission height are needed for the dispersion model.
2. The Gaussian slender plume equations. These should include, at a minimum, reflection from the ground surface but may also include reflection from the top of the boundary layer (See Box 1).
3. Parameterised chemical schemes are often employed for NO_2 applications.

4. A parameterisation that takes into account the initial dispersion caused by the traffic induced turbulence.
5. A numerical or analytical integration scheme that allows the calculation of the concentrations from continuously emitting straight line sources.
6. Some models may treat the road as an area or volume source, rather than a line source (e.g. OML-Highway). Though the dispersion equations are very similar the initial distributions of the plumes are treated differently.
7. These models may include wet and dry deposition.

Box 1: Mathematical formulation of Gaussian open road line source models

The basic equations describing Gaussian ORLS models gives the concentration (C) as a function of the total emissions of the line source (Q), divided by the wind speed (U) at the emission height (h), times the line integral of the Gaussian dispersion function (f).

$$C = \frac{Q}{U_h} \int_0^D f.dl \qquad (3.1)$$

The Gaussian dispersion function (f) is dependent on the dispersion parameters (σ) in the horizontal (y) and the vertical (z) direction.

$$f = \frac{1}{2\pi\sigma_y\sigma_z} \exp\left(-y^2/2\sigma_y^2\right)\left[\exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(z+h)^2}{2\sigma_z^2}\right) + \ldots\right] \qquad (3.2)$$

The function f describes the dispersion at a point in space (x,y,z). The dispersion parameters (σ) represent the size (standard deviation) of the Gaussian plume and are dependent on time travelled in the downwind direction (x). These increase at a rate proportional to x gradually decreasing to $x^{1/2}$, dependent on the time travelled. The two Gaussian terms in the square brackets describe the vertical dispersion from the source and from its reflection on the gound. The singularity that occurs in the equations when $U = 0$, *i.e.* $C \rightarrow \infty$, is usually avoided by setting a minimum wind speed.

Examples

There a number of such models available as well as a number of inter-comparison and validation studies. The following table provides an overview of some, but not all, ORLS models and relevant studies.

Table 2. Table showing a number of open road line source models (ORLS) used in Europe. When available the model names are linked to the EEA Model Documentation System (MDS). Often these models may be imbedded within modelling systems.

Model (MDS link)	Comments/description	Links to documentation, validation and inter-comparison studies
WORM	Gaussian line source model developed by the Norwegian Institute for Air Research (NILU)	Berger et al. (2010)
HIWAY	US EPA Gaussian line source model, best suited for at-grade highways	Petersen (1980) Berger et al. (2010)
CALINE 3 and 4	US EPA Gaussian line source model developed by the California Department of Transportation	http://www.dot.ca.gov/hq/InfoSvcs/EngApps/

CAR-FMI	Gaussian line source model developed by the Finnish Meteorological Institute (FMI)	Härkönen (2002) Berger et al. (2010)
IMMIS^{luft}	Multiple source Gaussian model for long term means. Uses statistical meteorology	http://www.immis.de/e IVU UMWELT (2008)
OML-Highway	Gaussian line source model developed by the National Environmental Research Institute (NERI), Denmark.	Berkowicz et al. (2007) Berger et al. (2010)
ROADWAY-2	Developed with support of the U.S. National Oceanic and Atmospheric Administration (NOAA)	Rao (2002)
SRM-2	Based on a roadside dispersion model developed by TNO. Legislated as the standard calculation method (2) in The Netherlands for a number of road configurations.	(van den Hout, 1988) http://wetten.overheid.nl/BWBR0022817/
ADMS-Roads	Multiple source Gaussian-type model developed by CERC, UK. Applicable for both open roads and street canyons.	www.cerc.co.uk/environmental-software/ADMS-Roads-model.html
IMPACT	Aria Impact is a second generation local scale Gaussian model that can be applied to industrial plumes and line source traffic emissions. It is applicable for calm wind situations, includes NO/NO_2 conversion and canyon effects in the urban environment.	http://www.aria.fr/english/aria_impact.php
PROKAS_V	Multiple source Gaussian model for long term means. Uses statistical meteorology	Flassak, Th., Bächlin, W., Bösinger, R. (1996) http://www.lohmeyer.de/eng/Software/default.htm

Application guidance

The following points should be noted when implementing Gaussian ORLS models:

1. These types of models are intended for use where there are no obstacles, e.g. buildings, surrounding the road. However, some Gaussian models may include a parameterisation of the effect of road side barriers, e.g. SRM-2 (van de Hout, 1988). They should not be used within street canyons unless the model is specifically designed to deal with that situation, since the meteorology and dispersion conditions around open roads and street canyons are quite different.
2. The slender plume approximation is not suitable for low wind speed conditions (< 1 m/s at emission height) as equation 3.1 approaches a singularity. The errors that occur with the model under such conditions may be significant. This is generally dealt with by assuming a minimum wind speed.
3. They often work optimally when the wind is blowing roughly perpendicular to the road. Significant errors or uncertainties can occur when the wind is blowing parallel to the road. The error may be dependent on the line source numerical integration scheme used but there is also a large intrinsic uncertainty to these models. Small changes in wind direction, when the wind is near parallel to the road, will lead to large changes in the predicted concentrations.
4. They are suitable for chemically inactive species, since the theoretical formulation behind this model cannot account for chemically reactive species. For the application of NO_2 this means that parameterised chemistry must be implemented separately.

5. They perform optimally when the stability is in the range of stable to unstable. Highly stable or highly unstable conditions may not be well modelled using the standard slender plume approximation. Some models may have special schemes to deal with these conditions, e.g. plume meandering in stable conditions or a non-Gaussian plume description for convective conditions.
6. The traffic induced turbulence parameterisations range from providing simple constant initial dispersion parameters to time decaying turbulent kinetic energy parameterisations. Studies have shown this to be an important element of the dispersion from traffic sources. When modelling near source traffic induced turbulence should be included in the model formulation in some way.
7. Though such models are often applied on an hourly basis the limitations of their formulation assumptions (e.g. steady state, slender plume, no obstacles, etc.) and on suitable input data (meteorology and emissions) can result in significant uncertainties at these short time scales. As a result they are more suitable for longer term average calculations and cannot be expected to provide accurate hourly predictions.
8. Due to the steady state nature of these models they cannot be applied over long distances with any certainty. In general such models should be used only at distances < 1000 m.
9. All of the available Gaussian ORLS models listed above will likely provide reasonable estimates of the dispersion from line source emissions for the application regions in which they were developed. This is usually the result of an iterative and adaptive process of model development by comparison with measurement data. If such a model is applied in a new application region then it should be reassessed using locally available observations whenever possible.

3.2 Street canyon modelling

Description

In general ORLS models cannot be used to determine concentrations near roads when building obstacles are present, i.e. within street canyons. Street canyon models are used to determine the concentrations when local buildings affect the meteorological and turbulence fields. There are three major types of street canyon models:

- Well mixed box models. Such models assume that pollutants are mixed within the street canyon and use parameterised forms of the vertical exchange of pollutants to determine these concentrations. These are very fast models and are suitable for long term means.
- Parameterised street canyon models. The wind field within the street canyon is described in a parameterised way, based on the local building geometry. Gaussian plumes then use this parameterised wind field to advect and disperse the pollutant. These models can be used to calculate hourly means but are also more suitable for longer time scales, as in the ORLS models.
- Obstacle resolving models. These are high resolution flow models that can resolve buildings. These are based on CFD (Computational Fluid Dynamics) models of two types; the more often applied RANS (Reynolds Average Navier Stokes) models and the LES (Large Eddy Simulation) models. RANS models represent a 'steady state' solution to the meteorological field whilst LES models represent the time evolving meteorological fields. Pollutants may be dispersed in the complex meteorological field using either Lagrangian particle models or Eulerian methods. These models are computationally intensive.

A thorough review of street canyon models was carried out by Vardoulakis et al. (2003). In that review a number of models are described in more detail. The models described include statistical models, box models, parameterised Gaussian models and obstacle resolving models. The authors provide a deeper discussion of their advantages and drawbacks.

Examples

Table 3 provides some examples of parameterised street canyon models that may be used for Directive related applications. In general obstacle resolving CFD models are not used for Directive related applications and are

not further described here. A complete list of CFD used for street canyon modelling may be obtained through the COST 728/732 model inventory system (www.mi.uni-hamburg.de/MicroMet.6553.0.html?&no_cache=1).

Table 3. Table showing a number of street canyon models used in Europe. When available the model names are linked to the EEA Model Documentation System (MDS). Often these models may be imbedded within modelling systems.

Model (MDS link)	Comments/description	Links to documentation, validation and inter-comparison studies
OSPM	Operational street-scale model, based on a combined plume and box model.	Berkowicz (2000, 2008)
AEOLIUS	Based on OSPM but with some adaptations. Used in the UK.	Buckland (1998)
IMMIS^{cpb}	for the calculation of complex individual case studies and short term calculations of air pollutant's transmission in street canyons	http://www.ivu-umwelt.de/e
SEP_SCAM	A combined plume and box model	Papathanassiou et al. (2008)
CAR II (SRM-1)	Developed by TNO and applied in URBIS, The Netherlands. Highly parameterised model for a range of street canyon configurations (annual means only). Also known as SRM-1, legislated as the standard calculation method (1) in The Netherlands near roads (< 60 m). Access through a web based version available.	(van den Hout, 1988), den Boeft et al. (1996), Wesseling and Sauter (2007) http://wetten.overheid.nl/BWBR0022817/ http://car.infomil.nl/Login/Login.aspx?ReturnUrl=%2fScenarios%2fNew.aspx
ADMS-Roads	Multiple source Gaussian model with street canyon module based on OSPM methodology.	www.cerc.co.uk/environmental-software/ADMS-Roads-model.html
MSS (Micro Swift_Spray)	Multiple source Lagrangian Particle Dispersion Model (MicroSpray) applicable at microscale with obstacles	www.aria-net.it www.aria.fr Tinarelli *et al.*, 2007, 2009
WINMISKAM	Three-dimensional prognostic flow model coupled with an Eulerian dispersion model.	Olesen, H.R., Berkowicz, R., Ketzel, M., Løfstrøm, P. (2009) http://www.lohmeyer.de/eng/Software/default.htm
PROKAS_B	Precalculated results (nondimensionalized) of the microscale flow- and dispersions model WINMISKAM for 21 building patterns are used for the calculation of complex individual case studies of air pollutant's transmission in street canyons	http://www.lohmeyer.de/eng/Software/default.htm Flassak, Th., Bächlin, W., Bösinger, R. (1996)

Application guidance

The following points should be taken into account when applying street canyon models:

1. Availability of accurate street and building geometry as well as the accuracy of the meteorological input data determines to a large extent the quality of the simulations.

2. Most simpler street scale models tend to perform better for intermediate wind directions, i.e. neither exactly parallel nor perpendicular to the street axis. Some models revert to separate parameterisations depending on the wind direction, i.e. use separate modules for parallel and perpendicular flows.
3. As in ORLS models the model assumptions and the available representative input data limits the quality of the model results. Hourly calculations with all the above mentioned street canyon model types will be prone to significant uncertainty and as such these models are also most suitable for longer term means.
4. Some models in this category require meteorological input either in the form of vertical wind profiles, extending from the ground level up to at least the roof level for the particular simulation area. In the case of CFD models, such input data are required in order to calculate the necessary inflow boundary conditions. This information is rarely available as part of routine meteorological measurements, therefore appropriate meteorological/flow models need to be used for obtaining the required inflow fields.
5. The models listed above can provide reasonable estimates of pollutant dispersion in street canyons for a range of street configurations and traffic loads. However, the ability to resolve details of the local geometry (e.g. lateral openings), either parametrically or explicitly, varies to a great extent between model formulations. For obtaining yearly averages and inter-annual trends, anyone of the models should be suitable given appropriate input conditions.
6. Despite their superior physical resolution, CFD models are usually limited to simulations of steady state flows over periods of a few hours and for spatial scales covering a few hundreds of meters. This is to a large degree due to their computational intensive nature. However, the improved physical representation of the wind fields in street canyons using such models make them the most suitable model for street canyon modelling. Further efforts to apply such models should be undertaken.
7. If CFD models are to be applied for Directive applications they must be applied in a way that can represent longer periods of time. A method for achieving this is to pre-process a suite of 'steady state' wind fields based on discretely varying meteorological input data. Such fields can then be used, based on the actual meteorological conditions, to calculate average concentrations in the period under consideration, e.g. Parra et al. (2010).
8. It is worth noting the following concluding statement in the street canyon review carried out by Vardoulakis et al. (2003) which provides a recommendation for authorities using the results of street canyon modelling: *"It should be stressed that all mathematical models need thorough validation against experimental data. The accuracy of their predictions is bounded by the accuracy of input data such as emission factors, traffic and meteorological data, street geometry, etc. Therefore, decision-makers should use modelling results cautiously, especially when relevant field measurements are not available."*

3.3 Lagrangian particle modelling

Description

In Lagrangian particle models (LPMs) the concentration is computed by counting 'particles' in a user defined volume (e.g. the cell of a regular grid). Each 'particle' represents a particular mass of one or several pollutants emitted from a given source. Time-dependent trajectories of particles are computed by stochastic differential equations (Langevin equations), which aim at describing turbulence properties (e.g. standard deviations of wind velocity fluctuations, ensemble average of dissipation of turbulent kinetic energy, well-mixed criterion, meandering in low wind speed conditions). Flow and turbulence fields have to be provided either by Eulerian models (e.g. CFD models in built-up areas) or by meteorological pre-processors (e.g. in flat terrain without significant influence of buildings).

Generally a large number of particles are necessary to derive concentration values with a high statistical accuracy. This can be from thousands to millions of particles. Though the advection schemes are quite efficient, the large number of model particles released implies that computation time is usually significantly higher than for the Gaussian models previously discussed. The computation time is directly linked to the number of particles

within the model domain, which in turn is determined by the number of particles released, the size of the model domain and the wind speed (transport time).

Chemical conversions of first order (exponential decay of the particle mass) can be modelled directly, likewise wet and dry deposition and sedimentation processes. More general chemical reactions cannot be carried out directly with these models.

Examples

There are only a few models of this type designed for the local and urban scales, whilst there are a number that are applied on regional scales. A list of these is provided in Table 4.

Table 4 Table showing a number of Lagrangian particle models used in Europe. When available the model names are linked to the EEA Model Documentation System (MDS).

Model (MDS link)	Application region	Comments	Links to documentation, validation and inter-comparison studies
AUSTAL2000	Local/urban Flat and complex terrain Built-up areas Point, line, area, volume sources	Official reference model of the German Regulation on Air Quality Control (TA Luft, 2002). Dry deposition, sedimentation, NO-NO_2 conversion, odour hours. Automatic estimation of the sampling error.	Janicke and Janicke (2002) www.austal2000.de
DIPCOT	Local-regional Flat and complex terrain Point sources	Dry and wet deposition.	Davakis et al. (2003) http://milos.ipta.demokritos.gr/DIPCOT.htm
FLEXPART	Long range	Global scale model linked to ECMWF. Wet and dry deposition.	Stohl and Wotawa (1997) Stohl et al. (2005) http://transport.nilu.no/flexpart
GRAL	Local/urban Flat and complex terrain Built-up areas Point, line, area, volume sources	Odour hours, explicit treatment of low wind speed conditions and tunnel portals.	Oettl and Uhrner (2010)
KFZ.LAG	Local/urban Flat and complex terrain Built-up areas Point, line, area, volume sources	NO-NO_2 conversion.	Schorling (1989, 1999)
LASAT	Local/regional Flat and complex terrain Built-up areas Point, line, area, volume, grid sources	Dry and wet deposition, sedimentation, chemical reactions, radioactive radiation, odour hours, 3-dimensional exhaust dynamics. Automatic estimation of the sampling error. Commercial software	Janicke and Janicke (2007) www.janicke.de/en/lasat.html
MILORD	Long range	Chemical reactions, wet and dry deposition, sedimentation.	Anfossi et al. (1995)

NAME	Long range	Chemical reactions, wet and dry deposition, sedimentation.	Jones et al. (2007)
SPRAY	Local/regional Flat and complex terrain Built-up areas Point, line, area, volume sources	Explicit treatment of low wind speed conditions. Dry and wet deposition, sedimentation, dense gas dispersion.	Tinarelli et al. (1994)
HYPACT	Local to regional scale applications	Used in connection to the RAMS meteorological model	http://www.atmet.com/html/docs/documentation.shtml
TAPM	Local scale to urban scale	Integrated meteorological and air quality model containing a hybrid LPM that uses Lagrangian particle motion in the vertical and Gaussian puffs in the horizontal	http://www.cmar.csiro.au/research/tapm/

Application guidance

LPMs are able to account in detail for three-dimensional wind and turbulence fields. In the near field of emission sources, LPMs can provide a more accurate description of the atmospheric dispersion as compared to models based on the classical diffusion equation. LPMs have been used extensively on local and long range transport applications but have, up to now, only had a limited application in urban scale air quality modelling. The following points should be noted:

1. The models are only as good as the meteorological and turbulence fields used to describe them. In homogenous meteorological conditions, without obstacles and surface complexities, and for moderate vertical variations of the meteorological parameters, LPMs will provide similar results to the faster Gaussian models.
2. Due to the large number of model particles released, the computational time required by these models can be quite large. However, with increasing number of emission sources and higher spatial resolutions, differences in computation times between Lagrangian particle models and Gaussian models become smaller. Even so, the large number of calculations necessary for simulating concentrations restricts the number of sources as well as the domain size that can be used.
3. This type of model should provide a better description of the dispersion and transport of pollutants than the simpler Gaussian models. If higher quality in complex situations is required then such models are often more suitable.
4. LPMs provide a time-dependent description of the dispersion process (in contrast to Gaussian plume models) and a consistent methodology for dealing with both simple and complex dispersion situations. This is an advantage as one does not need to switch from one model to another, if for example different kinds of sources have to be handled simultaneously or if the level of modelling detail needs to be increased.
5. These models are effective when combined with complex wind field models, such as CFD models or mesoscale meteorological models.
6. For standard applications they are restricted to chemical conversions of first order, i.e. linear decay.

3.4 Urban scale modelling

Description

Urban scale models are models that cover an entire city or agglomeration and have a spatial resolution of around 1 – 3 km, which is capable of resolving the variability in the urban background concentrations. These

models do not resolve the variability in the concentrations that occur at street level within the city. There are generally two types of these models:

- Eulerian grid models, that calculate concentrations based on a solution of the advection diffusion equations and chemical equations. These models may have complex chemical schemes.
- Gaussian/Lagrangian type models, that calculate concentrations using multisource Gaussian based models. The Gaussian plumes, emitted from point, line or area sources, may follow Lagrangian trajectories. These models do not generally contain complex chemical schemes and may be limited to steady state or parameterised chemical schemes.

Both types of models require meteorological fields and regional scale boundary conditions.

There are also a set of urban scale models using the Gaussian approximation that are based on long term statistical meteorology (climatology) and provide annual mean concentrations of inert species. These models are often referred to as statistical Gaussian models.

Examples

Table 5 provides examples of Gaussian/Lagrangian type models used for the urban scale and Table 6 provides examples of Eulerian type models applied to the urban scale.

Table 5 Table showing a number of Gaussian/Lagrangian models used in Europe for urban scale air quality applications. When available the model names are linked to the EEA Model Documentation System (MDS).

Gaussian / Lagrangian models (MDS link)	Comments/description	Links to documentation, validation and inter-comparison studies
ADMS-Urban	Multiple source Gaussian model for hourly concentrations including GRS chemical scheme. Developed by CERC, UK	http://www.cerc.co.uk/environmental-software/ADMS-Urban-model.html McHugh et al. (1997)
ATEM	Multiple source Gaussian model for long term means. Applied exclusively in the Czech Republic	Brechler (2000) www.atem.cz
OPS	Multiple source Gaussian-Lagrangian model for long term means developed by RIVM in The Netherlands. Applied to all of The Netherlands.	van Jaarsveld et al. (2004)
URBIS	Multiple source Gaussian model for long term means developed by TNO, The Netherlands	www.tno.nl/urbis
UDM-FMI	Multiple source Gaussian model for hourly means developed by the Finnish Meteorological Institute	Karppinen et al. (2000), http://www.fmi.fi/
IMMIS^{net}	Multiple source Gaussian model for long term means	http://www.immis.de/e
IFDM	Multiple source Gaussian model for hourly means. Can also be applied to line sources	Bultynck and Malet (1972), Olesen (1995)
PROKAS_V	Multiple source Gaussian model for long term means. Uses statistical meteorology	http://www.lohmeyer.de/eng/Software/default.htm Flassak, Th., Bächlin, W., Bösinger, R. (1996)

Table 6 Table showing a number of Eulerian models used in Europe for urban scale air quality applications. When available the model names are linked to the EEA Model Documentation System (MDS).

Eulerian models (MDS link)	Comments/description	Links to documentation, validation and inter-comparison studies
MODELS3/ CMAQ	Eulerian nested chemical transport model. US community model	http://www.cmaq-model.org/
CAMx	Eulerian nested chemical transport model	ENVIRON (2010) http://www.camx.com
MATCH	Nested chemical transport model	Robertson et al. (1999)
AURORA	Eulerian chemical transport model	Mensink et al. (2001)
MARS	Eulerian chemical transport model	Moussiopoulos et al. (1995)
WRF-CHEM	Nested and coupled meteorological and chemical transport model	http://cprm.acd.ucar.edu/Models/WRF-Chem/
CHIMERE	Nested chemical transport model	http://www.lmd.polytechnique.fr/chimere/ Vautard et al. (2005)
TAPM	Nested and coupled meteorological and chemical transport model	http://www.cmar.csiro.au/research/tapm/
AIRQUIS-EPISODE	Chemical transport model with sub-grid elements	www.airquis.com
FARM	Eulerian nested (one way and two-way) chemical transport model	www.aria-net.it http://www.mi.uni-hamburg.de/index.php?id=539 Gariazzo *et al.*, 2007a

Application guidance

There is a wide range of urban scale models available and most will provide reasonable estimates of the annual mean concentrations within the urban area for both NO_x and NO_2, given appropriate input data. All models should be validated against local measurements and all models should provide an estimate of their uncertainty.

When applying the Gaussian/Lagrangian type of model it is important to note the following:

1. Not all of these models use Lagrangian trajectories for their Gaussian dispersion models (i.e. spatially and temporally varying wind fields) and as such assume a homogenous meteorology for the entire domain. In this case these models should only be applied in flat and homogenous terrains
2. These models generally do not include temporarily varying chemical reaction schemes, though a number of these models post process the chemical species to provide an estimate of the final concentration of reactive species such as NO_2. Eulerian grid models are more suitable for calculating chemical reactions.
3. Gaussian models provide the possibility of unlimited resolution. Indeed these models provide the possibility of mapping concentrations to a resolution of just a few metres. Though this can produce convincing spatial distributions, Gaussian models may be less suitable for calculating hourly means than they are for annual mean concentrations in an urban region. This is due to the assumptions in their

formulation (e.g. steady state) but it will also depend on how they are applied, e.g. using back trajectories with non-homogenous wind fields or using straight line homogenous wind fields. In addition in cases where episodes of poor air quality occur due to recirculation of air masses then Gaussian models, based on homogenous wind fields, will not be able to represent these important episodes properly.

When applying Eulerian type models it is important to note the following

4. The vertical resolution of the model may have significant impact on the ground level concentrations in a Eulerian model. This should be assessed in any application of the model, especially when applying elevated emissions.
5. Many of the vertical dispersion schemes applied in Eulerian models, e.g. K theory, are based on steady state solutions and developed using measurements in flat and homogenous terrain. Such schemes are generally applicable for grid sizes > 5 km, rather than grids of 1 km or less.
6. In many meteorological and air quality models a range of vertical dispersion schemes are available. As with the vertical resolution of the model, the impact of the different vertical dispersion schemes should also be assessed to help partially quantify the model uncertainty.

3.5 Regional scale modelling

The focus of this guidance document is on local and urban modelling. However, urban modelling will always require either boundary conditions for the models or background concentrations. When nested models are applied then both regional and urban scales will often be modelled by the same model. The information in this section is to a large extent based on the regional scale assessment review carried out by Air4EU (Air4EU D5.1, 2007).

Description

Regional air quality models, or Chemical Transport Models (CTM's), describe the functional relation between emissions and concentration/deposition. The change in time of a concentration of species in a certain grid volume is described by the (changes in):

- mean wind speed
- turbulent dispersion
- chemical and physical transformation
- dry and wet deposition
- emissions

The required input to these models are the meteorological conditions, the land use and land cover to determine the dry deposition and also the anthropogenic, biogenic and natural emissions. Regional scale models furthermore need boundary conditions at the borders of the model domain.

Most of these models are 3D Eulerian grid models, containing in principle the most relevant processes needed to calculate the concentrations of regulated pollutants, but some are Lagrangian type models (See table 7). Grid resolutions of these models are generally 50 km or less. Currently around 10 km is the highest feasible grid resolution for calculating concentrations across all of Europe for time scales of a year. The suitability of the model for AQ Directive applications is often related to its computational run time. Some models may require restrictively long run times and cannot be effectively used to carrying out a large number of scenario assessments.

Regional scale models are used in a range of applications, from European scale health and eco-system impact assessments to air quality forecasting applications. Regional scale models are also applied on the national scale for assessment and planning purposes in relation to the AQ Directive.

Recently the GEMS and MACC (http://www.gmes-atmosphere.eu/services/raq/) projects, the PROMOTE project (http://www.gse-promote.org/) and the CityDelta and Eurodelta initiatives, have resulted in important

developments in regional scale air quality modelling. In particular most of the operational models currently in use in Europe are included in the MACC project, which will produce a number of products including ensemble reanalysis and forecasting of European air quality.

Examples

In Table 7 a number of regional scale chemical transport models (CTMs) are listed. Some of these are nestable and can also be used on the urban scale. All these models are Eulerian and include extensive chemical schemes.

Table 7. Table showing a number of Eulerian chemical transport models used in Europe for regional scale air quality applications. When available the model names are linked to the EEA Model Documentation System (MDS).

Model (MDS link)	Comments/description	Links to documentation, validation and inter-comparison studies
EMEP	Eulerian chemical transport model for regional and continental scales	http://www.emep.int
CAMx	Eulerian tropospheric photochemical dispersion model for spatial scales ranging from urban to regional.	ENVIRON (2010) http://www.camx.com
DEHM	Eulerian 3-D model covering the Northern Hemisphere. Two-way nesting capability with 4 domains (150 km resolution down to 5.6 km). Includes photochemistry, particles, Hg, POPs and pollen.	http://dehm.dmu.dk
MATCH	Nested chemical transport model developed by SMHI	Robertson et al. (1999)
LOTOS-EUROS	Eulerian chemical transport model for regional and continental scales developed by TNO	Schaap et al. (2008) http://www.lotos-euros.nl/
MODELS3/ CMAQ	Eulerian nested chemical transport model. US community model	http://www.cmaq-model.org/
REM-CALGRID	Regional Eulerian Model - California Grid Model. Further developed and applied in Germany by FU-Berlin	Stern (2003) http://www.geo.fu-berlin.de/met/ag/trumf/RCG/
CHIMERE	Nested chemical transport model	http://www.lmd.polytechnique.fr/chimere/ Vautard et al. (2005)
EURAD	Nestable European scale model with potential for inverse emission modelling.	Elbern et al. (2007) http://www.eurad.uni-koeln.de/index_e.html
MOCAGE	Multi-scale Chemistry and Transport developed at Météo-France for both research and operational applications.	Peuch et al. (1999) http://www.cnrm.meteo.fr/gmgec/spip.php?article87&lang=fr
SILAM	Both Lagrangian and Eulerian methods are available on the European scale	http://silam.fmi.fi

Beleuros	3-D Eulerian model covering large parts of Europe; base grid cells of 60 x 60 km, regional grid refinement to 15 x 15 km or 7.5 x 7.5 km.	http://pandora.meng.auth.gr/mds/showlong.php?id=166
OPS	Multiple source Gaussian-Lagrangian model for long term means developed by RIVM in The Netherlands. Applied to all of The Netherlands.	van Jaarsveld et al. (2004)

Application guidance

1. As with Urban scale Eulerian models there are a number of regional scale models available that will provide reasonable estimates for NO_2 concentrations. All models should be validated against local measurements and all models should provide an estimate of their uncertainty.
2. A number of operational models are implemented in the MACC (GMES Atmosphere pre-operational service) and these represent the state of the art in regional scale CTMs.
3. Regional scale models using data assimilation methods are recommended over other models for air quality assessment purposes or when used to provide background concentrations for urban scale models.
4. In regard to local and urban scale modelling regional scale models can be used to provide background concentration levels or boundary conditions for urban scale models. Indeed, if the regional scale contribution is significant then regional scale models should be applied when carrying out planning assessments.

3.6 Linking models of different scales

Description

There are a number of methods in use that can directly link the different scales of assessment. These may be separated into four major types. These are:

Nesting of models

The nesting of higher resolution models of limited spatial coverage, in lower resolution models of larger areal coverage. This is typically the case where Eulerian models are applied, both meteorological and chemical transport models. One way nesting means that information from the larger scale model is passed to the smaller scale model in one direction only, as boundary conditions for that model. Two-way nesting means that the high and low resolution models pass information in both directions. Nesting can have many levels, all the way from global models to local street level models.

Sub-grid modelling

This addresses the inclusion of 'sub-grid' models within gridded Eulerian models. This typically involves the placement of Gaussian type line or point source models, e.g. 'Plume in grid' or PIG models, inside a gridded Eulerian model. This is a similar concept to nesting but in practical terms the methodology is different as it goes from one type of model, Eulerian, to another, Gaussian. This will provide improved resolution in areas where large gradients occur and can provide, in the case of PIG models, an improved description of the dispersion and chemistry of a plume before placing the pollutant into the Eulerian grid.

Downscaling methods

Downscaling methods that redistribute concentrations according to specific parameterisations may also be used within Eulerian grids. The use of nested models is often called 'dynamic' downscaling but other types of downscaling will involve the use of parameters that are available at higher resolution than the model itself, e.g. population and land use, and these will be used to redistribute the concentrations within grid squares. Downscaling may provide higher resolution concentration fields or it may provide statistical information of the grid that can be used for further assessment.

Boundary conditions

Boundary conditions, or background concentrations, from observations or climatological data may also be used to link larger scales to smaller scales. It is common in urban scale air quality modelling to use observational data, from the up-wind direction, to provide the input data for these models. It is also common to use climatological data to drive regional scale models.

One general aspect of nesting and 'sub-grid' models is the need to avoid double counting of the emissions in the models, i.e. that the concentrations from a particular source within a larger scale model are added to the concentrations that result from that same source within the smaller scale model. When consistent meteorological and chemical transport modelling is carried out using nesting then this will not be a problem. However, if a small scale model is applied using boundary conditions from a different, and perhaps meteorologically inconsistent, larger scale model then concentrations resulting from emissions within the smaller region can be returned to the smaller region. In addition, there are a number of models that calculate urban concentrations and use these as urban background for local line source or street canyon models. In principle the emissions from that source should not be included in the urban background concentrations used. This is in practice difficult to obtain if calculations over many thousands of line sources are required.

Examples

There are a number of examples of nested modelling systems and models using downscaling or sub-grid models.

Table 8 Table showing a number of chemical transport models or modelling systems used in Europe that link different scales. When available the model names are linked to the EEA Model Documentation System (MDS).

Model/System (MDS link)	Comments/description	Links to documentation, validation and inter-comparison studies
THOR	Nested modelling system going from global model (DEHM) to urban (UBM) to street level (OSPM) concentrations	http://thor.dmu.dk
MATCH	Chemical transport model capable of one way nesting from regional to urban scale	Robertson et al. (1999)
CAMx	Chemical transport model capable of both one and two way nesting between regional and urban scales	ENVIRON (2010) http://www.camx.com
MODELS3/ CMAQ	Chemical transport model capable of one way nesting from regional to urban scale	http://www.cmaq-model.org/
AURORA	Chemical transport model capable of one way nesting from regional to urban scale	Mensink et al. (2001)
AURORA- IFDM	Coupling of the regional scale model AURORA (3 km) with the local Gaussian model IFDM (500 m). Deals with double-counting of emissions. Currently only applied for tracers.	Lefebvre et al. (2011)
EMEP	Chemical transport model capable of one way nesting from regional to urban scale	http://www.emep.int/
CHIMERE	Chemical transport model capable of one way nesting from regional to urban scale	http://www.lmd.polytechnique.fr/chimere/ Vautard et al. (2005)
WRF-CHEM	Chemical transport and meteorological model capable of two way nesting between regional	http://cprm.acd.ucar.edu/Models/WRF-Chem/

	and urban scales	
SIMAIR-road	Combines regional (MATCH) with local scale and (OSPM)	http://www.smhi.se/en/services/professional-services/Environment/simair-road-1.7647 Omstedt et al. (2009)
AIRQUIS-EPISODE	Contains sub-grid Gaussian models for line and point sources in an urban scale Eulerian grid	www.airquis.com
FARM	Eulerian nested (one way and two-way) chemical transport model	www.aria-net.it Gariazzo *et al.* (2007a)
BelEUROS/RIO-Corine	Statistical downscaling of gridded concentrations using land use regression	Janssen et al. (2008a; 2008b)

Application guidance

1. When carrying out urban scale assessment it is recommended to use background concentrations obtained from observations, or from regional scale models using data assimilation, rather than from models that do not use data assimilation. This is to avoid the propagation of model errors from the regional to the urban scale.
2. When carrying out modelling for planning purposes, regional scale model calculations that are relevant for the future concentrations should be applied rather than the use of current observations or climatology as boundary conditions.
3. Downscaling methods can be attractive methods for redistributing the concentration data. However, with any disaggregation method the uncertainty in the concentrations will increase. This should be taken into account when carrying out any downscaling method.
4. It is recommended to use consistent models, particularly meteorological models, when nesting if possible. This will provide continuous wind vectors across nested grids and provide similar boundary layer heights and dispersion characteristics.
5. When an urban scale model is nested within a different regional scale model, i.e. boundary conditions for the urban model are taken from a different source, then the urban scale model domain should be sufficiently large to avoid double counting. Though there are no set rules on this, domains of 300 km for large cities/agglomerations should be sufficient to avoid this (Air4EU D6.2, 2007).

4 Chemistry modelling

Some atmospheric compounds have such slow chemical reaction rates that they can essentially be treated as inert tracers when applying air quality modelling on the urban scale. However, this is not the case for nitrogen dioxide (NO_2) since it is a species that is rapidly formed from nitrogen oxide (NO) through its reaction with ozone (O_3) (Equation 4.1, Box 2). Indeed, if this were the only important reaction, as it is at night time under most urban conditions, then NO will be transformed to NO_2 until all the NO has been converted to NO_2 or until all the ozone has been used up (ozone limited). Given typical concentrations in the urban atmosphere this reaction takes place on a time scale of just a few minutes but this is dependent on concentrations and also on temperature. The rate of this reaction is important for modelling since the time it takes for exhaust emissions of NO (and NO_2) to reach a traffic monitoring station is only a few seconds whilst the time it takes for emissions to reach an urban background station is a few 10's of minutes – 1 hour. As a result the proportional concentration of NO_2, compared to NO, near to traffic sources is generally lower than at urban background stations.

In addition to the conversion of NO to NO_2 the other major daytime reaction that affects NO_2 is its destruction by photolysis, i.e. sunlight breaks the NO_2 molecule into an NO molecule and a ground state oxygen molecule (Equation 4.2, Box 2). This reaction rate may also be quite fast (10 – 30 minutes) in sunny environments. As a result a balance can be reached within an hour such that the production of NO_2 is balanced by its destruction. This is called the photo-stationary state or the photochemical steady state (Box 2).

On longer time scales, and dependent on the amount of hydrocarbons emitted in the atmosphere, there may also be a chemical production of NO_2 that results from the reaction of peroxyl radicals (the result of the photolysis of hydrocarbons) and NO (Equation 4.4, Box 2). This reaction takes place with a time scale of hours and can have an important impact on both NO_2 and eventually ozone concentrations in the urban environment.

The eventual fate of NO_2 is determined by a number of reactions but the most important of these is the conversion of NO_2 to nitric acid (HNO_3) (Equation 4.5, Box 2) which occurs at the rate of several hours to a day. Nitric acid is readily removed from the atmosphere through deposition and is involved in aerosol processes leading to the formation of ammonium nitrate. As a result of this time scale, NO_2 has an atmospheric lifetime of just a day or two, and as such is not transported over large distances.

Box 2: Important chemical reactions for NO_2 in the urban environment

The creation of NO_2 by the oxidation of NO with O_3 (Equation 4.1) is a very fast reaction that takes only a few minutes under typical urban atmospheric conditions. The production of NO_2 is balanced by its decay in the presence of sunlight (Equation 4.2), where *hv* is the energy from a photon with a wavelength $\lambda < 420$ nm, and the subsequent creation of ozone (Equation 4.3) with the generated ground state oxygen molecule (O). This last reaction requires some other molecule (M) for the reaction to occur.

$$O_3 + NO \rightarrow NO_2 + O_2 \quad (4.1)$$

$$NO_2 + h\nu \rightarrow NO + O \quad (4.2)$$

$$O + O_2 + M \rightarrow O_3 + M \quad (4.3)$$

In urban environments the emission of hydrocarbons, mostly in the form of volatile organic compounds, and the subsequent production of peroxy radicals RO_2 (including hydrogen peroxide HO_2), generated by the oxidation of these hydrocarbons, will also affect the oxidation of NO to NO_2. Here R represents any configuration of a range of hydrocarbons after the removal of a hydrogen atom H. The NO will react with the peroxy radicals in the following way.

$$RO_2 + NO \rightarrow NO_2 + RO \quad (4.4)$$

The rates of these reactions will vary dependent on the concentrations of the peroxy radicals but in urban areas the reaction rates are typically of an hour or more. As a result they can make a significant contribution to the NO_2 concentrations, and eventually O_3 concentrations through equations 4.2 and 4.3.

Though there are a number of alternative reactions, the eventual loss of NO_2 will go through reactions such as

$$NO_2 + OH + M \rightarrow HNO_3 + M \quad (4.5)$$

This last reaction has a rate of between a few hours and a number of days, dependent on the availability of OH. This is one of the reactions that is responsible for the short lifetime of NO_2 in the atmosphere (~ one day).

As a consequence of equations 4.1 – 4.5 the rate at which NO_2 will change its concentration is given by its production (Equations 4.1 and 4.4) minus its loss (Equations 4.2 and 4.5):

$$\frac{d[NO_2]}{dt} = k_1[NO][O_3] + k_4[NO][RO_2] - J[NO_2] - k_5[NO_2][OH] \quad (4.6)$$

where k_i is the reaction rate for Equations *i* and *J* is the photolysis rate coefficient for Equation 4.2. *J* depends on the amount of sunlight and this often represented by the height of the sun in the sky, or its inclination.

The chemical scheme applied will depend on the needs of the application and on the model type being used. Table 9 lists the major different types of chemical schemes indicating their suitability (fitness for purpose) for the different applications. In this chapter we look at the major chemical schemes and their related applications as follows:

1. Steady state chemical models
2. Distance dependent parameterised chemical models
3. Statistical/empirical parameterised chemical models

4. Photochemistry models

Table 9. Fitness for purpose matrix for NO_2 chemical schemes. Shown are the major chemical scheme types (columns) and spatial scales (rows) for both assessment and planning applications. Fitness for purpose is indicated by colour and appropriate comments. Green = 'fit for purpose'; Orange = 'conditionally applicable'; Purple = 'not fit for purpose'.

Chemical schemes and applications	Empirical schemes	Photo-stationary and ozone limitting schemes	Distance from source and mixing schemes	Reduced photochemical schemes	Full photochemical schemes
Assessment					
Street level	Given sufficient observations	Overestimates NO_2 in the presence of ozone		Difficult to apply at this scale. CFD only	Only reduced schemes necessary
Urban scale	Given sufficient observations	Suitable for winter or low hydrocarbons			
Regional scale				Missing significant chemistry	
Planning					
Street level	Only if scheme includes ozone and NO_2 primary emissions	Sensitivity to ozone and NO_2 emissions represented		Difficult to apply at this scale. CFD only	Only reduced schemes necessary
Urban scale	Only if scheme includes ozone and NO_2 primary emissions	Suitable with low light or hydrocarbons	Suitable with low light or hydrocarbons		
Regional scale				Missing significant chemistry	

4.1 Steady state chemical models

Description

Some local and urban scale models make use of the steady state solution to equations 4.1 – 4.3. This is shown in Box 3. The steady state solution is quite convenient because it allows the modelling of O_x ($O_x = NO_2 + O_3$) and NO_x ($NO_x = NO_2 + NO$) concentrations as non-reactive tracers, since these are conserved in Equations 4.1 – 4.3. After transport and diffusion of the O_x and NO_x concentrations, NO_2 concentrations can be calculated using the steady state solution, Equation 4.9. This makes the method attractive since Gaussian type models, which only work on non-reactive species, can transport and diffuse O_x and NO_x and then calculate NO_2 from their resulting values.

In order to include the effect of turbulent mixing, which occurs on similar time scales to the chemical reactions, Berkowicz et al. (1997) introduced a mixing (exchange) rate into Equation 4.6 for the street canyon model OSPM. This parameterisation reduced the amount of available ozone that was exchanged with the ambient air outside of the street canyon, thus reducing the conversion of NO to NO_2. In this parameterisation a steady state was still assumed to occur. The method has been further applied to a range of other models employed by NERI Berkowicz et al. (2011). When applied to open roads the mixing rate of the ambient air becomes distance dependent (see Section 4.2).

A special case of the steady state solution is when the daytime photolysis is ignored (J=0), e.g. at night or in weak sunlight conditions, and the steady state becomes simply the complete conversion of the available NO to

NO_2 by the consumption of ozone. i.e. all NO will be converted to NO_2 until all the ozone is consumed. This is known as the 'ozone limited' approximation.

Box 3: Photochemical steady state solution

The basic photochemical steady state solution for NO_2 is obtained by ignoring the contribution of the peroxyl radicals (RO_2) and the hydroxyl radical (OH) to the production and loss of NO_2 and by setting the rate of change of NO_2 in Equation 4.6 to be zero, i.e. no change in NO_2. Under this condition we find a relationship between the compounds of NO_2, NO and O_3 that is given by the following:

$$[NO_2] = \frac{k_1}{J}[NO][O_3] \qquad (4.7)$$

Equation 4.7 tells us the relative concentrations but does not provide us with a solution for NO_2. To find a solution we make use of the fact that NO_x ($[NO_x] = [NO] + [NO_2]$) and O_x ($[O_x] = [NO_2] + [O_3]$) concentrations are conserved in Equations 4.1 –4.3 and these can be treated as non-reactive tracers. As a result we can write a steady state solution for NO_2 as:

$$f_{NO2}{}^2 - f_{NO2}(1 + f_{Ox} + J') + f_{Ox} = 0 \qquad (4.8)$$

where

$$f_{NO2} = \frac{[NO_2]}{[NO_x]},\ f_{Ox} = \frac{[O_x]}{[NO_x]} \text{ and } J' = \frac{J}{k_1[NO_x]}$$

This has a solution of the form

$$f_{NO2} = \frac{(1 + f_{Ox} + J') - \sqrt{(1 + f_{Ox} + J')^2 - 4f_{Ox}}}{2} \qquad (4.9)$$

Solutions to this are shown in Figure 3.

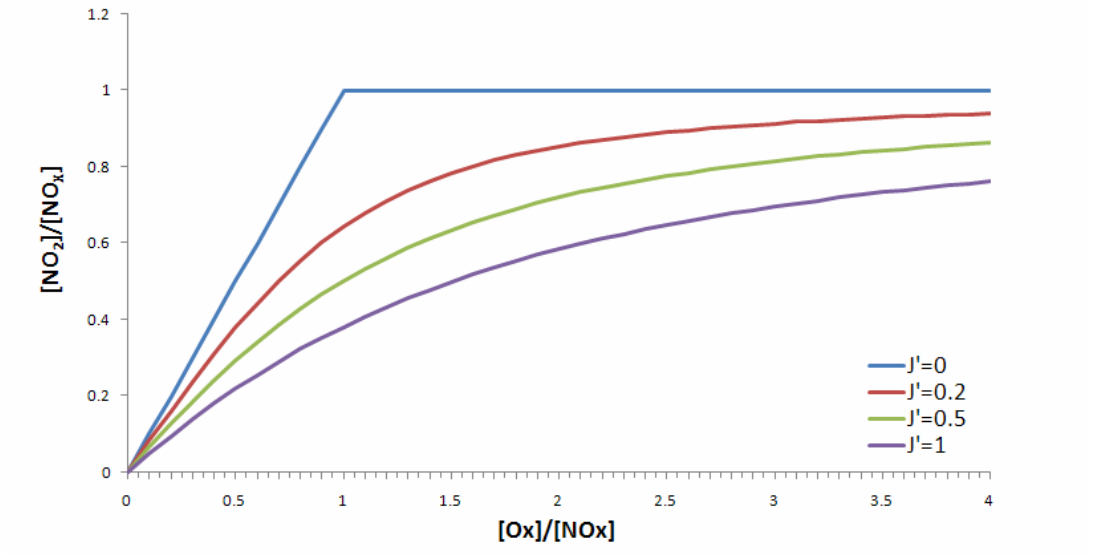

Figure 3. NO_2 concentrations, as a fraction of NO_x, plotted against O_x concentrations, as a fraction of NO_x for four different J' values. J' = 0 corresponds to night time (ozone limited), J' = 0.2 - 0.5 to a normal urban daytime

situation and J' = 1 to a low NO_x daytime situation. Based on Equation 4.9, Box 2. No initial concentration of NO_2 is assumed in this calculation.

Examples

In Table 9 a number of examples where steady state models are used are provided. This list is non-exhaustive. Note that the steady state approximation used in OSPM has an additional term that represents the limited mixing of the background concentrations with the emission plume.

Table 9 Table showing models used in Europe that apply the photo-stationary assumption. When available the model names are linked to the EEA Model Documentation System (MDS).

Models using the photostationary assumption (MDS link)	Comments/description	Links to documentation, validation and inter-comparison studies
AIRQUIS-EPISODE	Applied in conjunction with Gaussian line, point and Eulerian models.	www.airquis.com
IMMIS^{cpb}	A simple physico-chemical model based on the photochemical equilibrium is used to calculate total NO_2 concentrations	http://www.ivu-umwelt.de/e
IFDM-POLCA	The hourly NO_2-concentrations resulting from NO_x emissions near line sources and in street canyons are calculated using the photochemical equilibrium between NO, NO_2 and O_3.	Bultynck and Malet (1972)
OSPM	Steady state solution applied to street canyons with an additional relaxation term representing the limited mixing in the street canyon with the ambient urban air.	Berkowicz et al. (1997, 2011)
OSPM approach applied to annual averages	Adaptation of OSPM method for application to annual means. Photochemical equilibrium with limited mixing within a street canyon or open road (no distance dependence).	Düring et al. (2011)

Application guidance

The steady state approximation is physically correct, even if not all of the assumptions in its derivation are always applicable. As such there is a limit to its application.

1. The photo-stationary approximation for modelling NO_2 and ozone is a physically based description that is suitable for use during day time conditions in urban areas where transport times are longer than the reaction time scales. In many such situations the approximation will provide good estimates of hourly mean NO_2 concentrations given the correct NO_x and ozone concentrations. The method requires that ozone at background level be known.
2. The time scale at which the reactions take place make the steady state approximation more suitable for urban scale applications than for local traffic applications since the reaction is far from finished in the few seconds required to transport the exhaust emissions to the kerbside.
3. In the case of street canyons, where the residence time is longer than for open roads, the photostationairy assumption is likely to be more valid but it is recommended to use the approach used in OSPM to account for the limited mixing with the ambient air.

4. Alternative methods should be used, see next section, for determining NO_2 concentrations near to source, e.g. close to traffic, and particularly for open roads.
5. The steady state solution is most useful in urban environments with high NO_x concentrations, relative to those of hydrocarbons.
6. In Nordic winter conditions the steady state is often found to be a good approximation. This is chiefly due to the lack of available ozone that is very quickly consumed in the poor dispersion conditions, which also lead to high NO_x concentrations. As a result there is little conversion of NO to NO_2 possible and the modelled NO_2 concentrations will primarily depend on the primary emissions of NO_2.
7. In mid-latitudes the increased availability of ozone makes the approximation less applicable for traffic stations, however the method should work well for concentrations further from the source if the emissions of Hydrocarbons do not strongly affect the balance.
8. Ozone limiting method (OLM) generally over predicts NO_2, makes assumptions concerning the availability of O_3 and assumes stationarity. It is not recommended for general use but will provide a simple estimate for the maximum NO_2 available.

4.2 Distance dependent or limited mixing parameterised chemical models

Description

In addition to the photo-stationary state approximation there are also some other physically based models for determining NO_2 concentrations, particularly when using Gaussian type dispersion models. A method employed for line source models is the Discrete Parcel Method (DPM) originally developed by Benson (1984, 1992) and also adapted by Karpinnen et al. (2000). This method takes into account non-stationarity, solving the basic photochemical reactions in a box that is based on the initial concentrations of the well mixed plume. The advantage of the method is that it takes into account the travel time of the plume but the disadvantage is that it does not include the dispersion of the plume as a time varying factor in the concentration.

Another method described by Janssen et al. (1988) and applied to industrial plumes, uses the analytical solution to Equations 4.1 – 4.3 based on the assumption of constant ozone and then introduces an empirical fit to the resulting equations to account for turbulent mixing and ozone depletion in the plume. This method has not been applied to traffic sources. A similar method for industrial plumes, that parameterises the turbulent mixing of ozone in the plume but applies the ozone limiting assumption, known as the Plume Volume Molar Ratio Method (PVMRM) has been developed by Hanrahan (1999). This method is applied in the US EPA models CALPUFF (www.epa.gov/ttn/scram/dispersion_prefrec.htm#calpuff) and AERMOD (www.epa.gov/ttn/scram/dispersion_prefrec.htm#aermod) as one of the chemistry options for NO_2.

The method developed by Berkowicz et al. (1997, 2011) for OSPM, that applies the steady state assumption but includes limited mixing with the ambient air in a street canyon, has also been applied to open roads and other plume sources (OML-Highway and UBM models). In that case the the mixing rate of the ambient air becomes distance dependent and this limits the chemical reaction within the plume. Note this method, though distance dependent, still assumes that photochemical equilibrium is instantaneously reached within the plume.

On the urban scale ADMS-Urban, a Gaussian type model, applies the GRS chemical scheme to the concentrations of the primary emitted pollutants after transport and dispersion. It carries out a chemical transformation of these final concentrations over a period of time defined by the average ageing of the pollutants within the urban domain. As such it does not take into account changes in concentrations of the emitted pollutants during the transport and dispersion process.

Application guidance

These methods assume a time, or distance, dependence on the production of NO_2. As such they are very suitable for assessing NO_2 concentrations near source, e.g. near roads and industry, and are preferable to steady state descriptions.

1. For local scale modelling of open roads the discrete parcel method (DPM), e.g. Benson (1984, 1992) and Karpinnen et al. (2000), which takes into account non-stationarity and the basic photochemical

reactions, is recommended as an appropriate analytical technique, though it does not include the effect of limited turbulent mixing

2. In addition to the DPM the parameterisation implemented in both OSPM and OML-Highway has been shown to produce reliable results for the studies it has been applied to. This includes the limited turbulent mixing but assumes steady state and hence no time dependent chemical reaction dependence.
3. In general distance or time dependent schemes are preferred over basic photo-stationary schemes when concentrations are to be determined close to the source.

Examples

Table 10. Table showing models used in Europe that apply distance dependent or limited mixing chemical schemes for NO_2. When available the model names are linked to the EEA Model Documentation System (MDS).

Model (MDS link)	Comments/description	Links to documentation, validation and inter-comparison studies
CAR-FMI	R-DPM. Based on travel time of a discrete parcel to a receptor from a source. Determines the concentration based on travel time.	Härkönen (2002), Karppinen et al. (2000)
CALINE 4	Original version of the discrete parcel method (DPM)	Benson (1984, 1992)
ADMS	Option to apply the Generic Reaction Scheme (GRS). This is a time stepping scheme with a time scale based on an age profile. The chemical scheme is applied after advection of the primary pollutants to each receptor point.	www.cerc.co.uk/environmental-software/ADMS-Urban-model.html
OML-HIGHWAY	Steady state solution with an additional relaxation term related to the travel time from the source, representing limited mixing with the ambient urban air (Similar to OSPM method, Table 9).	Berkowicz et al. (2011)

4.3 Statistical/empirical parameterised chemical models

Description

There are a number of statistical or empirical models used to convert NO_x concentrations to NO_2 concentrations. To some degree these may be based on the photo-stationary state description given in Equation 4.9 but where some factors are changed to provide a better fit to the observed concentrations. Some parameterisations, however, are highly empirical and represent best fits to arbitrarily selected functions. Most of these empirical models are developed for annual mean concentrations, though some may be applied to hourly values, and most depend solely on the modelled NO_x concentration to determine NO_2. As such they are very site specific and to some extent year specific as well since they do not usually take into account changes in ozone or in other pollutants. In general the methods also do not differentiate between near or far from source concentrations, though some of the parameterisations may be conceptually based on a background NO_2 level and a traffic source increment when used in proximity to local line sources.

Despite, or as a result of, their empirical nature such models can give quite satisfactory results for annual mean concentrations as there is a clear dependence of NO_2 on NO_X concentrations. In Figure 4 annual mean concentrations of NO_2 and NO_X have been extracted from the AirBase database for all of Europe in the period 2006 – 2008 and plotted against one another. The data has been separated into Traffic and Background stations.

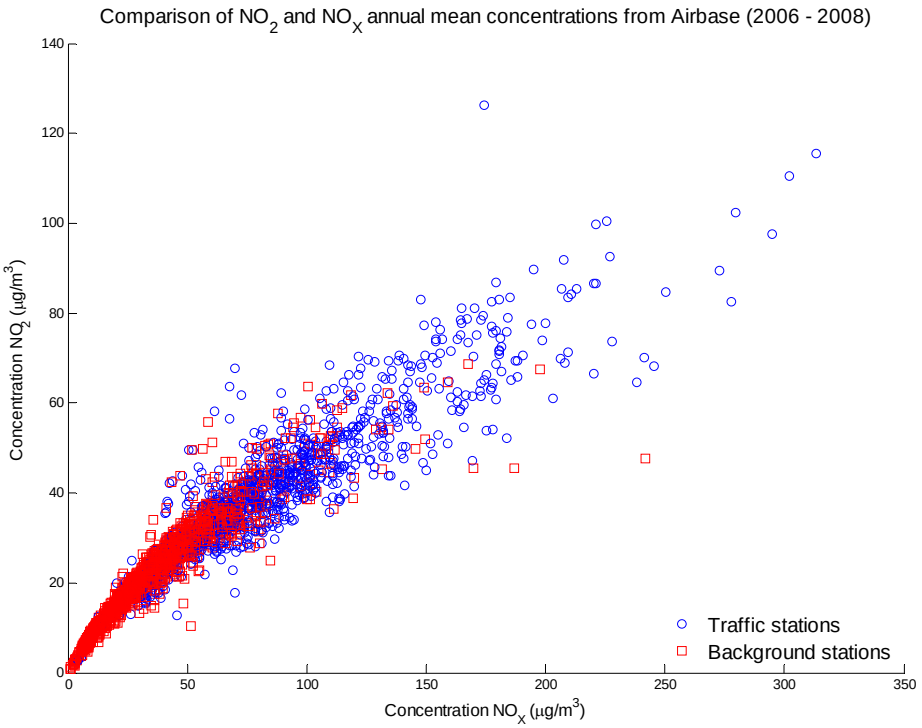

Figure 4. Relationship between the annual mean concentration of NO_2 and NO_x ($\mu g/m^3$). Data taken from AirBase (2006-2008). In total 991 traffic station data points and 1539 background station data points are shown.

Examples

There are a large range of examples available for empirical chemical schemes. Some use arbitrary functional fits and others physically based functions. Table 11 provides a summary table of these methods. Since these methods are often used in Directive related application Appendix 1 provides some more details concerning the methods listed in Table11

Table 11. Table showing models or empirical schemes used in Europe that provide and empirical relationship between NO_X and NO_2. When available the model names are linked to the EEA Model Documentation System (MDS).

Model or method (MDS link)	Comments/description	Reference
Ambient Ratio Method (ARM)	The simplest empirical relationship applied to convert modelled NO_x concentrations to NO_2 concentrations (US EPA). This method simply involves calculating the locally observed NO_2: NO_x ratio and applying that to all the model NO_X values. $[NO_2] = a[NO_x]$ where a is a constant.	(Chew and Meyer, 1991)
Methodologies developed for DEFRA	A number of empirical methods have been developed in the UK including polynomial fits to hourly data and methods that separate the background and local road NO_x emissions, see below. These methods are not operationally applied to models. $[NO_2(road)] = (-0.068\log([NO_x(total)]) + 0.53)[NO_x(road)]$	Derwent and Middleton (1996) Dixon et al. (2000) Laxen and Wilson (2002)

Airviro	Airviro modelling system applies the following parameterisation to convert from modelled NO_x concentrations NO_2 concentrations. $[NO_2] = 0.73[NO_x]\exp(-0.00452[NO_x] + 0.003014[NO_x]^2)$	www.smhi.se/airviro/
Romberg method	The Romberg method has been used for several years in Germany within a number of models (e.g. PROKAS, IMMIS and MISKAM). It uses the following straight forward parameterisation: $[NO_2] = \frac{A[NO_x]}{[NO_x] + B} + C[NO_x]$	Romberg et al. (1996) Bächlin and Bösinger (2008) Pongratz et al. (2010)
Standard Calculation Method in the Netherlands	Empirical relation, using the same basis as used for the Romberg method, but includes ozone concentrations, the fraction of NO_2 emitted in NO_X and differentiates between near road and urban background factors. $\overline{\Delta NO_2} = F \cdot \overline{\Delta NO_x} + \beta \cdot \overline{O_3^a} \cdot \frac{(1-F) \cdot \overline{\Delta NO_x}}{(1-F) \cdot \overline{\Delta NO_x} + K}$	Van den Hout en Baars (1988) Wessling and Sauter (2007) http://wetten.overheid.nl/BWBR0022817/
SAPPHO	This algorithm is based on the photo-stationary state (Box 3) and is applied to determine background concentrations of NO_2, based on NO_X, in The Netherlands (developed by RIVM). The algorithm tunes J' and $[O_x]$ to 8 years of annual mean measurements in The Netherlands as follows: $J' = 0.27[NO_x] + 4.5$ and $[O_x] = 1.3\sqrt{[NO_x]} + 27.4$	Erens and van Dam (2000)
Keller	Swiss empirical model for converting NO_x to NO_2. Was also applied by Erens and van Dam (2000) in The Netherlands and found to perform poorly in the Netherlands. $[NO_2] = 0.055[NO_x] + 55(1 - \exp(-(0.7 - 0.055)[NO_x]/55))$	Keller et al. (1997)
Oxidant Partitioning Model	This method is based on empirical fits (using up to fourth order polynomials) of the NO_2:O_X ratio for both 'near' and 'far' sources. The basic formulation is given as: $[NO_2] = (A[NO_x] + B).f(NO_X)$	Jenkin (2004) Murrells et al. (2008)

Application guidance

These types of models are strongly empirically based. They are useful in the areas in which they were developed when assessment is the only application. The methods will not provide the correct dynamic response to changes in emissions, boundary conditions or changes in meteorology unless these influences are implicitly included in their formulation, and they are therefore limited in usefulness for planning purposes when alternative emission or boundary condition scenarios are being assessed.

1. Statistical or empirically based conversion algorithms will generally provide a good approximation to the available measured annual mean concentrations of NO_2 in the domains and for the sites from which they were derived. However, it is never certain to what extent the available monitoring data is representative of the entire domain being modelled. Even so empirical algorithms can provide a good estimate of the annual mean NO_2, based on NO_x, in urban regions where sufficient measurements are available for their establishment.

2. It is not recommended to apply an empirical algorithm in an area for which it was not developed or assessed since the coefficients are generally site specific.

3. It is recommended to use more physically based empirical equations that include both near and far from source factors, ozone in some form and the NO_2:NO_X emission ratio. In this way the dynamic sensitivity of the algorithms to changes in these factors can be included. If these are not included then such algorithms are generally not appropriate for planning purposes since they will not provide the correct dynamic sensitivity to changes in emissions or boundary conditions

4.4 Photochemistry models

Description

Time evolving photochemical schemes are usually only applied in Eulerian models where each grid cell is treated as a box in which the chemistry takes place. There is a range of photochemistry schemes available that include NO_2 as one of the compounds. The simplest scheme would be Equations 4.1 – 4.3, with similar equations to Equation 4.6 for NO and O_3 also describing their temporal evolution.

Further to this the next simplest scheme in regular use is the generic reaction scheme (GRS) from Azzi et al. (1992) and Venkatram et al. (1994). This scheme includes hydrocarbons in a single lumped term known as R_{smog} and allows these lumped hydrocarbons to produce a pool of radicals, through photolysis, leading to enhanced oxidation of NO to NO_2 and a subsequent increase in ozone. This represents Equation 4.4.

Further to this there are a range of more 'complete' photochemical models and schemes, and variations on these, including the EMEP photochemical scheme (Simpson et al., 2003), MELCHIOR (Derognat, 2003), Carbon Bond-IV (CBM-IV) photochemical mechanism (Gery et al., 1989, Whitten et al., 1980), CBM IV with updated isoprene chemistry (Carter, 1996 and Whitten et al, 1996), CB05 photochemical mechanism (Yarwood et al, 2005b), and SAPRC99 mechanism (Carter, 2000). Though many of the gaseous phase reactions are similarly described in these schemes the lumping of hydrocarbons is often carried out differently.

These schemes can be solved numerically, using standard numerical methods, but even these methods will differ from model to model. One problem that is often encountered is that some reactions occur very quickly and some very slowly. The numerical methods become computationally expensive if the fast reactions are included. In general this leads to a number of 'steady state' approximations where reactions that achieve equilibrium within the typical time resolution of the model output are solved using a steady state approximation. This may also include the reactions for NO_2 given in equations 4.1 – 4.3. An example of an often used software for solving such schemes is the Kinetic Preprocessor (KPP), Damian et al. (2002).

Photochemistry schemes are included in all regional scale and most urban scale gridded chemical transport models, used for both assessment and planning purposes. The major drawback of these methods is that the near source chemical reactions may not be well described, i.e. emissions are instantaneously diluted into the volume of the model grids. When chemical reactions are non-linear this dilution may not be a good representation of the mean concentrations within grid cells that have high emissions.

Generally chemical schemes are not applied to Gaussian type models. The reason for this is the non-linear nature of the reactions, for which Gaussian forms are unsuitable, and the complexity of attributing chemical reactions and species to multiple Gaussian plumes. There are however a limited number of industrial plume models (CALPUFF, SCIPUFF/SCICHEM) that do incorporate photochemical schemes in some form. Of these the SCICHEM approach (Karamchandani et al., 2000) is the most physically based method allowing for complex chemistry by incorporating second order effects in both the plume dispersion and the chemistry.

Examples

There are a range of photochemical schemes available and implemented in both regional and urban scale models. Some models may apply a number of different chemical schemes. Some of these schemes are listed in Table 12.

Table 12. Table showing a number of the photochemical schemes applied in regional and urban scale models in Europe. When available the model names are linked to the EEA Model Documentation System (MDS).

Chemical scheme	Comments/description and example models (MDS link)	Links to documentation, validation and inter-comparison studies
GRS	Generic Reaction Scheme (10 reactions). This scheme includes hydrocarbons in a single lumped term. Applied in TAPM and ADMS.	Azzi et al. (1992) Venkatram et al. (1994)
EMEP	EMEP photochemical scheme (140 reactions). Applied in the EMEP Unified model and MATCH.	Simpson et al. (2003) http://www.emep.int
SAPRC99 and SARPC07 mechanism	The SAPRC99 and the updated SARPC07 gas-phase mechanism developed by University of California (CE-CERT) and applied in CMAQ, FARM and CAMx. Extensive descriptions of VOC (more than 700 species).	Carter (2000) http://www.cert.ucr.edu/~carter/SAPRC/
CBM-IV	Widely used chemical scheme known as the Carbon Bond Mechanism (28 species, 82 reactions). The mechanism is applied in various forms in a number of models, e.g. CAMx, LOTOS-EUROS, CHIMERE, REM-CALGRID,	Gery et al. (1989)
CB05	The 2005 version of the Carbon Bond mechanism developed for use in EPA atmospheric modeling studies. This mechanism optionally includes aerosol and mercury chemistry. 156 reactions and up to 89 species (54 state gases, up to 22 state particulates, and 13 radicals).	(Yarwood et al., 2005a; 2005b).
RACM	Regional Atmospheric Chemistry Mechanism (77 species and 237 reactions). Applied in EURAD-IM	Stockwell et al.(1997)
MELCHIOR2	Adapted from the EMEP photochemical scheme (44 species, 120 reactions) and applied in CHIMERE.	Schmidt et al. (2001) Derognat (2003)

Application guidance

The choice of chemical scheme is usually linked directly to the air quality model. We make the following notes in regard to their application.

1. It is recommended in large and polluted urban areas that a chemical scheme that includes the reactions of nitrogen oxides, ozone and hydrocarbon radicals be used for modelling the concentration of NO_2 at the urban scale. This is particularly important when carrying out planning activities, where the effect of emission changes on NO_2 concentrations is to be determined, since changes in VOC emissions may have an impact on these.
2. Many of the atmospheric chemistry schemes developed for regional and global models include reactions on time scales much longer than the resident time scales of the pollutants in urban areas and as such introduce an additional complexity and computational time that is unnecessary. However, when nesting urban scale models with regional or global models it is always useful to apply the same chemical schemes to ensure continuity with the regional scale models. As such it is recommended to use the same chemical schemes in both the urban and the regional scale models.

5 Emission data and inventories

Emissions are an essential part of any air quality model. However, the importance of emission data is not restricted to their use as input for air quality simulations. Emissions, and the physical characteristics of the emitting sources, are the key factor in the analysis of the alternatives to improve air quality in a given region in future years, as a result of the implementation of pollutant emissions abatement strategies.

A complete and accurate emission data set, should account for all the important emission sources within the area of interest. This is challenging from the perspective of the emission inventories needed to simulate urban NO_2 levels since ambient NO_2 concentrations depend on a series of atmospheric reactions involving several spatial and temporal scales, so a multi-scale approach is often required. Consequently, several emission inventories must be combined so the particular requirements of the different models are met.

From a modelling perspective, which is the view taken in this document, emissions are typically classified into three types of sources according to their spatial characteristics:

- Stationary or point sources: emissions originating from large point sources at fixed locations (such as power plants and industrial facilities)
- Mobile or line sources: emissions from all types of transport should ideally be included in this category, in particular vehicle emissions from road transport, railways, inland navigation, shipping, aviation, etc.
- Area sources: emissions from other stationary or mobile sources that are transient and widespread are represented and provided on an area basis often in administrative areas, such as counties and regions.

Anthropogenic emissions of oxides of Nitrogen (NO_X) are mostly related to combustion processes. They are produced from the chemical reaction of nitrogen and oxygen gases, within air or fuels, at high temperatures through a number of complex phenomena. NO_X emissions, and the individual NO and NO_2 emissions determining NO_2/NO_X ratios, depend on combustion conditions (temperature, residence time, fuel type, fuel/oxygen mixture, etc.) and are thus specifically determined by the interaction of chemical and physical processes occurring within a given combustion device. From the same combustion sources other emissions, such as hydrocarbons or volatile organic carbons (VOC) are also produced which may impact on NO_2 concentrations. Such sources include power plants, factories, domestic and industrial heating (gas, oil, wood), cars, trucks, buses and all other combustion sources.

A variety of methods are used to estimate emissions depending on their specific features, on the purpose of the inventory and on the sources of interest. One of the major challenges when modelling air quality is to consistently compile or combine emission data using different methods and inventories. For example, emissions from large point sources often come from direct measurements in the stacks while emissions estimates from area sources are frequently based on top-down methods. Emissions from traffic are very specific and emission computation often relies on specialized models or bottom-up methods.

An emission estimate, when direct measurements are not available, is usually the product of at least two variables, such as an *activity operation rate* and an average *emission factor* for the activity. Emissions may be more complex, requiring other information such as meteorological data, or in the case of traffic emissions road slope, driving speed, etc.. As such emission models are sometimes required that describe the functional dependence between time varying quantities and emissions. Apart from emission estimates, emission inventories should also contain relevant supporting data (metadata), such as the location of the emission sources, emission measurements where available, production or activity rates, methodology of measurements or calculations, emission factors, uncertainties, etc.

5.1 Methods, tools and sources

There are a variety of methods available for compiling emission inventories. The three major methods are:

- Direct measurement of specific emissions (usually only available for large industrial sources)
- Bottom up emissions (based on specific activity data and emission factors)

- Top down emissions (based on aggregated activity data and emission factors)

Of these the bottom up and top down methods are required for emission inventories in urban areas and both of these require information concerning activity rates and emission factors per activity. The emission factors are typically defined as the amount of pollutant emitted per activity unit or per a defined task performed. For example emission factors for a particular vehicle type may be in terms of mass of pollutant emitted per distance driven or, for the case of domestic heating, may be in terms of mass of pollutant emitted per mass of fuel consumed. In some cases emission factors will also be dependent on a range of other parameters, e.g. vehicle speed and age or meteorological factors.

The terms 'bottom up' and 'top down' refer to the direction in which activity data is spatially aggregated with the aim of preparing spatially distributed emissions. In the top down approach activity data, such as the amount of petrol sold in a particular administrative region, is collected. This is then distributed (disaggregated) over smaller areas, usually corresponding to grids suitable for air quality modelling, based on information that is representative of the activity, e.g. traffic volume in a grid square or a statistical relation to population distribution. In the bottom up approach activity or emission data is collected on a fine spatial scale, e.g. measured emissions or known activities of individual sources, and aggregated up to the required spatial scale. Inherent in the bottom up and top down concepts is that there is a form of aggregation towards a specified spatial resolution, which should correspond to the air quality model requirements. However, bottom up approaches may not be aggregated at all, e.g. when line source models are applied the basic data source is the traffic volume and fleet composition for individual road segments, but the basic methodology remains the same.

For urban scale modelling both methodologies will be required, particularly if the urban scale model is nested within regional scale models and if sub-grid local scale modelling is also employed. One of the challenges in constructing an emissions inventory for urban areas is to harmonise the two approaches as they do not necessarily provide the same results.

There is also a need to provide not just the spatial distribution of the emissions but also the temporal distribution. This is often approached through the development of representative temporal profiles down to a temporal resolution of an hour. Often temporal profiles are aggregated data, e.g. average hourly daily traffic volumes for a particular road category are provided as daily and weekly monthly/seasonal temporal profiles. In urban applications where the hour by hour development of pollutants is important, an accurate temporal profile is also required for the various source categories.

In Europe one of the most widely used sources of information on emission inventory methodology and data is the EMEP/EEA Emission Inventory Guidebook (EMEP/EEA, 2009). This guidebook is intended as both a general reference for good practice in emission inventories but also for meeting national emission reporting obligations under the LRTAP Convention and its protocols (ECE/EB.AIR/2008/4; ECE, 2008). It must also be used by the Member States of the European Union to fulfil emission reporting requirements under the National Emission Ceiling Directive (EC, 2001). It is important to note that the reporting requirements for these emissions are at national level, so the methodologies described are intended to produce national emission data. So while the general methodologies described in the guidebook are relevant for any emissions inventory, the aim of the guidebook is not commensurate with the aim of the urban scale air quality modeller who requires emission data for point, line and areas sources at a high spatial and temporal resolution.

Box 4: Basic methodology for calculating emissions (EMEP/EEA)

A basic general methodology for the estimation of emissions is described in the Atmospheric Emission inventory Guidebook (EMEP/EEA, 2009). The Quantity (Q) of each air pollutant emitted depends on the Activity Level (AL) and the Emission Factor (EF), a factor which defines the linear relationship between Q and AL according to the general formula:

$$Q_{s,i,j,k} = EF_{s,i,j,k} \times AL_{s,i,j}$$

Where s refers to the pollution source examined, i refers to the technology (industrial boiler, vehicle type, airplane type, solvent-related products, etc), j refers to the activity (combustion of diesel, vehicle km driven, airplanes landing, commodities production, etc.) and k refers to the pollutant examined. $Q_{s,i,j,k}$ is the quantity of pollutant k, from source s, technology i and activity j (in t yr^{-1}). $EF_{s,i,j,k}$ is the emission factor for the same pollutant, source, technology and activity per unit of activity. Finally, $AL_{s,i,j}$ is the activity level of the emission source, the technology and the activity (in t yr^{-1} or in GJ yr^{-1} or in veh km yr^{-1} or in LTO yr^{-1}, etc.).

Though the methodologies may be common there are a variety of tools and emission 'models' available for producing emission data for air quality models. In the table below a number of relevant handbooks, models, data sources and documents that can be used to determine emissions and establish emission inventories are given. A summary description of some of these is provided in Appendix 2.

In the following sections of this document a number of particular source categories relevant for NO_2 modelling are discussed. These are approached with a view to the establishment of urban emission inventories for both local (e.g. line source) and urban (e.g. aggregated emissions) sources.

Table 13. Sources of information, tools, data and other documents relevant to the establishment of emission inventories.

Source (year)	Comments/description	References and links to documentation
EMEP/EEA (2009)	Air pollution emission inventory guidebook, in previous versions known as EMEP/CORINAIR, has been developed over many years in cooperation between EEA and the UNECE/EMEP Task Force on air Emission Inventories and Projections (TFEIP). Contains guidance and most information necessary for establishing national air emission inventories, spatial emissions mapping and emission projections	http://www.eea.europa.eu/publications/emep-eea-emission-inventory-guidebook-2009
CEIP	Centre on Emission Inventories and Projections. Provides national air emission inventories and 50 x 50 km gridded emission data of the UNECE region.	http://www.ceip.at/
E-PRTR	European Pollutant Release and Transfer Register (166/2006/EC). Database which includes air emissions of large industrial point sources from EU-27, Iceland, Lichtenstein, Norway and Switzerland since the year 2007. The reporting threshold for NO_X is 100 tonnes/year.	http://prtr.ec.europa.eu/
LCP-D	European Large Combustion Plant Directive (2001/80/EC). Database which includes air emissions, fuel input and flue gas information of fuel combustion plants exceeding 50 MW of rated thermal input.	http://www.eea.europa.eu

Eurostat	Provides European statistics at national and regional level, e.g. national energy consumption data or regional population statistics. Additional statistics may be retrieved from national or regional statistics offices.	http://epp.eurostat.ec.europa.eu/
EDGAR	Emission Database for Global Atmospheric Research located at the Joint Research Centre IES. Includes sectorial gridded data on 0.1 x 0.1 degree grid.	http://edgar.jrc.ec.europa.eu/
HBEFA 3.1 (2010)	The Handbook of Emission Factors for Road Transport. Developed on behalf of the Environmental Protection Agencies of Germany, Switzerland, Austria, Sweden, Norway and France. JRC (Joint Research Center of the European Commission) also supports the development of the HBEFA. HBEFA 3.1 was released in January 2010	http://www.hbefa.net/e/index.html
TREMOVE (2007)	TREMOVE is a policy assessment model, designed to study the effects of different transport and environment policies on the emissions of the transport sector. Emission factors are based on COPERT.	http://www.tremove.org/
COPERT 4 (2010)	MS Windows based traffic emissions model on which the EMEP/CORINAIR emission factors are based. JRC and EEA support the development of COPERT.	http://lat.eng.auth.gr/copert/
TREMOD (2008)	Transport emission model developed in Germany. Based on the HBEFA emission factors. Includes scenarios up to 2030	http://www.ifeu.de/english/index.php?bereich=ver&seite=projekt_tremod
TRENDS (2003)	TRansport and ENvironment Database System. Contains national level trends in transport emissions for EU15 countries from 1970 – 2020. Established in 2003.	http://air-climate.eionet.europa.eu/databases/TRENDS/index_html
ARTEMIS	European project aimed at the development of a harmonised emission model for road, rail, air and ship transport to provide consistent emission estimates at the national, international and regional level. The resulting transport emission inventories model, is now underpinning the further development of the HBEFA.	http://www.trl.co.uk/ARTEMIS/
AIR4EU (2007)	Report on 'Emissions and data needs': Contains a review and guidance on the establishment of emission inventories. 'Final Recommendations for AQ assessment': contains recommendations for modelling and emission inventories at local and urban scales.	http://www.air4eu.nl/reports_products.html
E-MAP	Interpolates coarse (EMEP) emissions to a finer model grid and employs proxy data (land cover, population density) E-MAP (Emission Mapper) is a GIS based tool developed by VITO to spatially disaggregate emissions over different air quality model grids	Maes et al. (2009)
EMIT (2010)	Emissions Inventory Toolkit contains emissions factors for transport, industrial and domestic sources. Developed by CERC, UK.	http://www.cerc.co.uk/environmental-software/EMIT-tool.html
FAIRMODE (2010)	Background document on the emission needs at local scale for AQ modelling (Working Group 2, subgroup 3). Preliminary discussion on emissions and projections. issues related to modelling activities in the scope of the	http://fairmode.ew.eea.europa.eu/fol404948/sg3_background_doc ument_oct10_draft.pdf

	AQD.	
GEIA (2010)	Global Emissions Inventory Activity. This is an international network dealing with emissions inventories. It provides links to data set and organises conferences, mostly related to global data but also to regional-urban data.	www.geiacenter.org/
MIMOSA 4	Generates hourly output for different types of emissions for Flanders (or other flat countries), using COPERT 4. The model is geographically distributed, using as input modelled amounts of vehicles for each road. Vehicle category distribution is based on statistical data of the vehicle fleet in Flanders.	Mensink et al. (2000) Vankerkom et al. (2009)
TREFIC	Software program for the estimation of air pollution emissions related to road traffic (Italy)	http://www.aria-net.it/front/ENG/codes/files/7.pdf
GAINS	Energy/emission module of the GAINS integrated assessment model is used to generate scenarios up to 2030 at European or sub-national level (e.g. GAINS-Italy)	GAINS-Europe: http://gains.iiasa.ac.at/index.php/gains-europe GAINS-Italy: http://gains-it.bologna.enea.it/gains/IT/index.login
HERMES (2004)	The High-Elective Resolution Modelling Emission System (HERMES) generates the emission for Spain at a spatial and temporal resolution of 1 km^2 and 1 hour. HERMES considers both anthropogenic (power generation, industrial activities, on-road traffic, ports, airports, solvents use, domestic and commercial fossil fuel use, agriculture and livestock) and biogenic (vegetation) emissions using a bottom-up approach.	Baldasano et al. (2008) http://www.bsc.es/caliope

5.2 Emission inventories for traffic

One of the major sources of NO_2 in urban areas is traffic. The most widely used emissions model for traffic in Europe is the COPERT emissions software system developed for the European Commission (Gkatzoflias et al., 2007) in the framework of the CORINAIR project and can be used free of charge by most European countries. It is currently integrated in the EMEP/EEA methodology for emission computation (Ntziachristos and Samaras, 2009). The use of this system is recommended in order to promote a harmonised approach in the development of national transport emission inventories in Europe.

The Handbook Emission Factors for Road Transport (HBEFA) is also a complete emission database that provides emission factors for all vehicle categories (heavy goods vehicles, light commercial vehicles, passenger cars, urban buses, coaches and motorcycles), each divided into different categories, for a wide variety of traffic situations. Emission factors for all regulated and the most important non-regulated pollutants as well as fuel consumption and CO_2 are included. The HBEFA also contains a database for vehicle mileage and traffic situations based on the vehicle mileage for a number of specific countries (Germany, Austria, Switzerland and Sweden), but the data can be used after adaptation in other countries as well.

Emission factors from HBEFA are already included in the COPERT model and vice versa. It should be taken into account that emission calculations based on HBEFA NO_X emission factors produce higher values compared to previously used emission factors (http://www.hbefa.net/e/index.html). The COPERT model has recently (November 2010) been upgraded with new emission factors for heavy duty vehicles. The new emission factors

take into account the even higher NO_X emission factors caused by 'Selective Catalytic Reduction' (SCR) equipped trucks in urban driving environments. The HBEFA 3.1 and COPERT 4 models share many similarities, as the input emission factor data for all three models is based to a large extent on the emission factor database compiled in the frame of the ARTEMIS and COST 346 projects. The fundamental difference between the two models lies in the methodology for converting raw (input) emission factors to operational (output) emission factors. More specifically, HBEFA 3.1 applies a traffic situation (kinematics) approach, whereas in COPERT 4 an average speed approach is used (Sjödin and Jerksjö; 2008).

As part of the effort to harmonise emission inventories for transport sources the ERMES (European Research on Mobile Emission Sources) group has been established, supported by JRC. This group was created in 2009 in an effort to bring together all European groups working on Transport emission inventories and models, with a first focus on road transport. Objectives of this group include the coordination of research and measurement programmes for the improvement of transport emission and fuel consumption modelling in Europe, a Clearinghouse for European mobile emission modelling tools, and relevant guidance and training.

5.3 Traffic emissions and the NO_2:NO_X ratio

Since NO_X is usually the only emission reported there is generally no 'official' information concerning the split of NO_X into NO and NO_2. This is to a large extent the result of legislation (National Emission Ceilings Directive 2001/81/EC, Articles 4.1 and 7.1; EC, 2001) that requires only NO_X emissions to be reported (ECE, 2008). This is an important issue for air quality modelling purposes if an accurate estimation of NO_2 concentrations from road vehicles is to be made. Prior to 2000 this ratio was assumed to be around 5% but more recent results show that this ratio has been increasing and in 2010 it is in the range of 10 – 20%. The change in ratio is attributed to changing technologies and of course changing traffic fleet configurations with the introduction of these new technologies.

5.3.1 Assessment of the NO_2:NO_X ratio by measurement

In general two methods can be employed to determine this ratio. These are direct exhaust emission measurements, either in the laboratory or in the field, or ambient air concentration measurements in the field, after compensation for chemical processes. The last of these is frequently used by national and local authorities as these indicate the impact of 'real world' total emissions and trends can be established based on long term measurement data.

Analyses of air quality and remotely sensed emissions, e.g. Carslaw et al. (2011) and Carslaw and Beevers (2005b), and of routine air pollution measurement data in the UK, AQEG (2007), have demonstrated an increase of the volumetric NO_2:NO_X emission ratio from road traffic. The Air Quality Expert Group (AQEG, 2007) reported a decrease of NO_X by a factor of two in monthly data for busy roadsides in outer London, whereas no change was recorded in NO_2 emissions. This shift in the NO_2:NO_X ratio could be attributed to the introduction of strong oxidation catalysts used on light duty diesel vehicles (Diesel Particle Filters) to meet Euro III limits, resulting in higher oxidation rates of NO to NO_2. Another reason could be the deliberate production of NO_2 in heavy duty vehicles, such as buses, to facilitate the oxidation of particles. Therefore, the increased use of diesel as a fuel in cars and increased flows of buses, as is the case for London, would lead to higher NO_2 road traffic emissions.

In a similar study (Kessler et al., 2006) the same method has been applied to four cities in Baden-Württemberg using hourly air quality data for the years 1995 to 2005. Results for NO_2:NO_X ratios of traffic emissions start at 5 % (1995) and show a continuous increase since 1999/2000 to values of over 20% (2005).

Using data from the measuring network of the RIVM (The Netherlands) the fraction of NO_X that is emitted by road traffic as NO_2 was deduced for the period 2001-2007 (Mooibroek, 2009). The average fraction was found to slowly increase from 7% in 2001 to 15% in 2007. On busy highways, with a relatively large amount of new cars, the percentage is slightly higher.

In a study by Grice et al. (2009) the NO_2:NO_X ratio was assessed for a number of measurement sites throughout Europe. They found that the NO_2:NO_X ratio has increased, on average, from 8.6% in 2000 to 12.4% in 2004.

Measurement of exhaust emissions carried out in real world conditions for a range of driving cycles of Euro3 and Euro4 vehicles (Alvarez et al., 2008) have shown that real world emissions of NO and NO_2 may be poorly represented by standard laboratory driving cycles.

5.3.2 Emission models and the NO_2:NO_X ratio

The HBEFA database provides separated NO and NO_2 emission factors while COPERT produces aggregated NO_X emissions (as NO_2 equivalent) that are split afterwards according to predefined fuel/technology-specific ratios. These ratios range from less than 5% for gasoline vehicles to up to 55% in Euro4 diesel passenger and light duty vehicles. NO_2:NO_X ratios for newer technologies (Euro5 and Euro6) remain unclear at the moment.

An emission inventory was developed for road traffic emissions for London based on recent exhaust emission measurements of NO_X and NO_2 from a range of vehicle types and technologies (Carslaw and Beevers; 2005a). The results suggested that the mean NO_2:NO_X ratio on road links in London was 10.2%, but this percentage was not uniform: the lowest proportion of NO_2 (less than 5% by volume) was observed on major roads, where the vehicle speed is high, and it significantly increased to 12% on the congested roads of Central London, where emissions from diesel vehicles such as taxis and buses are high. This spatial variation of NO_2:NO_X ratio, which depends on the vehicle type, the fuel used and vehicle operating conditions, has to be considered in the development of road traffic emission inventories, particularly for urban areas.

In the study by Grice et al. (2009) An analysis of the predicted national level emissions, based on changing fleet composition and technologies, showed a similar trend to those measured in Europe, going from 6.3% in 2000 to 10.6% in 2005. That study was based on the TREMOVE traffic and emissions model.

5.4 Other transport related emissions

5.4.1 Shipping

NO_X emissions released by ships at high local concentrations into the marine boundary layer may give rise to local environmental impacts, particularly around harbours, and participate in O_3 reactions over larger distances. Total shipping NO_X emissions contribute by 5–12% to the total anthropogenic NO_X emissions. Thus, NO_X shipping emission patterns and fluxes should be accounted for in a complete emission inventory, but particular emphasis should be placed on harbour emissions when dealing with urban scale modelling. Existing global emission inventories for shipping have used a top-down analysis to estimate emissions based on global fuel burn. However, the discrepancy of the results compared to data published by the international marine bunker industry highlight the degree of uncertainty in these emission estimates. This can be primarily attributed to the limited data availability regarding maritime and inland water transport. A detailed methodology to calculate shipping emissions for regional scale applications is described in the EMEP/EEA Emission Inventory Guidebook.

European regional ship emission inventories use a bottom-up approach and calculate emissions on the basis of geo-spatial information, which means the distance each ship covers. ENTEC (Whall et al., 2002), EMEP (Vestreng et al., 2007), CONCAWE (2007) and IIASA (Cofala et al., 2007) are regional European inventories which have the same spatial distribution and methodology in common and hence lead to comparable emission estimates for all compounds. In contrast, global emission inventories calculate emissions based on the total fuel consumption (energy statistics data or estimated from fleet activity) including a technology split accounting for different ship types and sea and port activities (Marmer et al., 2009).

5.4.2 Harbours

In coastal urban areas, emissions related to a number of activities in the harbour area may constitute a significant emission source (Cooper; 2003). Therefore, harbour emissions should be accurately estimated in urban emission inventories including emissions from loading/unloading activities and combustion processes during wharf operations that can be calculated on the basis of mercantile fleet data, average in-port idling time and type of transported goods. For estimating ship harbour emissions three different stages should be considered, namely manoeuvring, hotelling and cruising. Data on shipping movements in the studied area, vessel data (e.g. engine power, fuels), fuel consumption and emission factors from the existing literature (Entec, 2002; Lloyd's Register, 1995, 1999) are required for the calculations (Gariazzo et al. 2007b and MARIN 2010, 2011).

The realistic estimation of emissions from ships at berth requires reliable information on fuel consumption while at berth and associated fuel characteristics, which is usually scarce. In some cases, such as in a relevant study for the Port of Rotterdam, an on-board survey including the filling in of a questionnaire by the chief engineer of

the ship can provide the necessary data (Hulskotte and Denier van der Gon, 2010). Particular attention should be given to shipping emissions from the smaller auxiliary engines when ships are stationary (with main engines shut down) and at berth in ports close to population centres. In the relevant paper by Cooper (2003) a methodology for addressing emissions from ships at birth is suggested, based on empirically derived, emission formulae using dead weight tonnage.

5.4.3 Airports

For European countries, an airport emission inventory based on a daily evaluation of LTO cycle (Landing and take off) for each aircraft category, derived from data recorded in every airport can be developed (Caserini et al.; 2001). Emission factors for the main aircraft types can be derived from the EMEP/EEA Emission Inventory Guidebook, expressed as mass of pollutant emitted during every stage of LTO cycle. Emission factors in the EMEP/EEA Guidebook are available for 45 aircraft types and 8 pollutants (including SO_2, NO_x, CO, CO_2 and TSP). However, emission factors are not provided for a significant number of aircraft categories characteristic for European airports, mostly for the small models. In this case, emissions from these aircrafts can be estimated based on their size and engine characteristics after grouping them into representative categories, and using available emission factors from similar aircraft types.

Associated with airport activities are the emissions from ground support equipment (GSE) and the enhanced transport activities delivering freight and people to and from the airport. The contribution from these sources may be much more significant to NO_2 concentrations than the aviation activities themselves. In regard to the calculation of emissions from GSE, fleet data (type and number of vehicles, fuel feeding, total working hours) and annual fuel consumption are required, as well as average emission factors for non-road mobile sources and machinery as proposed in the EMEP/EEA Emissions Inventory Guidebook.

5.5 Home heating and domestic emissions

Emission factors and methods can be found in the EMEP/EEA Emission Inventory Guidebook or from specific national studies based on representative measurements, when available. Small combustion installations are mainly intended for heating and provision of hot water but some of these installations are also used for cooking. The applications can be conveniently sub-divided by considering the general size and the combustion techniques applied:

- residential heating — fireplaces, stoves, cookers, small boilers (< 50 kW);
- institutional/commercial/agricultural/other heating including:
    - heating — boilers, space heaters (> 50 kW),
    - smaller-scale combined heat and power generation (CHP).

In small combustion installations a wide variety of fuels are used and several combustion technologies are applied. In the residential activity, smaller combustion appliances, especially older single household installations are of very simple design, while some modern installations are significantly improved. Emissions strongly depend on the fuel, combustion technologies as well as on operational practices and maintenance.

For the combustion of liquid and gaseous fuels, the technologies used are similar to those for production of thermal energy in larger combustion activities, with the exception of the simple design of smaller appliances like fireplaces and stoves. The technologies for solid fuels and biomass utilization vary widely due to different fuel properties and technical possibilities. Small combustion installations employ mainly fixed bed combustion technology, i.e. grate-firing combustion (GF) of solid fuels.

As with other emission inventories, information concerning the emission factors and activity/consumption data is required to calculate emissions. For domestic emissions this can be a complex task as heating technologies, fuel types and consumption patterns may vary widely from country to country, city to city and even within a city. Though guidelines and standard emission factors are available in the EMEP/EEA Emission Inventory Guidebook, national or local activity surveys and emission measurements will often be required to build up a suitable emissions inventory.

In most cases the activity data for residential combustion is provided on a statistical basis, a top-down methodology is recommended for emission estimation. In the example of the urban emission inventory developed for the Antwerp area (Moussiopoulos; 2003), the emissions due to space heating were assessed by a collective registration per km². The inventory was based on a top-down approach and included private households and non-private buildings. The emissions were calculated depending on the type of building (e.g., apartment, family residence), the type of heating system and type of fuel used. The emissions for private households were based on a 10 yearly census of houses per statistical unit, consisting of similar building types.

5.6 Industrial and other combustion sources

The energy industries activity covers combustion and conversion of fuels to produce energy, for example electricity or heat. The combustion activities undertaken in manufacturing industries generally provide process heat (directly or indirectly usually via steam, water or oil), electricity, or the fuel may be transformed. Emission factors and methods can be obtained from the EMEP/EEA Emission Inventory Guidebook, from the US Environmental Protection Agency (USEPA) emission factor handbook (http://www.epa.gov/ttn/chief/ap42/) or derived from plant specific data reported within national and international frameworks (e.g. E-PRTR, ETS, LCP-D).

Details of technologies used can be found within the Best Available Techniques Reference Documents (BREF) for energy installations, refining installations and Iron and Steel production European Integrated Pollution Prevention and Control Bureau (EIPPCB; http://eippcb.jrc.es/reference/).

Information on the production of power and production of power is available from national statistics agencies or the International Energy Agency (IEA). Typical sources for this data might be industrial or regulatory organisations within the country or from specific questionnaires to the individual combustion installations.

Since E-PRTR generally does not report activity data, such data in relation to the reported facility-level emissions are sometimes difficult to find except for facilities reported under the LCP-D. Another possible source of facility-level activity might be the national registries of emission trading systems. In many countries national statistics offices collect production data on facility level, but these are confidential. However, in several countries, national statistics offices are part of the national emission inventory systems and the extrapolation, if needed, could be performed at the statistics office, ensuring that confidentiality of production data is maintained. National production data may be also retrieved from national or international industrial associations. A power plant database including information on location, type, size and emission abatement technologies may also be commercially obtained through sources such as www.platts.com.

In many cases emissions from the industrial sector are dominated by a limited number of large companies, which are usually obliged by legislation to report their emissions yearly to the regional authorities. Based on these registrations and on additional calculations using statistical data and emission factors from the literature, a yearly emission inventory of industrial point sources can be compiled with satisfactory accuracy.

5.7 Future trends in traffic emissions

In order to develop plans to improve air quality, information concerning future emissions is required. For the most part local authorities responsible for air quality in any city or zone will have a number of abatement strategies available. Structural changes, such as car free zones and implementation of public transport, are measures that are to some degree under the control of local authorities. However, local authorities have no control over long range transport or over technological changes that will result in changing emission factors, e.g. Euro standards. Changes in fleet composition are to some degree affected by local or national policies but even these are driven by other external factors, e.g. climate change mitigation, particle emission reductions and economics. It is thus necessary to find future estimates for these elements in order to provide realistic planning scenarios.

In particular, future emission factors are quite uncertain. Experience has shown (e.g. Sturm et al., 2001; Alvarez et al., 2008; TNO, 2009; and Carslaw et al., 2011) that real world emissions and promised emission rates are rarely the same. Currently the Euro V fleet is being introduced and the Euro VI fleet will be introduced in the near future. Current estimates indicate that NO_X emissions for heavy duty vehicles should reduce significantly, by a factor of around two. However, direct emissions of NO_2 may not be reduced by the same amount.

The studies by Grice et al. (2009) has shown that the combination of new technologies and changes in vehicle makeup will lead to an average increase in the NO_2:NO_X ratio, in a selected number of European cities, from 10.6% in 2005 to 19.6% in 2010 and 32.0% in 2020. These increases in ratio could be offset by reductions in total NO_X emissions with a small decline in NO_2 concentrations between 2010 and 2020. However, for the most relevant emission sectors, the NO_2:NOx ratio for future technologies are not available. For instance, in the road transport sector, Euro V and Euro VI ratios are not available in COPERT. Those corresponding to Euro4 technologies can be used as a default, but more specific values should be used if available (based on local studies, etc.). An estimation of the impact that alternative NO_2 ratios have on the total NO_2 emission may be included to try to account for the uncertainty of this critical assumption for emission scenarios and abatement options related to road traffic, most particularly Low Emission Zones (LEZ).

Strategies to improve air quality, especially in the mid-term, include new technologies and behavioural changes that are difficult to model in terms of emissions both at the emission factor reduction level and the penetration rate. For instance, inclusion of electric vehicles in a significant percentage of the fleet leads to uncertain figures for vehicle sales, vehicle mileage, proportion of hybrid vs. pure battery technologies, charging performance, etc. Therefore, future scenarios should be accompanied by appropriate uncertainty estimates.

Other critical property for emission projections is their consistency with the past series. Likewise, the inventory should be adequate to the needs of emission scenario definition allowing the establishment of any relevant hypothesis. The design of the emission inventory must be able to give a good representation of any of the abatement options envisaged and to allow the quantification of the impact of individual measures and policies to understand the reasons of the achievement or exceedance of NO_2 limit values.

Application guidance

The following points are noted when developing and applying emission inventories for the use of NO_2 modelling in urban areas.

1. The use of established, supported and harmonised emission databases and methods is highly recommended for applications across Europe. For this the EMEP/EEA Guidebook (EMEP/EEA, 2009), which includes the COPERT methodology for road traffic, constitutes the basic reference. However, this guidebook concentrates on national level reporting and urban air quality modelling applications may require a substantial adaptation of inventories already in existence or the development of new ones.
2. The modelling requirements for the AQ Directive imply the need to cover both local and urban scales, which often means the combination of different model types (e.g. nested Eulerian models and sub-grid street level models). Therefore the consistency of emission data and methods across the scales/models should be controlled and checked.
3. The emission inventory development/adaptation process has to be transparent and well documented so the representativeness of the results can be fully understood and the main deficiencies can be addressed in future updates (e.g. emission factors, NO_2:NO_X ratios, statistics used as activity data, etc.). An emissions inventory must therefore contain not just emission factors and activities but the data on which these are based and the methods used to calculate these.
4. Essential to any emissions inventory are the uncertainties in the method and the data used to create them. The same is true for any scenario calculations. Without such emission uncertainties it is not possible to apply the results of air quality modelling in a meaningful way.
5. Continuous updates of emission factors are necessary and new emission factors need to be updated regularly in emission inventories.
6. Emission inventories for just NO_X are not sufficient. Both NO and NO_2 emissions are required.
7. For urban emission inventories in particular, which require accurate and detailed data, representative measurements at different source types (traffic, point sources) of the required area should be conducted to ensure that European emission factors and emission data also reflect the specific case. In some cases, in order to ensure the representativeness of the European databases, some adjustments should be made, for example for local fuel properties (Tsilingiridis et al.; 2002).

8. The temporal and spatial variation of emissions from all sources should be well considered in the preparation of emission inventories. Therefore, wherever possible the collection of data on the temporal variation of transport activities and fuel consumption of all emission sources is recommended, in order to produce temporal profiles (hourly, daily, weekly, monthly) to be used as input in the emission inventory.
9. The top-down approach for calculating time-dependent emissions data, based on the disaggregation of the annual mean value and producing time-dependent functions for monthly, weekly and daily periods, is typical for industrial or residential combustion sources. The bottom-up approach, where the calculation starts on an hourly basis and the annual value is produced as the sum of the hourly results, should be used for traffic emissions, where activity data is usually known on an hourly basis (Sturm et al.; 1999). Similarly, for addressing the spatial resolution of the emission sources, particularly in high-resolution urban emission inventories, bottom-up approaches are applied for line sources (traffic) or point sources (big industrial units), whereas the top-down methodology where statistical information from bigger units is disaggregated is suggested for residential sources and small business activities.

6 Meteorological data

All air quality models require some form of meteorological data to drive them. However, the types of inputs can vary significantly from model to model. Typical meteorological data used in air quality modelling are:

1. *Observed meteorological data:* Various forms of data are available that may be used directly for air quality models or processed further in conjunction with models. These include a range of surface synoptic measurements, turbulence measurements, radiation measurements and vertical profile measurements.
2. *Statistical meteorological data:* Some models use observed statistical meteorological data (e.g. wind roses). These data are usually based on observations.
3. *Diagnosed meteorological data:* Meteorological wind fields (2D or 3D) may be determined using simplified models based either on linearized techniques or on interpolation of available wind observations applying a mass conservation constraint. The latter are widely used in air pollution models and are referred to as diagnostic (or mass-consistent) wind models.
4. *Prognosed meteorological data:* These are outputs from numerical models that solve the relevant physical equations using some form of Eulerian grid, e.g. Numerical weather prediction models (NWP). Originally prognostic models for the wind field did not take into account observations, but in the last decade this type of model makes increasingly use of meteorological observations through data assimilation techniques.
5. *Meteorological pre-processors:* Often meteorological data does not contain the required information for the dispersion model, e.g. concerning turbulence, or the output from a NWP model has to be adapted to the requirements of the chemical transport model in terms of grids and variables (e.g. mixing height and eddy diffusivity). The meteorological pre-processors, or interface models, estimate all relevant meteorological variables for the calculation of concentrations and provide consistency between the meteorological and the chemical transport models.
6. *Object resolving models:* When high resolution wind fields are required, e.g. in street canyons or built up areas, then micro-scale meteorological models that resolve the individual buildings can be applied. Obstacle resolving models for the flow field and the dispersion are of two main types: complex codes, like computational fluid dynamics models (CFD), and simplified ones, also known as fast response dispersion models. The former are rarely used for AQ Directive applications due to their computational expense. The latter were developed primary for emergency response systems (e.g. QUIC for urban dispersion), but are used recently also for long term simulations (e.g. Moussafir et al., 2010). This second type of model is also diagnostic in nature.

The need for meteorological data is strongly dependent on the application. In general for urban scale assessment either diagnostic or prognostic models are used. Usually Eulerian based air quality models (Section

3.4 and 3.5) have a prognostic model as meteorological driver, while Lagrangian models may be driven by a diagnostic wind model and a relevant meteorological pre-processor. Gaussian based models can generally be applied using both diagnostic and prognostic fields. Local scale models may require only a single observational site as input.

The advantage of prognostic models is that they can be used for forecasting purposes and give full spatial coverage, though they will generally have a higher uncertainty than observed data. The advantage of observational data is that they reflect the real situation, but have limitations in terms of spatial coverage and variables measured. Diagnostic models take into account available observations, but have very simplified physical basis (one conservation equation). These limitations may be addressed by data assimilation techniques that combine in an optimal way atmospheric dynamics and observational data by solving the primitive equations system (e.g. Seaman, 2000). Data assimilation is a common approach in NWP models on the regional or synoptic scale and has been demonstrated to improve significantly the accuracy of modelled meteorological fields and concentrations (e.g. Zhang et al., 2009). However, the application of data assimilation methods on the urban or local scale is more complex and is rarely carried out, mostly due to the limited spatial representativeness of the urban observations.

In the recent COST728 action (COST728, 2010) the application of meso-scale meteorological models for air quality applications in Europe was extensively investigated. In the Air4EU project a review and set of recommendations concerning meteorological data for air quality modelling is given (Air4EU, 2007). Both COST710 (1998) and COST715 actions (2004) have provided reviews and recommendations concerning meteorological modelling and pre-processors.

6.1 Observational data

Description

Often the best meteorological data is that which is observed, with the disadvantage that observations may not be representative of a larger area. Only when the terrain is relatively homogenous can point data be extrapolated and used to represent a wider area. There are several types of observational data that are commonly used, either as direct input to air quality models, or in conjunction with diagnostic models. Regional scale (meso-scale) meteorological models may also assimilate these data into the model itself using data assimilation techniques.

The accessibility and quality of meteorological data varies but there are standard protocols and methodologies for a wide range of these, particularly those reported to the WMO (www.wmo.int/pages/prog/www/IMOP/IMOP-home.html).

Examples

Typical meteorological data used in air quality modelling are listed in Table 14.

Table 14. Different types of observed meteorological data that may be used for air quality modelling.

Observation	Comments/description	Application
SYNOPTIC ground based measurements	These are standard meteorological measurements carried out and reported to the WMO (World Meteorological Organisation). These measurements are standardised and are most readably available through national meteorological bureaus. Generally these will record hourly data but may only report 3 – 6 hourly data.	Used for validation and assimilation in weather prediction models. Can be used for diagnostic models to produce wind fields.
Radio sonde data	Sonde releases are carried out regularly throughout the world at either 6 or 12 hour intervals. Vertical profiles of wind, temperature, pressure, and humidity are standard. These data are available through the national weather bureaus. They are less widely distributed spatially	Used for validation and assimilation in weather prediction models. Can be used for diagnostic models to produce wind fields. Vertical profiles are particularly useful for initialising boundary layer height and providing

	than the ground based synoptic measurements.	upper boundaries for diagnostic models.
Sonic measurements of turbulence	These types of measurements, often using sonic anemometers, sample wind speed and temperature at high frequency (e.g. 20 Hz). From these high frequency samplings turbulent parameters can be derived. Such 'sonic' instruments are often used for special measurement campaigns but are nowadays used more regularly.	These measurements may provide turbulence parameters for Gaussian type models when local assessments or model development/validation are being carried out
Radiation measurements	In addition to standard meteorological measurements, both long and short wave radiation are often measured. Most commonly measured is global radiation, which is the total incoming shortwave radiation per surface area.	Radiation measurements are often used to determine atmospheric stability and is also required when the surface energy balance must be determined. This type of data is often requested/required in meteorological pre-processes used for Gaussian models
Wind profilers (SODAR)	Both radar, lidar or sonic methods may be used to derived high resolution vertical profiles of wind and temperature remotely. These types of measurements are often limited to the boundary layer and are regularly used for monitoring the development of the boundary layer.	Few operational air quality models use these data directly. They are often used for validation of boundary layer development or for particular campaigns
Masts and roof tops	When vertical profiles are required using normal measurement techniques then instrumentation may be fitted to masts or on roof tops. This is common for campaign measurements but less often used as routine observations.	These data provide vertical profiles in the lowest 10 – 200 m (depending on the mast height). Very useful in the urban canopy where vertical profiles of wind and temperature may differ significantly from standard profiles. Roof top observations are often required as input data for street canyon models.

Application guidance

1. If air quality data is to be used for validation purposes then it is extremely useful, almost absolutely necessary, that meteorological measurements also be performed at the air quality site. This aids interpretation of the modelling results significantly.
2. If roof top observations are required then these should be made well above the roof top itself to avoid undue influence form the local obstacles.
3. If diagnostic models are to be used in urban areas then urban meteorological measurements must be representative of an area equivalent to at least the model resolution, before they should be included in the model. This means that measurements in built up areas need to be made at least twice as high as the surrounding building height. Radio towers are one possible site for such observations.

6.2 Statistical meteorological data

Description

Some statistically based models, e.g. long term Gaussian models, use climatological or statistical meteorological data as input. Such data is generally derived from available observations and is often presented as wind rose data, where the frequency of particular wind speeds, wind directions and atmospheric stability are binned and tabulated and used as meteorological input. Use of these data should ideally be limited to areas that are homogenous since there is no physically meaningful way of interpolating wind roses in space.

Application guidance

1. Statistical meteorological fields are often sufficient for annual mean calculations, when applied to steady state dispersion models.

6.3 Diagnostic wind field models

Description

This type of meteorological model uses observational data to generate quasi-steady state 3D wind fields, in general under the constraint of mass conservation (known also as mass-consistent wind models). The 3D wind field is reconstructed starting from available observations through a two-step procedure. At the first step observations are interpolated to the computational grid employing some form of objective analysis and/or appropriate parameterisations, while at the second step this "initial" wind field is adjusted to satisfy mass conservation by minimum possible modifications. Originally mass consistent models were developed to describe the circulation over complex terrain on a regional scale. Later this approach was adapted to local / urban scale in order to account for building's modifications on the flow and the dispersion.

Mass-consistent models simulate the dynamic effects of the surface terrain or obstacles on the flow; all other physical phenomena (e.g. thermal circulations) are implicitly included either in the observations, or through parameterisations in preparing the "initial" wind field. For regional scale models parameterisations may refer to sea breeze or down slope circulations (e.g CALMET), while for local scale models the parameterisations are based on semi-empirical expressions for the flow regimes around a single building or typical arrangement of buildings (Röckle, 1990). These semi-empirical expressions have been continuously improved based on wind tunnel experiments or field campaigns in built up areas. At both scales the effect of the atmospheric stability on the flow can be included through the mass consistent adjustment step. However, results show that the "initial" wind field plays a crucial role for the accuracy of the final 3D wind field.

Input data for a regional scale diagnostic model includes surface based measurements, upper air data, wind profiles (measured or calculated by a coarser grid prognostic model) and terrain data (topography and land use). Input data for a local scale model usually only includes an upstream vertical profile, or the wind speed above roof level, but 3D building geometry in the modelled domain has to be well represented.

A number of regional scale diagnostic models have been developed and are described in both research papers and user manuals. Available reviews can be found in Ratto et al (1994) and in the COST710 review from Finardi et al., (1997). Their most common feature is a methodology for interpolating wind fields at some defined height, and some parameterised description of the vertical profiles. They usually differ in the numerical solver to adjust the wind field. On the local scale the models are only a few and the differences are in the semi-empirical parameterisations for the wind speed in different zones around a single building (upstream, lee-side cavity, recirculation, etc) and between buildings (street canyons, intersections etc.).

Regional scale mass consistent wind models are regularly used for air quality modelling due to the fact that they require less complicated input data than do prognostic models, are less demanding in terms of computational power and generally require little specialized training. Their disadvantage is that they require a large number of representative measurements to produce realistic fields over the entire model domain. Extrapolation of the wind data based on very few measurements, or non-representative measurements, can lead to significant errors in the wind fields they produce. To overcome this shortcoming to some degree, a diagnostic model may be run as postprocessor of a prognostic model in down scaling the wind field to a finer grid.

Local scale (building aware) diagnostic models are used mainly as part of emergency response systems, where the urban flow should be modelled quickly with sufficient accuracy. Recent model intercomparisons for modelling the flow in a complex urban environment show that a diagnostic model performs only slightly worse than CFD codes but 2-3 orders of magnitude faster (Neophytou et al., 2010). Long term simulations for air quality assessment are reported in some recent publications (e.g. Moussafir et al, 2010). The main disadvantage of microscale mass-consistent models is that the urban structure is highly variable, while the parameterisations in the models are based on idealized building structures. Before using such types of models for regulatory applications in a complex urban environment further evaluation would be necessary.

Examples

A number of examples of diagnostic wind field models are listed in Table 15 below, which is adapted from Air4EU, D4.1 (2007).

Table 15. Different types of diagnostic meteorological models that may be used for air quality modelling. Both terrain resolving and obstacle resolving models are listed

Meteorological model	Example AQ model or system (MDS link)	Short description	References
Regional and urban scale, terrain resolving models			
FLOWSTAR	ADMS	Linearized diagnostic model developed by CERC and applied in ADMS.	www.cerc.co.uk/environmental-software/FLOWSTAR-model.html
MATHEW	AirQUIS	Mass-consistent diagnostic model using surface observations as input.	Sherman (1978) www.airquis.no
Airviro wind model	Airviro	Diagnostic, using prognostic model run to equilibrium. Applied in the Airviro air quality system.	Danard (1977) www.smhi.se/airviro
CALMET	CALPUFF	Diagnostic model using synoptic, upper air or model input. U.S. EPA recommended model.	www.src.com/calpuff/calpuff1.htm
WINDS	SAFE_AIR	Mass consistent model using different type of wind observations and/or modeled vertical profiles.	Canepa and Ratto (2003) http://www.fisica.unige.it/atmosfera/www_eng/vento/safe_air.htm
MINERVE	SPRAY FARM	Mass-consistent model using surface observations or prognostic model output .	Geai (1987) www.aria.fr/new_aria/index.php
Local/Urban scale, obstacle resolving models			
ABC	LASAT	Mass-consistent model with semi-empirical parameterisations of flow regimes around buildings.	Röckle (1990) http://www.ima-umwelt.de/ausbreitungsrechnung/modellpalette/abc.html?L=1
QUIC-URB	QUIC	Mass-consistent model. Assimilates multiple meteorological data sources or mesoscale meteorological model	Singh et al (2008) http://www.lanl.gov/projects/quic/quicurb.shtml

		data.	
Micro SWIFT	MSS (Micro Swift_Spray)	Mass consistent model, deals with nested domains. Assimilates available data.	Tinarelli at el (2007) http://www.aria-net.it/ http://www.aria.fr/

Application guidance

1. It is recommended to use diagnostic fields, rather than prognostic ones, for air quality assessment when there is a good spatial coverage of meteorological data and the terrain is not highly complex. Use of observed data is more certain than prognostic data.
2. When using diagnostic models it is recommended to have as extensive an observational network as possible since interpolation between, or beyond, measurements with diagnostic models is quite uncertain. It is particularly important to have meteorological measurements both in- and outside the built up urban region to provide information on both areas.
3. The area of representativeness and the spatial distribution of the meteorological data are also important. The measurements should be representative of an area significantly larger than the grid into which they are interpolated. Better performance is also achieved when the monitoring stations are uniformly distributed. Thus, a critical selection of the observations should be carried out before inclusion in a diagnostic model.
4. For a non dense measurement network diagnostic models perform better in neutral, or unstable, situations since in stable atmospheres the wind at the surface may be decoupled from the upper wind and thus the observations will be representative of a very shallow local flow and not representative of larger areas.
5. When very few monitoring sites are available, or even only one, diagnostic models can still be used as they will still reflect some of the dynamics of the flow resulting from the terrain. Under such situations the observation(s) should be representative of a large area and it is recommended to combine these with either upper air data or prognostic model fields.
6. In general it is recommended to include wind profile data (e.g. from SODAR, Lidar) or, as an alternative, modelling profiles from prognostic models may be used.
7. An assessment of the quality of the meteorological fields calculated by diagnostic models should be carried out. This may be done using cross-validation methods when sufficient observational data are available. Understanding the limitations in the meteorological fields can help to understand discrepancies and uncertainties in the eventual concentration fields.

6.4 Prognostic meteorological models

Description

Most prognostic meteorological models used for air quality applications are also used for numerical weather prediction (NWP). There are a large number of prognostic meteorological models available to produce wind fields for use in air quality assessment. Commonly used models, such as WRF, MM5, ALADIN, HIRLAM, TAPM and RAMS have been used in many applications. See COST728 (2010) for a list and description of such models. These models require more detailed input data and information, especially in regard to surface types and boundary conditions, than do diagnostic models but they do not necessarily require in situ measurements. As such prognostic models can be used for forecasts and are considered to give the best description of the physical processes.

For applications to urban air quality the impact of the urban canopy, i.e. buildings, can be significant. Several of the commonly used NWP models include some form of parameterisation to account for changes in surface fluxes. Schemes such as those from Marttilli et al. (2002) take into account changes in heat fluxes and adapt the

wind profiles in the lower layers to account for building obstacles and the typical surface conditions found in urban areas.

In general prognostic mesoscale meteorological models have been developed for weather prediction applications. These models often focus on prediction of temperature, precipitation and extreme weather events rather than on stable and stagnant weather conditions that are important for air quality modelling. For this reason turbulence parameterisations that are developed to deal with such conditions are preferred when using such models, e.g. Luhar et al (2009).

Prognostic meteorological models are complicated systems that require a good knowledge of meteorological modelling in order to apply them. This makes their application often more costly and less accessible for local authorities or modellers to apply. Simplified prognostic models imbedded in user friendly interfaces, such as TAPM, have been developed so that knowledgeable non-experts may apply them.

Examples

A number of examples of NWP models are provided in the table below.

Table 16. Different types of prognostic meteorological models (weather prediction models) that may be used for air quality modelling.

Meteorological model	Example AQ model or system (MDS link)	Short description	Links and references
MM5	Models-3, CMAQ CAMx	Widely used multi-scale U.S. community meteorological model developed at NCAR. Replaced by WRF	www.mmm.ucar.edu/mm5/
WRF	Models-3, WRF-CHEM, CMAQ, CAMx EMEP	Widely used multi-scale U.S. community meteorological model. Replaces MM5.	www.wrf-model.org/
RAMS	CAMx	Prognostic meteorological model developed at Colorado State University	Pielke et al. (1992) www.atmet.com/
TAPM	TAPM	Combined prognostic meteorological and air quality model. Simpler PC based model interface	www.cmar.csiro.au/research/tapm/
ETA	DMU-ATMI THOR, SKIRON	Prognostic meteorological model developed at NCEP	http://etamodel.cptec.inpe.br/
ALADIN	CHIMERE	French meteorological bureaus forecast model 10 x 10 km resolution	www.cnrm.meteo.fr/aladin/
HIRLAM	Enviro-HIRLAM	European mesoscale prognostic meteorological model.	http://hirlam.org/
ECMWF	MACC ensemble forecasts FLEXPART	Model from the European Centre for Medium-Range Weather Forecasts producing global and European forecasts and analysis	www.ecmwf.int/

COSMO-CLM	AURORA	Non- hydrostatic operational weather prediction model applied and further developed by the national weather services joined in the COnsortium for SMall scale MOdelling (COSMO).	www.clm-community.eu/index.php?menuid=1

Application guidance

1. For almost all regional and urban scale applications the use of prognostic meteorological models is recommended as they provide the most physically consistent method of assessing meteorological fields. This is particularly important when episodic poor air quality occur due to recirculation of air masses. Though diagnostic wind fields may capture stagnation events they do not usually capture recirculation events, as prognostic models are capable of doing.
2. A description of the impact of the urban canopy on meteorological fields is preferred in a meteorological model as this can significantly influence the wind profiles in the lower levels of any prognostic model.
3. Models, or schemes within models, that include turbulence parameterisations that are suitable for low wind conditions are preferable for urban scale air quality applications over models, or schemes, that are developed only for weather prediction.
4. Prognostic meteorological models require application by expert users. It is highly recommended that only trained experts run and interpret these types of models.
5. Meteorological models should be validated against observations when applied to air quality assessment.

6.5 Meteorological pre-processors

Description

Though prognostic models produce surface turbulence parameters, which are essential for calculating dispersion characteristics, observed or diagnosed meteorological data may not contain this information. For this reason so called 'pre-processors' are used in a number of air quality models to derive these turbulence characteristics. Generally each Gaussian type model will have its own type of pre-processor that will derive these near surface turbulence parameters from wind speed, temperature gradients and/or radiations measurements, and surface characteristics such as roughness lengths, see COST710 (2004) and COST728 (2010) for an overview of these

Application guidance

1. Often pre-processors are an integral part of a dispersion model and the model performs optimally with that particular pre-processor, particularly Gaussian type models. For that reason the use of pre-processes is recommended even when turbulent data is directly available from prognostic models.

6.6 Obstacle resolving models

Description

Obstacle resolving meteorological models can in principle provide wind fields and turbulence within the urban canopy at high resolution (< 1 m). Even so, there is large uncertainty in such fields as they can be highly sensitive to input data as well as model assumptions and formulation. Such models are generally not used for AQ Directive applications due to their intensive calculation requirements. They are generally limited to particular process studies or to emergency/accidental release calculations for a short period of time and for a limited region. These models are most often coupled to Eulerian dispersion models or Lagrangian Particle models in order to assess concentration levels. Such models may also be used to assess simpler parameterisations.

Examples

The most extensive overview of these models in Europe was carried out during the COST732 project (http://www.mi.uni-hamburg.de/Home.484.0.html). A model inventory was established during that project (http://www.mi.uni-hamburg.de/Model-Inventory.6295.0.html?&no_cache=1). This type of modelling has been briefly presented in Section 3.2 and is not further extensively discussed as they are rarely used for AQ Directive applications, though this may change in the future.

7 Quality control and evaluation

Quality control and evaluation has been extensively presented and discussed in Chapter 5 of the 'Technical reference guide' for modelling (EEA, 2011). A number of points made in that guidance document are reiterated here but these are further refined with more specific recommendations, relevant to NO_2 modelling.

Though some quality objectives are defined for modelling in the Air Quality Directive (EC, 2008) Annex I, there is no defined quality objective for planning applications of models. The assumption is that any model used for assessment, fulfilling the assessment quality objectives, can also be usefully applied for planning purposes. Since validation of models is not possible for future scenarios this is a generally reasonable position to take.

However, as previously discussed the dynamic sensitivity of a model, i.e. its response to changes in emissions, is not guaranteed even if the current air quality situation is well modelled. This may be the result of parameterised forms of the chemical schemes or incorrect source apportionment of the emissions. For this reason quality assurance of the dynamic sensitivity of the model to changes in emissions should also be assessed.

7.1 Directive related quality objectives

In Section 3.6 of the 'Technical reference guide' (EEA, 2011) a presentation and interpretation of the AQ Directive modelling quality objectives, (EC, 2008) Annex I, is provided. This is summarised here in relation to NO_2.

The quality objectives are given as a relative uncertainty (%). For NO_2 this percentage uncertainty is given to be 50% for hourly means and 30% for annual means. Uncertainty is then further defined in the AQ Directive to mean the following:

> *'The uncertainty for modelling is defined 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.'*

As in the previous Directives the wording of this text remains ambiguous but since values are to be calculated a mathematical formulae is required. The following interpretation is suggested that is called the Relative Directive Error (RDE) and it is defined mathematically at a single station as follows:

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

where O_{LV} is the closest observed concentration 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). It is this value that should be less than the specified uncertainty.

This formulation is similar to that recommended by Stern and Flemming (2004) called the Relative Percentile Error (RPE), which is defined at a single station as:

$$RPE = \frac{\left|O_p - M_p\right|}{O_p}$$

where O_p and M_p are the observed and modelled concentrations at the percentile (p), used to define the exceedance percentile.

The two major differences between the formulations are 1) in the choice of using the closest value to the limit value or using the defined percentile as reference and 2) in the choice of using the limit value or the observed concentration of the percentile as the normalising factor in the denominator. When the observed percentile concentration is the same as the limit value then these two formulations are equivalent. When dealing with annual means the concept is the same, but only one value is available for the calculation, i.e. $O_{p,LV}$ and $M_{p,LV}$ are replaced by the observed and modelled annual means.

There can be arguments for or against the RDE interpretation. For instance if observed annual mean concentrations are well above the limit value then the use of the limit value concentration in the denominator, rather than the observed concentration as in RPE, can lead to large relative errors, e.g. RPE will be satisfied but not RDE. However, the opposite is true when the observed and modelled concentrations are well below the limit value. In such cases the use of RPE can lead to high, and unacceptable, relative errors that would otherwise have been acceptable using the RDE interpretation.

In regard to NO_2 modelling the requirement that the 19'th highest hourly mean modelled concentration be within 50% of the 19'th highest observed hourly mean NO_2 concentration is a very demanding request for a model. However, the 30% target for annual mean concentrations should be achievable for a number of models.

7.2 Validation of models

A model can be validated in several ways, usually depending on the type of application it is intended to serve. In general terms four types of evaluations modes (Dennis et al. (2010) and Rao et al. (2011), Galmarini et al. (2010)) can be identified which, when applied, should provide a rather comprehensive overview of the quality of model results. Namely:

- *Operational Model Evaluation:* Operational evaluation involves the direct comparison of model output with analogous observations. An operational evaluation makes use of routine observations of ambient pollutant concentrations, emissions, meteorology, and other relevant variables.
- *Diagnostic Model Evaluation:* Diagnostic evaluation examines the ability of a model to predict the outcome state pollutant by correctly capturing the physical and chemical processes incorporated in the model.
- *Dynamic Model Evaluation:* Dynamic evaluation focuses on the model's ability to predict changes in air quality concentrations in response to changes in either source emissions or meteorological conditions. This exercise requires historical case studies where known emission changes or meteorological changes occurred that could be confidently estimated.
- *Probabilistic Model Evaluation:* Probabilistic evaluation attempts to capture the uncertainty or level of confidence in model results for air quality management or forecasting applications. This approach is necessarily based on knowledge of uncertainty in both model predictions and observations.

"Operational, diagnostic, and dynamic evaluation approaches complement one another by not only characterizing how well the model captured the air quality levels at that time, but how well the model captures the role and contributions of individual inputs and processes and the ability of the air quality model to respond properly to changes in these factors." (Dennis et al, 2009). While it is true that all evaluation approaches use a statistical formalism, and statistical techniques, the framework conceives probabilistic evaluation as a rather more comprehensive approach than the mere application of statistical tools.

7.2.1 Data requirements

The validation of models requires suitable quality controlled air quality monitoring data, emission data, and meteorological data. In addition information concerning the uncertainties of these data and their spatial representativeness (in the case of monitoring data) is also required. This current document does not provide direct guidance on determining these, as this is largely related to monitoring data. However, FAIRMODE is currently engaged in aspects concerning representativeness of measurement data. Discussions and guidance on this topic will be available in the future

7.2.2 Qualitative analysis

Many types of diagrams (scatter plot, quantile-quantile, time series, etc.) have been used for qualitative model evaluation, each focusing on particular aspects of model performances (distribution, mean values, correlation, etc.). In the FAIRMODE WG2/SG4 proposed template for reporting model performances (http://fairmode.ew.eea.europa.eu/models-benchmarking-sg4/wg2_sg4_benchmarking_v2.pdf), the use of the Target diagram as a main instrument for qualitative assessment purposes is proposed. This diagram (Jolliff et al., 2009) (Figure 6) combines information on standard deviation, bias, root mean square error and model efficiency as well as some information on the correlation coefficient and is an attractive tool to qualitatively summarize model performances.

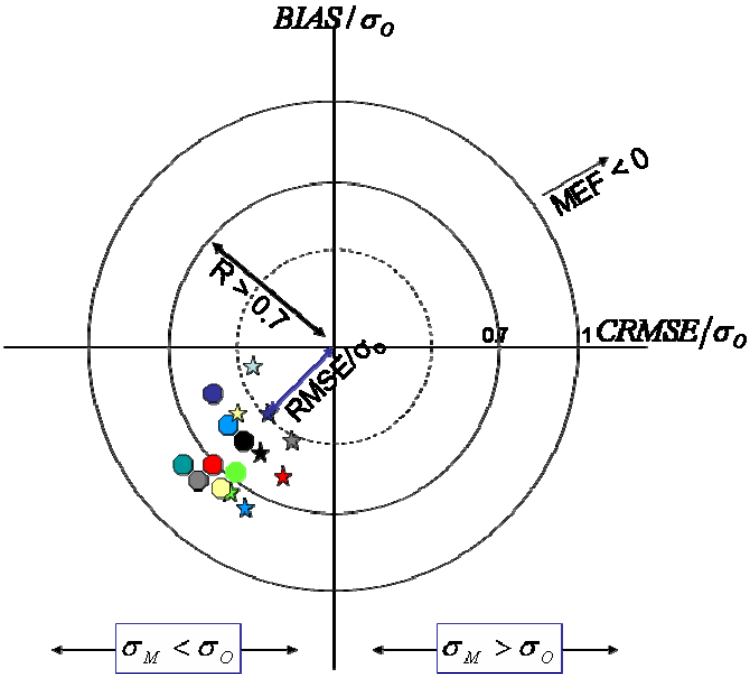

Figure 6. The target diagram displays the model to observation field bias (Y axis) and the model to observation unbiased RMSE (CRMSE) (X-axis). Both axes are normalized by the standard deviation of the observations. The distance between any point and the origin is then the value of the total normalized RMSE. The outermost circle establishes also that all points between it and the origin represent positively correlated model and observations, and also have a better than average MEF score. A second circle may be added to indicate another positive R value, such as R=0.7, for which all points between it and the origin have a correlation greater than 0.7. Finally a dashed line can be used to indicate the average observational uncertainty. If the CRMSE is multiplied by the sign of the standard deviation observation-model difference then the target diagram provides also information about whether the model standard deviation is larger (X>0) or smaller (X<0) than the observations standard deviation.

7.2.3 Quantitative indicators

A wide variety of performance indicators, proposed for different fields (e.g. meteorology, oceanography, air quality), for different applications (e.g. assessment, forecasting, research studies) may be found in the literature. Regardless of the model application, scope and type, it is generally recommended to apply multiple performance indicators since each one has its advantages and disadvantages.

Within WG2/SG4 of FAIRMODE a series of performance indicators have been selected based on a review of various work dealing with model evaluation (see http://fairmode.ew.eea.europa.eu/models-benchmarking-sg4/wg2_sg4_benchmarking_v2.pdf). The following reduced set of indicators has been proposed to capture the main aspects of model performances for a given model application.

- MFB (mean fractional bias)
- RMSE (root mean square error)
- R (correlation coefficient)
- T (Target indicator)
- FAC2 (Factor of two)
- RDE (relative directive error)
- RPE (Relative percentile error)

In this procedure, the approach suggested by Boylan and Russel (2006) which has been adopted in the EPA methodology for model evaluation (EPA 2007 & 2009) has been followed and extended. For each statistical indicator, two quality bounds are proposed: a performance criterion which states whether sufficient quality for policy application is reached and a performance goal which points to the optimum quality level a model is expected to reach. These two literature-based quality bounds, which still need to be reviewed and updated on the basis of modelling exercises and/or expert judgment, are available to help the user assess the quality of its model performances for a given application (see http://fairmode.ew.eea.europa.eu/models-benchmarking-sg4/delta_tool_concepts_userguide.pdf).

7.3 Dynamic evaluation and trend assessment

Dynamic evaluation (Dennis et al. (2010) and Rao et al. (2011), Galmarini et al. (2010)) consists of assessing the response of models to particular aspects of, and changes in, the model inputs. This is particularly important for emissions but is also relevant to boundary conditions and meteorology. When applying the model for planning purposes, it is the response of the model to changes in input data that is the most important. Examples of dynamic evaluation can be found in Gego et al. (2007, Gilland et al. (2008), Godowitch et al. (2008), Hografe et al. (2006), Pierce et al. (2010). Dynamic evaluation is one of the four types of evaluations which should guarantee a comprehensive evaluation of the modelling system. Such an evaluation is crucial as it helps to specify the boundaries of applicability of a model and define the response to changes in input.

The model quality objectives (MQO) describe in the AQ Directive apply only to the assessment of the current air quality when reporting exceedance. There are no MQO for other applications in the AQD such as planning or forecasting. However, there is clearly an expectation when using models for these applications that they have been verified and validated in an appropriate, but unspecified, way. It is therefore important to have a specific assessment for planning applications (emission scenario analysis) because there is no guarantee that a model which performs well on a given base case (assessment) also performs well when used to perform emission scenarios. This is a clear conclusion from previous inter-comparison exercises (e.g. CityDelta, Eurodelta, POMI) which have been focusing on the analysis of model responses to emission scenarios. This may be particularly relevant to NO_2 chemistry modelling, since some of these chemistry schemes do not contain the appropriate process descriptions to reflect changes in emissions or ozone concentrations (Chapter 3).

Although the truth for scenario modelling is unknown some methodologies can be applied to increase confidence and robustness for this type of applications. As mentioned above one possible way is to perform inter-comparison exercises. These exercises deliver some estimate of model variability, and allow for the identification of outlying behaviours. Although an outlying performance does not necessarily mean poor performance, once

the causes for model divergences are understood, the inter-comparison exercise generally results in a reduction of the model variability and an increased robustness and confidence in model results.

An alternative is to assess model performance on emission data that has known differences in the emissions. These can be differences in weekday/weekend traffic types and volumes or it can be differences from year to year over longer periods, e.g. a decade. In essence the model trend over several years should be able to capture the observed trend, assuming the emission trends are well established. If this is not the case then the model will not be suitable for long term planning activities

7.4 The model operator and user

The level of expertise required to run a model and interpret its results varies according to the complexity of the application and the complexity of the model involved. It also depends on the allowable level of interaction between the model operator and the model. There are a range of software packages available that capsulate what may be fairly complex models into a limited user interface. The model user, who will provide both input data and output interpretation, can be a significant source of uncertainty in any model application. This is particularly the case when errors in the input data, or misinterpretations in the model input requirements, are allowed to propagate, unnoticed, to the interpretation/communication phase of modelling. The likelihood that such errors are picked up is higher for an experienced model user or developer than for a less experienced operator.

Quality assurance of model users is not a requirement in any quality assurance routine. However, many of the errors that can befall both experienced and inexperienced model users alike can be mitigated by the following:

- Always carry out a validation exercise for any new application or data set. If the model shows serious or suspicious disagreement with the observations then input data and model settings (if applicable) should be reassessed. If validation is not possible then a comparison with a similar application is recommended.
- In general the results should be presented to a second party, or wider group if possible. Often, during such presentations, problems may be noticed with the results.

In regard to model users, we make the following distinctions concerning levels of expertise in applying models. These three levels are indicative only as there is a continuous range of expertise. These levels also reflect those presented in Schluenzen and Sokhi (2008) in regard to model users and developers as well as those contained in the EEA Model Documentation System which provides a similar ranking of 'basic', 'intermediate' or 'advanced' expertise.

1. Model operator: This refers chiefly to people (e.g. consultants) who apply 'off the shelf' software packages (e.g. industrial stacks) or other user interfaced model software packages. Model operators will provide both input data and interpretation of the results and thus should have a basic understanding of the processes that the model represents. They are not expected to be able to change any of the model code.
2. Model user: This refers to people who have a more advanced level of understanding of the model and its processes. They should be able to change simple elements of the code, e.g. input and output, to accommodate different needs. They should be able to install, run and if necessary compile the model. They should be literate in programming languages.
3. Model developers: This refers chiefly to researchers who are actively involved in model development, be it the programming, numerical implementation or the development of the mathematical descriptions used in the models. They should have a good insight into the model, both numerically and physically and should be able to assess and improve models.

7.5 Documentation

For the most effective implementation of air quality models good documentation is required. The following elements should be provided and be accessible in any set of model documentation. This is true even if the model is not intended for general use. Good documentation is a pre-requisite for any model quality assurance.

1. Model description: This includes physical, mathematical and numerical descriptions of the model.

2. User manual: User manuals are essential for the correct implementation and interpretation of both the input and output data. This should also include limitations on the applicability of the model.
3. Model code and comments: When the model code is available (this is preferable) then the code should be readable and reflect the existing model description. Comments throughout the code are necessary as code may not always be intuitively interpretable.
4. Test datasets: At least one input dataset and the model results should be available for other users to aid in implementation of the model.
5. Validation and verification documentation: Documents(s) reflecting validation or verification studies using the model should be available.
6. Update information: If there are updates to the model by the model developer then these should be clearly documented and both new and old updates should be available.

Currently the EEA Model Documentation System (http://air-climate.eionet.europa.eu/databases/MDS/index_html) provides model developers and users with the possibility of providing 'descriptive' text and 'keywords' concerning the models, covering a number of topics. As such, the MDS provides a portal to models and their documentation rather than a detailed documentation of each model. We list these topics here to indicate the type of data contained in the MDS.

Table 17. List of the different model topics assigned in the EEA Model Documentation System

MDS descriptive topics	Sub-topics
Basic information	Model name Model version and status Latest date of revision Contact information Level of knowledge needed to operate model
Intended field of application	
Model type and dimension	
Model description summary	
Model limitations/approximations	
Resolution	Temporal resolution Horizontal resolution Vertical resolution
Schemes	Advection & Convection Turbulence Deposition Chemistry
Solution technique	
Input	Availability and Validation of Input data Emissions Meteorology Topography Initial condition Boundary conditions Data assimilation options
Output quantities	
User interface availability	
User community	
Previous applications	
Documentation status	Ranking levels 1 - 5
Validation and evaluation	Ranking levels 1 – 5 Model intercomparison
Frequently asked questions	

Portability and computer requirements	Portability CPU time Storage
Availability	
References about model development	
Other references	

Within the 'Documentation Status' topic a choice is provided for ranking the level of documentation, following the suggested levels in Moussiopoulos et al. (1996):

1. *Complete documentations available, ranging from the scientific description down to users manuals with details on the machine code.*
2. *Rather good scientific documentation and less complete user's manuals.*
3. *Worse scientific documentation as compared to "2"*
4. *Generally, incomplete or messy documentation.*
5. *No documentation at all.*

For models to be applied by the model developers a ranking of 2 or better is suitable, for models being used by other model users a ranking of 1 is necessary.

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Appendix 1: Short description of parameterised chemistry schemes

In Section 4.3 empirically based parameterised forms of NO_2 chemistry are discussed. Because a large number of urban and local scale air quality models are based on Gaussian dispersion models these forms of chemical parameterisations are frequently used. In this appendix we review a number of these methods listed in Table 11.

A1.1 Ambient Ratio Method (US EPA)

The simplest empirical relationship is the Ambient Ratio Method (ARM) suggested by the US EPA (Chew and Meyer, 1991) to convert modelled NO_x concentrations to NO_2 concentrations. This method simply involves calculating the locally observed NO_2: NO_x ratio and applying that to all the model values.

$$[NO_2] = a[NO_x]$$

This method is intended for use for annual mean concentrations. The US average ratio was determined to be *a* = 0.75.

A1.2 Methodologies developed for DEFRA (UK)

More complex empirically based model parameterisations have also been applied. The empirical model applied by Derwent and Middleton (1996) provides a polynomial fit to one stations hourly mean data. Further investigations of similar data in the UK by Dixon et al. (2000) showed the original fits to be very site specific with different coefficients necessary for different sites. Laxen and Wilson (2002) added more stations and years to the dataset and provided a simpler parameterisation for annual mean NO_2 based on NO_x measurements. The renewed version attempts to represent the proximity of the stations to NO_x emissions by separating the near road and urban background contributions as described by:

$$[NO_2(road)] = (-0.068\log([NO_x(total)]) + 0.53)[NO_x(road)] \text{ (units ppb)}$$

where

$$[NO_x(total)] = [NO_x(road)] + [NO_x(background)]$$

A1.3 Airviro (SE)

Airviro (www.smhi.se/airviro/) recommends applying the following parameterisation to convert from modelled NO_x concentrations NO_2 concentrations.

$$[NO_2] = 0.73[NO_x]\exp(-0.00452[NO_x] + 0.003014[NO_x]^2) \text{ (units } \mu g/m^3)$$

A1.4 Romberg method (DE)

The Romberg method (Romberg et al. 1996) has been used for several years in Germany within a number of models (e.g. PROKAS, IMMIS and MISKAM). It uses the following straight forward parameterisation:

$$[NO_2] = \frac{A[NO_x]}{[NO_x] + B} + C[NO_x] \qquad \text{(units } \mu g/m^3)$$

Where the constants A, B and C are regression parameters determined by fitting the available observational data.

The above formula was first proposed by Van den Hout en Baars (1988). At that time there was a physical meaning to the constants. The constant C is indicative of the NO_2:NO_X emission ratio, but not exactly the same since annual means are calculated, and the constant A represents the background ozone levels. The constant B was empirically determined, particularly for ensuring the correct slope to the curve for low NO_X concentrations. The above equation was recently assessed and fitted for German stations by Bächlin and Bösinger (2008). The

data and fits are shown in Figure 5. The same formulation, but different empirical constants, has been applied in Graz (Pongratz et al., 2010).

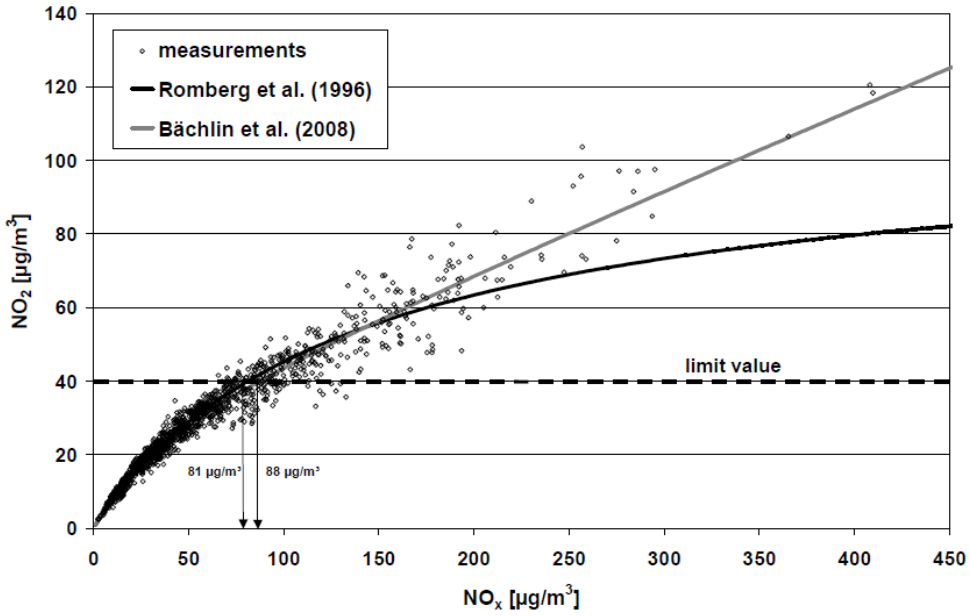

Figure A1.1. Empirical fits of the Romberg et al. (1996) parameterisation of NO_2 concentrations. Shown is the original Romberg fit (A=103, B=130, C=0.005) and the updated fit from Bächlin and Bösinger (2008), (A=29, B=35, C=0.217) made on the German station data shown (years 2004 – 2006). Source: Bächlin and Bösinger (2008).

A1.6 Standard Calculation Method in the Netherlands (NL)

In the Netherlands a simple but effective empirical relation is used for the calculation of NO_2 contributions due to traffic emissions. It was first proposed by Van den Hout en Baars (1988). Since then it has been refined, tested and modified and presently it is used in several Dutch legislated models where it is part of the "Standard Calculation Method (SCM)" applied in The Netherlands (http://wetten.overheid.nl/BWBR0022817/).

For yearly averaged calculations in an urban environment, converting a calculated average NO_x concentration contribution ($\overline{\Delta NO_x}$) into a NO_2 contribution ($\overline{\Delta NO_2}$), the relation is:

$$\overline{\Delta NO_2} = F \cdot \overline{\Delta NO_x} \quad + \beta \cdot \overline{O_3}^{\,a} \cdot \frac{(1-F) \cdot \overline{\Delta NO_x}}{(1-F) \cdot \overline{\Delta NO_x} + K}$$

with $\overline{O_3}$ the yearly average ambient ozone concentration, F the fraction of the NO_x that is emitted as NO_2 and the constants K=100 and β=0.6. Originally β was estimated from limited data, it was later shown to result from the non-linearity in the NO_x/NO_2 chemistry, combined with the correlation between hourly NO_x concentrations and ozone concentrations. In Dutch street canyons it varies between 0.6 and 0.9. Results from calculations with the Dutch standard method for the urban environment have been compared to OSPM calculations (Nguyen and Wesseling, 2009) as well as to measured concentrations (Wessling and Sauter, 2007), the agreement is considered satisfactory.

When yearly average concentrations around open roads are calculated the above relation is also used, but with β=1.0 and the concentration contributions being determined in 12 wind sectors and subsequently the appropriate weighted average of these is determined. In case hourly concentrations are calculated (urban streets and open roads) the above relation is also used with β=1.0, to convert calculated hourly average NO_X contributions into hourly average NO_2 contributions. Benchmarks have shown that, with the above scheme, simple Gaussian models produce yearly average NO_2 concentrations that are quite similar to those of more

advanced models, performing hourly calculations. Furthermore, (limited) available measurements show a satisfactory agreement between calculated and measured NO_X and NO_2 concentrations along roads (Figure A.2). Close to the road the calculated contributions overestimate the measured data by roughly 5% whereas at the larger distance the calculated contributions underestimate the measured data by roughly 15%. The optimal value of K (normal value 100) was found to depend somewhat with the distance to the road.

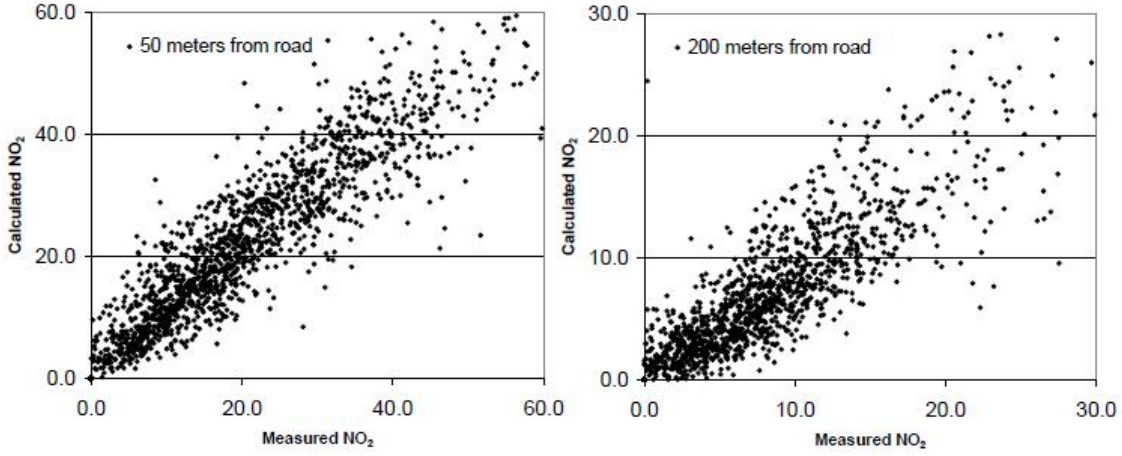

Figure A1.2. Measured and calculated hourly NO_2 concentration contributions along a highway (Wessling, 2004).

A1.7 SAPPHO (NL)

In the Fifth national environmental report from RIVM (Erens and van Dam, 2000) an algorithm (SAPPHO) for calculating NO_2 from NO_x is presented that follows the same form as the steady state equation given in Equation 4.8 but where the factor J' and $[O_x]$ are tuned to 8 years of annual mean measurements in The Netherlands and given as follows.

$$J' = 0.27[NO_x] + 4.5 \qquad \text{(units ppb)}$$

$$[O_x] = 1.3\sqrt{[NO_x]} + 27.4$$

The results of the model fit are shown in Figure A1.2.

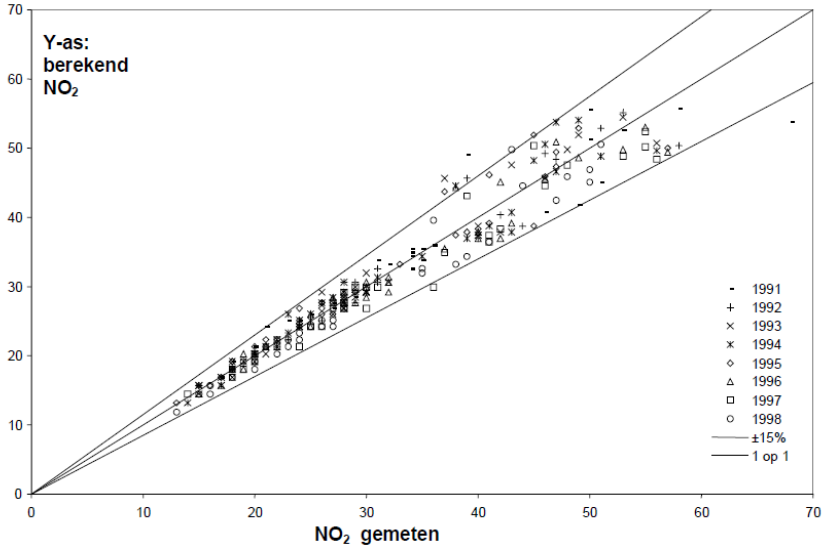

Figure A1.2 Scatter plot of measured (gemeten) NO_2 concentrations verses calculated (berekend) NO_2 concentrations using SAPPHO applied to annual mean data from a number of Dutch sites for the years 1991-1991 (N=280). Figure taken from Eerens and van Dam (2000).

A1.8 Keller (CH)

In the same report (Erens and van Dam, 2000), a Swiss empirical model for converting NO_x to NO_2 (Keller et al., 1997) was also applied and found to perform poorly in the Netherlands. Though its performance in Switzerland was considered satisfactory.

$$[NO_2] = 0.055[NO_x] + 55\left(1 - \exp\left(-(0.7 - 0.055)[NO_x]/55\right)\right) \text{ (units µg/m}^3\text{)}$$

A1.9 Oxidant Partitioning Model (UK)

For assessment of NO_2 concentrations in the UK (Murrells et al., 2008), maps of NO_2 concentrations are derived from the NO_X concentrations maps using the oxidant-partitioning model from Jenkin (2004). This model describes the inter-relationships between NO, NO_2 and ozone as a set of chemically coupled species, giving some physical basis to the approach but using site specific empirical relationships. Empirical fits (using up to fourth order polynomials) of the NO_2:O_X ratio are used. Different fits are used for 'near' and 'far' from source contributions. NO_2 is then calculated for model applications using the following form

$$[NO_2] = (A[NO_x] + B).f(NO_X) \qquad \text{(units ppb)}$$

Where *A* is an empirical and site specific (inside or outside London) parameter representing the local oxidant contribution, *B* is the regional oxidant [O_X] concentration and the function *f(NO$_X$)* is the empirically fitted NO_2:O_X ratio. The method is applied to annual mean concentrations. The inclusion of O_X in the functional dependency makes this method more suitable than other NO_X based methods for describing changes in primary emissions and ozone background concentrations.

A1.10 Limited mixing steady state approach applied to annual averages (DE)

In Germany a simplified chemistry model based on the annual average values is used for the calculation of NO_2 contributions due to traffic emissions. It was proposed by Düring et al. (2011). This is based on the methodology applied in OSPM, Berkowicz et al. (2011) but applied to annual average concentrations. Assuming that the equilibrium of NO, NO_2 and O_3 is quickly reached, the three differential equations turn into three algebraic conditional equations. Solving them yields the analytical equation for the concentration:

$$[\mathrm{NO_2}] = 0.5\left(\mathrm{B} - \sqrt{\mathrm{B}^2 - 4\left([\mathrm{NO_x}]\,[\mathrm{NO_2}]_\mathrm{O} + [\mathrm{NO_2}]_\mathrm{n}/\mathrm{k}\tau\right)}\right)$$

With the variables

$$[\mathrm{NO_2}]_\mathrm{n} = [\mathrm{NO_2}]_\mathrm{V} + [\mathrm{NO_2}]_\mathrm{B}$$

$$[\mathrm{NO_2}]_\mathrm{O} = [\mathrm{NO_2}]_\mathrm{n} + [\mathrm{O_3}]_\mathrm{B}$$

$$\mathrm{B} = [\mathrm{NO_x}] + [\mathrm{NO_2}]_\mathrm{O} + \frac{1}{\mathrm{k}}\left(\mathrm{J} + \frac{1}{\tau}\right)$$

$[NO_x]_V$ and $[NO]_V$ are calculated from the difference in NO_x between the traffic station and the background station as

$$[\mathrm{NO_2}]_\mathrm{V} = \mathrm{p}\left([\mathrm{NO_x}] - [\mathrm{NO_x}]_\mathrm{B}\right)$$

$$[\mathrm{NO}]_\mathrm{V} = \left([\mathrm{NO}] - [\mathrm{NO}]_\mathrm{B}\right)$$

with p = NO_2/NO_x being the fraction of NO_2 in the direct traffic emissions and $[NO_x]$ as the concentration, which is estimated at the traffic station by measurements or a dispersion model.

Strictly speaking, the above equations of the chemistry model can only be used in time series calculations, because the parameters J and k are dependent on meteorological parameters. Based on research projects these equations can also be applied for annual mean concentrations using the following parameters:

J = 0.0045 s^{-1}

K = 0.00039 $(ppb\ s)^{-1}$

τ = 100 s (street canyons) or 40 s (free dispersion)

Appendix 2: Short description of manuals and tools for calculating emission inventories

In Section 5.1 a list of information, tools, data and other documents relevant to the establishment of emission inventories is provided in Table 13. Some of these are provided in more detail here.

A.2.1 EMEP/EEA Emission Inventory Guidebook and reporting guidelines

According to the Convention on Long Range Transboundary Air Pollution (LRTAP), member states are required to report annual national emissions data of main atmospheric pollutants (SO_2, NO_x, NMVOC, CH_4, CO, NH_3 and various heavy metals and POPs) using the NFR nomenclature. The EMEP reporting guidelines (ECE/EB.AIR/2008/4) are setting down the rules for reporting under the LRTAP. The EMEP reporting guidelines as well as the EU directive on national emission ceilings (2001/81/EC) determine the use of the EMEP/EEA Emission Inventory Guidebook (GB) which determines the principles, methodologies and default calculation values (e.g. emission factors) to be used. Previous versions of the GB used the CORINAIR methodology based on the SNAP nomenclature which has since been replaced by the NFR nomenclature. However, in the current GB a link to the SNAP nomenclature is still provided. The GB is structured by the 115 NFR categories as specified in Annex III of the EMEP reporting guidelines. The NFR categories are merged into 20 GNFR categories which have to be used for reporting gridded data. The GB also determines "good practice" principles to ensure the transparency, consistency, comparability, completeness, and accuracy of national air pollutant inventories. The GB provides different levels of "tier" methods which may be used for emission estimates. Higher tier methods need more specific input data but provide a higher accuracy of the estimation results. An important underlying principle is the "key source analysis", which means that emission sources contributing most to the national total emissions or to the emission trends need higher tier methods in order to keep overall uncertainty as low as possible. The GB additionally provides guidance for preparing emission projections and some basic methods for natural emissions.

A set of public tools for preparing emissions inventories (CollectER software) is available at http://www.air.sk/en/reporter.php. The CollectER software may be used for data collection of large point sources and area sources as well as for disaggregating data at e.g. NUTS2 level. Most countries use similar principles but developed their own software, ranging from spreadsheets solutions to multiuser databases which have also integrated reporting of green house gases.

A2.2 COPERT

COPERT is an MS Windows software programme for calculating air pollutant emissions from road transport. The technical development of COPERT has been funded by the European Environment Agency (EEA). Since 2007, the European Commission's Joint Research Centre has undertaken the further scientific development of the model. COPERT was initially intended to be used by national experts to estimate road transport emissions to be included in official annual national inventories, but it is also available at no cost for use in other research, scientific and academic applications. The methodology for the calculation of exhaust emissions and fuel evaporation in COPERT is based on the relevant methodology in the dedicated chapters in the EMEP/CORINAIR Atmospheric Emission inventory Guidebook. The use of a common software tool to calculate road transport emissions is encouraged by EU legislation, as it facilitates a transparent and standardised procedure for data collecting and emissions reporting, that allows for direct comparison of emissions between member states.

The latest version, COPERT 4, estimates emissions of all major air pollutants (CO, NO_x, VOC, PM, NH_3, SO_2, heavy metals) produced by road transport (passenger cars, light duty vehicles, heavy duty vehicles, mopeds and motorcycles) as well as greenhouse gas emissions. The programme also provides speciation for NO/NO_2. The previous version, COPERT III, contains a separate module that estimates exhaust emissions from internal combustion engines used in off-road applications (agriculture, forestry, household, industry, waterways and railways).

The calculated emissions are classified in three sources:

- Hot emissions: produced during thermally stabilised engine operation
- Cold-start and warming-up emissions: occurring during engine start from ambient temperature, and
- NMVOC emissions produced due to fuel evaporation

The total emissions are calculated as the product of activity data provided by the user and speed dependent emission factors calculated by the software.

The COPERT 4 software system can be applied for the compilation of annual national inventories, however it can also be used with a sufficient degree of certainty for the compilation of urban emission inventories requiring a higher spatial (1 km×1 km) and temporal resolution (1 hour).

More information on COPERT methodology and downloads of the COPERT modules can be found at: http://lat.eng.auth.gr/copert/

A2.3 TREMOD: Transport Emission Model

The transport emission model "TREMOD" was developed by the IFEU-Institute on behalf of the Federal Environmental Agency in Germany to calculate national emissions of all transport modes (road traffic, railroad traffic, water and air traffic) between 1960 and 2030. The TREMOD Emission Inventory is being employed for air quality management and reporting purposes by relevant authoritative bodies and other organisations, particularly the automotive industry association and the association of mineral oil companies.

TREMOD version 4.0 includes direct energy consumption and tailpipe emissions for the entire Germany, as well as the respective shares of energy expenditure and emissions from energy provision, from the point of primary energy carrier production onwards. Currently the emissions of the main air pollutants (hydrocarbons, NO_2, CO, SO_2, NH_3, N_2O, dust and diesel particulates) are calculated by TREMOD. Emission rates are calculated on the basis of the following data:

- the number of vehicles in road traffic
- the mileage driven
- the performance of passenger (person/km) and goods (ton/km) transportation

The emission factors for road traffic have been prepared for the Handbook Emission Factors of Road Traffic (HBEFA) 2.1 /INFRAS 2004a/ which is the basis for TREMOD. The most important change has been the update of emission factors for diesel passenger cars and heavy-duty vehicles up to stage Euro 3, resulting to higher calculated NOx emissions compared to the previous version.

More information on the methodology used in TREMOD and downloads are available at: http://www.ifeu.de/english/index.php?bereich=ver&seite=projekt_tremod

A2.4 EMIT: Emission Toolkit

EMIT is an Emissions Inventory Tool developed from CERC (http://www.cerc.co.uk/environmental-software/EMIT-tool.html) which can be used to store, manipulate and assess emissions data from the full range of source types. It is applied in the United Kingdom. The sources fall into two categories. Firstly there are the explicit sources such as major roads, rail and industrial sources. Secondly, EMIT can hold data from sources that may be too small to be considered explicitly, and instead are treated as average emissions on a 1 km^2 grid, such as minor roads and commercial and domestic sources.

Emissions can be calculated either by scaling a national emissions figure by a local statistic or from activity data, such as traffic flows or fuel consumption. Emissions factors are included from a number of databases including the UK Emission factor Database (UK EFD), UK Greenhouse Gas Emission Inventory (UK GHG), ICAO database, IPCC inventories.

A2.5 TREFIC

TREFIC ("TRaffic Emission Factor Improved Calculation") is a software program, provided with a simple GUI, that has been developed to estimate atmospheric pollutant emissions from road sources in terms of pollutant mass per trip unit. The software is primarily applied in Italy but has also been used European wide (for regional

modelling) and has been explicitly applied in Australia, Brazil, France, India, Israel, Morocco, Qatar, Romania, Russia and Tunisia for local emission inventories. A brief overview can be found at http://www.aria-net.it/front/ENG/codes/files/7.pdf and example applications have been presented by Nanni et al. (2004, 2011).

The calculation of road vehicles emission factors (EF), is based on COPERT 4 methodology according to vehicle types, fuel consumption, average travelling speed and road types. This methodology, implemented in a computing module which is the core of TREFIC software, is the base of a system built in order to:

- pre-process the input to make it compliant with CORINAIR specifications;
- determine, from the EF, total emissions for each link of the road network;
- produce synthetic and GIS output of these emissions;
- produce input files for dispersion models (at present SPRAY and ARIA ImpactTM models are directly covered).

Input data consist of 4 groups of files, related with:

- road network: for each link of the network, provide information about length, traffic flows, etc.;
- vehicle fleet: for each of three road types (urban, rural, highway), split in COPERT 4 categories;
- time modulations: tables that help to quantify the time profiles of emissions factors (for traffic flows, for average traveling speeds and for temperature);
- emission factors: .mdb tables implementing COPERT 4 methodology.

A2.6 GAINS

The Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model, developed by IIASA APD Programme, is an integrated assessment model dealing with costs and potentials for air pollution control and greenhouse gas (GHG) mitigation, and assessing interactions between policies. In 'scenario analysis' mode, GAINS can be employed to generate consistent emission projections and cost implications of alternative emission control scenarios for air pollutants (SO2, NOX, VOC, PM, NH3) and greenhouse gases, through projections of underlying activity data, collections of emission factors and lists of control measures and their costs. These projections can be either used on-line in GAINS to estimate impacts on environment and human health, or to feed off-line air quality models. Documentation can be found at http://gains.iiasa.ac.at/index.php/documentation-of-model-methodology/supporting-documentation-europe

Implementations exist for various world regions and countries. The European version of GAINS and its predecessor, the RAINS model, have been applied to assist key policy negotiations on improving air quality in Europe, under the UNECE Convention on Long-range Transboundary Air Pollution and for the European Union. The Italian version (D'Elia *et al.*, 2009), managed by ENEA (National agency for new technologies, Energy and sustainable economic development), allows to generate national and sub-national emission scenarios up to 2030, taking consistently into account the expected of activities scenarios and European, national and regional legislation and air quality plans. Emission projections for administrative regions, combined with the finer spatial distribution provided by the reference national emission inventories, have been used to feed national simulations of MINNI atmospheric modelling system (Zanini *et al.*, 2010) used to assess the impact of future emission reductions. This approach has been also used to prepare the accompanying documentation supporting the notification of a postponement of the deadline for attaining the limit values for NO_2 under Directive 2008/50/EC.

A2.7 HERMES: High-Elective Resolution Modelling Emission System

The HERMES emission model is capable of estimating the emissions for Spain with a spatial resolution (1 km x 1 km) and temporal resolution (1h) for chemistry transport modelling applications. Furthermore, HERMES constitutes the emission core of the operational air quality forecasting system for Spain developed under the CALIOPE project funded by the Spanish Ministry of the Environment (http://www.bsc.es/caliope), whose operational domain corresponds to Europe, the Iberian Peninsula-Balearic Islands, Canary Islands and Barcelona Great Area with spatial resolution of 12 km, 4 km, 2 km and 1 km, respectively.

Some of the characteristics of HERMES model are summarized as follows:

1. Use of emission estimation methodologies compiling the state-of-the-art in different sectors and activities. HERMES generates results according to the European Environmental Agency's Selected Nomenclature for Air Pollution (SNAP).
2. Definition of the emission spatial patterns coming from primary gaseous (NO_x, NMVOC, CO, SO_2) and particulate pollutants (TSP, PM_{10}, $PM_{2.5}$) together with greenhouse gases emissions (CO_2, CH_4, N_2O).
3. Selective chemical speciation of emissions. The chemical mechanisms currently included in HERMES are the CB-IV (Gery et al., 1989), CB05 (Yarwood et al., 2005a) and MELCHIOR2 (Derognat et al., 2003).
4. Capability of generation of graphic and alphanumeric information with high spatial and temporal resolution (1 km x 1 km, 1h). (http://www.bsc.es/caliope/?q=node/27).
5. Development of the model following a quality protocol that guarantees the reliability of the results.
6. Implementation of the emission methodologies for their easy revision and actualization.
7. Combination of emissions from different sources to perform sensibility analysis or to study the sectorial contribution of each source to the emissions.
8. It includes a specific module to calculate the emissions resuspended particle matter from paved roads.

More information on HERMES model and results can be found in Baldasano et al., (2008) and Pay et al., (2010). Results and discussion of the HERMES application in the CALIOPE system evaluation can be found in Baldasano et al., (2011).