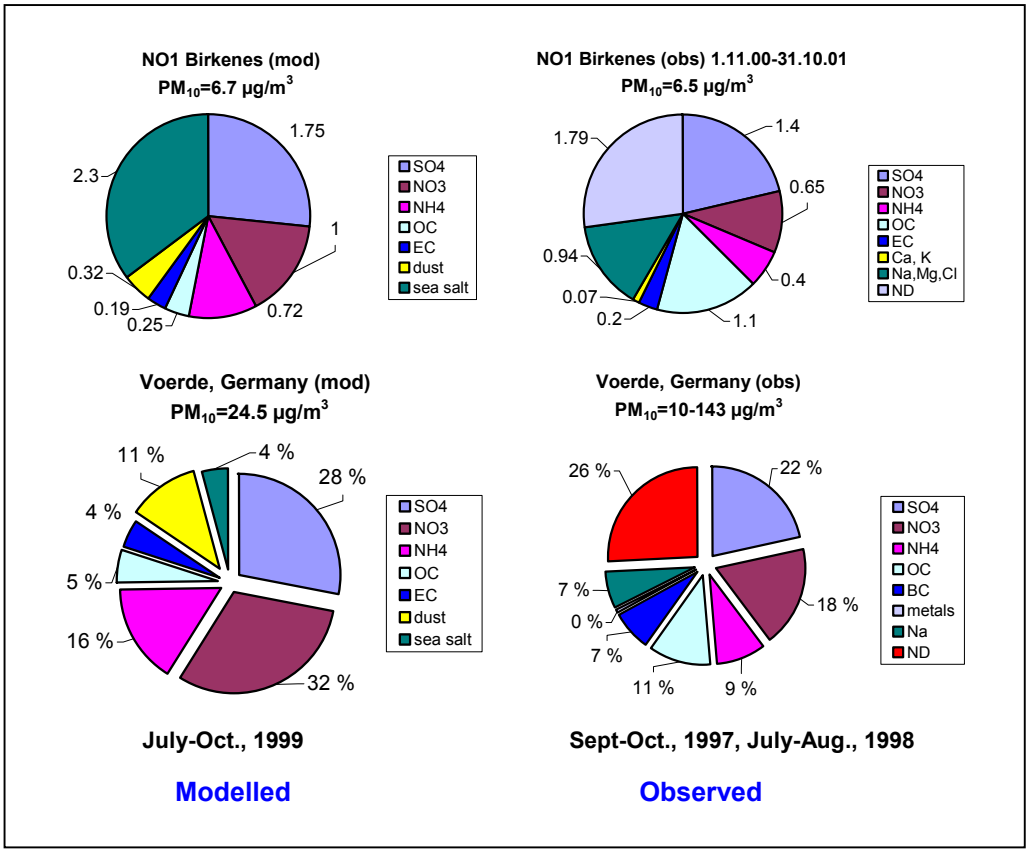
EMEP Report 5/2002

EMEP Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe

Transboundary Particulate Matter in Europe: Status Report 2002



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Transboundary Particulate Matter in Europe: Status Report 2002

Joint CCC & MSC-W &CIAM Report 2002

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Contents

Page

Executive Summary

Long-range transboundary transport is responsible for a significant fraction of the particulate pollution in European cities as well as in rural areas (EMEP-WMO, 1999). It is apparent that particulate matter is not a single pollutant and its mass includes a mixture of many pollutants in a complex multiphase system. The traditional EMEP approach is to find how the air quality and the deposition of pollutants are affected by emissions in the different countries in Europe and involves emission inventories, emission-based model simulation of atmospheric processes, transport and deposition, and verification and assessment by observations and measurements. This report summarises the recent developments in the particulate matter work of EMEP.

The establishment of an adequate monitoring programme is a crucial step in the development of abatement strategies for particulate matter. After the inclusion of particulate matter in the workplan of EMEP, a monitoring strategy has been developed in close cooperation with the scientific community and national experts through the Task Force on Measurements and Modelling. Chapter 1 of this report presents the current status with respect to the implementation of the monitoring programme as well as it presents a broad overview of measurement results for year 2000. The number of sites monitoring PM mass in year 2000 was still low, but a number of countries have initiated measurements during 2001 and 2002. Measurements of parameters other than PM mass are still sparse and countries are strongly encouraged to expand their activities to meet the requirements given by the monitoring strategy (www.nilu.no/projects/ccc/pm_strategy.html).

A summary of PM emissions data is given in chapter 2. Twenty of the forty-eight Parties to the Convention submitted PM emission data for the year 2000 to the UNECE. It is encouraging that as many as eleven Parties (23%) submitted national total and level 1 sector PM_{10} emissions. The reporting of fine fraction ($PM_{2.5}$) was somewhat lower (17%). Five Parties submitted PM data at NFR (Nomenclature For Reporting) level 2. In addition, France submitted PM data at SNAP 2 level. Six Parties provided gridded total emissions and three Parties reported gridded sector PM emissions.

Despite the encouraging number of initial reporting, the number of submissions of officially reported data is far from complete and steps should be made to allow for the completion of the official UNECE/EMEP PM emissions inventory for the year 2000. There are still important discussions on the type of emission sources to be considered in a European based PM emission inventory and further analysis is necessary in order to establish appropriate emission factors for all identified sectors.

In chapter 2 we present an evaluation of the differences in available emission information for four different countries (Austria, France, the Netherlands and United Kingdom) for the year 1995. The evaluation is intended as an illustration of the type of discussion that would eventually lead to the creation of a harmonized European-wide PM emission inventory. The comparison of differences is made between the results of international inventory (CEPMEIP), national submission, and modelling work of CIAM.

Advances in aerosol modelling within EMEP are presented in Chapter 3. The EMEP aerosol model has recently been extended to include aerosol dynamics, implying that, preliminary size resolved results on aerosol mass, number concentrations and chemical composition are available. The initial testing of the model results has focused on the analysis of the effect of aerosol dynamic processes on primary PM concentrations. A significant conclusion so far is that accounting for size dependent emissions and depositions of aerosols in the model has a larger impact on the calculated primary PM concentrations than introducing coagulation processes. These results stress again the importance of appropriate information on the size distribution of primary PM emissions for assessing PM concentrations. They also highlight the need for accurate descriptions of size dependent dry and wet removal processes, and further studies on these topics are envisaged for the future. Some preliminary comparisons with observations are presented in chapter 3, while further validation on validation is under work.

The EMEP monitoring programme still provides insufficient data for model validation. This is particularly the case for information on the chemical composition of the aerosol. Although secondary inorganic aerosol components such as nitrate and ammonium have been part of the programme for many years, only few countries report data obtained from measurements that allow for the separation of gas and aerosol phase for these compounds. Apart from their essential role in developing an improved description with respect to acidification and eutrophication, these compounds are also needed for the ongoing development of PM modelling. For other parameters even less data are available. In particular for carbonaceous species information on the ambient concentration and chemical composition is generally lacking. Thus a comprehensive campaign aiming at determining concentrations of elemental and organic carbon concentrations at a number of sites across Europe has thus recently been initiated. Detailed information about this campaign can be found in EMEP CCC-Report 4/2002. A comprehensive description of the atmospheric particles also requires the evaluation of particle number, surface and volume distributions in addition to their mass and their chemical composition. Size distribution measurements combined with chemical speciation are also necessary for identifying also the sources of atmospheric particulate matter. Further, the model validation requires sufficient measurement data both in terms of site density, data quality and chemical/physical parameters determined.

Comparison of the EMEP model with available measurement data indicates a general underestimation of ambient PM_{10} concentration levels, and for some regions the discrepancies are large (e.g. in the Mediterranean area). It should be noted however that natural and re-suspended anthropogenic mineral dust and biogenic aerosols are currently not incorporated in the model.

A methodology to estimate reduced life expectancy due to particulate pollution in Europe is presented in Chapter 4. The methodology integrates population data, findings from epidemiological studies, information about the formation and dispersion of fine particles in the atmosphere and estimates of present and future levels of emissions of fine particles and their precursors. Chapter 4 also presents a preliminary implementation of the methodology for Europe based on presently available models and data. Awaiting further refinements in these scientific

disciplines, the quantitative implementation should be considered as preliminary and needs to be revised as soon as more substantiated scientific information becomes available.

Keeping these imperfections in mind, the preliminary results of this assessment suggest that the life expectancy in Europe is significantly shortened by particulate pollution, namely (based on the present assumptions) between three months in Scandinavia and more than two years in central Europe. The 95 percent confidence interval of these estimates due to uncertainties in the evidentiary epidemiological studies ranges between 1.5 and eight months in Norway and eight months and 3.5 years in Central Europe. However, this situation is expected to change profoundly in the future due to the recent agreements on agreed emission control. The Gothenburg Protocol of the Convention on Long-range Transboundary Air Pollution and the Emission Ceilings Directive of the European Union should bring significant reductions of the precursor emissions of secondary aerosols and also primary emissions of $PM_{2.5}$ are expected to decline by 57 percent as a consequence of stringent controls for stationary and mobile sources. These emission controls will thus reduce the average loss of life expectancy in Europe to somewhat more than seven months in Europe, spreading from one month in Norway to more than 13 months in Bulgaria, Romania and the Ukraine. Full application of all available technical control measures could further reduce these losses on average by another 33 percent.

1. Particulate matter measurements in Europe

by Michael Kahnert, Steinar Larssen, Kjetil Tørseth and Wenche Aas

This chapter provides an overview over the current state of EMEP's measuring activities of particulate matter (PM), over recent developments, and over future directions. A summary of PM_{10} mass concentration results obtained in the year 2000 is presented, while the detailed presentation of PM monitoring data can be found in the EMEP/CCC-Report 4/2002.

In 2001 at its 25th session the EMEP steering body adopted the PM monitoring strategy (a detailed description of the strategy can be found at www.nilu.no/projects/ccc/pm_strategy.html). At the third meeting of EMEP's task force on measurements and modelling (TFMM) in March 2002 the revised draft of the EMEP Manual for Sampling and Chemical Analysis (EMEP/CCC, 1996) for PM_{10} mass measurements and chemical speciation was adopted. The mass measurement part is based on standard EN 12341 of the European Committee for Standardization (CEN) (CEN, 1998) and recommends employing the gravimetric method, which has proven to be the most accurate method. Gravimetric methods also have the advantage of allowing chemical analysis of the collected PM_{10} sample after weighing. The application of the methods and quality assurance procedures recommended by the manual is important in order to harmonise the ongoing PM_{10} measurements (EMEP, 2001; Lazaridis et al., 2002) throughout the EMEP network.

Nevertheless, there is different measurement equipment presently in use, which poses a challenge with respect to quality assurance. It should be noted that parties can decide to use sampling equipment other than the high- or low-volume reference samplers listed in the EMEP manual, provided that their sampling equipment has been demonstrated to give results comparable to those obtained with the reference methods.

As illustrated in Figure 1.1 only three countries have reported PM_{10} data in the year 2000 and from a total of 26 stations (five Swiss, eight German, and 13 Spanish stations). However, it is expected that the number of stations will double in the near future, and that the spatial coverage of Europe will improve substantially. As in 2002, Austria, the Czech Republic, Denmark, Ireland, the Joint Research Centre (JRC) in Ispra (Italy), The Netherlands, Norway, Portugal, Sweden, Slovakia, and Great Britain were measuring PM_{10} in addition to Switzerland, Germany, and Spain. In the near future, PM_{10} monitoring is also expected to start in Finland, Greece, Slovenia, and possibly in Kazakhstan. Also shown in Figure 1.1 are current activities of $PM_{2.5}$ measurements. Table 1.1 provides an overview over stations and measurement equipment in different European countries. Not all the collected data have been reported to EMEP-CCC.

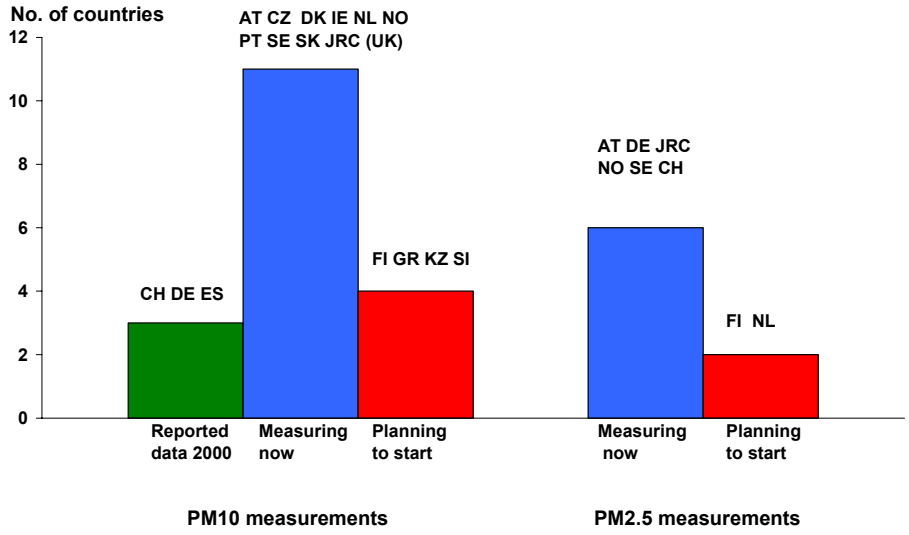

Figure 1.1: Number of countries measuring PM_{10} and $PM_{2.5}$ (by August 2002).

Table 1.1: List of countries measuring PM_{10}, and of equipment used.

Country	Stations measuring PM_{10}	Start measuring	Time resolution	Instrument
Austria	AT02	July 1999	daily	Digitel High Vol. DHA80
	AT04 and AT05	Jan. 2001		
Belgium				
Croatia				
Czech Republic	CZ01 and CZ03		30 min	radiometry (Verewa)
Denmark	DK05	Oct. 2001	24h	SM200 (ADAM)
Estonia				
Finland	FI04 or FI17	spring 2002	daily	Kleinfiltergerät
France				
Germany	DE01, 02, 03, 04, 05, 07, 08, 09	1. Jan. 1999	daily	Digitel High Vol. DHA80
Greece	GR1, Aliartos	autumn 2003		not decided
Hungary				
Iceland				
Ireland	IE31 Mace Head	Aug. 2001	daily	U Miami Hi Volume
Italy (IT1)				
JRC -Ispra	IT04	March 2000	daily	Kleinfiltergerät
Kazakhstan	Borovoe	Jan. 2003		SKC or Casella
Latvia				
Lithuania				
Macedonia	no			
Moldova	no			
Netherlands	NL09 and NL10	1992/93	hourly	beta-dust FH 62 I-N
Norway	NO01	Jan. 2000	daily	Kleinfiltergerät
Poland				
Portugal	PT03 and PT01	1979	daily	HiVol, Sierra Andersen
Russia				
Slovakia	SK04	Oct. 2001	weekly	Partisol Plus R&P
Slovenia	SI03	2003	daily	Kleinfiltergerät
Spain	ES01, 03, 04, 05, 06, 07, 08, 09, 10, 11, 12	1997	daily	MCV
Sweden	SE12	March 1990	24 h (particles)	R&P ACPM and
	SE11 and SE35	Sep. 99 and Jan. 02	1h, 3h and 24 h	R&P TEOM
Switzerland	CH01, 02, 03, 04, 05	Jan. 1997	daily (48h CH01)	Digitel high vol. DHA80
Turkey				
UK	UK43 and UK06	1997	hourly	
Yugoslavia				

Annual averages of PM_{10} concentrations measured at the different EMEP stations during 2000 are presented in Figure 1.2. Only those stations that have a data coverage of more than 50% are shown.

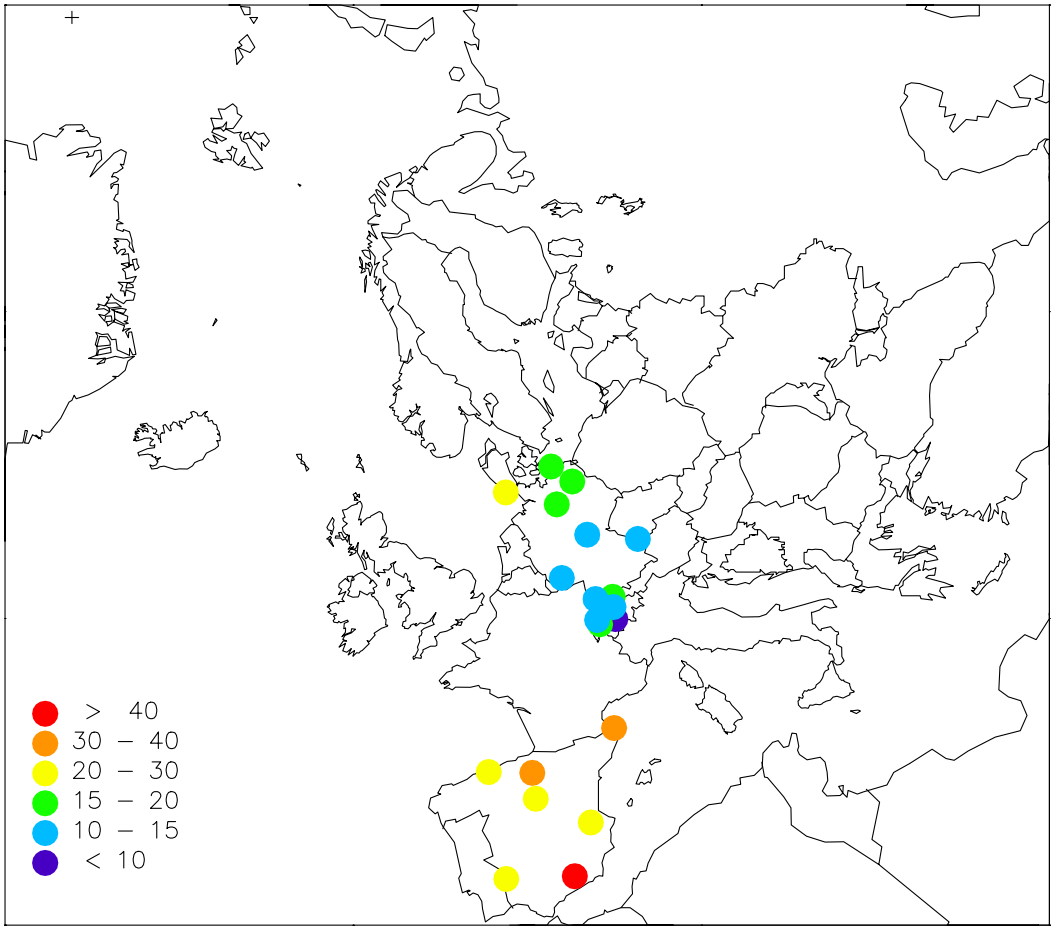

Figure 1.2: Annual averages of PM_{10} concentrations at EMEP sites in 2000.

Figure 1.3 depict results of PM_{10} measurements collected in the AIRBASE data system[1] operated by the European Topic Centre on Air and Climate Change (ETC/ACC) under contract of the European Environmental Agency. Due to the large number of different methods employed in obtaining the AIRBASE data, the results are mainly meant to indicate the spatial trends and show the geographical coverage. Figure 1.3 shows annual averages of PM_{10} concentrations at different rural stations included in AIRBASE.

[1] AIRBASE data have been extracted by F. de Leeuw (RIVM, The Netherlands). Figure 1.3 was produced by P. Kurfurst and J. Fiala (Czech Hydrometeorological Office).

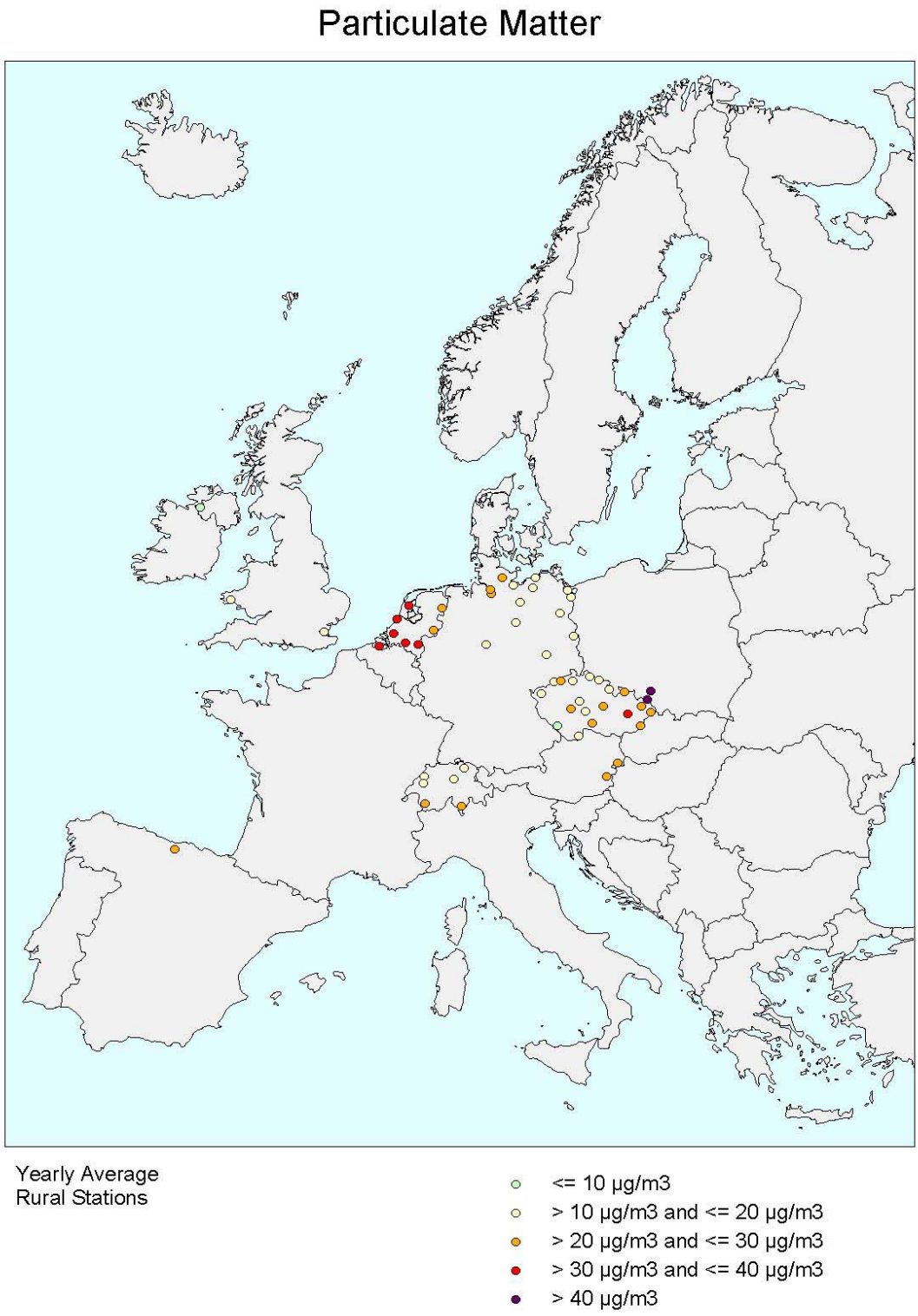

Figure 1.3: Annual averages of PM_{10} concentrations at AIRBASE rural sites in 2000.

A summary of annual averages of PM_{10} concentrations measured in 2000 in different countries is presented in Figure 1.4. The data shown have been obtained by averaging for each country over all stations that reported PM_{10} data in that country, and obviously the site locations etc. make direct comparisons of concentration levels difficult. All reporting stations have been included into the average regardless of their temporal data coverage. EMEP data are represented by blue columns. The other results represent AIRBASE PM_{10} data from rural (green), urban (red), and traffic (black) sites. Also indicated in the figure are the indicative value according to the EU directive of an annual PM_{10} concentration average of 40 µg/m^3 for the target year 2005 (dotted line), and the corresponding indicative value of 20 µg/m^3 for the target year 2010 (dashed line).

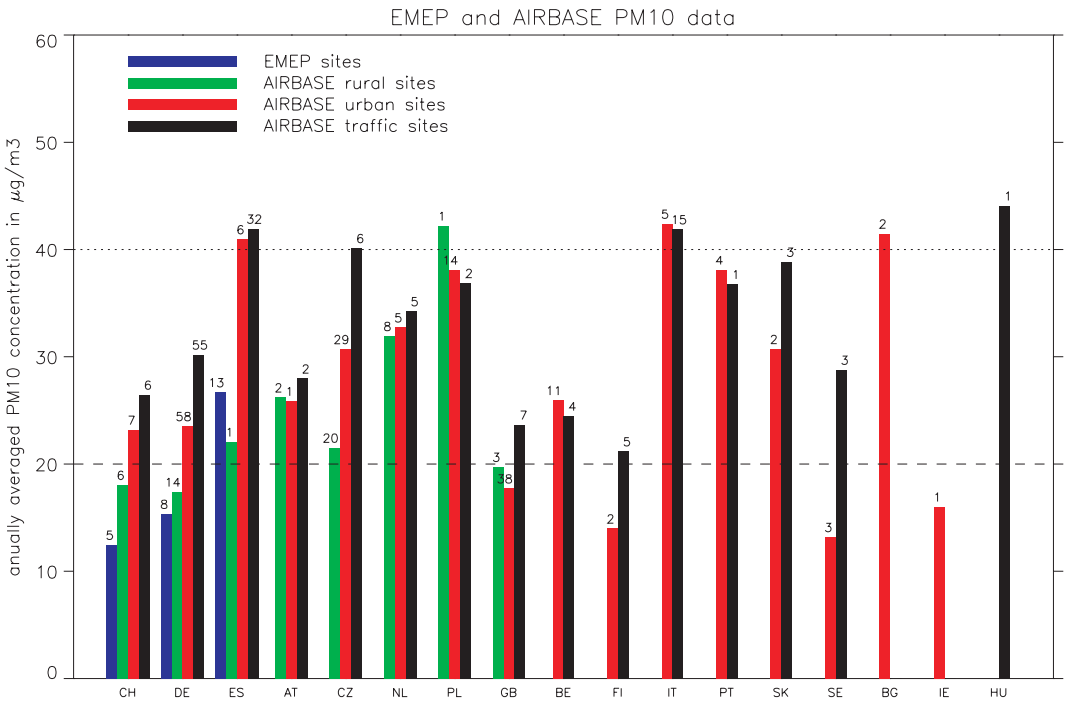

Figure 1.4: Annual and country-averages of PM_{10} concentrations at EMEP background sites (blue), and at rural (green), urban (red), and traffic sites (black) in 2000. Numbers over each column indicate the number of stations over which the country-average has been taken. The data for rural, urban, and traffic sites have been extracted from AIRBASE.

Figure 1.4 shows for some countries rather small differences in PM_{10} concentrations between urban and rural sites. Even though local sources in urban centres or agricultural sources in rural areas can be important contributors to the observed PM_{10} concentrations, a significant contribution usually originates from aerosols that have been transported over regional scales. This results in small differences between the time-averaged PM_{10} concentrations observed at urban sites and at rural background sites. The annually averaged PM_{10} concentrations in Figure 1.4 are for most countries still far above the limit value set for 2010, in many cases even twice as high as the limit value.

There is accumulating evidence indicating that monitoring PM_{10} mass is not sufficient for answering the relevant questions concerning the formation and behaviour of particulate matter in the atmosphere. More information about the physical and chemical properties of aerosols is needed, which places additional requirement on the monitoring programme. The PM strategy thus requires monitoring of other parameters to complement the measurements of PM_{10} mass. More specifically, a three-level approach has been adopted by the EMEP steering body. Level 1 activities shall consist of PM_{10} measurements that at least one station in each country will perform on a daily basis. In addition, a number of sites will send once a week samples to a common laboratory for the purpose of determining the fraction of elemental carbon (EC) and organic carbon (OC). Also PM mass of fine particles ($PM_{2.5}$ or $PM_{1.0}$) as well as the concentrations of sulphate, nitrogen species and base cations are to be measured on level one sites. Determination of gas/particle distribution for nitrogen species using "artefact-free" methods, mineral dust analysis, and chemical speciation as a function of particle size shall be conducted at a limited number of sites (level 2 sites). At level 3 sites it is envisaged to undertake research campaigns or other advanced research activities usually not available at EMEP sites, such as OC-characterisation, measurement of number and area size distribution, light scattering measurements, determination of aerosol optical depth, etc. Figure 1.5 provides an overview over the number of countries occasionally or routinely conducting more detailed aerosol studies at certain sampling sites (in 2002) and over the kinds of measurements.

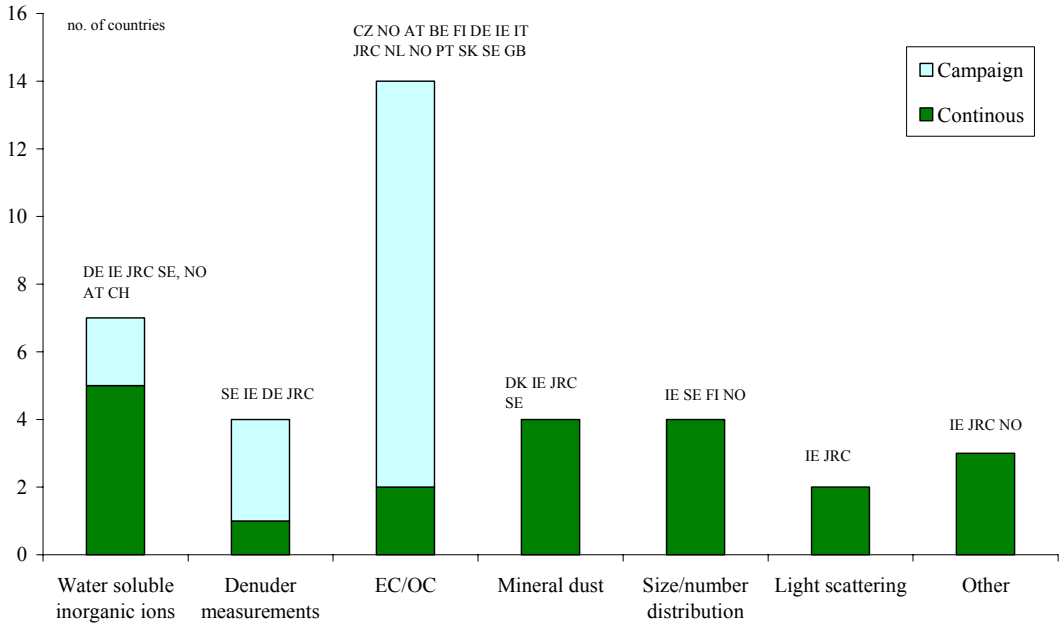

Figure 1.5: Overview over various aerosol measurement activities in different countries in 2002.

Elemental and organic carbon can constitute a significant portion of the total aerosol mass. However, this fraction is currently the most severe limitation of our ability to obtain mass closure for aerosols. Therefore a more accurate

determination of the EC/OC fraction is presently of high priority. Accounting for both organic and inorganic constituents of aerosol particles in models is also essential for accurately describing the physical and chemical processes in clouds and aerosols (Jakobsen et al., 2000). However, our knowledge about sources and composition of the EC/OC fraction of aerosols is still highly incomplete. Fossil fuel combustion (in the Northern Hemisphere) and biomass burning seem to be the major anthropogenic sources of OC. Oxidation of volatile organic compounds (VOC) is another, however poorly quantified source of OC. Very little is known about the impact of natural biological and marine sources. Due to the high significance of the EC/OC fraction of aerosols, a one-year EC/OC campaign within the EMEP network has been launched in the summer of 2002. In the future EMEP activities in this field are likely to be intensified.

1.1 References

CEN (1998) Air Quality. Determination of the PM10 fraction of suspended particulate matter. Reference method and field test procedure to demonstrate reference equivalence of measurement methods. Brussels, European Committee for Standardization (EN 12341).

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Jakobsen, M.C., Hansson, H.-C., Noone, K.J. and Charlson, R.J. (2000). Organic atmospheric aerosols: Review and state of the science. *Rev. Geophys., 38,* 267-294.

Lazaridis, M., Semb, A., Larssen, S., Hjellbrekke, A.-G., Hov, Ø., Hanssen, J. E., Schaug, J. and Tørseth, K. (2002). Measurements of particulate matter within the framework of the European Monitoring and Evaluation Programme (EMEP) I. First results. *Sci. Total Environ., 285*, 209-235.

2. Officially reported PM emissions

Zbigniew Klimont, Vigdis Vestreng and Leonor Tarrasón

This year, Parties to the Convention on Long Range Transboundary Air Pollution (CLRTAP) were requested, for the first time, to report particulate matter (PM) emissions. Parties were requested to report PM emission data for the year 2000 according to the Draft Guidelines for Estimating and Reporting Emission data (EB.AIR/GE.1/2001/6 and Add.1).

Twenty of the forty-eight Parties to the Convention submitted PM emission data to the UNECE. It is encouraging that as many as eleven Parties (23%) submitted national total and level 1 sector PM_{10} emissions. The reporting of fine fraction ($PM_{2.5}$) was somewhat lower (17%). Five Parties submitted PM data at NFR (Nomenclature For Reporting) level 2. In addition, France submitted PM data at SNAP 2 level. Six Parties provided gridded total emissions and three Parties reported gridded sector PM emissions.

Despite the encouraging number of initial reporting, the number of submissions of officially reported data is far from complete and steps should be made to allow for the completion of the official UNECE/EMEP PM emissions inventory for the year 2000. There are still important discussions on the type of emission sources to be considered in a European based PM emission inventory and further analysis is necessary in order to establish appropriate emission factors for all identified sectors.

The information compiled and distributed under the "Coordinated European programme on Particulate Matter Emission Inventories, Projections and guidance" (CEPMEIP) and the emission inventory produced at CIAM by the RAINS model are presently available but the completion of a European-wide PM emission inventory would require further collaboration with national experts, possibly through a series of arranged workshops.

In this chapter we present an evaluation of the differences in available emission information for four different countries (Austria, France, the Netherlands and United Kingdom) for the year 1995. The evaluation is intended as an illustration of the type of discussion that would eventually lead to the creation of a harmonized European-wide PM emission inventory.

2.1 Primary PM emission data reported in 2002

For the first time this year, Parties to the Convention on Long Range Transboundary Air Pollution (CLRTAP) were requested to report particulate matter (PM) emissions according to the Draft Guidelines for Estimating and Reporting Emission data (EB.AIR/GE.1/2001/6 and Add.1).

A total of 20 Parties to the Convention submitted some PM emission data to the UNECE, 19 Parties submitted some PM data for the year 2000 and 15 Parties reported some PM emission information for 1995. Actual emission PM data for 2000 is available via Internet at http://webdab.emep.int. Reported annual totals

can be found in Vestreng and Klein (2002). An overview of officially reported PM data available from the UNECE/EMEP database can be found in Annex I, Table A1-Table A3.

The spatial distribution of PM_{10} and $PM_{2.5}$ emissions for use in model assessments at MSC-W for year 2000 model runs are shown in Figure 2.1 and Figure 2.2.

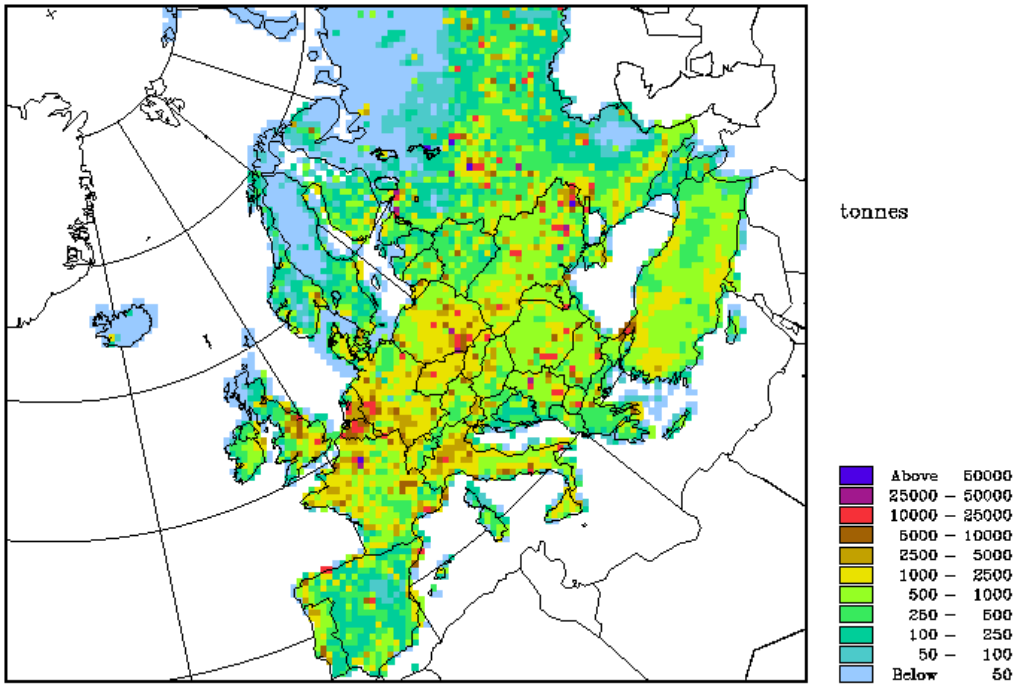

Figure 2.1: Emissions of PM_{10} in 2000 at 50 km resolution (Mg).

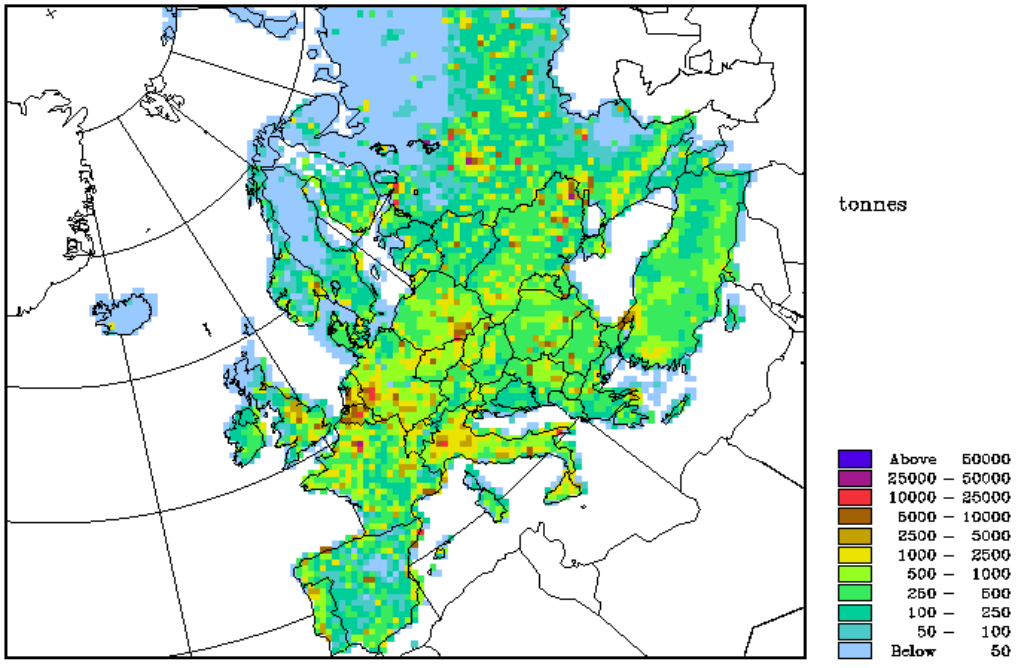

Figure 2.2: Emissions of $PM_{2.5}$ in 2000 at 50 km resolution (Mg).

The gridded 1995 emission distributions from the CEPMEIP project were scaled with emission data reported for 2000 by Austria, Denmark, Finland, France, Hungary and United Kingdom, otherwise the emission data input represents the 1995 emission level as retrieved from the CEPMEIP web site: http://www.air.sk/tno/cepmeip/emissions.php. Officially reported 2000 emission data from other Parties could not be included due to lack of completeness and/or consistency of the submission.

2.2 The way to an official 2000 primary PM inventory

Despite the encouraging number of initial reporting, the number of submissions of officially reported data is far from complete and steps should be made to allow for the completion of the official UNECE/EMEP PM emissions inventory for the year 2000. The year 2000 is envisaged already as reference year in future negotiations under the Convention and therefore deserves further attention.

A vast amount of information has been already collected during the CEPMEIP project. An extensive database of activity data and emission factors for several important sources has been established. However, there are still important discussions on the type of emission sources to be considered in a European based PM emission inventory and further analysis is necessary in order to establish appropriate emission factors for all identified sectors. Although the experience gained in CEPMEIP project and parts of collected data could be directly used in compilation of the inventory for the year 2000, a number of improvements/ extensions are necessary to adapt CEPMEIP approach to the needs of the inventory for 2000, in particular collection of the activity data is generally considered more accurate when compiled by the individual countries.

A questionnaire was circulated among the Parties to the Convention that reported PM emission for 1995, the year of reference of the CEPMEIP Inventory, in order to evaluate the usefulness of the CEPMEIP initiative (see also EMEP Note 2/2002). Of the 10 Parties that responded to the questionnaire, only one Party, Hungary, was not aware of the existence of the CEPMEIP. All Parties found the information in the CEPMEIP website easy to find and considered the project useful, in particular with respect to the availability of emission factors. Information on activity data from CEPMEIP was only partially used by the countries, mostly for comparison purposes, as the general view was that the CEPMEIP inventory could be completed and improved by using the activity data provided by the Parties. The most useful information from the CEPMEIP project was the availability of emission factors. However, the Parties recognized the need to further document and update the emission factor information provided by the CEPMEIP. This update can only be carried out in cooperation with national experts, through comparisons addressed to identify and fill in gaps in their system or update information for some of the sectors.

In the following section, a comparison of national reported data, CEPMEIP data and results from the RAINS PM model is presented. The RAINS PM model that has been recently extended and updated (Klimont et al., 2002) taking into account the results of CEPMEIP study (CEPMEIP, 2002), several national (e.g., APEG, 1999; Winiwarter et al. 2001; CITEPA, 2001; BUWAL, 2001; EWE, 2000; UBA, 1998; Pfeiffer et al., 2000, etc.) and international PM inventories. The RAINS

activity databases for 1995 use information from international statistics and modeling studies (e.g., UN, 2002; EC, 1996; EC, 1999a,b,c; FAO, 2002; IEA, 1998; etc.) as well as CEPMEIP project database available from the Internet (CEPMEIP, 2002).

The inter-comparison for 1995 data is intended as an example of the discussions, possibly thorough different organized workshops, that would eventually lead to the creation of a harmonized European-wide PM emission inventory. Cooperation with CIAM is encouraged to verify and update the databases used in the RAINS model.

2.3 Discussion of differences between 1995 inventories

As indicated in the previous section, an analysis of PM emission data has been performed for selected countries. In order to explain the reasons for the observed differences in the 1995 inventory, an analysis of emissions reported by Parties and calculated by CEPMEIP has been carried out on a lower level of aggregation, i.e., SNAP level 1 and 2 or NFR level 2. Additionally, results of the RAINS PM model, developed at CIAM/IIASA (Klimont et al., 2002), were used.

Only those Parties that reported own estimates of national total, sector level 1 (SNAP or NFR), and level 2 (SNAP or NFR) data on emissions of particulate matter were selected for the inter-comparison. This excludes Czech Republic (national total for TSP), Estonia, Finland, Hungary and Switzerland (national, sector level 1 only, or based on CEPMEIP) (Annex I, Table A1-Table A3). Therefore an in depth analysis can only be made for Austria, France, Netherlands and United Kingdom that meet all the criteria.

Table 2.1 shows, for selected countries, a comparison of the national total emissions reported to UNECE under the LRTAP in 2002, with emission estimates from the CEPMEIP and the RAINS model. All estimates are for the year 1995. CEPMEIP and national estimates were already compared in the previous section. The RAINS model calculation for TSP is similar to CEPMEIP, with slightly better match for Austria and the Netherlands but even worse, compared to the national estimate, for France. Also for PM_{10}, RAINS estimates are significantly lower than CEPMEIP and national source, while the differences for other countries between all inventories are lower than ten percent. The same pattern, as for PM_{10}, is observed for fine particles. In order to evaluate further the differences, it was decided to look at PM_{10} estimates disaggregated at SNAP 1 and lower levels. This is because only for PM_{10} disaggregated national data are available for all discussed countries.

Table 2.1: National total 1995 emissions comparison.

Year: 1995	UNECE	CEPMEIP	RAINS	UNECE	CEPMEIP	RAINS	UNECE	CEPMEIP	RAINS
Units: Gg	PM2.5			PM10			TSP		
Austria	27.6	33.5	31.0	46.8	46.2	43.5	75.8	82.7	77.5
France	336.0	350.6	205.3	587.7	449.7	289.0	1525.0	693.3	527.3
Netherlands	n.a.	41.4	37.6	60.9	64.4	62.0	75.0	127.3	117.7
United Kingdom	132.3	164.4	154.7	237.9	259.6	261.1	n.a.	473.4	555.6

Tables 2.2 and 2.3 provide data on SNAP 1 level for all four considered countries and inventories. Below a discussion of differences observed between inventories is provided.

2.3.1 Austria

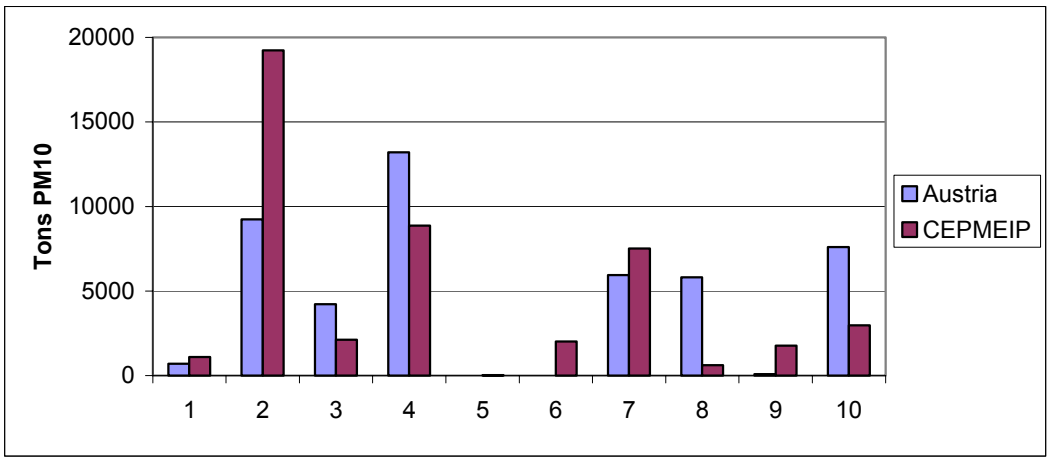

Figure 2.3: ***Austria:*** *Comparison of PM_{10} level 1 sector data 1995 reported with CEPMEIP.*

Although the total PM_{10} emissions differ between the studies by only about five percent, the sectoral comparison reveals great variation in estimates. A brief discussion below concentrates on the sectors for which major differences occur. A study by Winiwarter et al. (2001) that is the background document for the Austrian national contribution to EMEP, is used to investigate detailed assumptions behind Austrian estimates.

For *residential combustion plants* CEPMEIP estimates twice as much as RAINS or national inventory. Analysis of detailed assumptions (actually beyond the SNAP level 2) shows that in all studies major contribution comes from wood combustion (between 70 and 85 percent) and there are two factors that affect the estimates, activity data and emission factors. There is a significant difference in activity data, CEPMEIP having nearly 30 percent higher wood consumption in this sector than the other studies. Also emission factors differ; while national inventory and RAINS distinguish between emission factors for households (stoves, single-family boilers) and residential plants, CEPMEIP has one category "non-industrial combustion plants' for which one emission factor is used. The CEPMEIP emission factor is about the same as the one used in Austrian inventory for households. Emission factor for larger installations, often controlled, is typically lower. The difference between RAINS and national inventory is mostly due to different assumptions about the distribution of fuel used in households and residential plants.

For *combustion in manufacturing industry*, both CEPMEIP and RAINS seem to underestimate emissions. There is some difference in the calculation for processes with contact (SNAP level 2, 030300), where national study reports higher emissions, but the majority of the observed discrepancy (about 1.6 Gg) has its

roots in underestimation of emissions from combustion (030100). This is only to a limited extend linked to differences in activity data, the more important reason are emission factors or rather assumptions about the level of abatement in industry. Full explanation is only possible in direct consultation with national experts.

In case of *production processes*, there is a factor of two difference between studies, at least when comparing SNAP 4. However, in case of RAINS, some adjustments were made using the Winiwarter et al. (2001) study that was available at the time of updating the RAINS model and therefore about 6 Gg were added to category 'other' (sector 12 in the table below) to represent emissions from activities that were missing from the database, e.g., production of limestone, silicate, dolomite, sand, etc. CEPMEIP could not have done it, as the results of the Austrian inventory were not available at that time. Therefore RAINS has a fairly good agreement but still overestimates emission from iron and steel industry by nearly 30 percent. More detailed look into CEPMEIP inventory shows an overestimation of emissions from iron and steel by a factor three and missing emissions from mineral industry, similarly to the first RAINS iteration. This example shows how important it is to collaborate closely with national experts to include all sources that are relevant for a given country. Otherwise they might be easily overlooked when attention is paid to activities that are relevant only in European scale. Another difference in estimated for this sector is linked to the fact that Winiwarter et al. (2001) includes emissions from construction works in this category, while CEPMEIP allocated it to SNAP 6 (solvent use) and RAINS into category '12' (not included in RAINS). However, overall contribution of this particular source to PM_{10} emissions is relatively small.

Although agreement for emissions from *road transport* is very good on an aggregated level there are significant differences between SNAP 2 categories, mostly for heavy-duty truck and non-exhaust emissions; the latter being a very uncertain source in terms of overall emissions and size distribution. Additionally it was observed that national inventory neglects exhaust PM emissions from gasoline vehicles, while CEPMEIP and RAINS estimate them for Austria at about 0.6 Gg.

For *other mobile sources and machinery*, there is a striking difference between CEPMEIP and other studies. Verifying data on a lower aggregation level shows that CEPMEIP included only emissions from railway and air traffic, while major contribution seems to come from agriculture and forestry (Winiwarter et al., 2001; UNECE/EMEP database). RAINS estimates are quite consistent with national inventory, although lower by about 0.5 Gg. The reason for that is missing estimate from air traffic in RAINS.

Although overall contribution of *waste treatment* to PM_{10} emissions is expected to be relatively small, a large discrepancy is observed between the studies. It seems that CEPMEIP and RAINS overestimate emissions from open burning of residential waste (the only source of emissions from this sector in RAINS) and additionally CEPMEIP reports 1.3 Gg of emissions from municipal waste incineration. Austrian inventory and RAINS do not show any emissions from this source as it is assumed that a vast majority of the waste is burned in well controlled plants and generated energy (heat) is used and therefore it is shown

under SNAP 1. This is remark is valid for all countries that will be discussed further. CEPMEIP assumes large quantities of municipal waste that is incinerated, i.e., for Austria it is 43 million tons; a very large number. We believe that this needs to be verified by national experts.

For *agriculture*, national estimate is twice as high. As discussed in Winiwarter et al. (2001) there is significant difference in emission rates used between CEPMEIP and national inventory for livestock and Austrian study includes also emissions from arable farming which dominate the total. RAINS relies also on a different background information for livestock, somehow similar to CEPMEIP, and, although includes emissions from arable farming, the contribution of this source to PM_{10} is by orders of magnitude smaller than in the national inventory. One has to bear in mind that these are very uncertain sources of emissions and there are only few studies that investigated them. Even less is known about the size distribution of PM emissions from these sources.

Finally, CEPMEIP and RAINS include *other sources of emissions* that are often not included in national inventories, i.e., cigarette smoking, barbeques, and fireworks. In case of RAINS they are reported in category '12' (Other, not included in CORINAIR) while CEPMEIP reports them under SNAP 6 (solvent use). The estimates are the same as RAINS estimates are derived from the CEPMEIP database. The mentioned category '12' (also included in the tables below) should not be confused with sector 12 (Excluded) that is reported in the CEPMEIP inventory (compare CEPMEIP results available on the Internet) and includes emissions from bunker fuel.

2.3.2 France

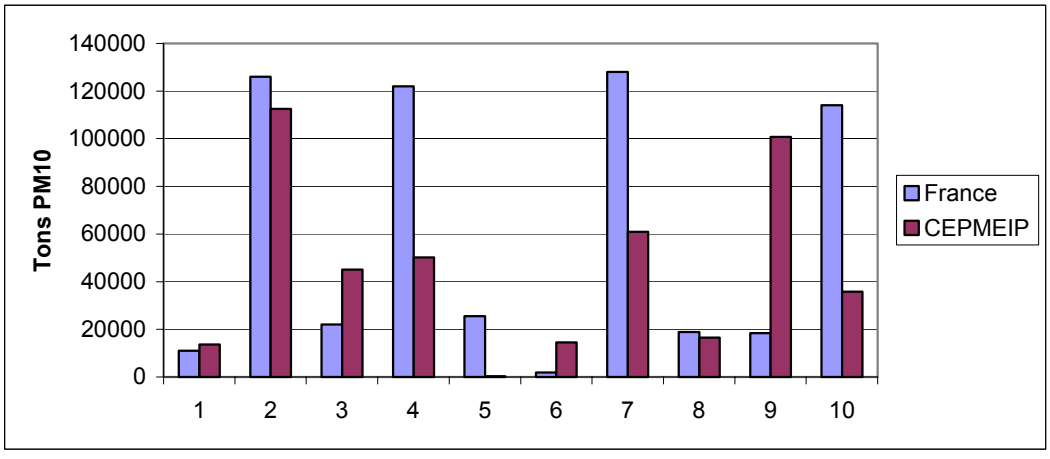

Figure 2.4: ***France****: Comparison of PM_{10} level 1 sector data 1995 reported with CEPMEIP.*

There are very significant differences in total PM_{10} emissions between the studies. CEPMEIP estimate is by about 25 percent and RAINS by 50 percent lower than the national submission that is based on the report by CITEPA (2001). This study and SNAP 2 level submission as well as detailed CEPMEIP and RAINS results

are used to analyze the differences. The sectoral comparison reveals even greater variation in estimates.

For *residential combustion plants* CEPMEIP and national estimate is about the same, while RAINS calculates about half the amount. Similarly to Austria, the main contribution comes from wood combustion (between 80 and 95 percent) and there are two factors that affect the estimates, activity data and emission factors. There is a difference in activity data of about 12 percent, i.e., RAINS has about 45 PJ less wood burned in this sector than other studies. But this is not making up for the large discrepancy. The principal reason is the use of different emission factors. RAINS emission factors assumed for France (similar to values used for Austria) are nearly 40 percent lower than what is used in CEPMEIP study and most likely by CITEPA (2001)[2].

For *combustion in manufacturing industry*, there is a pretty good agreement found for RAINS and the national inventory (also on the SNAP 2 level). CEPMEIP, however, estimates 100 percent higher emissions than the national work. CITEPA (2001) calculates total emissions from combustion in boilers at about 10 Gg while CEPMEIP has more from plants in pulp and paper industry burning black liquor and bio-waste (about 6 Gg) and from use of heavy fuel oil in various industries (above 5 Gg), the rest coming primarily from coal combustion. Also for processes with contact (030300) CEPMEIP estimate is twice as high, cement production alone being responsible for as much as total emissions from this category reported in the national study, i.e., 10 Gg. Other major process is sinter plant where, according to CEPMEIP, nearly 6 Gg of PM_{10} are emitted.

In case of *production processes*, there is a factor of two differences between the national inventory and the other studies. While a significant discrepancy was found for iron and steel industry, CEPMEIP having 100 percent higher emissions than both CITEPA and RAINS, the major difference arises from category 040600, where a large number of processes is included. CEPMEIP and RAINS estimate emissions from this sector at about 20 Gg and CITEPA reports nearly 108 Gg. Since no more detailed information is available from the CITEPA (2001) report, it was not possible to trace back the reasons for such a big variation. It is possible that, similarly to Austria, both international studies omit important sources of emissions and therefore a consultation with national experts is necessary to explain this. Additionally, an error in the CEPMEIP emission factor database was found for one of the sub-sectors in this category. It does not affect emissions of PM_{10} as only emission factor for TSP has been mistyped (it is actually lower than emission factor for $PM_{2.5}$) and it leads to an underestimate of TSP emissions from small industrial sources (fugitive) by about 30 Gg.

A large difference was found for *extraction and distribution of fossil fuels* sector, i.e., national inventory reporting an order to two orders of magnitude higher emissions than other studies. However, lack of more detailed information from the CITEPA study did not allow for explanation of this difference.

[2] Unfortunately, report by CITEPA does not include emission factors assumed for this source. Also for other sources only few emission factors are reported in that study.

The difference for *"solvent use"* sector between national inventory and other studies is large but on the basis of the available it is not possible to explain. CEPMEIP and RAINS do not calculate PM emissions from solvent use, however, CEPMEIP reports here emissions from construction, barbeques, cigarette smoking, etc.

There is no good agreement for emissions from *road transport*. CEPMEIP estimates about half the amount of the national study. The main differences are found in passenger cars and light duty vehicles (half of the CITEPA estimate) and non-exhaust sources, where the difference is about 45 Gg. Although, RAINS estimate for exhaust emissions matches the national inventory pretty well, there is a large difference in non-exhaust estimate, i.e., 40 Gg. According to CITEPA estimates, a major source of PM_{10} from road transport is 'road wear' contributing 45 Gg and making up for most of the variation observed between the studies. This is not confirmed in RAINS (CEPMEIP does not include this source) where emissions from road abrasion are estimated at only about ten percent of CITEPA value. Emissions from road abrasion and re-suspension are very uncertain and needs to be evaluated carefully in collaboration with national experts.

For *other mobile sources and machinery*, the differences are not very large on aggregated level but a closer look at the data on a lower aggregation level shows few important differences. RAINS emissions are larger than other estimates mainly due to relatively high emissions from construction and industrial machinery as well as two stroke engines and CEPMEIP estimate is lower than other studies owing to lack of emissions from agricultural tractors that is only partly compensated by inclusion of military sources that are not included (at least not directly reported) in the national inventory and RAINS.

Although overall contribution of *waste treatment* to PM_{10} emissions is expected to be relatively small, a large discrepancy is observed between the studies. It seems that CEPMEIP and RAINS underestimate emissions from open burning of agricultural waste, simply by assuming that they are zero, i.e., effective ban on this activity is present. CITEPA reports about 13.7 Gg of PM_{10} from this source. The largest discrepancy, however, can be traced back to the municipal waste incineration for which CEPMEIP reports emissions of nearly 98 Gg. French inventory suggests emissions of 4.7 Gg from this source. CEPMEIP assumes large quantities of municipal waste that is incinerated in France, i.e., 975 million tons. We believe this needs to be verified by national experts (see also discussion of this sector for other countries).

For *agriculture*, national estimate is higher by a factor four to five. Although there is a difference in estimates for emissions from livestock (the only category included in CEPMEIP), the main reason for larger emissions in CITEPA's estimate is sector 100100 (cultures with fertilizers); a source of 98 Gg. RAINS estimates that PM_{10} emissions from arable farming are below 2 Gg, although they might be very high for TSP. One possible explanation is that CITEPA has used American studies where very high emission rates were reported. It is, however, important to consider to what extend the results of these studies are transferable to the European situation. RAINS calculation for arable farming relies on work done in the UK (ICC and SRI, 2000) and may represent a lower estimate.

Finally, CEPMEIP and RAINS include *other sources of emissions* that are often not included in national inventories, i.e., cigarette smoking, barbecues, and fireworks. In case of RAINS they are reported in category '12' (Other, not included in CORINAIR) while CEPMEIP reports them under SNAP 6 (solvent use). The estimates are the same as RAINS estimates are derived from the CEPMEIP database. Although CITEPA reported additional sources of PM (forest and other vegetation fires), they were excluded from this comparison.

Table 2.2: Comparison of PM_{10} estimates for Austria and France on SNAP 1 level, Gg.

SNAP 1 sector	Austria			France		
	UNECE	CEPMEIP	RAINS	UNECE	CEPMEIP	RAINS
1: Combustion in energy industries	0.75	1.10	1.16	10.96	13.66	10.33
2: Non-industrial combustion plants	9.20	19.23	10.32	125.55	112.50	58.89
3: Combustion in manufacturing industry	5.15	2.13	2.88	22.13	45.00	20.48
4: Production processes	12.25	8.87	6.06	122.35	50.09	43.79
5: Extraction and distribution	-	0.03	0.43	25.46	0.21	2.60
6: Solvent use	-	2.02	-	1.87	14.44	-
7: Road transport	7.14	7.52	7.96	127.71	60.89	87.03
8: Other mobile sources and machinery	3.09	0.61	2.58	18.76	16.47	23.43
9: Waste treatment	0.09	1.77	0.48	18.41	100.72	3.13
10: Agriculture	7.60	2.97	3.25	114.45	35.75	24.84
12: Other (not included in CORINAIR)	-	-	8.42	-	-	14.49
TOTAL	45.27	46.24	43.53	587.74	449.73	289.01

2.3.3 Netherlands

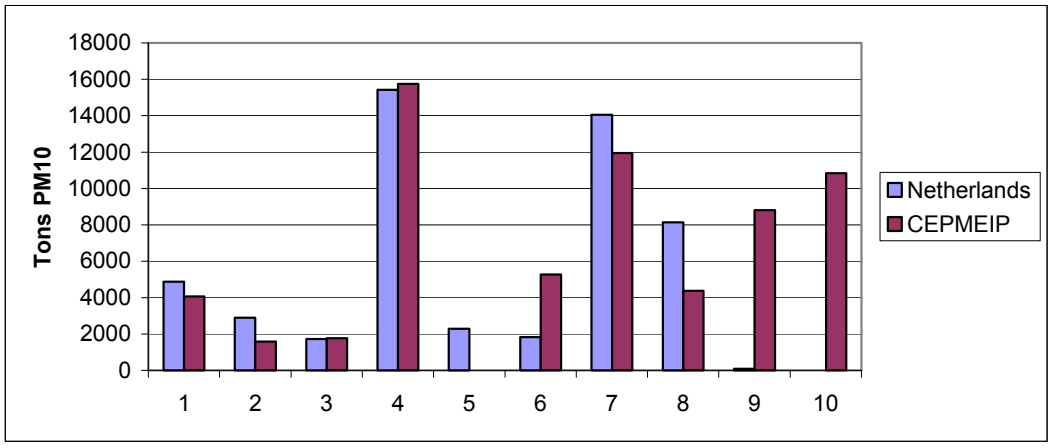

*Figure 2.5: **Netherlands**: Comparison of PM_{10} level 1 sector data 1995 reported with CEPMEIP.*

The total PM_{10} emissions differ between the studies by only a few percent and, in fact, the Netherlands is the only country where also sector estimates match very

well for majority of sectors. It was more difficult to explain some of the differences, though, since we had no access to background documentation where information about activity data and emission factors used would be available.

For *residential combustion plants*, the national inventory reports higher emissions than CEPMEIP and RAINS but it is not possible to explain them on the basis of available information. It is very likely that the reasons for differences are similar to other countries, i.e., wood combustion and assumptions associated with this activity. According to CEPMEIP and RAINS about 70 percent of emissions from this sector originate from wood combustion.

A difference was found for *extraction and distribution of fossil fuels* sector, i.e., national inventory reporting similar emissions to RAINS, while CEPMEIP reporting none at all. RAINS emissions originate from storage of coal, while the national inventory estimate is associated with the NFR category 5C (Geothermal energy and other). No additional data is available.

For the difference in *"solvent use"* sector please see the discussion for France and comments to 'other sources' (category '12') below.

Emissions from *road transport* reported by CEPMEIP differ by about 2 Gg (15 percent) from other studies and the sole reason for that is the lower estimate of emissions from passenger cars by CEPMEIP.

For *other mobile sources and machinery*, the national inventory suggests much larger emissions. The difference to RAINS is easy to explain, i.e., RAINS estimates only 0.3 Gg PM_{10} from agriculture and forestry while in the national study as much as 2.5 Gg are emitted from this source. In CEPMEIP, several categories are missing: agriculture, forestry, industry, and households, while at the same time military sources are included. The latter are not included (at least not directly reported) in the national inventory and RAINS model.

For *waste treatment*, the story repeats itself (see discussion for Austria and France). CEPMEIP assumes large quantities of municipal waste that is incinerated, i.e., 276 million tons, and consequently estimates emissions of 8.3 Gg of PM_{10}. We believe this needs to be verified by national experts.

For *agriculture*, there seem to be fairly good agreement, bearing in mind uncertainties for this source. Verifying the allocation of emissions on a lower level of aggregation, however, reveals that in the national inventory all emissions are associated with NFR10A (agricultural soils) rather than livestock. This needs to be explained.

Finally, CEPMEIP and RAINS include *other sources of emissions* that are often not included in national inventories, i.e., cigarette smoking, barbecues, and fireworks. In case of RAINS they are reported in category '12' (Other, not included in CORINAIR) while CEPMEIP reports them under SNAP 6 (solvent use). The estimates are the same as RAINS estimates are derived from the CEPMEIP database. The national inventory reported also emissions of PM_{10} from solvent use and it remains to be explained if they are really associated with this

sector or, similarly to CEPMEIP, some other sources where allocated into that category.

2.3.4 United Kingdom

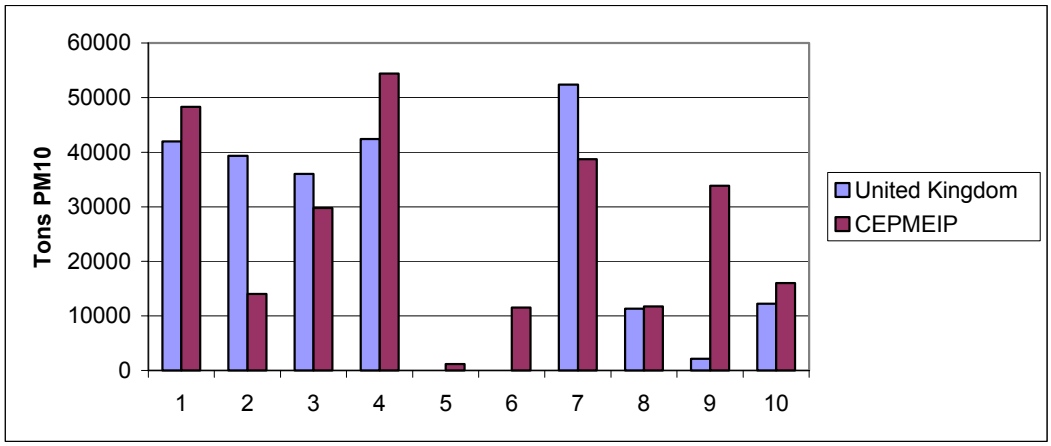

Figure 2.6: ***United Kingdom****: Comparison of PM_{10} level 1 sector data 1995 reported with CEPMEIP.*

CEPMEIP and RAINS estimates of total PM_{10} emissions for UK are higher than the national inventory by about ten percent. Similarly to the Netherlands, we have not received a detailed background documentation, which makes it more difficult to explain some of the differences.

For *power plants*, CEPMEIP estimates higher emissions than other studies. The main reason seems to be significantly higher estimate for petroleum refining plants as well as about ten percent higher emissions from public power plants.

For *residential combustion plants*, the CEPMEIP estimate is much lower than the national inventory and RAINS. In contrary to other countries discussed, a major source of PM from residential sector in UK in 1995 is coal combustion. The activity data information seems to be the same but there is a difference between emission factors. We do not have such information for the national inventory but comparing CEPMEIP to RAINS, shows a difference by up to a factor three. RAINS distinguishes between stoves, single-family boilers and larger residential plants, differentiating emissions factors and level of control. CEPMEIP, on the other hand, assumes only one set of emission factors for hard coal, and they are significantly lower.

Large differences are found for emissions from *production processes*. CEPMEIP estimate is about two times higher than the national inventory and it can be traced back to the discrepancy for iron and steel industry, i.e., CEPMEIP estimate is about four times higher. RAINS, on the other hand, calculates about 40 percent less PM_{10} emissions from this source than the national study. In this case, iron and steel industry is about the same but there are a number of sources that are included in the national inventory but are missing in RAINS, e.g., sulphuric acid, pulp and paper, road paving with asphalt. Emissions from these particular sectors were

added to the RAINS database in category 'Other' but it does not explain all the difference found between studies. The reason is a number of processes that are included under SNAP category 040600 but owing to lack of more detailed information it is not possible to explain it further.

A difference was found for *extraction and distribution of fossil fuels* sector, i.e., national inventory not reporting emissions from this source, while RAINS estimates about five times more than CEPMEIP. Overall, it is not a major source but needs to be investigated further. RAINS emissions originate primarily from storage and handling of coal.

For the difference in *"solvent use"* sector see the discussion for France and comments to 'other sources' (category '12') below.

Emissions from *road transport* reported by CEPMEIP are nearly 30 percent lower than the national inventory; on the other hand RAINS estimate is by nearly 10 percent higher. The reasons for differences are not the same. CEPMEIP estimate for passenger cars and light duty vehicles is about half of the national estimate, resulting in absolute difference of about 12 Gg. RAINS estimates much higher emissions from non-exhaust sources, which explains most of the variation.

For *other mobile sources and machinery*, RAINS deviates from the other studies. The reason for that are much higher emissions from inland navigation and coastal shipping calculated in RAINS. This needs to be evaluated in close collaboration with national experts.

For *waste treatment*, the same discrepancies are observed as for other countries. CEPMEIP assumes large quantities of municipal waste that is incinerated, i.e., 292 million tons, and consequently estimates emissions of 29.2 Gg of PM_{10}. We believe that this needs to be verified by national experts. It is also worth noting that from about the same amount of waste (276 Mt) only about 8.3 Gg of PM_{10} were calculated in CEPMEIP inventory for the Netherlands.

For *agriculture*, there seem to be fairly good agreement, bearing in mind uncertainties for this source. Only CEPMEIP estimate is a bit higher than national inventory and RAINS.

Finally, CEPMEIP and RAINS include *other sources of emissions* that are often not included in national inventories, i.e., cigarette smoking, barbecues, and fireworks. In case of RAINS they are reported in category '12' (other, not included in CORINAIR) while CEPMEIP reports them under SNAP 6 (solvent use). The estimates are the same as RAINS estimates are derived from the CEPMEIP database.

Table 2.3: Comparison of PM_{10} estimates for the Netherlands and United Kingdom on SNAP 1 level, Gg.

SNAP 1 sector	Netherlands			United Kingdom		
	UNECE	CEPMEIP	RAINS	UNECE	CEPMEIP	RAINS
1: Combustion in energy industries	4.88	4.07	4.69	42.70	48.34	42.03
2: Non-industrial combustion plants	2.90	1.59	1.65	39.35	14.02	39.62
3: Combustion in manufacturing industry	1.72	1.77	2.16	36.00	29.80	31.1
4: Production processes	15.40	15.75	14.69	42.40	54.39	26.42
5: Extraction and distribution	2.29	-	1.48	-	1.19	9.78
6: Solvent use	1.82	5.28	-	-	11.54	-
7: Road transport	14.05	11.95	13.84	52.40	38.72	57.08
8: Other mobile sources and machinery	8.14	4.37	5.10	11.30	11.72	18.53
9: Waste treatment	0.09	8.81	0.53	1.44	33.82	4.62
10: Agriculture	9.56	10.83	12.51	12.26	16.02	13.73
12: Other (not included in CORINAIR)	-	-	5.29	-	-	18.14
TOTAL	60.85	64.42	61.96	237.9	259.56	261.05

2.3.5 Summary of main differences

Below a short summary of major discrepancies observed between the discussed inventories for 1995 are summarized, indicating major sources of differences and reasons for that. Please note that reasons for a particular country may be slightly different or that more/other factors might contribute.

Residential combustion; the differences originate primarily for wood combustion. The origins of the observed discrepancies are different emissions factors and activity data (for wood it is very difficult to account for wood that is not purchased but 'collected' and these estimates are both uncertain and not always available). It is also important to note that in this sector there is a variation in how the activities are represented in considered inventories, i.e., including distinction between for example fireplaces, stoves and boilers or not.

Production processes; a variety of reasons for discrepancies in estimates ranging from different assumptions on activity data and processes included to the level of control assumed.

Road transport; the differences are mostly for non-exhaust emissions and their size distribution. A more general problem with emissions from transport is reporting/estimating emissions from two-stroke engines (mopeds and other machinery).

Off road; the differences stem primarily from the difficulty in finding appropriate activity data. There are a number of studies reporting on emission factors from various types of machinery but the accounting/reporting practices as far as fuel consumption is concerned vary across the countries and statistical sources.

Waste treatment; estimates of emissions from open burning of agricultural and residential waste are very uncertain as both activity data and emission factors are not well documented. However, and important element that contributes to the

differences in estimates is “municipal waste incineration” reported in CEPMEIP study. The assumptions about the amount of waste burned are very high. This needs to be verified by the national experts; especially that none of the countries discussed estimated any significant emissions from this category in their national studies.

Other sources; there is also an issue of missing sources or sources accounted in other SNAP/NFR categories that cannot be easily compared. For example, CEPMEIP decided to use sector “solvent use” for reporting emissions from construction activities, cigarette smoking, barbeques, fireworks, etc. This leads to confusion when viewing the emission results on SNAP 2 level where emissions from these activities appear under “paint application” or “use of HFC, N_2O, NH_3, PFC, and SF_6” categories of the SNAP nomenclature.

In addition, we should keep in mind that semi-anthropogenic and natural sources of particulate matter like forestry, natural soil dust emissions and soil dust emissions from agriculture and biological pollen emissions are not considered in the CEPMEIP inventory. Neither is re-suspension of road dust included in the emission. Further separate effort should be made in order to estimate the extent of these emission sources.

The completion of the official UNECE 2000 primary PM emission inventory would require further collaboration with national experts, possibly through a series of arranged workshops. This collaboration is essential both to complete the overview of emission sources and to verify the assumptions about the emission factors used for specific countries. Further cooperation with national experts is also necessary in view of further developments in control technology, better operating practices and introduction of new laws requiring more efficient abatement.

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Annex I

(Annex to Chapter 2)

Table A1: PM2.5: Overview of national emission totals, sector data and gridded data reported and stored at the UNECE/EMEP emission database at MSC-W.

Totals	NFR level 1	NFR level 2	Gridded 50km x 50km
X	X	X	X

Party/Year	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2010	2020
Armenia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Austria	X---	X---	X---	X---	X---	X---	X---	X---	X---	X---	XXX-	X---	X---	X---	X---	XXX-	X---	X---	X---	XXX-	XXX-	----	----
Belarus	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Belgium	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Bosnia and Herzegovina	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Bulgaria	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Canada	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Croatia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Cyprus	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Czech Republic	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Denmark	----	----	----	----	----	----	----	----	----	----	---X	----	----	----	----	---X	----	----	----	----	XXXX	----	----
Estonia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----	----	----	----	----	----
Finland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----	----	----	XXXX	----	----
France	----	----	----	----	----	----	----	----	----	----	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	----	----
Georgia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Germany	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Greece	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Hungary	X---	----	----	----	----	X---	----	----	X---	----	X---	X---	X---	X---	X---	XX--	XX--	XX--	XX--	XX--	XX--	----	----
Iceland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Ireland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Italy	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Kazakhstan	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Kyrgyzstan	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Latvia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Liechtenstein	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Lithuania	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Luxembourg	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Malta	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Monaco	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Netherlands	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Norway	----	----	----	----	----	----	----	----	----	----	--X-	--X-	--X-	----	--X-	--X-	--X-	--X-	--X-	--X-	--X-	----	----
Poland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----
Portugal	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Republic of Moldova	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Romania	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Russian Federation	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Slovakia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Slovenia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Spain	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Sweden	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Switzerland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	X---	----	----	----	----	----	----	----
The FYR of Macedonia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Turkey	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Ukraine	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
United Kingdom	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	----	----
United States	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----
Yugoslavia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
European Community	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----

Table A2: PM10: Overview of national emission totals, sector data and gridded data reported and stored at the UNECE/EMEP emission database at MSC-W.

Totals	NFR level 1	NFR level 2	Gridded 50km x 50km
X	X	X	X

Party/Year	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2010	2020
Armenia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Austria	X---	X---	X---	X---	X---	X---	X---	X---	X---	X---	XXX-	X---	X---	X---	X---	XXX-	X---	X---	X---	XXX-	XXX-	----	----
Belarus	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Belgium	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Bosnia and Herzegovina	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Bulgaria	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Canada	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Croatia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Cyprus	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Czech Republic	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Denmark	----	----	----	----	----	----	----	----	----	----	---X	----	----	----	----	---X	----	----	----	----	XXXX	----	----
Estonia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----	----	----	----	----	----
Finland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----	----	----	XXXX	----	----
France	----	----	----	----	----	----	----	----	----	----	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	----	----
Georgia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Germany	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Greece	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Hungary	X---	----	----	----	----	X---	----	----	X---	----	X---	X---	X---	X---	X---	XX--	XX--	XX--	XX--	XX--	XX--	----	----
Iceland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Ireland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XXXX	----	----
Italy	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Kazakhstan	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Kyrgyzstan	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Latvia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Liechtenstein	----	----	----	----	----	----	----	----	----	----	XX--	----	----	----	----	XX--	----	----	----	----	XX--	XX--	----
Lithuania	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Luxembourg	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Malta	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Monaco	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Netherlands	----	----	----	----	----	----	----	----	----	----	XXXX	----	----	----	----	XXXX	----	----	XXX-	----	----	----	----
Norway	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Poland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----
Portugal	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Republic of Moldova	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Romania	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Russian Federation	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Slovakia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Slovenia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Spain	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Sweden	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Switzerland	----	----	----	----	----	----	----	----	----	----	XX--	----	----	----	----	XX--	----	----	----	----	XX-X	X---	----
The FYR of Macedonia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Turkey	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Ukraine	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
United Kingdom	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXXX	XXX-	----	----
United States	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----
Yugoslavia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
European Community	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----

Table A3: TSP: Overview of national emission totals, sector data and gridded data reported and stored at the UNECE/EMEP emission database at MSC-W.

Totals	NFR level 1	NFR level 2	Gridded 50km x 50km
X	X	X	X

Party/Year	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2010	2020
Armenia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Austria	X---	X---	X---	X---	X---	X---	X---	X---	X---	X---	XXX-	X---	X---	X---	X---	XXX-	X---	X---	X---	XXX-	XXX-	----	----
Belarus	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Belgium	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Bosnia and Herzegovina	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Bulgaria	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Canada	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Croatia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Cyprus	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Czech Republic	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	X---	X---	X---	X---	X---	XXX	----	----
Denmark	----	----	----	----	----	----	----	----	----	----	---X	----	----	----	----	---X	----	----	----	----	XXX	----	----
Estonia	----	----	----	----	----	X---	X---	X---	X---	X---	X---	X---	X---	X---	X---	XX--	X---	X---	X---	X---	XXX	----	----
Finland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----	----	----	XXX	----	----
France	----	----	----	----	----	----	----	----	----	----	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	XXX-	----	----
Georgia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Germany	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Greece	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Hungary	X---	----	----	----	----	X---	----	----	X---	----	XX--	XX--	XX--	XX--	XX--	XX--	XX--	XX--	XX--	XX-X	XX--	X---	X---
Iceland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Ireland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Italy	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Kazakhstan	----	----	----	----	----	----	----	----	----	----	X---	X---	X---	X---	X---	X---	X---	X---	X---	X---	X---	----	----
Kyrgyzstan	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Latvia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	X---	----	----
Liechtenstein	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Lithuania	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----
Luxembourg	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Malta	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Monaco	----	----	----	----	----	----	----	----	----	----	XX--	XX--	XX--	XX--	XX--	XX--	XX--	XX--	XX--	XX--	XX--	----	----
Netherlands	----	----	----	----	----	----	----	----	----	----	XXXX	----	----	----	----	XXXX	----	----	XXX-	----	----	----	----
Norway	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Poland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	XX--	----	----
Portugal	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Republic of Moldova	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Romania	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Russian Federation	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Slovakia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Slovenia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Spain	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Sweden	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Switzerland	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	X---	----	----	----	----	----	----	----
The FYR of Macedonia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Turkey	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	-X--	-X--	-X--	-X--	-X--	-X--	----	----
Ukraine	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
United Kingdom	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
United States	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
Yugoslavia	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----
European Community	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----	----

3. The Unified EMEP aerosol model version: initial results on particle mass, number and chemical composition

by Svetlana Tsyro and Leonor Tarrasón

For the first time, an aerosol dynamics model has been implemented in EMEP as integrated part of the Unified EMEP Eulerian modelling system developed at MSC-W. A short presentation of the model formulation and initial results on aerosol mass and number concentrations and chemical composition are presented in this chapter.

Previous calculations of PM mass within EMEP did not include the effect of aerosol dynamic processes and only for primary particles allowed the description of size dependent deposition processes. Instead, the new Unified EMEP aerosol model version (UNI-AERO) takes into account aerosol dynamics and allows for size dependent description of removal processes.

The initial testing of UNI-AERO has focused on the analysis of aerosol dynamic processes and of the effect of size-resolved dry and wet deposition on primary PM concentrations. A significant conclusion so far is that the partitioning of the fine aerosol between the Aitken and accumulation modes and the choice parameterisation of dry and wet deposition processes for the different size classes are the most relevant variables for the description of the transport of primary PM concentrations. Accounting for size-dependant emissions and depositions of aerosols in the model has a larger impact on the calculated primary PM concentrations than introducing coagulation processes into the model. These results stress again the importance of appropriate information on the size distribution of primary PM emissions for assessing PM concentrations. They also focus the need for accurate descriptions of size dependent dry and wet removal processes, and further studies on these topics are envisaged for the future.

To support the progress in the model development it is important to complement these sensitivity studies with validation of model calculations against observations. Some preliminary comparisons with observations are presented in this chapter, while further work on validation of UNI-AREO is envisaged for the autumn of 2002. For this reason, the results presented here should be regarded as interim and subject to possible changes as work on model development and testing progresses.

3.1 Model description

The aerosol version of the Unified EMEP Eulerian model system (UNI-AERO) distinguishes 7 gaseous components and 7 particulate matter components in 4 different size modes. UNI-AERO is flexibly designed so that the model can be extended as necessary to include more chemical and aerosol components.

The present version of UNI-AERO solves 27 prognostic equations (7 for gases and 20 for particle number and masses). As indicated in Table 3.1, work is under

progress to extend these to 32 prognostic equations by including new aerosol components. The EMEP Aerosol model version has shown to be relatively cost-efficient in terms of CPU usage.

The gaseous components used explicitly in the aerosol model version are primarily those described by the acidification version of the Unified EMEP model, namely the following sulphur and nitrogen compounds: SO_2, H_2SO_4, NO, NO_2, HNO_3, PAN, NH_3. The seven chemical aerosol components are: sulphate (SO_4), nitrate (NO_3), ammonium (NH_4), organic carbon (OC), elemental carbon (EC) mineral dust and sea salt (NaCl). The four size modes are: the nucleation mode (diameters below 0.02 μm), Aitken mode (between 0.02 μm and 0.1 μm), accumulation mode (between 0.1 μm and 2.5 μm) and the coarse mode (between 2.5 μm and 10 μm). In addition, aerosol water content is included as a diagnostic variable.

An overview of the aerosol prognostic variables used in UNI-AERO is given in Table 3.1. All particles within each mode are assumed to have the same size (monodisperse) and chemical composition (internally mixed aerosols).

Aerosol water is considered in the model as a diagnostic parameter and is calculated based on a scheme developed by Binkowski and Shankar (1995). In UNI-AERO, hygroscopic particles are assumed to be in stable equilibrium with the surrounding water wapor, such that they grow by absorbing water if the relative humidity increases and they shrink due to water evaporation if the humidity decreases.

Please note that secondary organic aerosol (SOA), natural dust (e.g. Saharan dust) and bio-aerosols are not currently included in the model. SOA are not included in the present model version because their formation mechanisms are still subject to large uncertainties (Andersson-Sköld and Simpson, 2001). The introduction of natural dust is under consideration.

Table 3.1: Aerosol prognostic variables used in UNI-AREO: N-number and M-mass concentration.
(Element presently included (X) and will be included (O).)

	N	M SO_4	M NO_3	M NH_4	M EC	M OC*	M Dust**	M Sea salt	Water
Nucleation D< 0.02 μm	X	X				O			
Aitken 0.02 <D< 0.1 μm	X	X	X	X	X	X	O	O	
accumulation 0.1 <D< 2.5 μm	X	X	X	X	X	X	X	O	Diagnostic parameter
coarse 2.5 <D< 10 μm	X	X	X	O			X	X	

*) Only primary OC is currently considered.
**) Only anthropogenic mineral dust is currently considered.

The present UNI-AERO version is an integrated part of the Unified EMEP Eulerian modelling system and therefore uses common advection and diffusion schemes and shares the same meteorology and emissions treatment as in all other Unified model versions. Initial and boundary conditions for gaseous components are the same as in the acid deposition version of the Unified model. For aerosols, initial and boundary concentrations are presently put to zero. An overview of the emission, transformation and removal processes included in UNI-AREO are schematically presented in Figure 3.1.

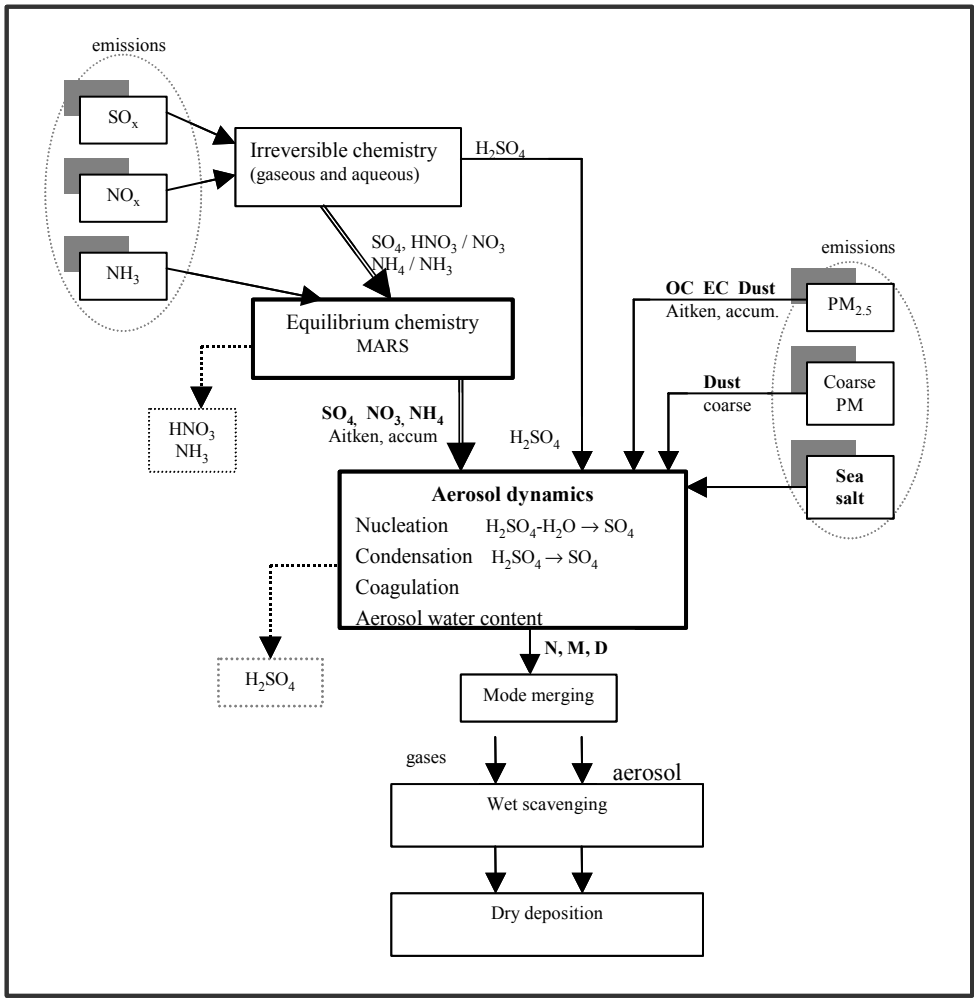

Figure 3.1: Schematic computational structure of the Unified EMEP aerosol model version (UNI-AERO).

UNI-AREO distinguishes primary and secondary aerosols. The present model version considers both emissions of precursor gases and of anthropogenic primary particles. It describes chemical reactions between gaseous components according to EMEP acid deposition chemistry. Equilibrium reactions are parameterised following the Model for Aerosol Reacting System MARS that is also used in the EPA Models-3 Modelling System and in the MADE model developed in Ford Aachen (Ackermann et el., 1998). Aerosol dynamic processes are coupled to the chemistry and parameterised following the MM32 version of MULTINOMO (Tsyro et al., 2002; Pirjola and Kulmala, 2000) except for nucleation processes

that are parameterised according to the rates used in the MADMAcS model from the Institute of Tropospheric Research in Leipzig. Finally, removal by dry and wet deposition is considered to be size dependent process. The parameterisation of dry and wet removal processes is at present very crude and given the relevance of such processes in the transport and distribution of PM we envisage a series of separate studies to test and provide more accurate descriptions.

3.1.1 Emissions

Emissions of primary $PM_{2.5}$ and PM_{10} used in the initial model runs are taken from the 1995 emission inventory compiled by the Netherlands Organisation for Applied Scientific Research (TNO) within the CEPMEIP project (Co-ordinated European programme on Particulate Matter Inventories, Projects and Guidance). Since no information on the chemical speciation or particle size distribution of PM emissions is presently available, assumptions have been made for distribution of the primary PM emissions between the size modes and chemical components.

As detailed in Tsyro (2002), primary $PM_{2.5}$ mass emissions in each source category have been apportioned between organic carbon (OC), elemental carbon (EC) and mineral dust. The carbonaceous OC and EC emissions have been distributed between Aitken and accumulation modes in the same proportion: 15% of the mass has been assigned to the Aitken mode and 85% to the accumulation mode. The mineral dust emissions in the fine size range have all been assigned to the accumulation mode. Primary coarse PM emission has been assumed to consist of dust particles and have been assigned to the coarse mode. From these assumptions, the particle number emissions in the different Aitken, accumulation and coarse modes have been derived from the mass emissions (see Table 3.2)

Table 3.2: Size and chemical speciation of primary PM emission (in %) and component densities (kg/m^3) used in UNI-AERO.

$PM_{2.5}$ Source sectors	**OC**	**EC**	**Mineral dust**
Power generation	33	33	33
Residential and other combustion	50	20	30
Industrial combustion	33	33	33
Industrial processes	0	20	80
Storage and handling	40	0	60
Heavy duty vehicles	40	50	10
Light duty vehicles	40	20	40
Motorised bikes	40	20	40
Passenger cars	40	20	40
Non-road transport	10	60	30
Agriculture	70	0	30
Waste incineration	33	33	33
Size distribution (Aitken/accum)	15 / 85	15 / 85	0 / 100
Coarse PM = PM_{10} - $PM_{2.5}$	-	-	100
Density, ρ (kg/m^3)	2000	2200	2600

Emissions of SO_2, NO_x* and *NH_3, the gaseous precursors of secondary inorganic aerosols (SIA) are 1999 EMEP emissions estimates for the model calculations presented here.

Generation of sea salt aerosol over ocean is driven by the surface wind. A simplified bulk parameterisation for the production of sea salt aerosol is presently implemented in the EMEP Aerosol model version (Jonson et al., 2000). All the sea salt particles are assigned to the coarse mode.

3.1.2 Chemistry

The chemical scheme in UNI-AREO is based on the chemical scheme in the EMEP Unified acid deposition model version (UNI-ACID) with two differences (EMEP Report 1&2/2002). The first difference is that sulphur dioxide homogeneous and heterogeneous oxidation reactions are described separately, thus allowing the explicit description of sulphate (SO_4) formation by nucleation and condensation of gaseous sulphuric acid (H_2SO_4). The second one is that the equilibrium reactions for NO_3 and NH_4 are now performed with the thermodynamic model MARS (Model for an Aerosol Reacting Systems) developed by Saxena et al. (1986) and Binkowski and Shankar (1995).

3.1.3 Aerosol dynamics

The aerosol dynamics module implemented in the Unified Aerosol model version accounts for particles nucleation and growth by coagulation and condensation of vapours. More detailed information on the parameterisation of aerosol dynamic processes can be found in Tsyro (2002).

Nucleation

The empirical parameterisation for the rate of binary homogeneous nucleation of H_2SO_4-H_2O from the MADMAcS model at the Institute of Tropospheric Research, Leipzig (Berndt *et al.* 2000) has been used in the current calculations. Newly formed particles have been assigned to the nucleation model so that homogeneous nucleation in the model increases the nucleation particle number and SO_4 mass in the nucleation mode. Formed by nucleation, these particles coagulate very efficiently and/or can grow by condensation of gases to the Aitken size.

Condensation

Particles grow by condensation of vapours on their surfaces. Condensation of sulphuric acid is explicitly included in UNI-AERO, where the condensation rate of H_2SO_4 on particles is calculated according to Fuchs and Sutugin (1970). The condensation of H_2SO_4 results in an increase of SO_4 mass, while it does not affect particle number. For nitrate and ammonium, equilibrium reactions are parameterised according to the thermodynamic model MARS.

Coagulation

Currently, only coagulation due to particle Brownian motion, which is a dominating process for sub-micron particle, has been accounted for in UNI-AERO following the parameterisation already used in MULTIMONO (MM32

version), in which Brownian coagulation coefficients are calculated according to Fuchs (1964). Coagulation modifies the aerosol size distribution, and the overall effect is a reduction of particle number. This process is particularly efficient for the smallest particles in the nucleation and Aitken modes, which rapidly coagulate with each other and particularly efficiently with larger particles.

Mode merging

The scheme tested in this work is based on the "mode merging by renaming" algorithm by Binkowski (1999). To apply this method, a log-normal particles size distribution has been imposed within each size mode. Constant values for standard deviations of particle distribution have been assigned in each mode based on observation data. The complementary error function has been used to find the fractions of particles greater than and smaller than the limit value. The portions of particle number and particle mass remaining in the mode and the corresponding portions to be transferred to the larger mode have been derived. Then, new number and mass concentrations and diameters have been calculated in each of the modes.

Aerosol water content

Water mass in each mode has been computed at every time step based on the particles chemical composition and relative humidity. In this way, particles containing a fraction of hygroscopic components (SO_4, NO_3, NH_4 and sea salt) are allowed to change their sizes depending on the ambient relative humidity. Particle wet diameters are calculated in each size mode from the total particle mass, including water. Particle wet diameters are then used in calculations of aerosol dynamics and in the dry and wet deposition routines. Consistently, the particle density is recalculated as the mass weighted average of the densities of all aerosol components (including aerosol water) in each mode.

3.1.4 Dry and wet removal

Rather simple parameterisations for calculating dry and wet removal rates depending on the particle size have been adopted in the present version of UNI-AERO (Tsyro, 2002) Different deposition velocities at 1 m height have been assigned to particles in each size mode. For wet deposition, in-cloud scavenging efficiencies are prescribed to particles in each size mode, independently of their chemical composition. Below clouds, size-dependent collection efficiencies of aerosols by raindrops are used (see overview in Table 3.3).

Table 3.3: Particle dry deposition velocity at 1 m (vd_{1m}*), in-cloud scavenging efficiency (*ε*) and sub-cloud collection efficiency (*E*)*

	nucleation mode	Aitken mode	Accumulation mode	coarse mode
Vd_{1M} (cm/s)	1.0	0.5	0.1	1.5
ε	0.0	0.2	0.7	0.7
E	0.4	0.3	0.1	0.4

3.2 Initial tests of the effect of aerosol dynamics on aerosol mass concentrations

Previous calculations of PM mass within EMEP did not include the effect of aerosol dynamic processes, and they only allowed the description of size dependent deposition processes for the primary particles. In EMEP/MSC-W Note 5/2002, initial sensitivity tests to systematically determine the relevance of different dynamic and removal processes in the calculation of PM mass concentrations have been presented. This type of analysis is particularly significant to determine the type of input data required for integrated assessment modelling for the evaluation of health impacts. A short summary of the main results and preliminary conclusions from the sensitivity tests are presented here.

Primary $PM_{2.5}$ and PM_{10}

The first test compares the results from UNI-AERO for the distribution of primary particles with previous results from the EMEP AEROMADE model (Tsyro and Tarrasón, 2001; Tarrasón et al., 2000). AEROMADE did include a size dependent description of dry and wet deposition but only for fine (below 2.5 μm) and coarse particles, and it did not include aerosol dynamic processes. In UNI-AERO, instead, primary PM_{10} has been distributed between Aitken, accumulation and coarse modes, and aerosol dynamic processes are explicitly included. In this case, only anthropogenic primary PM (i.e. organic carbon (OC), elemental carbon (EC) and mineral dust) is present. In this test, no gaseous emissions, boundary conditions or chemistry has been included. Therefore, neither nucleation nor condensation will occur, and coagulation is the only aerosol dynamics process that is active. The comparison of primary particle mass from these two models allows testing the importance of coagulation processes.

The comparison of $PM_{2.5}$ and PM_{10} concentrations derived from these two models shows that accounting for aerosol dynamics, namely coagulation, has no significant effect on the mass of coarse PM. However, $PM_{2.5}$ concentrations calculated with size-resolved UNI-AERO are on average 2-15 % lower than $PM_{2.5}$ concentrations obtained by using the mass model AEROMADE. The reason for the reduction of primary $PM_{2.5}$ concentrations seems to be related to the introduction of additional modes in the size-dependent description of dry and wet deposition of fine particles rather than to the introduction of aerosol processes. In fact, the effect of introducing coagulation of Aitken and accumulation particles has increased $PM_{2.5}$ concentrations only by 1-2%. Therefore, the conclusion is that size-dependent dry and wet deposition of particles is the most important reason for the reduction of primary $PM_{2.5}$ concentrations in the aerosol model.

The sensitivity tests performed by Tsyro (2002) show that the most significant input for the description of the transport of primary $PM_{2.5}$ concentrations is the partitioning of the fine aerosol between the Aitken and accumulation modes and the parameterisation of dry and wet deposition processes for the different size classes. Therefore, appropriate information on the size distribution of primary PM emissions is essential for adequate PM modelling.

The results also focus the need for accurate descriptions of size dependent dry and wet removal processes and further studies in these topics are envisaged for the future.

Secondary Inorganic Aerosol (SIA)

The second series of tests focussing on secondary inorganic aerosols has just been initiated and is expected to continue in order to establish the effect of nucleation and condensation on the calculated mass concentrations of SIA. Preliminary concentrations of secondary inorganic aerosol, sulphate (SO_4), nitrate (NO_3) and ammonium (NH_4), calculated with the EMEP Unified Aerosol model version (UNI-AERO) have been compared to calculations from the Unified Acid Deposition model version (UNI-ACID). Differences in the model formulation of several important processes can be expected to cause differences in the SIA concentrations predicted by UNI-AERO and UNI-ACID. The main differences in the Aerosol model version compared to UNI-ACID are:

1) UNI-AERO accounts for aerosol dynamics,
2) UNI-AERO uses the thermodynamic equilibrium model MARS,
3) UNI-AERO uses size-dependent dry deposition and wet scavenging rates.

The differences in model calculations of SIA concentrations due to the differences in the model formulations should be investigated separately.

The results for SIA from the aerosol and acid deposition model versions are generally in a reasonable agreement (Figure 3.2), though seasonally varying discrepancies have been found for individual components. Generally, UNI-AERO calculates somewhat lower particulate mass concentrations than UNI-ACID for nitrate and ammonium but not for sulphate. For all months, overall concentrations of NO_3 obtained by using UNI-AERO are lower, while concentrations of gaseous HNO_3 are higher compared to those calculated by using UNI-ACID. The MARS equilibrium model used in UNI-AERO appears to distribute less of the total NO_3 to the particle phase and more to the gas phase than UNI-ACID does. This is an interesting result and should be validated against observations. Annual mean NH_4 concentrations are slightly lower than those predicted by UNI-AERO, but there are differences between the model comparison for warm and cold seasons that need to be investigated further. The same can be said for sulphate for which UNI-AERO calculates larger SO_4 concentrations compared to UNI-ACID. Systematic testing of UNI-AERO with respect to all relevant chemical and physical processes in order to explain their individual effects on calculated SIA concentrations has just been initiated, and further analysis is required and should be reported in due time.

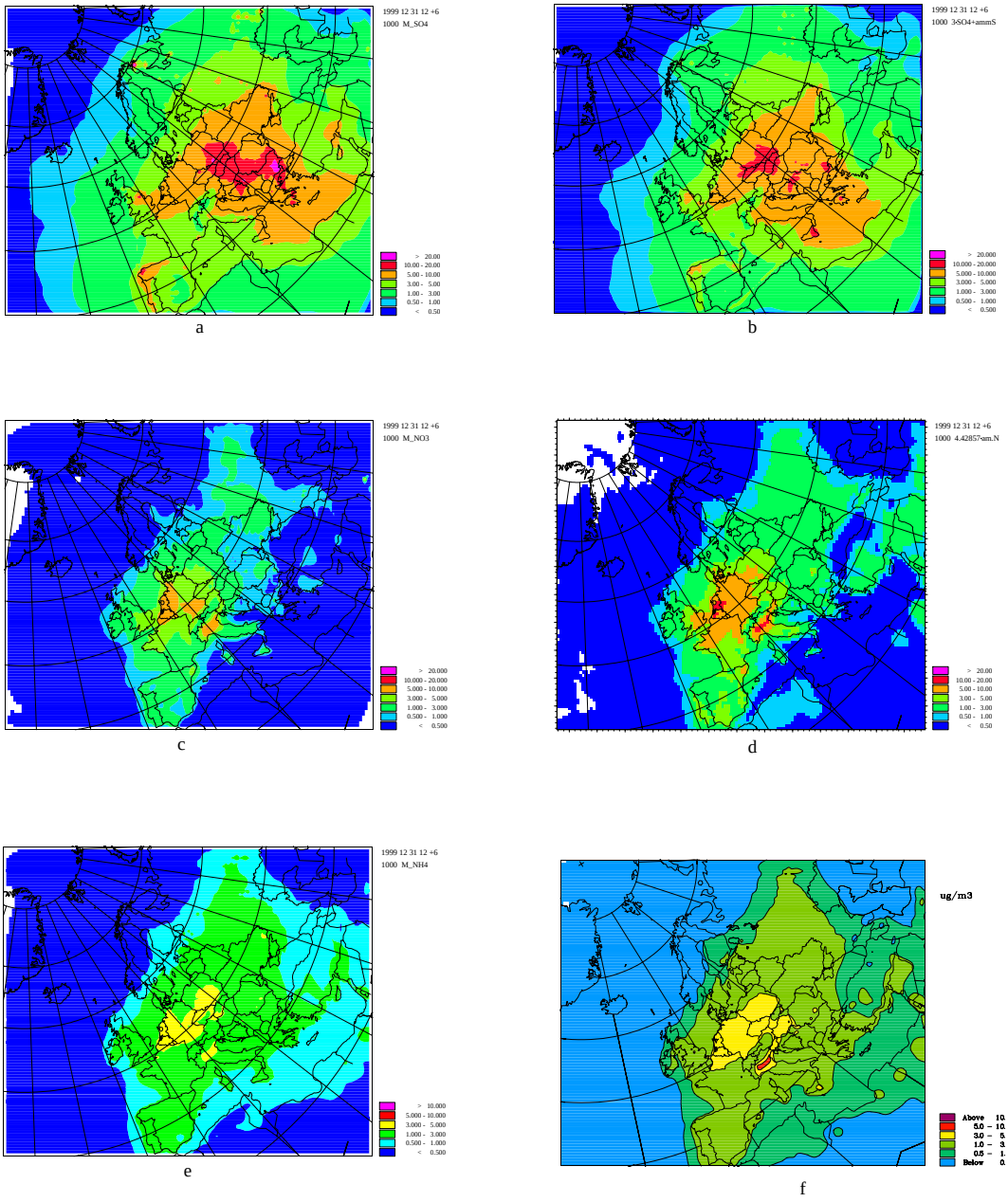

Figure 3.2: Annual mean concentrations of SIA calculated for 1999 by UNI-AERO: a) SO_4, c) NO_3, e) NH_4, and by UNI-ACID: b) SO_4, d) NO_3, f) NH_4.

3.3 Preliminary calculations of the aerosol mass and number with UNI-AERO

The EMEP Unified Aerosol model version provides calculations of aerosol mass, chemical composition, as well as particle number concentration and size distribution. Preliminary results on particle mass and number concentrations, as well as concentrations of individual aerosol components are presented in this section. It is important to keep in mind that all model results shown here should be regarded as interim and subject to possible changes as work on model development and testing progresses.

3.3.1 $PM_{2.5}$ and PM_{10} concentrations

Concentrations of $PM_{2.5}$ and PM_{10} in 1999 calculated with the Unified Aerosol model version are presented in Figure 3.3a and Figure 3.3b.

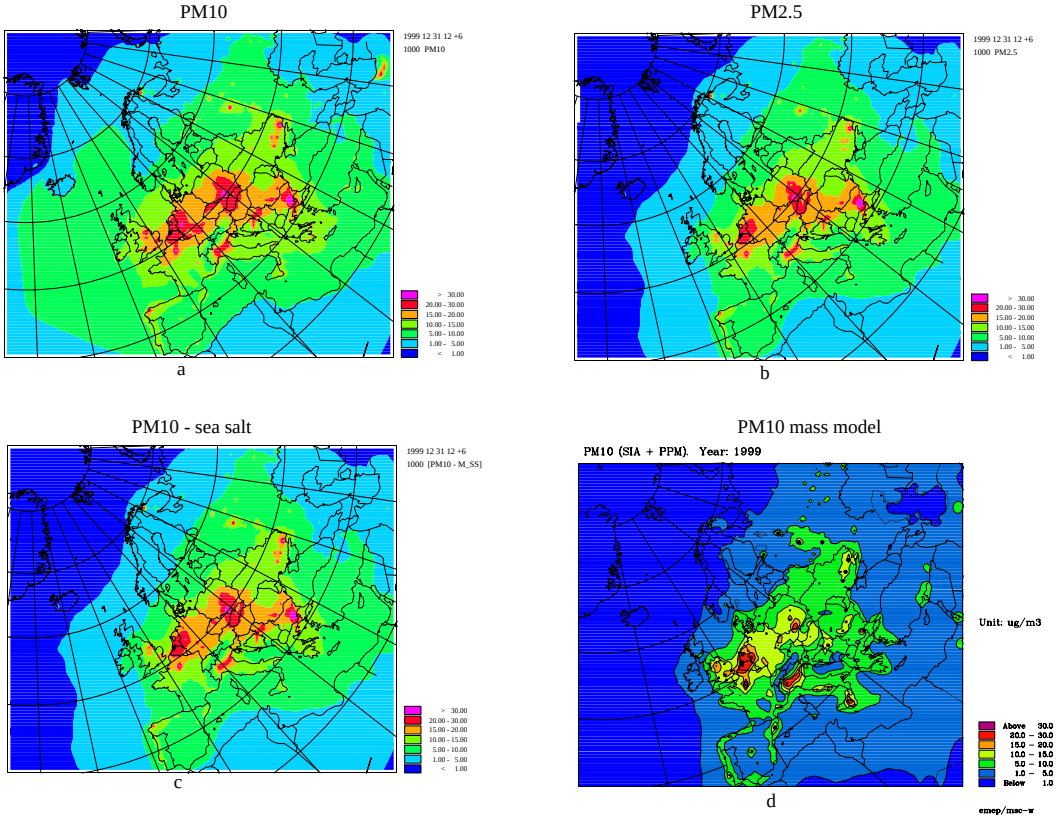

Figure 3.3: Concentrations of PM_{10} and $PM_{2.5}$ calculated for 1999 with UNI-AERO: a) PM_{10}, b) $PM_{2.5}$, c) PM_{10} without sea salt contribution, and with the EMEP Eulerian models: d) PM_{10}.

Figure 3.3 shows that overall PM_{10} concentrations calculated with UNI-AERO are higher than PM_{10} values from the last year calculations (Figure 3.3d). Sea salt was not considered in previous years PM modelling, while it contributes to the total PM_{10} mass in UNI-AERO. To make the old and new results more compatible, sea salt mass has been subtracted from the PM_{10} mass by UNI-AERO. Figure 3.3c reveals that the general level of PM_{10} concentrations from UNI-AERO is higher compared to the earlier calculations with the EMEP Eulerian models.

The differences in PM_{10} concentrations are not only due to implementing the aerosol dynamics and the MARS thermodynamic model in the new Unified EMEP aerosol model. They are also due to modifications and refinements in the common Unified model routines for dry and wet deposition and for turbulent exchange (EMEP Reports 1&2/2002). The modifications in the common Unified EMEP modelling system have resulted in a general increase of the concentrations of SIA (see Chapter 3 and 4 in EMEP Reports 1&2/2002) and also in primary PM. Another important reason for the larger PM_{10} concentrations calculated recently with UNI-AERO is the inclusion of sea salt particles in the aerosol model version. Accounting for sea salt contribution has caused the increase in the total PM_{10} mass, especially in coastal areas. For example, in Southern Norway PM_{10}

modelled concentrations have increased from 1-3 to 5-15 μg/m^3, which compares better with measurements (see section 4).

3.3.2 Total number concentrations

As an example of the new possible output from UNI-AERO, annual mean concentrations of the aerosol total number concentrations calculated with UNI-AERO are presented in Figure 3.4. Large number concentrations are associated with the areas of particle production from emission and/or nucleation. Note that these data still need to be validated against measurement data from research networks.

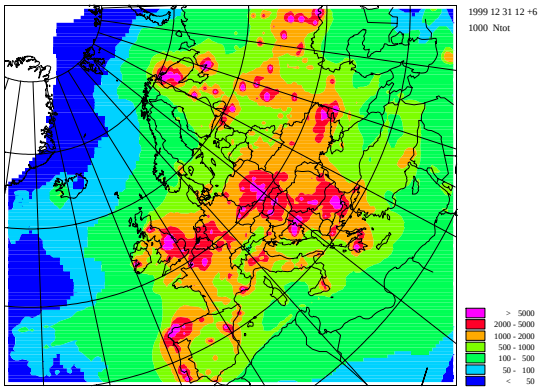

Figure 3.4: Total particle number concentration calculated with UNI-AERO (1999 annual mean).

3.3.3 Mass concentrations of aerosol components (aerosol chemical composition)

Annual mean in 1999 concentrations of individual aerosol chemical components considered in UNI-AERO are presented in Figure 3.5.

As mentioned earlier, concentrations of secondary inorganic aerosol (SO_4, NO_3 and NH_4) are different in UNI-AERO compared to UNI_ACID, and further analysis is necessary to establish the validity of these results. The relative importance of these components to the PM_{10} mass concentrations also needs further validation. Initial results show that SO_4 is the dominating aerosol component, accounting for 30-50% of concentrations in western, central and northern Europe and for 50-70% in Eastern Europe and parts of Russia. NO_3 is the next important components to the ambient aerosol mass. Its contribution to the PM_{10} mass is largest in central Europe (20-30%) and decreases to 5-15% in the Eastern Europe and Russia, and to below 5% in the Northern Europe. NH_4 contributes generally 5-20%.

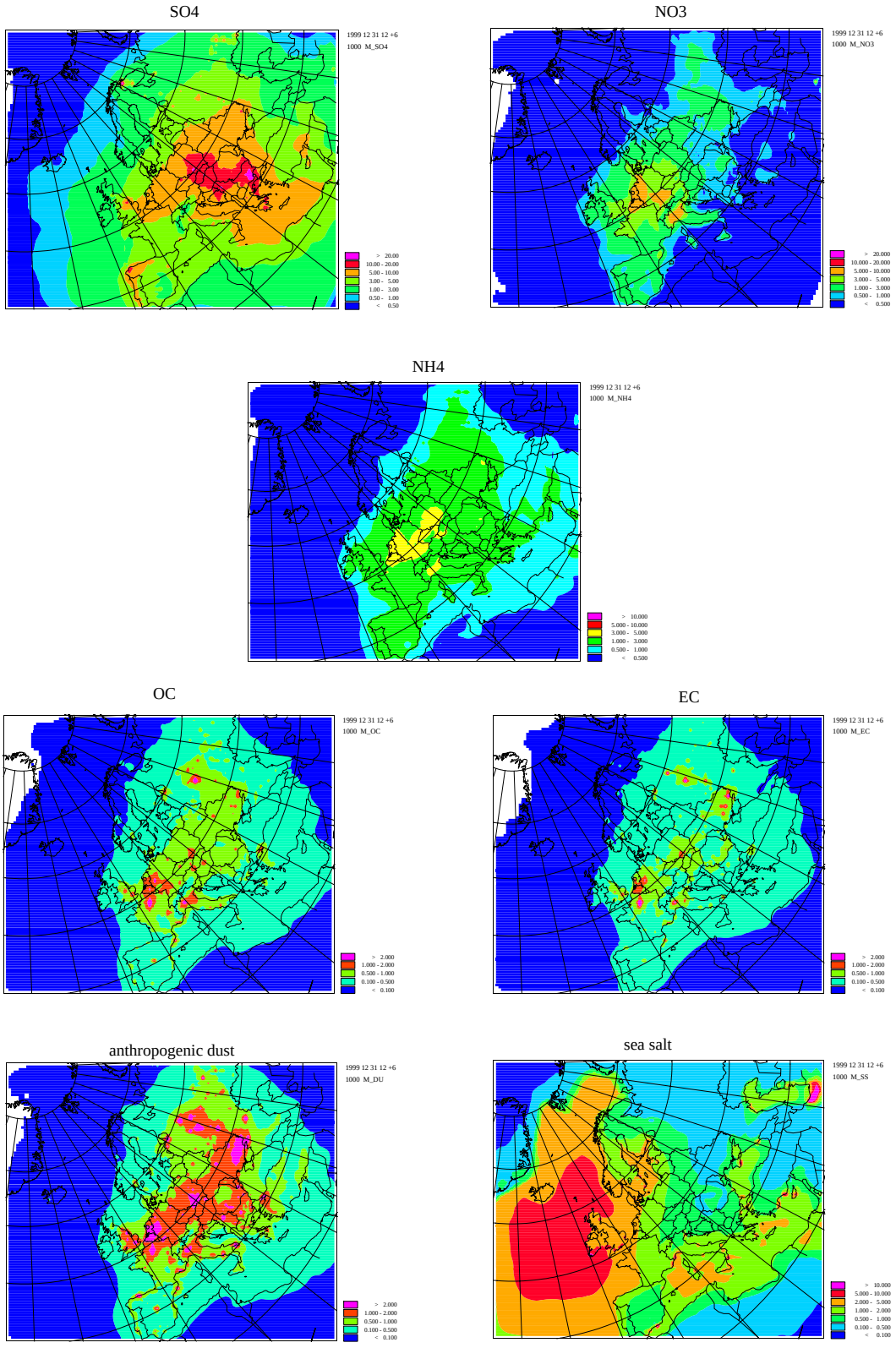

Figure 3.5: Annual mean in 1999 concentrations of aerosol components calculated with UNI-AERO.

The geographical distribution of primary PM components reflects the distribution of their emission sources and the assumptions made on emissions chemical composition and size distribution. Primary particles have been considered to be composed of OC, EC and dust, so that their calculated distribution and relative concentrations are determined by the assumed chemical speciation and particle size distribution of the primary emissions. Differences between the spatial distribution of organic and elemental carbon correspond mostly to the distribution of PM emissions by emission sector. Anthropogenic dust has been allocated in the model to all coarse primary emissions and a considerable part of the fine PM emissions. It is thus the most abundant chemical component of the primary PM. Contributions of carbon particles to PM_{10} mass vary from 3 to 15% for primary OC and from 1 to 7% for EC. Primary anthropogenic dust contributes with 5-10% in most areas. Its contribution is greatest in Ukraine (exceeding 60% in some grid squares), Russia, and Turkey, which is associated with areas of large primary PM emissions combined with lower SIA concentrations.

Finally, the annual mean concentration of sea salt is derived from the wind driven source term and an effective deposition rate as all sea salt aerosols have been assigned to the coarse mode. The effective dry deposition term is the reason why sea salt concentration decreases relatively fast as it is transported from the sea. A more elaborated parameterisation of sea salt generation in different size modes based on the work by Monahan et al. (1986) is being tested in the Unified Aerosol model version.

3.4 Initial comparison of some UNI-AERO results with available measurements

Verification of the EMEP Unified Aerosol model version against available measurements is an essential requirement for progress in model development. Only an initial comparison of some modelled parameters with available measurements is included in this report as the work on validation of UNI-AREO is planned for the autumn of 2002.

Figure 3.6(a) compares model calculated and measured PM_{10} concentrations at EMEP sites in 1999.

PM_{10} concentrations from UNI-AERO compare better with measurements at all Swiss and at most of the German sites than the earlier results with the EMEP Eulerian mass models (EMEP Report 5/2000). For a number of German stations UNI-AERO slightly overestimates $PM_{10.}$ An improved agreement between the calculated and measured PM_{10} mass compared to previous years is seen at Birkenes station. This is mainly due to the introduction of sea salt aerosol in the model. However, there are still significant differences in chemical composition (see also Figure 3.7). A rather coarse parameterisation of sea salt generation used in the calculations for 1999 predicts apparently too high values of sea salt concentrations. A new parameterisation for sea salt aerosol formation has recently been tested and first model results for year 2000 are compared with measurements in Figure 3.6b.

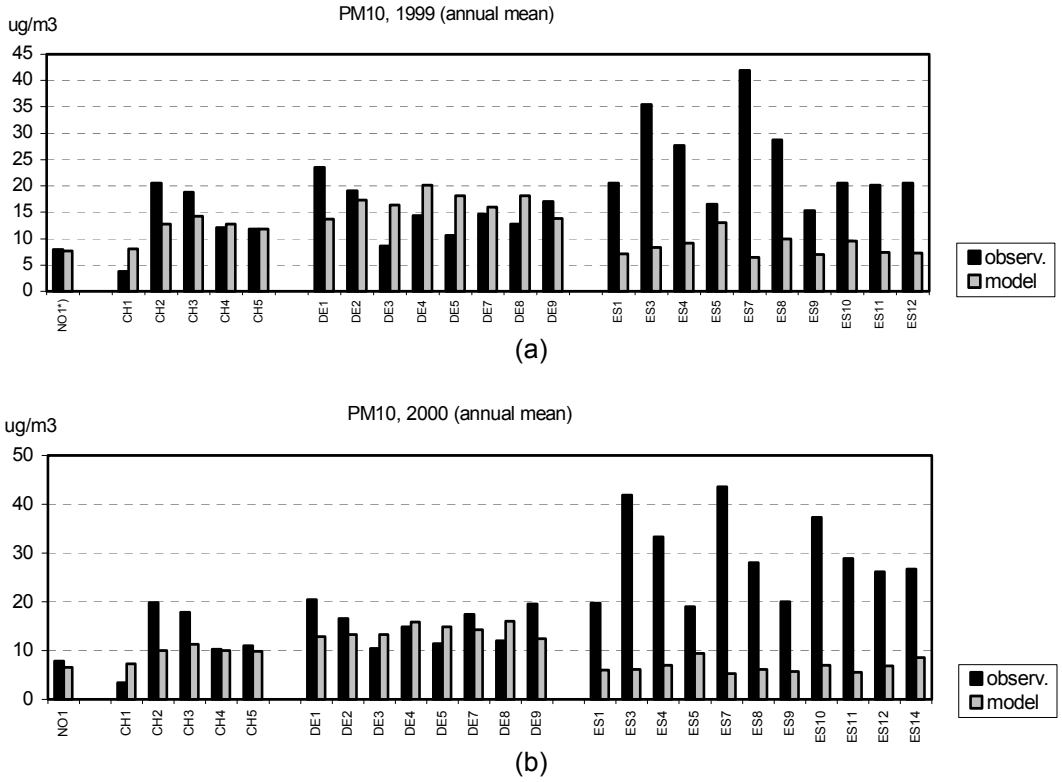

Figure 3.6: Calculated with UNI-AERO and measured at EMEP stations annual mean concentrations of PM_{10} in 1999 ($^{*)}$ in 2000 in Birkenes) and 2000.

A considerable underestimation by the model of PM_{10} concentrations in both 1999 and 2000 at a number of Spanish stations is still seen. In Spain, pollution episodes with very high PM_{10} concentrations (sometimes exceeding 100 $\mu g/m^3$) are experienced due to the transport of Saharan dust. Therefore, in Spain and other areas with a large dust component, we can expect a general underestimation of observation by UNI-AERO since natural mineral dust is not yet included in the model. Accounting for natural dust as well as for bio-aerosols and their re-suspension is believed to be crucial for accurately predicting PM concentrations.

Aerosol chemical composition at two sites predicted by UNI-AERO is presented in Figure 3.7. PM_{10} composition measured at the same sites during different campaigns is also shown. The modelled and measured particle composition cannot be directly compared as the measurements were made in different years and time periods than the model calculations and different chemical components were analysed. Therefore, this comparison is not meant to be a rigorous validation of the UNI-AERO prediction, but only to give some indication on the performance of the model.

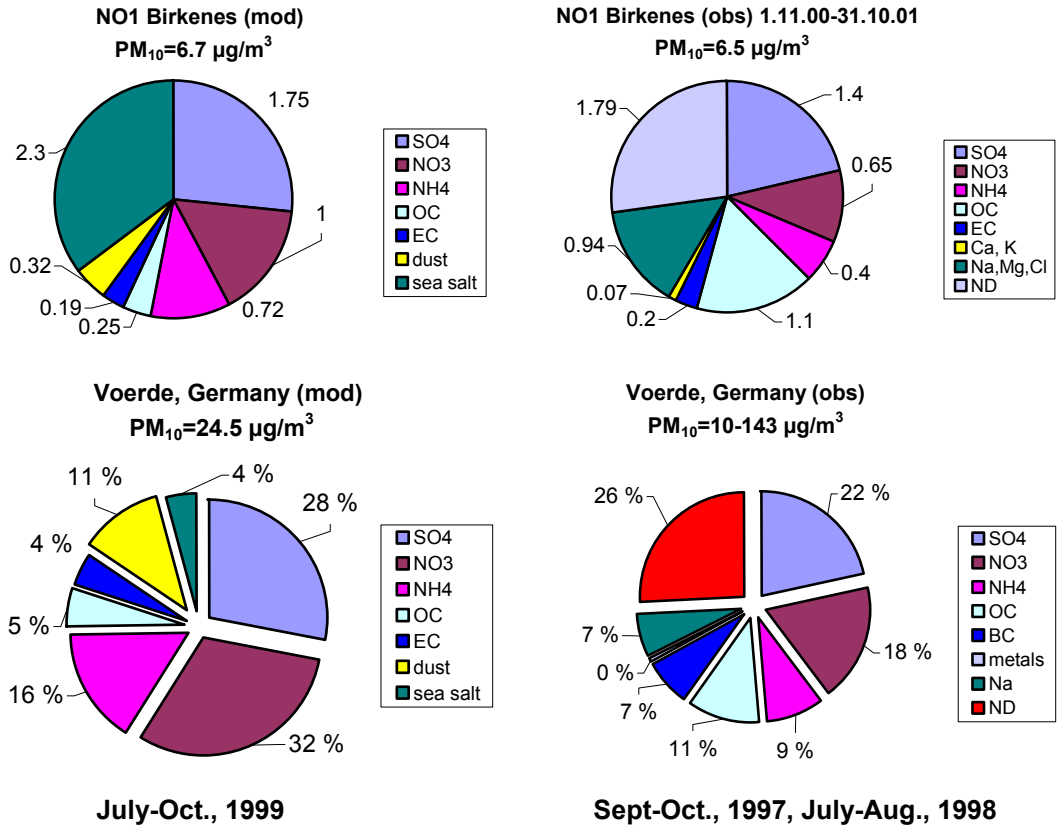

Figure 3.7: Chemical composition of PM_{10} as calculated for 1999 by UNI-AERO and measured in Birkenes, Norway (Tørseth et al., in press) and in Voerde, Germany in 1997-98 (Kuhlbusch et al., 1999).

Largest differences between model results and measurements in Birkenes are found for OC and sea salt. Too low OC concentrations calculated with UNI-AERO are because the model does not include yet secondary organic and primary biogenic aerosols. Model calculations of sea salt aerosol need further verification.

Measurements of the size distribution of PM mass and number are presently being compiled under different programs associated with EMEP. This type of information is extremely important for model verification and development purposes. Much effort will be dedicated to the comparison of aerosol model results with PM mass, chemical composition, aerosol number and size distribution in the next few months in order to secure the further development of the model and establish the soundness of its initial results.

3.5 Outlook

The testing and validation of the EMEP aerosol model results for PM mass, chemical composition, and particle number concentration and size distribution has just been initiated and further progress will be reported in the near future. The validation of model results involves mostly process studies and one of the main priorities is to analyse the chemical and dynamic parameterisation of secondary inorganic aerosols. The analysis will continue to determine the influence of nucleation, condensation and coagulation on particulate mass concentrations.

As particulate mass concentrations is presently the basic input data for health impact assessment, the modelling effort within EMEP will put priority on the calculation of PM mass. This implies that aerosol mass sources presently not included in the model, such as natural dust sources and re-suspension processes have to be accounted for.

The modelling work suffers from missing emissions speciation and monitoring data. A number of parameters have to be covered by making assumptions due to the lack of available information, although the processes determined by these parameters need to be included in the model in order to ensure its physical soundness. There is a strong need for measurement data for verification of the calculated particle mass, number concentrations, chemical composition and size distributions.

3.6 References

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4. Preliminary estimates of changes in statistical life expectancy due to the control of particulate matter air pollution in Europe

by Reinhard Mechler, Markus Amann, Wolfgang Schöpp and Zbigniew Klimont

4.1 Introduction

Over the past decade epidemiological studies in Europe and worldwide have measured increases in mortality and morbidity associated with air pollution. Studies in the United States have shown that those living in less polluted cities live longer than those living in more polluted cities (Dockery et al., 1993; Pope et al., 1995). After adjustments for other factors, an association remained between ambient concentrations of fine particles and shorter life expectancy. These findings were confirmed by a reanalysis of the original studies published by the Health Effects Institute (HEI, 2000) and by a recently published large-scale assessment of mortality based on data collected by the American Cancer Society (Pope et al., 2002).

With accumulating evidence about health effects of air pollution, interest is growing to use data from these studies to inform environmental policies. The World Health Organization (WHO) has produced a guideline document ("*Evaluation and use of epidemiologic evidence for environmental health risk assessment"),* providing a general methodology for the use of epidemiological studies for health impact assessment (WHO, 2000). In 2001, WHO convened a working group to examine several of the aspects introduced in this report as they apply specifically to air pollution health impact assessment (WHO, 2001).

Following these guidelines from WHO, this report presents an initial estimate of implications of present and future exposure of the European population to particulate matter on statistical life expectancy. The methodology integrates population data, findings from epidemiological studies, information about the formation and dispersion of fine particles in the atmosphere and estimates of present and future levels of emissions of fine particles and their precursors. Awaiting further refinements in the scientific disciplines, the quantitative implementation should be considered as preliminary and needs to be revised as soon as more substantiated scientific information becomes available.

This assessment of health impacts of air pollution provides an extension of the Regional Air Pollution Information and Simulation (RAINS) model developed at the International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria. To the extent possible, the methodology relies on calculations and data already implemented in the RAINS model for the assessment of emission control strategies focusing on acidification, eutrophication and ground-level ozone (Schöpp et al., 1999) and on the extension to particulate matter, which is presently under development (Amann et al., 2001).

4.2 The conceptual framework

This study uses the following basic steps to estimate health impacts of air pollution control scenarios:

1. Obtain, for all European countries, information (a) on current mortality rates from UN population statistics and (b) on future baseline mortality rates that are implied by the UN world population projections.
2. Estimate exposure of the European population to particulate matter pollution (a) for 1990, (b) for 2010 assuming implementation of presently decided emission controls, and (c) for the lowest PM levels that could hypothetically be achieved by full application of present-day technical emission controls. This requires (i) spatially explicit information about population densities, and (ii) spatially explicit information of PM levels resulting from the three emission scenarios.
3. Using associations between particulate matter pollution and mortality found by epidemiological studies, determine the modification of mortality rates due to PM pollution.
4. Calculate changes in life expectancy (compared to the baseline UN scenario) resulting from the modified exposures to PM pollution of the three emission scenarios.
5. Examine how sensitive these estimates are to changes in the underlying assumptions.

With this approach, the study combines information about

- results from epidemiological studies that quantify mortality impacts of exposure to air pollution,
- demographic structures in the various European countries and their expected development over time,
- geographically explicit estimates of exposure to air pollution, based on gridded population data and concentration fields of fine particulate matter, distinguishing urban and rural areas,
- the formation and dispersion of aerosols (fine particles) in the atmosphere from primary emissions of fine particles as well as the precursor emissions (sulphur dioxide, nitrogen oxides, ammonia, volatile organic compounds) leading to secondary aerosols,
- the situation estimated for 1990, the predicted conditions in the year 2010 if presently decided emission control strategies were fully implemented and the maximum technically feasible emission controls that could be achieved in the year 2010, taking into account the presently envisaged economic development in the various European countries.

4.2.1 Population data

For all European countries considered in the analysis, demographic information on total population, cohort size and expected deaths was extracted from the recent (medium fertility) world population projections of the United Nations (UN, 2000). These projections provide data in five-years intervals up to 2050. Population data for 2000 and 2010 are listed in Table 4.1.

The probability that an individual will die at a certain age depends both on him/her not dying before that age, and on a probability (or risk) that in adults increases with age. Age-related differences can be described in form of a "life table". For each country, the age-specific baseline, non-accidental mortalities contained in such life tables are calculated from population statistics as the quotient of deaths to population for five-year time-periods for 2000-2050. These calculations were carried out on the national level, using statistics and projections of cohort sizes and death numbers provided by the UN Population Division (2000).

For estimating losses in life expectancy, this study follows all cohorts at least aged 30 years in 2010 over their whole lifetime, i.e., from 2010 to 2075. The mortality rates projected by the UN scenario for 2050 were assumed prevail constant between 2050 and 2075. Younger cohorts are not followed, since they were not addressed in the supporting epidemiological studies. Further analysis is necessary to explore the impacts of including younger cohorts, especially since recent studies indicate a correlation between air pollution and infant mortality (see also Englert, 1999).

For this study, estimates of losses in life expectancy are carried out for all of Europe with a spatial resolution of 50*50 km, corresponding to the grid system defined by the European Monitoring and Evaluation (EMEP) Programme (www.emep.int). While the dispersion model used for this analysis calculates ambient concentrations of PM at this resolution, the present spatial distribution of population in Europe had to be compiled from a variety of sources.

For the EU countries, basic population data on NUTS3 level are taken from the EUROSTAT statistics for the year 1997 (EUROSTAT, 2000). Administrative borders were updated with information from the Austrian Statistical Office (Statistik Österreich, 1999). These population data were spatially allocated to the administrative boundaries provided by the Environmental Systems Research Institute (ESRI, 2000a). For a number of countries (Austria, Belgium, Denmark, Finland, Ireland, Germany, Luxemburg, Netherlands, Portugal, Sweden and the UK) recent information on the NUTS3 boundaries was taken from NUTS databases (Statistik Österreich, 1999). For Poland, information on population distribution was acquired from national sources (http://www.stat.gov.pl/english/index.htm), and data for Russia were taken from the UNEP-GRID database http://www.grid.unep.ch). Information for other countries was extracted from the ESRI demographic database (ESRI, 2000b).

While this information originates from different points in time between 1996 and 1999, for 2010 the assumption was made that, within each country, the spatial distribution of population will remain unchanged.

In this way, for each 50*50 km grid cell total population was derived. The age group distribution as well as the life tables for the population in a grid cell were deduced from the UN national data set.

An attempt was made to distinguish population living in cities and in rural areas. As a first step, population density was calculated for all administrative districts.

All territorial units with a population density of more than 2.5 persons per hectare were classified as cities. For the remaining administrative regions, city population was taken from www.citypopulation.de and the corresponding cities were allocated using their geographical location given by ESRI (ESRI, 2000a). As mentioned above, no changes in urbanization were assumed for the future (i.e., up to 2075).

The resulting spatial distribution of population (including the locations of the cities considered in the analysis) is displayed in Figure 4.1, and the allocation to urban and rural population is listed in Table 4.1.

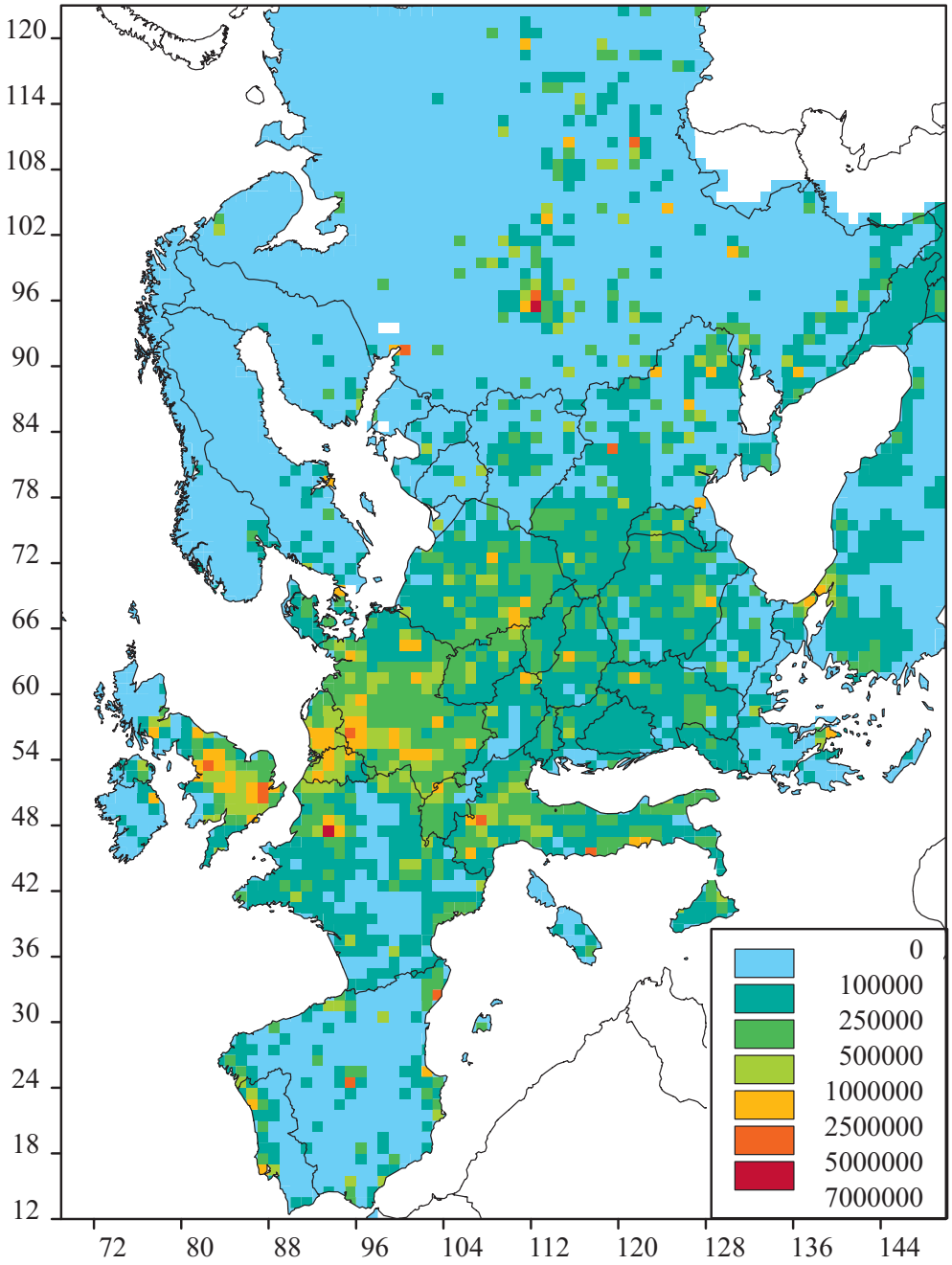

*Figure 4.1: European population in 2000 (persons per 50*50 km grid cell).*

Table 4.1: Rural and urban population in Europe in 2000 and 2010 (1000 persons).

	Total population in 2000			2010	
	Rural	Urban	Total	Total	older than 30 years
Austria	4,341	3,731	8,072	7,953	5,475
Belgium	1,976	8,205	10,181	10,296	6,903
Denmark	3,742	1,543	5,285	5,374	3,548
Finland	2,442	2,699	5,140	5,187	3,426
France	31,774	30,162	61,936	61,203	38,904
Germany	36,100	45,875	81,976	81,353	56,638
Greece	4,956	5,543	10,499	10,579	7,345
Ireland	2,145	1,515	3,660	4,201	2,390
Italy	20,392	37,121	57,512	56,390	40,379
Luxembourg	421	0	421	490	304
Netherlands	2,502	13,109	15,611	16,313	10,757
Portugal	4,308	5,379	9,687	10,082	6,678
Spain	15,024	22,456	37,480	39,569	27,696
Sweden	2,168	6,678	8,846	8,703	5,864
United Kingdom	10,378	48,835	59,213	60,262	38,956
Total	**142,668**	**232,851**	**375,519**	**377,955**	**255,263**
Albania	2,422	826	3,248	3,311	1,650
Belarus	7,088	3,148	10,236	9,819	6,162
Bosnia and Herzegovina	4,299	79	4,377	4,269	2,744
Bulgaria	4,730	3,655	8,385	7,185	4,869
Croatia	2,882	1,902	4,784	4,650	2,968
Czech Republic	6,734	3,582	10,315	10,138	6,870
Estonia	785	677	1,462	1,253	812
Hungary	5,355	4,820	10,175	9,489	6,334
Latvia	954	1,467	2,421	2,288	1,504
Lithuania	1,482	2,214	3,696	3,594	2,301
Norway	2,556	1,837	4,393	4,614	2,948
Poland	24,395	14,265	38,660	38,253	23,919
Republic of Moldova	2,969	1,348	4,318	4,190	2,366
Romania	12,735	9,946	22,681	21,819	13,889
Russian Federation	46,567	70,998	117,565	136,976	87,026
Slovakia	3,059	2,329	5,388	5,430	3,384
Slovenia	1,255	732	1,987	1,955	1,342
Switzerland	3,117	3,964	7,081	7,073	4,906
TFYR Macedonia	924	1,060	1,983	2,072	1,233
Ukraine	28,247	21,999	50,245	45,239	29,222
Yugoslavia	5,831	4,107	9,938	10,404	6,416
Total Non-EU	168,454	154,954	323,407	331,417	212,866
Total Europe	**311,122**	**387,805**	**698,926**	**709,372**	**468,129**

4.2.2 Endpoint: Loss in life expectancy

Exposure to outdoor air pollution is associated with a broad spectrum of acute and chronic health effects ranging from irritant effects to death (American Thoratic Society (ATS), 1996a,b). While all these outcomes are potentially relevant for health impact assessment, this study restricts itself to the quantification of changes in mortality resulting from alternative air pollution control scenarios.

Associations between air pollution exposure and mortality have been assessed through two types of epidemiological studies:

- Time series studies of daily mortality measure the proportional increase in the daily death rate attributable to recent exposure to air pollution.
- Cohort studies follow large populations for years and relate their mortality to their exposure to air pollution over extended periods.

Both designs provide estimates of relative risk of mortality that can be associated with exposure to air pollution. It is important to point out that the relative risks derived from time series and cohort studies have different meanings, but refer to similar effects of air pollution: in both cases, pollution-related mortality reflects a combination of acute and chronic effects (Englert, 1999).

The WHO working group on health impact assessment (WHO, 2001) concluded that both designs could contribute useful, albeit different, information. Through their design, time series studies yield estimates of "premature" deaths due to recent exposure, in all likelihood among those who are frail due to either chronic disease, or to some transient condition. Because such studies cannot quantify chronic effects of long-term exposure, some deaths attributable to air pollution will be missed and the extent to which air pollution advances the time of death cannot be quantified (Kuenzli, 2001; McMichael, 1998). For this reason, the use of risk estimates from time series studies of daily mortality will in most cases underestimate the impact of pollution exposure on both the attributable numbers of deaths and average lifespan in a given population.

Cohort studies can provide the most complete estimates. Such studies include not only those whose deaths were advanced by recent exposure to air pollution, but also those who died from chronic disease cause by long-term exposure.

Review of cohort studies

Due to the complexity of conducting cohort studies, only few analyses are available that examine the relation between long-term exposure to air pollution and mortality. These studies quantify relative risks (RR) of mortality that can be attributed to changes in exposure to air pollution. Table 4.2 summarizes these studies.

Table 4.2: Available cohort studies.

Study	Study object	Relative risk (RR) for all-cause mortality
Abbey et al., 1991 (Seventh-Day Adventists study)	TSP 6303 non-smoking Seventh-Day Adventists in California from 1977-1986 Al-cause mortality	No correlation between TSP and all-cause mortality found
Abbey et al., 1999 Update of Seventh-Day Adventists study	PM_{10} 6338 non-smoking Seventh-Day Adventists in California from 1977-1992 Al-cause mortality	RR=1.12 (1.01-1.24) for 10 µg/m^3 PM_{10}
Dockery et al., 1993 (Six Cities Study)	$PM_{2.5}$ 8000 adults in 6 cities in USA followed up for 14-16 years from 1974-1991, Age: 25-74 at enrolment (max. 90 at end) All-cause mortality	RR=1.13 (1.04-1.24)
Pope et al., 1995 (American Cancer Society, ACS Study)	$PM_{2.5}$ Cohort of >552,138 living in 151 cities in US for 7 years from 1982-1989 Age: 30+ at enrolment Average annual all-cause mortality	RR=1.07 (1.04-1.11)
HEI, 2000 (HEI Re-analysis)	$PM_{2.5}$ Re-analysis of Pope *et al*. (1995) and Dockery *et al*. (1993)	Re-analysis of Dockery *et al.* : RR=1.14 Pope *et al.* (1995): RR=1.07
Pope et al., 2002	$PM_{2.5}$ Analysis of ACS data for 116 cities in the US for 16 years Age: 30+ at enrolment All-cause mortality, cardiopulmonary mortality, lung cancer	For 1979-1983: RR=1.04 (1.01-1.08) For 1999-2000: RR=1.06 (1.02-1.14) For 1979-2000: RR=1.06 (1.02-1.11)

An early attempt was made in 1991 by Abbey et al. to look for relationships between air pollution and mortality using health data of Californian Seventh-Days Adventists communities. At that time, statistical analysis was hampered by the non-availability of measurements of fine particulate matter ($PM_{2.5}$), so that only relations with total suspended particles (TSP) could be examined. No consistent associations between TSP and mortality were found. The study was updated in 1999, following 6,338 subjects from 1977 to 1992 and extending it to PM_{10} (Abbey et al., 1999). After corrections for age, past smoking, education, occupation and body mass index, a positive association between all-cause mortality and the number of days with PM_{10} above 100 µg/m^3 was found for males, but not for females. No associations were found with mean PM_{10}, and with cardiopulmonary and respiratory mortality.

In 1993, Dockery et al. analysed the mortality of 8,000 adults living in six cities in the USA. This "Six Cities Study" followed cohorts of adults aged 25-74 over 14-16 years. The study estimated relative risk (RR) of 1.14 for a 10 µg/m^3

increase in PM_{10}, which corresponds to an 11% change in mortality for each 10 μg/m^3 change in $PM_{2.5}$. The 95 percent confidence interval of RR was determined at 1.04-1.24.

The largest study using data of the American Cancer Society (ACS) examined the linkage between air pollution and mortality for more than 500,000 people aged older than 30 years in the USA over a time period of eight years (Pope et al., 1995). For fine particulate matter ($PM_{2.5}$), a relative risk of 1.07 for all-cause mortality (equivalent to a 6.8 percent change in mortality per 10 μg $PM_{2.5}$/m^3) was found. The 95 percent confidence interval of RR was estimated at 1.04 to 1.11.

In the year 2000, the Health Effects Institute (HEI, 2000) conducted a reanalysis of the original Six City (Dockery et al., 1993) and ACS (Pope et al., 1995) cohort studies. This reanalysis assured the quality of the original data, replicated the original results, and tested those results against alternative risk models and analytic approaches without substantively altering the original findings of an association between indicators of particulate matter air pollution and mortality. In particular, it reconfirmed the relative risks found in the original studies for associations with $PM_{2.5}$. Smaller associations with mortality were shown for PM_{15} and $PM_{15\text{-}2.5}$ (coarse particles).

A recent study (Pope et al., 2002) extended the time span of the ACS study to 16 years and tested possible associations of mortality with a wide range of explanatory variables (age, sex, race, smoking, education, marital status, body mass, alcohol consumption, occupational exposure and diet). It was found that fine particulate ($PM_{2.5}$) and sulphur oxide pollutions were associated with all-cause, lung cancer and cardiopulmonary mortality (Table 4.3). Using the Cox proportional hazard model, the study conducted separate analyses for PM observations of the period (1979-1983) of the first ACS study, for the follow-up period (1999-2000) and for both periods combined.

Table 4.3: Adjusted mortality relative risks (RR) associated with a 10 μg/m^3 change in $PM_{2.5}$ (Source: Pope et al., 2002).

	Adjusted RR (95% confidence interval)		
Cause of mortality	1979-1983	1999-2000	Average
All-cause	1.04 (1.01-1.08)	1.06 (1.02-1.10)	1.06 (1.02-1.11)
Cardiopulmonary	1.06 (1.02-1.10)	1.08 (1.02-1.14)	1.09 (1.03-1.16)
Lung cancer	1.08 (1.01-1.16)	1.13 (1.04-1.22)	1.14 (1.04-1.23)
All other cause	1.01 (0.97-1.05)	1.01 (0.97-1.06)	1.01 (0.95-1.06)

Consistent associations were found between ambient levels of $PM_{2.5}$ and all-cause mortality, cardiopulmonary mortality and lung cancer. For the first period, the relative risks were found to be slightly smaller than those determined in the original study, while the RR resulting from the extension up to the year 2000 match the original estimates. Measures of coarse particle fraction and total suspended particles were not consistently associated with mortality.

Personal exposure versus cohort exposure

It is often suggested that personal exposure of individuals may not be well represented by ambient concentrations of pollutants in urban background air, which are usually monitored on a routine basis. As shown by a number of studies, the relation between personal exposure and background concentration depends on the pollutant under consideration, particularly on its dispersion characteristics and whether significant indoor pollution sources exist (e.g., gas cooking for NO_2). While for individuals such relationships were found to be weak, for larger groups of people ambient background concentrations of $PM_{2.5}$ represents well the characteristic exposure (Boudeta, 2001).

For purposes of health impact assessment WHO (2001) has pointed out that, while it is common to refer to the results of epidemiological studies of air pollution as providing estimates of the exposure-response relation, most epidemiological studies actually measure the relation between ambient concentration and response. Thus a health impact assessment, to be consistent with the original evidentiary studies, must relate to ambient concentrations rather than to actual personal exposure.

The Cox Proportional Hazards Model

For estimating the concentration-response function, the epidemiological studies described above used the Cox proportional hazards model (Cox, 1972). The proportional hazards model postulates that changing the stress variable (here the change in PM concentrations) is equivalent to multiplying the hazard rate (here the mortality rate) by a proportionality factor, which is here the relative risk function. The fatalities due to PM impacts are usually assumed to be Poisson-distributed, thus the concentration-response function is of log-linear type. The Cox proportional hazard model expresses the number of fatalities in a time period as a function of the baseline fatalities and PM concentrations:

$$y = y_0 * e^{\beta * PM} \tag{1}$$

with
- y number fatalities
- y_0 baseline fatalities
- PM PM concentrations
- β functional parameter

With the baseline mortality rate μ_0 defined as the quotient of baseline fatalities y_0 and population size P, the adjusted mortality rate $\overline{\mu}$ is calculated as

$$\overline{\mu} = \frac{y}{P} = \frac{y_0}{P} * e^{\beta * PM} = \mu_0 * e^{\beta * PM} \quad . \tag{2}$$

The factor multiplying the baseline hazard rate is also termed “relative risk” RR, which is determined as

$$RR(PM) = e^{\beta * PM} \quad . \tag{3}$$

Calculating life expectancy from mortality rates

Using the Cox proportional hazards model, a methodology was developed to calculate impacts of various scenarios of precursor emissions of fine particles on the life expectancy of the European population.

The methodology starts from the cohort- and country-specific mortality taken from the life tables and calculates for each cohort the survival function over time. The survival function is modified by exposure to PM pollution using the RR found in epidemiological studies, and can then be converted into reduced life expectancy for an individual person. The mathematical formulation of the approach is provided in Mechler et al. (2002).

Transferability

A health impact assessment applies air pollution effect estimates derived from one population to estimate impacts in another (target) population, based on the assumption that these estimates can be transferred. Care must be taken if one cannot assume that the contribution of various causes of deaths is similar, if the mixture of pollutants differs, if the baseline health statuses of the populations are not the same or if exposure ranges do not overlap.

Currently, only the cohort studies listed in Table 4.2 are available and provide the basis for numerous impact assessments. Since all these cohort studies were conducted in the United States, the generalization of their results to populations in Europe and elsewhere is a concern. Recent studies have begun to explore effect modifiers that may explain the variation in air pollution effect estimates observed among locations in Europe and the United States (HEI, 2000; Katsouyanni et al., 2001). However, results are not yet available and the present knowledge is quite limited, so that it is difficult to include other factors in a practical impact assessment at this point in time.

Extrapolations beyond the range of observational evidence

As pointed out by WHO (2001) caution must be used in extrapolating beyond the range of pollutant concentrations reported in the evidentiary study. The study of Pope et al. (2002) to which this assessment refers, covers annual mean $PM_{2.5}$ concentrations from approximately 5[3] to 33.5 $\mu g/m^3$. The analysis in this paper finds, for the European situation, annual mean $PM_{2.5}$ concentrations from 1 to 60 $\mu g/m^3$ (at a 50*50 km resolution).

Although recent analyses suggest that there is no discernable threshold for the effects of particulate air pollution on longer-term average mortality from cardio-respiratory disease (HEI, 2000), this paper adopts a conservative assumption and does not extrapolate the calculation of health impacts below the lower range reported in Pope et al. (2002) i.e., below 5 $\mu g/m^3$. For this preliminary implementation, it must be kept in mind that the present calculations of $PM_{2.5}$

[3] The exact value of the lower range of $PM_{2.5}$ concentrations was not published in Pope et al. (2002). In Figure 1 (Pope et al., 2002: p. 1136) the lower value for PM2.5 concentrations for 1999-2000 is around 5 $\mu g/m.^3$. The exact value will be used once more data are made available from the Pope study.

concentrations do not include contributions from natural sources and secondary organic aerosols.

Similarly, there are a number of grid cells for which for 1990 $PM_{2.5}$ concentrations are calculated to exceed the upper range of 33.5 $\mu g/m^3$ analysed by Pope et al. (2002). For these situations the assumption is made that the linear response identified for the study domain does also hold, at least up to annual mean concentrations of 60 $\mu g/m^3$.

4.2.3 Estimates of ambient concentrations of fine particles

Fine particulate matter in ambient air is composed of a large variety of particles with different sizes and physical and chemical properties (e.g., Visser et al., 2001). One may distinguish directly emitted primary particles and secondary aerosols that are chemically formed in the atmosphere from several precursor emissions. Primary particles originate from energy combustion, material handling, industrial activities, surface corrosion, and from natural sources (desert dust, sea salt, pollen, organic material, etc.). A certain fraction of secondary aerosols is of inorganic nature (ammonium salts of nitrates and sulphates) and is generated from sulphur dioxide (SO_2), nitrogen oxides (NO_x) and ammonia (NH_3) emissions, while secondary organic aerosols are a product of complex photochemical processes in the atmosphere involving, inter alia, emissions of volatile organic compounds (VOC).

Long-range transported PM

This study estimates, for selected emission scenarios, fields of $PM_{2.5}$ concentrations with the help of atmospheric dispersion models that describe the formation and transport of primary and secondary aerosols over the European continent. The European perspective is necessary because $PM_{2.5}$, which has been associated by the evidentiary studies with adverse health effects, has a mean atmospheric residence time of about 50 hours, during which it can be transported over several hundreds up to 1000 kilometres away from its sources.

For this study the EMEP unified Eulerian modelling system was used, for meteorological conditions of the year 2000, to calculate secondary inorganic aerosol and primary particulate matter. The concentrations of secondary inorganic aerosols, i.e., the sum of particulate sulphate (SO_4), nitrate (NO_3) and ammonium (NH_4), have been calculated with the unified acid deposition model version. The gridded emission fields of SO_2, NO_x and NH_3 were derived from the country total emissions provided by RAINS (Table 4.4) by scaling with the present-day emission distribution stored in the EMEP database. Secondary organic aerosols are not included in the results.

Concentration fields of primary $PM_{2.5}$ were calculated with the unified aerosol model for primary PM. In the present calculations, aerosol dynamics have not been included since results from the EMEP aerosol dynamics model still need more analysis and validation. While using national total emissions of $PM_{2.5}$ as given in Table 4.4, the geographical distribution of emissions within countries is based on quality-controlled and corrected results from the CEPMEIP emission inventory that was developed by TNO.

PM concentrations in urban areas

The EMEP Eulerian European-scale atmospheric dispersion models estimate $PM_{2.5}$ concentrations with a spatial resolution of 50*50 km and are therefore considered representative for rural background concentrations. It is clear from monitoring data that concentrations within cities are usually higher than at regional background sites. Even within cities, certain gradients of $PM_{2.5}$ levels are found, with higher concentrations at curbsides than at typical urban background stations. As the evidentiary epidemiological studies identified relationships between mortality and PM concentrations measured at urban background sites, use of the results of these studies must obviously relate to urban background concentrations and not to PM levels at street level.

In absence of spatially more resolved dispersion calculations covering the European continent, the preliminary assumption was made that in urban background air primary particle concentrations are 25 percent higher than in the surrounding rural background air shed. This should reflect the higher exposure to primary PM emissions from traffic in cities. Due to the longer chemical reaction time of secondary particle formation, no differences in the levels of secondary particles (ammonium sulphates and ammonium nitrates) are assumed between urban and rural sites. Work is underway to derive more accurate estimates of PM concentrations in cities (see http://rea.ei.jrc.it/netshare/thunis/citydelta/).

4.2.4 Emission scenarios

This study explores losses in statistical life expectancy for three emission scenarios:

- the situation in 1990,
- the situation as it can be expected for the year 2010 assuming implementation of the presently decided emission control policies (the 'current legislation' (CLE) scenario), and
- the hypothetical situation in the year 2010, if all technically available emission control measures were fully implemented (the maximum feasible reductions (MFR) scenario).

Emission estimates are taken from the database of the Regional Air Pollution Information and Simulation (RAINS) model developed at the International Institute for Applied Systems Analysis (IIASA). The 2010 CLE scenario follows the economic and energy development as outlined by the "Shared Analysis Project" of the European Union and documented in the European Union Energy Outlook to 2020 (CEC, 1999). On this basis, the CLE scenario assumes implementation of the existing national and international regulations for emission controls, while the MFR scenario explores the full scope of all technical emission control measures contained in the RAINS databases (Cofala and Syri, 1998a,b). For $PM_{2.5}$, emissions estimates were taken from the recent PM databases of the RAINS model. Emission data for each country are listed in Table 4.4.

The preliminary estimates suggest that present policies will bring primary emissions of $PM_{2.5}$ down by 57 percent in the year 2010 compared to 1990. Full

application of today's technology would enable a cut of $PM_{2.5}$ emissions by 86 percent.

For SO_2, the Directive on National Emission Ceilings together with the requirements of the Large Combustion Plant Directive implies an 82 percent reduction of the sulphur emissions in the EU-15 between 1990 and 2010. The full technical potential, though at high costs, would allow for a 91 percent cut. In non-EU countries, SO_2 emissions are projected to decline by 61 percent up to 2010, with a technical potential similar to that in the EU-15.

In the EU-15, emissions of nitrogen oxides will be reduced with present legislation by about 53 percent in 2010, while technically 64 percent would be possible. In non-EU countries, a 34 percent decline is foreseen. For ammonia, the recent international agreements stipulate a 19 percent reduction, while the full technical abatement potential is estimated at about 40 percent.

It needs to be emphasized that these estimates must be seen as preliminary and 'work in progress' since they were not yet discussed with national experts. They are used here as illustrative estimates to test the methodology for calculating losses in life expectancy and need to be refined and validated in the future.

Resulting changes in ambient concentrations of $PM_{2.5}$ are presented in Figure 4.2.

Table 4.4: Emissions of $PM_{2.5}$, SO_2, NO_x and NH_3 (in kilotons).

	$PM_{2.5}$			SO_2			NO_x			NH_3		
Country	1990	2010 CLE	2010 MFR	1990	2010 CLE	2010 MFR	1990	2010 CLE	2010 MFR	1990	2010 CLE	2010 MFR
Austria	38	26	17	93	39	25	192	103	96	77	66	48
Belgium	56	27	11	370	99	57	344	176	144	97	74	57
Denmark	22	13	6	182	26	18	274	127	104	122	69	40
Finland	29	17	10	242	82	68	280	145	83	40	31	23
France	214	126	56	1256	375	170	1867	810	726	810	780	528
Germany	513	119	75	5291	520	386	2717	986	758	757	550	353
Greece	58	42	19	492	188	57	331	326	196	80	73	59
Ireland	15	8	4	178	42	21	113	65	62	127	116	111
Italy	193	100	47	1651	311	153	2037	924	649	462	419	282
Luxembourg	4	2	1	21	3	1	22	10	6	7	7	7
Netherlands	47	28	15	200	50	49	571	260	199	233	128	104
Portugal	31	21	9	342	152	50	303	250	190	77	73	52
Spain	141	104	39	2062	570	181	1166	847	680	352	353	225
Sweden	47	18	9	117	64	48	338	148	136	61	57	44
UK	184	84	44	3812	461	182	2839	1167	777	329	297	218
EU-15	**1593**	**736**	**361**	**16308**	**2982**	**1465**	**13394**	**6344**	**4805**	**3631**	**3093**	**2149**
Albania	10	5	1	72	56	7	24	35	16	32	35	25
Belarus	75	40	10	843	405	51	385	255	107	219	158	103
Bosnia-H	25	16	3	487	416	22	80	57	19	31	23	17
Bulgaria	134	75	15	1842	846	143	355	229	120	141	108	86
Croatia	26	14	3	180	70	18	80	87	36	40	30	22
Czech Republic	171	39	20	1873	246	171	546	238	128	107	101	72
Estonia	40	11	3	261	70	19	83	33	17	29	29	16
Hungary	67	19	7	966	191	21	219	104	58	120	90	73
Latvia	12	4	2	121	37	6	116	38	24	43	35	19
Lithuania	18	7	2	222	61	10	151	80	49	80	81	49
Norway	48	40	31	52	22	17	220	156	125	23	21	17
Poland	271	128	61	3001	1185	403	1217	610	324	505	468	367
Moldova	20	12	2	197	117	19	87	62	25	47	42	29
Romania	208	109	22	1331	594	100	518	349	184	292	210	206
Russia	1547	629	123	5012	2360	562	3486	2718	1095	1282	894	571
Slovakia	52	19	6	548	110	85	219	102	69	60	39	30
Slovenia	14	6	3	200	22	14	60	35	25	23	21	12
Switzerland	14	10	7	42	26	12	155	74	60	72	63	54
Macedonia	14	8	2	107	81	5	39	27	9	17	16	11
Ukraine	638	227	33	3706	1345	380	1888	1222	530	729	592	406
Yugoslavia	63	32	7	585	269	31	211	144	47	90	82	54
Non-EU	**3468**	**1449**	**365**	**21648**	**8530**	**2096**	**10140**	**6655**	**3070**	**3980**	**3138**	**2237**
Total	**10122**	**4369**	**1451**	**75912**	**23025**	**7123**	**47068**	**25997**	**15750**	**15223**	**12461**	**8771**

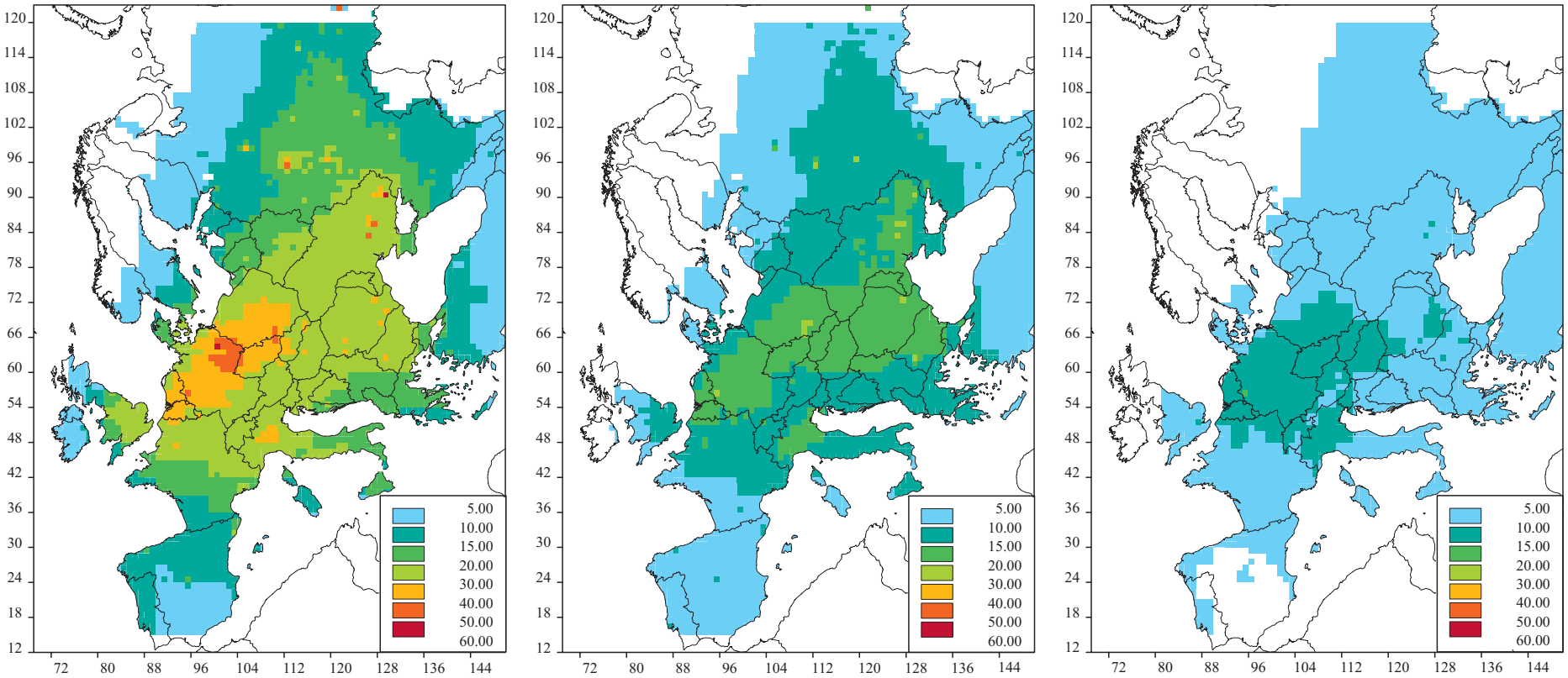

Figure 4.2: Preliminary estimates of rural background concentrations of $PM_{2.5}$ in 1990, for the "current legislation" scenario in 2010 and for the technically feasible emission reductions in the year 2010 (annual mean concentrations, $\mu g/m^3$).

4.3 Results

With the methodology, data and assumptions outlined above losses in life expectancies were calculated for the three emission scenarios. Assuming that 1990 emission levels would be kept constant beyond 2010, the largest life shortening due to anthropogenic $PM_{2.5}$ in ambient air is calculated for the Czech Republic (704 days), Germany (including the new Bundesländer, 664 days), Poland (655 days), Belgium (652 days) and Netherlands (643 days, see Table 4.5). Least impacts are calculated for Ireland (154 days) and the Scandinavian countries (Norway 136 days, Finland 194 days and Sweden 266 days). These preliminary calculations do not include natural sources such as windblown dust, thus they suggest low effects for the Iberian Peninsula (Spain 254 days and Portugal 236 days). These estimates need to be revisited with additional information on the contribution of natural sources, e.g., from the Sahara. On average, life expectancy of Europeans was reduced by 491 days.

By 2010, the implementation of the presently decided emission controls is calculated to improve life expectancy on average by about 214 days, so that PM pollution would cause life shortening of only 277 days on average. Largest improvements are expected for Germany, UK, the Netherlands, the Czech Republic and Belgium, where losses in life expectancy will be cut by more than 50 percent compared to 1990. Full implementation of technically available emission controls would restrict losses to about 184 days. This means that after implementation of the presently decided emission controls there remains scope for improving average life expectancy by more than two months in the EU-15 countries and by 2.5 months in the non-EU countries through further technical emission controls.

Losses in statistical life expectancy on grid level are presented in Figure 4.3 for rural areas. A graphical representation of the country-level results is provided in Figure 4.4.

The results discussed above relate to the statistical life expectancy of all people that are older than 30 years in 2010. These numbers are calculated with the assumption that the pollution level of the selected scenario will remain constant after 2010, so that each cohort will be exposed to this level until the end of its lifetime. This implies that the actual gain in life expectancy will be larger for the 30 years old cohort than, e.g., for the 80 years old cohort, and Table 4.5 lists the average for all cohorts older than 30 years. For comparison, Table 4.6 lists the gains in life expectancy of the cohort aged 30 in 2010.

Table 4.5: Preliminary estimates of reduced average life expectancy due to $PM_{2.5}$ exposure in 1990, for the current legislation case in 2010 and for the maximum technically feasible emission reductions in 2010 (central estimates).

	Country average $PM_{2.5}$ concentrations			(Population weighted country average) reduction in statistical life expectancy		
	1990	CLE	MFR	1990	CLE	MFR
	(annual mean concentrations, $\mu g/m^3$)			(days lost)		
Austria	25.3	14.0	10.4	513	289	210
Belgium	32.2	16.6	12.1	652	334	236
Denmark	18.7	9.7	7.1	362	190	137
Finland	7.5	4.9	3.3	194	132	0
France	20.2	11.5	8.6	432	243	176
Germany	31.3	15.1	11.3	664	314	235
Greece	15.2	11.3	7.7	308	231	152
Ireland	7.7	4.1	3.2	154	110	0
Italy	20.5	12.2	9.1	437	257	186
Luxembourg	29.5	15.5	11.5	568	299	221
Netherlands	32.9	16.6	12.2	643	328	238
Portugal	10.5	7.6	4.9	236	171	108
Spain	11.0	7.5	5.1	254	175	117
Sweden	10.3	5.9	4.3	266	147	113
United Kingdom	14.5	7.2	5.4	431	213	155
EU-15 *)	19.1	10.6	7.8	464	251	185
Albania	17.3	12.2	7.8	331	234	149
Belarus	19.8	12.3	7.5	445	275	164
Bosnia and H.	20.3	13.0	8.3	413	265	168
Bulgaria	24.4	16.1	9.4	528	344	191
Croatia	22.4	13.7	9.1	471	283	188
Czech Republic	34.0	16.1	11.4	704	322	227
Estonia	12.7	8.0	5.1	264	163	111
Hungary	27.3	15.9	10.3	606	342	220
Latvia	14.9	9.4	6.0	328	204	130
Lithuania	18.1	11.0	7.1	416	248	157
Norway	7.1	4.3	3.3	136	102	0
Poland	29.5	15.4	10.2	655	341	222
Rep. of Moldova	25.2	15.9	9.3	557	352	203
Romania	26.1	16.5	9.7	547	347	199
Russian Federation	13.5	9.1	5.8	475	289	156
Slovakia	29.5	16.4	10.7	623	338	222
Slovenia	24.9	14.3	10.1	497	282	200
Switzerland	23.9	13.5	10.3	458	261	199
TFYR Macedonia	18.6	13.0	8.1	371	258	159
Ukraine	24.0	14.6	8.6	593	341	188
Yugoslavia	22.7	14.6	8.8	454	291	174
Non-EU *)	21.0	12.9	8.2	525	308	182
Europe *)	20.1	11.8	8.0	491	277	184

*) weighted average

Table 4.6: Losses in life expectancy due to particulate pollution for the age cohort 30-34 years in 2010 (central estimate, number of days).

	1990	CLE 2010	MFR 2010
Austria	591	333	242
Belgium	742	380	269
Denmark	419	220	158
Finland	210	140	0
France	499	280	203
Germany	753	356	267
Greece	354	266	175
Ireland	167	119	0
Italy	515	302	219
Luxembourg	640	336	249
Netherlands	733	373	271
Portugal	275	200	124
Spain	299	204	135
Sweden	276	159	129
United Kingdom	480	243	176
EU-15	531	289	213
Albania	356	251	160
Belarus	515	318	190
Bosnia and H.	462	297	188
Bulgaria	615	400	222
Croatia	538	323	215
Czech Republic	772	353	249
Estonia	315	195	132
Hungary	690	389	250
Latvia	391	244	156
Lithuania	490	293	185
Norway	216	130	0
Poland	718	374	244
Rep. of Moldova	621	393	226
Romania	632	401	230
Russian Fed.	554	339	183
Slovakia	684	371	244
Slovenia	546	310	219
Switzerland	533	304	232
TFYR Macedonia	406	282	174
Ukraine	706	406	224
Yugoslavia	511	328	196
Non-EU	604	356	209
Average	563	319	211

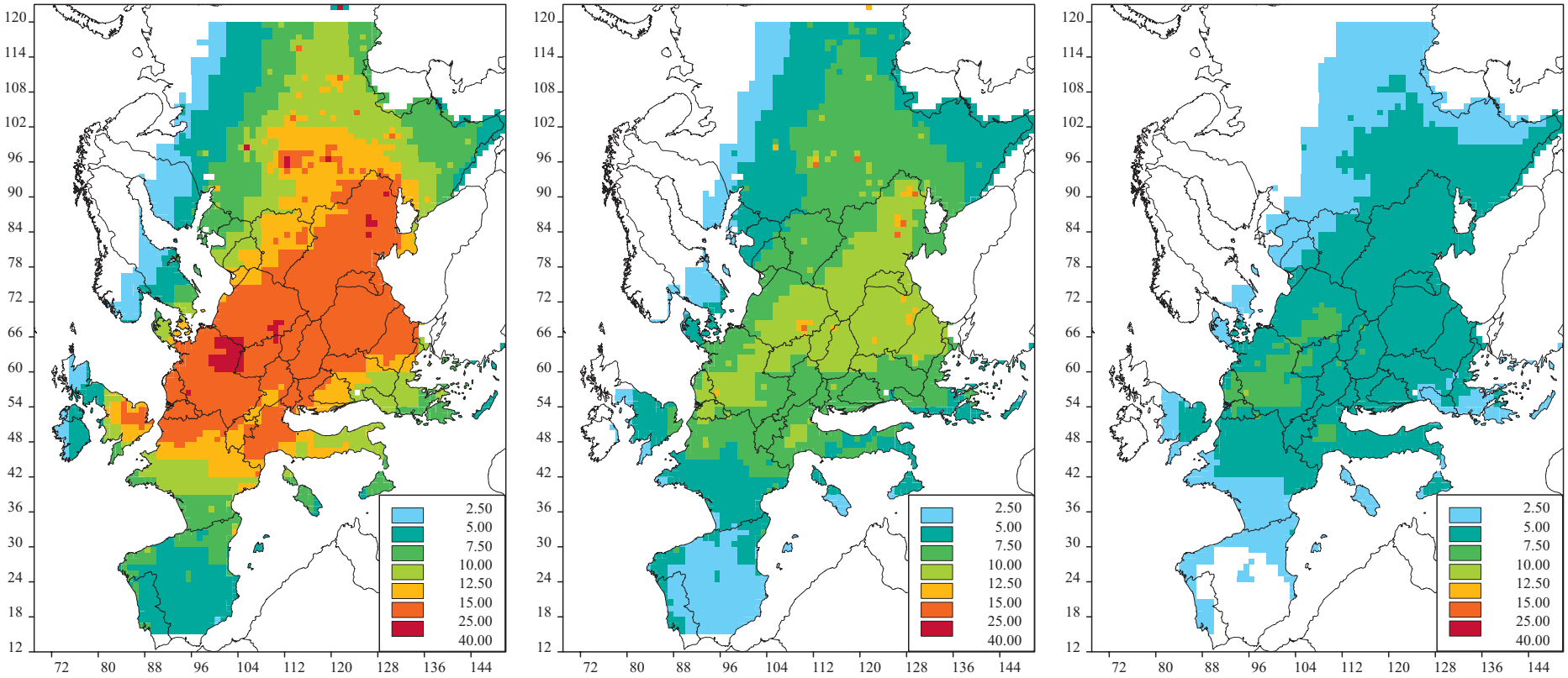

Figure 4.3: Losses in statistical life expectancy in rural areas due to particulate pollution, in 1990 (left panel), for the current legislation scenario in 2010 (central panel) and the maximum technically feasible reductions (right panel), in months

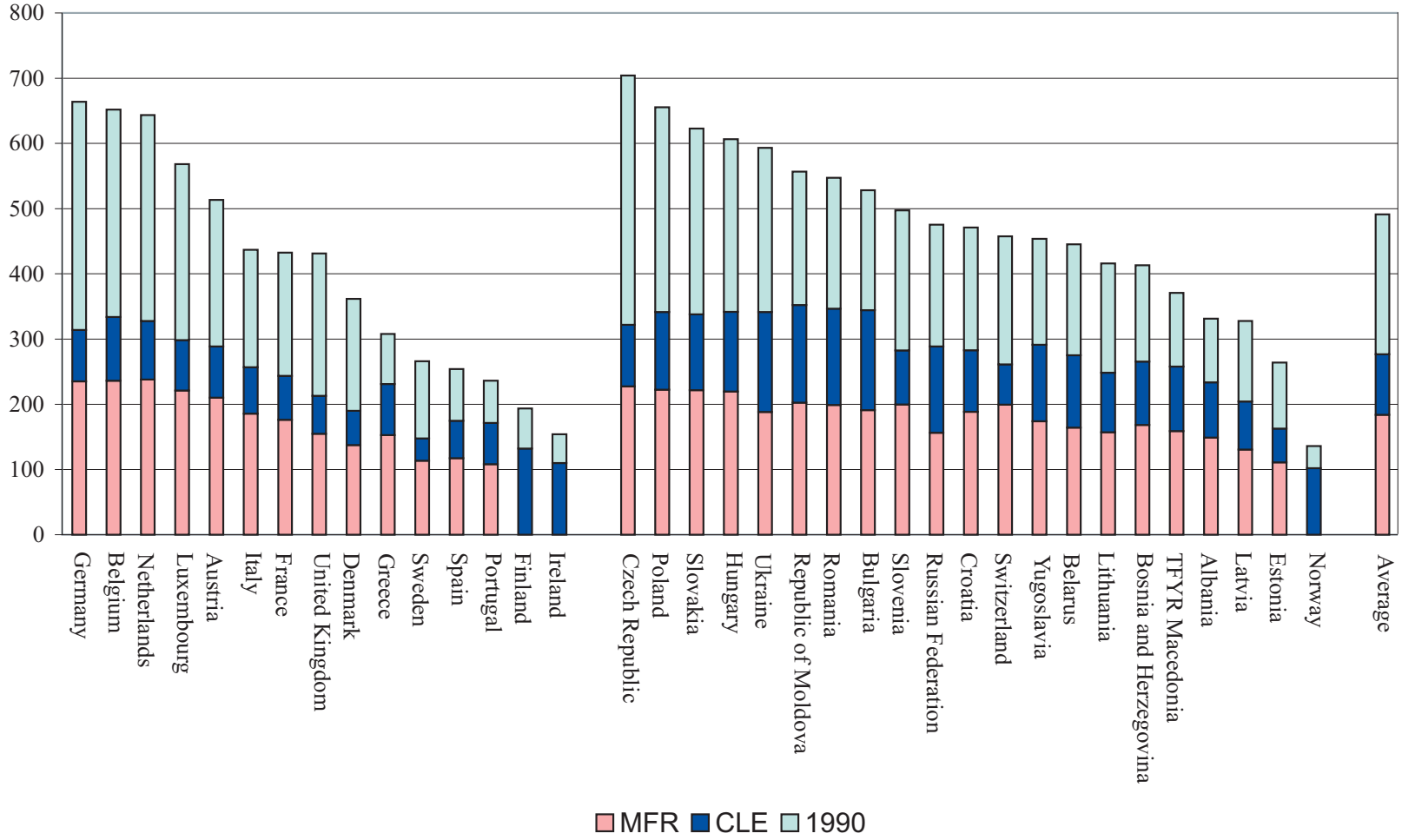

Figure 4.4: Losses in statistical life expectancy for the emission scenarios (in days).

4.4 Uncertainties

It is the objective of this paper to present a methodology for estimating losses in life expectancy due to air pollution at the European scale and to explore the potential order of magnitude of this effect. Many of the data, models and assumptions used for these illustrative calculations are preliminary and need refinement and further validation before robust quantitative conclusions could be drawn. This applies, *inter alia*,

- to the estimates of primary $PM_{2.5}$ emissions in Europe,
- to the projections of emission levels of PM and other pollutants in the year 2010 in Europe,
- to calculations of the formation and atmospheric dispersion of primary and secondary aerosols in Europe,
- to estimates of ambient PM levels in urban air sheds,
- to the use of appropriate dose-response curves derived from epidemiological studies,
- to the question which property of particulate matter is causally linked with mortality.

Each of these aspects is associated with considerable uncertainties. By linking this information, the methodology to estimate losses in life expectancy combines these uncertainties. Suutari et al. (2001) developed a methodology to propagate uncertainties through a similar chain of model calculations aiming at determining ecosystems protection from alternative emission control scenarios. It was shown that, as long as the uncertainties in different elements of the model chain (e.g., the estimates of emissions and of ecosystems sensitivities) are statistically independent from each other, uncertainties do not accumulate, but compensate each other to a large extent.

In principle, this methodology could equally well be applied to the calculation of losses in life expectancy to quantify uncertainties of the overall results, although in practice such an implementation would take considerable time and resources. Instead, a partial sensitivity analysis was conducted using the upper and lower bounds of the 95 percent confidence interval of the relative risk function identified by Pope et al. (2002). Thus the sensitivity analysis explored the losses in life expectancy resulting from relative risks of 1.02 and 1.11, compared to the central estimate of 1.06 per 10 $\mu g/m^3$. Results suggest for 1990 the loss in average life expectancy ranging from 167 to 919 days with 491 days as the central estimate (Table 4.7). For the CLE scenario, the range extends from 94 to 496 days, while the maximum feasible reduction case results in 62 to 329 days.

Table 4.7: Sensitivity analysis: Reduced life expectancy due to $PM_{2.5}$ exposure for the three emission scenarios, for the lower and upper ranges of the 95 percent confidence interval of the relative risk identified in Pope et al., 2002.

	Lower confidence range (RR=1.02)			Upper confidence range (RR=1.11)		
	1990	CLE	MFR	1990	CLE	MFR
	days			days		
Austria	174	98	71	919	517	376
Belgium	221	113	80	1167	598	423
Denmark	123	65	47	648	340	246
Finland	66	45	0	347	236	0
France	147	83	60	774	436	315
Germany	225	107	80	1188	562	421
Greece	105	79	52	551	414	273
Ireland	52	37	0	276	197	0
Italy	148	87	63	783	460	332
Luxembourg	193	101	75	1018	535	396
Netherlands	219	111	81	1152	587	426
Portugal	80	58	37	423	307	194
Spain	86	59	40	455	313	210
Sweden	90	50	39	476	264	203
United Kingdom	146	72	53	772	381	277
EU-15 *)	158	85	63	831	450	332
Albania	113	79	51	593	419	267
Belarus	151	94	56	798	493	294
Bosnia and H.	140	90	57	740	475	301
Bulgaria	179	117	65	946	616	342
Croatia	160	96	64	843	506	337
Czech Republic	239	109	77	1261	577	407
Estonia	90	55	38	473	291	199
Hungary	206	116	75	1086	612	393
Latvia	111	69	44	587	366	234
Lithuania	141	84	53	745	445	281
Norway	46	35	0	243	182	0
Poland	223	116	76	1174	611	398
Rep. of Moldova	189	120	69	997	631	363
Romania	186	118	68	980	621	356
Russian Federation	161	98	53	851	517	280
Slovakia	212	115	75	1115	605	397
Slovenia	169	96	68	890	506	358
Switzerland	156	89	68	820	468	357
TFYR Macedonia	126	88	54	664	462	284
Ukraine	202	116	64	1063	611	337
Yugoslavia	154	99	59	812	522	312
Non-EU *)	178	105	62	940	552	326
Europe *)	167	94	62	879	496	329

*) weighted average

4.5 Discussion and conclusions

4.5.1 Discussion

This paper introduces a methodology to estimate reduced life expectancy due to particulate pollution in Europe. It combines epidemiological evidence about a systematic association between fine particulate matter ($PM_{2.5}$) and increased mortality with inventories and projections of emissions of $PM_{2.5}$ in Europe and calculates the implications of mortality changes due to population exposure to $PM_{2.5}$ concentrations on statistical life expectancy in the various European countries.

The paper also presents a preliminary implementation for Europe based on presently available models and data. This implementation should be considered illustrative, demonstrating that, in principle, all information required for the health impact assessment is available and to explore the order of the magnitude of effects. Many elements of the calculations presented in this paper have to be considered as preliminary placeholders. It is envisaged that within the next few years the work programs of the Convention on Long-range Transboundary Air Pollution and the Clean Air for Europe (CAFE) program of the European Commission will improve many important ingredients of the calculations. Work is progressing to enhance emission inventories of particulate matter, to refine the Eulerian dispersion models of aerosols and further validate them with more monitoring data, to improve estimates of ambient levels of particulate matter in urban air, and to obtain comprehensive advice from WHO about the use of epidemiological evidence for health impact assessment. Such improved knowledge will reduce many uncertainties of the calculations presented in this paper.

However, there are other uncertainties, which are not expected to disappear as a result of the improved information that is expected to become available within the next few years. For instance, there might not be complete certainty about the causal factor in particulate matter leading to increased mortality, and a range of alternative hypotheses might prevail.

There are also a number of methodological uncertainties in the calculations of reduced life expectancy that will most likely not be completely resolved within the next few years. Questions about the transferability of results from the evidentiary studies conducted under conditions in the United States to the European situation, taking into account, e.g., differences between Western European, Eastern European and Mediterranean countries. This also applies to the range of PM pollution that is covered by the US studies. It turns out that considerable areas in Europe have significantly higher PM pollution levels than found in the US studies, and assumptions need to be adopted about the extrapolation of the response curves found in the US to the higher PM levels in Europe.

At the same time, the atmospheric calculations applied in this analysis suggest for areas in Scandinavia PM levels below the lower bound of the US studies (however, these preliminary calculations do not include natural emissions). The assumption of a lower cut-off threshold for PM effects (in order to remain within

the validity of the statistical analysis of the evidentiary studies) has implications about potential benefits of improving air quality even at relatively clean sites. This needs to be discussed in view of the absence of a no-effect threshold concentration for PM as suggested in the literature (e.g., HEI, 2000).

Transferability is also an issue in the context of a potential confounding role of other pollutants present in European air sheds. This study calculates mortality effects exclusively for PM pollution. At the same time, certain effects are also demonstrated in the literature for sulphur dioxide (Pope et al., 2002), ozone, and nitrogen oxides (Katsouyanni et al., 2001). While care must be taken to avoid double counting of effects by adding up responses from multiple pollutants, some studies indicate potentially independent effects, e.g., for SO_2 and ozone.

As explained above, the calculations in this paper are limited to population of age 30 years and older. There is, however, some recent evidence about impacts of particulate pollution on infant mortality (Bobak and Leon, 1999; Woodruff et al., 1997), which could have very strong effects on statistical life expectancy.

4.5.2 Preliminary results

Keeping these imperfections in mind, the preliminary results of this assessment suggest life expectancy in Europe to be significantly shortened by particulate pollution, with the present assumptions between three months in Scandinavia and more than two years in central Europe. The 95 percent confidence interval of these estimates due to uncertainties in the evidentiary epidemiological studies ranges between 1.5 and eight months in Norway and eight months and 3.5 years in Central Europe.

This situation is expected to profoundly change in the future due to the recently agreed emission controls. The Gothenburg Protocol of the Convention on Long-range Transboundary Air Pollution and the Emission Ceilings Directive of the European Union should bring significant reductions of the precursor emissions of secondary aerosols: SO_2 will be cut by 82 percent compared to 1990, NO_x by 53 percent and ammonia by 18 percent. Primary emissions of $PM_{2.5}$ are expected to decline by 57 percent as a consequence of stringent controls for stationary and mobile sources.

These emission controls will reduce the average loss of life expectancy in Europe to somewhat more than seven months in Europe, spreading from one month in Norway to more than 13 months in Bulgaria, Romania and the Ukraine. Full application of all available technical control measures could further reduce these losses on average by another 33 percent.

The results obtained from this study can be compared with other work in the literature. A report of the Committee on the Medical Effects of Air Pollutants has applied a similar methodology to quantify long-term effects of particles on life expectancy (COMEAP, 2000), based on Hurley et al. (2000). This study focused on the UK and did not quantify scenarios of potential improvements in air quality, but analysed gains in life expectancy for a hypothetical 10 $\mu g/m^3$ improvement of PM_{10}. The study found that for the entire population alive in 2000, if the 10 $\mu g/m^3$ improvement were maintained for the rest of their lives, the population would

gain between 4 and 26 million life years, which is equivalent to about 1-6 months per person. For comparison, the analysis presented in this paper suggests for the UK cohorts older than 30 years a range from 2.4 to 13 months with 7.3 months (218 days) as the central estimate for a country-average (not population-weighted) 7.3 $\mu g/m^3$ reduction of $PM_{2.5}$ levels between 1990 and the CLE scenario. Such a 7.3 $\mu g/m^3$ improvement in $PM_{2.5}$ is roughly equivalent to a 10 $\mu g/m^3$ change in PM10 concentrations, so that the numerical results can be directly compared with each other. Obviously, the exact magnitude of the effect depends crucially on the assumption of the relative risk taken by a particular study, which is slightly lower in the UK study conducted in the year 2000 than that of this paper, which relies on the recent 2002 study of Pope et al.

The effect of particulate air pollution on life expectancy in the Netherlands was explored by Brunekreef et al. (1997). This study explores the response in statistical life expectancy for a Dutch cohort of 100,000, using a relative risk of 1.1 for a 10 $\mu g/m^3$ change in $PM_{2.5}$. This results in a gain in life expectancy of 1.11 years. This is consistent with the gain of one year found for the Dutch population in this study, which is based on a relative risk of 1.06 found in Pope et al. (2002).

Nevalainen and Pekannen (1998) conducted a similar analysis for Finland. While the authors rely on the same evidentiary studies available at that time (i.e., Dockery et al., 1993; Pope et al., 1995), they explore the implications on life expectancy for Finland by using Finnish demographic data. They do not, however, estimate ranges of PM pollution that are realistic for Finland, but conduct the analysis for a hypothetical 10 $\mu g/m^3$ change.

4.5.3 Outlook: Loss of life expectancy as an additional endpoint in a multi-pollutant/multi-effect assessment

The RAINS model offers a framework to address multi-pollutant control strategies that simultaneously address several environmental endpoints. For the scenario analyses conducted for the Gothenburg Protocol of the UN/ECE Convention on Long-range Transboundary Air Pollution and the Directive on National Emission Ceilings of the European Union, the model identified cost-effective balances of emission reductions of SO_2, NO_x, VOC and NH_3 in order to reduce acidification, eutrophication and harmful effects of ground-level ozone (Amann and Lutz, 2000). With these emissions as precursors for the formation of secondary aerosols, health impacts of PM pollution could be additionally included in such a multi-pollutant/multi-effect framework (Table 4.8).

Table 4.8: The multi-pollutant/multi-effect framework of the RAINS model.

	SO_2	NO_x	NH_3	VOC	Primary PM
Acidification	√	√	√		
Eutrophication		√	√		
Ground-level ozone		√		√	
Health impacts from fine particles	√ (via secondary aerosols)	√	√	√	√

While the calculations presented in this paper have to be considered as illustrative at the present stage, the availability of the methodology and its integration in the RAINS model opens the possibility to introduce gains in statistical life expectancy as a further environmental endpoint in the multi-pollutant/multi-effect analysis. Thereby, emission control strategies could be explored that balance emission controls over primary and secondary particles (and thereby address health effects from particulate matter), while keeping full account of benefits to and requirements from improvements of acidification, eutrophication and ground-level ozone.

4.6 References

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