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Cross-Cutting Issue 1: Emissions & Data Needs

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Glossary

ARTEMIS	Assessment and Reliability of Transport Emission Models and Inventory Systems, EC project
BAT	Best available techniques
BREF	Best Available Techniques Reference Document
CEPMEIP	Co-ordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance
CLRTAP	UN-ECE Convention on Long Range Transboundary Air Pollutants
COPERT	Computer Programme to calculate Emissions from Road Transport
CORINAIR	Core Inventory of Air Emissions, European emission inventory of the European Topic Centre on Air Emissions (ETC/AEM)
CORINE	Coordination of Information on the Environment, EU Programme
CRT	Continuously Regenerating Trap
CTM	Chemistry Transport Model
DMS	Dimethylsulfide
ECE cycle	Economic Commission of Europe, Urban Driving Cycle
EMEP	Co-operative Programme for the Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe
EPA	United States Environmental Protection Agency
EPER	European Pollutant Emission Register
EUDC	Extra Urban Driving Cycle
EUROTRAC	EUREKA Project on the Transport and Chemical Transformation of Environmentally Relevant Trace Constituents in the Troposphere over Europe; Second Phase
GENEMIS	Generation and Evaluation of Emission Data, EUROTRAC-2 subproject
GIS	Geographic information system
GHG	Green house gases
HC	Hydrocarbons (sometimes referred to as "VOC")
HBEFA	Handbook Emission Factors for Road Transport
HDV	Heavy duty vehicles
IIASA	International Institute for Applied Systems Analysis
IER	Institute of Energy Economics and the Rational Use of Energy, University of Stuttgart
IPCC	Intergovernmental Panel on Climate Change
LDV	Light duty vehicles
LOTOS	Long Term Ozone Simulation, Eulerian grid model of TNO
LPS	Large point sources reported to CORINAIR
LSD	Low sulphur diesel
MEET	Methodologies to Estimate Emissions from Transport, EC project
MERLIN	Multi-pollutant, Multi-Effect Assessment of European Air Pollution Control Strategies: an Integrated Approach, EC project
NAEI	UK National Atmospheric Emissions Inventory

NATAIR	Improving and Applying Methods for the Calculation of the Natural and Biogenic Emissions and Assessment of Impacts on Air Quality, EC project
NEDC	New European Driving Cycle
NFR	UN-ECE Nomenclature for Reporting
NGV	Natural gas vehicle
NUTS	Nomenclature of Units for Territorial Statistics
OSCAR	Optimised Expert System for Conducting Environmental Assessment of Urban Road Traffic, EC project
PARTICULATES	Characterisation of Exhaust Particulate Emissions from Road Vehicles, EC project
PC	Passenger cars
PRTR	Pollutant Release and Transfer Register
PTW	Powered two-wheelers
RAINS	Regional Acidification Information and Simulation model
SATURN	Studying Atmospheric Pollution in Urban Areas, EUROTRAC-2 subproject
SCENES	European Transport Forecasting model
SCR	Selective catalytic reduction system
SCRT	Combination of SCR and CRT systems
SNAP	Selected Nomenclature for Air Pollution
SUTRA	Sustainable Urban Transportation for the City of Tomorrow, EC project
TFEIP	United Nations Economic Commission for Europe (UNECE) - Task Force on Emission Inventory and Projection
TNO	Institute of Environmental Sciences, Energy and Process Innovation
TREM	Transport Emission Model for Line Sources
TREMOD	German Transport Emission Estimation Model operated by UBA
TREMOVE	Combination of TRE (TRENEN) and MOVE (FOREMOVE)
TRENDS	Transport and Environment Database System
UBA	German Federal Environmental Agency
ULSD	Ultra-low-sulphur diesel
ULP	Unleaded petrol
USGS	United States Geological Survey
VOC	Volatile organic compounds

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Executive Summary

An emission inventory which is able to provide accurate emissions with a high resolution in space, time and substances is a prerequisite for the operation of dispersion and chemistry transport models (CTMs). The generation of emission data firstly requires the development of a detailed source inventory that leads to an identification of major sources. Furthermore the generation of emission maps based on a source inventory requires a geographic source and emission allocation and the analysis of temporal emission variations.

Within this report, methodologies of emission data generation are reviewed and recommendations are derived how to assess and reduce uncertainties. The report focuses on selected issues that are relevant for the Air4EU scope and priorities. Recommendations made are partly of a general sense and mostly deal with data availability, data assignment and treatment and possible level of detail. Recommendations for the usage of specific data such as activity rates or literature sources for emission factors are in most cases not reasonable as there is always an individual data choice depending on a review of the current state of knowledge and on national methodologies and data bases.

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1. Introduction

Emission data are an important input to air quality assessment and the development of abatement strategies. They provide comprehensive information on emission sources and emission fluxes in the area under consideration. In this context, an emission inventory which is able to provide accurate emissions with a high resolution in space, time and substances is a prerequisite for the operation of dispersion and chemistry transport models (CTMs).

Spatial resolution of emission modelling depends on scale of air quality assessment. Methodologies, data need and key sources to be considered depend on pollutant and on scale as well. On regional/continental scale, point, area and line sources have a comparable significance and biogenic/natural emissions should be included as they may contribute significantly to the concentrations in non-urban areas. For urban/agglomerate scale, anthropogenic sources such as road traffic, industrial activities and - in middle and northern Europe - residential combustion are major emission sources. Exhaust and non-exhaust emissions from road traffic and emissions from significant industrial point sources are of special importance for air quality on local/hotspot scale.

Within this Milestone report, methodologies of emission data generation are reviewed for each scale of air quality assessment. The report focuses on selected issues that are relevant for the Air4EU scope and priorities. Examples of emission inventories are described and recommendations for emission modelling derived. These recommendations are partly of a general sense and deal with accuracy and data quality. Recommendations for a certain scale mostly deal with data availability, data assignment and level of detail that should be reached in order to minimize uncertainties of emission modelling. Recommendations for the usage of specific data such as activity parameters or literature sources for emission factors are in most cases not reasonable as there is always an individual data choice. This expert judgement should be based on a detailed analysis of source specifications/technologies and an extensive review of the current state of knowledge on source specific emissions.

A significant amount of work on the generation and evaluation of emission data and the composition of emission inventories has been conducted in the frame of the EUROTRAC 2 subproject GENEMIS (<http://genemis.ier.uni-stuttgart.de>), with final results having been published in (Friedrich & Reis 2004).

2. General aspects of emission inventories

In the following some general requirements are given for the generation of emission inventories. It is based on the work done for the GENEMIS final report (Friedrich & Reis 2004). The following information in this chapter with regard to general data structure and requirements is largely based on (Sturm & Winiwarter 2004). They provide comprehensive information about methods for compiling an urban emission inventory in a harmonised way. Requirements on emission inventories concern the pollutants and emission sources covered and the temporal, spatial and substance resolution of annual sectoral emission data. Different methodologies can be applied to establish an emission inventory. While an inquiry for single sources – based on in site information on emissions and activity data – is defined as bottom-up methodology, a use of statistic data results in a top-down estimation on e.g. national scale. The big effort for bottom-up methodologies and the inaccuracy of top-down approaches are limitations for both methods. Therefore a combination of both methods is normally used for national or urban emission estimates.

2.1. Pollutants

Council Directive 96/62/EC of 27th September 1996 on ambient air quality assessment and management provides in Annex I a list with atmospheric pollutants that are to be taken into consideration in the assessment and management of ambient air quality. Pollutants to be studied at an initial stage, including pollutants governed by existing ambient air quality directives are:

1. Sulphur dioxide (SO₂)
2. Nitrogen dioxide (NO₂)
3. Fine particulate matter (PM₁₀, PM_{2.5}), and precursors SO₂, NO₂, Ammonia (NH₃)
4. Suspended particulate matter (TSP)
5. Lead (Pb)
6. Ozone (O₃) and precursors nitrogen oxides (NO_x) and volatile organic compounds (VOC)

Other air pollutants to be considered are

7. Benzene
8. Carbon monoxide (CO)
9. Polycyclic-aromatic hydrocarbons (PAH)
10. Cadmium (Cd)
11. Arsenic (As)
12. Nickel (Ni)
13. Mercury (Hg)

Since "assessment" is described as any method used to measure, calculate, predict or estimate the level of a pollutant in the ambient air, it is important that emission inventories include at least the pollutants that are referred to in the first part of the list. With regard to health effects of urban air quality, benzene and CO are relevant as well. This first group of air pollutants has thus the highest priority within air quality assessment and air quality modelling. In addition, the quality and availability of emission data for these pollutants is much better than for the rest (except CO₂). Pollutants with regard to regional or global impact (climate

forcing) that are not included in the list are CO₂, CH₄, N₂O and others. These pollutants might not be considered for urban chemistry.

2.2. Emission sources

2.2.1. Anthropogenic sources

Emission inventories usually classify emissions into three source types, namely point, area and line sources. These source types and their respective source groups can be defined as follows.

Point sources: emission data are provided on an individual plant or emission outlet (usually large) usually in conjunction with data on location, capacity, operating conditions etc.

Source groups: energy and transformation industry; production and processing of metals; mineral industry; chemical industry; processes in wood, paper pulp, food, drink and other industry; waste incineration.

Area sources: smaller stationary or fugitive sources are provided on an area basis.

Source groups: small combustion plants in households, agriculture, enterprises and institutions; material handling and storage; livestock and other processes in agriculture; solvent and other product use; other mobile sources and machinery (except road transport); waste treatment and disposal; small industrial plants; extraction and distribution of fossil fuels.

Line sources: vehicle emissions from road transport, railways and inland navigation can be provided for sections along the road, railway-track or waterway.

Source groups: road transport on highways, rural and urban roads (hot emissions, cold starts, gasoline evaporation, tyre and brake abrasion, suspension of road dust); other transport.

Emissions from aviation below 1 000 m height (landing, taxiing and take-off) are usually modelled as point or area source emissions. Cruise flights above 1 000 m can be modelled as line sources but are mostly not included in emission inventories because emissions are released above the mixing height of the atmospheric boundary layer.

2.2.2. Biogenic and natural sources

Biogenic/natural emissions should be taken into account especially on regional scale because they can contribute significantly to ambient emissions. Biogenic emissions (in principle related to living material) are especially relevant to model O₃, and to a lesser extent to model NO₂. For O₃, biogenic NMVOC emissions from forests are the major source group. For NO₂, biogenic NO-emissions from soils are important. Although on European average, biogenic NO emissions account for only 4 % of anthropogenic NO emissions, they can be relevant in rural areas. Natural emissions (in principle related to non-living material, sometimes also called geogenic emissions) are especially important for PM. Important sources are sea-salt and windblown dust, including Sahara-dust.

Methods for the quantification of biogenic/natural emissions on regional/EU scale still have high uncertainties. Within the UN-ECE Task Force on Emission Inventory and Projection (TFEIP), the Nature Expert Panel has compiled guidelines for the assessment of natural emissions in Europe (Simpson & Winiwarter 1998 resp. EEA 2003). Source categories dealt with are emissions from forests and forest fires, emissions from soil, grassland emissions, emissions from wetlands and waters, from wild animals, volcanoes, natural gas seeps and from lightning.

2.3. Differentiation of emission data

The use of emission data as input for air quality assessment requires a spatial, temporal and substance resolution. These differentiations allow to combine emission data via a chemistry transport model (CTM) with meteorological information and the modelling of chemical formation and transformation mechanisms. The comparison of resulting modelled and monitored concentrations might lead to a validation and improvement of emission inventories.

2.3.1. Spatial resolution

An accurate spatial resolution resp. spatial allocation of emissions is required in order to simulate emission situations in areas under investigation in an appropriate way and in order to generate spatial emission patterns as input for the mapping of air quality using CTMs. The smaller the region of interest is the finer the spatial resolution of the emission inventory has to be. For micro-scale models it is required to represent the location and emission behaviour as accurate as possible. Therefore a bottom up approach should be applied and a disaggregation of statistical data may not be sufficient. An accurate spatial allocation of emissions requires distinguishing between point, area and line sources. Methodologies to generate spatial emission data are usually based on statistical data for administrative units as well as digitised maps of land use data, roads and point sources that are intersected and linked to annual sectoral emission data using a geographic information system (GIS).

In order to ensure usability of gridded emission information a reference for the origin of the grid and a conversion to the national or an international grid or coordinate system/projection has to be given (e.g. Latitude/Longitude, Lambert conformal conic projection, Gauss-Krueger projection).

2.3.2. Temporal resolution

The distinction between high and low time resolution of emission data is important. For micro-scale or local scale estimation as well as for the simulation of air pollution episodes, high time resolution emission inventories are needed. To estimate the background concentration of primary pollutants, a lower temporal resolution is sufficient. To this aim, the distinction between continuous and discontinuous sources, taking into account seasonal variations, can be appropriate. In principle the time variation of the emission quantity follows the time dependency of the activity pattern. The standard for urban air modelling is a time resolution of one hour. This time resolution is necessary for activity parameters like traffic density, as well as for ambient parameters, like temperature. It should be noticed that time dependent activity

patterns are a function of local time (day light saving time), while ambient parameters like temperature are often given for standard time (UTC).

2.3.3. Substantial disaggregation

A speciation of NMVOC emissions is required by CTMs for the modelling of ozone formation as well as for air quality assessment (toxicity, direct health effects). Different mechanisms and corresponding substance resolutions are used within existing CTMs. In the case of PM, the investigation of source specific particle size distributions or size fractions like PM₁₀ and PM_{2.5} is important because atmospheric properties and potentially health effects vary with particle size. Also the chemical composition of particulate matter affects atmospheric processes and chemistry and may influence health effects. Although mainly consisting of NO, nitrogen oxide emissions are given in emission inventories in terms of the equivalent NO₂. For the application of photo-chemical models it is necessary to specify the fractions of NO and NO₂ for each individual emission source.

2.4. General approaches for emission calculations

The following methodology/data structure was defined by (Sturm & Winiwarter 2004) as harmonized method for the compilation of urban emission inventories for urban air modelling. Emission calculations can be done either with a bottom-up or a top down approach or a combined methodology according to data availability and project capacities.

2.4.1. Bottom-up approach

The bottom-up methodology is based on a source oriented inquiry of all activity and emission data needed for describing the emission behaviour of a single source. Due to the fact that this is connected to a big effort in data collection, real bottom-up approaches are limited to certain emission sources and types. Although the following requirements and recommendations are listed under bottom-up methodology, they include also information for top-down approaches.

The data of an emission inventory can be categorized as 'static lookup data', 'lookup data' and 'dynamic input data'. 'Static lookup data' are data such as units, unit conversion factors, compounds, etc. These data are normally entered into the database before installation. 'Lookup data' include emission factors, building registers, road link definitions, source complex and company registers, administrative district boundaries, etc. Although the 'lookup data' may change over time, they are considered to change less frequently than the 'dynamic input data'. 'Dynamic input data' are data that need to be updated regularly as they have only a limited validity period. This group includes all dynamic data that are not considered to be lookup data, such as e.g. area distributed consumption data, population data, point source emission/consumption/production data, time variations, etc.

Point source data

The data describing point source emissions can be divided into several groups. Some of the data describe the stacks through which the emissions are released to the atmosphere. Other data describes any cleaning devices that may be installed to reduce the emissions, and yet other data describe the activity that generate the emission, in the form of emission, consumption or production data and operating schedule for a given process.

- **Stack data:** The information of stack data contains standard parameters such as coordinates, identification numbers, name, stack height, diameter, gas flow rate, gas flow temperature, altitude and emissions.
- **Cleaning device data:** The following data may be given for each cleaning device, and for each component of which the emission is reduced by the cleaning device: identification number, physical links between units, cleaning efficiency factor etc.
- **Process data (activity):** Industrial activities can be divided into different processes. For each process, data can be collected on consumption, production, operation or directly on emissions. The parameters to be collected for the three alternatives are similar, such as identification numbers of the process. For energy consumption and production, fuel type or product type, respectively, must be defined.
- **Measured emission data:** In addition to or instead of consumption or production data, one may have measured data for the process emissions. These data may be in the form of measured emission, or as measured flue gas concentration and flow rate.

Area source data

Area source data may include emission data and energy consumption data distributed in administrative sub districts or in a grid. Necessary grids or administrative district level must be defined before import of these data. This includes the geographical boundary of each district.

Population or employment data may be defined as spatially distributed on administrative districts in the same way as area source data. Population data is normally used for distribution of data which is related to human activities. Land use data is usually used to further allocate emissions spatially.

Line source data

For the definition of line source data, a number of 'lookup data' concerning traffic data and the modelling of traffic induced emissions must be defined.

- **Road classes:** Road classes are used to categorize road sections. A number of traffic parameters may be assigned default values according to the road class to which the individual road belongs. Typical road classification can be: major roads / transit roads, city centre streets, residential area streets, industrial area streets and secondary streets.
- **Vehicle classes:** Before importing data for traffic volumes, it is necessary to define the relevant vehicle classes. Such classes may be: personal cars, buses, two-stroke engine motorcycles, trucks etc. The division is such that it is natural to assign emission

factors to each vehicle class separately, as well as it is possible to obtain traffic counting of such vehicles on the various roads.

- Road link definition: The road network consists of several road sections which are called road links. These must be described with identification number, co-ordinates and name. In addition, physical description such as number of lanes, width in each direction, road class type, must be entered.
- Dynamic traffic data: For each road link or classes of road links, detailed information on the traffic flow must be collected. Examples of such data are: annual daily traffic, free flow speed, cold start ratio and vehicle class distribution. Most of the parameters above must be given for each lane and direction.

2.4.2. Top-down approach

On continental or national scale so called top-down emission inventories are usually used for describing the emission situation based on national statistics. E.g. on a Europe-wide basis, EMEP provides annual emission information (EMEP 2004) reported by the member countries. Information on emissions is very often being derived on the scale of an administrative unit. For this administrative unit the respective statistical and socio-economical data is available. Within international inventories, administrative units used are countries. Starting from national figures, a top-down approach allows attributing the emissions to smaller units (for example, national data can be broken down to the level of NUTS 3 regions, which have the size of a British county, a French département or a German Kreis). Parameters used for such a disaggregation are listed in Table 2-1, based on (Orthofer and Winiwarter 1998).

Table 2-1: Parameters for the disaggregation to smaller administrative units

SOURCE CATEGORY	DISAGGREGATION PARAMETER
Industrial fuel use	industrial employment; industrial statistics: nominal production, energy consumption, fuel consumption, etc.
Industrial processes	industrial employment by branch; industrial statistics: nominal capacity of production, raw material consumption
Domestic combustion	population or heating equipment and specific heat demand, domestic fuel consumption or sales
Domestic solvent use	population
Regional road traffic	car registrations, population, fuel consumption or sales
Nature and agriculture	statistics: total area or specific for forest uses of the administrative units

The top-down approach is characterized by the fact that the emissions are derived for the largest unit, with uncertainty increasing the further down emissions are broken. Note that, despite increasing uncertainty, specific emission attribution within an area still increases the overall accuracy for that area with respect to using just averages. Nevertheless such approaches lack of including local data, as they use typical country-wide behavioural patterns which may not be reflected by an urban area to be considered specifically.

With respect to bottom-up emission assessments, a trade-off between effort spent on assessing single sources and accuracy of specific information has to be performed in all cases. Notably, this trade-off between top-down and bottom-up approaches is quite different for application to different scales. From the previous, it becomes clear that the widely available national CORINAIR figures might not be useful for urban inventories. Even the emissions on NUTS 3 level are usually derived from the national totals by disaggregation and therefore do not carry much additional information.

2.4.3. Combined method

Exclusive use of "bottom-up" methods fails due to lack in available input data, while exclusive use of "top-down" methods will only lead to an undesirable level of accuracy for e.g. urban emission inventories. For this reason, both methodical approaches are always combined. In order to achieve the necessary level of accuracy required for urban air modelling, it is recommended by (Sturm & Winiwarter 2004) to use the following methodical approaches:

Mobile sources

It is recommended to use bottom-up approaches for traffic on the main road network, regular railway traffic on the rail network and scheduled traffic for ferry boats. Top-down approach might be used for traffic on the secondary road network (area sources), cold start emissions and evaporation losses, off-road traffic, shunting activities in railway traffic, maritime and inland traffic on waterways, harbour activities, residential combustion, solvent use etc.

Stationary sources

Bottom-up approaches are applied to point sources while area sources are mostly treated with top-down methodologies. If the activity data is given on a statistical basis (e.g. for residential combustion), a top-down methodology must be applied, e.g. for SNAP sectors 2 (heat demand), 6 (solvents), 10 (agriculture) and 11 (nature).

If enough information on location, activity data and emission factors is available it is possible to use a bottom-up approach instead of a top-down method. If for instance detailed information on land use cover and location is available for emission sources from nature and agriculture, and specific meteorological data and emission factors can be used the approach can be considered as bottom-up.

3. Review of existing methods for generating emission data

Emission inventories are developed using different types of information that describe source specific activities and emissions, their location and their temporal variation. In the following, approaches to calculate and allocate emission data are described. Several aspects of and methodologies for emission calculation concern all scales of emission inventories. Common methodologies are presented for the usage of emission factors, activity data and trend information as well as for data to characterise emissions with regard to time and chemical or physical properties. Additionally, there are special requirements and aspects for the urban/agglomerate and local/hotspot scale with regards to calculation methodologies and spatial resolution.

A number of air quality models on various scales are currently applied which all require emissions in spatial and temporal resolution. The Model Documentation System of the European Topic Centre on Air and Climate Change contains basic key words and descriptions for several dispersion models (see <http://pandora.meng.auth.gr/mds/mds.php>). Methodologies used to generate emission data within existing inventories and models always depend on data availability and project capacities.

In addition to emission calculation based on a source oriented approach, there are several receptor oriented methodologies for an identification of emission sources. Based on an analysis of measured concentrations and their chemical composition several statistical methods and inverse modelling techniques were developed to provide an apportionment of major source groups such as exhaust from diesel engines, brake and tyre wear, wood, coal and oil combustion, suspension of crustal material, metal industries refineries etc. The sectoral resolution of these approaches is rather low as for each source group a unique tracer is required. Thus, technological specifications of comparable processes can not be considered (e.g. to distinguish between diesel engines from passenger cars, heavy duty vehicles and off-road vehicles). However, because of the use of measured local concentrations source apportionment provides important information that can be used for a comprehensive validation of emission data and source inventories.

3.1. Methodologies for the calculation of emissions

Emissions are usually calculated based on statistical or estimated activity rates and technology specific emission factors, i.e. emission per unit activity (s. equation below). Activity rates and emission factors need to have the same level of detail and relation to be applicable. Thus, it is often necessary to aggregate or disaggregate these parameters with the help of statistical data or other information. If for instance mileage data are only available for different vehicle categories and average urban, rural and highway road classes, more detailed emission factors have to be averaged using statistical distributions e.g. of slope, driving conditions, share of diesel and gasoline engines, EURO emission standards and vehicle capacity/weight. Availability and level of detail of activity rates depend on national/regional conditions.

$$E_{ij} = EF_i * A_i * F_{ij} \quad (1)$$

where:

E_{ij} = pollutant emission from source group i and (optional) component j

EF_i = pollutant emission factor of source group i

A_i = annual, monthly, daily or hourly activity rate of source group i

F_{ij} = (optional) factor (e.g. particle size factor, factor for chemical speciation of PM, VOC, NO_x) for emission component j from source group i

3.1.1. Stationary anthropogenic sources

General methodologies and data bases

Simple or more detailed methodologies and basic data for the calculation of most anthropogenic emissions can be found within the joined EMEP/CORINAIR Atmospheric Emission Inventory Handbook (EEA 2003). Information within the US EPA emission data compilation AP42 (EPA 1995) is continuously updated and widely used for emission calculations as well. The Factor Information REtrieval (FIRE) Data System is a database containing EPA's emission estimation factors for criteria and hazardous air pollutants in an easy to use Windows program (EPA 2004). Other useful EPA tools are TANKS for the estimation of volatile organic compound (VOC) and hazardous air pollutant (HAP) emissions from fixed- and floating-roof storage tanks (EPA 2005) and SPECIATE, EPA's repository of Total Organic Compound (TOC) and Particulate Matter (PM) speciated profiles for a variety of sources for use in source apportionment studies (EPA 2002). However, these data bases partly consist of measurement results that represent technologies of the eighties or earlier years. Nevertheless, useful information is provided for stack as well as fugitive emissions. Several other national or international studies are currently available dealing with the availability and current state of knowledge regarding emission factors that can also be used for sectoral emission calculation (e.g. BUWAL 2000a, TNO 2001, Klimont et al. 2002, AEAT 2001a, DMU 2006, CITEPA 2006). Furthermore, information on specific emissions from industrial sources can be found within the Reference Documents on best available techniques (BREFs) (see <http://eippcb.jrc.es>). BREFs present results of an exchange of technical information organised in specific Technical Working Groups to determine and describe "best available techniques" in accordance with Council Directive 96/61/EC concerning integrated pollution prevention and control (IPPC Directive). The Organisation for Economic Co-operation and Development (OECD) publishes free-of-charge Emission Scenario Documents considering emissions from chemical industries within the framework of the Inter-Organisation Programme for the Sound Management of Chemicals (IOMC) (s. <http://www.oecd.org/>).

In addition, a large amount of data on industrial processes that have to be submitted by the operating companies to regional or national administrative authorities might be used for emission modelling. These data often include activity rates such as production volume, fuel input etc., measured or calculated annual/monthly stack emissions and additional point source information but have a restricted availability due to data security concerns. If accessi-

ble these data can be included as anonymised information, especially for regional meso-scale and urban/local scale inventories in order to significantly reduce uncertainties.

Special requirements and aspects for urban and local scale

If significant point sources have to be included into an urban or local emission model, industrial emission data are often available from administrative authorities. The usage of emission factors from literature does usually not reflect local characteristics and can lead to significant under- or overestimation of single source emissions. Sampling and analysis at individual sources require standard methods and standard facilities (see e.g. ISO standards (www.iso.org), US EPA Emission Measurement Center (www.epa.gov/ttn/emc/), German VDI guidelines (www.vdi.de)) and need significant experience and funds depending on the air pollutant considered. Sampling methods for fine particulate matter are still at the development stage, estimated inaccuracies are commonly in the range of $\pm 50\%$ (Sloss 2004).

For local area sources, emissions normally have to be calculated with a top down approach, using statistical parameters such as population data to derive urban and local activities based on national or regional information. Statistical data often exist on urban scale as well, e.g. for fuel consumption in households and industrial combustion. In some cases small scale data e.g. the fuel consumption per block/square of residential area are available. If emission calculations have to be done for a local area, simple or more detailed methodologies and basic data can be found within the above mentioned reports and data bases.

3.1.2. Mobile anthropogenic sources

General methodologies and data bases

For mobile source exhaust emissions plenty of measuring data and several extensive data bases or models exist that can be used for calculations on all scales. The calculation of hot emissions is usually done by multiplying annual, monthly, daily or even hourly mileage data and emission factors on various levels of detail. Emission calculations for road traffic should be done for different combinations of vehicle categories (e.g. PC, LDV, HDV, buses, two wheelers), EURO emission standards, road classes, slopes, driving conditions, vehicle capacities/weights. The calculation of excess cold start and evaporation emissions from road traffic is more difficult because emissions and temporal variation are temperature dependent and should therefore be considered with an approach that includes regional or even local ambient temperature. In general, ambient temperature can be taken from observations or from the same meteorological models that are used as input to CTM's. A detailed hourly bottom up approach for excess cold start and evaporation emissions was for example carried out for Germany by (Wickert 2001) including hourly temperature data and hourly activity profiles on a highly detailed sectoral level (road/vehicle classes).

A simple methodology based on average emissions factors and a more detailed methodology including relevant regional/local parameters can be found for all mobile source emissions within the Atmospheric Emission Inventory Handbook (EEA 2003). For road transport, emission factors are provided by COPERT III and HBEFA (among others, see Table 3-1). The COPERT III model (Computer Programme to calculate Emissions from Road Transport) follows a harmonised European approach and contains speed dependent emission factors per vehicle technology for hot, cold start and gasoline evaporation (Ntziachristos & Samaras

2000). Additionally, other basic data is given such as monthly national temperatures, fuel quality, average speed and driving shares per country and road class. The model enables the user to calculate national total emissions for EU 15+ (with activity rates provided by the user). Emission factors for passenger cars up to EURO I were derived from the project MEET (Samaras & Ntziachristos 1998) and the COST 319 action (Joumard 1999). Emission factors for heavy duty vehicles, coaches and busses were taken from HBEFA, as well as emission factors for motorcycles and mopeds (further processed by Rijkeboer 1997). The model has a spatial differentiation of three road classes (urban, rural, highways). Emission factors for non-road transport are provided in an additional data base (as a function of power, not country specific). Currently, a new model version is developed (COPERT IV) that contains updated emission factors up to EURO IV based on real-world driving cycles for passenger cars and heavy duty vehicles from the COST 346 action and the project ARTEMIS.

The HBEFA (Handbook Emission Factors for Road Transport) is a highly detailed emission factor data base and was published as separate versions for Germany, Switzerland and Austria (e.g. UBA 2004). It provides emission factors for hot, excess cold start and evaporation emissions and uses specific information on slope, average trip length and traffic situations from those countries. Emission factors for passenger cars, light and heavy duty vehicles were derived in measurement campaigns with real world test cycles up to EURO III (in the case of HDV in cooperation with COST 346 and ARTEMIS). Cold start excess emission factors depend on ambient temperature and reduction factors for short trips. Options to assess influences of air conditions and diesel particulate filters are also included. The model has a spatial differentiation of 30 regular traffic situations (urban, rural, highways for different vehicle speed, type of road etc.) plus additional country specific situations including acceleration, deceleration, multilane-roads, serpentines, slope etc. Methodologies for road traffic emission calculation based on HBEFA were published in (VDI 2003). German road traffic emissions are officially calculated with HBEFA implemented into the German TREMOD model.

Table 3-1: Emission factor data bases for the calculation of road traffic emissions

EMISSION MODEL	SECTORAL RESOLUTION	CALCULATION METHODOLOGY	POLLUTANTS COVERED
HBEFA, e.g. for Germany (UBA 2004)	hot, cold start and evaporation emission factors for detailed source categories: e.g. ~6.000 combinations of vehicle category, EURO emission standard, road class, slope and driving conditions. Aggregation is possible or a further distinction between vehicle capacities/weights (~16.000 combinations).	emission factors are based on detailed measuring data and derived emission functions that include vehicle acceleration. Country specific fleet information is used for aggregation.	SO ₂ , CO, NO _x , NMVOC, CH ₄ , CO ₂ , N ₂ O, NH ₃ , diesel PM, xylene, toluene, benzene, Pb
COPERT III (Ntziachristos & Samaras 2000)	hot, cold start and evaporation emissions from PC, LDV, HDV, urban buses, 2 wheelers differentiated in urban, rural and highway driving.	emission factors from COST 319, MEET and other European projects.	SO ₂ , CO, NO _x , NMVOC & split, CH ₄ , CO ₂ , N ₂ O, NH ₃ , diesel PM, 23 PAH & POP, Dioxins/Furans, HM from fuel (Pb, Cd, Cu, Cr, Ni, Se, Zn)

Because of the different methodologies both emission factor data sets are based on (emission factors as a function of speed in the case of COPERT and as a function of traffic situations in the case of HBEFA), the same assumptions for vehicle category, technology and road class will lead to different results (see Figure 3-1). The user has to decide individually which emission factors are suitable for his scope. In addition, there are several other emission factor data sets having a local or national scope (see e.g. AEAT 2002, DMU 2006).

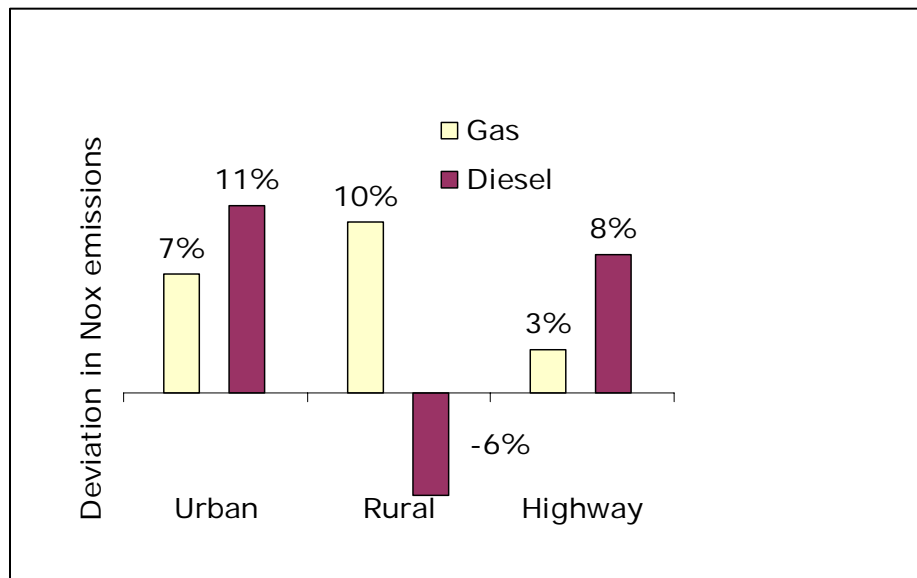


Figure 3-1: Deviation of NO_x emissions calculated with COPERT III from NO_x emissions calculated with HBEFA (example: passenger cars in Sweden in the year 2000)

In addition to exhaust emissions, non-exhaust emissions due to tyre and brake wear and road dust suspension should be considered for an accurate calculation of fine particulate matter emissions. However, knowledge on specific non-exhaust emissions per different road classes, vehicle categories and driving conditions is scarce and still in the process of scientific discussion. Available information on tyre and brake wear emission factors was recently published in (EEA 2003). For road dust suspension, the US EPA published an empiric equation derived from measurements in the USA (EPA 2003) that is often used for emission estimations. EPA methodology was often criticised because it lacks mechanistic background (see e.g. Venkatram 2000). Meanwhile first measurement and derived emission factors have been published for different European driving conditions that give a more accurate assessment of non-exhaust emissions (e.g. Düring et al. 2005, Gehrig et al. 2003). Further information is given in Chapter 3.1.6.

Off-road-vehicles and other transport (transport by rail, air planes and ships) may cause even higher emissions than road traffic, depending on the local/regional activity rates. Specific emissions of non-road diesel engines (e.g. of construction machinery, agricultural vehicles or ships) are usually much higher than emission factors from modern heavy duty vehicles on the road. Specific emissions of these utilities were studied e.g. in (BUWAL 2000b), (AEAT 2001) and (Klimont et al. 2002) considering all types of nonroad sources, (Jorgensen & Sorenson 1997) for rail transport, (Trozzi & Vaccaro 1998a), (Trozzi & Vaccaro 1998b), (Dor-

land & Olsthoorn 1998), (Dings et al. 1997), (Lloyd's Register 1995) for ship emissions. Specific fuel consumption for various engines according to (ICAO 1999) can be used for the calculation of air craft emissions during the LTO cycles (landing and take-off cycles).

Special requirements and aspects for urban and local scale

Urban and local road traffic emissions are usually calculated based on site specific census data from automatic counting of small and large vehicles and/or manual counting of different vehicle classes. A detailed analysis of traffic flow, traffic volume and fleet composition is required for an accurate emission modelling. There exist two different calculation methodologies on urban and local scale:

- Emission calculation using emission factors and parameterised hourly or half-hourly traffic flow data; assignment of a typical average driving condition that present road class, slope and travel patterns as speed and flow.
- Emission calculation using emission functions and online traffic flow data from a traffic flow model; vehicle speed and acceleration are taken into account for single road segments; emission factors are based on engine emission maps taking into account speed and engine load; model calibration is often done with site specific information on vehicle speed, composition etc.

Direct consideration of acceleration by the use of emission functions is usually not possible, as these data (e.g. used for the development of HBEFA) are not publicly available. Thus, emission factors are usually assigned according to the road class, driving condition/speed, slope and other site-specific conditions like presence of traffic lights, crossroads or pedestrian crossings. In addition to hot emissions, excess cold start emissions have to be taken into account especially for urban traffic for the first 5 km driven after a cold engine start. Information on the average travel distance distribution per vehicle category and road class can be found in literature or derived from local traffic models. In addition, hourly or half hourly ambient temperature data are required for the calculation of cold start emissions, as cold start factors (e.g. g/start) depend on temperature. VOC emissions due to gasoline evaporation (hot soak losses, diurnal losses, running losses) should be taken into account as well. Again, if available, local information on ambient temperature and parking time distributions should be used. Figure 3-2 shows as an example a possible detailed methodology for road traffic emission calculation on local scale.

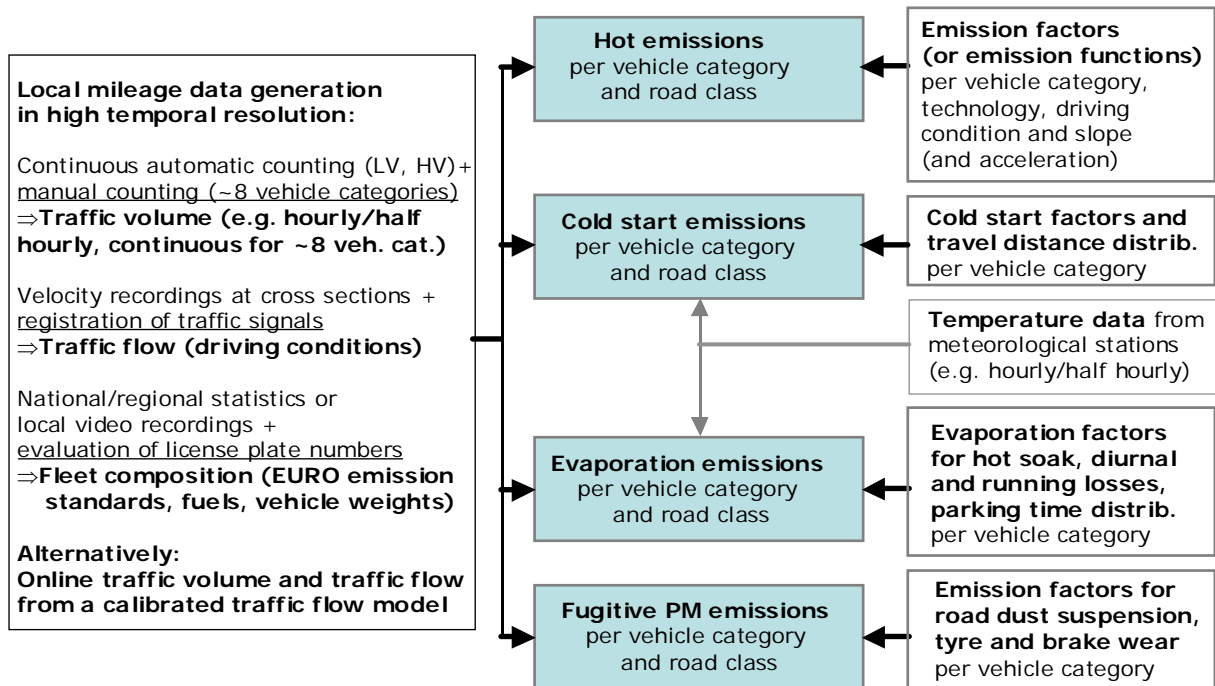


Figure 3-2: Example of methodology for road traffic emission calculation on local scale

3.1.3. Biogenic/natural emissions

Biogenic and natural emission sources are relevant to model O_3 , NO_2 and PM. Most regional CTM follow the approach by (Simpson 1995) to model biogenic NMVOC emissions from forests. To be able to calculate these forest emissions an appropriate land use and land cover data base is required. Recent studies by (Lenz et al. 2001) have provided detailed information about forest types in Europe. For NO_2 , most model approaches follow the work by (Yienger and Levy 1995) and (Stohl et al. 1996). For sea-salt, most models follow the approach by (Monahan et al. 1986). Concerning windblown dust several ongoing studies try to come to a coherent approach; also in this case a proper land use and land cover data base is of primary importance. For the calculation of Sahara-dust, most modellers follow the approach by (Marticorena et al. 1997).

It should be noted that there the CTM all follow slightly different approaches to determine biogenic and natural emissions, and that no recent studies have been performed to compare the different methods which are in use. Within the EC-project NATAIR a review and recommendations for an improvement of these methodologies will be done until the end of 2007 (see <http://natair.ier.uni-stuttgart.de>).

3.1.4. Substantial disaggregation

NMVOG

A disaggregation of total NMVOG in defined substance classes is required according to the ozone formation mechanism of the CTM that will be used for atmospheric modelling. Hundreds of different substances are usually lumped in classes according to different schemes and atmospheric chemistry mechanisms. The following schemes are often used: EMEP MSC-W, RADM2, RACM and CBM4. A brief description of gas-phase chemistry mechanisms can be found in the Air4EU Deliverable 5.1. NMVOG speciation schemes for significant sources were published by (Rudd & Marlowe 1998), (Passant 2002) and (Theloke & Friedrich 2007).

Particulate matter

Emissions of airborne particulate matter cover a wide range of diameter between a few nm up to about 100 μm and thus have various atmospheric properties and health effects. Calculations of fine particulate matter emissions are usually done by assigning source specific particle size factors (e.g. for PM_{10} and $\text{PM}_{2.5}$) to total particulate matter (PM) emissions, calculated with emission factors and activity rates. Particle size factors and PM emission factors used in emission inventories do often not originate from the same emission measurements. This increases uncertainties due to the mass losses that occur during size selective impactor measurements in comparison to the measurement of total particulate matter emissions. Compiled particle size factors can be found e.g. in (EPA 1995), (Dreiseidler et al. 1998), (TNO 2001), (Klimont et al. 2002) and on national scale e.g. in (AEAT 2001), (CITEPA 2006) and (Pregger 2006).

Emissions of elements or chemical compounds as a portion of emitted particles are often estimated by combining PM emission data with analysed source specific information on the substance content of these emissions. In addition, especially for heavy metals (HM) and persistent organic pollutants (POP) a lot of measured specific emission factors are available in the literature to calculate total emissions of these substances (sum of solid, liquid and gaseous emissions). Except mercury (Hg) atmospheric concentrations of these substances are mainly in the solid state. (Berdowski et al. 1997) provided a first emission estimation for heavy metals and persistent organic pollutants (POP) in Europe. Estimated and officially reported country totals are provided amongst others for particulate matter as well as HM and POPs by EMEP WEBDAB (EMEP 2004). A comprehensive compilation of PM speciations can be found in (EPA 2002). An improved European emission inventory for heavy metals was developed within the ESPREME project (ESPREME 2005) using officially reported EMEP emissions as a basis and additional calculations to improve the data base.

Emissions of elemental carbon (EC) and organic carbon (OC) are usually estimated based on a quantification of particulate matter emissions and source specific EC/OC content factors. Only little information exists on these fractions. Specific information can be found e.g. in (EPA 2002). For PM from wood combustion in small stoves or fire places, (McDonald et al. 2000) cites measured EC fractions of 7 to 15 % and OC fraction of 58 to 71 %. In the nineties estimations of EC from road traffic in Germany were 60 % for passenger cars and 40 % for heavy duty vehicles. The implementation of oxidation catalysts as well as diesel particulate

filters might already have changed the fleet emissions. Black Carbon (BC) - the principal light-absorbing component of soot - is usually quantified based on emission factors that are derived using specific methods (e.g. light absorption, analysis of dissolved filter samples). The main sources for primary carbonaceous aerosols are incomplete combustion of fossil fuel and biomass. There currently exist two global emission inventories for BC (Bond et al. 2004), (Liousse et al. 2004) with a statistical approach and rather low sectoral resolution.

Nitrogen oxides

So far, only little information was available to specify the fractions of NO and NO₂ for individual emission sources. NO oxidizes to NO₂ in the atmosphere, and thus NO_x emission data are usually calculated and shown as NO₂. NO is rather harmless to human health, whereas NO₂ causes health risks at certain concentrations. In addition, there are other harmful effects of nitrogen oxides, e.g. NO₂ plays an important role in ozone formation via complex reactions with VOCs. NO₂ concentration levels in the urban air can exceed limit values and can cause health effects, especially local problems may occur, e.g. beside heavily-loaded roads. NO_x emissions from combustion plants usually consist of ~95 % NO and ~5 % NO₂. For diesel engines without oxidation catalyst or a catalyzed PM filter the NO ratio is usually > 90 %. Engine-out NO_x portion contain typically some 5% of NO₂, but even higher than 20% of NO₂ may exist in raw exhaust at some running conditions. The engine-out NO₂/NO_x ratio depends on the engine design, calibration and the operating conditions. Diesel engines have higher NO₂/NO_x ratio than gasoline engines.

Average NO and NO_x concentrations near roads in Europe decreased during the last decade while NO₂ concentration remained stable or even increased. For instance, (Höpfner et al. 2004) reported that in Germany NO₂ concentrations are at the same level as in 1987, whereas NO_x concentrations have decreased by 50% at the same time period. One reason for this development may be an increase of the direct NO₂ emissions. Since the implementation of EURO 3 emission standards, oxidation catalysts became a standard equipment of diesel passenger cars. Due to NO oxidation to NO₂, the NO₂ ratio is much higher in the exhaust of diesel vehicles. In addition, municipal buses were equipped with CRT-systems in several cities generating and using NO₂ for soot oxidation. The consequence on urban air quality is observed by monitoring stations. NO₂ emissions from stoichiometric catalyst-equipped gasoline cars or NGVs have been reported to be extremely low (Sjödin et al. 2004). Properly balanced diesel particulate filters using the creation of NO₂ in an upstream oxidation catalyst to enhance regeneration should not produce more NO₂ than needed for the soot oxidation. A review of information on source and technology specific NO₂/NO_x ratio can be found in Table 6-1 in the Annex.

3.1.5. Scenario calculations

Starting from a base year emission inventory a "business as usual" scenario (trend scenario) can be developed assigning specific trend information or assumptions for each source group. National emission projections are required by NEC Directive reported to the EU Commission, by CLRTAP reported to UNECE and for GHG emissions reported to IPCC.

Trend factors for activities can partly be derived from available prognosis or projection studies or can be estimated by extrapolating time series of activity data or available secondary

statistical data from the past in the future. Trend factors for emission factors have to be based on an analysis or estimation of effects that have to be expected in consequence of environmental regulations (e.g. emission limits of Large Combustion Plant Directive, EURO emission standards for vehicles) and technological trends (e.g. implementation of new plants with low specific emissions). An abatement scenario uses information about effects of additional technical or non technical measures on emission factors or activity rates. Scenario calculations are mainly based on European/national trend information but should also include regional/local conditions, which may change the emission situation in the future (e.g. changes in road network, land use, industrial structure).

Trend scenarios for mobile sources (road motorways, road urban, road non-urban, rail, air, inland waterways) can be found as country totals up to the year 2020 in the TREMOVE model (<http://www.tremove.org/>). TREMOVE is based on COPERT emission factors and transport volume and cost data from the SCENES transport forecasting model. Resulting emissions can be downloaded for free.

Trend scenarios for energy consumption are provided by the PRIMES model, developed by the National Technical University of Athens (NTUA) (see www.e3mlab.ntua.gr). The model provides fully detailed EUROSTAT energy balance sheets per country and per year and scenarios for the primary energy production, net imports, gross inland consumption, electricity generation, final energy demand by sector and CO₂ emissions. Main characteristics of PRIMES are the partial equilibrium energy model, 2000-2030 time horizon, a very detailed techno-economic characterisation of energy technologies, EU policy analyses (DG RTD, ENV and TREN) and European energy and transport trends up to 2030. The optimisation is done with market-related mechanisms (prices) modelling all relevant demand and supply sectors in individual countries.

National/regional emission trend scenarios can be taken from CAFÉ baselines (IIASA 2004). Scenarios are given for 30 European countries covering the activity rates and emissions for NO₂, SO₂, NH₃, NMVOC as well as TSP, PM₁₀ and PM_{2.5}. The CLE (current legislation) CAFE baseline relies on the baseline energy projection of the 'European energy and transport – Trends to 2030' outlook (CEC 2003) as a starting point assuming continuation of current trends in the energy sector. As an alternative projection, the CAFE baseline NAT employed the national energy projections of the EU Member States. For both trend scenarios IIASA quantified for each Member State the impacts of the legislation on future emissions. In addition, in the MTFR scenario (Maximum Technically Feasible Reduction) was assumed full implementation of the presently available most advanced technical emission control measures in the year 2020, although excluding premature retirement of existing equipment before the end of its technical life time (see reports <http://www.iiasa.ac.at/rains/caf.html>).

IIASA global "Scenarios of World Anthropogenic Emissions of Air Pollutants and Methane" were developed up to 2030 (IIASA 2005). Global scenarios with regard to GHG emissions can be found in the IPCC SRES report (IPCC 2000b). The scenarios cover a wide range of the main demographic, economic and technological driving forces of GHG and sulphur emissions.

The development of action plans and strategies to reduce urban emissions requires trend and abatement scenarios for ambient air concentrations. Thus, emission trends have to be transferred to the local/urban as well as regional source contributions. This is usually done by

using results of source apportionment studies analysing monitoring data and the origin of locally measured concentrations (see e.g. John & Kuhlbusch 2004).

3.1.6. Special topics considered in Air4EU case studies

Emissions from two wheelers

PTW (powered two-wheelers such as scooters, mopeds and motorcycles) have so far not been playing a dominant role in transportation and have therefore been included only recently in EU-wide emission standards (Euro 1-3). However, in urban traffic their contributions to total emissions are remarkably high. A comparison of Swiss fleet emissions from two wheelers and passenger cars in urban areas (in tons per year) shows that CO and HC emissions caused by PTW are higher by a factor of 2.7 and 16, respectively (Vasic & Weilenmann 2006).

The highest number of PTW can be found in Italy, Germany, France and Spain (ACEM 2004). Projections until 2020 show a decrease in stock of mopeds and motorcycles all over Europe with the exception of Italy, Sweden (Ntziachristos et al. 2004) and Greece (TRENDS 2003). Their contributions to air pollution, especially by VOC (e.g. Benzene), CO and PM will thus continue to be of relevance in certain areas. Because of rich combustion, NO_x emissions of PTW (even when not equipped with a catalyst) are relatively low and comparable to modern passenger cars.

Mopeds and motorcycles have a wide range of technologies (2-stroke, 2-stroke direct injection and 4-stroke), engine characteristics (less than 100 cm³ up to 2000 cm³), driving patterns (from daily transport mainly in southern Europe to recreational use mainly in northern Europe) and driving dynamics. Real world driving cycles like CADC (Common ARTEMIS Driving Cycle) and WMTC (Worldwide Harmonized Motorcycle Emissions Certification Procedure) were thus developed to get an improved representation of motorcycle emissions (Gense & Elst 2003).

As still a considerable number of PTW is equipped with a 2-stroke engine, their PM emissions can be even higher in concentration than particulate matter emitted by diesel passenger cars. A lot of the fraction consists of semi-volatile hydrocarbons originating from fuel that is not properly combusted. Effective control measures like direct injection are therefore able to not only reduce HC but also PM emissions (by up to 90%, Ntziachristos et al. 2004).

Emission factor data sources for two wheelers are described in Table 3-2. Scooters and mopeds (two-stroke and engine displacement <50 cm³) are mostly used in urban and rural environments, therefore emission factors for highway driving are not given.

Table 3-2: Sources of emission factors for two wheelers

SOURCE	SECTORAL RESOLUTION	SPATIAL RESOLUTION	POLLUTANTS COVERED
Czerwinski et al. 2000, 2002, 2003	Euro 1 hot and cold emission factors for mopeds and motorcycles (with and without catalyst)	Urban, suburban, rural, highway	CO, HC, NO _x , CO ₂ , PM
HBEFA UBA 2004	Pre-Euro and Euro 1 for mopeds (<50cm ³) and motorcycles (<150 and >150 cm ³ for 2-stroke engines, <150, 150-250, 250-750 and >750 cm ³ for 4-stroke engines). Hot and evaporation emission factors for detailed source categories. Cold start emission factors are not given.	Evaporation emission factors as national mean, hot emission factors per road class and driving situation (can be aggregated to urban, rural and highway driving).	SO ₂ , CO, NO _x , NMVOC, CH ₄ , CO ₂ , N ₂ O, NH ₃ , xylene, toluene, benzene
Ntziachristos et al. 2004	Pre-Euro, Euro 1, Euro 2 and Euro 3 emission factors for 2-stroke (mineral and synthetic oil), 4-stroke engines and compression ignition	None	PM
Ntziachristos et al. 2006	Pre-Euro, Euro 1, Euro 2 and Euro 3 emission factors for 2-stroke and 4-stroke engines	Urban, rural, highway	CO, HC, NO _x , PM
Vasic & Weilenmann 2006	Euro 1 hot emission factors for motorcycles, evaporation and cold start emission factors are not given.	Urban, rural, highway	CO, HC, NO _x , CO ₂

Tables 6-2 and 6-3 in Annex I provide results of a data review for measured two-wheeler emission factors for benzene and PM. Annex II provides additional information on two-wheeler emissions with main focus on particulate matter, emission measurement techniques and uncertainties. This annexed report from the LHTEE of Aristotle University of Thessaloniki comprises a detailed review of methodologies and studies in the field of PTW emission assessment and specific recommendations that are not referred to in Chapter 4.

Non-exhaust emissions from road traffic

Non-exhaust emissions from mobile sources are caused by the abrasion of brakes, tyres and road pavement and by the traffic induced suspension of road dust. Suspended particles can have various sources such as atmospheric deposition, dirt from side walks, trees etc. and sand or salt for ice control (Figure 3-3). Non-exhaust particulate matter mostly has an aerodynamic diameter of at least 1-2 μm as it is mechanically formed (particles from combustion processes tend to be smaller).

Tyres are made out of a variety of rubber types as well as metal and organic additives (mostly zinc). Brakes also consist of a variety of metallic, mineral and ceramic materials. Size distribution of particulate matter from tyre and brake wear is still uncertain, a fact that is reflected in the range of particle sizes given in literature. For brake wear (Garg et al 2000) found that 63 % of total suspended particles (TSP) can be attributed to the $\text{PM}_{2.5}$ fraction (and 86 % to PM_{10}), whereas in (TNO 2001) it was assumed that 40 % of PM_{10} represent the $\text{PM}_{2.5}$ fraction. For tyre wear, (Rauterberg-Wulff 1998) found that particles were always $> 2.5 \mu\text{m}$, while (Lükewille et al. 2001) estimate that 5% of PM_{10} belong to the $\text{PM}_{2.5}$ fraction. Partly much higher PM_{10} and $\text{PM}_{2.5}$ fractions were assumed in (EEA 2003). References and examples of emission factors can be found in Table 3-4 and Table 6-4 (Annex I).

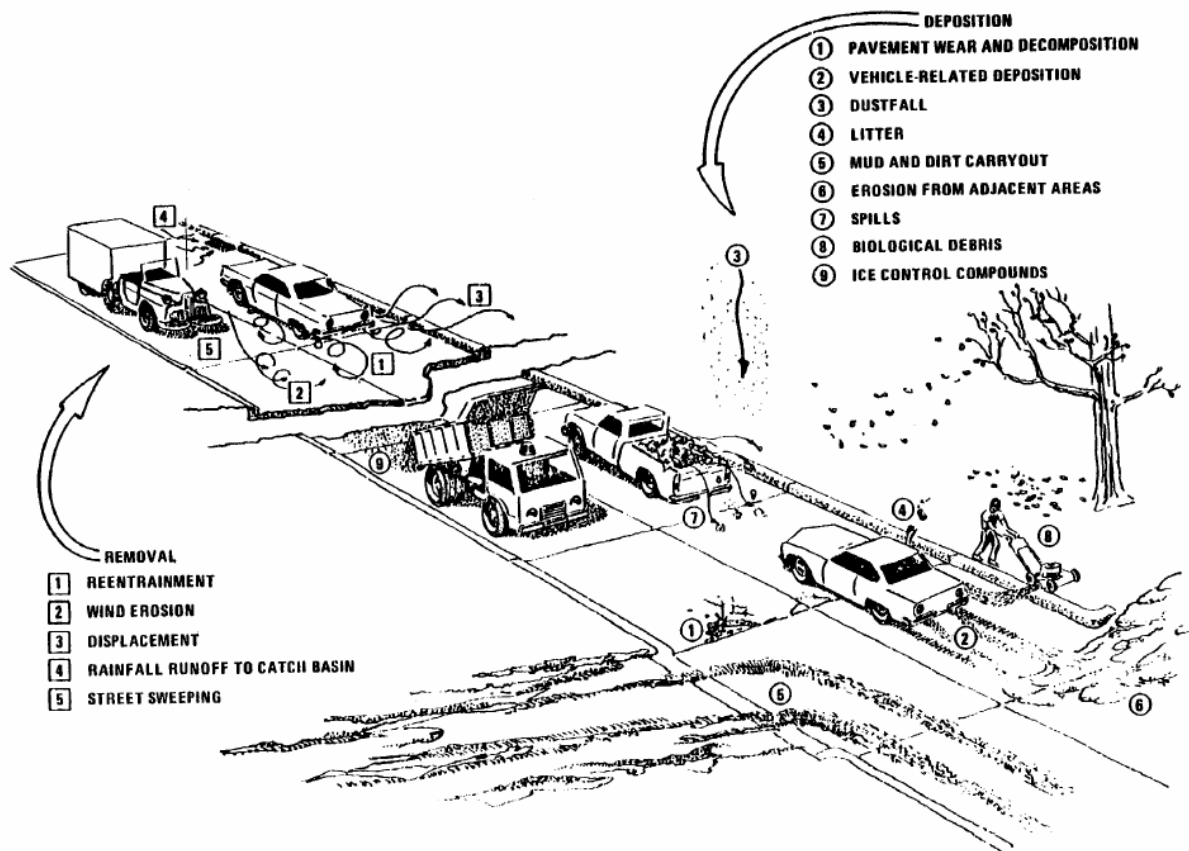


Figure 3-3: Non-exhaust emission sources (EPA 2003)

There are several models to describe the suspension of road dust. The US EPA developed an empirical model that incorporates silt load and average weight of the vehicle fleet as main parameters based on an analysis of 65 measuring data sets from the US (see EPA 2003 and equation 2). It was assumed that PM emissions from transport are mostly due to dust suspension. However, this purely statistical methodology was criticised by (Venkatram 2000) and outside of the US because it lacks mechanistic background and it is based on local data sets that were partly measured under conditions (silt loads, humidity, vegetation, weight of vehicle fleet) that are far from usual driving situations especially in middle and northern Europe. An application of a modified EPA-model was done for Germany in (Pregger 2006). For that, factors and model constants for usual road conditions derived in (Venkatram 2000) were combined with information on average vehicle weight, silt load of road surfaces and average mileage on different road categories in Germany to derive emission factors as shown in Table 3-3. Even this modified EPA methodology results in significant higher emission estimations compared to German measurement results of (Düring et al. 2005) (see Table 6-4). Whereas for urban roads the fleet emission factors turn out to be quite similar, the application of the modified US EPA equation for rural and highway traffic results in much higher specific emissions and indicates a large overestimation and non-applicability of the model.

$$EF = k * (sL)^p * (W)^b \quad (2)$$

where:

- EF = emission factor PM (unit according to unit of k)
- k = factor depending on PM fraction:
 (EPA 2003): PM₃₀: k = 2.94 g/km PM₁₀: k = 0.56 g/km PM_{2.5}: k = 0.135 g/km
 modification derived from (Venkatram 2000): PM₁₀: k = 0.18 g/km
- sL = silt load on road surface (PM₇₅) in g/m²
- W = average weight of vehicle fleet in US short tons
- p, b = empirical constants:
 (EPA 2003): p = 0.65 b = 1.5
 modification derived from (Venkatram 2000): p = 0.52 b = 2.14

Table 3-3: Emission factors for total PM from transport in Germany 2000 including exhaust, abrasion and suspension derived from a modified US EPA model (Pregger 2006)

in g/Vehicle-km	EF PM	EF PM ₁₀	EF PM _{2.5}
Highways	0.387	0.165	0.068
Urban roads	0.506	0.215	0.089
Main rural roads	0.374	0.159	0.066
Small rural roads	0.468	0.199	0.082

Nordic countries like Sweden, Norway or Finland show significant non-exhaust emissions because of the use of studded tyres during winter and early spring months. Studies like (Omstedt et al. 2005) for Sweden or (Tønnesen 2005) as part of the AirQUIS modelling system for Norway take into account road wetness and the interaction of suspension with road dust to calculate additional tyre and road surface wear.

(Tønnesen 2005) developed an emission model for particulate matter from traffic sources, providing hourly emission rates for PM₁₀ and PM_{2.5} from vehicular traffic. It is used as a part of the AirQUIS modelling system. For exhaust particles, emission factors are used. For the road dust, data from measurements close to roads are used to derive empirical factors for the emission. An emission reduction algorithm for non-dry conditions is also included. Equation 3 is used to calculate PM₁₀ emissions per vehicle for dry road surface conditions

$$QPM = Q_{EP} + (QR_{2.5} + C * (A * TT + B) * (VD / VD_{ref})^2) * (0.98 * ST + 0.02) \quad (3)$$

Here, QPM is the emission per vehicle (g/vkm), Q_{EP} is the average exhaust particle emission for the actual road (g/vkm), $QR_{2.5}$ is the emission of fine particles from the road (g/vkm), C is the emission of fine particles derived from measurements, A and B are derived linear constants for heavy traffic, TT is the percentage of heavy vehicles, VD is the driving speed (km/h), VD_{ref} is a reference driving speed and ST is the fraction of studded tyres.

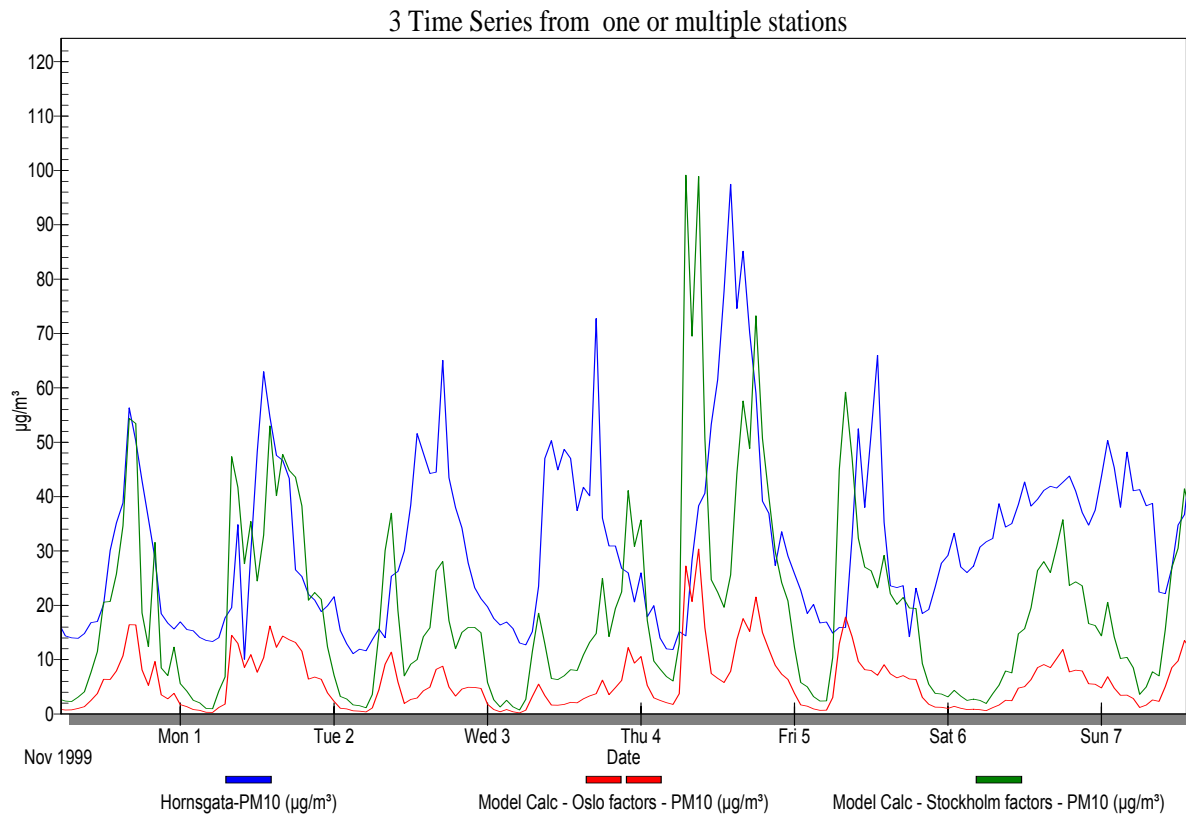


Figure 3-4: Modelled and measured concentrations of PM_{10} at a roadside location in Stockholm using Oslo (red) and Stockholm parameters (green) in the emission model. Measurements are shown in blue.

Measurements made in both Oslo and Stockholm give significantly different values for the empirical constants used in the above formula (see Figure 3-4), a factor of 2, indicating the variation due to local conditions. In addition the model requires adjustment of the parameters during periods after road side snow deposits have disappeared, which leaves behind a large reservoir of dust. The dust emission model still requires further development in order to describe these high intensity periods. In addition, a better description of the fine fraction particles originating from the road is also needed. When more measurements of roadside concentrations in the summer season become available, a better expression for the dependency on use of studded tyres can also be formulated. Despite its acknowledged shortcomings, the emission model provides crucial input to the air quality modelling.

Some of the Air4EU city partners developed own approaches for non-exhaust emissions in their inventories. Rotterdam is using traffic emission factors that take into account 50% suspension of road dust for street canyon modelling. The calculations compare reasonably well with monitoring data. Oslo is using local emission factors that were developed empirically (from monitoring data analyses) and calculations based on (Tønnesen 2005).

Several studies have summarised the literature of studies of resuspension PM emission factors related to roads (e.g. Gustavsson 2003, Düring & Lohmeyer 2001).

Table 3-4: Emission factors for road traffic non-exhaust sources

SOURCE	TYPE	SECTORAL RESOLUTION	SPATIAL RESOLUTION	POLLUTANTS COVERED
Düring et al. 2005	Measurements at German roads, literature review	Sum of non-exhaust: Suspension of road dust including brake and tyre wear for PC/LDV and HDV	Urban, rural, highway, tunnel (urban under different driving conditions and traffic situations)	PM ₁₀
Gehrig et al. 2003	Measurements at Swiss roads	Sum of non-exhaust: Suspension of road dust including brake and tyre wear for PC/LDV and HDV	Urban, rural and highway	PM ₁₀
Fitz & Bufalino 2002	Measurements at Californian roads	Suspension of road dust, PC, LDV	Urban, rural and highway	PM ₁₀ , PM _{2.5}
Garg et al. 2000	Measurements at test benches	Brake wear for PC, LDV, HDV, 2-wheelers	None	TSP, PM ₁₀ , PM _{2.5} (30 % of abrasion is TSP)
Gebbe et al. 1998	Measurements at test benches	Tyre wear for PC, LDV, HDV	Urban	TSP
Omstedt et al. 2005	Empirical model for daily mean concentrations	Road dust suspension	Urban (street canyon)	PM ₁₀
Rauterberg-Wulff 1998	Measurements at German roads	Tyre & brake wear for HDV and PC/2-wheelers	Urban (street canyon, tunnel)	TSP, PM ₁₀ , PM _{2.5}
Rauterberg-Wulff 2000	Measurements at German roads	Road dust suspension	Urban (street canyon)	TSP, PM ₁₀ , PM _{2.5}
Tønnesen 2005	Measurements at Norwegian roads	Road dust suspension		PM ₁₀ , PM _{2.5}
Johansson 2002	Measurements at Swedish roads	Road dust suspension	Urban (street canyon)	PM ₁₀ , PM _{2.5}
EEA 2003	Data compilation for Atmospheric Emission Inventory Guidebook	Tyre, brake abrasion, road dust suspension	None	TSP, PM ₁₀ , PM _{2.5}
Klimont et al. 2002	Data compilation for RAINS PM module	Tyre, brake and road abrasion for PC/LDV, HDV, 2-wheelers	None	TSP, PM ₁₀ , PM _{2.5}
TNO 2001	Data compilation for CEPMEIP	Tyre and brake wear for PC, LDV, HDV, 2-wheelers	None	TSP, PM ₁₀ , PM _{2.5}

Fine particulate matter emissions from stationary small combustion plants

Solid fuel combustion in small residential, commercial and institutional combustion plants is a significant source for fine particulate matter emissions. This source group consists of millions of small plants operated without flue gas cleaning and with a high share of old plants installed within the seventies or eighties and before. A plant inventory for Germany shows that in the year 2000 71 % of all solid fuel combustion plants were installed before 1990 (Struschka et al. 2003). Emissions of small combustion plants significantly depend on

- plant type (single stove, central heating, open fire etc.)
- fuel specification (ash, sulphur content, moisture, heating value, pellets/chips/wood)
- plant technology (old/conventional/modern/advanced, small/large, manual/automatic)
- plant operation (thermal load, air/fuel ratio)

Wood combustion plays a major role in air quality for large areas in middle and northern Europe. A significantly increasing activity rate is expected for the future because the use of regenerative energies is a target of climate policy. In addition, high oil and gas prices will result in an increasing use of wood as alternative fuel. Several recent studies can be found where emission measurement data was analysed and assessed and particle size factors were derived from size selective measurements at wood combustion plants. Table 3-5 shows collected examples of these studies that originate from UK, Switzerland, Austria, Germany and the USA. For an accurate emission calculation the plant inventory and the activity rates have to be determined and representative emission factors have to be derived from available measuring data from single plants. A detailed inventory for Germany was developed by (Struschka et al. 2003) based on comprehensive emission measurements, including wood, coal, lignite and oil combustion plants. Examples of PM emission factors for small wood combustion plants are shown in the Annex I in Tables 6-5 and 6-6 as result of a data review.

Table 3-5: Emission factors for small wood combustion plants

SOURCE	SOURCES, PLANTS	POLLUTANTS COVERED
Baumbach et al. 1999	3 commercial underfeed stokers, 2 residential wood stoves (pellets, logs), 1 fireplace; 8 small industrial underfeed stokers and grate firing systems (incl. flue gas cleaning)	PM, PM ₁₀ , PM _{2.5} , PM _{1.0}
Struschka et al. 2003	Measuring data for several plants (commercial/residential, manual/automatic, old/modern), complete inventory (stock, fuel consumption, emissions) for Germany	PM, PM ₁₀ , PM _{2.5} , PM _{1.0}
Mohn 2000	11 commercial plants: 6 automatic underfeed stokers, 4 automatic grate firing systems. 1 two-stage furnace	PM
AEAT 2001b	small industrial wood-burning furnace	PM, PM ₁₀ , PM _{2.5}
Spitzer et al. 1998	Summary/analysis of emission measuring campaign incl. 28 single wood stoves and 77 central heating systems (residential)	PM

SOURCE	SOURCES, PLANTS	POLLUTANTS COVERED
EPA 1995	Summary/analysis of available emission measuring data within AP-42	PM, PM ₁₀ , PM _{2.5}
Wieser & Gaegauf, 2000	2 manual wood stoves and 2 automatic stoves (pellets/chips) (residential)	PM, PM ₁₀ , PM _{0.4} (SMPS)
Wieser et al. 2001	1 fireplace, 2 manual stoves, 2 pellet stoves (residential); 2 underfeed stokers, 1 grate firing and 1 two-stage system (commercial)	PM, PM _{0.6} (SMPS)
Purvis et al. 2001	2 residential wood stoves (conventional/modern)	PM, PM ₁₀ , PM _{2.5}
AEAT 2004	Summary/analysis of available emission data from CITEPA and AEAT for different wood combustion systems (open/closed, conventional/advanced, manual/automatic)	PM, PM ₁₀ , PM _{2.5}
Gullett et al. 2003	Fireplace (oak, pine, artificial log)	PM
Fisher et al. 2000	16 residential wood stoves (5 catalytic and 11 non-catalytic stoves)	PM

3.2. Methodologies for the generation of emission data in spatial and temporal resolution

Emissions sources are spatially allocated to a region using the actual locations of the emissions sources, and/or using spatial surrogate data which are physical parameters that can be associated with emissions activities. Emissions sources are temporally allocated by assigning a temporal profile, e.g. a distribution of emissions activity over a 24-hour period, to each source category.

3.2.1. Spatial allocation of sources and emissions

High or low spatial resolution of emission data is one of the most important dimensions which characterize emission inventories. According to their source strength or to the degree of detailing of the inventory, emitters can be defined and spatially allocated as point, line or area sources.

Required spatial resolution

The smaller the region of interest is the finer the spatial resolution of the emission inventory needs to be. A characteristic of regional/continental emission inventories is the rather low spatial resolution. Emission data for Europe are usually provided on macro-scale (e.g. EMEP grid with 50 km x 50 km resolution) in order to model the regional and background emissions in a feasible way. On regional/national scale, emission data are often generated in meso-scale resolution below 10 km x 10 km so that a better distinction between urban and non-

urban areas and a better consideration of industrial and traffic-influenced areas can be done. For regional problems such as the Ozone (O₃) concentration a resolution of e.g. 4 km x 4 km is sufficient. When focusing on urban air quality, a higher spatial resolution of modelled emissions than the one chosen on regional scale has to be reached. A high resolution is required to simulate sources as well as ambient air concentrations in a complex and heterogeneous area. The spatial resolution of an urban inventory may range between 250 m x 250 m and 2 km x 2 km. Existing inventories often have a spatial resolution of 1 km x 1 km or 2 km x 2 km. To provide a micro-scale or a local scale assessment, an inventory with high spatial resolution is needed. A typical area under investigation is 1 km x 1 km or certain streets with a spatial resolution of some meters (e.g. 15 m x 15 m). Hotspot scale modelling is mainly carried out in single street canyons or near roads where exceedances of ambient air quality limit values mostly occur.

Allocation of point sources

The allocation of point source emissions e.g. from power plants, industrial sources, landfills and airports requires information on position (coordinates) and the effective height of stacks. Modelling point sources is often limited due to nondisclosure rules for industrial data. EPER data (EPER 2004) provide the location of 8.082 major sources in the EU, Norway and Hungary that can be used if national/regional industrial data is not available. The future reporting for PRTR will include as well point sources from further Eastern European countries. However, only those activities which are listed in Annex A3 of the EPER Decision are included, resulting in coverage of approximately 90 % of the emissions from industrial facilities (EN-TEC 2005). The data available was reported for 2001, the first reporting year for the EPER. Other industrial sources have to be considered as area sources.

Point source emission data should be linked to a vertical distribution/effective emission height, because this significantly affects the atmospheric dispersion. As EPER does not contain information required for the calculation of the effective emission height (stack height plus plume rise), general information has to be used that estimates the vertical distribution for different source groups. Plume-rise models or calculations of effective emission heights use stack height, temperature and exit velocity (s. e.g. German VDI guideline 3782 Part 3 (VDI 1985). The effective emission height depends on the downwind distance from the source, the emitted heat flux and the atmospheric stability (class).

If single source information is not available for the calculation of effective emission heights, sector estimations might be used for the vertical allocation to model layers. The vertical emission distribution used for the EMEP emission inventory is shown in Table 3-6 as an example. A distribution of the effective emission height is given for SNAP source groups level1.

Table 3-6: Distribution of gas and aerosols in the EMEP emission inventory according to different height levels based on the source sector (de Meij et al. 2006)

Source category (SNAP)	Height emission gases				Height emission aerosols			
	ground	~150m	~250m	high	ground	~150m	~250m	high
1. Combustion in energy and transformation industries			8%	92%	20%	20%	40%	20%
2. Non-industrial combustion plants	50%	50%			100%			
3. combustion in manufacturing industry	50%	50%			70%	7.5%	15%	7.5%
4. Production processes	90%	10%			100%			
5. Extraction of fossil fuel	90%	10%			20%	20%	40%	20%
6. Solvents	100%				100%			
7. Road transport	100%				100%			
8. Other mobile sources and machinery	100%				100%			
9. Waste treatment and disposal	80%	20%			100%			
10. Agriculture	100%				100%			
11. Nature	100%				100%			

Allocation of line sources

Mobile sources on the main road and rail network, ships and air traffic are treated as line sources. Traffic on major urban roads is often modelled based on traffic census data that is provided by a digitised traffic map. Main input for such models are automatic traffic counters. Results from manual traffic counting are sometimes used to specify traffic volume per different vehicle categories and approximate the fraction of heavy goods vehicle traffic. Emissions from traffic on side roads are often allocated according to the road lengths per grid cell. In a regional/continental inventory, road traffic emissions on highways/motorways can be allocated as well with the help of digitised road maps and the length of each road section. Urban road transport and traffic on smaller rural roads are usually allocated as area sources on EU scale using statistical data such as vehicle stock, mileage or population and land use data. On national level, a digitised road map might be available that includes as well annual traffic census data for each road section of highways and rural roads.

Allocation of area sources

Many small emitters (e.g. small stacks from residential combustion) can be treated as area sources. Surrogate factors should be identified for each area source category, used to spatially allocate the emissions. On EU scale, the spatial allocation of area source emissions to administrative units (NUTS 1, 2 or 3) can be done with the help of statistical data such as employees, land use or population (e.g. from EUROSTAT Regio database, see <http://epp.eurostat.ec.europa.eu>). Area sources can then be distributed using land use data (e.g. CORINE) with high spatial resolution distinguishing residential areas, industrial areas, agricultural areas, forests etc. The area source must be defined as a closed polygon line according to the location where the emission takes place (e.g. fugitive emissions on a waste disposal site) including the emission height. The accuracy of the boundaries of the area source should be 1‰ from the scale of the map used (Sturm & Winiwarter 2004). Area sources on urban/local scale are usually distributed using land use data with high spatial

resolution (e.g. 100 m x 100 m). In addition, data in further detail could be useful (e.g. data on fuel input or population per block/square of residential area). A much finer resolution is needed for micro-scale models in order to represent the location and emission behaviour as accurate as possible. But it has to be noted that for such applications a bottom up approach is necessary. A top-down approach (a disaggregation of statistical emission data) may not be sufficient.

Intersection of point, line and area sources with a defined emission grid

As described above, different geographical information is needed for the spatial allocation of country or regional total emissions depending on the source type. Emission data can be distributed in resolutions that are only limited by the availability and level of detail of proxy information used for distribution. Methods for allocation and grid intersection are shown for point, line and area sources in Figure 3-5. The geographic position of point sources is defined by the coordinates (x, y). Line sources can be intersected with the emission grid by vector data for e.g. road sections. On regional scale (macro-scale) and if no other information than section length is used for the allocation of road transport emissions, vector data of a road map can alternatively be converted into raster data. Thus, grid intersection is done according to area sources in a resolution of e.g. 1 km x 1 km. Two alternative methods are shown in Figure 3-5 for area sources (see last two pictures). The first shows the intersection of an administrative unit (polygon, e.g. NUTS level 3) with the emission grid. Annual emissions within the administrative unit are directly allocated to the grid cells according to the intersected areas. The second shows that a much higher accuracy can be obtained if the emissions of an administrative unit are allocated to a land use category (e.g. urban area) first. This can be realised by an intersection with a land use map (raster data, e.g. 1 km x 1 km resolution). In a second step, emissions allocated to land use area are intersected with the emission grid and allocated to grid cells.

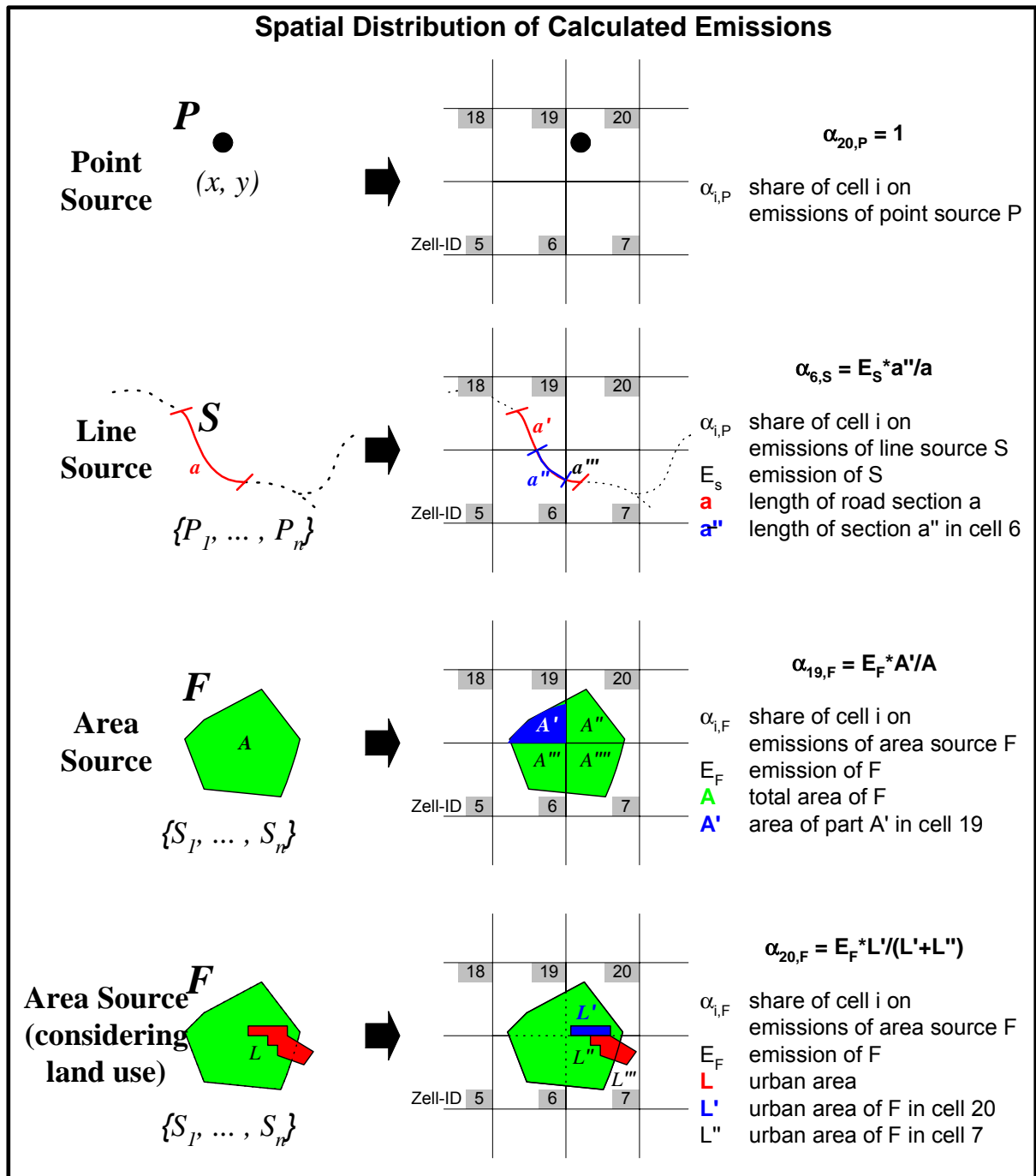


Figure 3-5: Methodologies for the spatial distribution of emissions (Wickert 2001)

3.2.2. Temporal resolution

Most of the activities which cause air emissions are time dependant. Normally, emission values as well as activity rates are averaged over longer periods such as a month or a year. In order to describe the emissions to air as a function of time of the day, week, month and year, source or process specific temporal profiles have to be generated and assigned to the sectoral annual emission data.

Temporal profiles are usually developed for and assigned to emission sources or source groups as hourly shares of annual emissions or averaged monthly, weekly and diurnal profiles in the following way:

- Monthly profile: share of annual emission for each month
- Weekly profile: share of weekly emission for each day
- 3 diurnal profiles: share of daily emission for each hour (working day, Saturday, Sunday)

Taking into account country specific or local conditions/information is often not possible on regional/continental scale due to limited resources. Source specific information that could be collected for one specific plant/process in one specific region usually has to be applied to other sources and regions. For most of the source categories, temporal profiles depend on the activity and the emissions follow the activity profile. Temporal profiles for some sources depend on climatic conditions (e.g. small combustions in households, gasoline evaporation, vehicle cold starts). Regional or local temperature data is often taken into account when generating temporal profiles for these sources. Not only in the case of temperature dependence, temporal profiles have a much higher accuracy if they consist of hourly shares of the annual emission i.e. 8760 or 8784 values for a year. In contrast, the use of average monthly, weekly and diurnal profiles can lead to high differences if hourly emissions are generated without considering seasonal variations of the weekly and diurnal profiles.

On urban and local scale, the temporal profiles for significant point sources might be generated based on diurnal and weekly operating schedules of the plants. Information on specific operation conditions such as holiday seasons, time of shifts or electricity consumption/production might be available and taken into account for large industrial sources as well as some area sources. For road traffic there often exist hourly or half hourly data on traffic volume from automatic counting on major roads or manual counting for selected episodes. These data can be used to derive general profiles for all comparable roads and episodes in an urban area and/or to directly calculate hourly emission data with a bottom up method. It might be not feasible to consider local conditions for some sources as e.g. small industrial/commercial processes. Available source specific information then has to be applied that has been collected in another region.

Figure 3-6 shows as an example hourly emission shares for the heat demand of small combustion plants in households. The use of average profiles is not recommended for this source group because of the significant temperature dependence. Methods used to generate temporal profiles for the IER GENEMIS emission model are described e.g. in (Friedrich et al. 2003). For the calculation of the heat demand from households as well as industrial/commercial

consumers it was assumed, that the required heating energy is proportional to the difference between indoor and ambient temperature. According to the German Guideline VDI 2067 heating only occurs when the daily mean temperature is lower than 12.5 °C and the highest energy demand for heating occurs at the design temperature of -15 °C. The dependency of heating energy demand E_{heat} from temperature is described as:

$$E_{\text{heat}}(T) = E_{\text{max}} * 100 / 35 * (20 - T) \quad \text{in [kWh] for } T \leq 12.5 \text{ °C} \quad (4)$$

$$E_{\text{heat}}(T) = 0 \quad \text{in [kWh] for } T > 12.5 \text{ °C} \quad (5)$$

The share of energy required for heating and additional energy demand that is not temperature dependent (e.g. process heat, water heating) should be known in order to include the temperature dependence for considered processes (e.g. coal fired small commercial combustion plants) in an accurate way.

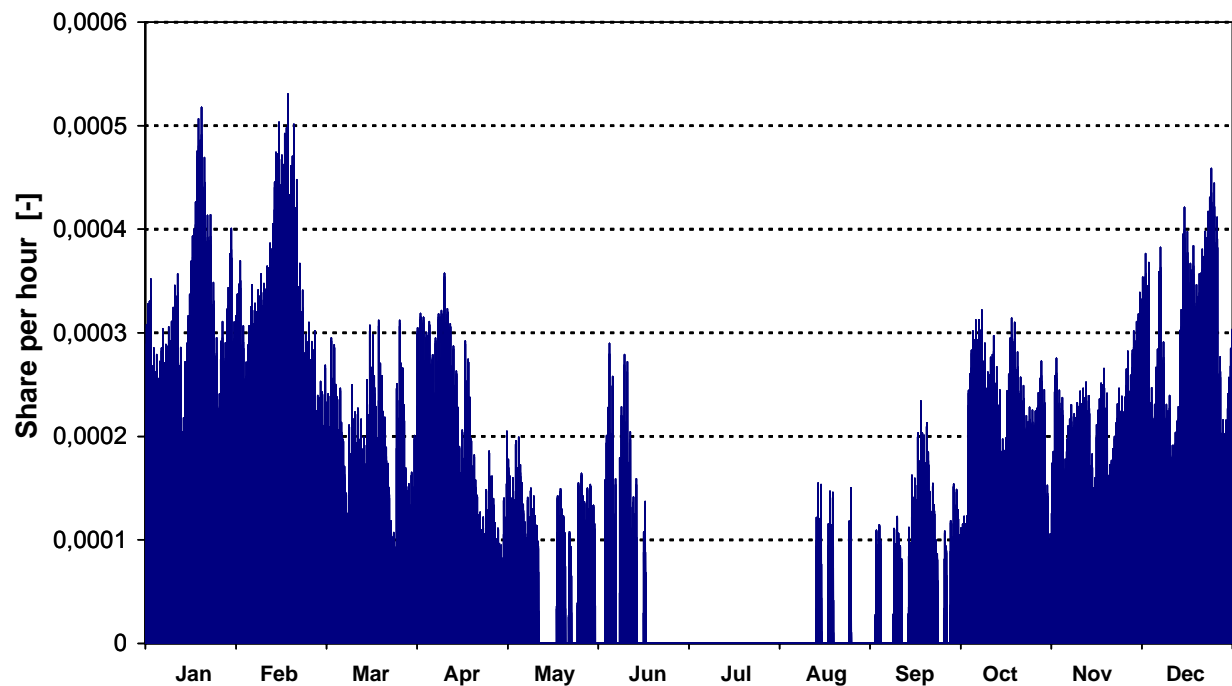


Figure 3-6: Temporal resolution of heat demand from households (based on Guideline VDI 2067 and measured temperature data of Augsburg in 1994)

In Figure 3-7 calculated hourly NMVOC emissions are shown based on the work of (Wickert 2001). Temporal profiles for hot emissions are based on hourly traffic census data for different vehicle types and road classes. Hourly temperature data of several meteorological stations and an average urban mileage profile for passenger cars and light duty vehicles were used for gasoline evaporation and excess cold start emissions.

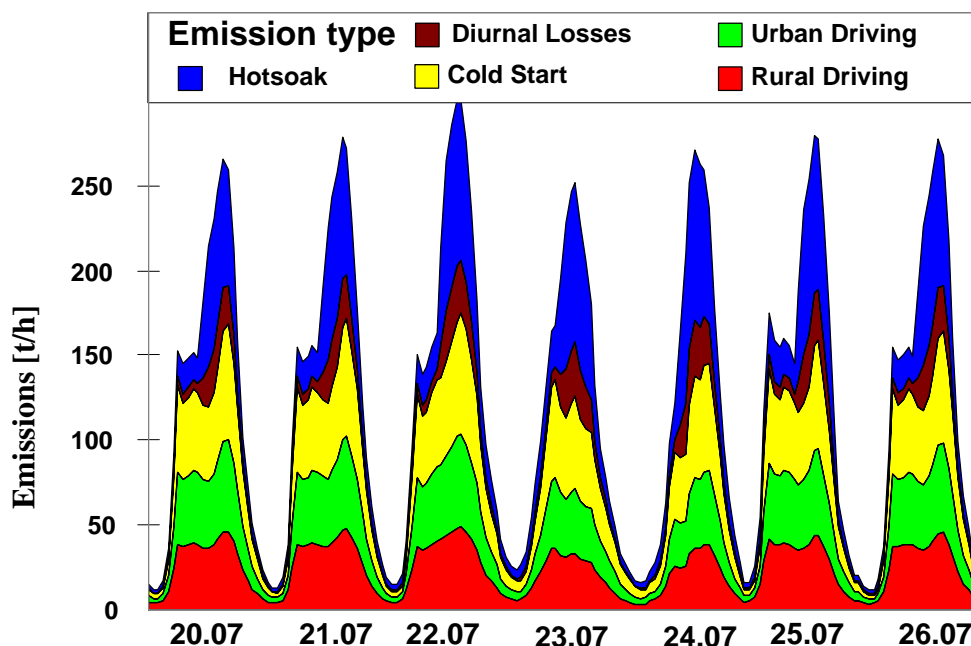


Figure 3-7: Hourly NMVOC emissions from road traffic for a typical week in July in Germany (Wickert 2001)

Table 3-7 shows information that is often used for the generation of temporal profiles. As far as possible, specific regional (e.g. national), urban or local information should be included. Local/regional hourly temperature data is required if processes are temperature dependent.

Table 3-7: Data need for the generation of temporal profiles

SOURCE GROUP	DATA NEED FOR PROFILE GENERATION
Road traffic – hot emissions	Hourly traffic census data for different vehicle categories, road classes, manual and/or automatic counts
Road traffic – cold start emissions	Hourly local/regional temperature data, hourly traffic census data for different vehicle categories and urban roads
Road traffic – gasoline evaporation	Hourly local/regional temperature data, hourly traffic census data for different vehicle categories and road classes
Railways, ships, air traffic	Train/ship/flight schedule information, manual counting
Power plants	Monthly, weekly, diurnal profiles of activities (e.g. energy consumption or production, load curves)
Industry	Monthly, weekly, diurnal profiles of activities (e.g. working days/hours, energy consumption, production volume)
Small combustion in households	User behaviour, hourly local/regional temperature data to calculate heat demand
Small commercial and institutional combustion, solvent use, other manufacture processes	Monthly, weekly, diurnal profiles of activities (e.g. working days/hours, energy consumption), hourly local/regional temperature data to calculate heat demand
Agriculture	Monthly, weekly, diurnal profiles of activities (e.g. working hours, energy consumption, seasonal variations, temperature, production cycles)

3.3. Application of emission data

3.3.1. Access to emission data (data, reports, documentations)

A broad range of information on emissions is accessible via internet (methodologies, tools, measured and modelled data, emission factors). Regional (EU/national) data is normally provided due to official reporting obligations, whereas urban or local data can be harder to gain access to. In the following chapter, examples for emission data on local, urban, regional, European and global scale are given.

Access to urban emission data

City Delta is a modelling exercise within the CAFE programme (Clean Air for Europe) to compare long-term model responses to emission reduction scenarios on urban scale. The pollutants in focus are particulate matter (PM₁₀, PM_{2.5}) and ozone; the participating cities are Berlin, Prague, Katowice, Milan, London, Paris and Copenhagen. Information about the city's emission inventories including spatial and temporal resolution is available at the website, where data can be downloaded (<http://aqm.jrc.it/citydelta/>).

As another urban example, the Digital Environmental Atlas of Berlin provides data and maps on local and urban scale sources and emissions (http://www.stadtentwicklung.berlin.de/umwelt/umweltatlas/edinh_03.htm, in English). Traffic related air pollution is covered for hydrocarbons, diesel particulates, NO₂ and benzene for 1993. Recent data and reports are given at the German version of the website and also at the Berlin Air Quality pages, where among others measurements and modelling on hotspot scale dealing with NO_x and PM₁₀ are illustrated (see link "Werkstatt Feinstaub").

Access to national emission data

With emission reporting obligations towards the UNECE (Geneva Convention on Long-Range Transboundary Air Pollution), the UNFCCC (United Nations Framework Convention on Climate Change) or the EU (e.g. the NEC - National Emission Ceiling - Directive), national emission data is mostly accessible via a country specific internet portal. Additional information concerning calculation methodologies, emission factors and uncertainties is often provided.

For the UK, NAEI (National Atmospheric Emissions Inventory) provides access to emission datasets and reports at <http://www.naei.org.uk/emissions/index.php> with emission factors, activities and maps by 1 km x 1 km resolution (see also Table 3-8). Annual national emission data is available since 1970, greenhouse gases since 1990. Formats used for the sectoral resolution are NFR (Nomenclature for Reporting) and CRF (Common Reporting Format). Emission factors for the different sectors are derived from UK based measurements and experts estimates. Uncertainties in the national emission inventory are described in (Passant 2003).

CITEPA (Centre Interprofessionnel Technique d'Etudes de la Pollution Atmosphérique, http://www.citepa.org/emissions/index_en.htm) covers national annual emissions for mainland France and the overseas areas for SO₂, CO, CO₂, NH₃ and NMVOCs since 1990. Further substances and previous years are available for mainland France only. A more de-

tailed spatial resolution is provided down to Arrondissements and sixty urban areas for 1994 and down to Départements for 2000. Sectoral resolution is presented according to SECTEN (Secteurs Economiques et Energie, comprising seven economic sectors) and other official formats such as UNECE, EMEP or UNFCCC. An overview of inventory results for different pollutants is given in English and can be viewed on the page “national annual data”; the full report in French contains more information on figures, sectors and uncertainties (CITEPA 2005).

In Germany, the Federal Environmental Agency (UBA) provides access to emission inventories at <http://www.umweltbundesamt.de/emissionen/publikationen.htm> (in German and English). Annual national emission data is available since 1990, sectors are presented in CRF and NFR formats. Within UBA, the Central System of Emissions (CSE) is used for centralised data storage of all detailed information required for emission calculation such as methods, activities and emission factors. CSE is not accessible via the internet. A more detailed spatial and temporal resolution is provided by each federal state individually (e.g. the emission inventory of Bavaria see <http://interl.bayern.de/emissionskataster/php/start.php>).

Access to EU-wide emission data

On European level, EMEP offers the emission database WEBDAB (<http://webdab.emep.int/>, see also Table 3-8) that consists of officially reported annual emissions of the UNECE CLRTAP member states and additional expert estimations. So far, uncertainties are not considered. Publications about emission data and other background reports can be downloaded at http://www.emep.int/index_facts.html.

The CAFE programme (Clean Air for Europe) is set up by the European Commission to develop the Thematic Strategy on Air Pollution, meaning that measures to improve European air quality are assessed regarding their potential for further improvements beyond present legislation. The baseline projections for CAFE are provided by the IIASA and used for their model RAINS (Regional Air Pollution Information and Simulation) (see <http://www.iiasa.ac.at/rains/> and Table 3-8).

TNO (Netherlands Organisation for Applied Scientific Research) supplies in CEPMEIP (Coordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance) a European emission inventory for particulate matter (PM_{2.5}, PM₁₀ and TSP) in 1995. Emission data are available at SNAP level 1 and 2 for all known anthropogenic sources (<http://www.air.sk/tno/cepmeip/>, see Table 3-8).

Access to global emission data

Also on global scale, efforts are made to develop emission inventories. EDGAR (Emission Database for Global Atmospheric Research) is an emission inventory for direct and indirect greenhouse gases including halocarbons and aerosols from anthropogenic sources (s. <http://www.mnp.nl/edgar/>). In EDGAR 3.2, emissions such as CO₂, CH₄, N₂O, HFCs, PFCs, SF₆ and the precursor gases CO, NO_x, NMVOC, SO₂ are accessible for 1990 and 1995 (Olivier and Berdowski 2001). Spatial resolution is given per country (for fossil fuel and biofuel sources, industries) and on a 1 x 1° grid (for landuse sources, partially also available per country, and for natural sources). A recent dataset, EDGAR 3.2 Fast Track 2000, gives estimates of the abovementioned gases for the year 2000 based on the 1995 dataset and trend analyses per country and sector (in CRF).

In the joint EU project POET (Precursors of Ozone and their Effects in the Troposphere) several global emission inventories (e.g. EDGAR) are brought together to assess annual emissions of Ozone precursors on a 1 x 1° grid (<http://nadir.nilu.no/poet/> and <http://www.aero.jussieu.fr/projet/ACCENT/POET.php> for data retrieval). Anthropogenic emissions are available for 1990-2000, scenarios are calculated up to 2020 (Olivier et al. 2003).

For the project RETRO (Reanalysis of the tropospheric chemical composition over the past 40 years), global anthropogenic and vegetation fire emissions of the trace gases NO_x, NMVOC, CO, SO₂ (among others) were generated. Data sets cover the years 1960-2000 with monthly time resolution on a 0.5 x 0.5° grid (<http://retro.enes.org/emissions/index.html>).

GEIA (Global Emissions Inventory Activity) is a scientific network of the IGBP (International Geosphere-Biosphere Program) to develop inventories of global gas and aerosol emissions from natural and anthropogenic sources. On their data portal (<http://www.geiacenter.org/>), GEIA and ACCENT (Atmospheric Composition Change, EU Network of Excellence, http://www.accent-network.org/farcry_accent/) provide gridded data collected from several publicly available inventories (POET, RETRO, GEIA version 1, and soon IIASA). Emissions such as ozone precursors, greenhouse gases, organo-halogens, several heavy metals, aerosols and their precursors are available in the categories total anthropogenic, total biomass burning, biogenic, and oceans. Spatial and temporal resolution varies depending on the data source and covers global or regional data on a 1 x 1° and a 0.5 x 0.5° grid per season and year for 1960-2000.

The joint initiative AEROCOM (Global Aerosol Model Intercomparison) was set up to study global aerosols and their impact on climate and to compare several global aerosol models. An emission inventory for 2000 and pre-industrial years (representing anthropogenic emissions in 1750 to quantify anthropogenic forces) is available at its website (<http://nansen.ipsl.jussieu.fr/aerocom/>). There, datasets on fossil fuel and biofuel related organic and black carbon (Bond et al. 2004), sea salt (Gong et al. 2003), DMS (dimethyl sulfide), SOA (secondary organic aerosol) and others cover a 1 x 1° grid. Temporal resolution is one year, in some cases (e.g. DMS) one day.

3.3.2. Applications on regional/EU scale

Several data bases for annual emissions exist on European scale (see Table 3-8). EMEP WEBDAB (EMEP 2004) provides annual emission data for 50 countries as country totals reported by the Parties to the Convention on Long-Range Transboundary Air Pollution (CLRTAP) or estimated in addition by (Vestreng 2003). EMEP data base includes annual emissions for the year 1980 to 2002. TNO CEPMEIP data base contains activity data, emission factors and resulting particulate matter emissions for the base year 1995 (TNO 2001). The MERLIN emission data base (base year 2000 and projection for 2010) was developed for 27 European countries as basic input for an integrated assessment model that calculates emissions, achievable reduction and abatement costs for each combination of technology and measure (IER 2005). The calculations are based on a unique data set of unabated emission factors. Resulting abated emission factors and emissions differ from officially reported data. Internal and external validation is continuing. Emission data for the RAINS model of IIASA (IIASA 2004) were generated for 30 countries as basic input for the analysis of reduction strategies with a detailed approach. Emission data are available for the base

year 2000 and projections for 2005, 2010, 2015, 2020. The TNO emission data base used for LOTOS-EUROS chemistry-transport model is based on EMEP country totals, EPER point sources and additional calculations and point source information collected by TNO. Natural/biogenic emissions are calculated online within the CTM.

Table 3-9 lists emission tools that were developed to generate highly resolved emission data for regional/EU scale air quality modelling. Methodologies for the calculation of sectoral emissions may be quite different within each inventory. The choice of emission factors is not harmonised. Also methods for and quality of spatial allocations and temporal distributions might be quite different. A crucial point for the accuracy of emission modelling on meso-scale is an accurate representation of line sources for road traffic and the availability and implementation of industrial point source data. In addition there exist gridded 1 x 1 ° global emission data over Europe with low spatial resolution from the EDGAR project (Olivier & Berdowski 2001), often used to fill in data gaps for the eastern part of Europe.

Table 3-8: Examples of EU scale sectoral emission data bases

EMISSION DATA BASE	SECTORAL RESOLUTION	CALCULATION METHODOLOGY	POLLUTANTS COVERED
EMEP WEBDAB EMEP 2004	Officially reported emissions for 87 source groups UNECE NFR level 2. Expert estimations for SNAP level 1 source groups	Officially submitted data by the parties to the CLRTAP to the EMEP project via the UNECE secretariat. In addition expert estimations to fill gaps (see Vestreng 2003).	SO ₂ , NO _x , CO, NH ₃ , NMVOC, PM, PM ₁₀ , PM _{2.5} , several heavy metals and POP
LOTOS e.g. Schaap et al. 2004 Bultjes et al. 2003	14 aggregated source groups used as model input, SNAP level 1 incl. natural/biogenic + traffic subdivided into diesel exhaust, gasoline exhaust and fuel evaporation	EMEP data and TNO calculations with activity data and emission factors, TNO CEPMEIP data for particulate matter. TNO point source database supplemented with EPER data. Natural emissions are calculated online.	CH ₄ , SO ₂ , NO _x , CO, NH ₃ , NMVOC, PM, PM ₁₀ , PM _{2.5}
TNO CEP-MEIP TNO 2001	~235 detailed or ~30 aggregated source groups SNAP level 2	Officially submitted national data and TNO calculations based on activity rates and abated emission factors.	PM, PM ₁₀ , PM _{2.5}
IER MERLIN IER 2005	~255 detailed or 10 aggregated source groups SNAP level 1	Based on activity rates, unabated emission factors, measure efficiencies and implementation degrees or abated emission factors for each technology and measure, country and year.	CO ₂ , CH ₄ , N ₂ O, SO ₂ , NO _x , CO, NH ₃ , VOC, Benzene, PM ₁₀ , PM _{2.5}
IIASA RAINS Baseline Emissions IIASA 2004	~240 detailed or ~45 aggregated source groups UNECE NFR format 2	Based on activity rates, unabated emission factors, removal efficiency, abated emission factor and capacities controlled for detailed source groups and year. Data was aligned to officially reported EMEP emission data.	SO ₂ , NO _x , NH ₃ , VOC, PM, PM ₁₀ , PM _{2.5}

Table 3-9: Methodologies for the spatial disaggregation - examples of national/EU scale emission models

EMISSION DATABASE	AREA SOURCES	LINE SOURCES	POINT SOURCES
EU SCALE			
LOTOS e.g. Schaap et al. 2004	NUTS data, high resolution population density maps, CORINE land use data	Digital road maps of mayor roads and traffic intensity	Point source coverage built over the years from various questionnaires and supplemented with EPER data
IER GENEMIS for Europe (without Germany) Wickert 2001 Schwarz 2001	NUTS 0 to 3 currently using (extrapolated) CORINAIR 94 emission data and/or statistics; 1 km x 1 km land use data of CORINE and USGS	Digital road maps (~118000 road sections of motorways, national highways) and railway net converted into 1 km x 1 km land use data	Point source coverage based on CORINAIR 90/94 (5466 LPS)
EMEP Vestreng 2003	50 km x 50 km gridded emissions reported by countries. Additional expert estimations based on population maps and distribution maps from related pollutants	50 km x 50 km gridded emissions reported by countries. Additional expert estimations based on population maps and distribution maps from related pollutants	50 km x 50 km gridded emissions reported by countries. Additional expert estimations based on CORINAIR 90 point source coverage (5823 LPS)
NATIONAL SCALE			
NAEI UK AEAT 2001a	Based on surrogate statistics including population and land use data	Annual average daily flows on a link by link basis on major roads, railway tracks and vehicle kilometres data	Point sources based on partly accessible industrial data
IER GENEMIS for Germany Wickert 2001	NUTS 3 using statistical data; 1 km x 1 km land use data of CORINE	Digitised road net incl. traffic census data (~83000 road sections of motorways, national and other highways)	Point source coverage based on industrial data (~48000 point sources in 9 of 16 Federal States), in addition CORINAIR 90/94 data (357 LPS)

In the UK for example, the National Atmospheric Emission Inventory (NAEI) has been developed with funds from the Department for Environment Food and Rural Affairs (DEFRA), the National Assembly for Wales, the Scottish Executive and The Department of Environment, Northern Ireland. The atmospheric emissions have been developed for various UK sources, e.g., cars, trucks, power stations and industrial plant. Figure 3-8 shows an example of the gridded annual NO_x as NO₂ emissions for the entire United Kingdom from NAEI.

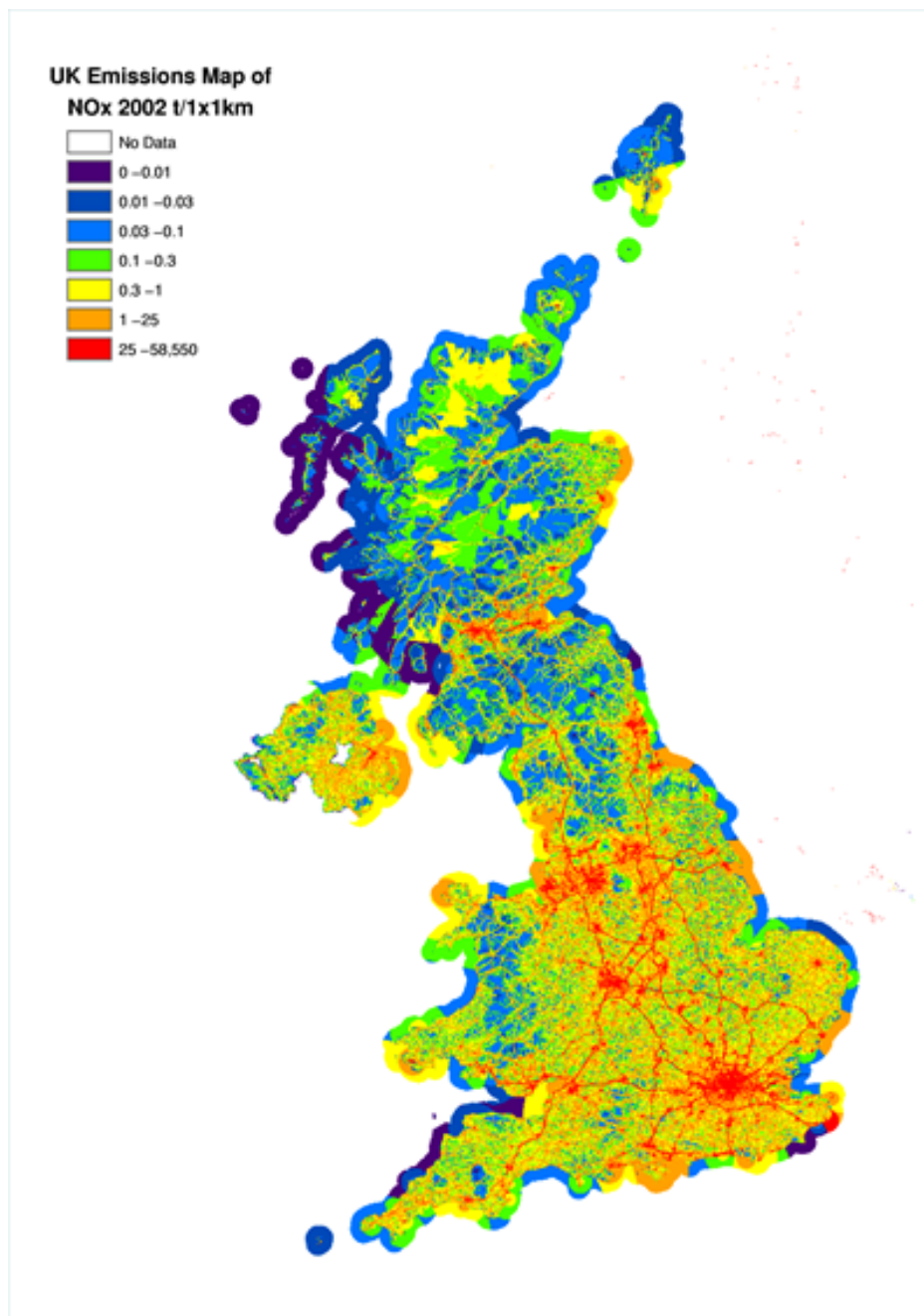


Figure 3-8: Annual NO_x emissions (2002), 1×1 km grid spacing, from NAEI (http://www.naei.org.uk/mapping/mapping_2002.php).

3.3.3. Applications on urban/agglomerate scale

Table 3-10 lists some examples of urban scale emission models/inventories. As an example, the detailed Stuttgart emission inventory of 1996 (UMEG 1998) could be based to a large extent on local information. Industrial emission declarations for 416 licensed plants were included as well as an additional data collection for 1955 not licensed commercial plants such as printing, wood processing, metal processing, gasoline distribution, paint-spray lines, chemical cleaning etc. Road traffic emissions were modelled based on 3453 major road sections and 1117 side road sections, 31 vehicle categories, 12 road classes and emission fac-

tors from HBEFA. Emissions from small combustion were calculated based on energy demand distinguished between gas, oil, wood and coal and fuel specific emission factors. Energy demand was modelled based on information on building use and heating structure (central heating, single stove) from urban administration, statistics of the chimney sweepers and energy suppliers.

Table 3-10: Examples of urban/agglomerate scale emission inventories and models

CITY	SOURCE GROUPS	POLLUTANTS COVERED	SPATIAL RESOLUTION	TEMPORAL RESOLUTION
Milan	11 SNAP sectors	SO ₂ , NO _x , CO, NH ₃ , NMVOC, CH ₄ , PM, PM ₁₀ , PM _{2.5}	5 km x 5 km + point sources	2772 profiles (4 seasons, 3 day-types, 7 pollutants, 3 climatic classes, 24 hours)
Berlin	5 SNAP sectors (1, 2, 3, 4, 7)	SO ₂ , NO _x , CO, NMVOC, CH ₄ , PM, PM ₁₀ , PM _{2.5}	2 km x 2 km / 1 km x 1 km + 620 point sources (2000)	1008 profiles (12 months, 7 days, 24 hours)
London	10 SNAP sectors	SO ₂ , NO _x , CO, NMVOC, PM, PM ₁₀	1 km x 1 km + point sources	420 profiles (12 months, 7 days, 24 hours, 4 pollut.)
Paris	Point, area, line sources	SO ₂ , NO _x , CO, NH ₃ , NMVOC, CH ₄ , PM, PM ₁₀ , PM _{2.5}	3 km x 3 km	18 profiles (6 months, 3 day-types)
Stuttgart (agglom.)	9 SNAP sectors	SO ₂ , NO _x , CO, VOC, PM, HCl, HF	1 km x 1 km + 3289 point sources (1996)	-

The Air4EU cities Oslo, Paris, Prague and Rotterdam have developed individual approaches for the generation of their emission inventories that are due to varying source contributions, different levels of data availability and types of air quality models that require emissions as an input.

Point, line and area sources were considered for Oslo's emission inventory as well. Point sources are evaluated by using consumption data, production data (and emission factors) or by using emission data directly from the national database of Statistics Norway, available at www.ssb.no/english. Parameters such as stack height, diameter, flue gas temperature and velocity for each individual high stack are further input in the emission generation and dispersion modelling. Overall, however, emissions from point sources contribute very little to total emissions in Oslo. Line sources are evaluated by traffic counts (for road links) in combination with slope, speed and vehicle types. Data bases for traffic emission factors are COPERT III and national Norwegian sets. Suspended PM is calculated in an additional model taking into account road conditions and the number of automobiles with studded tyres (daily average limits for PM₁₀ are exceeded especially during winter and spring). Results for non-exhaust emissions are considered to be uncertain, though. Area sources include traffic with an ADT

(average daily traffic) of less than 3000, home heating and shipping. A lot of homes in Oslo are heated with wood which contributes to PM₁₀ exceedances (firewood for home heating is increasingly used also in other European cities, e.g. Rotterdam). Emissions from wood burning are estimated with the help of wood consumption data and an emission factor for Oslo from Statistics Norway.

AIRPARIF set up an emission inventory for Paris in 2000 (last update in 2004) that considers point, line and area sources in a 1 x 1 km resolution. Declarations made for the Polluting Activities General Tax are taken as a basis for emission calculation of major point sources, the other industries are modelled with the help of fuel consumption factors per employees. The regional model takes into account plume rise (by the parameters exhaust velocity and temperature and stack heights) to distribute emissions vertically. Line sources are traffic-related and the emissions are calculated based on real-time observed traffic data (with a system that was developed within the project HEAVEN). 35000 road segments are considered to provide information on traffic emissions for the Paris and Ile-de-France road network, traffic on minor roads is considered with a simple approach (aggregated on fictive roads). The system is updated every hour. Emission factors are taken from COPERT III, suspension of particulate matter is not included. Other emission sources that are part of the Paris emission inventory are residential heating, agriculture, forestry and biogenic contributions (all treated as area sources) and other mobile sources (including Roissy and Orly airport as area and point sources).

Point sources in the emission inventory of Prague are represented by measured emissions for the largest stacks corresponding to the Czech classification REZZO 1 and calculated emissions for smaller point sources corresponding to REZZO 2 (the calculations are based on fuel consumption and CORINAIR emission factors). Combustion characteristics are described by stack height and diameter, flue gas temperature, flow rate and velocity. Source specific information for that is provided by the plant operators and validated by the Czech National Environmental Inspectorate. Temporal variation of emissions from large combustion sources is accounted for by annual profiles. In addition to that, some sources have weekly, daily, shift or irregular profiles, highest temporal resolution is on a daily basis. Similar to Oslo, traffic as classified in REZZO 3 is modelled as line source for major roads and area source for minor roads. Emission factors for traffic are taken from the Czech data base MEFA and are depending on the catalytic type, speed, slope and year. They are, however, considered to be too low. A cross-check with COPERT III emission factors has not been undertaken. Non-exhaust PM emissions from suspension are not included in the modelling. Area sources include small combustion and are allocated using land use data and population depending on ambient temperature and season.

In Rotterdam and the Rijnmond area, point sources such as industrial stacks and refineries are important emitters but contribute little to air quality due to their vertical distribution. Large plants have to report to national and regional authorities, so information is available in detail. Small industries also have to report, but in order to avoid expensive accurate monitoring some plants report the maximum emission that is in their operation license. So emissions could be overestimated. Activity data for traffic emissions is derived from counts (by loops in highways) and traffic flow models with origin/destination matrices for inner-urban driving. Emission factors are taken from the Dutch national data set. However, vehicle speed, fleet

composition, the amount of congestion and PM from suspension are still uncertain. As Rijnmond contains one of the largest ports in the world, emissions from ships are another important source although exact figures, especially for PM, are barely known. Likewise, emissions from transshipment, from storage of coal and ore (PM), from leaking (VOCs) and from the container terminals (NO_x) are hard to quantify.

For London, some local authorities, such as Greater London Authority (GLA), have developed a regional emissions database, called London Atmospheric Emission Inventory (LAEI). The LAEI emissions are annual emissions, updated annually considering the yearly changes in the economy, technology, environmental legislation and others. The following figures show the horizontal variations of annual NO_x emissions (Figure 3-9) and annual PM₁₀ emissions (Figure 3-10) across Greater London for year 2001 with a fine resolution of 1 km grid spacing. The figures are produced based on the summation of emissions from all emission source types.

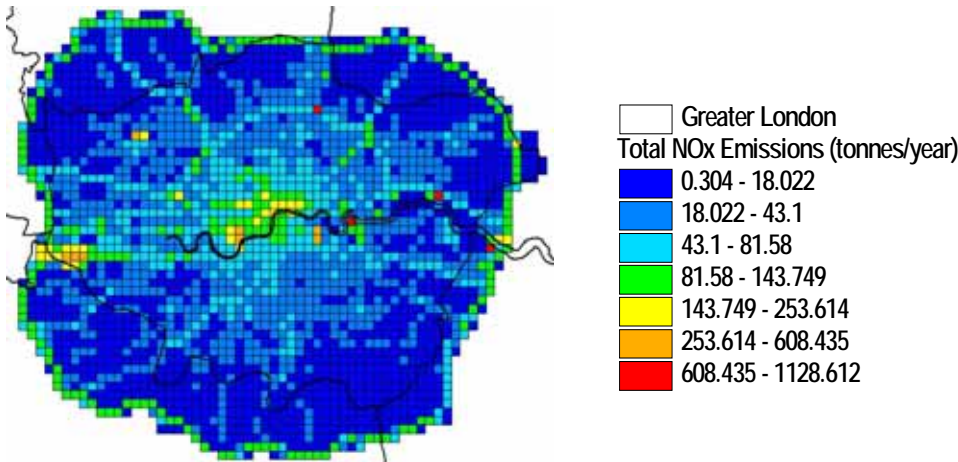


Figure 3-9: Annual gridded NO_x emissions (2001), 1×1 km grid spacing, developed by LAEI over Greater London.

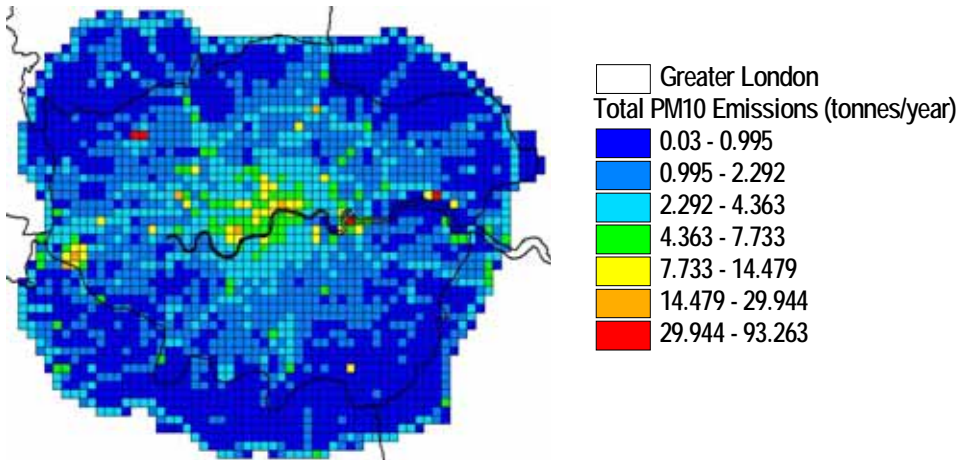


Figure 3-10: Annual gridded PM₁₀ emissions (2001), 1×1 km grid spacing, developed by LAEI over Greater London.

The emission databases mentioned above has been widely used in urban scale air quality modelling studies in the UK (i.e. Luhana et al. 1998; Kitwiroon et al. 2002). These atmospheric emission databases developed by NAEI or LAEI are available to public. However, the emission inventories are generally available with an annual-total emissions value for each emission source and are not in a model ready format. Air quality models (AQMs) typically require emissions data on an hourly basis, for each model grid cell (and perhaps model layer), and for each model species. Consequently, the raw emissions inventories are generally processed using emissions data processing system to transform an emission inventory through temporal allocation, chemical speciation, and spatial allocation, to achieve the input requirements of the AQMs. Some emissions data processing systems have been developed to meet the requirement of different AQMs. For example, SMOKE (Sparse Matrix Operator Kernel Emissions) has been used to prepare emission input files for CMAQ (Community Multiscale Air Quality modelling system) (Qiu et al. 2001) and EMIMO (EMission model for MA-drid area) has been used in ANA (Atmospheric Mesoscale Numerical Pollution Model for regional and Urban Areas) (San Jose et al. 1998) but users are responsible for providing the appropriate temporal profile for temporal allocation and surrogate files for spatial allocation for these processing systems. Usually point, area and line sources are treated separately in these emission processing system. The purpose of any emissions processor is to convert the resolution of the emission inventory data to the resolution needed by an air quality model. However, for mobile and biogenic sources, some processors, like SMOKE do offer emission inventory preparation functions. For mobile sources, SMOKE computes an emission inventory from mobile-source activity data, using emission factors from the MOBILE6 model. For biogenic sources, SMOKE includes both the BEIS2 and BEIS3 models for computation of hour-specific, meteorology-based biogenic emissions from vegetation and soils.

Apart from using existing emission inventories, some air quality models or modelling systems have their own emission database or emission inventory set up tool. For example, the Airviro dispersion simulation system uses the dynamic emission database (EDB) with the EDB handles time and temperature variations in emission rates from point, area and line (traffic) sources and ADMS-Urban (McHugh et al., 1997a, 1997b) imports emissions data from CERC's Emissions Inventory Toolkit, called EMIT which sets up emission inventory using activity data and the imbedded emission factor database. The Norwegian AirQUIS integrated environmental information system (<http://www.nilu.no/airquis/>) is an advanced example of an air quality management tool including an emission module and model that enables to calculate/implement detailed point, line and area emission data and to provide hourly emission data in spatial resolution.

As can be seen from these examples, cities normally have access to a lot of data for their emission inventories, but as they usually come from a lot of sources, they have to be checked for accuracy and consistency to avoid mistakes and double-counting. Furthermore, some processes and sources still bear high uncertainties (e.g. suspension of road dust, shipping emissions, wood burning) and are therefore hard to assess. More information on emission modelling on urban/agglomerate scale can be found within the final reports of the EUROTRAC-2 subprojects GENEMIS (Friedrich & Reis 2004) and SATURN (Moussiopoulos 2003).

3.3.4. Applications on local/hotspot scale

Emission modelling at local scale is mainly done within research projects to improve or validate emissions data as well as micro-scale dispersion modelling. Local/regional authorities usually do not use micro-scale modelling approaches for air quality assessment at hotspots but analyse monitoring data to determine contributions of road traffic, fossil fuel combustion etc. HBEFA and COPERT III are often used as data sources for emission factors and other information for road traffic. Table 3-11 lists as examples two different methodologies that were applied to generate emission data on local/hotspot scale from road traffic. Whereas the BEBAS-methodology represents best practice used within a scientific validation project, TREM is an emission model with a more feasible approach that is used for air quality modelling.

Within the EU project OSCAR (Optimised Expert System for Conducting Environmental Assessment of Urban Road Traffic) an optimised Expert System was developed to assess the environmental impact of road traffic in terms of traffic flows, emissions and air pollution. OSCAR aims for providing realistic estimates of urban/local congested conditions and resulting emissions (s. <http://www.eu-oscar.org/>).

Figure 3-11 shows an example of a small scale inventory of road traffic emissions around the Goettinger Strasse in Hanover based on local traffic census data. The grid covers an area of 1 km x 1 km with a resolution of 15 m x 15 m. Daily NO_x emissions were calculated based on HBEFA emission factors. Other comprehensive local scale analyses were done e.g. for the Marylebone Road in London and the Hornsgatan in Stockholm (see Air4EU D3.1 Review of Methods for Assessing Air Quality at the Local/Hotspot Scale and Johansson et al. 1999).

Table 3-11: Examples of methodologies to generate emission data from road traffic on local/hotspot scale

EMISSION MODEL/STUDY	SHORT DESCRIPTION	EXAMPLE APPLICATION
TREM TREM 2002	Transport Emission Model for Line Sources (TREM): developed at the University of Aveiro to support quantification of hot and cold emissions induced by road traffic, with high temporal and spatial resolution. The model is based on emission factors per road class and vehicle type, engine type and capacity, model year and average speed, based on MEET/COST methodology and is implemented in Geographical Information System environment.	Lisbon Metropolitan Area
BEBAS IER 2004	Calculation of High Resolution Emission Data for a Conurbation and Street Canyons (IER project): site specific traffic volume data half-hourly (inductive loops and video recordings), speed and acceleration measurements (Laser Traffic Speed Meter), analysis of license plate numbers (vehicle fleet composition). Emission calculation with emission factors of HBEFA (UBA 2004) per vehicle category and capacities/weights, EURO emission standard, road class, slope and driving conditions. In addition, use of emission functions for present vehicle driving conditions (speed, acceleration and vehicle mass). Calculation of hot, excess cold start and evaporation emissions based on local information.	Hanover (Göttinger Straße)

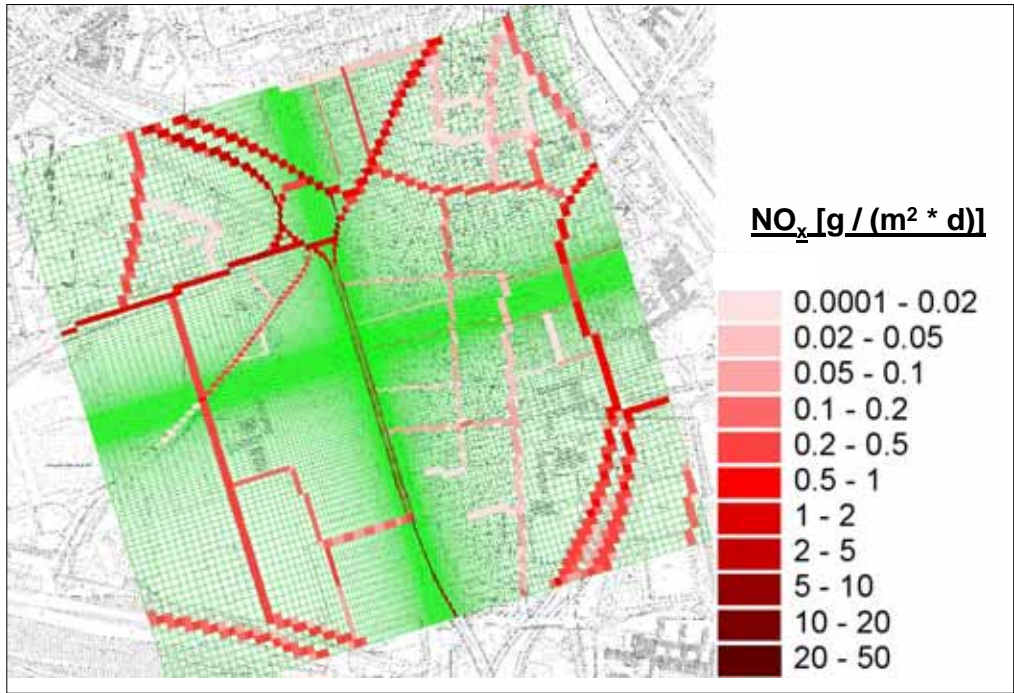


Figure 3-11: NO_x emission rates as average daily values for working days on small scale grid around the Goettinger Strasse (IER 2004)

Figure 3-12 shows results of a local bottom up emission calculation in a 30 min temporal resolution based on manual traffic counting (IER 2004). The vehicle fleet and driving conditions could be characterized in high level of detail. Vehicle mileage data were linked with emission factors from HBEFA (UBA 1999).

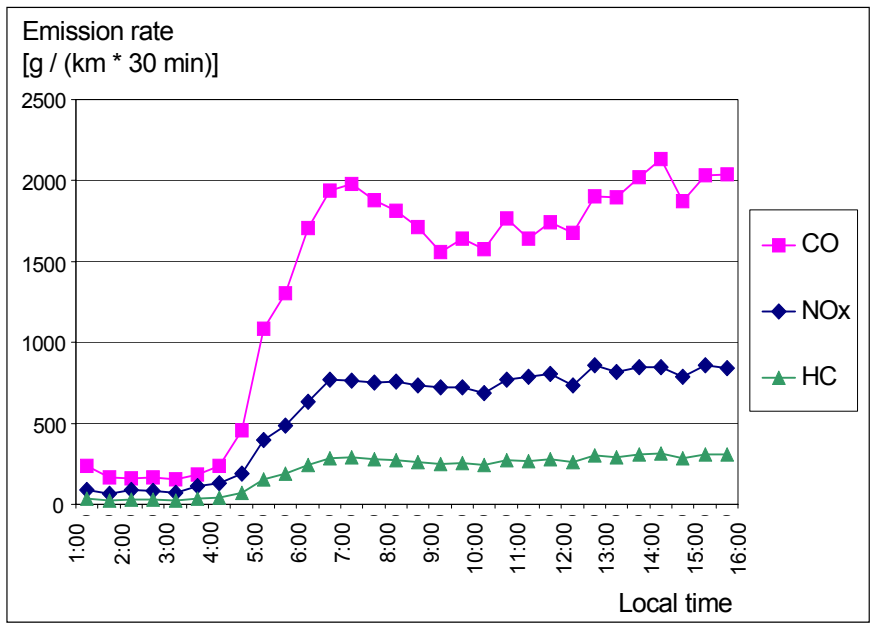


Figure 3-12: Temporal resolution: Modelled emission rates at Goettinger Strasse on Wednesday, August 8th 2001 (IER 2004)

3.4. Quality assessment and uncertainty analysis related to emission data

Uncertainties in emission models typically originate from a number of different causes. One of the sources of uncertainty is the modelling approach itself as it assumes to describe the real process by using several assumptions and may be based on incomplete knowledge about the emission process itself. Another source of uncertainties is input data used by the emission model (statistics, emission factors, activity rates). Also, natural variability in the process can significantly contribute to the total uncertainty (Borrego & Tchepel 1999).

Total uncertainty = Model uncertainty + Data Uncertainty + Variability

The variability is inherent to the processes that produce emissions. If all other sources of uncertainty were removed, the natural variability would still make it impossible to make precise emission estimations at a certain point in time and space (EIIP 1996). There are two major components of the variability: (i) spatial component that introduced by variation from source to source and (ii) temporal component that could be observed within the same source during some time period. An incomplete understanding of the variability in a process can lead to systematic errors in the estimation (Borrego & Tchepel 1999).

3.4.1. Documentation and archiving

It is good practice to document and archive all information required to produce emissions inventory estimates. For example, IPCC has defined several requirements as important precondition for uncertainty analysis in GHG inventories. According to (IPCC 2000a) documentation and archiving should include:

- Assumptions and criteria for selection of activity data and emission factors;
- Emission factors used, including references for default factors or to published references or other documentation for emission factors used in higher tier methods;
- Activity data or sufficient information to enable activity data to be traced to the referenced source;
- Information on the uncertainty associated with activity data and emission factors;
- Rationale for choice of methods;
- Methods used, including those used to estimate uncertainty;
- Changes in data inputs or methods from previous years;
- Identification of individuals providing expert judgement for uncertainty estimates and their qualifications to do so;
- Details of electronic databases or software used in production of the inventory, including versions, operating manuals, hardware requirements and any other information required to enable their later use;

- Worksheets and interim calculations for source category estimates and aggregated estimates and any re-calculations of previous estimates;
- Final inventory report and any analysis of trends from previous years;
- QA/QC plans and outcomes of QA/QC procedures.

Documentation is one important precondition for uncertainty analysis, validation and verification and should consist of records that are unambiguous. Documentation should be maintained for every annual inventory produced and provided it for review. It is good practice to include the checks/audits/reviews that were performed, when they were performed, who performed them, and corrections and modifications to the inventory resulting from the QA/QC activity (IPCC 2000a). Additional requirements for documentation and quality assurance can be found in (IPCC 2000a). Guidelines for estimating and reporting emission data under the Convention on Long-range Transboundary Air Pollution can be found in (UNECE 2003).

3.4.2. Quality Assurance / Quality Control

One of the approaches to estimate and to reduce the uncertainties is the Quality Assurance / Quality Control (QA/QC). QA/QC procedures play an increasingly important role in environmental studies, especially when those studies are conducted to support decision in environmental problems. QA/QC procedures are designed to ensure that the appropriate methods and data are used, that errors in calculations and measurements are minimised, and that documentation is adequate to meet the project objectives. Focus has to be placed on five specific goals: transparency, consistency, comparability, completeness and accuracy (IPCC, 2000). Striving for all five of these qualities allows for meaningful comparison of data submitted by parties.

Planning of QA/QC procedures assure development of the highest possible quality results. The first step in planning any environmental project is to define the purpose and intended use of the results. It is important to realise that quality of the project result is fitness-for-purpose. Not always it is possible to achieve the desired quality level because of the state-of-the-science or unforeseen problems. In any event, an important step is to qualify the final result. This means to provide an assessment of how the desired level of quality, or quality objectives, is met. The target quality can be evaluated using quality indicators (QI). The determination and evaluation of the uncertainty is also an important step in QC.

- **Quality Assurance:** An integrated system of management activities involving planning, implementation, assessment, reporting and quality improvement to ensure that a process, item, or service is of the type and quality needed and expected by the customer.
- **Quality Control:** The overall system of technical activities that measures the attributes and performance of a process, item, or service against defined standards to verify that they meet the stated requirements established by the customer, operational techniques and activities that are used to fulfil requirements for quality.

Quality Assurance applied to the emission modelling is designed to guarantee an adequate data compilation and using in order to provide a complete, representative and accurate estimation. Also, QA is implemented to ensure that an emission model is used within the range

of its applicability, i.e. for the conditions the model was developed for and the modelling results correspond to the expected quality.

Implementation of Quality Assurance/Quality Control procedures to the emission modelling provides an additional approach to the data quality assessment. The main objectives of QA/QC are focused on (i) detection of error sources, (ii) the error quantification and, finally, (iii) minimisation of error comprised to emission estimation results.

Three main sources of error could be identified for the emission model:

- Methodology (aggregation, simplification, resolution)
- Model (code error),
- Data (not representative, not complete).

Detailed information about Quality Assurance/Quality Control procedures are given in (IPCC 2000a). Also practical considerations in developing QA/QC systems and guidance how to use, document and verify emission data can be found there.

3.4.3. Uncertainty assessment and validation/verification of emission data

There exist several techniques to evaluate emissions data including “common sense” review of the data, source-receptor methods, bottom-up evaluations that begin with emissions activity rates and emission factors, and top-down evaluations that compare emissions estimates to ambient air quality data. Each evaluation method has strengths and limitations.

Comparisons with available other independently compiled emissions datasets are an option to evaluate completeness, approximate emission levels and correct source category allocations. These comparisons are also recommended by (IPCC 2000a). A verification process helps to estimate uncertainties and to improve and harmonise basic data used for emission calculations. In addition to this internal validation, an external review of calculations or assumptions by experts in relevant technical fields (expert peer review) assures that the current state of science is taken into account and helps to avoid systematic errors due to the choice of inappropriate or unrepresentative values. Expert judgements can be obtained by systematic methods (Morgan and Henrion 1990), in order to reduce the subjectivity. Based on the results of the emissions evaluation, recommendations can be made on possible improvements to the emission inventory. Local agencies responsible for developing the inventory can then make revisions to the inventory data prior to air quality modelling.

Displaying emission data graphically is also a useful means for quality assuring them. A tile plot of gridded emissions is an effective tool for identifying misplaced sources and for assuring oneself that spatial patterns of emissions are consistent with the location of sources. Pie charts are useful for assessing whether distribution of emissions among source types or categories is plausible. Displays of time series enable to look at diurnal or weekly patterns in emissions to see whether these appear logical (e.g. comparisons for night vs. day or week-ends vs. weekdays).

A validation of emission data as well as emission factors can be done by comparing modelled and measured concentrations for selected areas where a high resolution bottom-up emission inventory exists and different source contributions can be identified based on local

monitoring data. Validation of emission data includes as well the spatial and temporal variation. For that purpose, several validation studies were carried out for traffic emissions and emission factors at roads, in tunnels or street canyons. Modelled road transport contributions to ambient air concentration values near roads are compared to local upwind/downwind, tracer or hotspot/background measurements.

One example for a validation study is the project VALIUM (Schatzmann et al. 2005), where a data set in high detail was produced in 2001-2003 to validate the model system M-SYS as well as local emission calculation based on HBEFA as tools for urban air quality assessment. For that, field studies, tracer experiments and wind tunnel experiments were carried out at a street canyon in Hanover, Germany. The studies showed that due to the strong temporal variations of driving behaviour and driving patterns the common procedure to assign a rigid "traffic situation" to a given road segment results in considerable errors and should not be applied in numerical simulations with high temporal resolution. Another example for a recent emission factor validation in Germany was the project BAB II (Corsmeier et al. 2005). The Special Issue of Atmospheric Environment gives an overview of BAB II, its measuring concept, the experimental setup, and the quality assurance and control program. Results in terms of measured and model-calculated emissions of gaseous and particulate species are reported in 10 papers.

Recently an emission validation study for $PM_{10}/PM_{2.5}$ exhaust and non-exhaust emission factors for street pollution modelling was carried out by (Ketzler et al. 2005). The study collected and compared current available emission factor estimates for PM_{10} and $PM_{2.5}$ from emission databases, and validating these with street pollution measurements in Denmark, Sweden and Germany. All models consistently indicate that a large part (about 50% - 85% depending on the location) of the total PM_{10} emissions originates from non exhaust emissions.

A comprehensive verification study for road-traffic induced primary particle emissions in Switzerland for different traffic regimes and processes (exhaust pipe emissions, emissions from abrasion and suspension) was jointly realised by the Swiss Federal Laboratories for Materials Testing and Research (EMPA) and the Paul Scherrer Institute (PSI) (s. Gehrig et al. 2003). Concentration measurements of the ambient air were performed on both sides of busy roads. During meteorological conditions with winds across the street it was possible to determine the contribution of the local traffic from upwind-downwind differences. At sites, where this concept could not be realised, these contributions were calculated from the differences of the kerbside sites and nearby background sites. Hourly dilution factors were calculated from the measured concentration differences of nitrogen oxides (NO_x), the number of vehicles, and published NO_x emission factors from HBEFA. The emission factors for particles were calculated from the measured concentration differences, assuming that these undergo the same dilution as nitrogen oxides. Two vehicle categories were distinguished: LDV (light duty vehicles < 6 m, i.e. petrol and diesel passenger cars, vans, motor cycles) and HDV (heavy duty vehicles > 6 m, i.e. lorries and coaches). In order to distinguish between exhaust pipe emissions and emissions from abrasion and suspension the PM_{10} and $PM_{1.0}$ fractions were measured separately. $PM_{1.0}$ was interpreted as direct exhaust pipe emissions, and PM_{10} as total fine particle emissions. The difference $PM_{10} - PM_{1.0}$ thus represented the emissions from abrasion and dust suspension.

Another recent Swiss study compared calculated road traffic emissions based on HBEFA with emissions derived from measurements (Colberg et al. 2005). The study applied two different techniques: (1) Trace gas measurements in a tunnel provided real-world emission factors of entire vehicle fleets and, after statistical regression analysis, additionally emission factors of different vehicle classes. (2) Based on dynamometric test measurements and engine maps at different conditions, emission factors of single vehicles are combined with emission factors of HBEFA road traffic emission model (UBA 2004). The results demonstrate good agreement for the trends of time series measurements. However, a deviation for the absolute emission factors of NO_x is observed for light- and heavy duty vehicles (LDV and HDV, respectively). Emission estimates based on HBEFA were larger than 50% for LDV and 15% for HDV compared to the emission factors derived from the tunnel study. The deviation is even larger for the CO emissions (75% overestimation for LDV and 130% overestimation for HDV). The VOC emissions could only be compared for LDV. There the deviation was within the confidence interval of the measurements.

In Austria, HBEFA emission factors were validated as well based on tunnel measurements (e.g. Hausberger et al. 2003). New model input data for heavy duty vehicles were gained from extensive measurement campaigns on HDV engines at TU Graz and a European data collection program. The comparison of the new emission factors with the values given in the HBEFA version 1.2 shows that emissions of modern HDVs are underestimated clearly by HBEFA. Road tunnel measurements were used to validate the new model.

The quality of currently published emission factors (including systematic errors) has been assessed by (Kühlwein 2004) as well by comparisons between the emission rates, modelled on the basis of the extended traffic measurements, measured pollutant concentrations and the data from the tracer gas experiment. The results are shown in Figure 3-13.

Several studies deal with the evaluation of COPERT emissions factors as well. In (Mensink 2000) two emission validation methods for urban emission inventories are presented. Emission factors derived from the COPERT II methodology were compared with on-board emission measurements and modelled traffic flow rates were compared with observations. The second validation method focuses on the completeness of the inventory, i.e. coverage of all sources, comparing measured pollutant fluxes in the urban plume with the downwind transported and dispersed emissions integrated over plume width and mixing height. Several validation studies have been done in the ARTEMIS project as well (Assessment of Road Transport Emission Models and Inventory Systems, see <http://www.trl.co.uk/artemis/>). In (Mellios et al. 2006) road traffic urban emission inventories were validated by means of concentration data measured at air quality monitoring stations in Europe.

A critical evaluation of US on-road vehicle emission inventories based on the MOBILE6 model was recently published by (Parrish 2006). US Environmental Protection Agency estimates of on-road vehicle emissions were compared with ambient measurements and a fuel-based emission inventory. The following significant weaknesses and strengths were identified: (1) The emission estimates have varied considerably over the past 15 years and are not clearly converging to progressively more accurate and certain results. (2) The most recent emissions estimate accurately captures the rapid decrease in CO and VOC emissions, but overestimates the magnitude of CO emissions by about a factor of two. (3) The NO_x emission estimates for the mid to late 1990s are reasonably accurate, but NO_x emissions have

increased through that decade rather than decreased as indicated in emission estimates. (4) The most recent emissions estimate more accurately apportions NO_x emissions between diesel and gasoline fueled vehicles than did earlier reports. (5) The ratio of two specific VOC species that has been characterized by ambient measurements suggests that the inventory speciation of the VOCs is inaccurate by factors of 3 to 4.

In a few cases the uncertainties of emission factors due to measuring errors were determined and published. In general, sources of gaseous pollutant emissions are better characterised and hence inventories for gaseous pollutants are less uncertain than inventories for particulate matter and metals, which, in turn, are less uncertain than the inventories for persistent organic pollutants. Further refinement of the uncertainty analysis is desirable, especially the need to improve the understanding of the characterisation of emission sources and uncertainty in emissions data provided directly by industry or regulators (Passant 2003).

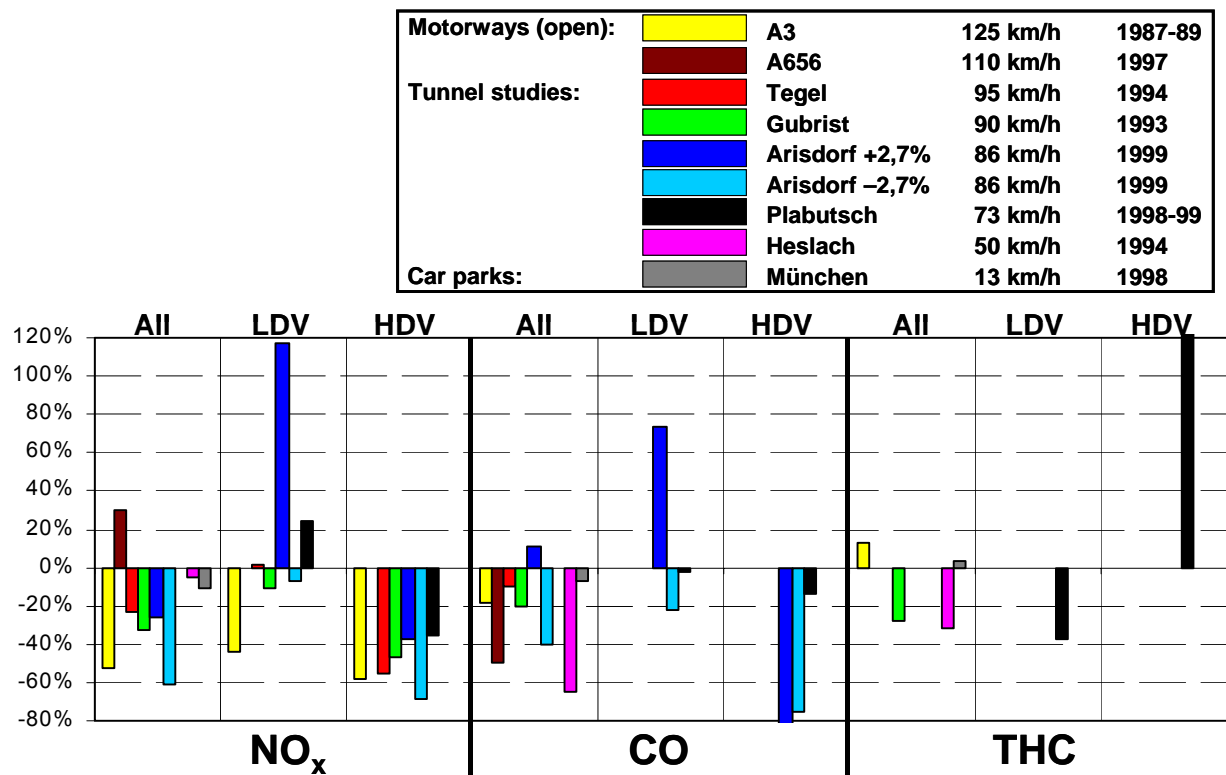


Figure 3-13: Deviation of measured real-life emission factors of various studies from corresponding HBEFA emission factors (Kühlwein 2004)

A comprehensive work on uncertainties of emission data and methods for uncertainty assessment was published by (Van Aardenne 2002). An analysis of procedures for verification of emission inventories can be found in (Saeger 1994). Useful information on uncertainties, validation and verification can be found as well in (Friedrich & Reis 2004). Uncertainty analyses based on bootstrap simulation or similar “Monte Carlo” methods were performed in e.g. (Borrego et al. 2003), (Rypdal & Zhang 2000) and (Winiwarer & Rypdal 2001). An assessment of the uncertainties of highly resolved emissions estimated by an emission model was done on urban scale within the experiment EVA (Evaluation of Highly Resolved Emission In-

ventories). An overview of this experiment executed in Augsburg, Germany in the year 1998 and a presentation of the major results was published in (Slemr et al. 2002). An example of top down emission validation is (Hov et al. 1997). Observations and model calculations of the concentration of hydrocarbons at five Scandinavian rural sites during March–June 1993 were reported and compared. A Lagrangian numerical model was used to calculate the concentrations of the individual hydrocarbons at the five sites based on EMEP emission data. The calculated concentrations for NMVOC with quite long chemical lifetimes agree well with the observations. For the sum of observed and calculated hydrocarbons the correlation coefficients are in the range of 0.65–0.88 for the five sites and the ratio between calculated and measured concentrations was 0.72–0.97, indicating that the European VOC emission inventory is quite well estimated.

Numerous source apportionment studies were done within the last decades using different approaches, for instance within the SAPPHIRE project (s. <http://www.gees.bham.ac.uk/research/sapphire/>) and the EUROTRAC-2 subprojects SATURN (s. <http://aix.meng.auth.gr/saturn/>). Results of source apportionment studies can be used as well to analyse if calculated source contributions correspond to measured source contributions in a selected area. Exemplary results of a source apportionment study are shown in Figure 3-14. The approach used allows distinguishing between regional and urban backgrounds and local road traffic contributions. A combined analysis of monitoring data from different specific locations and of information on source specific tracers allows identifying source contributions to the ambient air concentration.

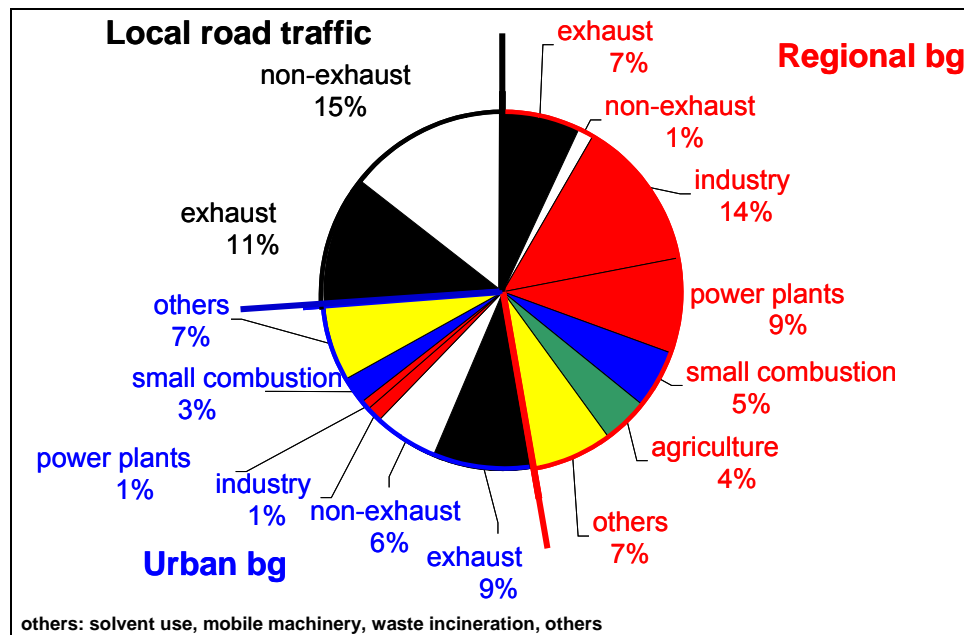


Figure 3-14: Results of the urban scale air quality assessment in Berlin: source apportionment of PM₁₀ for the traffic station Frankfurter Allee (based on John & Kuhlbusch 2004).

3.4.4. Most important gaps and research needs in present emission data generation

Emission inventories have several uncertainties and knowledge gaps that significantly affect accuracy and validity of this information. Therefore research is still needed in this area. Most important gaps and research needs are described in the following.

- **Harmonisation and data documentation:** The use of emission factors and other source specific information for the development of sectoral emission inventories is not harmonised and sufficiently documented. It will be difficult or impossible to achieve a full harmonisation because there is always an individual data choice especially for top down inventories. Another problem might be existing country specific or sector specific interests that influence emission calculations and reporting. Therefore at least a detailed and standardised documentation of emission inventories and used activity rates and emission factors should be demanded.
- **Knowledge of specific local/regional information:** Data availability on urban scale is usually limited with regard to local/regional information on emission factors, activity rates and other parameters. Therefore, a significant lack of source specific information usually exists for bottom-up analysis, especially for major sources such as road traffic or industry. Data has often to be used that were derived from other regions/technology stocks and national statistical distributions are often the only possibility to specify sources. As a result of possibly particular conditions, uncertainties of generated emission data on a high spatial resolution and a small scale area can be much higher than on a regional scale and a coarser grid.
- **Integration of meso-scale emission inventories:** The integration of regional meso-scale or urban scale emission inventories in regional/continental inventories would significantly increase their quality and applicability with regard to the spatial source allocation and resolution. However, an integration is usually not feasible due to different data structure, limited data availability and documentation and limited personnel capacities. Harmonised or at least transparent data structures and methodologies do often not exist and the availability and usability of information for the spatial disaggregation (point source information, line source coverage, area source emissions/activities for smaller administrative units) is not given. This would initially need an analysis and specification of requirements and possibilities of integration and the identification of available data sources. Alternatively, different and complementary emission data sets are often integrated in CTMs based on already gridded data.
- **Uncertainty analysis:** There is a lack of uncertainty analysis of generated emission data as systematic methodologies for uncertainty assessment do not exist. For uncertainty analysis of spatial and temporal emission data comprehensive validation studies have to be performed analysing modelled and measured concentrations for certain locations and episodes. To derive the uncertainty of spatial emission data is quite complicated because concentration measurements as well as atmospheric dispersion modelling are uncertain as well. Therefore a further development of methodologies of and experiences on emission data validation is needed esp. on regional scale.

- **Integration of point and line sources into emission inventories:** Emission inventories do often have a lack of industrial point sources due to limited data access. Available data from EPER include a small number of major sources in Europe but are not completely validated and therefore highly uncertain information. A lack of accurate line source modelling often exists due to limited project capacities. Accurate inclusion of line sources requires traffic census data at least for major roads and different vehicle categories. For regional/continental inventories a consistent digitised road map and traffic census data linked to road sections are not available. Insufficient representation of line and point sources increases uncertainties of spatial emission data especially on regional meso-scale below 10 km x 10 km spatial resolution.
- **Vertical profile of point sources emissions:** The effective emission height is required for an accurate use of point source emissions within atmospheric dispersion models. Therefore information is required about location, stack height, altitude, flue gas temperature, flow rate and velocity. There is a lack of generalised specific information for different source types. The point source information provided by EPER does unfortunately not include information on the emission height (EPER 2004).
- **NM VOC speciation:** There is still a lack of harmonised and accurate NM VOC emission speciation. A definition of a standard speciation depends on the requirements of the atmospheric modelling community on the one hand and the AQ assessment on the other hand. Often used schemes are RADM2, RACM, CBM4 and EMEP.
- **Temporal profiles:** More accurate information for a temporal disaggregation of emissions is needed with regard to source characteristics and regional/local dependence. Especially a better methodology for the incorporation of specific local/regional temperature data is required for source groups as residential combustion, gasoline evaporation and cold start emissions. Effects of the liberalisation of the EU energy market are usually not yet taken into account for the temporal profiles of large combustion plants.
- **Particulate matter emission characterisation:** Source specific information on size and chemical content of anthropogenic particulate matter emissions are required for AQ assessment and atmospheric dispersion modelling. Large knowledge gaps still exist for PM_{2.5} and finer size fractions and especially for fugitive emission sources as e.g. tyre and brake wear from road traffic, suspension of road dust and fugitive industrial emissions. Specific emissions of fugitive sources and their significance as local, urban or regional emission source are usually unclear.
- **Emission calculation for certain pollutants and sources:** Large knowledge gaps exist for the quantification and characterisation of emissions of certain pollutants and from certain sources. Beside the already mentioned fugitive anthropogenic PM emissions there still exists a lack of information for biogenic/natural emissions such as NM VOC from certain plant types (e.g. species in boreal forests), windblown dust from different soil types or methane emissions from animals or swamps/seepage. As well for agricultural emissions e.g. of NH₃ or pesticides there are still high uncertainties depending on animal species resp. crop types, agricultural methodologies or other regional parameters. Uncertainties of anthropogenic emissions exist especially with regard to heavy metals and other elements or POP and other organic compounds.

- **Validation of emission factors:** Emissions factors used for emission calculations in Europe should be further validated, especially for major sources such as road traffic. Further validation studies should start with a comprehensive analysis of the work already done in several studies (see examples before) and should directly lead to a review and improvement of the emission factors used.

4. Recommendations for the generation and use of emission data for air quality assessment

This chapter outlines recommendations of good practice for emission modelling on regional/continental, urban/agglomerate and local/hotspot scale. Emission data with a high resolution in space, time and substances are a prerequisite for the operation of air quality models on all scales. Uncertainties in emission data are strongly related to uncertainties in the source specific input data for emission calculations (activities, emission factors) as well as in the usage/availability of spatial allocation data and temporal profiles. The following recommendations have been the basis for contributions to the Air4EU Recommendation Document D 6.2 (Final recommendations for AQ assessment).

4.1. General recommendations for the generation of emission inventories

Compiling an emission inventory requires a multitude of input parameters in order to model reality as accurate as possible. Data collection, data analysis and data choice are important procedures for this compilation. As a consequence of political agreements several quality criteria have been introduced for greenhouse gas emission inventories submitted by countries to the UNFCCC (IPCC 2000a). These quality criteria specifically include accuracy, completeness, consistency and comparability and can be applied to all emission inventories. The following general recommendations mainly refer to these requirements.

- It is good practice to document and archive all information, assumptions and methodologies used to generate an inventory as well as highly resolved emission data. A systematic documentation and archiving are important preconditions for a constant improvement and update as well as an uncertainty analysis.
- Emission inventories should include summaries of methods used and references to source data such that the emissions estimates are transparent and steps in their calculation may be retraced. A documentation should consist of records that are unambiguous. This is an important precondition for a constant improvement and update as well as for an uncertainty analysis/verification. Examples of specific documentation and reporting can be found in (IPCC; 2000).
- Emission inventories should be compared with available other independently compiled emission datasets. This is an option to internally evaluate completeness, to approximate emission levels and to correct source category allocations. It helps to check key sources and plausibility of source contributions. As far as possible and reasonable, used basic data and methodologies should be harmonised with already existing inventories. For ex-

ample urban inventories should be harmonised with existing inventories on regional or national level.

- An independent review of calculations or assumptions by external experts in relevant technical fields is recommended as well. A real expert peer review calls for a systematic approach to calculate emission data. Although harmonised methodologies do often not exist, it is recommended to discuss data choice e.g. for emission factors with external experts in order to avoid the choice of inappropriate and incorrect values. National or international research institutes should be contacted as well to ensure the current state of knowledge.
- Emission inventories are “living databases” that require constantly updating and improving if new information/knowledge gets available. If calculation methodologies were changed, emission time series should be recalculated in order not to present incorrect emission trends.
- Emission inventories should include all anthropogenic sources, which may contribute significantly to the ambient air concentrations. Coarse estimations – if transparent – are better than to omit relevant sources because there seems to be no established methodology and data for an emission calculation. Especially for fugitive fine particulate matter emissions, estimations should be done for sources like e.g. agriculture, construction, industry and road dust suspension. On the other side, a high number of coarse estimations increase the overall uncertainty of the inventory. This should be kept in mind when assessing and interpreting emission inventories.
- A validation of emission data is desirable but often not possible because it requires extensive project capacities. Emission data in high spatial and temporal resolution have to be generated and used to model atmospheric concentrations with the help of an atmospheric dispersion model and highly resolved meteorological data for the year and area under investigation. A comparison with measured concentrations allows to validate the calculated total emission level and its spatial and temporal variation. Using information about the specific chemical content/species of emissions (e.g. PM₁₀, NMVOC) and measured concentrations allows to validate the contribution and specific emissions of selected source groups (also by using receptor models). Numerous of these studies were carried out on different scales in the past. It is recommended to carry out a source apportionment study for at least one selected site in order to quantify the primary emissions and to compare calculated and measured source contributions. If direct validation of an inventory is not possible results of such validation studies for similar areas can be used to further assess the inventory and used emission factors.
- Biogenic/natural emissions should be taken into account if they may contribute significantly to ambient air concentrations. Their significance depends on pollutant (e.g. NMVOC, PM₁₀) as well as scale of air quality assessment. Monitoring data are usually used to quantify biogenic/natural contributions on local scale. There exist several applied methods to quantify and model these emissions on regional scale based on activities and emission factors or functions (see D5.1). The improvement of these methods is currently a research topic within e.g. NATAIR (Improving and Applying Methods for the Calculation

of Natural and Biogenic Emissions and Assessment of Impacts on Air Quality) and other projects.

- An uncertainty assessment and analysis should be done for calculated emissions at least qualitatively. The EMEP/CORINAIR Guidebook (EEA 2003) presents a default table for quality ratings that can be used for uncertainty discussion and assessment. More valuable than coarse uncertainty estimations is transparency of basic data and methodologies used for emission calculations.
- Source groups within emission inventories should be categorised according to international nomenclatures in order to compare, integrate or review resulting sectoral emission data. This categorisation should be done as detailed and transparent as possible. It is recommended to use NOSE (Nomenclature of Sources of Emissions) of EUROSTAT (based on CORINAIR SNAP level 3 and other categorisations) and additionally NAPFUE (Nomenclature for Air Pollution Fuels) fuel codes to distinguish between different combustion processes (see UBA 2002, EEA 2003).

4.2. Recommendations for the calculation of annual emissions

The first step of generating emission data is the calculation of sectoral emissions based on source specific basic data (activity rates, emission factors and source specifications). This leads to a sectoral source and emission inventory usually as an annual emission table for the whole area or already subdivided into different administrative units. Calculation methodologies as well as data quality are mainly a function of data availability and data choice. Increased requirements on the accuracy of emission calculation exist on the urban and local scale modelling compared to regional/continental scale. Key source groups and processes have to be analysed with high level of detail and as far as possible based on site specific information on activities and specific emissions.

- Emissions from road traffic should be calculated based on detailed data on technology level that can be used for all scales and areas in Europe. HBEFA and COPERT III are often used as data source for emission factors and additional information (see UBA 2004, Ntziachristos & Samaras 2000). Therefore the modelling of road traffic emissions should distinguish at least between different road classes and vehicle categories. If possible, a bottom-up approach should be used for an inventory on national or smaller scale based on annual traffic volume per road section and vehicle category. Information about fraction of diesel and gasoline engines and distribution of EURO emission standards of the fleet are usually available from national or regional statistics and should be used to aggregate detailed emission factors for emission calculations. For research projects on local/hotspot scale, this information should be derived locally e. g. by automatic license plate number registration using video recording and consultation of vehicle register. On larger scale it is recommended to use national inventories or official EMEP country totals on NFR level 2 (EMEP 2004).
- It is recommended to distinguish between NO and NO₂ emissions and emission factors in order to be able to model the increasing NO₂/NO_x emission ratio on roads in Europe during the last years (see e. g. data provided in the Annex).

- For urban and local scale, it's recommended to calculate and model road traffic emissions as far as possible with a bottom-up approach based on local information. Census data from automatic counting should usually be available for sections of major roads. This provides traffic volume distinguished between large (heavy) and small (light) vehicles. In addition, manual traffic counting is recommended for selected roads and episodes (typical working day, Saturday, Sunday of different seasons). This enables to distinguish between several vehicle categories, to determine travel patterns and fraction of heavy-duty vehicles and to specify traffic on side roads where no automatic counting is done.
- Driving conditions such as road characteristic and speed should be analysed and defined in detail within urban and local models in order to assign appropriate emission factors. Acceleration plays an important role for emission generation as well. As acceleration is due to slope, side roads and traffic signals, these conditions should be taken into account as well.
- Non-exhaust emissions of particulate matter due to tyre wear, brake wear and road dust suspension due to traffic should be included in urban emission inventories as they contribute significantly to the PM₁₀ concentration. Recently a new chapter within (EEA 2003) was published that provides suitable methodologies as well as emission factors for tyre and brake wear emissions. For road dust suspension the use of the empiric equation derived from measurements in the USA (EPA 2003) is not recommended as this can lead to overestimations of a factor of two or more. Meanwhile measuring results and derived emission factors have been published for different European driving conditions that enable quite better estimations for urban and non-urban road dust suspension (e.g. Düring et al. 2005 and Gehrig et al. 2003).
- It is recommended to include available point source data in emission models on all scales e.g. for power plants and large industrial plants. If industrial data is not available, EPER data (EPER 2004) provide emissions from 8.082 major sources in the EU, Norway and Hungary that can be used alternatively and in addition e.g. for landfills. The future reporting for PRTR will include as well point sources from further Eastern Europe countries. Especially for urban and local models, activities of and emissions from power plants and large industrial combustion and production plants should be included as individual point sources. These data should be provided by plant operators and available from regulatory authorities. Emission calculations for single point sources should be done using detailed emission factors on technology level taking into account local plant and abatement technologies. The usage of average emission factors from literature does usually not reflect local characteristics and leads to significant under- or overestimations of single source emissions. Emission measurements at single sources should be carried out by certified institutions with standardised methods and facilities (see e.g. ISO standards (www.iso.org), US EPA Emission Measurement Center (www.epa.gov/ttn/emc/), German VDI guidelines (www.vdi.de)).
- Emissions from area source groups can be calculated with a top-down approach, e.g. by using official EMEP country totals on NFR level 2 (EMEP 2004). For a higher sectoral resolution it is recommended to use original national inventories instead of aggregated EMEP data. In order to significantly improve accuracy and transparency it is recom-

mended to calculate emissions from urban or local key area sources based on a bottom-up determination of activities as well. Therefore it should be checked whether there exist suitable information within municipal registers. Fuel consumption of small combustion plants might be quantified using energy and sales statistics and information from energy supply companies. Type and capacity of solid fuel combustion plants can be derived from building registers that often includes information collected from chimney sweepers or administration. Default emission factors per fuel type and calculation methodologies from (EEA 2003) or (EPA 1995) are recommended if more recent and more specific information is not available from national or regional experts (see also data for wood combustion in the Annex).

- Available statistical and socio-economical data used for allocating national figures to smaller administrative units/areas should correspond well to the emission causing activities. Using population data is recommended only for selected source groups such as consumer goods or households. Other useful parameters for disaggregation to smaller administrative units may be for example
 - car registrations, fuel consumption or car sales for road traffic emissions,
 - employment by branch, nominal production or capacity, energy consumption, fuel consumption, raw material consumption for industrial source groups or
 - land use statistics, animal numbers and crop production for agriculture.Especially on urban scale, a multitude of statistical parameters should be available for top-down emission calculations. Nevertheless top-down approaches lack of including local data, as they use typical country-wide behavioural patterns which may not be reflected by the urban area under consideration.
- The development of action plans and strategies to reduce urban emissions requires trend scenarios in addition to emission inventories for previous years. National trend scenarios can be taken e. g. from CAFÉ baselines (IIASA 2004) or national experts. Prognosis studies for specific source groups on a regional scale might exist that should be taken into account. This information can be used for urban or local trend scenarios as well. In addition to that, it is recommended to include into scenario calculations also urban/local conditions, that may change the emission situation in the future (e.g. changes in road network, land use, industrial structure).
- Source apportionment studies are recommended to compare and verify calculated source contributions within urban areas. Monitoring data of regional and urban background stations as well as local traffic stations can be combined and analysed following e.g. the approach of (Lenschow et al. 2001). A comparison of regional background concentrations with urban concentrations leads to the urban source contributions. If urban background concentrations are compared to local concentrations near major roads, contributions from local road traffic can be estimated and compared to model results.
- It is recommended to carry out a systematic analysis of the impact of technical and non-technical measures such as low emission zones, improving traffic flows, speed limits, gas engines or particle traps for public transport, street sweeping etc. on PM_x emissions in urban areas.

4.3. Recommendations for the spatial disaggregation

Based on a sectoral emission inventory, a spatial mapping can be achieved by allocating source emissions to smaller geographic units/areas or cells of a model grid. Often a geographic information system (GIS) and an intersection with geographic information such as road maps, point source coordinates and land use data are used for this purpose. Air quality assessment on the regional/EU scale is usually done in macro-scale ($> 10 \times 10$ km) and meso-scale ($< 10 \times 10$ km) resolution with a maximal spatial resolution of 1×1 km. Air quality assessment on urban scale usually requires a higher spatial resolution than available emission data from regional inventories have. In order to minimise uncertainties of gridded emission data, a detailed spatial allocation of point and line sources should be done. The following approaches can be recommended.

- The spatial allocation of emissions should be based on point, line and area sources. It is recommended to allocate area source emissions by statistical and land use data and to use a GIS to intersect source contributions with a model grid.
- In general, road traffic should be modelled as line sources for emission data in meso-scale resolution ($< 10 \times 10$ km). It is recommended to model at least non-urban traffic on major roads (highways/motorways) as line sources based on a digitised road net, individual road sections and - if available - linked mileage data. On national scale it might be possible to allocate complete traffic on non-urban roads as line sources depending on data availability. Urban road traffic is usually modelled on regional scale as area source based on statistic data on vehicle stock or population and settlement area. On urban scale, emissions from major roads should be modelled as line sources based on local traffic census data (automatic counting). The spatial allocation within an emission model requires a digitised road net and linked mileage data. Automatic traffic counters usually distinguish between small and large vehicles. There should be a verification of the fraction of heavy-duty vehicles by manual traffic counting for selected major roads and episodes. Manual traffic counting for selected side roads should be used as well to allocate emissions from light and heavy-duty vehicles on e.g. side road length per grid.
- For the inclusion of point source data, information on source location (coordinates) and effective height of stacks (stack height plus plume rise) should be available. In addition to industrial sources, airports and landfills should be modelled as well as point sources on regional scale. Local industrial data should be used on urban or local scale. EPER data can be used alternatively for major sources. The vertical distribution of EPER emissions has to be estimated as EPER does not comprise information on stack height, flue gas temperature and velocity.
- For area sources it is recommended to use statistical data for a “first stage” distribution on smaller administrative units. In some cases small scale data for e.g. the fuel consumption per block/square of residential area is available for urban areas. Land use data (e.g. CORINE) with high spatial resolution should be used to further disaggregate and allocate area source emissions before intersecting with a grid.
- When focusing on urban problems it is recommended to generate emission data on the highest possible spatial resolution in order to improve the accuracy and spatial differentiation. A resolution between $250 \text{ m} \times 250 \text{ m}$ and $500 \text{ m} \times 500 \text{ m}$ can possibly be

achieved. For regional problems (e.g. Ozone concentration) a resolution of e.g. 1 km x 1 km or 2 km x 2 km is sufficient. As land use data of CORINE 2000 have a resolution down to 100 m x 100 m, emission data might be generated in a higher spatial resolution without extensive efforts on data collection.

4.4. Recommendations for the temporal disaggregation

Source or process specific temporal profiles have to be generated and assigned to annual emissions to generate emission data in high temporal resolution. The following recommendations can be given for the temporal disaggregation.

- Temporal profiles of activities and emissions strongly depend on the processes. Therefore profiles should be defined and assigned to source groups on a detailed sectoral structure in order to include partly available specific data and to easily update and further develop the profiles. The use of average profiles on e.g. SNAP level 1 is not recommended.
- If source specific information for one specific plant/process or one specific region is applied to other sources resp. regions comparability should be assessed with regard to technology, operation and other significant boundary conditions (e.g. temperature/climate, holiday seasons, shift operations, harvest times, idle times).
- Temporal profiles for some sources depend on climatic conditions (e.g. small combustions in households, gasoline evaporation, vehicle cold starts). If possible, (local) hourly/daily temperature data should be taken into account for these sources.
- Temporal profiles have a much higher accuracy if they consist of hourly shares of the annual emission. In contrast, the use of average monthly, weekly and diurnal profiles leads to possibly high differences if seasonal variations of the weekly and diurnal profiles are not taken into account.
- It is recommended to use local data if possible (e. g. traffic counts, working hours in local plants, holiday seasons). Hourly temperature data should be taken into account for sources like residential combustion plants, gasoline evaporation and vehicle cold starts.
- It is recommended to use monitoring data in temporal resolution for a validation of the temporal emission profiles.

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6. Annex I – Basic data for calculating emissions for special topics

Table 6-1: Source and technology specific NO₂/NO_x ratios

TECHNOLOGY/PROCESS	NO ₂ /NO _x RATIO	DATA SOURCE
Stationary combustion plants		
Small underfeed furnaces, wood waste, 1.8 MW	0,090	Mohn 2000
Small underfeed furnace, nature-led-wood, 930 kW	0,040	Mohn 2000
Small stoker-fired furnace, wood waste, 3 MW	0,050	Mohn 2000
Small underfeed furnaces, wood waste (chips), 186 / 210 kW	0,090	Mohn 2000
Diesel engines		
Light-duty vehicles, average EURO 0	0,160	Latham et al. 2001
Light-duty vehicles, average EURO 1	0,140	Latham et al. 2001
Light-duty vehicles, average EURO 2	0,090	Latham et al. 2001
Light-duty vehicles EURO 0, v=30 km/h	0,181	Latham et al. 2001
Light-duty vehicles EURO 1, v=30 km/h	0,144	Latham et al. 2001
Light-duty vehicles EURO 2, v=30 km/h	0,243	Latham et al. 2001
Light-duty vehicles EURO 0, v=50 km/h	0,171	Latham et al. 2001
Light-duty vehicles EURO 1, v=50 km/h	0,149	Latham et al. 2001
Light-duty vehicles EURO 2, v=50 km/h	0,269	Latham et al. 2001
Light-duty vehicles (ULSD), average for Leicester UK	0,245	Latham et al. 2001
VW TDI, 1500rpm, ULSD S<10ppm, NO ₂ /NO _x after Oxicat, temperature before catalyst: 300°C	0,262	Czerwinski et al. 2006
VW TDI, 1500rpm, ULSD S<10ppm, NO ₂ /NO _x after Oxicat, temperature before catalyst: 400°C	0,300	Czerwinski et al. 2006
VW TDI, 2500rpm, ULSD S<10ppm, NO ₂ /NO _x after Oxicat, temperature before catalyst: 300°C	0,062	Czerwinski et al. 2006
VW TDI, 2500rpm, ULSD S<10ppm, NO ₂ /NO _x after Oxicat, temperature before catalyst: 400°C	0,237	Czerwinski et al. 2006
VW TDI, 3000rpm, ULSD S<10ppm, NO ₂ /NO _x after Oxicat, temperature before catalyst: 300°C	0,137	Czerwinski et al. 2006
VW TDI, 3000rpm, ULSD S<10ppm, NO ₂ /NO _x after Oxicat, temperature before catalyst: 400°C	0,275	Czerwinski et al. 2006
Passenger car Peugeot 406 with catalyst and particulate trap FAP, 1715 rpm, 80 km/h, on roller test bench, temperature before catalyst: 350°C	0,580	Czerwinski et al. 2006
Passenger car Peugeot 406 with catalyst and particulate trap FAP, 1715 rpm, 80 km/h, on roller test bench, temperature before catalyst: 400°C	0,310	Czerwinski et al. 2006
Passenger car EURO 2, Lti, Taxi, Nissan IDI, standard, total	0,076	AQEG 2006
Passenger car EURO 2, Lti, Taxi, Nissan IDI, w. Oxicat, total	0,200	AQEG 2006
Passenger car EURO 2, Lti, Taxi, Nissan IDI, w. Oxicat, urban	0,062	AQEG 2006
Passenger car EURO 2, Lti, Taxi, Nissan IDI, with Oxicat, motorway	0,241	AQEG 2006
Passenger cars EURO 3, ECE, average of 15 cars	0,150	Petit 2006
Passenger cars EURO 3, EUDC, average of 15 cars	0,330	Petit 2006
Passenger cars EURO 3, NEDC, average of 15 cars	0,250	Petit 2006
Passenger cars EURO 4, ECE, average of 11 cars	0,300	Petit 2006
Passenger cars EURO 4, EUDC, average of 11 cars	0,540	Petit 2006
Passenger cars EURO 4, NEDC, average of 11 cars	0,430	Petit 2006

TECHNOLOGY/PROCESS	NO ₂ /NO _x RATIO	DATA SOURCE
Liebherr 914T, LSD S<50ppm, 2000 rpm full load - hot	0,187	Czerwinski et al. 2006
Liebherr 914T, LSD (S<50ppm, 2000 rpm 378 Nm - hot	0,357	Czerwinski et al. 2006
Liebherr 914T, LSD (S<50ppm, 1400 rpm full load - hot	0,132	Czerwinski et al. 2006
Liebherr 914T, LSD (S<50ppm, 1400 rpm 445 Nm - hot	0,362	Czerwinski et al. 2006
Liebherr 914T, LSD (S<50ppm, 1400 rpm 297 Nm - hot	0,661	Czerwinski et al. 2006
Liebherr 914T, LSD (S<50ppm, 2000 rpm full load - cold	0,039	Czerwinski et al. 2006
Liebherr 914T, LSD (S<50ppm, 2000 rpm 378 Nm - cold	0,059	Czerwinski et al. 2006
Liebherr 914T, LSD S<50ppm, 1400 rpm full load - cold	0,044	Czerwinski et al. 2006
Liebherr 914T, LSD (S<50ppm, 1400 rpm 445 Nm - cold	0,052	Czerwinski et al. 2006
Liebherr 914T, LSD (S<50ppm, 1400 rpm 297 Nm - cold	0,098	Czerwinski et al. 2006
Liebherr 914T, ULSD S<10ppm, 1400 rpm, FBC Octel Octi-max 4810a, 300°C, CRT	0,500	Czerwinski et al. 2006
Liebherr 914T, ULSD S<10ppm, 1400 rpm, FBC Octel Octi-max 4810a, 300°C, Oxicat	0,320	Czerwinski et al. 2006
Heavy-duty vehicles, EURO 0, v=30 km/h	0,093	Latham et al. 2001
Heavy-duty vehicles, EURO 1, v=30 km/h	0,112	Latham et al. 2001
Heavy-duty vehicles, EURO 2, v=30 km/h	0,140	Latham et al. 2001
Heavy-duty vehicles, EURO 0, v=50 km/h	0,072	Latham et al. 2001
Heavy-duty vehicles, EURO 1, v=50 km/h	0,089	Latham et al. 2001
Heavy-duty vehicles, EURO 2, v=50 km/h	0,143	Latham et al. 2001
Heavy-duty vehicles (ULSD), average for Leicester UK	0,121	Latham et al. 2001
Heavy-duty vehicles, EURO 2	0,083	DC 2006
Heavy-duty vehicles, EURO 2, Oxicat	0,047	DC 2006
Heavy-duty vehicles, EURO 2, CRT	0,357	DC 2006
Heavy-duty vehicles, EURO 3	0,050	DC 2006
Heavy-duty vehicles, EURO 3, CRT	0,600	DC 2006
Heavy-duty vehicles, US EPA 2004, with after treatment	0,043	DC 2006
Heavy-duty vehicles, EURO 4, SCR	0,020	DC 2006
Heavy-duty vehicles, EURO 5, SCR	0,020	DC 2006
Heavy-duty vehicles, EURO 4, PM Cat (open system)	0,405	DC 2006
Heavy-duty vehicles, EURO 5, SCRT (SCR + CRT)	0,080	DC 2006
Bus, EURO 0, v=30 km/h	0,151	Latham et al. 2001
Bus, EURO 1, v=30 km/h	0,208	Latham et al. 2001
Bus, EURO 2, v=30 km/h	0,179	Latham et al. 2001
Bus, EURO 0, v=50 km/h	0,130	Latham et al. 2001
Bus, EURO 1, v=50 km/h	0,142	Latham et al. 2001
Bus, EURO 2, v=50 km/h	0,175	Latham et al. 2001
Bus (ULSD), average for Leicester UK	0,159	Latham et al. 2001
Bus with CRT	0,10-0,50	Ling & Helden 2003
Gasoline engines		
Passenger car EURO 3, NEDC, average of 2 cars	0,060	Petit 2006
Passenger car EURO 4, NEDC, average of 5 cars	0,030	Petit 2006
Vans in general	0,050-0,38	Soltic 2003
Passenger car (ULP), average for Leicester UK	0,100	Latham et al. 2001
Passenger car, small (<1400cc) average 3 cars EURO 0,1,2	0,025	Latham et al. 2001
Passenger car, medium (1400-2000cc)	0,080	Latham et al. 2001
Passenger car, large (>2000cc)	0,095	Latham et al. 2001
Passenger car (ULP), Euro 0, v=30 km/h	0,008	Latham et al. 2001
Passenger car (ULP), Euro 1, v=30 km/h	0,041	Latham et al. 2001
Passenger car (ULP), Euro 2, v=30 km/h	0,059	Latham et al. 2001
Passenger car (ULP), Euro 0, v=50 km/h	0,007	Latham et al. 2001
Passenger car (ULP), Euro 1, v=50 km/h	0,030	Latham et al. 2001
Passenger car (ULP), Euro 2, v=50 km/h	0,027	Latham et al. 2001
Gasoline engines in general under normal driving conditions	0,020-0,05	Hilliard 1979

Table 6-2: Average urban two-wheeler emission factors for benzene from HBEFA (UBA 2004) for Germany in 2000

TECHNOLOGY	DISPLACEMENT [cc]	EMISSION FACTOR [g/veh.-km]
Moped without catalytic converter	<=50	0.242
Moped with catalytic converter	<=50	0.186
Scooter: before EURO1	<=50	0.219
Scooter: EURO1	<=50	0.166
Scooter: EURO2	<=50	0.060
Motorcycle: 2 stroke/ before EURO1	50-150	0.985
Motorcycle: 2 stroke/ EURO1	50-150	0.249
Motorcycle: 2 stroke/ EURO2	50-150	0.132
Motorcycle: 2 stroke/ EURO3	50-150	0.082
Motorcycle: 2 stroke/ before EURO1	>150	0.748
Motorcycle: 2 stroke/ EURO1	>150	0.323
Motorcycle: 2 stroke/ EURO2	>150	0.179
Motorcycle: 2 stroke/ EURO3	>150	0.088
Motorcycle: 4 stroke/ before EURO1	50-150	0.038
Motorcycle: 4 stroke/ EURO1	50-150	0.038
Motorcycle: 4 stroke/ EURO2	50-150	0.055
Motorcycle: 4 stroke/ EURO3	50-150	0.034
Motorcycle: 4 stroke/ before EURO1	151-250	0.026
Motorcycle: 4 stroke/ EURO1	151-250	0.026
Motorcycle: 4 stroke/ EURO2	151-250	0.053
Motorcycle: 4 stroke/ EURO3	151-250	0.026
Motorcycle: 4 stroke/ before EURO1	251-750	0.078
Motorcycle: 4 stroke/ EURO1	251-750	0.062
Motorcycle: 4 stroke/ EURO2	251-750	0.042
Motorcycle: 4 stroke/ EURO3	251-750	0.020
Motorcycle: 4 stroke/ before EURO1	>750	0.078
Motorcycle: 4 stroke/ EURO1	>750	0.062
Motorcycle: 4 stroke/ EURO2	>750	0.042
Motorcycle: 4 stroke/ EURO3	>750	0.020

Table 6-3: Measured two-wheeler emission factors for PM

TECHNOLOGY	EMISSION FACTOR [g/veh.-km]	DATA SOURCE
motorcycles 2-stroke: Yamaha EW 50 Slider, with catalyst, 2.5 kW, v=30 km/h, with cold start	0.090	Czerwinski et al. 2000
motorcycles 2-stroke: Yamaha EW 50 Slider, with catalyst, 2.5 kW, v=30 km/h, without cold start	0.024	
motorcycles 2-stroke: Yamaha EW 50 Slider, with catalyst, 2.5 kW, Swiss driving cycle cold ZUS 98 (30 min cooling time)	0.074	
motorcycles 2-stroke: Yamaha EW 50 Slider, with catalyst, 2.5 kW, Swiss driving cycle warm ZUS 98	0.019	
motorcycles 2-stroke: Yamaha EW 50 Slider, without catalyst, 2.5 kW, v=30 km/h, with or without cold start	0.013	
motorcycles 2-stroke: Yamaha EW 50 Slider, without catalyst, 2.5 kW, Swiss driving cycle cold ZUS 98 (30 min cooling time)	0.055	
motorcycles 2-stroke: Yamaha EW 50 Slider, without catalyst, 2.5 kW, Swiss driving cycle warm ZUS 98	0.044	
motorcycles 4-stroke: Aprilia Leonardo 125 with catalyst, 8.5 kW, v=30 km/h, with cold start	0.009	Czerwinski et al. 2000
motorcycles 4-stroke: Aprilia Leonardo 125 with catalyst, 8.5 kW, v=30 km/h, without cold start	0.004	
motorcycles 4-stroke: Aprilia Leonardo 125 with catalyst, 8.5 kW, Swiss driving cycle cold ZUS 98 (30 min cooling time)	0.014	
motorcycles 4-stroke: Aprilia Leonardo 125 with catalyst, 8.5 kW, Swiss driving cycle warm ZUS 98	0.005	
motorcycles 4-stroke: Aprilia Leonardo 125 without catalyst, 8.5 kW, v=30 km/h, with cold start	0.017	
motorcycles 4-stroke: Aprilia Leonardo 125 without catalyst, 8.5 kW, v=30 km/h, without cold start	0.006	
motorcycles 4-stroke: Aprilia Leonardo 125 without catalyst, 8.5 kW, Swiss driving cycle cold ZUS 98 (30 min cooling time)	0.014	
motorcycles 4-stroke: Aprilia Leonardo 125 without catalyst, 8.5 kW, Swiss driving cycle warm ZUS 98	0.010	
Mopeds conventional	0.2	Ntziachristos et al. 2006
Mopeds EURO 1	0.08	
Mopeds EURO 2	0.04	
2-stroke motor cycles conventional	0.2	
2-stroke motor cycles EURO 1	0.08	
2-stroke motor cycles EURO 2	0.04	
2-stroke motor cycles EURO 3	0.012	
4-stroke motor cycles conventional	0.02	
4-stroke motor cycles EURO 1	0.02	
4-stroke motor cycles EURO 2	0.005	
4-stroke motor cycles EURO 3	0.005	

Table 6-4: Non-exhaust emission factors

ROAD CLASS / TRAFFIC SITUATION	VEHICLE CATEGORY	EMISSION FACTOR [g/km]			LITERATURE
		PM	PM 10	PM2.5	
<u>tyre wear:</u>					
urban traffic, Berlin	PC	0,053			Gebbe et al. 1998
urban traffic, Berlin	LDV	0,107			Gebbe et al. 1998
urban traffic, Berlin	HDV	0,539			Gebbe et al. 1998
Road traffic	HDV	0,045	0,027	0,019	EEA 2003
Road traffic	LDV	0,017	0,010	0,007	EEA 2003
Road traffic	PC	0,011	0,006	0,004	EEA 2003
Road traffic	2-wheeler	0,005	0,003	0,002	EEA 2003
Germany, mean value	HDV and buses	0,421	0,042		Klimont et al. 2002
Germany, mean value	LDV (4-stroke excl. GDI)	0,066	0,009		Klimont et al. 2002
Germany, mean value	motorcycles, mopeds	0,028	0,003		Klimont et al. 2002
"not specified"	HDV	0,371	0,019		TNO 2001 (CEPMEIP)
"not specified"	LDV	0,090			TNO 2001 (CEPMEIP)
"not specified"	PC	0,069			TNO 2001 (CEPMEIP)
"not specified"	motorcycles	0,035			TNO 2001 (CEPMEIP)
"not specified"	PC	0,097			Warner et al. 2002
<u>brake wear:</u>					
dynamometer tests, impactor	PC	0,003			Garg et al. 2000
dynamometer tests, impactor	LDV	0,005			Garg et al. 2000
dynamometer tests, impactor	HDV	0,009			Garg et al. 2000
dynamometer tests, impactor	mean value (PC/LDV/HDV)	0,006	0,001	0,004	Garg et al. 2000
tunnel, Berlin	PC		0,001	0,000	Rauterberg-Wulff 1998
tunnel, Berlin	HDV		0,025	0,007	Rauterberg-Wulff 1998
dynamometer tests	PC	0,015			Johansson et al. 1999
dynamometer tests	HDV	0,085			Johansson et al. 1999
dynamometer tests	buses	0,110			Johansson et al. 1999
urban traffic, Stockholm	PC	0,017			Westerlund 2001
urban traffic, Stockholm	HDV	0,084			Westerlund 2001
"not specified"	HDV	0,032	0,032	0,032	TNO 2001 (CEPMEIP)
"not specified"	LDV	0,008	0,008	0,008	TNO 2001 (CEPMEIP)
"not specified"	PC	0,006	0,006	0,006	TNO 2001 (CEPMEIP)
"not specified"	motorcycles	0,003	0,003	0,003	TNO 2001 (CEPMEIP)
Road traffic	HDV	0,033	0,032	0,013	EEA 2003
Road traffic	LDV	0,012	0,012	0,005	EEA 2003
Road traffic	PC	0,007	0,007	0,003	EEA 2003
Road traffic	2-wheeler	0,004	0,004	0,001	EEA 2003
Germany, mean value	HDV and buses	0,027	0,023	0,007	Klimont et al. 2002
Germany, mean value	LDV (4-stroke excl. GDI)	0,004	0,004	0,002	Klimont et al. 2002

ROAD CLASS / TRAFFIC SITUATION	VEHICLE CATEGORY	EMISSION FACTOR [g/km]			LITERATURE
		PM	PM 10	PM2.5	
<u>road dust suspension:</u>					
urban streets	PC/ LDV		0.030		Hüglin et al. 2000
urban streets	HDV		0.450		Hüglin et al. 2000
urban street (Hornsgatan) in winter	urban fleet		0.209	0.025	Johansson 2002
Switzerland, mean value	PC/ LDV		0.031		BUWAL 2001
Switzerland, mean value	HDV		0.475		BUWAL 2001
Switzerland, mean value	motorcycles		0.016		BUWAL 2001
Switzerland, mean value	mopeds		0.008		BUWAL 2001
highways	PC/ LDV		0.082		Fitz & Bufalino 2002
urban streets	PC/ LDV		0.098		Fitz & Bufalino 2002
interstate roads	PC/ LDV		0.129		Fitz & Bufalino 2002
other rural roads	PC/ LDV		0.064		Fitz & Bufalino 2002
<u>tyre and brake wear, road dust suspension:</u>					
urban streets (mean values) Nordic countries	average fleet (4% HDV)		0.205		Omstedt et al. 2005
highways, rural roads	PC/ LDV		0.022		Düring et al. 2005
highways, rural roads	HDV		0.2		Düring et al. 2005
urban streets (mean values)	PC/ LDV		0.05		Düring et al. 2005
urban streets (mean values)	HDV		0.45		Düring et al. 2005
highways, rural roads	PC/ LDV		0.04		Gehrig et al. 2003
highways, rural roads	HDV, coaches		0.2		Gehrig et al. 2003
highways, rural roads	public-transit buses		0.334		Gehrig et al. 2003
urban streets	PC/ LDV		0.054		Gehrig et al. 2003
urban streets	HDV, coaches		0.541		Gehrig et al. 2003
urban streets	public-transit bus		0.438		Gehrig et al. 2003

Table 6-5: Particulate matter emission factors for small commercial/institutional wood combustion plants

LITERATURE	YEAR OF REFERENCE	PROCESS	EMISSION FACTOR [kg/TJ]			
			PM	PM10	PM2.5	PM1.0
Baumbach et al. 1999	1999	underfeed stoker 1995 175 kW; wood chips (17% moisture)- chip boards (22% moisture); multicyclone	29	28	25	23
	1999	underfeed stoker modern 150 kW; wood chips (19-24% moist.); wood waste; particle depos. in furn. chamber	16	14	11	10
	1999	underfeed stoker modern 150 kW; wood waste (joiner's works.);8.1% moist.; particle depos. in furn. chamber	73	55	44	38
	1999	advancing grate, 450 kW, part load - full load; wood chips, 82.7% moisture; multicyclone	31 (28-34)	31	29	25
Struschka et al. 2003	2000	slow-combustion stoves, tiled stoves, chimneys, bathroom boilers, stoves; <15 kW; untreated wood	102	100	98	93
	2000	boiler 4-25 kW; untreated wood	23	22	18	17
	2000	boiler 25-50 kW, untreated wood	176	162	138	124
	2000	shaft furnace >50-1000 kW; untreated wood	21	20	17	15
	2000	shaft furnace >50-1000 kW; treated wood	63	58	49	44
	2000	injection furnace, underfeed stoker, dual-chamber furnace; >50-1000 kW; untreated wood	64	61	55	49
	2000	injection furnace, underfeed stoker, dual-chamber furnace; >50-1000 kW; treated wood	104	92	73	67
	2000	mean values for small commercial/institutional combustion plants in Germany in 2000	73	69	60	55
Mohn 2000	2000	automatic underfeed stoker 1.8 MW; wood waste 29 % moisture; multicyclone, DeNOx	128			
Mohn 2000	2000	grate furnace 2 MW; wood waste 19% moisture; cyclone, electric filter	2			
Mohn 2000	2000	underfeed stoker, dual-chamber; wood chips; 45% moisture; cyclone	70			
Mohn 2000	2000	grate furnace 3 MW; chips from wood waste; cyclone	104 (81-127)			
Mohn 2000	2000	underfeed stoker, wood chips (different contents of chip board); cyclone	93 (37-141)			
AEAT 2001b	2001	SNAP 020103, waste soft and hard wood	26	25	21	
EIIP 2001	2001	heating, wood-fired boiler; wood/bark/waste; no flue gas cleaning	364			
EIIP 2001	2001	bark-fired boiler; wood/bark waste; no flue gas cleaning	1943	703		
EIIP 2001	2001	wood/bark-fired boiler; wood/bark waste; no flue gas cleaning	298	267		
DMU 2003	2001	wood combustion in Denmark	143	143	135	
DMU 2006	2004	wood combustion in Denmark	143	143	135	

Table 6-6: Particulate matter emission factors for small residential wood combustion plants

LITERATURE	YEAR OF REFERENCE	PROCESS	EMISSION FACTOR [kg/TJ]			
			PM	PM10	PM2.5	PM1.0
EPA 1995	1996	open fireplace; dry wood 13 MJ/kg; no flue gas cleaning; measurement from a single fire place	1330	1330		
EPA 1995	1996	conventional wood stove; no flue gas cleaning	1176	1176		
	1996	catalytic wood stoves; combustor/converter	753	753		
	1996	non catalytic stoves; baffles/secondary combustion chamber	784	784		
	1996	pellet stoves; wood/sawdust pellets	250	250		
	1996	masonry heater, dry wood	215	215		
Hildemann et al. 1991		fireplace; softwood; no flue gas cleaning	861			
		fireplace; hardwood; no flue gas cleaning	334			
Gullett et al. 2003		woodstove; oak, 22% moisture; no flue gas cleaning	769			
		fireplace; oak, 20-28% moisture; no flue gas cleaning	428			
		fireplace; pine 11% moisture; no flue gas cleaning	215			
		fireplace; artificial log, 1% moisture; no flue gas cleaning	1277			
Fisher et al. 2000		wood stoves (catalytic and non catalytic)	748			
Houck et al. 1997		open fireplace <25 kW; dry wood 13 MJ/kg	907			
Houck et al. 2001		woodstove; wax-sawdust fire log, 3% moisture; no flue gas cleaning	416-708			
Fine et al. 2002	2001	masonry fireplace; yellow poplar, 33% moisture; no flue gas cleaning; (referring to PM2.5)	667		667	
	2001	masonry fireplace; mockernut hickory 12% moisture; no flue gas cleaning; (referring to PM2.5)	472		472	
	2001	masonry fireplace; soft wood (sweetgum, pine) ;12-14% moisture; no flue gas cleaning; (ref. to PM2.5)	199		199	
McDonald et al. 2000		woodstove; mixed hardwoods, oak; 10-15% moisture; no flue gas cleaning	247			
		fireplace; softwood (pine) 10% moisture; no flue gas cleaning	379			
		fireplace; hardwood (oak, mix) 15% moisture; no flue gas cleaning	407			
Spitzer et al. 1998	1997/98	various measurements at wood stoves in Austria, no flue gas cleaning	90 - 148			
Stanzel et al. 1995	1994	single stove (Austrian average); log, wood briquettes; no flue gas cleaning	30			
	1994	heating: central/covering one floor (Austrian average); log, wood briquettes; no flue gas cleaning	47 (30-70)			
Baumbach et al. 1999	1999	wood stove, 9 kW; log (beech, fir), briquettes	44 (31-56)	43	41	39
	1999	chimney stove, 6 kW; log (beech)	67	66	65	60
	1999	pellet stove, 8.5 kW; pellets, softwood	11	11	11	10
	1999	average value of measurements	46	45	43	41

Table 6-7: Examples of particulate matter emission factors for small residential wood combustion plants

LITERATURE	YEAR OF REFERENCE	PROCESS	EMISSION FACTOR [kg/TJ]			
			PM	PM10	PM2.5	PM1.0
Struschka et al. 2003	2000	slow-combustion stoves, tiled stoves, chimneys, bathroom boilers, stoves; (<15 kW), untreated wood	102	101	98	94
	2000	boiler: 4-25 KW; untreated wood	23	22	18	17
	2000	boiler: 25-50 KW, untreated wood	175	162	138	124
	2000	average values for wood combustion in Germany	116	112	104	98
Wieser & Gaegauf 2000	2000	batch, space heating; log 16% moisture	28 (18-38)			
	2000	storage heater (soapstone), wood 16% moisture	96			90
	2000	automatic pellet stove, wood 8% moisture	21			10
	2000	automatic chip stove (stoker), wood 60% moisture	135			26
Wieser 2001	2001	chimney stove, 6-7 kW; log (beech) 16-20 % moisture	24.5-111			
	2001	storage heater (soapstone), 6-3 kW, log (beech) 16-20% moisture	93-241			
	2001	boiler, 25-70 kW, log (beech – softwood 20% moisture)	16-39			
	2001	pellet stove 12 kW, wood 8% moisture	54			
	2001	pellet boiler, 16 - 25 kW, wood 8% moisture	20			
	2001	underfeed stoker (dry wood chips), 70 kW; wood chips 60% moisture	79			
	2001	underfeed stoker (dry wood chips), 120 kW; dry chips 35% moisture	109			
	2001	advancing grate furnace, 325 - 800 kW; wood chips 60% moisture	42-55			
Purvis et al. 2001	2001	dual-chamber furnace, 200 kW; wood waste (joiner's workshop) 10% moisture	64			
	2001	wood stove conventional, log (oak) 34.2% moisture	757 (457-1140)	88	79	
	2001	wood stove conventional, log (fir), 22-30% moisture	508 (282-734)	81	63	
AEAT 2004	2001	wood stove modern, log(oak) 34.2 % moisture; secondary air	514	81	66	
	2001	open fireplace <25 kW, wood	750	707		
	2003	advanced closed fireplace <25 kW, wood	310	295	288	
AEAT 2004 (CITEPA 2003)	2003	conventional stoves <25 kW, wood	310-400	223	194	
	2003	modern low emission stoves <25 kW, wood/pellets	30 (4-65)	23	14	
	2003	conventional manual feed boiler <50 kW, wood	250	229		
	2004	modern low emission boilers <50 kW, wood/pellets	34 (20-43)	34	27	
AEAT 2004 ("Biomass heating", Future Energy Solutions UK 2002)	2004	conventional manual feed boiler 50-1000 kW, wood	180	180	150	
	2004	modern auto- feed boiler 50-1000 kW, wood/pellets	32- 43	38		
	2004	large boiler 1-50 MW, wood, with abatement plant	2	2		
DMU 2006	2004	wood combustion in Denmark	715	679	643	

7. Annex II – PM emissions from two-wheelers

PM emissions from two-wheelers

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Executive Summary

This document has been compiled within the FP6 project Air4EU as a guidance document for the assessment and generation of PM emissions from two-wheelers. It describes the difficulties associated with PM assessment from two-wheelers and the reasons for these problems, and it provides recommendations on the technical procedure and instrumentation to be used, as well as further scientific research in regard to this topic. A number of particular issues are also dealt with, such as the influence of several factors on PM emissions from two-wheelers, including speed and engine technology and characteristics. The recommendations are intended to aid good assessment practice and to highlight areas that require further development.

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1. Introduction

1.1. Aim

The main aim of this report is to describe available methodologies for the assessment of PM emissions from two-wheelers, identify their main problems and provide recommendations for their best use.

1.2. Structure of this document

As outlined in the 'Overview and structure' section of the final recommendations document, each recommendation topic has been structured to provide access to the individual recommendations provided. This structure can be seen in the table of contents but is reiterated here for clarity.

The recommendations within this particular document are all relevant to the topic of the assessment of PM emissions from two-wheelers. Some general recommendations on PM emission assessment from traffic can also be found for the sake of completeness in the final chapter.

Within this topic a number of methods and issues for carrying out this assessment are discussed which include:

- *The reasons for increased PM emissions from two-wheelers*
- *The main PM characteristics that need to be monitored*
- *The factors influencing PM sampling and analysis from two-wheelers, and the main techniques used*
- *The difficulties associated with the current assessment methods and some recommendations for future assessment*

Each of these chapters is subsequently divided into particular relevant aspects of the different methodologies and recommendations. Most of them are future or scientific recommendations as the legislation on the PM emissions from two-wheelers and the associated research are relatively new.

Throughout the document recommendations are given in *Italics* and supporting text in normal font. In Chapter 6 (recommendations) some difficulties of the current assessment situation leading to the appropriate recommendations are given in bold.

1.3. The problem of PM emissions from two-wheelers in Europe

PM is not a single pollutant and its mass includes a mixture of many pollutants in a complex multiphase system. Particles can be 'primary' in the sense that they are emitted as particles from the source, or they can be secondary, meaning that they are produced in the atmosphere from gaseous precursors. Aerosol particles cover a wide range of diameter between a few nanometers for particles just produced from the gas phase, to tens of micrometers for coarse material, and thus have various atmospheric properties. The health effect of the particles is a strong function of the chemical composition of the PM as well as the physical properties of the particles, and therefore *the investigation of source specific particle size distribu-*

tions or size fractions like PM_{10} and $PM_{2.5}$ is important. It is also well known that PM is the sum of contributions from a variety of sources, each with its chemical and physical characteristics.

Calculations of fine particulate matter emissions are usually done by assigning source specific particle size factors (e.g. for PM_{10} and $PM_{2.5}$) to total particulate matter (PM) emissions, calculated with emission factors and activity rates. Emissions of elements or chemical compounds as a portion of emitted particles are often estimated by combining PM emission data with analysed source specific information on the substance content of these emissions. Estimated and officially reported country totals are provided amongst others for particulate matter by EMEP WEBDAB (EMEP; 2004). An improved European emission inventory for heavy metals was developed within the ESPREME project (ESPREME; 2005).

Two wheelers account for a significant proportion of vehicular air pollution in urban areas, particularly Hydrocarbons (HC) and Particulate Matter (PM), as well as CO and lead. The environmental impact from these two-stroke engines is pervasive. Carbon monoxide is a powerful respiratory irritant. Particulate emissions contribute to respiratory disease. Hydrocarbon emissions contribute to ozone formation. A few Asian countries still use leaded gasoline. Research has shown that the lead emitted in unburned hydrocarbons (i.e. scavenged fuel from carburetted 2-strokes) is much more damaging to the nervous system of children than the lead which is emitted as products of combustion. Thus, the importance of 2-stroke 2-wheelers emissions and their contribution to the air pollution in the cities is increasingly recognized and investigated in several countries.

Vehicle fleets in most Asian countries comprise two- and three-wheelers, a majority of which are powered by 2-stroke engines. Air pollution in many Asian cities such as Delhi, is increasing due to the proliferation of vehicles powered by simple two-stroke cycle engines (EP-ANCR; 1999). These engines produce high levels of carbon monoxide, unburned hydrocarbons, and particulates. Therefore, the problem of powered two-wheeler (PTW) emissions has been addressed in the past by Asian countries, while the European Directives on PTW emissions are relatively new. Throughout southern Europe and particularly in Italy, mopeds (two-wheeled vehicles equipped with a less than 50 cc two-stroke engine) are widely used. They represent important means of transport, especially in the large cities, helping to meet daily urban transport needs. In these countries, the use of mopeds is facilitated by the mild climate and it is linked to several factors including ease in parking, agility in traffic jams, inexpensive maintenance and in some countries the absence of any taxation on the sale or purchase of second-hand mopeds.

The exhaust gases of mopeds and two-stroke engine motorcycles emit various pollutants in quantities that vary according to the power (horsepower), age and condition of the engine and the vehicle's state of repair, use and modifications made to its various components after purchase. Vehicles with catalytic converters, that is, manufactured under EURO 1/EURO 2 standard emissions specifications, and ones without catalytic converters differ greatly in emissions of PM and other pollutants. European Union Directive 97/24/EC came into force on 17 June 1999; mopeds manufactured earlier than that date are referred to as conventional. European Union Directive 97/24/EC regulates emissions of carbon monoxide (CO), volatile organic compounds (VOC) and nitrogen oxides (NO_x), but not of particulate matter (PM). Therefore, the European institutions are preparing the amendment of the Directive

97/24/EC on "Characteristics of two or three-wheel motor vehicles". One of the objectives of the future legislation is to lower the particulate emissions from motorcycles, especially from the ones equipped with two-stroke engines. Nevertheless the legal limits for particle mass, or counts are still not taken into consideration, for this sensible market sector. The serious health effects of particle emissions from traffic are known from the discussions about diesel engines technology and legislation. In this context the particle emissions of small 2-stroke engines with lost oils lubrication cannot be neglected any more. Historically, two-stroke engines are known to emit higher levels of PM and HC, but lower levels of NO_x . A particular concern is about the 2-stroke scooters, small motorcycles and 2-stroke 3-wheelers, which in several countries are used very much in congested city centres. At present there is a demand for improved knowledge about particulate emissions from 2-stroke Scooters. Since emissions from other type of vehicles have been dramatically decreased as a result of more stringent emission regulations in many countries, the focus on 2-stroke Scooter emissions is becoming more obvious.

The principal cause of high emissions from 2-stroke two-wheelers is the simple scavenging process (called "crankcase scavenging") that allows 35%+ of the engine's fuel to escape unburned from the engine. Two-stroke cycle engines are utilized due to their rugged construction, low cost, and high power / weight ratio. Two-stroke-powered vehicles include scooters and mopeds ("two-wheelers") and three-wheeled motorized tricycles ("three-wheelers, tuk-tuks, tricycles") that are used for taxi service and as utility vehicles. Two-stroke cycle engines are widely used throughout Asia for personal transportation. A recent estimate in India calculates that 80% of the 2-wheelers are powered by two-stroke engines, although recent sales data for new 2-wheelers indicate a shift toward four-stroke engines. In 1992-93, two- and three-wheelers with two-stroke cycle engines were reported to account for 70% of the total unburned hydrocarbons (HC) and 46% of the carbon monoxide (CO) emissions in India and were a significant contributor to particulate emissions. This is due to the large number of vehicles with two-stroke engines and the particularly high emissions from these engines.

Over the years concerns have been expressed on the contribution of mopeds to urban air pollution through their two-stroke engines as well as to injuries, deaths and disabilities due to the high accident rates involving in most cases young people. One of the study's most important conclusions is that mopeds play a considerable role in producing Rome's urban pollution. Each of the city's estimated 443 000 mopeds covers an average of about 6 000 km annually for a total of 2.58 billion km. These 443 000 mopeds are responsible for 20% of the carbon monoxide and 21% of the PM10 concentrations measured by monitoring stations and represent 17% of the total circulating vehicles in Rome (about 2 600 000). PM emissions from mopeds are responsible for about 350 premature deaths annually, which comprise 1.4% of deaths of people aged 30 years or older in the City of Rome; about 450 people are admitted to hospital for respiratory illnesses (2.2% of total admissions) and 660 for cardiovascular problems (1.0% of total admissions), in addition to other minor health problems.

2. What are the causes of increased PTWs emissions?

Regarding PM emissions, significant differences between 2-stroke mopeds and 4-stroke scooters can be observed. 4-stroke scooters have in general very low emissions, while in the case of 2-stroke mopeds Pre Euro 1 vehicles have hot emissions of about 180 mg/km and Euro 1 have hot emissions of about 30 mg/km. However, experimental data show no clear evidences that Euro 2 mopeds have PM hot emissions lower than Euro 1. No significant differences exist between whole cycle emissions and hot phase emissions for 4-stroke scooters. A higher percentage of the total collected mass from 2-stroke mopeds than 4-stroke scooters shows an aerodynamic diameter less than $1\mu\text{m}$, making them a more significant health problem (Czerwinski and Schramm; 2005).

Two-stroke engines have high emission rates due to three causes: the engine emits high quantities of hydrocarbons including Benzene and other pollutants; it works on a mixture of oil and fuel rather than fuel alone, as with four-stroke engine; and a large quantity of the fuel is vented out unburned.

2.1. The effect of engine technology

Engine technology has a very strong influence on the particle emissions due to differences of mixture tuning, exhaust gas temperature, secondary air supply and catalyst. The particulate matter emitted by motorcycles equipped with 4-stroke engines, even the less modern ones, appeared to be of similar mass and size to that from the conventional gasoline passenger cars, at least in terms of mass per distance, which is the parameter considered by the policy maker. The particulate emitted by two wheelers powered with two stroke engines were much higher in mass and strongly depend on the engine technology. Conventional 2-stroke engines exhibit mass-size distributions with peak towards 200-300 nanometers whereas the direct injection technology produced smaller diameters (Czerwinski and Schramm; 2005).

The actual state of knowledge resulting from the research efforts of 2-stroke manufacturers confirms higher PM emissions and a higher part of solid PM at cold start. Also, the deterioration factor of PM and HC emissions over the lifetime of the engine is very low. Those emissions depend mainly on the actual conditions of the machine, such as lube-oil content, mixture tuning, mixture preparation, combustion, post-oxidation in the exhaust (secondary air system), exhaust gas temperature and scavenging losses (depending on engine construction). For different engines there is a large PM emission variation of about factor 5, thus often making the direct measurement of PM for legislation and control purposes too complicated as it requires too much time and costs. Therefore, for some assessment purposes where these resources are limited, it is proposed to replace the PM measurements using the correlations of PM with CO, HC values and lube-oil content. Engine technology has been found to exert a very strong influence on the particle emissions. The Carburettor scooter has a richer emission mixture, higher exhaust gas temperature until 400°C , a secondary air system (SAS) and quicker light off of the catalyst (at $\sim 100^{\circ}\text{C}$). The associated consequences for emissions include higher PM emissions at cold start, when the catalyst is not active at the beginning of the warm-up phase, and very low PM at warm operation due to post-oxidation, apart from

CM8 mopeds (with DI) that show lower cold than hot emissions, and the production of sulphate PM due to high exhaust gas temperature.

High lubricant and fuel concentrations, in combination with PM emissions, are also the origin of the white smoke formation in the exhaust of 2-stroke two-wheelers. The characteristic white colour is the result of the size and the nonsolid nature of the lubricant droplets in the exhaust of these vehicles. In that respect, Sugiura and Kagaya (1977) and Kagaya and Ishimaru (1988) reported a positive correlation between white smoke and PM concentration in the exhausts of two-wheelers. This correlation may be better established when studying the soluble organic fraction (SOF) of PM emissions, i.e. the particulate mass collected by extracting the PM collection filters in a solvent. Palke and Tyo (1999) mention that SOF analysis shows that only 5 % wt of the trapped PM consists of solid carbonaceous material. Similarly, for a white smoke emitting engine, Sakai et al. (1999) found SOF up to 98 % wt. An investigation by Ricardo, on behalf of ACEM, involving a total of 12 motorcycles, included two 50 cm³ 2-stroke bikes. One was a DI without catalyst, and the other a conventional 2-stroke with catalyst. Ricardo determined a lubricant fraction of > 97% for both. Sugiura et al. (1977) conducted infrared analyses of the extracted component of the PM collected on a filter and found a composition consisting largely of unburned or slightly oxidised lubrication oil. After visual inspection of the PM collected on a filter, a light tan colour was observed, indicating the absence of solid black soot-like material. Czerwinski et al. (2002a) confirmed the absence of soot-like material in the PM emissions of a 50 cm³ 2-stroke moped. They found that the ratio of the PAS (Photoelectric Aerosol Sensor) signal to DC (Diffusion Charger) signal was very low, indicating that the measured particles consisted mostly of SOF (little carbon and carbon-bound PAHs). Carroll et al. (2000) fitted a partial dilution system to the exhaust of a 500 cm³ 2-stroke engine and they compared the PM organic fraction, operating the engine with two different lubricants. The weighted average volatile fraction of PM for a mineral-based lubricant was found to be 73 % and for a biosynthetic lubricant 58 %. The weighted average unburned lubricant contribution to SOF was found to be 67 % and 49 % for the mineral and bio-synthetic lubricant respectively, confirming that lubricant is the primary source of 2-stroke engine PM.

2.2. The effect of after-treatment

Because of the relevant characteristics of the typical 2-stroke PM emission, oxidation catalysts have a significant influence on the reduction of this emission, more than in the case of diesel engines. In a study by the Italian National Agency for New Technologies, Energy and Environment and the Municipality of Rome it was found that particulate matter emissions from non-catalysed moped are strongly related to lubricant consumption. In the case of catalysed moped lubricant in the exhaust gases is oxidized according to catalyst temperature, the higher the catalyst temperature is, the more the lubricant is oxidised, and as a consequence, particulate matter emissions decline as catalyst temperature raises. Another result is that during ECE-47 cycle, particulate matter emissions from catalysed moped are about 75% lower than emissions from non-catalysed mopeds. Similarly, the results from the ANCMA investigation show PM emissions for Euro 1 vehicles in the order of 60-80 mg/km for the conventional 2-stroke with oxidation catalyst, 20-30 mg/km for the DI 2-stroke without catalyst and 15-25 mg/km for the 4-stroke. This should be compared with the current limit value of

50 mg/km for a diesel passenger car (Euro 3) or 25 mg/km for the Euro 4 stage of such vehicles. When coming legislation steps for mopeds will cause DI 2-strokes with oxidation catalysts to appear on the market, as it may do for Euro 2 and is likely to do for Euro 3, PM emissions of less than 25 mg/km may reasonably be expected. The oxidation catalyst can reduce PM of about 40 to 70% - this oxidation can be improved by the secondary air introduction in the exhaust pipe. Oxidation catalysts are 10 – 80 % effective in PM reduction depending on vehicle operation and catalyst formulation, with a mean of this range (40 – 50 %) more commonly reported. Although the catalytic converter appears to be efficient in the reduction of the soluble organic fraction of the particulate emissions, it has a moderate effect on the emissions of HC and CO (Czerwinski and Schramm; 2005).

Czerwinski et al. (2002a) compared the number distribution of particle emissions from a 2-stroke motorcycle with and without catalyst. The results showed that the mean diameter did not change significantly. Authors comment that "the catalyst has no influence, or increases only a little the particle counts". However, in the relevant comparison they demonstrated almost an order of magnitude increase in the maximum concentration with use of a catalyst. *Obviously, more research is necessary to draw any conclusions on the effect of a catalyst on 2-stroke particle size distribution.* However, adequate catalyst maintenance is required in order to achieve the emission reduction mentioned above (Iyer; 2002).

2.3. The effect of lubricant oil specifications

The major share of 2-stroke PM-emission originates from non-combusted lubrication oil. And this is all the more true when such engines are adjusted considerably leaner than was the case before Euro 1. Lubricating oil is introduced through intake air-fuel mixture either by mixing with petrol or by injection. The entire quantity of oil passes through the engine in every cycle of operation (the "once-through" system). Small oil droplets of incompletely combusted lubricating oil emitted through exhaust appear as visible smoke and PM. Over 98% of PM is soluble organic fraction (unlike largely carbonaceous in diesel PM). Lubrication rates have gone down over the years and are going down even further with modern technology. Targeted lubrication by means of an oil pump instead of mixed lubrication (where the lubrication oil is mixed with the fuel) enables the designer to adjust the lubrication rates much more precisely to the needs, and thereby to avoid unnecessary 'over-lubrication'. This situation is all the more true for DI 2-stroke technology.

Patschull and Roth (1994) studied the influence of lubrication oil specifications on the particle size distribution in 2-stroke two-wheeler emissions. Three lubrication oils from the same manufacturer but of different specifications were tested: one mineral oil and two synthetic ester based oils, one of which was biodegradable. Despite the difference in lubricant properties, no significant differences in particle size distribution were found for the three oils. This was observed for all engine operating conditions considered. The same conclusion was obtained by Carroll et al. (2000). In their study, a comparison between mineral and bio-synthetic lubricants showed that the mean diameter of the particle distribution was about the same for different engine loads. What Patschull et al. (1994) found significant for PM emissions was the oil/fuel mixing ratio, defined here as the volumetric ratio of the lubrication oil to fuel. The most significant effect is the reduction of the particle number concentration when the mixing ratio decreases (less lubrication). Furthermore the measured mean particle diameter also

decreased when the mixture became leaner in oil: less lubrication leads to a relatively larger share of smaller particles. Table 2-1 shows some results of this study.

Table 2-1: Effect of the oil/fuel mixing ration on the particle size distribution.

Load [kW]	Volumetric mixing ratio (lube : fuel)	Particle concentration at distribution mode [%]	Dp mode [nm]
3.8	1:25	125	200
	1:50	100	160
	1:75	90	120
	1:100	85	110

Conclusively, PM emission can be influenced to a limited extend by the fuel quality, but it largely depends on the air-fuel ratio, thus it is increased when a richer mixture is used. PM can therefore be reduced roughly proportional with the reduction of the lube-oil ratio.

The influences of lube oils on the particle emissions from several studies were confirmed on DI and gasoline scooters and they are slightly modified on the Carburettor scooter. Changing the fuel quality (Aspen) may increase the condensates with one oil and lower the condensates with another oil. There is a clear evidence of coinfluences of oil and fuel on the spontaneous condensation and on the particle emission parameters. However, it should be taken into account that the influence of oil quality on the (nano) particle emissions varies for different scooters.

Modern synthetic lubrication oils combine better lubrication performance with better combustibility of the exhausted oil, thereby even more reducing the resulting PM-emission.

2.4. Relevant emission reduction methods

From the discussion in the previous section some PM emission control measures for Powered Two Wheelers (PTW) become obvious. For example, the gradual substitution of 2-stroke with 4-stroke two-wheelers is an emission reduction strategy that has already been successfully applied in many Asian countries. As it was also shown that the oxidation catalytic converter can significantly result in lower emission rates, its adequate maintenance and care for its durability are particularly recommended.

2-stroke PM emissions can also be controlled by reducing the oil dosage and with the use of superior 'low smoke' oil. *Further studies are required to reaffirm these findings.*

The unfavourable emissions from carburetted two-stroke cycle engines are primarily due to: 1) short-circuiting of fuel during scavenging, 2) incomplete scavenging, 3) the use of waste lubrication, and 4) poor control of air/fuel ratio (Willson; 2002).

2.4.1. Short Circuiting

In a carburetted crankcase-scavenged two-stroke engine, the combustion products from the previous cycle are forced from the cylinder with a new air/fuel charge. This charge is compressed in the crankcase by the underside of the piston and then enters the cylinder when the piston uncovers the transfer port. Unfortunately, the exhaust port is open during the entire time that the transfer port is open – allowing part of the air/fuel mixture to “short circuit” through the cylinder during the scavenging process. This is the major source of the high hydrocarbon emissions from crankcase-scavenged engines, allowing 35%-40% of the fuel to be lost directly out of the exhaust ports during the scavenging process.

2.4.2. Incomplete Scavenging

The maximum volume change in the crankcase of a crankcase-scavenged engine is equal to the swept volume of the engine. Due to pumping losses, the volume of air/fuel mixture used for scavenging is significantly less than the swept volume of the cylinder (i.e., the delivery ratio is less than unity). There is not enough mixture to completely force the old exhaust products from the cylinder, resulting in high levels of residual exhaust, which remain in the cylinder. This is a major contributor to combustion instability that in turn contributes to high carbon monoxide and hydrocarbon emissions. In order to stabilize the combustion process, richer mixtures are used, again leading to high CO emissions. The efficiency of the scavenging process can be improved by increasing the scavenging volume, but this will increase the hydrocarbon emissions if the engine is scavenged with an air/fuel mixture.

2.4.3. Waste Lubrication

Crankcase-scavenged two-stroke cycle engines use a waste lubrication system. In small two-stroke engines, the oil is mixed with the fuel during refuelling, or the driver purchases “premix.” Some of the oil is deposited on the appropriate components (crank bearings, rod bearings, cylinder walls) while the mixture is in the crankcase. The remaining oil then travels with the air/fuel mixture into the cylinder where it is either short-circuited or trapped in the cylinder. The short-circuited oil contributes to the hydrocarbon emissions. The trapped oil does not burn readily and becomes a major source of the visible smoke produced by small two-stroke engines. Direct-injected two-stroke engines still use a waste lubrication system. However, since the oil is not dissolved in the fuel, it deposits more effectively on the walls and bearings where it is needed. This reduces the oil migration into the combustion chamber, which dramatically minimizes the smoke caused by combustion of the lubricating oil.

2.4.4. Air/Fuel Control

Although high hydrocarbon emissions are inevitable in carburetted two-stroke engines, high carbon monoxide emissions are not. They arise from rich carburettor tuning. In the range at which maximum power occurs, CO concentration levels are typically 1.5% - 2.5%. If a carburettor is set for leaner operation, misfire may occur during acceleration transients. Due to the time delays involved with crankcase scavenging, acceleration enrichment is generally ineffective, so the engine is typically tuned rich at all times. Carburettors are also tuned rich to provide “fuel cooling” of the cylinder to help prevent overheating of the piston. For Carburet-

tor-variant with high exhaust gas temperature the sulphates create spontaneous condensates and take a share of PM.

As it was also reported in the previous sections, high PM emission levels from two-wheelers are primarily caused by “scavenging losses” produced when the fresh air/fuel mixture is used to flush the exhaust gases from the previous stroke out of the engine. Over 35% of the fuel is typically lost in the scavenging process. The crankcase-scavenged two-stroke cycle engine was developed by Sir. Dugald Clerk in the 1890s and is still widely used due to its simplicity, durability, power density and low cost. Technology now exists to produce much cleaner two-stroke cycle engines. This technology relies on “direct in-cylinder fuel injection” (DI), which can be used to reduce fuel consumption by approximately 35% and dramatically reduces particulate emissions. The DI technology is being used by manufacturers in Europe for scooters. In a carburetted engine, there is ample time for the fuel to vaporize to form a homogeneous air/fuel mixture. In a direct-injected two-stroke engine, the fuel is injected into the cylinder late in the cycle, as the piston is returning. The time available for vaporization and mixing is short, so the fuel must be atomized into very fine droplets to allow the fuel to vaporize for combustion. The fuel is injected directly into the cylinder, so the injector must withstand combustion pressure and temperature. These factors provide challenges to designers of DI fuel injection equipment.

Particulates and visible smoke are dramatically reduced through the use of direct injection technology. The same direct-injection technology that reduces emissions also reduces fuel consumption. By eliminating the 35%+ of the fuel which escapes unburned, engine efficiency is increased. By reducing carbon monoxide production, combustion efficiency is further increased. The result is a $\approx 40\%$ + reduction in fuel consumption, producing a corresponding reduction in the production of greenhouse gases.

3. What should be monitored?

It is of particular importance in order to provide future recommendations on the PTW PM emission assessment to determine the PM properties that should be monitored. The characteristic level of PM-emissions from small 2-stroke engines should be established and the question of whether this indeed presents a problem (e.g. when compared to the levels from diesel engines) should be further investigated. The nature and physical properties of such emissions (again in comparison with diesel engine PM emission) should be determined, as well as their likely influence on the repeatability and reproducibility of the measurements. This is necessary to investigate whether such emissions can be measured with the equipment and methods in use for the measurement of diesel engine PM-emission, and if not to recommend in which way would they need to be adapted.

3.1. Physical properties

Several studies have focused on investigating the effect of two-wheelers engine technology upon the physical properties of particles (Faberi et al.; 2004). Mopeds emit particles because of the presence of unburned engine oil in their exhaust (Palke & Tyo; 1999, Santino & Picini; 2001). The PM (defined as the portions of exhaust gases trapped in a filter in the form of both aerosols and solid particles) in exhaust emissions comprises more than 95% unburned

or slightly oxidized (non solid) engine oil, and the remaining 5% comprises solid carbonaceous and inorganic PM (Palke & Tyo; 1999). A study by the Japan Automobile Research Institute on the composition of the PM emitted by two-stroke engines (Takayuki et al.; 1999) confirms Palke & Tyo's 95%, specifying that these are soluble organic particles, differentiating them from those emitted by diesel engines, also in terms of their higher carcinogenic characteristics. Very few studies have dealt with the measurement of this pollutant. Those available were done by the Italian National Agency for New Technologies, Energy and the Environment (Santino & Picini; 2001) and Society of Automotive Engineers (Laimboch; 1991, Palke & Tyo; 1999) and Labeco Italia Srl. The vehicles studied were both catalytic as well as conventional, two- and four-stroke and larger and smaller than 50 cc. What makes this study so interesting is the type of the test carried out, consisting of repeated trips along two typical tracts of urban roadway in Bologna, each requiring about 40 minutes of travel time. Although none of the studies examined indicates the size of the PM emitted by the two-stroke engine, the technicians who designed the tests indicate that all particles emitted were either PM10 or smaller. The bench-check technicians all agreed in this assumption, as a result of both the nature and size of the test filters and the nature of this unidentifiable PM as a fine hydrocarbon aerosol (interviews with the Italian National Agency for New Technologies, Energy and the Environment, National Research Council Institution Motori CNR and Labeco Italia Srl). A very close relationship between oil consumption and the PM emissions of non catalytic mopeds supports the hypothesis of the fine aerosol. It is not known how the particles are transformed in the atmosphere or their role in the formation of the PM10 concentrations measured in the city.

Table 3-1: Emission factors of two-stroke engine mopeds.

		Conventional	EURO 1 (catalytic converter)
CO	g/km	15.00	7.50
Non-methane VOC	g/km	9.30	4.20
NO_x	g/km	0.03	0.03
Benzene	mg/km	156.70	16.40
PM	mg/km	91.40	33.90
1,3-Butadiene	mg/km	48.00	0
Benzo[<i>a</i>]pyrene	µg/km	2.15	0.40
Formaldehyde	mg/km	15.00	0.50
Acetaldehyde	mg/km	5.50	0.30

(from: Effect of motorcycle engine technology upon physical properties of nanoparticles BONNEL Pierre, MARTINI Giorgio, KRASENBRINK Alois, DE SANTI Giovanni European Commission – Joint Research Center Via Enrico Fermi,1 – 21020 Ispra (VA) – Italy).

Though almost entirely dominated by oil volatility hydrocarbons, the chemical composition of motorcycle particulate matter also appears to be related to both cycle demand and to calibration.

3.2. Particle size distribution (number and mass weighted)

Various studies show concentration data. One should, however, be aware that concentration numbers are dependant on the definition of such numbers (as measured or 'normalised'), on the fact that concentrations may have been measured in either raw exhaust or in diluted exhaust, which obviously will affect the volumetric concentration, and that the basis for the size distribution may be either mobility diameter or aerodynamic diameter. It is also expected that the particle size distribution from 2-stroke vehicles may significantly differ according to sampling conditions.

Regarding the particle size distribution there seems to be a tendency for 2-stroke PM to contain greater numbers of particles of a smaller size for similar mass emissions. There is scarce information in the literature on PM and particle characteristics from 2-stroke engines. Nevertheless some consistent conclusions can be drawn:

- The PM emission level is found in the 10 – 500 mg/km range. This is a wide range because the emission rate depends on a number of factors (oil / fuel mixing ratio, use of a catalyst, etc.), as it was explained in the previous chapter.
- The larger fraction of PM consists of soluble organic non-carbonaceous species (SOF). SOFs are reported in the 55 – 98 % range of total PM. Larger values are reported for white-smoke emitting vehicles. Due to this reason, PM sample filters are tan- rather than black-coloured. It can be concluded that high SOF originates from unburned lubrication oil and fuel escaping combustion.
- There is also a large range of particle size distributions reported with log-normal peaks occurring in the 40 – 200 nm range. The upper size limit in this case is beyond what is usually reported even for Diesel vehicles. Particle concentration is typically high, in the 10^8 to 10^9 cm³ range.

As part of a UK Government programme to determine emissions factors for a variety of vehicles, further study was undertaken at Ricardo Consulting Engineers to measure individual chemical species of current and possible future concern (Czerwinski and Schramm; 2005). The aim of the study reported was to investigate the unregulated emissions from a range of twelve 2- and 4-stroke motorcycles with respect to various drive cycles, engine and after-treatment technologies. Particulate mass emissions from motorcycles were found to be generally low, with the exception of the carburetted two-stroke (0.63 g/km) these did not exceed 0.02g/km – inside the Euro 4 emissions levels (0.025g/km) for a diesel vehicle irrespective of drive cycle. The total particle number emissions from the 97/24/EC, 98/69/EC and WMTC cycles ranged from below 10¹²/km to greater than 10¹⁴/km – levels that are in line with the magnitude of emissions observed from light duty MPI and G-DI passenger cars over a range of operating conditions.

4. Monitoring methods for PTWs

4.1. Measurement considerations

The sampling devices: thermoconditioner and thermodesorber influence clearly the aerosol in the sense of elimination of condensates by increasing the sampling temperature – these influences depend on the composition of aerosol. The sampling place: tailpipe, or CVS influences strongly the particle size distributions (PSD) due to condensation – and coagulation effects.

Current CVS techniques used for diesel PM would be inadequate since large variations occur on account of dominant nucleation mode (Rijkeboer et al.; 2004).

4.2. Effect of engine load and speed

Patschull et al. (1994) conducted a series of tests to evaluate the effects of different engine operating conditions on the size distribution of exhaust particles of a 2-stroke engine. They used a single cylinder 80 cm³ engine and the exhaust gas was diluted in a mini-dilution tunnel. Particle size classification was performed by means of a Differential Mobility Particle Sizer (DMPS). The engine load was kept constant, and it was found that the mean particle diameter was about 150 nm for the lowest speed and decreased with increasing speed, reaching 90 nm at full speed. On the other hand the concentration increased from 108 cm³ at the lower speed up to 108 cm³ at maximum speed. These values are 'normalised'. Carroll et al. (2000) tested a 500 cm³ 2-stroke engine of a snowmobile using a partial dilution system. They too observed that as engine speed increased, the mean particle diameter decreased, in this case from 81 nm at idle to 23 nm at full speed. The particle concentration was highest at the mid-speed with a value of about 108 cm³, as measured (this would be approximately 13x108 cm³ normalised), whereas at high speed or idle the particle concentration reached 107 cm³ as measured (or 5x107 cm³ normalised). These higher particle concentrations at partial load were ascribed to higher scavenging losses and poorer combustion quality than at full-load operation, which is typical for 2-stroke engines.

The Italian National Agency for New Technologies, Energy and the Environment (Santino & Picini, 2001), Laimboch (1991) and Palke & Tyo (1999). The emissions of a two-stroke motor vary considerably during the transition time to reach optimal speed and revolutions per minute. This study therefore assumes the following values as PM emissions factors: 243.4 mg of PM per km travelled for conventional mopeds and 29.0 mg of PM per km travelled for mopeds with catalytic converters.

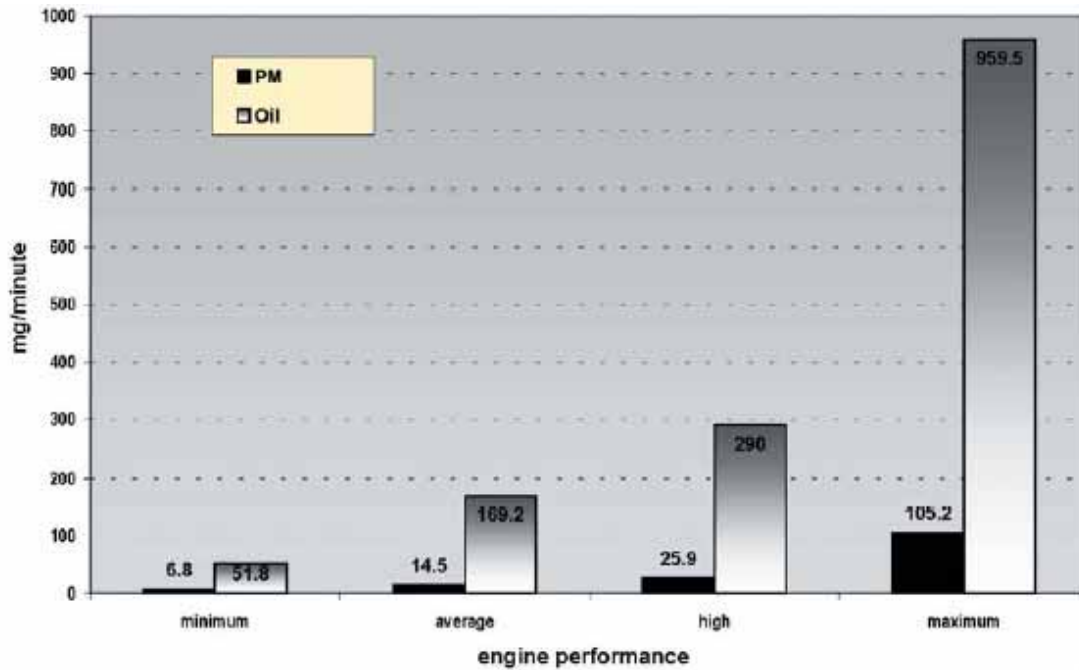


Figure 4-1: PM emissions and oil consumption of two-wheelers.

The dynamic tests were conducted on a chassis dynamometer following the regulated European test cycles (ECE R40 and 47). The sampling conditions were identical to those used for diesel passenger cars, i.e. a dilution tunnel whose flow rate was kept constant during the entire testing campaign. The total mass and the mass versus size distributions were measured using a Low Pressure Impactor (LPI). Some steady state measurements were also conducted with a Scanning Mobility Particle Sizer (SMPS) to investigate the effect of vehicle speed upon the particle concentration.

The detection limit (in km^{-1}) was checked for each vehicle and driving condition. There are some orders of magnitude difference between the detection limit and the actual measurement levels even for nano-sized particles. For all three vehicles there is a distinct difference between the low speed and high-speed condition: In high-speed driving a bimodal distribution is formed, while a log-normal distribution is seen for low-speed driving. For the two cases of low-speed, the distribution appears log-normal and at a much lower concentration than in the case of the high-speed driving. It is important to note that the concentration of the catalyst equipped vehicle appears at one to two orders of magnitude lower levels than that of the non-catalyst vehicles for similar driving conditions. It should also be noted that for the non-cat vehicles this is a wide-open throttle condition while for the larger and more powerful motorcycle it is only a partial load driving mode. Whether this creates a tendency for higher or for lower numbers seems to depend very much on the actual vehicle, however.

4.3. Test cycles

The fact that for motorcycles a completely new test cycle is in the course of development, since it was felt that the existing cycle according to ECE 40 should be regarded as by now totally inadequate, throws up the question to what extent the present test cycle for mopeds according to ECE 47 is still adequate. The ECE 40 test cycle was adopted in the late seventies because a test cycle was urgently needed, the cycle chosen did already exist for passenger cars, and there was no outlook at the time on anyone starting the development of a dedicated cycle for motorbikes. Furthermore also the industry recognised these shortcomings of the existing cycle and was actively co-operating in the development of its successor the WMTC (World Motorcycle Test Cycle). The situation with the ECE 47 cycle is significantly different. For the ECE 47 Regulation a special test cycle was developed right at the start (Rijkeboer et al.; 2003). And although the original proposal was subsequently subjected to far reaching adaptations by the ECE-GRPA (mainly for the fact that mopeds were totally different things in the different Member States), the final product still has some real relationship with the actual use of this class of vehicle. For this reason the test cycle may still be regarded as a sufficiently adequate representation of the real-world use of mopeds, or at the very least that its updating is far less urgent than that of the ECE 40 test cycle. This is also the feeling of the industry.

The emission characteristics of the PM emission were determined over a number of steady state conditions (constant speeds of 30, 50, 70 and 90 km/h) and a small number of driving cycles. These driving cycles were the standard ECE-40 test and two real-world driving cycles as determined by the 'Fachhochschule Biel' in Switzerland: an inner city cycle (FHB Zentrum) and a suburban cycle (FHB Peripherie).

Another question is, however, how to include a cold start. This is, however, of particular importance, as it has been confirmed, that the cold start (summer temperatures 20 - 25°C) causes a considerable increase of CO, HC and of nanoparticles. The emission-relevant warm up period of investigated scooters was 1.5 min and the emission-relevant cool down period at 25°C was about 30 min. As mentioned before, PM collected on filters is normally tan coloured. However, Palke and Tyo (1999) found on a 100 cm³ 2-stroke motorcycle that after a cold start and before the engine reaches thermal stabilisation, PM samples show a black colour, suggesting a high solid carbonaceous and/or inorganic content. In absolute levels the cold-start related PM emissions were much higher than during a warm ECE-40 procedure. It is plausible that poor combustion caused by cold operation, and a rich mixture during 'choke' conditions are responsible for an increased production of black carbonaceous particles. One needs to consider that this also occurs in 4-stroke engines and it is not a special characteristic of 2-stroke ones. An additional cold-start related effect was reported by Czerwinski et al. (2002a), who tested a catalytic 2-stroke moped scooter at a steady speed of 30 km/h following cold start. The authors report that during the warming-up period (until about 120 s after the start) there was a blow-out of solid nanoparticulates, which had previously been deposited in the catalyst. The same situation was also observed over a transient cycle.

Referring to the aerodynamic diameter mass distribution, 99% of total collected mass was found to have an aerodynamic diameter less than 1 µm. For EURO I mopeds results showed

that oil consumption during hot phase is close to that measured during cold phase but the oxy-cat reduced emissions by about 80%. Aerodynamic diameter mass distribution showed almost the same pattern as pre-Euro I mopeds. For EURO II mopeds, hot and cold emissions were different. Oil consumption and cold emissions of the CM7 moped were close to those of the EURO I mopeds, but its hot emissions were very low. In contrast, CM8 mopeds in spite of lower oil consumption had hot emissions higher than expected. Moreover cold emissions were lower than hot emissions.

At this moment the procedure starts from cold, after a prescribed preconditioning. The total test then consists of 8 sub-cycles, of which only the last 4 are sampled. Our proposal is to keep the preconditioning as it is, and to sample all 8 sub-cycles, rather than to sample only the first 4 sub-cycles, as might be an alternative. The length of a test of 8 sub-cycles, although not completely fixed, is about 5 km, and one cold start on a total test length of 5 km should be regarded as more representative than one cold start on a total test length of 2.5 km. In the old set-up a total test length of 2.5 km was adequate, since no cold start was involved. Table 4-1 gives an overview of the length of all known cold start tests. India is the only exception with a very short test.

Table 4-1: The test length per cold start for various tests.

Testcycle	Total length per cold start	Comments
70/220/EEC (current)	11.0 km	Passenger car cycle
ECE 15/ECE 40	4.05 km	As used in Taiwan
ECE 47	~5.0 km	Not currently used in combination with a cold start
IDC	2.5 km	India: 50 cc motorcycles
WMTC (draft)	5.2 km	Class 1 Motorcycles

Real data about actual trip lengths are hard to obtain. In a recent investigation for the Dutch authorities information obtained from the Dutch 'Central Bureau of Statistics' on the one hand suggested that the majority of trips fall in the category 5-7.5 km, whereas on the other hand the total annual mileage worked out at something like 1800 km. This would imply an annual number of trips lying between 240 and 360, or less than 1/day on average. Apparently there is a discrepancy between these figures. The industry pleads for a weighting between the cold start part (sub-cycles 1-4) and the hot part (sub-cycles 5-8) of 0.3 / 0.7, on the grounds that only the first start in the morning is a true cold start, whereas any subsequent start is not truly cold. The consultant feels that a straight addition (effectively resulting in a 50/50 weighting) might be the best approach in the absence of more accurate data. Alternatively more data would have to be obtained (Rijkeboer et al.; 2003).

4.4. Emission factors

PM emission factors can be represented as a function of amount and type of lubricant for well maintained 2-stroke motorcycles without any advanced emission control technologies. Emission factors are approximations based on limited available data and are indicative of trends.

Table 4-2 lists typical PM emission factors of a diesel passenger car and a range of current 2-stroke vehicles to provide a comparison of expected particulate emissions from powered two-wheelers (PTWs). The diesel vehicle emission factor originates from the COPERT III model (Ntziachristos and Samaras; 2000) and is obtained for an urban average speed of 25 km/h. PTW emission factors correspond to the ECE-47 cycle tests for mopeds (Santino et al.; 2001) and to the ECE-40 cycle for motorcycles (Palke and Tyo; 1999 and Kojima et al.; 2000).

Table 4-2: PM-emission factors comparison for different vehicles.

Vehicle	Average speed (cycle) [km/h]	Emission factor [mg/km]
Diesel Passenger Car Euro 1	25	91
Non-catalyst moped	~30 (ECE-47)	172
Catalyst moped	~30 (ECE-47)	43
Non-catalyst motorcycle 100 cm ³	~25 (ECE-40)	330 (190 – 470)
Catalyst motorcycle 100 cm ³	~25 (ECE-40)	10 - 20

It is obvious that there is a wide range of emission factors reported for PTWs, depending on their engine and emission control technology. However, it can be said that the average PM-emissions from 2-stroke two-wheelers are in the same order of magnitude as those of a diesel passenger car.

For estimate of emission factors driving cycle parts with different average speeds were performed. During the warm measurement these driving cycle parts are repeated one after the other, with sampling of the diluted exhaust gas in separate bags. The driving cycles, which were really performed on the chassis dynamometer were measured and the characteristic parameters (v_m , a_m , etc.) of those cycles were evaluated. This allows to consider the differences between the desired (conducting) and the realised (real) driving cycle. These differences can be particularly significant for the low-power vehicles, which cannot always follow the conducting (desired) driving cycle. The development of cycles and the relationships of average speeds and accelerations also have to be considered. The driving resistances of the test bench can be set according to the national exhaust gas legislation.

Diffusion charging is a relatively new technique for determining the active surface of particles. This technique has been used to record in real time the active surface of particles emitted by the 2-stroke vehicles (Rijkeboer et al.; 2003). From the investigation it is obvious that high surface concentrations occur mainly during accelerations and not at steady speed driving. This is consistent with the observation of white smoke emissions from the vehicles. The

high surface concentration per distance driven suggests that current two-wheelers may be significant contributors to atmospheric photochemical reactions and gas-to-particle transformations.

The particle measurement procedure and equipment feature a dedicated partial dilution system developed for particle characterisation analysis. The main part is the primary dilutor where a small portion of the exhaust gas enters and is diluted with dehumidified and filtered air. The primary dilutor is designed to provide rapid turbulent mixing of the exhaust gas. After the primary dilutor, the diluted exhaust gas stream is divided into 2 branches, called "dry" and "wet" branches by convention. The dry branch is designed to sample only non-volatile particles up to 250°C (solid particles) while the "wet" branch measurements correspond to the total (volatile and non volatile) concentration. The sample flow is led through a thermode-nuder to remove volatile material, before it is fed into an Electrical Low Pressure Impactor (ELPI). In the wet branch the main quantity of the sample enters a gravimetric impactor (DGI). The DGI was only used as a filter holder for cross-comparison of the particle mass with the PM determined at CVS conditions. A smaller quantity of the sample from the "wet" branch is further diluted and fed to a diffusion charger (ASMO) and a differential mobility analyser (DMA). The diffusion charger is used to record in real-time the active particle surface. The DMA is used to classify particles according to their mobility diameter. The number concentration of single charged particles is then determined by means of a condensation particle counter (CPC). Under steady state conditions, DMA and CPC can be linked (with the CPC located after the DMA and the ELPI over the ECE-47 cycle) to build a Scanning Mobility Particle Sizer (SMPS) which can provide the number-weighted size distribution of particles in the range 10 nm – 400 nm. The flux recorded corresponds to the ELPI filter stage which measures particles in the range 7 – 30 nm aerodynamic diameter. Because of the different width of the DMA and ELPI channels, both results have been normalised to the width of each channel (dN/dlogDp). However, this does not correct for the different expression of size (aerodynamic diameter for ELPI versus mobility diameter for DMA). In principle, the solids flux should never exceed the totals one. In cases where this occurs it should be attributed to the difference in the size expressions and size ranges reported for each case.

A more detailed description of the sampling and analytical techniques for PM emissions from two-wheelers is given in the following section.

4.5. Monitoring instruments

Sampling point for nanoparticle analysis and the possible conditioning of the sampled gas probe have a strong influence on the measured particle counts and particle mass due to the effects of spontaneous condensation (HC and/or sulphates), coagulation, diffusion losses, re-evaporation. These effects can overlap on each other differently for the different substances, which are present in the aerosol. *The sampling, dilution and conditioning should follow as closely as possible the real world atmospheric conditions.*

The methods and their main characteristics are as follows:

ELPI, Electrical Low Pressure Impactor: It is a 12 stage cascade impactor that measures particle number concentrations as a function of aero-dynamic diameter. The Particle size distribution is based on aerodynamic diameter and it features a low size resolution but a high

time resolution down to 1 second. An impactor is a size selective element where the aerosol is guided through a nozzle behind which a plate forces the gas flow to change direction abruptly. Due to their inertia, particles cannot follow the bent gas streamlines but are driven towards the plate. Only friction with the gas makes them accommodate with the new flow direction. Depending on their size-to-mass ratio - that is, their mass density the particles are deposited on the plate and thereby removed from the aerosol flow. The diameter defined by this separation process is called the aerodynamic diameter. The size limit between particles that pass the impactor and those being deposited on the plate – the cut-off diameter - depends on the velocity in and pressure drop across the nozzle, the geometry of the impactor and, as stated before, particle mass density. *By variation of these parameters, usually the pressure behind the nozzle, the cut-off diameter is adjusted to the desired value.* In the ELPI, 12 impactor stages operate in sequence, each with a smaller cut-off diameter. Thus, particles collected in one stage belong to one size class which is limited by the cut-off diameters of the two subsequent impactor stages. After size classification, the deposited particle load has to be quantified. In classical impactor design, the particle mass deposited on the plate is determined by gravimetric analysis, but this technique has shortcomings as it needs high particle loads to be accurate, and analysis is carried out after measurement - the method is off-line. In order to make ELPI an on-line instrument, aerosol particles are electrically charged before they enter the cascade impactor. Each impactor collector plate is connected to an electrometer amplifier. Thus, instead of collecting deposited particle mass, the plates take up the electric charge of the impinging particles and pass it on as measurement current. Number size distributions are calculated from the measured current, the size dependent average charge per particle, and the deposition probability per impactor stage. The time resolution is around one second, while the covered particle size range is between 30 nm and 10 μ m.

SMPS, Scanning Mobility Particle Sizer: It determines the Particle size distribution based on mobility diameter. It features a high size resolution but a low time resolution, typically 1 to 3 min scan time. SMPS provides a method to determine the number size distribution of sub-micron aerosol particles. It uses a measurement system consisting of an electric charger, a mobility classifier, a counter, and PC control. The aerosol is first guided into a neutralizer. This is a chamber in which the air is ionised by beta rays from a radioactive source (^{85}Kr). Positively and negatively charged ions diffuse onto the surface of the aerosol particles until a charge equilibrium is established. The probability for 100 nm particles to carry one elementary charge is about 20% for positive and 40% for negative charging, while the rest of the particles leave the neutraliser with zero electric charge - hence the name “neutraliser”. The charged aerosol particles are now classified by their mobility in a Differential Mobility Analyzer (DMA). The DMA is a long condenser through which a laminar flow of particle free air is guided. For practical reasons it has cylindrical geometry. The aerosol particles enter through a narrow slit at the upper end of the DMA. When a voltage is applied between the outer and inner electrodes, the charged particles are dragged towards the centre. Their velocity is determined by the equilibrium of electric force and friction with the suspension air - small particles move faster, larger ones are slower. While the particles move in radial direction, the laminar air flow carries them along the cylinder axis. Thus, they hit the centre electrode below the inlet, large particles later and further downstream than small ones. At its lower end the centre rod has a narrow outlet slit where particles of a certain size arrive. This size can be tuned by varying the voltage applied to the electrodes. The other particles are either de-

posited on the walls or they are transported out of the DMA with the sheath air. The number concentration of the size selected particles is determined using a Condensation Particle Counter (CPC). Since the particles are too small for optical detection, they have to be magnified by condensing volatile material onto their surface. This is accomplished by guiding the fine particles through saturated vapour of butanol (or vapour of another volatile substance) and subsequently cooling the mixture. The vapour molecules use the aerosol particles as condensation nuclei for the formation of droplets, one out of each aerosol particle. The final size of the butanol droplets is around one micron which is just large enough to be detected using light scattering. By computer control the DMA voltage is increased so that larger and larger particles are fed into the CPC. The software combines CPC counts with particle size calculated from the DMA voltage and calculates a size spectrum.

Thermodesorber: The thermodesorber is used to discriminate solid from volatile components in a mixture of aerosols. It consists of a heater where the aerosol mixture can be heated to 330 °C, and a charcoal trap that absorbs evaporated aerosol components. Due to their high mobility, vapour molecules diffuse into the pores of the charcoal granules and are absorbed while the solid, comparably heavy aerosol particles drift past the granules on almost straight trajectories. Therefore, solid, non-volatile particles are not affected by the thermodesorber. By lowering the heating temperature, less substances evaporate due to their individual boiling point. From the temperature at which certain particles appear, their material can be concluded. This operation mode of the thermodesorber is called thermogramm.

NanoMet: NanoMet is a modular measurement system for on-line characterization of aerosols, consisting of a diluter, an evaporation tube, two sensors and various accessories. Being both robust and user friendly NanoMet is also well suited for field use. The following NanoMet components are normally used:

- a mini-diluter to reduce the aerosol concentrations to levels within the nominal ranges of the NanoMet sensors
- a thermal conditioner (evaporation tube) to evaporate remaining droplets of volatile material still present after the diluter
- two real time sensors, the diffusion charging sensor (DC) for active surface and the photoelectric aerosol sensor (PAS) for soot surface

Evaporation tube: By definition, DPF efficiency is related to solid exhaust gas particles. In addition to solid soot and ash particles, exhaust contains many volatile substances that pass the DPF as vapour: hydrocarbons (HC), sulphate and water. When the exhaust cools down to ambient temperature these volatiles tend to nucleate and condense into nanosized droplets. As most particle analysing instruments cannot distinguish droplets from solid particles, the two fractions have to be separated in the sampling system in order to get a valid measurement of the DPF efficiency. A very straightforward method to remove droplets of volatiles from the exhaust gas sample is to evaporate them in a heated tube. If the exhaust gas sample contains volatiles in high concentration, they will re-nucleate into new droplets after the heated tube, and there will be no benefit of the thermal treatment. However, if the exhaust sample is diluted before entering the evaporation tube, concentration of the volatiles will be low enough to keep them in vapour phase after heating. This method is called post-dilution

thermo conditioning (PDTC). PDTC does not remove the volatile substances from the exhaust sample, but it keeps them in vapour phase rather than in particle phase.

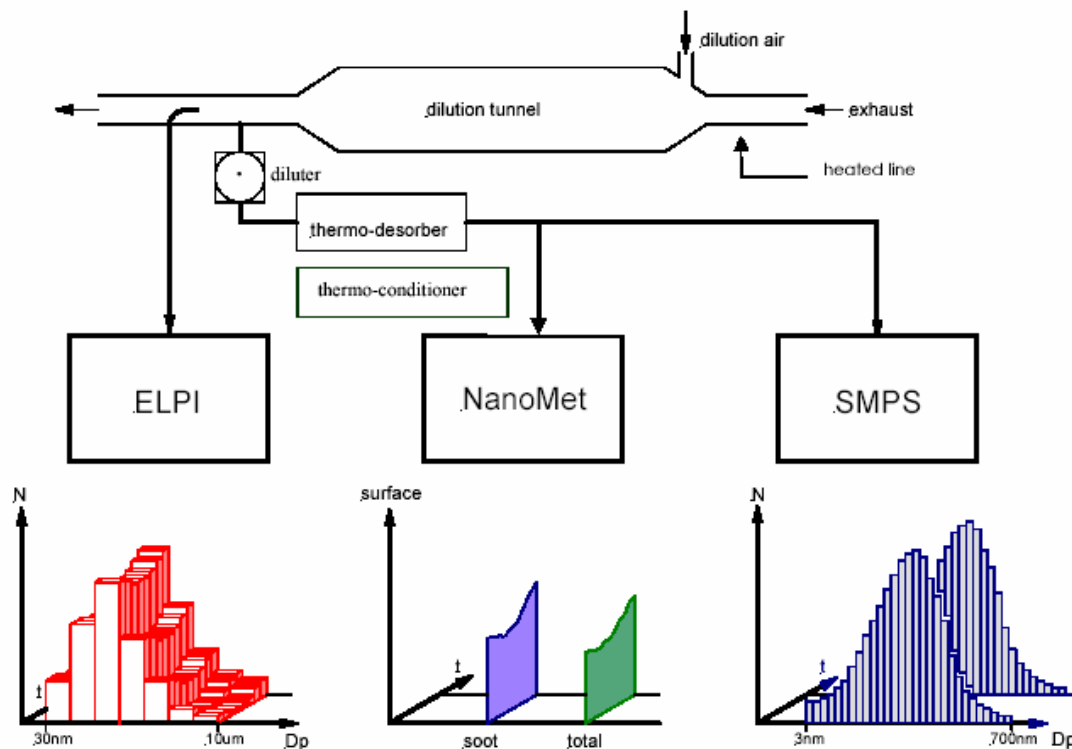


Figure 2: Nanoparticle measurement systems, particle characterisation and time resolution

4.6. Analysis of samples

4.6.1. Coulometric analysis of the PM-filtrate residue (giving EC+OC)

Carbon: In Germany, investigation of the filter residue with respect to elemental carbon EC and organic carbon OC is the official reference method (under guideline VDI 2465) for the measurement of soot in emission and workplace situations. In Switzerland the SUVA supervises diesel emissions at workplace using the same method. Measurement principle:

- The carbon and its compounds from the filter are burnt in a two stage process; the resulting carbon dioxide is determined by coulometry.
- The hydrocarbons adsorbed on the soot particles are purged in nitrogen flow and oxidized with CuO.
- The inorganic carbon fraction is burnt in oxygen flow.

In each step of the reaction, gaseous products including carbon dioxide are formed. They are guided into the measurement cell of the coulometer which is kept at constant temperature and is filled with highly alkaline barium perchlorate solution. The carbon dioxide falls out as barium carbonate, thereby decreasing the alkalinity. The resulting pH change is measured as change of the electric potential of a glass electrode. Finally the adsorption solution is titrated

back to the initial pH value. The amount of electric charge needed in the last step is proportional to the absorbed volume of carbon dioxide, and therefore proportional to the carbon content of the sample. The coulometric method detects elemental carbon (EC) as well as organic carbon (OC), i.e. the sum of both quantities, total carbon (TC) is determined.

4.6.2. Analysis of SOF/INSOF and Sulphates of PM

Solvent method: This is a 3-step procedure

- In order to determine the HC-content the sample (PM-sample on PALLFLEX glass fiber-filter) is extracted with dichloromethane and the weight of the extracted material (= sum of HC) is determined by comparing the remaining mass of the sample after evaporation of the solvent to the original total mass
- In order to determine the sulphate-content a similar extraction is performed using water/isopropanolmixture
- The remaining mass after these 2 extractions is insoluble material, usually taken as soot but mixed with other INSOF-materials like wear metals, possibly insoluble minerals and additive metal oxides. Standard deviation of this method is about 20-30 µg per filter for HC and Sulphate and 40 µg per filter for soot

4.6.3. Quasi-coulometric method (MEXA-1370 PM)

The MEXA-1370 PM can measure the main particulate fractions (soot, SOF, sulphates) separately by detecting the CO₂ and SO₂ gas that are generated from vaporization, oxidation and reduction processes of filtered PM samples. The first step of the measurement consists in putting a sample filter into a combustion furnace at 980°C where N₂ gas flows as a carrier. Under these conditions the volatile organic fraction of PM is completely vaporised and then oxidised into CO₂ by O₂ gas supplied into a dedicated compartment of the furnace. Thus SOF mass can be calculated by measuring the amount of CO₂ generated in this process. After completing the vaporization of the organic carbon fraction, the carrier gas is switched to O₂ so that the elementary carbon is converted into CO₂ by combustion. Soot mass can be calculated from the amount of CO₂ generated at this stage. Sulphates are also vaporized and converted into SO₂ at the same time: similarly, the sulphate mass can be calculated by measuring the amount of SO₂.

4.6.4. PAH-content in PM

Following analytical methods for detection of particulate-associated PAH are applied at JRC Analytical Laboratory: The principal stages in the measurement of PAH are sampling, extraction, clean-up and analysis. The method used in this work has been developed and validated in JRC laboratories on the bases of the EPA method TO13 and ISO/DIN 12844. In brief, the PAH in the PM sampled on the Teflon-coated glass fiber filters are extracted in an automatic Soxhlet extractor for 2 h with dichloromethane. The extracts are evaporated to 500 µL and solvent exchanged to hexane with a Turbo-vap™ instrument using a mild stream of nitrogen and a slight heating (30 °C) of the evaporation vessel. The extract was transferred to a solid phase extraction cartridge for clean-up. The bulk of nonpolar species is removed by elution with hexane and the PAH are eluted with hexane/dichloromethane (40:60) followed by ace-

tonitrile (0.1% triethylamine). After evaporation the samples are collected in toluene containing d12-chrysene as internal standard and analyzed by gas chromatography-mass spectrometry (GC-MS) using electron ionization (EI). The MS is run in the single ion monitoring mode and quantitative analysis is carried out by integrating the areas of the molecular ions (M+) of the internal standards and the PAH. A standard mixture spiked with a known amount of internal standard is used for response factor calculations and for determination of retention times. For data reduction the toxicity equivalency factor (TEF) an approach is used, in which each individual PAH is assigned a toxicity rating relative to benzo(a)pyrene that is set to unity. The benzo(a)pyrene toxicity equivalents (TEQ) of a given sample are calculated as the sum of the concentrations multiplied by TEF over all the measured compounds. Although the accuracy of this approach may be debatable, it is certainly based on vast toxicological data and represents a convenient way to reduce a multidimensional data set in a meaningful way. The TEF approach has found widespread use for chlorinated dibenzodioxins and dibenzofurans.

4.6.5. Non-volatile fraction (NVOF, or non-VF) of the collected particles

Non-VF is determined by heating the filters with collected PM in an oven under inert (N₂) atmosphere. This method differs from the standard Soxhlet method which involves extraction of the organic particulate fraction in an organic solvent (e.g. dichloromethane). Apart from the different practices, Soxhlet extraction also mainly separates the organic species and leaves most of the sulphates on the filter, while inert atmosphere evaporation removes also most of the hydrated sulphates from the particulate phase. Hence, the two methods mainly differ in the treatment of the sulphate fraction (so SOF is soluble organic fraction while VF is volatile fraction). In the case of 2-stroke engine emitted particles, it may hold that because of the large lubrication oil fraction in the particulate phase, higher temperatures and/or residence times are required to fully evaporate heavy hydrocarbons. If this is true, then some VF still remains on the particulate phase in this method and as a result non-VF is overstated. Another aspect is that especially mopeds driven over the ECE-47 cycle operate under full load conditions, which require fuel-rich combustion mixtures. In such cases, soot particles have been reported to be emitted also from premixed combustion engines such as 2-stroke ones.

5. Assessment difficulties in the case of PTWs

There are at least three operative difficulties in assessing the amount of pollution that can be attributed to mopeds. The first difficulty is a general one and lies in the complex relationships linking the PM emitted by the various classes of vehicles to the pollutant concentrations recorded by the monitors. The problem is how much of the PM₁₀ concentrations are produced by motor vehicles. Thus, the amount of natural or industrial fine dust not attributable to vehicles needs to be determined. Fillinger et al. (1999) have estimated that the proportion of PM₁₀ related to vehicular traffic increases with the increase in the concentrations found. *For concentrations greater than 40 µg/m³ the amount related to traffic is estimated to be 58%.* The second difficulty lies in the calculation of the specific pollutant emissions for a given configuration of the total vehicles circulating in a city. *Thus, the more calibrated the model for calculating vehicles and emissions, the greater the vagueness of the individual variables required by the model.* Emission assessment models require input data such as the average distance covered and average speeds for each category of circulating vehicle, and for each year of production, in the city under examination. The third difficulty concerns the main pollutant considered by the health impact assessment which is PM₁₀. The PM measured in vehicular emissions is composed of aerosols, which partly combine with the atmosphere and differ from what is collected and measured by the monitors. Moreover, the particles are carried even at a great distance, which makes establishing a direct relationship between emissions and concentrations measured in proximity to a tract of roadway difficult. Specific site studies do not offer much information in this sense, but it can be supposed that the monitors also measure particles produced far from the roadway site where they are located. The study concludes by maintaining that *PM measurements represent an overall picture of the city and not of a specific area (Faber et al.; 2004).*

Measurement of particle distribution characteristics from engine exhausts is prone to sampling artefacts because of the sensitivity of the exhaust aerosol to sampling conditions. Because of the fact that particle emissions from 2-stroke engines mostly consist of lubricant droplets, the condensation or evaporation of these droplets on the exhaust or transfer hoses may be an additional source of measurement artefacts. Depending on the dilution ratio and the temperature to which the line downstream of the exhaust pipe and the dilution tunnel is heated, these droplets can condense on the line walls before entering the measurement instruments. Therefore, the sampling devices, such as thermoconditioner and thermodesorber, influence clearly the aerosol particles in the sense of elimination of condensates by increasing the sampling temperature – these influences depend on the composition of aerosol. The sampling place, whether it is tailpipe or CVS, influences strongly the particle size distributions (PSD) due to condensation – and coagulation effects. Also in mass measurements, the oil samples condensed on filter papers can be lost as a result of the passage of gas through the filter (Kojima et al.; 2000). The most important processes affecting aerosol characteristics are outlined in the following list.

- Thermophoresis causes particle loss to the walls due to a temperature gradient from the exhaust gas to the colder wall of the pipe.

- Diffusiophoresis occurs from the Brownian motion of particles. The diffusion coefficient increases with decreasing particle size. Diffusion may lead to particle loss when particles attach on the wall due to the process.
- Coagulation typically occurs at particle concentrations exceeding 10^8 cm^{-3} and shifts the size distribution to larger diameters. The rate of coagulation depends on concentration, temperature and particle size.
- Condensation is another process that causes particle growth or even particle formation. When the exhaust gas cools down in the exhaust pipe, volatile hydrocarbons or sulphuric acid may condense onto the particles. Furthermore, if the concentration of the pre-existing nuclei is low, volatiles may even condense to form droplets (new particles).

Due to the liquid nature of 2-stroke particle emissions, such phenomena are more important and one needs to take their effect into account when sampling 2-stroke exhaust aerosol. All the processes mentioned here are promoted by longer sampling lines (longer residence time). *Deposits on the exhaust pipes and sampling lines from previous measurements further add to the problems.*

Because of these processes taking place, the sampling procedure (conditioning of the sample gas probe, dilution and sampling position) may influence the measured aerosol characteristics (PM, PSD, PAS, DC).

The experimental programme of TNO was performed on a typical current 125 cm^3 motorcycle with a conventional 2-stroke engine without catalyst. As far as the regulated exhaust components are concerned, it was completely in compliance with the EC 97/24 directives for exhaust sampling for motorcycles. For particle sampling, a probe was inserted in the full-flow dilution tunnel, leading to a Scanning Mobility Particle Sizer (SMPS). The sample flow was diluted twice before entering the SMPS. The monodisperse sample flow from the SMPS was then led into a Condensation Particle Counter (CPC). *Special effort was taken to keep the transfer lines from the sample point in the tunnel to the SMPS as short as possible.* In contrast to the usual car set-up, the gas from the exhaust tailpipe was led into the transfer hose using an open system. This is not actually specified in the EC 97/24 regulations, *but the original discussions in the ECE-GRPE assumed this to be necessary for PTWs and hence the pictures suggest to use an open system.* An uncontrolled dilution takes place here, which *may promote spontaneous condensation. On the other hand, this method has the advantage that coagulation, which is likely to occur when a certain threshold in concentration is exceeded, may be prevented.* It was assumed that the concentration of particles in the raw exhaust gas was beyond this threshold. *More research is necessary, however, to draw a conclusion on this point.* Furthermore, the exhaust transfer lines were not heated. *In a dedicated particle sampling set-up one would consider heated lines, since non-heated lines make the measurement set-up sensitive to thermophoresis.* However in the present experiment a sampling set-up in accordance with the Directive was specifically chosen. The vehicle is characteristic for a category of relatively highly tuned small sporty vehicles.

Obviously the repeatability of the gravimetric measurements is very poor. Further consideration of the measurement process suggested that (minor) differences in the run-up to the actual measurement may have caused at least some of this. This may be a result of the fact that the measurement process is very sensitive to spontaneous condensation and thermo-

phoresis. It is recognised, however, that more repeat tests are necessary to draw a firm conclusion on this point.

Total particle concentration is the most variable of all metrics reported. CoVs reach values which would normally be unacceptable (>50%) to support repeatable measurements. Relevant experiments suggest that the preceding operational history of the engine might be a significant factor. In general, the nucleation mode (< 50 nm) is more variable than the accumulation mode, and in some cases may even change the shape of the distribution. It is clear from the description of this phenomenon that, despite stabilised conditions; the particle emission rate follows a dynamic pattern. The actual size distribution shape is a function of the equilibrium conditions between the solid and vapour state of a number of volatile substances present in the exhaust gases. Especially in the case of 2-stroke engines, a large concentration of lubrication oil hydrocarbons and sulphates is found in the exhaust gases. Hence, depending on the operational state of the engine and its operational history, such substances may ad- or de-sorb from the walls and respectively suppress or promote the formation of nanoparticles. If this is true, then operation at 50 km/h can heat up the exhaust system, which subsequently desorbs condensable species to form nuclei and gradually formulate the nucleation mode. On the other hand, the extent of this process depends on the preceding history of the measurement, therefore *a larger nucleation mode is formed (before the equilibrium is reached again) when idle operation instead of a driving cycle has preceded the measurement*. Idling, because of the low exhaust line temperature, promotes volatile adsorption on the walls, which will be later desorb to form nuclei. Such a phenomenological mechanism seems to be responsible for the large dispersion of the values and the high CoVs values in all cases of steady speed driving. *It could be suggested that better repeatability would be reached if longer times (such as 15 minutes) are used for stabilisation of the vehicle, exhaust line and sampling system*. However, additional research is required for this to be proven. Although this would be possible, steady speed/load driving for more than a couple of minutes is impossible in actual vehicle usage, hence longer stabilisation times would lead to results not encountered in actual use.

In a comparative study by TNO and LAT, the TNO experience was that even after similar (although shorter duration) preconditioning, slight variations in idling time may undo the stabilising effect *when the actual measuring and sampling does not follow immediately*. Such effects were not encountered by LAT during *transient testing where no equilibrium condition is reached*, although they were by TNO. A not uncommon reason for rejecting a measurement is that no stabilised conditions were actually reached before the test, even if the gaseous pollutant measurements were all stabilised. In contrast, no measurement was rejected under the transient testing.

Table 5-1 clearly shows that there are many potential influencing factors that may determine the outcome of a PM measurement on small 2-stroke engines. Additionally it should be borne in mind that, as shown above, in the case of a determination of the particle size profile (at least on the basis of mobility diameter) the nucleation mode turns out to be the dominant mode for many operational conditions. LAT has conducted a short parametric campaign to examine what the effect of the dilution factor and the filter face velocity would be on the emissions (two different CVS flow rates and two filter face velocities. The first observation concerns the variability of the measurement, which is less in the case of the high filter face

velocity. With regard to the absolute level of emissions, the higher face velocity results in a marginally higher PM recording (especially in the case of the low CVS flow rate). However, such observations may also derive from instability of the vehicle emission rate as such. In the case of the solid fraction all results seem to overlap regardless of filter velocity and CVS flow rate. *The difference observed in the low CVS flow rate originates mainly from the volatile fraction.* This can cause a problem as in the case of 2-stroke engines it is exactly the volatile fraction that is the most important as far as mass emissions are concerned.

Table 5-1: Factors influencing the sampling of PM emissions from two-wheelers.

Parameter	Effect (especially with regard to 2-stroke PM)
Dilution factor / CVS total flowrate	Regulates PM concentration and hence sensitivity. Also regulates temperature and residence time in the dilution tunnel which may enhance/suppress condensation of volatiles.
Design of the dilution system	The actual design of the dilution system (open or closed, single stage or two-staged) does influence the temperature profile over the sampling system and hence the condensation profile.
Filter face velocity	Filter efficiency depends on face velocity. High velocities may tend to remove volatile material from the filter as this becomes saturated. Low velocities may result in particle loss in the probe before the filter.
Filter material	Filter material may show SOF-selective behaviour. E.g. quartz or glass fibre filters generally show higher volatile fraction concentrations than teflon (or teflon coated) ones. This may be critical for the high volatiles emitting 2-strokes.
Filter temperature	Sampling temperature at the filter position affects the SOF speciation due to volatile species evaporation/condensation. More precise control of this temperature may be necessary.
Backup filter	The back-up filter is mostly used to collect volatile species not captured by the primary one. However, experience shows that its use also tends to increase the measurement variability.
Filter loading	If the filter becomes saturated, a large fraction of volatiles may start to evaporate. On the other hand, a small loading may decrease the weighing accuracy.
Sample pre-treatment	It may be necessary to omit large particles (re-entrained from the walls) from the sample by cut-off techniques to increase repeatability.
CVS and sample lines cleanliness	A high emission rate of lubrication oil tends to build up deposits quickly on the sample line walls. Apart from producing a high background HC concentration, this may also damage parts of the installation and affect the operation of devices and sensors.

Establishing typical exhaust emissions of motorcycles involves much more variation than in the case of other vehicles, as the total field is much wider. This is because two-wheelers vary in engine characteristics from less than 100 cm³ and less than 5 kW to 2000 cm³ and close to 150 kW, in ready to ride mass from less than 100 kg to nearly 400 kg, and in performance from less than 100 km/h. Technologies include conventional 2-stroke, DI 2-stroke and 4-stroke (Gense and Elst; 2003). This wide range in vehicle characteristics inevitably reflects in the characteristic patterns of use, as characteristic riding styles and predominant use of certain road types will be heavily influenced by variations in performance level and in the class of the motorcycle. This fact has to be taken into account when establishing characteristic emission levels for powered two-wheelers (PTWs).

Because of this variability, work was undertaken in 1999 to develop a “real-world” driving cycle for use in PTWs certification. A relevant study by Rijkeboer (2000) showed that the European vehicle fleet can be roughly characterised as a north-European fleet, predominantly consisting of larger bikes mainly used for recreational purposes, and a south-European fleet, predominantly consisting of smaller vehicles mainly used for daily transport. A country like Germany would be a typical representative of the first category; countries like Greece and Spain would be typical for the second. Countries like Italy and France possess a combination of both categories. Further investigation revealed the following typical circumstances of use, presented in Table 5-2.

Table 5-2: Typical places and circumstances of use.

PTW Type	Type of road (and cold/warm started)			
	Urban - cold	Urban - warm	Country roads	Motorway
Scooters & small utility	important	important		
Medium class & Touring	important less important	less important important	very important very important	less important
Big and sporty	less important	less important	very important	less important

It should be noted that the ECE 40 type approval test cycle (dating from the very early days of car testing) only addressed the situation warm start/urban traffic. For the current Euro 2 a cold start has been included, but the urban character was maintained. Therefore, it was suggested to set up a test cycle in 3 parts (that is urban, country road and motorway) and to weight them according to the Table considerations (WMTC project). Based on the world-wide nature of the WMTC a world-wide weighting is needed, but a significant difference between Europe and the world only shows for class 3. The weighting factors have not yet been finally established, but many turn out something like 50/50 urban cold/hot for bikes of class 1 (e.g. <125 cc), 30/70 urban/country road for bikes of class 2 (e.g. 125-500 cm³) and 15/65/20 urban/country road/motorway for bikes of class 3 (e.g. > 500 cm³) in European traffic.

The main shortcoming of the EC 40 test is that it considers a driving pattern similar to the car procedure. However, two-wheelers, even small ones, show the dynamics of sportily driven cars. Bikes with very high power to mass ratios (>200 kW/tonne) surprisingly showed

increasing dynamics with increasing speeds, within the range of average speeds investigated. This has an influence on the emission behaviour of PTWs.

- The large range of emission values over the different cycles made it difficult to accurately measure the different road types in one package of settings. *Settings of sampling and analysing equipment should therefore as far as possible be adjusted to the emission level to be expected.*
- The brake loads to be set for PTWs are relatively small in comparison to those of cars, making them difficult to set accurately. *Internal frictions on the bench should be as low as possible or electrically compensated.* This problem increases when a chassis dynamometer is used that was actually designed to test cars. *“Outdated” chassis dynamometers are unsuited for acceptable measurements of PTW emissions, especially in the case of small bikes.*
- A highly dynamic driving cycle in combination with a high power to mass ratio of the vehicle tested can easily result in poor repeatability of the test. *This means that special attention should be paid to the response time of the brake load simulation device: this should be much faster than is acceptable for the ECE 40 (or the car type) test cycle. Especially in the case of high performance super sport bikes this is an important requirement for repeatable, and hence representative, results.*
- Many PTWs are essentially drive wind cooled, even if they do possess a form of liquid cooling of the cylinders and cylinder head. This puts more emphasis on the drive wind simulation than in the case of cars. As a rule the temperatures both of the coolant and as measured at the spark plug are significantly higher in the lab than under comparable conditions on the road. *It is to be expected that especially the emissions of catalyst equipped vehicles during the warm-up may be affected by this aspect.*

6. Recommendations

For future particle mass and size distribution measurements on PTWs, the following recommendations can be made:

1. The present gravimetric particulate measurement procedure as used for diesel PM measurement is very likely to be inadequate for reproducible PM measurements on small 2-stroke engines in the future. More research is needed to establish the special characteristics of PM-emissions from small 2-stroke engines, especially in the case of future technologies, and the consequences of these characteristics for the requirements to a measurement procedure that has sufficient repeatability and reproducibility to form the basis of a quantitative assessment of the PM-emission of a small 2-stroke engined vehicle. For example, already now the major part of the 2-stroke PM-emission consists of volatile components. With a further reduction in fuel/air ratios this percentage will increase, so the sampling will become more sensitive to sampling temperatures and condensation/evaporation effects on the sampling filter, that determine the extent of the volatile fraction. Likewise the sampling is likely to become more sensitive to phenomena like local condensation and thermophoresis, with the subsequent problems like temporary loss of PM on the one hand and scavenging of PM-material previously stored in the exhaust and/or the sampling equipment on the other (i.e. a bigger influence of the preceding ‘history’ of the exhaust and sampling system). For

these reasons, particular attention should be paid to the influence of the preceding operational history of the engine, the effects of variations in the way and degree of dilution, and the possible necessity of working with heated sample lines. *An extensive parametrical study of the necessary characteristics of the total CVS procedure is strongly recommended.* In the absence of such information, before such a thorough investigation has been made on a sufficiently extensive scale no serious recommendation can be made as to the relevance of the existing measurement procedure as used for diesel PM for use in the case of 2-stroke PM.

2. The measurement of particle size distributions is prone to sampling artefacts, and it is expected that the particle size distribution may significantly differ according to sampling conditions, up to a degree where even the measurement of mass emissions becomes insufficiently accurate. **Such variability is mainly caused by the behaviour of the nucleation mode.** As yet it is unclear if this nucleation mode should be regarded as largely an artefact of the sampling procedure, or rather as a typical aspect of real-world behaviour that may get lost by insufficient sampling conditions. *Thus, more understanding is needed about the reality of the observed nucleation mode and its behaviour under real-world circumstances.* Only in that way meaningful conclusions can be drawn concerning the way the sampling and measurement conditions should be optimised.

3. It should be pointed out that, together with considerations regarding the environmental effects, **a better understanding of the health effects of 2-stroke PM emissions ought to be the basis for any legislative limitation of such emissions.** Such understanding is still largely lacking even for the health effects of diesel PM, although some studies may start to appear soon, and this is all the more true for the physically considerably different PM from 2-strokes. Given the difficult task of proposing a reproducible method to establish a sensible representation of the adverse effects of 2-stroke PM, more understanding of the actual behaviour of the various fractions of this PM and their respective effects are urgently needed so as to be able to target the really harmful fractions and to avoid the risk of 'drowning' these in a method that targets a possibly less harmful bulk with the unwanted secondary effect that the really harmful fraction gets lost in its variation. If, for example, it could be decided that from an environmental and health point of view the nucleation mode has no unacceptable effects, one could afford to work out a procedure that avoids the inclusion of this mode and that way arrive at a much more reproducible measurement. Hence, *it can be recommended that, given the significant differences in reproducibility in the measurement of the different modes, more understanding of the real health effects of 2-stroke PM emissions is needed before it can be decided which characteristics of 2-stroke PM, or which fractions of the total PM-emissions, are really of importance and should therefore be targeted by a sampling and measurement procedure.*

4. As long as there is little or no understanding of the health effects of 2-stroke PM **there is obviously no basis to establish a sensible threshold for this PM. Still less is there any basis to 'translate' this threshold into an acceptable mass emission** (assuming that the actual health criterion will be expressed in another way and for another characteristic). When on the basis of the research recommended above such a threshold has been established, a working relation between that and a 'corresponding' mass emission is still needed so as to establish a meaningful mass emission limit for type approval. Thus, *it is recommended that a*

working relationship is established between the likely harmful characteristics of these characteristics and the overall mass emissions, as a workable basis for limit setting.

5. As it was previously mentioned, the PM emission from 2-strokes is in fact already largely included in the measurement of the HC emission. This means that this emission is already under control to a considerable extent. When on the basis of the considerations and recommendations above it is decided to postpone a possible dedicated limitation of this emission, it is all the more important to measure it in its HC guise as completely as possible. *It could be suggested that the way heavy HC from diesel engines is measured may be used for guidance.* It should be ascertained that the fraction concerned will indeed be measured in full as HC. This could require that the HC emissions from 2-stroke engines are measured in an analogous way as the HC-emissions from diesel engines, namely by means of a heated sample line and a heated FID instrument, if necessary further to be worked out. This possibility should be investigated with priority. The validity of such an approach could be checked in an easy way, and in a relatively short time frame. *Adaptation of the HC measurement procedure in such a way is likely to remove much of the immediate urgency of a dedicated PM measurement.*

6. Again as pointed out previously, **both the emissions of total-HC and those of PM are likely to decrease with future legislative limits more or less in parallel** and to a significant degree. Again this could mean that the urgency of a dedicated PM limit is much reduced by the legislation already in place, and/or contemplated for the near future. *It is therefore recommended that more information should be collected about characteristic PM emissions from Euro 2 and later vehicles and their characteristic technologies, both as to their level and as to their composition and characteristics.* Such information is expected to confirm the acceptability of a postponement of a short term dedicated limitation of 2-stroke PM-emissions until sufficient knowledge has been collected to establish both a sampling and measurement procedure and limit values for such a limitation.

7. The cost for society as a whole would benefit from an approach where, if a dedicated limitation is introduced, the type approval procedure is based on a simplified procedure, while a parallel monitoring process acts as a safeguard that the hoped for empirical relation (as indicated under recommendation 4) between the simplified result (presumably mass emission) and the actually important characteristics (presumably expressed in different terms and units) is maintained in practice, and that the real objectives are actually obtained. It is recommended that, if and when a dedicated legislative limitation of 2-stroke PM is established, *this will take the form of a simplified procedure, and that the effectiveness of such an approach will be monitored by a smaller scale (i.e. concerning less vehicles) but more thorough (targeted at more meaningful but more difficult to measure characteristics) monitoring programme.*

As explained previously, the typical patterns of use of motorcycles differ significantly from those of passenger cars. Motorcycles show a much greater variation in characteristics than cars. These variations contribute even further to differences in typical patterns of use. The typical motives of use vary per class of vehicle, and hence per typical, country specific fleet composition. They in turn are determining the typical places (road types) and circumstances of use, which vary significantly, both between each other and in comparison to cars. *The testing of motorcycles on a chassis dynamometer is much more dependant on the*

characteristics of the specific test equipment and the test circumstances of the laboratory than the testing of cars. Special care has to be taken to avoid the risk of misleading test results. As a result of all of these aspects an inventory of different vehicles and engine technologies will result in significantly different conclusions depending on whether they are based on the emissions as produced over the type approval test cycle or on a programme that did take the above considerations into account.

With regard to the concentration of PM emissions – at least with regard to currently operational two-wheelers - the concentration is such that the actual emission rates of current two-wheelers can reach or even exceed the respective concentration of today's diesel passenger cars, even in the case of catalyst equipped vehicles. If this conclusion would be established over a larger sample of vehicles, this would mean that current CVS installations for passenger cars may provide the necessary sensitivity for PM emission recordings also from two-wheelers. What current technology shows, however, is that near-future two-wheelers may be equipped with catalyst systems and direct injection engines, whereas lubricants are being produced which fully combust in the chamber. If this is the case in the near future, then the **current CVS procedure may not be sufficiently sensitive**. What already needs to be tackled at the moment for today's 2-stroke vehicles, is all those measures that need to be taken to obtain a representative emission rate of this volatile aerosol. *In case that a CVS procedure is adopted for two-wheelers PM sampling, a specific project aiming at a parametrical study of the effect of these parameters would be strongly recommended.*

Some relevant scientific recommendations on the technical issues of PM assessment from two-wheelers are the following:

- *More effort should be made to draw conclusions on the repeatability of gravimetric particle mass measurements. The results here indicate that they may also be very sensitive to thermophoresis and spontaneous condensation. These effects seem to be very sensitive to the preceding operating history of the vehicle.*
- *To prevent particle loss due to thermophoresis, the use of heated exhaust transfer lines should be investigated. In this way a more quantitative analysis of the effect of dynamic driving behaviour might be made.*
- *The use of an open exhaust gas collection system gives an uncontrolled dilution of the raw exhaust gas, which might be favourable to the process of spontaneous condensation. It is recommended to investigate the dilution system in more detail.*

For additional general recommendations on performing good practice monitoring, that have to be followed in all air quality assessment procedures, the reader is referred to the Chapter 3.1.6 of the Milestone Report.

7. References

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