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Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe

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CHAPTER 1

Introduction

1.1 Overview

This report presents the progress of activities on acidification, eutrophication and ground level ozone within EMEP during 2003-2004. The Unified EMEP model was reviewed last year under the Task Force on Measurements and Modelling. The review concluded that the Unified model represented a substantial development and enhancement compared to the previous EMEP models and that the model was currently state-of-art (EB.AIR/GE.1/2004/6). For policy applications, the review recommended that additional attention should be given to the calculation of emission data within EMEP and further analysis on the underestimation of particulate matter should be carried out before engaging into the calculation of source-receptor calculations.

The update of Unified EMEP model following the recommendations from the review workshop is documented in Chapter 2. The model updates are mostly relevant to the parametrisation of aerosols and to the calculation of ecosystem impacts as they include a revision of the land-use data sets done in cooperation with the Coordinating Centre for Effects (CCE). The updates concerning particulate matter analyse further the possible contributions to the undetermined PM mass, that is, the part of PM mass that is observed but not explained by modelling approaches. This involves evaluating how much of the un-determined PM mass could be particle-bound water and how important the contribution from organic aerosols could be. The progress in modelling PM is reported in EMEP Status Report 4/2004.

Following the recommendations from the evaluation of EMEP Unified model and in preparation of the revision of the EU National Emission Ceilings Directive, there has been a considerable effort to update and review the emission data used as basis for scenario analysis and impact calculations. In particular, the spatial distribution of the emissions used as input to the Unified EMEP model has been thoroughly revised and a new methodology for allocating emissions by sector has been proposed and tested. The basic national sector distribution has been revised and updated by IIASA through bilateral discussions with the Parties. Scenario runs and source-receptor calculations have used the 2010 and 2020 national projections developed by IIASA under the EU CAFE_BASELINE project (Amann et al. 2004). Status calculations for 2002 and model runs for previous years have used national emission totals as reported by the Parties and revised by MSC-W in co-operation with ETC/ACC (Vestreng et al. 2004). The revision of emission data is documented in Chapter 3 in this status report.

The main focus of this status report is on the calculation of source-receptor relationships. After the review of the EMEP model and the subsequent updates, MSC-W has engaged in a considerable number of scenario runs. The scenario runs are centred around 2010 CLE to provide the basis for the development of integrated assessment models and have been carried out for three different years. The goal is to proceed with two new meteorological years in order to characterise the expected meteorological variability of the source-receptor relationships for use in integrated assessment. The methodology used for the calculation of source-receptor (SR) relationships has been revised with respect to previous calculations and the new methodology is presented and evaluated in Chapter 4. The conclusions from this investigation result in the set of SR tables presented in the Appendix for depositions, air concentrations and relevant indicators.

While the source-receptor relationships have been centred this year in 2010 scenarios to support the preparation of the revision of the National Emission Ceilings Directive and the Gothenburg Protocol, the status calculations on air concentrations and depositions have been updated for 2002. The status in 2002 of transboundary air concentration and depositions is presented in Chapter 5 for acidifying and eutrophying compounds and in Chapter 6 for photo-oxidant pollutants. In addition, country specific reports have been prepared with presentation of the most significant features on transboundary pollution from sulphur, nitrogen, ozone and PM for 2002 (Klein et al. 2004).

Finally, in Chapter 7, updated estimates of the risk for acidification and eutrophication area presented. The risk levels for ecosystem damage derived from the new calculations are considerably higher than those estimated back in 1998. The study in Chapter 7 systematically analyses which factors have determined the largest changes in the calculation of risk damage to ecosystems and concludes that the general increase in the risk calculations is the result of a series of individual improvements and that the increases can not be attributed to a single update of the data or methods used for evaluating ecosystem damage.

All data included in this report will be available at the EMEP web site after its presentation at the 28th session of the EMEP Steering Body. Countries are encouraged to analyse the data and provide their own conclusions. Reactions and comments are both welcome and encouraged.

Descriptions of the EMEP models, and their results, including source-receptor calculations, can be obtained from the EMEP web-site, http://www.emep.int.

1.2 Definitions, statistics used

For sulphur and nitrogen compounds, the basic units used throughout this report are μg (S or N)/m^3 for air concentrations and mg (S or N)/m^2 for depositions.

This report includes also concentrations of particulate matter (PM). The basic units throughout this report are μg/m^3 for PM concentrations and the following acronyms are used for different components to PM:

SIA - are secondary inorganic aerosols and are defined as the sum of sulphate (SO_4), nitrate (NO_3) and ammonium (NH_4). In the Unified EMEP model SIA is calculated as the sum: SIA= SO_4 + NO_3(fine) + NO_3(coarse) + NH_4

PPM - denotes primary particulate matter, originating directly from anthropogenic emissions. It is usually distinguished between fine primary particulate matter, $PPM_{2.5}$, with dry aerosol diameters below 2.5 μm and coarse primary particulate matter, PPM_{co}, with dry aerosol diameters between 2.5μm and 10μm.

PM$_{2.5}$ - denotes fine particulate matter, defined as the integrated mass of aerosol with dry diameter up to 2.5 μm. In the Unified EMEP model $PM_{2.5}$ is calculated as the sum: $PM_{2.5}$ = SO_4 + NO_3(fine) + NH_4 + $PPM_{2.5}$

PMcoarse - denotes coarse particulate matter, defined as the integrated mass of aerosol with dry diameter between 2.5μm and 10μm. In the Unified EMEP model PMcoarse is calculated as the sum: PMcoarse = NO_3(coarse) + PPM_{co}

PM$_{10}$ - denotes particulate matter, defined as the integrated mass of aerosol with dry diameter up to 10 μm. In the Unified EMEP model PM_{10} is calculated as the sum: PM_{10} = SO_4 + NO_3(fine) + NH_4 + $PPM_{2.5}$+NO_3(coarse) + PPM_{co}

For ozone, the basic units used throughout this report are ppb (1 ppb = 1 part per billion by volume) or ppm (1 ppm = 1000 ppb). At 20°C and 1013 mb pressure, 1 ppb ozone is equivalent to 2.00 μg m^{-3}.

A number of statistics have been used to describe the distribution of ozone within each grid square:

Mean of Daily Max. Ozone - First we evaluate the maximum modelled concentration for each day, then we take the 6-monthly mean of these values, over the 6-month period 1 April - 30 September.

SOMO35 - The Sum of Ozone Means Over 35 ppb is the new indicator for health impact assessment recommended by WHO. It is defined as the yearly sum of the daily maximum of 8-hour running average over 35 ppb. For each day the maximum of the running 8-hours average for O_3 is selected and the values over 35 ppb are summed over the whole year.

If we let A_8^d denote the maximum 8-hourly average ozone on day d, during a year with Ny days (N_y = 365 or 366), then SOMO35 can be defined as:

$SOMO35 = \sum_{d=1}^{d=Ny} max\big(A_8^d - 35\ \text{ppb}, 0.0\big)$

where the max function ensures that only A_8^d values exceeding 35 ppb are included. The corresponding unit is ppb.days.

AOT40 - the accumulated amount of ozone over the threshold value of 40 ppb, i.e..

$AOT40 = \int max(O_3 - 40\ \text{ppb}, 0.0)\, dt$

where the max function ensures that only ozone values exceeding 40 ppb are included. The integral is taken over time, namely the relevant growing season for the vegetation concerned. The corresponding unit are ppb.hours (abbreviated to ppb.h). The usage and definitions of AOT40 have changed over the years though, and also differ between UNECE and the EU. Mills (2004) give the latest definitions for UNECE work, and describes carefully how AOT40 values are best estimated for local conditions (using information on real growing seasons for example), and specific types of vegetation. Further, since O_3 concentrations can have strong vertical gradients, it is important to specify the height of the O_3 concentrations used. In previous EMEP work we have made use of modelled O_3 from 1 m or 3 m height, the former being assumed close to the top of the vegetation, and the latter being closer to the height of O_3 observations. In the new Mapping Manual (Mills 2004) there is an increased emphasis on estimating AOT40 using ozone levels at the top of the vegetation canopy.

Although the EMEP model now generates a number of AOT-related outputs, in order to allow great flexibility in later analysis, we will concentrate in this report on four “practical” definitions:

$\mathbf{AOT40^{3m}}$ - AOT40 calculated from O_3 concentrations at 3 m height. This AOT40 is close to that derived from measurements. (Technically, the 3 m is above the displacement height, and so close to the top of a forest canopy, but well above a crop canopy).

$\mathbf{AOT40_f^{3m}}$ - AOT40 calculated as above, but over April-September in analogy with previous calculations of AOT40f for forests.

$\mathbf{AOT40_f^{uc}}$ forests using estimates of O_3 at forest-top (uc: upper-canopy). This AOT40 is that defined for forests by Mills (2004), but using a default growing season of April-September.

$\mathbf{AOT40_c^{uc}}$ calculated for agricultural crops using estimates of O_3 at the top of the crop. This AOT40 is close to that defined for agricultural crops by Mills (2004), but using a default growing season of May-July, and a default crop-height of 1 m.

In all cases only daylight hours are included, and for practical reasons we define daylight for the model outputs as the time when the solar zenith angle is equal to or less than 89°. (The proper UNECE definition uses clear-sky global radiation exceeding 50 W m^{-2} to define daylight, whereas the EU AOT definitions use day hours from 08:00-20:00. Model outputs are also available using the EU definition, but not presented here).

The AOT40 levels reflect interest in long-term ozone exposure which is considered important for vegetation - critical levels of 3 000 ppb.h have been suggested for agricultural crops and natural vegetation, and 5 000 ppb.h for forests (Mills 2004).

1.3 Country Codes

Many tables and graphs in this report make use of codes to denote countries and regions in the EMEP area. Table 1.1 provides an overview of these codes and lists the countries and regions included in the present 2002 source-receptor calculations. NAT and BIC are discussed further in section 4.9.

All Parties to the LRTAP Convention, except four, are included in the calculations. These are: Canada and United States of America, Monaco and Liechtenstein. The first two countries are not included because they lie outside the EMEP area domain. Monaco and Liechtenstein are not included because their emissions and geographical extents are below the accuracy of the source-receptor calculations.

Although Albania is not a Party to the LRTAP Convention, its situation in Europe and the extent of its estimated emissions justify a separate study of this country as emitter and receptor.

Malta is introduced as a receptor country. The estimated emissions from Malta are below the accuracy limits of the source-receptor calculations and do not justify a separate study of Malta as a emitter country.

1.4 Other Publications

This report is complemented with EMEP Status Report 4/2004 (Tørseth, ed.) on Transboundary Particulate Matter in Europe and by country specific reports on the 2002 status of transboundary acidification, eutrophication, ground level ozone and PM (Klein et al. 2004).

A number of other reports and papers of relevance to transboundary air pollution and involving EMEP/MSC-Wand CCC staff have become available in 2003/2004:

Peer-reviewed

- Dillon, P.J., Skjelkvåle, B.L., Somers, K.M. and Tørseth, K. Coherent responses of sulphate concentration in Norwegian lakes : relationships with sulphur deposition and climate indices. *Hydrology and Earth Sys. sci.*, 7 (4), 596-608, 2003
- Hindar, A., Tørseth, K., Henriksen, A. and Orsolini, Y. The significance of the North Atlantic Oscillation (NAO) for sea-salt episodes and acidification-related effects in Norwegian rivers. *Environ. Sci. Technol.*, 38 (1), 26-33, 2004.
- Kahnert, M., M. Lazaridis, S. Tsyro, and K. Tørseth. Requirements for developing a regional monitoring capacity for aerosol in Europe within EMEP. *J. Environ. Monit.*, (6): 646–655, 2004.
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Other

- W. Aas, A.-G. Hjellbrekke, S. Mano, and H. Uggerud. Data quality 2002, Quality Assurance, and Field Comparisons. EMEP/CCC Report 4/2004, The Norwegian Institute for Air Research, Kjeller, Norway, 2004.

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Code	Country/Region
AL	Albania
AM	Armenia
AT	Austria
ATL	Remaining N.E. Atlantic
BA	Bosnia and Hercegovina
BAS	Baltic Sea
BE	Belgium
BG	Bulgaria
BIC	Boundary and Initial conditions
BLS	Black Sea
BY	Belarus
CH	Switzerland
CS	Serbia and Montenegro
CY	Cyprus
CZ	Czech Republic
DE	Germany
DK	Denmark
EE	Estonia
EMC	EMEP Land Areas (all)
ES	Spain
EU	European Community
FI	Finland
FR	France
GB	United Kingdom
GL	Greenland
GE	Georgia
GR	Greece

Code	Country/Region
HR	Croatia
HU	Hungary
IE	Ireland
IS	Iceland
IT	Italy
KZ	Kazakhstan
LT	Lithuania
LU	Luxembourg
LV	Latvia
MD	Republic of Moldova
MED	Mediterranean Sea
MK	The FYR of Macedonia
MT	Malta
NAT	Natural+other emissions
NL	Netherlands
NO	Norway
NOS	North Sea
PL	Poland
PT	Portugal
REM	Remaining Land Areas
RO	Romania
RU	Russian Federation
SE	Sweden
SI	Slovenia
SK	Slovakia
TR	Turkey
UA	Ukraine

Table 1.1: Country/Region codes used in the source-receptor calculations

Russian Federation means the part of the Russian Federation inside the EMEP domain of calculations. The same applies to the Remaining N.E. Atlantic region and natural marine emission area. Remaining Land Areas refer to parts of North Africa and Asia within the model domain (REM=NOA+ASI). For North Africa this concerns parts of Morocco, Algeria, Tunisia, Libya and Egypt. With respect to Asia it includes Syria, Lebanon, Israel, parts of Uzbekistan, Turkmenistan, Iran, Iraq and Jordan. The European Union includes Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Italy, Luxembourg, The Netherlands, Portugal, Spain, Sweden, United Kingdom, Cyprus, Czech Republic, Estonia, Hungary, Latvia, Lithuania, Malta, Poland, Slovakia and Slovenia.

CHAPTER 2

Unified EMEP Model: Updates

Hilde Fagerli, David Simpson and Svetlana Tsyro

2.1 Introduction

The Unified EMEP model which was fully documented in Simpson et al. (2003) had model version number rv1.8. As presented and approved by the in-depth review of the model during the task Force on Measurement and Modelling meeting in Oslo in November 2003, a number of changes have been made to improve the model's performance in some respects. We outline here the main changes made between model versions rv1.8 and the model version used in this report, rv2.0.

Additionally, work has proceeded to improve the "ACID" version of the model, so that it can closely mimic some of the results of the full model at much lower CPU cost. This work is briefly summarised also.

2.2 Night time production of HNO_3

The scheme for night time production of HNO_3 has been revised and several modifications have been introduced. We describe here the new scheme and underline the differences with respect to the old scheme.

The night time production of total nitrate (defined as the sum of HNO_3 in the gas phase and NO_3^- as ammonium nitrate in particulate form) is initiated by the gas phase reaction:

$$NO_2 + O_3 \longrightarrow NO_3 + O_2$$

Then, N_2O_5 is formed in equilibrium with NO_3:

$$NO_3 + NO_2 \rightleftharpoons N_2O_5$$

N_2O_5 may further react with water on deliquescent aerosols, producing two HNO_3 molecules:

$$N_2O_{5\,(g)} + H_2O_{(l)} \longrightarrow 2HNO_{3\,(aq)}$$

HNO_3 formed in the reaction above is assumed to evaporate and will take part in the formation of ammonium nitrate or coarse nitrate.

Since in daylight NO_3 is rapidly photolysed total nitrate is only produced through this path in the absence of sunlight. Thus in winter, with low OH concentrations and many hours of darkness, the above reactions are believed to be the major source of total nitrate in the atmosphere (Dentener and Crutzen 1993).

It can be noted that the rate limiting step for the overall night time production of total nitrate is the initial reaction between NO_2 and O_3. However, at low humidities and/or low aerosol burden, the overall reaction can be limited by the availability of aerosols.

The first modification concerns the calculation of aerosol surface area available for heterogeneous reactions. Previousely, it was assumed similar as in Dentener and Crutzen (1993) that all water-soluble aerosol consisted of ammonium (bi)sulphate. In the old model version, the availability of deliquescent aerosols was assumed to be proportional to the sulphate concentration and the volume occupied by the aerosols (the dimensionless volume fraction, V, units: cm^3 aerosol/ cm^3 air) where the hydrolysis take place was approximated by the volume of sulphate aerosol mass;

$$V = \frac{S \times M_s}{A_0 \rho} \tag{2.1}$$

where S is the concentration of sulphate (molecules cm^{-3}), M_s is the molecular weight of SO_4^{2-}, A_0 is Avogadros number and ρ is the aerosol density (g cm^{-3}). This approach might lead to the underestimation of the reaction surface offered by atmospheric aerosol.

In the modified approach we also consider nitrate and ammonium particles. Thus the aerosol volume is approximated by the volume of ammonium plus (fine-mode) nitrate plus sulphate. We assume a specific aerosol density of 2 g cm^{-3}. This is appropriate for dry aerosols with relative humidities less than approx 75% (Warneck 1988). At higher relative humidity, the salts undergo deliquescence, the water content increases, and the density decreases towards values near 1 g cm $^{-3}$. The particles grow by absorbing water and hence the surface available to heterogeneous reactions increases. To account for the increased surface area, we apply

$$\rho = \begin{cases} 2 & \text{RH} \leq 75 \\ 2 - \frac{RH-40}{60} & \text{RH} > 75 \end{cases} \tag{2.2}$$

where relative humidity, RH; is given in %.

From the volume fraction V, the aerosol surface A can be derived. The second modification concerns the calculation of surface area for monodisperse aerosol. In the old model version we assumed spherical, monodisperse aerosols with radius 0.034 μm. This gave a surface to volume ratio of 88.2, found as $4\pi r^2/(4/3\ \pi r^3) = 3/r$.

In the new model version, we apply the size integrated aerosol surface. Whitby (1978) determined the parameters defining the trimodal lognormal size distribution (see e.g. Seinfeld and Pandis 1998) of sulphate. He also observed that most sulphate was present in accumulation mode. The modal parameters for the accumulation mode for sulphate was determined as 0.078μm for the number mean diameter and a standard deviation of 2. Thus, the aerosol surface can be derived from;

$$A = V \times \frac{\int_0^\infty 3r^2(dN/dr)}{\int_0^\infty r^3(dN/dr)} \tag{2.3}$$

Integration gives

$$A = V \times \frac{3}{r_i} e^{-\frac{5}{2}(ln\sigma_i)^2} \tag{2.4}$$

As an approximation, we apply the modal parameters for accumulation mode for sulphate (number mean diameter 0.078 μm and standard deviation of 2), giving

$$A = V \times \frac{3}{0.034} e^{-\frac{5}{2}(ln2)^2} = V \times 26.54 \tag{2.5}$$

Correcting the pseudo first order reaction coefficient for the effects of gas phase diffusion limitations, as outlined by Schwartz (1986), K_a is given by

$$K_a = A \times \frac{\nu \times \alpha}{4} \tag{2.6}$$

where ν is the mean molecular speed for N_2O_5 and α is the reaction probability.

The third, and last, modification concerns the reaction probability of N_2O_5 with H_2O. The reaction probability has been measured for surfaces of different aqueous solutions. Typical values are of the order of 10^{-2} (Mozurkewich and Calvert 1988, Hu and Abbatt 1997). In the old model version we used α=0.1. Mentel et al. (1999) showed that $\alpha_{N_2O_5}$ is on the order of one magnitude lower for nitrate aerosols compared to sulphate aerosols. To include this effect, we follow the parametrization suggested by Riemer et al. (2003), where the reaction probability is weighted according to the composition of the aerosol:

$$\alpha_{N_2O_5} = f \times \alpha_1 + (1 - f) \times \alpha_2 \tag{2.7}$$

with α_1=0.02, α_2=0.002, and

$$f = \frac{m_{SO_4^{2-}}}{m_{SO_4^{2-}} + m_{NO_3^-}} \tag{2.8}$$

$m_{SO_4^{2-}}$ and $m_{NO_3^-}$ are the aerosol mass concentrations of sulphate and nitrate. This modification gives a much lower night time production of nitrate in air with high nitrate aerosol content.

2.3 Gas/aerosol partitioning

The EMEP aerosol dynamics model (UNI-AERO) used already last year the module EQSAM (Metzger et al. 2002, Metzger 2000) to calculate the partitioning between gas and aerosol phase of HNO_3 and NO_3^- aerosol and NH_3 and NH_4^+ aerosol, respectively (Tsyro et al. 2003). In revision 2.0 this is also standard in the full model and in the ACID version.

2.4 Boundary conditions for ammonium and nitrate

Boundary conditions for ammonium and nitrate have been added in version 2.0, and the boundary conditions for nitric acid has been updated.

As described in Simpson et al. (2003), boundary conditions for a number of species are described with simple functions. These have been designed to enable concentration values that correspond to observations. The concentrations are adjusted in the vertical and for latitude and time of the year (monthly) to match the observed distributions. The functions used to describe the seasonal changes in the ground level boundary conditions and in the vertical were described in Simpson et al. (2003), and in table 2.1 we only give the parameters for the additional species.

Table 2.1: Parameters used to set prescribed boundary conditions

	C_{mean}	d$_{max}$	ΔC	H_z	C^v_{min}	C^h_{min}
	ppb	days	ppb	km	ppb	ppb
HNO_3	0.07	180	0.03	∞	0.025	0.03
fine nitrate	0.07	15	0.03	1.6	0.025	0.02
coarse nitrate	0.07	15	0.0	1.6	0.025	0.02
NH_4^+	0.15	180	0.0	1.6	0.5	0.03

Notes: See text in Simpson et al. (2003) for definition of terms.

2.5 Landuse

The landuse data sets used within the EMEP model were revised in March of 2004, making use of new data provided by the Coordinating Centre for Effects CCE (Jaap Slotweg, pers. comm.). This new data consisted of separate maps of landuse from the CORINE system (de Smet and Hettelingh 2001) and from the Stockholm Environment Institute at York (SEIY) system (`www.york.ac.uk/inst/sei/APS/projects.html`). (Note that the SEI database is updated compared to that referred to by de Smet and Hettelingh (2001)). These two data-sets did in fact form the basis of the landuse maps used in the 2003 EMEP reports, but the new data format provided by CCE enabled a better merge of the two. The basic principal used was to apply CORINE data wherever available, thereafter SEIY data. In addition, the more detailed SEIY data (especially on agriculture) was used to guide the split of the broader CORINE categories into the EMEP land-classes needed by the model.

2.6 Sea salt

Sea salt aerosol has now been included in the Unified model. Two parameterisations for the generation of sea salt spray generation, which were previously tested within the EMEP Aerosol model (Tsyro et al. 2003), have now been implemented in both OZONE and ACID model versions. The model employs the empirical expression from Monahan et al. (1986) to calculate the generation of sea salt aerosols with diameters larger than ca. 1 μm and parameterisation from Mårtensson et al. (2003) for production of sea salt aerosols smaller than 1 μm.

2.7 PM-water

PM2.5 and PM10 mass determined with gravimetric methods is likely to include particle-bound water, which does not get completely removed under filters conditioning at temperature 20°C and relative humidity 50%. The possibility of accounting for particle water in calculated $PM_{2.5}$ and PM_{10} concentrations has now been introduced into the Unified model.

Particle water content in $PM_{2.5}$ and PM_{10} is calculated with the Equilibrium Simplified Aerosol Model (EQSAM) (Metzger et al. 2002, Metzger 2000) for the conditions required for filters conditioning, i.e. temperature 20°C and relative humidity 50 %. The mass of aerosol liquid water content (LWC) is found as the sum:

$$LWC = \sum_{i}^{N} (M_i/m_i) \tag{2.9}$$

where LWC (kg m^{-3}) is the liquid water content of aerosol, N is the total number of single-salt solutions, M_i (mol m^{-3}) is the molar concentration and m_i [mol kg^{-1}] is the molality of salt i.

The soluble aerosol compounds in the Unified model are SO_4^{2-}, NO_3^-, NH_4^+. The calculated aerosol water content depends on the mass of soluble PM fraction and on the type of salt mixture in particles.

Accounting for particle water in calculated $PM_{2.5}$ and PM_{10} has been shown to improve the general correspondence between model results and observations. However, there are caveats to the model esimates of particle-bound water as no verification of the calculated water content is presently available. Further details as well as results and initial evaluation of model calculation of particle water can be found in Tsyro (2004a) and Tsyro (2004b).

2.8 An improved ACID model

Historically the EMEP project has used a hierarchy of models for different purposes. For the work leading up to the UN-ECE ‘Gothenburg’ Protocol and EU NEC Directive, acidification issues were tackled with the 10-reaction ACID model (Hov et al. 1988, Iversen 1990) and ozone tackled with the 140-reaction model of Simpson (1993, 1995).

In contrast, the EMEP model version discussed in the majority of this report, and used in the source-receptor calculations, make use of fully-coupled ozone and acidification chemistry. Over 140 reactions between 71 species are used to model both ozone and acidification chemistry. Although this model version can be presumed to represent the most realistic and thorough description of atmospheric chemistry within the EMEP model system, it is very demanding on computer resources. In order to keep options open in the future we are currently exploring the ability of a simplified model to reproduce the acidification and particulate modelling results of the more complex model. In some circumstances this simplified model may provide results of acceptable accuracy at much lower CPU cost, and thus possibly enable simulations over more years than is possible with the full model.

The changes made to accomplish this simplified model are:

- Procedure added to read in 3-D fields of OH, O_3 and H_2O_2 from prior runs of the full model. Thus, we would typically run the full model for the emissions and meteorology year in question, in order to obtain monthly average 3-D fields of these variables for use as ‘background’ concentrations in the ACID model. This ensures that the chemistry of the ACID model is driven by levels of for example OH and O_3 that are consistent with the OZONE model for the scenario in question. (The older ACID models uses 3-D fields generated by quite independent models or from rather crude extrapolations of measured data. This lead for example to the ACID model having quite different OH concentrations to the full model, leading inevitably to different chemical-turnover times.)

- The dominant oxidation pathway for SO_2 to sulphate is via the aqueous phase reaction with H_2O_2. This reaction is very fast, so that the least abundant of the two (SO_2 and H_2O_2) is depleted within a few minutes after the air enters the cloud. In the old scheme, the H_2O_2 concentrations were kept fixed at a certain average, not taking into account the lowered concentrations in polluted air. In the modified ACID version, we simulate this oxidation limitation by reducing H_2O_2 concentrations according to the expression

$$H_2O_2^* = \frac{H_2O_2 \times H_2O_2}{H_2O_2 + SO_2} \tag{2.10}$$

Thus, with low SO_2 concentrations, H_2O_2 is left virtually unaltered, whilst high SO_2 concentrations reduce the H_2O_2 concentrations substantially.

In summary, the new ACID model is now run in a mode which is far more consistent with the results of the full-chemistry OZONE model than has previously been possible. For example, the results for sulphur components from this ACID version have been compared to the results from the full OZONE version for 1997 and the deviations has been found to be of the order of $\pm$ 5 $\%$.

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CHAPTER 3

Emission distributions used for source-receptor calculations and CAFE scenario analysis

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In preparation of the revision of the National Emission Ceilings Directive, there has been a considerable effort to update and review the emission data used as basis for scenario analysis and impact calculations. In particular, the spatial distribution of the emissions used as input to the Unified EMEP model has been thoroughly revised and a new methodology for allocating emissions by sector has been proposed and tested. This effort responds to the new challenges in environmental policies, where as effects are oriented towards population and the non-compliance areas become discrete, the proximity to sources becomes more important.

This chapter documents the emission data used in source-receptor calculations and scenario calculations carried out by EMEP/MSC-W this year and under the EU CAFE-BASELINE project. The data is characterised by national totals, sector distributions and by the spatial distribution of the emissions. Special attention has been given to the identify changes in the 2004 emission data with respect to emission estimates from previous years.

3.1 National Emission Totals

National emission totals used for 2002 model calculations are based on official submissions of the Parties to UNECE/EMEP. These data have been compiled and verified by national experts and have been revised as documented in Vestreng at al, (2004).

National emission data are presented in Appendix A, Tables A:1 to A:5 for gaseous main pollutants and Tables A:6 and A:7 for primary particle emissions.

The tables contain also scenario projections for 2010 and 2020 as provided by IIASA in April 2004. These estimates, referred to as IIASA_April2004 scenario estimates, are very similar to those used for MSC_W source-receptor calculations (referred to as IIASA_March2004) and are those used for the EURODELTA project. The scenario values differ somewhat from those presented in EMEP Status Report 4/2004 (Amann et al., 2004) which correspond to new updates by IIASA_May2004. This continuous update in scenario estimates reflects the progress of work under the CAFE-BASELINE project and new estimates are expected again by the end of august (IIASA_August2004).

Emission totals over the whole EMEP domain remain almost constant in 2002 with respect to 2001, both for main compounds and particulate matter primary emissions. Changes in emissions are below 1% for all compounds, although the reported changes can be more significant for individual countries and regions. We have distinguished the *European Union* (EU25) from *EMEP Eastern Europe* (EEE[1]) and *Other Areas*, the later group including Iceland, Norway and Switzerland. Europe (*Total*) corresponds to all anthropogenic emissions inside the EMEP domain. For the European Union, we have distinguished EU15 member states previous to the 2004 extension from the new member states that are included under EU10+ in order to highlight significant differences between these countries, when occurring. Figure 3.1 shows the changes in national totals for 2002 with respect to 2001 for these different regions. There is a general decrease in the emissions of all compounds for most regions, except in Eastern European countries, where emissions of most components increase in average with respect to 2001. Changes considered by region are generally below 5%. The only exception is for *Other Areas*. The reason for larger changes in *Other Areas* is that this group includes only 3 countries, so that weighted average values are closer to individual country variations. The reported emission changes for individual countries can be larger than the regional weighted average. However, individual national changes do not normally exceed 20% and are generally below 10%. The only two exceptions are for VOC emissions from Armenia and for PM_{10} emissions from Switzerland, where reported emissions are about $^1/_2$ of those reported in 2001. Emission changes from the later can be easily visualised in Figure 3.1, and they are the reason for the high PM_{10} changes calculated in *Other Areas.*

Figure 3.2 shows the decrease in reported emissions for different regions since 1990, for main compounds. Since 1990, emissions of sulphur dioxide are reported to have decreased by 49% in the EMEP area, the decrease of emissions being larger in EU25 (66%) than in Eastern European countries (42%). However, this decrease of sulphur emissions in EU25 has been achieved to a large extent because of stronger

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[1] EEE, EMEP Eastern Europe, has been defined to include: Albania, Armenia, Azerbaijan, Belarus, Bosnia and Herzegovina, Bulgaria, Croatia, Georgia, Kazakhstan, Republic of Moldova, Romania, Russian Federation, Serbia and Montenegro, The Former Yugoslavian Republic of Macedonia, Turkey and Ukraine.

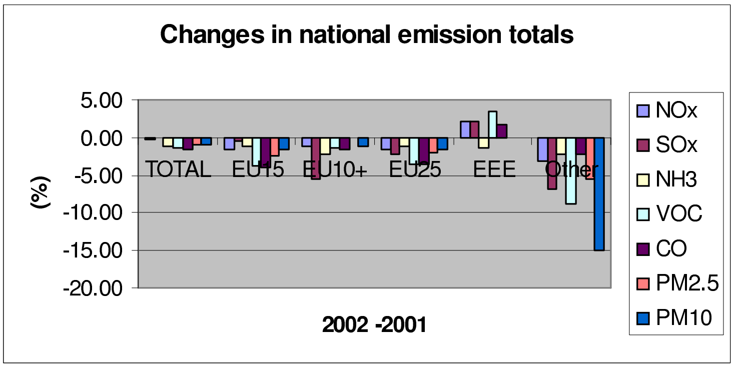

Figure 3.1: Percentage changes in national emissions of main compounds and primary particulate matter for different European regions. Negative values indicate decrease of emissions in 2002 with respect to 2001.

reductions in the new European Union member states (EU10+). For ammonia, the decrease of emissions in Western Europe since 1990 is generally smaller (7% in EU15, 3% for Other Areas) than in Eastern European countries (43% in EU10+, 33%in EEE). The new EU countries have reduced ammonia emissions far more effectively than the EU15 countries, bringing the average reduction for EU25 down by 15.5%.

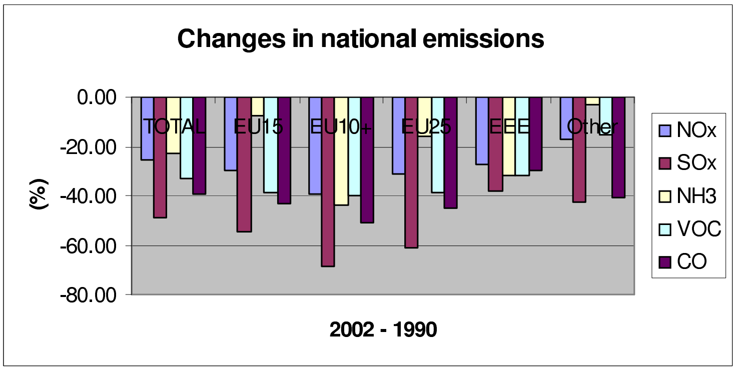

Figure 3.2: Percentage changes in national emissions of main compounds for different European regions. Negative values indicate decrease of emissions in 2002 with respect to 1990.

3.2 Sector distributions

The sector distributions used throughout this report for status calculations, scenario runs and source-receptor calculations with the Unified EMEP model have been revised by IIASA and are based on official submissions by the Parties and bilateral consultations with emission experts carried out under the EU CAFE_BASELINE project. The process is documented in EMEP Status Report 4/2004 (Amann et al., 2004).

The NFR sector disaggregated data reported by the Parties and revised by IIASA has been the basis for the conversion to the levels used as input in the EMEP Unified model. For modelling purposes, the final aggregation levels by sector follow the SNAP code nomenclature at level 1.

Figure 3.3 shows aggregated sector distributions for SO_x, NO_x, NH_3, VOC, $PM_{2.5}$ and PM_{10} anthropogenic emissions. The data is presented for the five different area regions introduced in last section. In addition, emissions from ship traffic in sea areas are also presented as an independent group. All regions are represented with two different emissions columns: the column to the left shows the sector distribution used in 2003 (old) and the column to the right is the 2004 (new) estimate by IIASA. Emission values in Figure 3.3 correspond to year 2000. The picture shows the relative importance of the different regions and sectors to total emissions. It is interesting to note that in 2000, emissions from EU15 dominate the emissions from the European Union for all pollutants. The European Union emissions of NO_x, NH_3, VOC and CO (not shown) are about twice as large as emissions from the rest of countries in EMEP (EMEP Eastern European countries, EEE). For SO_x and PM_{10} emissions, EU25 and EEE contribute similarly to the total EMEP emissions. For SO_x and NO_x, emissions from ship traffic are larger than those from the whole group of countries under EU10+, the new member states of the European Union.

Although differences between the old and new estimates can be significant for single sectors, the emission totals for all components agree within 3%. This justifies the use of the new sector distribution as a percent of total emissions. For model simulations for other years than 2000, we have used the new sector distribution as percent of the actual national emissions for the simulated year.

Figure 3.4 shows the percentage contribution of each sector to the total emissions. Again, the data is presented per component and for the five main regions. Both estimates identify the same main contributor sectors to total emissions of all pollutants, differences affect mainly on the relative importance of main contributor sector. Differences are generally larger for individual countries than for regions and groups of countries, as indicated also in Figure 3.4 where the largest differences between estimates are seen in the smaller group of countries, *Other areas*. Differences are largest for primary PM emissions than for the main gaseous components (also for CO that is not shown). The largest changes are related to ground-based sectors for primary PM emissions: residential combustion and traffic emissions, both in the vicinity of population centres. As the change in the emissions of PM with the new estimate is between source sectors that are co-located and emitted at the same height, we do not expect these differences to affect significantly the Unified EMEP model estimates.

More interesting are the changes that the new distribution introduces in sectors with relatively small contribution to total emissions. The new sector distribution resolves inconsistencies in the reporting from the countries. For example, countries report in

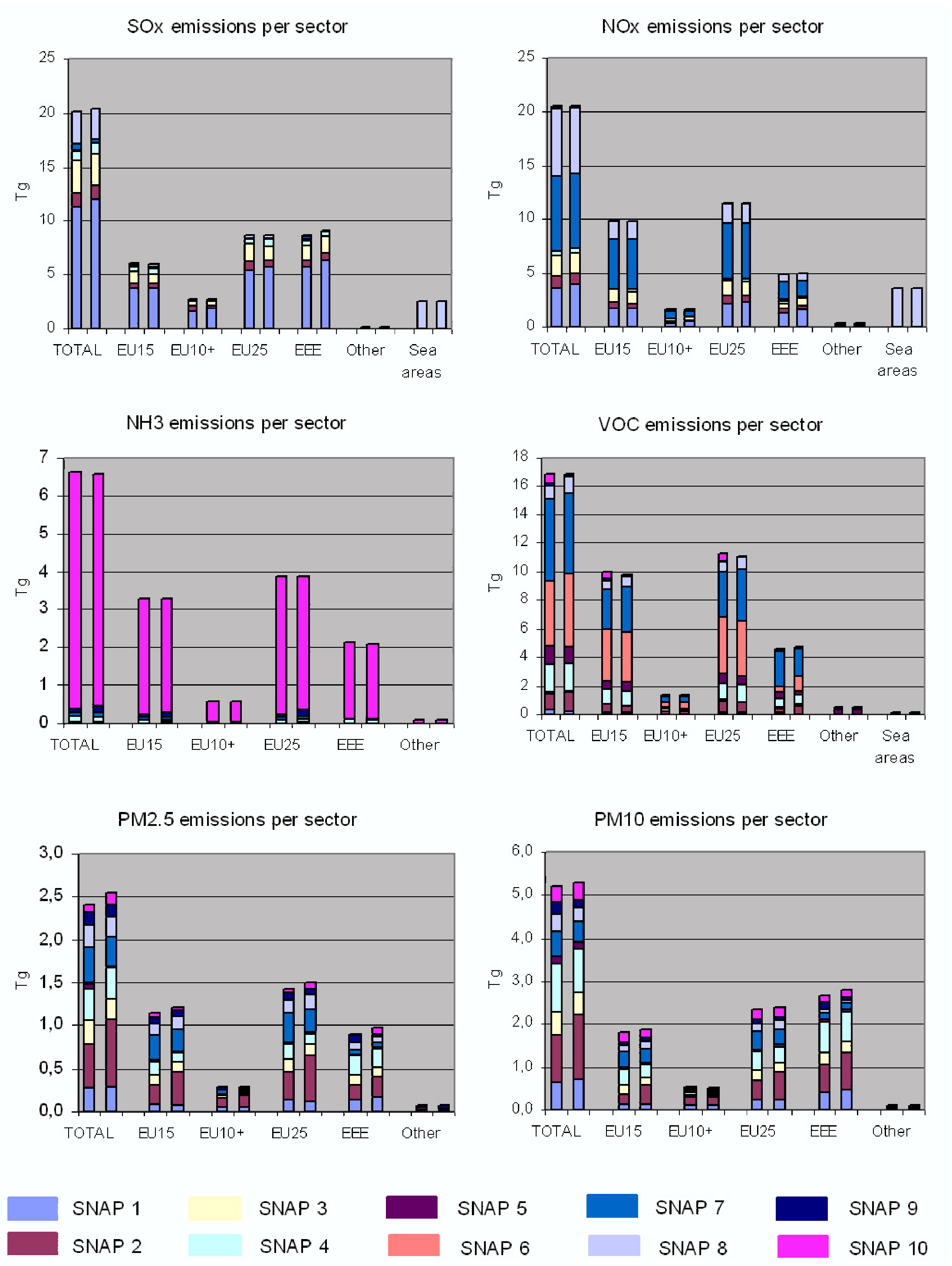

Figure 3.3: Sector emissions from anthropogenic sources aggregated at SNAP level 1 for different components and different regions over Europe (see text for further explanation).

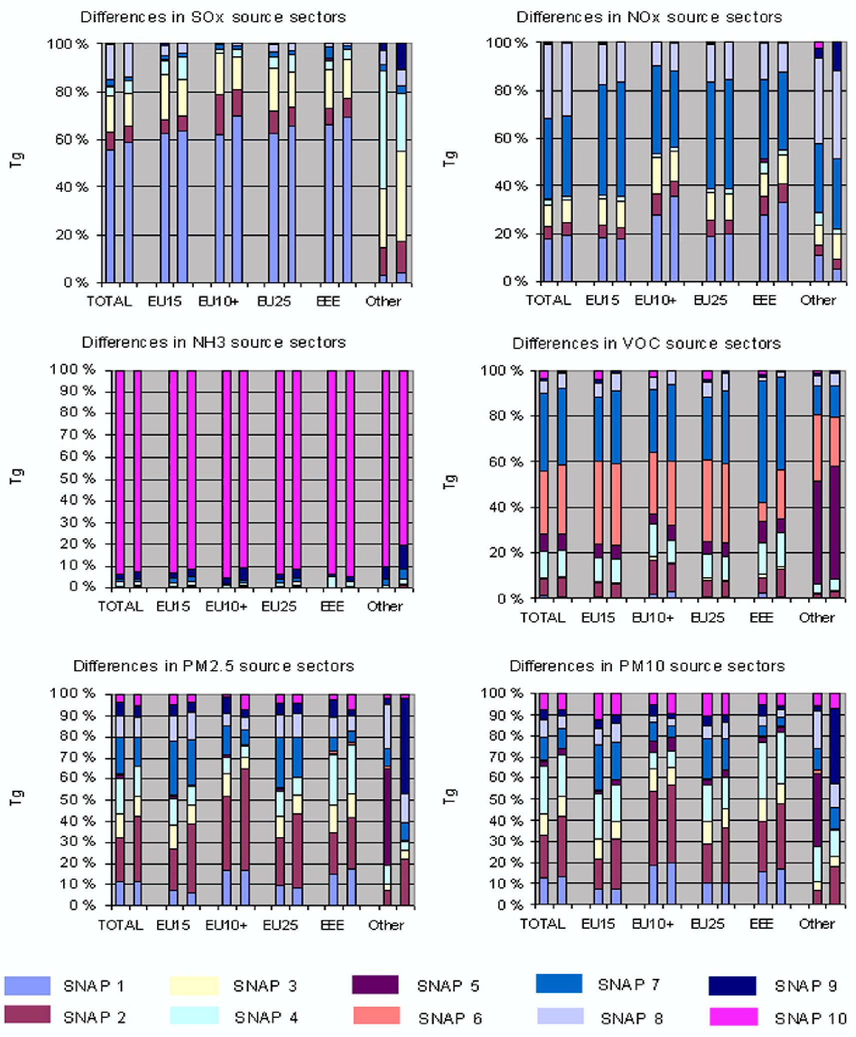

Figure 3.4: Differences in the relative contribution of the anthropogenic source sector to total emissions. In each graph, columns to the left are sector contributions used in 2003 EMEP results, the right columns are corrections made by IIASA under the CAFE BASELINE project and used as basis for 2004 EMEP/MSC-W results.

different sectors emissions from machinery and off-road transport related to main activities like agriculture or fossil fuel extraction. While these emissions of should be reported in sector 8 (off-road transport and machinery), some countries report instead NO_x and CO emissions in sector 10 (agriculture and forestry) and sector 5 (fossil fuel extraction). These are now corrected in the IIASA sector distribution. Another example is related to the reports of VOC emissions in sector 10 (agriculture and forestry).

Some countries include emissions of isoprenes and terpenes from forests as anthropogenic emissions. Other countries consider instead forest emissions as natural emissions and report these under nature (SNAP 11). The inconsistency in reporting VOC emissions has consequences for the model results as it might lead to a duplication of the VOC emissions from forests. This is because the Unified EMEP model calculates its own biogenic VOC emissions (Simpson et al, 1995,1999,2003) in a way that is consistent throughout the whole model domain and depends on the actual meteorological conditions of the simulated year. Duplication problems are now avoided by using IIASA new sector distributions.

3.3 Spatial distribution of national sector emissions

The spatial distribution of emissions is a determining input for atmospheric transport and dispersion calculations. The modelled air concentrations and depositions are intrinsically linked to spatial location of the emissions. As the study of effects from air pollution becomes more oriented towards population and the areas non-compliance with existing international agreements become more discrete, the proximity to sources becomes more important. Thus, an accurate description of the spatial distribution of emissions is even more relevant at present.

The methodology to determine the spatial distribution of emissions used in EMEP/-MSC-W modelling has been revised and updated in the past two years, in cooperation with CONCAWE and the JRC-EI through the CITY DELTA project. The new methodology has now been implemented and tested. In the following we describe the basic principles of the new methodology, present its results and compare them with previous estimates. At the end of this section, some illustrative examples are presented on the significance of the spatial distribution of emissions for air pollution dispersion calculations.

3.3.1 Methodology used for gridding national sector emissions

The main requirements for the new methodology to provide the spatial distribution of the emissions are that the method can be applicable for the whole EMEP domain and that it should guarantee consistency among different compounds. The new methodology follows an aggregated sector approach, so that emissions over Europe from the same sector are distributed according to the same principles. This also guarantees consistency in the spatial distribution of the different pollutants as the same source can emit different compounds.

The sector information has been aggregated following SNAP level 1 because it is at this level of aggregation that most gridded information presently exists. For each sector, we have identified a series of ancillary information that can be used as indicators of the spatial distribution of the emission in the sector. The quality of the ancillary data and their appropriateness as indicators will determine the accuracy of the emission distribution. The new methodology is flexible on its implementation, so that more accurate information can be incorporated as it becomes available to the method. Table 3.1 gives an overview of the ancillary information used for gridding the emissions in each SNAP sector.

Table 3.1: Overview of the ancillary data used to derive the spatial distribution of sector emissions in the new 2004 methodology.

Sector aggregation	**Gridded according to following ancillary data (2004 methodology)**	**Notes**
SNAP 1: Energy Combustion	LPS information for NO_x , SO_x (IER) LPS from countries, when available	Both spatial positions and intensities are presently used
SNAP 2: Residential Combustion	Population (IIASA)	
SNAP 3: Industrial Combustion	50% Population (IIASA) 50% LPS NO_x, SO_x (IER, countries)	Only 4 countries have reported LPS data
SNAP 4: Production Processes	LPS NO_x, SO_x (IER, countries)	Both spatial positions and intensities are presently used
SNAP 5: Extraction Fossil Fuels	GS data for S5 for PM (TNO, CEPMEIP)	
SNAP 6: Solvent and Product Use	Population (IIASA)	
SNAP 7: Road Transport	GS data for S7 for NO_x, if available: or GS data for S7 for PM (TNO, CEPMEIP)	Only 11 countries have reported gridded sector data for NO_x
SNAP 8: Other Mobile Sources	GS data for S8 for NO_x, if available; or GS data for S8 for PM (TNO, CEPMEIP)	Only 11 countries have reported gridded sector data for NO_x
SNAP 9: Waste	XX% Population (IIASA) XX% LPS (IER, countries) XX% Agriculture (S10,TNO, CEPMEIP)	Fractions per country based in CEPMEIP information (see Table 3.3)
SNAP 10: Agriculture & Forestry	GS data in S10 for PM (TNO, CEPMEIP)	

Information on Large Point Source (LPS) locations and emission intensities is used for identifying the position of emissions from sector 1 and sector 4, for all countries. It is also considered that 50% of the emissions from combustion in industry (sector 3) can be allocated to iron, steel and non-ferrous metal industry, thus also distributed according to the information on LPS. The LPS data used at present has been compiled by the University of Stuttgart (IER) and includes both SO_x and NO_x emissions. While LPS SO_x information is used to distribute SO_x emissions, all other gaseous compounds and primary particle emissions are distributed according to information on LPS NO_x. The main difference with respect to the gridding method described last year in Vestreng (2003) is that, in the new methodology, information on the actual intensity of the LPS is used to differentially distribute the sector emissions over a country. When LPS information is reported by the countries, it is checked for consistency with the information from IER and used as ancillary data to distribute emissions from sector 1, 4 and 3. Table 3.2 provides an overview on national LPS data available in 2004. Only four countries have reported data on LPS and not for all components. The accuracy spatial distribution of sectors 1, 4 and 3 is expected to increase as better and more complete information on large point sources is made available. For example, we are aware that the present method introduces a systematic bias in the spatial distribution emissions in sector 4, as it locates production process emissions following energy combustion. We expect that this bias will be reduced when we are in position to use more refined LPS data that distinguishes between sector emissions and includes all different compounds. Other sources of ancillary data that could be used in the future are information from the EPER database and further LPS data from national reports.

Table 3.2: Overview of official information on Large Point Sources (LPS) reported to UNECE/EMEP and available in 2004. (LPS are sources over the following yearly intensities: 500Mg for SO_x, 500Mg for NO_x, 500Mg for CO, 10Mg for NMVOC, 1Mg for NH_3 and 50Mg for PM.)

Country	CO	NH_3	NMVOC	NO_x	SO_x	PM_{10}	$PM_{2.5}$	Total
Czech Republic	1		1	1	1			4
Spain	1	1		1	1			4
TFYR of Macedonia	1			1	1			3
Slovenia	1		1	1	1			4
Total no. countries	4	1	2	4	4	0	0	15

Information on Population (POP) numbers and distribution is a good indicator for emission sources in the surroundings of urban centres. Therefore, population data is used supporting information to distribute emissions from residential sources (sector 2) and emission from solvent and other product use (sector 6). Population is also used to map 50% of sector 3 emissions, as it can be used as a good indicator of the location of electricity and heat production. The population data presently used has been provided by IIASA and therefore it has the additional advantage to be consistent with the data used in the evaluation of impact from air pollution in human health.

Information from the CEPMEIP project (TNO) for PM emissions has been tested for different sectors and found to be a good ancillary indicator for emissions from road traffic (sector 7), other mobile sources (sector 8) and agricultural sources (sector 10) when no other information is available. TNO has used mostly population and to a limited extent also roadmap information to establish the spatial distribution of sectors 7 and 8. Emissions from agriculture are distributed based on farm activity data at high spatial resolution and land use information. Since the information compiled by TNO for the CEPMEIP project (CEPMEIP, 2002) focuses on particulate matter, the activity data is mostly related for poultry farms. The extrapolation of these emission distribution data to other pollutants, introduces a bias for ammonia emissions that we know are mostly related to dairy and pig farms. We hope to be able to correct for this bias in follow-up versions of the gridding methodology, for instance by using FAO statistics and land-use information.

Information on national gridded sector emissions (GS) should be reported every five years to the CLRTAP. However, very few countries report gridded sector emissions. In 2004, only 12 countries have reported gridded sector data and not for all pollutants. When countries have reported GS data, we have compared the official reports with the results of the methodology explained above. The result of the comparison has generally been reassuring for both cases. However, it showed that the national GS data for road traffic emissions and other mobile source generally reproduced better the roadmap network in the country than TNO S7 and S8 data. For this reason, when countries have reported GS data for NO_x (11 countries), that data is used instead of TNO data to consistently derive sector 7 and sector 8 emission distributions for all other pollutants.

The method to map emissions from waste treatment and disposal is necessarily more complex than for other sectors since the activities in sector 9 can include both waste incineration in urban areas and open burning of agricultural waste. In three countries, Azerbaijan, Norway and the United Kingdom, emissions in this sector include also flaring activities in oil platforms although it is an open question whether the emissions should be allocated in sector 9 or rather in sector 5. Population is used as an indicator to distribute waste incineration sources in the country. The distribution of emissions from open burning of agricultural waste is based on the distribution of sector 10. The proportion of sources to either one waste category varies from country to country. We have used CEPMEIP (2002) data on waste activities detailed at SNAP level 2 to determine that proportion of waste emissions. The results are summarised in Table 3.3, where the recommended height of the emissions is also established. For population related waste, low height sources correspond to open waste and high sources correspond to incinerators.

The method summarized in Tables 3.1, 3.2 and 3.3 has been applied to all countries that have not reported gridded sector data for all components. If a country has reported GS data for a component, we compare the reported gridded sector (GS) data with our method results. If there are no obvious inconsistencies, the officially reported GS sector data is directly used. Table 3.4 provides an overview of the countries that have reported in the last four years GS data that has been seen to be used directly in EMEP model applications.

Table 3.3: Overview of indicators used for the spatial distribution of emissions from Sector 9, waste treatment and disposal related activities in each country.

Country	Agriculture-related waste		Population-related waste		Flaring (LPS)-related[‡]	
	Percentage	Height	Percentage	Height[†]	Percentage	Height
AL	75	Low	25	Low	0	High
AM	50	Low	50	Low	0	High
ASI	50	Low	50	Low	0	High
AT	0	Low	100	High	0	High
AZ	25	Low	20	Low	55	High
BA	50	Low	50	Low	0	High
BE	0	Low	100	High	0	High
BG	75	Low	25	Low	0	High
BY	100	Low	0	Low	0	High
CH	0	Low	100	High	0	High
CY	75	Low	25	Low	0	High
CZ	100	Low	0	Low	0	High
DE	0	Low	100	High	0	High
DK	25	Low	75	High	0	High
EE	100	Low	0	Low	0	High
ES	50	Low	50	High	0	High
FI	50	Low	50	Low	0	High
FR	0	Low	100	High	0	High
GB	0	Low	90	High	10	High
GE	50	Low	50	Low	0	High
GR	75	Low	25	Low	0	High
HR	75	Low	25	Low	0	High
HU	60	Low	40	High	0	High
IE	0	Low	100	Low	0	High
IS	0	Low	100	Low	0	High
IT	0	Low	100	High	0	High
KZ	100	Low	0	Low	0	High
LT	100	Low	0	Low	0	High
LU	0	Low	100	High	0	High
LV	100	Low	0	Low	0	High
MD	100	Low	0	Low	0	High
MK	100	Low	0	Low	0	High
NL	0	Low	100	High	0	High
NO	0	Low	0	High	100	High
PL	25	Low	75	Low	0	High
PT	25	Low	75	Low	0	High
RO	75	Low	25	High	0	High
RU	75	Low	25	Low	0	High
SE	0	Low	100	High	0	High
SI	60	Low	40	Low	0	High
SK	50	Low	50	High	0	High
TR	25	Low	75	Low	0	High
UA	100	Low	0	Low	0	High
YU	75	Low	25	Low	0	High

Notes: [†]: High= Incinators, Low=Open waste [‡]: Oil and Gas Production

Table 3.4: Overview of official submissions on the spatial distribution of sector emissions (gridded sector data, GS) available in 2004.

Country code	CO	NH_3	NMVOC	NO_x	SO_x	PM_{10}	$PM_{2.5}$	Total
AT - Austria	1	1	1	1	1	1	1	7
CH- Switzerland	1	1	1	1	1			5
DE- Germany	1	1	1	1	1			5
DK- Denmark	1	1	1	1	1	1	1	7
ES - Spain	1	1	1	1	1			5
FI - Finland	1	1	1	1	1	1	1	7
GB - United Kingdom	1	1	1	1	1	1		6
LT- Lithuania	1	1	1	1	1			5
NL - Netherlands	1	1	1	1	1			5
NO- Norway	1	1	1	1	1			5
RU - Russian Federation		1						1
SE - Sweden	1	1	1	1	1			5
Total no. countries	11	12	11	11	11	4	3	63

3.3.2 Results and differences with respect to previous years

The new methodology to distribute sector emissions in the EMEP domain has been used in all calculations by the EMEP Unified model carried out in 2004. In the following, we highlight differences in the spatial distribution of emissions with respect to estimates from previous years. It should be noted that the comparison is carried out for two estimates using the exactly the same sector totals and the same total national emissions, so that the differences presented in this section correspond only to the spatial distribution of emissions.

Sector 1

The distribution of sources from power plants and energy combustion has changed significantly in Italy, particularly around Milan area; in France around Paris; in Portugal, Ireland and Belgium. The largest differences however are in Eastern European countries: Croatia, Romania, Serbia and Montenegro, Czech Republic, Slovenia, Slovakia, Poland, Belarus, Ukraine and the Russian Federation. Figure 5 shows the distribution and extent of the changes in spatial distribution of power combustion sources with respect to previous years. The figure shows ratios between the old and the new spatial distribution of sources. Red areas indicate places where emissions were assumed significantly higher (by a factor of 5) last year than in the present 2004 estimate. Black points correspond to areas with significantly higher emissions with the present 2004 allocation methods. Blue depicts areas with no significant changes in the spatial distribution of emissions, usually relating either to areas where there exist official reports of gridded sector data or to regions outside the UNECE domain where even ancillary data is difficult to find.

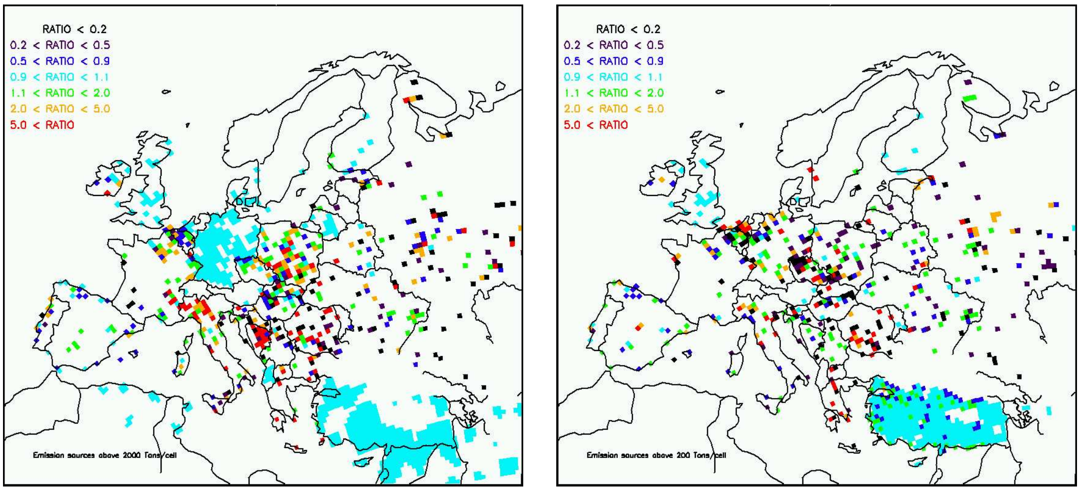

Figure 3.5: Energy Combustion. Differences in the distribution of sector 1 emissions with the new grid methodology: a) SO_x (left panel) and b) PM_{10} (right panel). See text for further explanation.

Figure 3.5a shows changes in emissions of sulphur dioxide. Since sector 1 is the predominant sector for SO_x emissions, changes in the spatial distributions of this sector are particularly relevant for the total SO_x emissions. For other compounds, however, the map of changes look similar to Figure 3.5a. This is because changes in NO_x distributions are in this case similar to SO_x and the distribution of CO and VOC emissions from energy combustion have been distributed in the new methodology to be consistent with NO_x emissions, so that the ratio of NO_x/VOC and NO_x/CO remains constant. In the future we might change this, as information from Large Point Sources is updated per component.

The largest difference in the spatial distribution of sector 1 with respect to other components is for PM emissions. For PM_{10} and $PM_{2.5}$, the distribution of combustion sources was before based on TNO-CEPMEIP distributions while now they are based on reported GS and the LPS data either from IER or directly reported by the countries. This secures that primary PM emissions are now distributed consistently with the emissions of the gaseous precursors of PM. Since 95% of PM_{10} emissions from combustion sources is in the fine mode, the changes in Figure 3.5b are also representative for $PM_{2.5}$. The reason for changes illustrated in Figure 3.5a and Figure 3.5b is the update in the methodology applied to distribute the emissions form energy combustion. Last year, a method was used to re-distribute the emissions from sector 1 according to the position of Large Point Sources, but the emissions were still homogenously distributed among the different LPS in each country. The updated methodology used this year, redistributes all emissions in the sector according to information on the actual intensities of emissions from each LPS. This means that the new distribution of sector 1 differentiates emissions within the countries according to the extent of each individual LPS.

Sector 2

Residential combustion has been gridded according to population for all compounds, as we expect these emissions to occur in urban centres. Differences with previous calculations are evident from Figure 3.6a in Portugal, Italy, France, Ireland, Slovenia, Poland, Belarus, Czech Republic, Slovakia, Slovenia, Hungary, Romania, Ukraine and the Russian Federation. All these countries have not reported GS data, but in the former methodology the distribution of emissions from this sector was made according to the sector share of reported gridded totals emissions. The use of gridded total emissions even though they are reported by the countries gives rise to inconsistencies. An example is shown in Figure 3.6a concerning the large changes (red points) in Sicily (Italy). The maximum SO_2 emission area from Italy is situated in Sicily, corresponding to Mount Etna volcanic emissions. In the old methodology, sector emissions were distributed according to grid totals, scaled by the share of each sector to national totals. As a result, a significant part of Italy's residential combustion emissions were placed in Sicily. The new methodology is more correct as it allocates the emissions of sector 2 in urban centres, following population. This is done so even if countries have reported

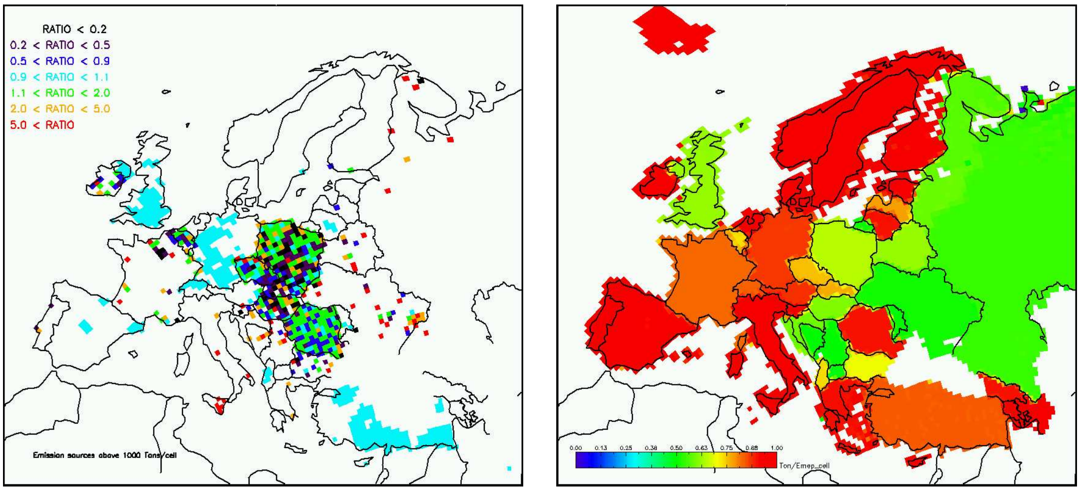

Figure 3.6: Residential Combustion. a) Differences in the spatial distribution of sector 2 SO_x emissions due to the new gridding methodology (left panel). b) Ratio $PM_{2.5}/PM_{10}$ illustrating the consistency of the PM emission in sector 2 (right panel). See text for further explanation.

gridded emission totals.

For PM, emissions from sector 2 follow also population distribution and are consistent with the gaseous pollutants. To analyse the consistency of the PM emissions across Europe we have checked the ratio between $PM_{2.5}$and PM_{10}. The $PM_{2.5}/PM_{10}$ ratio from sector 2 emissions gives an indication of the relative importance of combustion of gas, oil, wood and coal burning for residential purposes in each country. We can expect a larger fraction of $PM_{2.5}$ in gas and oil burning, and a larger content of fly ash (PM_{10}) in wood and coal burning. Therefore, we can expect a lower ratio in Eastern European countries that in Western European countries. The results of the test depicted in Figure 6b) show an average ratio of 0.5 in Eastern European countries and of 0.95 in Western European countries, as expected for sector 2.

Sector 3

The same type of differences in the spatial distribution of sector 1 and sector 2 are found also in the distribution of industrial combustion sources. Changes affect the same countries as in Figure 5 and Figure 6, and for the same reasons as explained above: the new methodology makes a consistent use of ancillary data for all components and countries that have not reported GS data, it does no longer use gridded totals information and it explicitly uses information on LPS intensities instead of only their locations.

Sector 4

Figure 3.7 shows differences in the spatial distribution of emissions from non-combustion production processes. The differences respond to the present use the total information on LPS, allocating also the intensities, and not only positioning as it was done before. However, the methodology used still needs to be refined for emissions from this sector: we are allocating non-combustion processes according to energy combustion information and this can give rise to inaccuracies. It is important to compile more detailed ancillary data from LPS (IER, country reports, EPER) so that it includes also sector emission information.

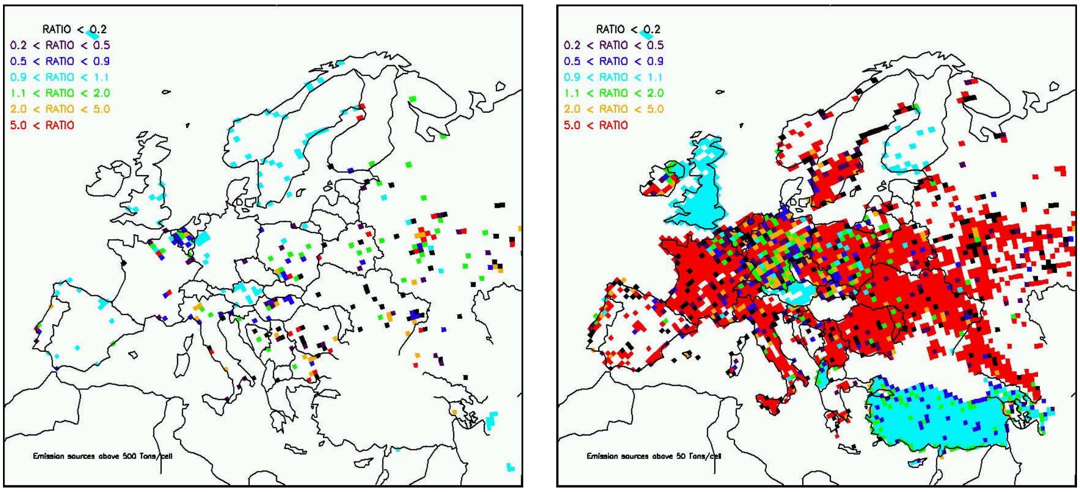

Figure 3.7: Production Processes. Differences in the distribution of sector 4 emissions with the new grid methodology: a) NO_x (left panel) and b) PM_{10} (right panel). See text for further explanation.

Emissions from production processes are less significant for total SO_x, NO_x emissions than for PM_{10} and $PM_{2.5}$. However, it is for PM emissions that we found the largest differences between the two gridding methods (see Figure 3.7b). The reason for the largest differences for PM_{10} (and $PM_{2.5}$) data is that the old methodology used TNO-CEPMEIP data while we are now using NO_x LPS data from IER or countries. TNO-CEPMEIP included a series of area sources in this production sector that are now concentrated in LPS areas. As mention above, it is difficult to say how accurate is the new description of sector 4 sources because of the ancillary data used has its obvious limitations. Figure 3.7b shows, for example, that the new methodology misplaces cement factories in Spain while the TNO-CEPMEIP distribution seemed to be more in line with the actual source distribution. How important these problems are for the modelled concentrations fields of PM matter, needs to be investigated further.

Sector 5

Sector 5 emissions are related to mining and fuel extraction. Emissions from this sector are significant for VOC and PM. We do not expect emissions from NO_x and SO_x and NH_3 reported in this sector and only low values of CO emissions from oil fields. Figure 8 shows the differences in the distribution of VOC emissions due to the use of the new methodology. The new methodology uses gridded sector emissions for sector 5 from TNO-CEPMEIP emissions of PM to distribute the VOC, thus securing the consistency of the emission spatial distribution among pollutants. In the old method, distribution of pollutant emissions in sector 5 was done according to gridded total distributions. The old methodology systematically located VOC emissions close to urban centres. This was because the main contributors to VOC emissions are solvent and paint industry and transport emissions, all of them normally situated around city/urban centres and adjacent roads. Figure 3.8 shows that the new methodology consistently moves VOC emissions from sector 5 away from city centres. PM emissions from sector 5 are mostly due to dust fugitive emissions from mining and are dominantly in the coarse mode. Only a small fraction of the PM emissions from sector 5 are emitted as $PM_{2.5}$. Again, the analysis of the ratio between $PM_{2.5}$and PM_{10} is a good test of the consistency of PM emissions reported in both modes. The average value for $PM_{2.5}/PM_{10}$ emissions from sector 5 over Europe is 0.15. Figure 3.8 identifies France, Finland and Norway as outliers in the estimated PM emissions for sector 5. Bilateral discussions with national emission experts from these countries are further required to clarify the possible inconsistencies.

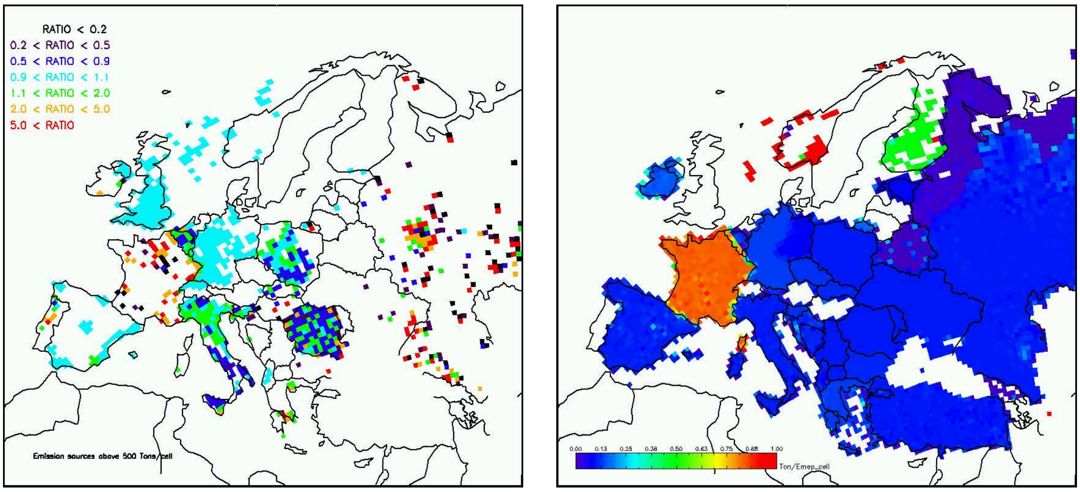

Figure 3.8: Extraction and Distribution of Fossil Fuels. a) Differences in the distribution of Sector 5 emissions with the new grid methodology for VOC (left panel) and b) Ratio $PM_{2.5}/PM_{10}$ illustrating the consistency of the PM emission in sector 5 (right panel). See text for further explanation.

Sector 6

Emissions from solvents and product use generally represent about 30% of the total VOC emissions. Emission from this sector are located in the new methodology according to population, and therefore systematically moved towards urban centres. The differences with the old methodology that located these emissions according to grid total emissions are shown in Figure 3.9. Since about 40% of VOC emissions originate

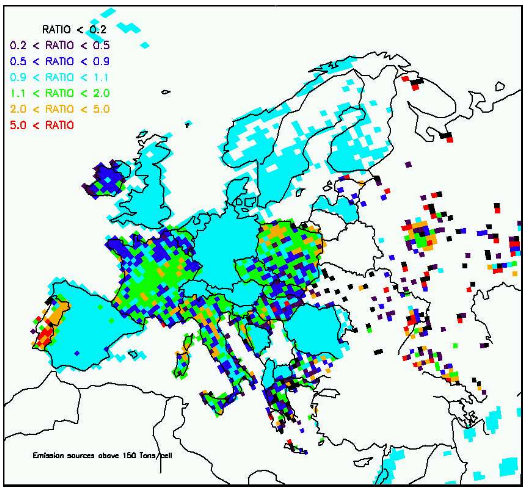

Figure 3.9: Solvents and Product Use. Differences in the distribution of sector 6 emissions with the new grid methodology for VOC emissions. See text for further explanation.

from traffic (both road and off-road), gridding by total emissions involved systematic biases that are now corrected. We do not expect any significant contribution to emissions from other pollutants in this source sector.

Sector 7

The present methodology to allocate emissions from road traffic uses reported gridded sector data for NO$_x$ for distributing all other pollutant emissions in this sector. However, only 11 countries have reported gridded sector emissions from NO$_x$. For countries that have not reported the gridded sector data for NO$_x$, TNO-CEPMEIP emission distribution for PM$_{10}$ in sector 7 are used instead as tracers of the road traffic distributions. Figure 3.10 shows examples on the changes in the gridded emission distribution when using different methods. The upper left panel in Figure 3.10a shows the distribution of PM$_{10}$ emissions from sector 7 with TNO-CEPMEIP. It can be seen these emissions are spatially correlated with population by comparing with the population map in the lower left panel. The upper right panel in Figure 3.10a shows the spatial distribution of PM$_{10}$ emissions using official gridded sector 7 data for NO$_x$ (NO$_x$ GS7) to allocate the emissions. The NO$_x$ GS7 method is not highly correlated with population as indicated in the lower left panel in Figure 3.10a. In fact, the distribution of PM$_{10}$ emissions using the official NO$_x$ GS7 data corresponds much better to the distribution of traffic volumes and roadmaps from Spain. The same applies to all other pollutants: NO$_x$ GS7 is better indicator to grid road transport emissions than the TNO S7 method that relates PM in traffic more to population than the actual roadmaps. Still, only 11 countries have reported gridded sector NO$_x$, so the TNO-CEPMEIP distributions have been generally used. Despite its limitations, the use of the new method is

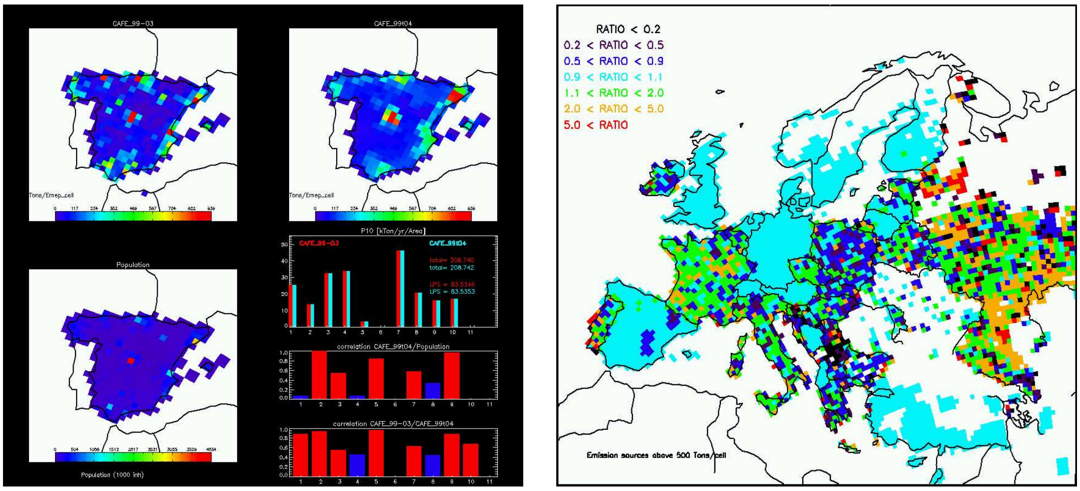

Figure 3.10: Road transport. a) Illustration of methods used to distribute PM_{10} emissions from road traffic in Spain, following TNO S7 data or NO_x GS7 reported emissions b) Differences in the distribution of sector 7 emissions with the new grid methodology for NO_x emissions (right panel).

more adequate than the previous use of gridded total data, as illustrated for NO_x in Figure 3.10b.

The reason for the superiority of the new method is that about 30% of NO_x emissions are related to combustion and not only to traffic emissions. The old method distributed traffic emissions according to gridded total data and could therefore erroneously locate traffic emissions in areas with power plants. Such erroneous distribution of NO_x emissions in some particular areas gave rise to inconsistencies with VOC emissions and had consequences for the calculation of ozone concentrations. The new method avoids this type of inconsistencies and secures that emissions from the same sources are distributed equally for all pollutants.

To test the robustness of the new method and check the consistency of the results, we have calculated the ratios between NO_x, PM_{10}, VOC and CO all from road traffic emissions and compared these to COPERT (2003) results.

The first ratio analysed is NO/VOC. This ratio is significantly affected by the proportion of diesel powered vehicles (e.g., heavy duty vehicles or diesel passenger cars) in the overall road transport fleet. A higher presence of diesel will tend to increase the ratio of NO_x to VOC. For the mix of vehicle technologies in the EU in year 2000, COPERT results indicate a range of between 4.5 (100% Diesel) and 0.75 (100% Gasoline). The upper left panel in Figure 11 shows the values of the NO_x/VOC ratio when applying the new methodology for gridding emissions. There are interesting variations in the ratios within European countries that have reported emission GS data indicating the differences on the type of vehicles circulating in different roads. Averaged values over EU15 are 1.1-2.0 and about 0.8-1.3 in EU10+, corresponding well with COPERT estimates.

In Eastern Europe, the situation is different as the vintage of the vehicles is old thus lowering the NO_x/VOC ratio in EEE countries down to 0.5 (typical of pre-catalyst passenger cars). Armenia appears to be a special case, with NO_x/VOC ratios up to 5, which could be due to a dominating presence of old diesel trucks in the country.

The next ratio analysed and compared to COPERT results is NO_x/PM_{10}. The ratio NO_x/PM_{10} is again indicative of the type of vehicle and type of fuels used. Gasoline cars generate NO_x emissions but little PM_{10} (at least in terms of mass)[2] while for diesel vehicles both PM_{10} and NO_x are emitted. Consequently, the overall NO_x/ PM_{10} ratio will be higher in situations with a larger proportion of gasoline powered vehicles in the overall fleet. The vintage of the vehicles also a role, since older heavy duty diesel technology, of the type still present in Eastern Europe; emit significantly emissions of PM_{10}.

Typical NO_x/PM_{10} ratios in Western Europe are about 20; somewhat higher in countries with lower percentages of diesel vehicles (like the United Kingdom, where the ratio is about 25) and lower for countries with high penetration of diesel vehicles (like Spain, with 15; and France with 11). The old diesel vehicles from Armenia and other Eastern European countries tend to emit significantly more PM_{10} (but not NO_x) and the ratios can then be about 4, according to COPERT. In the Russian Federation, a combination of use of old vintage gasoline cars and smaller old diesel trucks could explain a NO_x/ PM_{10} ratio of about 15. The right upper panel in Figure 11 shows the results for the emission estimates used in EMEP that correspond well with the COPERT estimates.

The last ratio analysed is NO_x/CO, which again is indicative of the type of vehicles and fuels used in a country and should be consistent with the other two ratios. Diesel vehicles emit considerably less carbon monoxide than gasoline cars. Typical ratio values in EU25 according to COPERT are NO_x/CO=1 (100% diesel) and 0.1 for (100% gasoline). The values derived from EMEP are lower but show the higher values in countries where diesel powered vehicle are more dominant, and lower values in countries where this is not so.

In Eastern Europe, the vintage of the cars is older. For old gasoline cars (pre-catalyst), COPERT estimates a significantly lower NO_x/CO ratio, down to 0.05. The values in EMEP in Eastern Europe are consistent with this low ratio, consistent with the NO_x/VOC and NO_x/PM_{10} ratios that already indicated a high presence of old gasoline vehicles. The only inconsistency is in Armenia. The NO_x/VOC ratio in Armenia was very high which indicated a generalised use of old diesel trucks in the country. According to COPERT, the NO_x/CO ratio for old diesel trucks should be around 2. However, Armenia reports NO_x/CO of 0.02, more in line with other Eastern European countries. Bilateral discussions with national experts should help to clarify these values.

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[2] In many cases, the mass emissions of particulate matter from gasoline powered vehicles are ignored in determining overall emissions from road transport. For example COPERT III does not provide an emission correlation for particulates from gasoline vehicles.

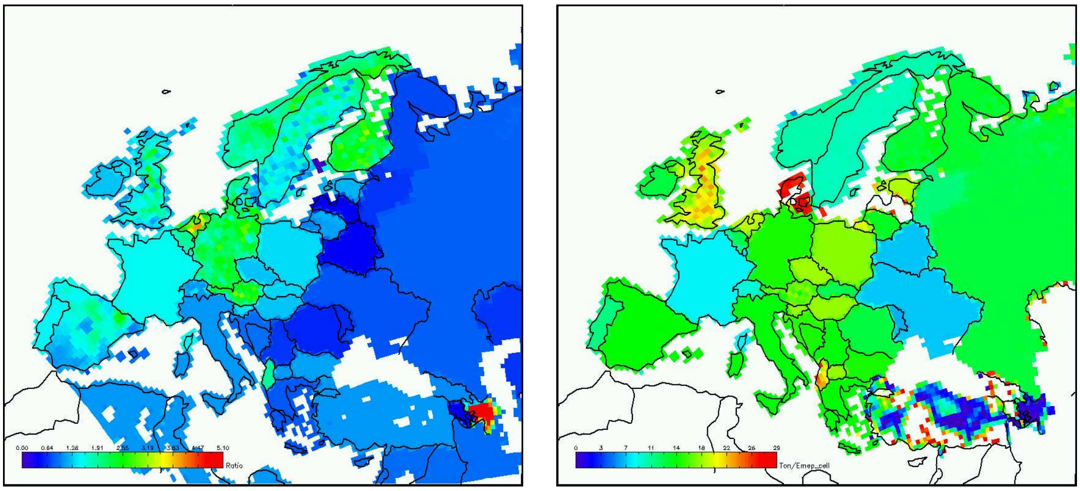
In general, the ratio values derived from EMEP emissions correspond well with COPERT estimates. An overview of the comparison between COPERT estimates and the ratio values derived from EMEP emissions is given in Table 5 for road transport emissions.

Table 3.5: Summary comparison of COPERT and EMEP derived estimates for pollution emission ratios from road traffic.

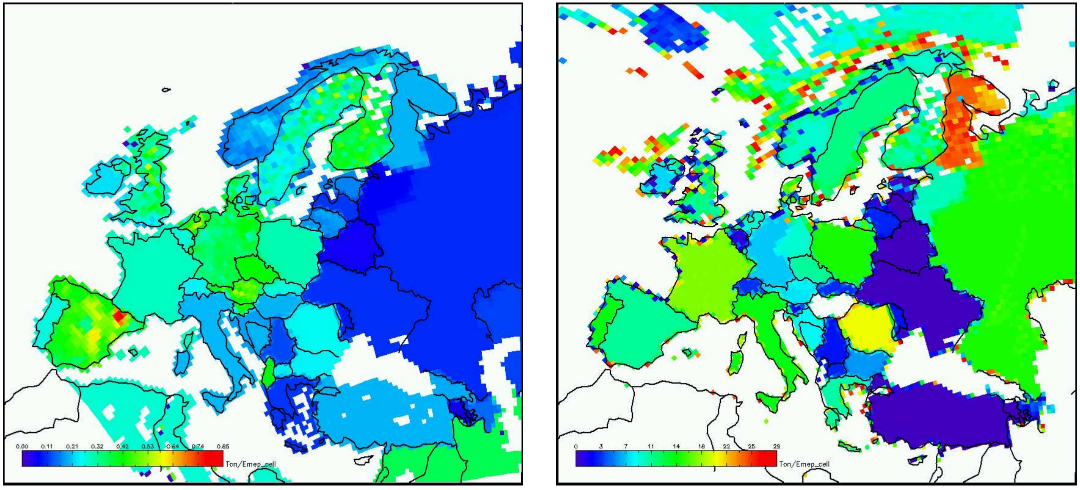
	NO_x/VOC	NO_x/PM_{10}	NO_x/CO
EU 15 - COPERT	1.1-2.0	10-25	0.3 - 0.4
EU 15 – EMEP use	1.1-3.0	8-25	0.1- 0.5
EU10+ - COPERT	0.8-1.3	8-15	0.2 -0.4
EU10+ – EMEP use	0.6-1.2	8- 25	0.2-0.5
EEE -COPERT	0.5- 0.75	4 -10	0.05 – 1 (2)
EEE – EMEP use	0.5 -0.8 (5)	4 -15	0.01-0.3
Expected ranges	0.5- 5.0	4 - 25	0.01-2

Sector 8

The methodology to distribute emissions of off-road traffic and machinery is the same as for road traffic emissions. If countries have reported gridded sector information for NO_x, the distribution is used as basis for distributing the other pollutants, otherwise PM_{10} information from CEPMEIP (sector 8) is used. The resulting differences between the new and old methodology resemble those for sector 7 and therefore are not shown. Emissions from ship traffic are included in this sector but the present methodology does not imply any changes in the spatial distribution of shipping sources. Both the spatial distribution and the intensity of international sources in sector 8 require a careful re-evaluation as there are identified inconsistencies in the reporting of these emissions from the countries. However such re-evaluation is beyond the purpose of this study. As for traffic emissions, we have calculated NO_x/VOC, NO_x/PM_{10} and NO_x/CO ratios and compared them to COPERT results in order to check the consistency of the results. For emissions in sector 8, we expect a more generalised use of diesel in off-road transport than in road traffic emissions. Thus, we expect lower values in the ratio NO_x/PM_{10} in all countries than those calculated for sector 7. The lower right panel in Figure 3.11 shows the ratio NO_x/PM_{10} in sector 8. Comparing this with the upper right panel in Figure 11, we see that the ratio decreases in general, as expected. It is interesting to note the anomalies in port areas and also in Turkey. Turkey



NO$_x$/VOC (S7, range 0.5-5.0) NO$_x$/PM$_{10}$ (S7, range 4-25)



NO$_x$/CO (S7, range 0.01-1) NO$_x$/PM$_{10}$ (S8, range 4-25)

Figure 3.11: Indicator ratios used to test the consistency of the emissions from different compounds for traffic sector 7 and sector 8. See text for further explanation.

is the only country where the new methodology has not been applied consistently because of lack of ancillary information. Further efforts should be dedicated in the short term to fully include Turkey in the new gridding methodology. The high NO$_x$/PM$_{10}$ ratios in port areas are another example of the inconsistencies in international shipping data in sector 8 and require, as already mentioned, a special separate study.

Sector 9

This is the sector where the new methodology has introduced the largest variations with respect to previous year's estimates. The contribution from waste treatment and disposal to emission totals is below 5% for most gaseous emissions, and about 5% for primary PM emissions. Consequently, the change from a gridded total scaling approach to the new methodology has introduced significant changes in most countries, even though the influence of such changes in the distribution of total emissions will be small. The new methodology uses population as indicator to distribute waste incineration in urban areas and agricultural activities from CEPMEIP as indicator for agricultural waste. In addition, some few countries include flaring activities in this sector. The resulting changes between the two methods are presented in Figure 3.12. A significant feature in this picture, presently under bilateral discussion with TNO, is that agricultural activities from the Russian Federation are moved with the new methodology towards the European border line. Further refinement of sector 9 gridded distribution will depend on the availability of ancillary data and/or national information on gridded sector data.

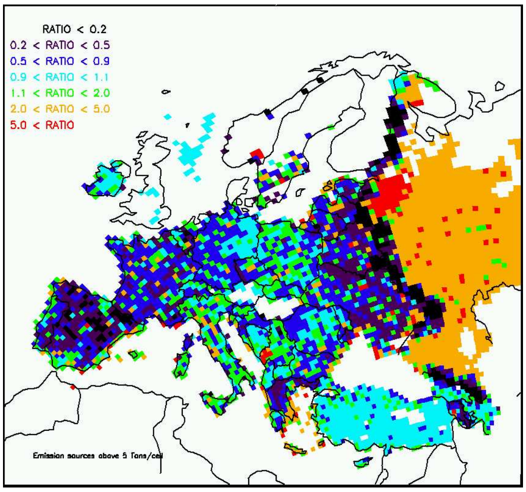


Figure 3.12: Waste. Differences in the distribution of sector 9 emissions for PM_{10} with the new grid methodology. See text for further explanation.

Sector 10

In the new methodology, emissions from agricultural sources are re-arranged according to TNO-CEPMEIP data for sector 10 which in turn is based on farming activities and land-use information. We do not expect emissions of other gases than NH_3 emitted in this sector, as explained in section 2 above when discussing IIASA's revised sector distribution. Ammonia emissions from agriculture are mainly related to dairy and pig farms while for PM, about 2/3 of all emissions originate in this sector from poultry farms. This difference can give rise to a systematic bias in the spatial distribution of ammonia emissions but such bias is considered to be small, as in most countries farming activities areas are collocated. The advantage of the new method is that it can be applied to the whole of Europe and that is guarantees the consistency of primary PM emissions and gaseous precursors.

In 2004, TNO elaborated a new estimate of the spatial distribution of agricultural

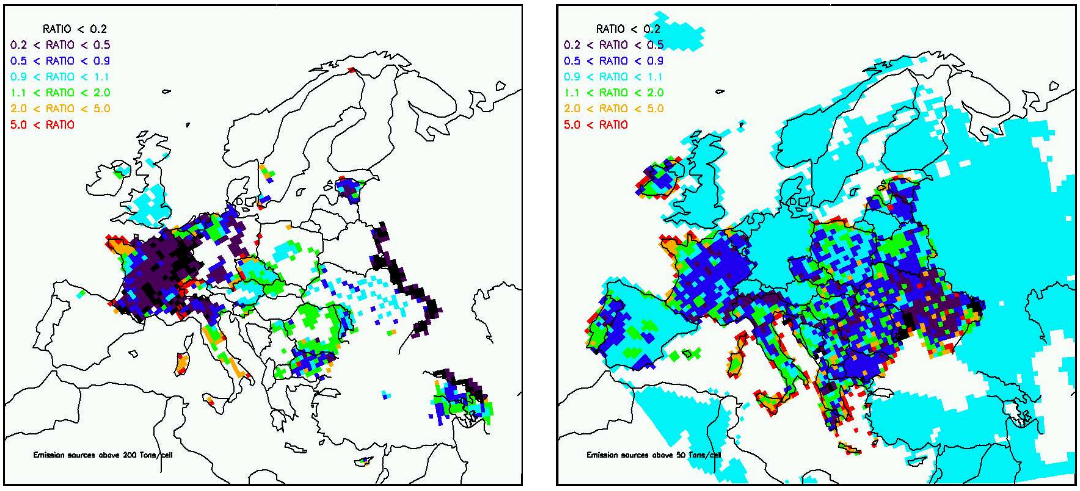


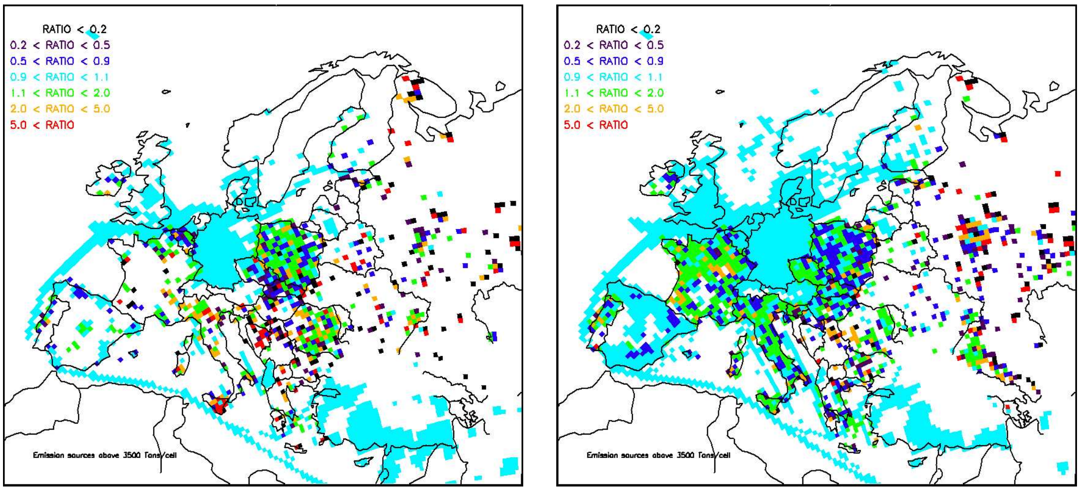
Figure 3.13: Agriculture & Forestry. Differences in the distribution of sector 10 emissions with the new grid methodology for a) PM_{10} - TNO S10 update (left panel) and for b) NH3 emissions (right panel) and. See text for further explanation.

emissions (Visschedijk, pers. comm.) for use as ancillary data in the new gridding methodology. The new TNO S10 estimate differs considerably from previous estimates in France, Italy and the Russian Federation, as illustrated in Figure 3.13a. In particular, for the Russian Federation, official gridded data for NH_3 is available to EMEP and the distribution of agricultural sources differs with the new TNO S10 but is more in agreement with previous CEPMEIP distributions. For NH_3, the spatial distribution of emissions follows the reported data in the Russian Federation and therefore, no changes are made in this country with respect to the previous distributions (Figure 3.13b)). However this implies that PM_{10} and NH_3 emissions are not consistent in Russian Federation, especially along its borders with other Eastern European countries. As already mentioned, the updates in S10 emission distributions are presently under discussion and we trust that bilateral discussions with TNO will provide a solution and an explanation for the Russian Federation emissions.

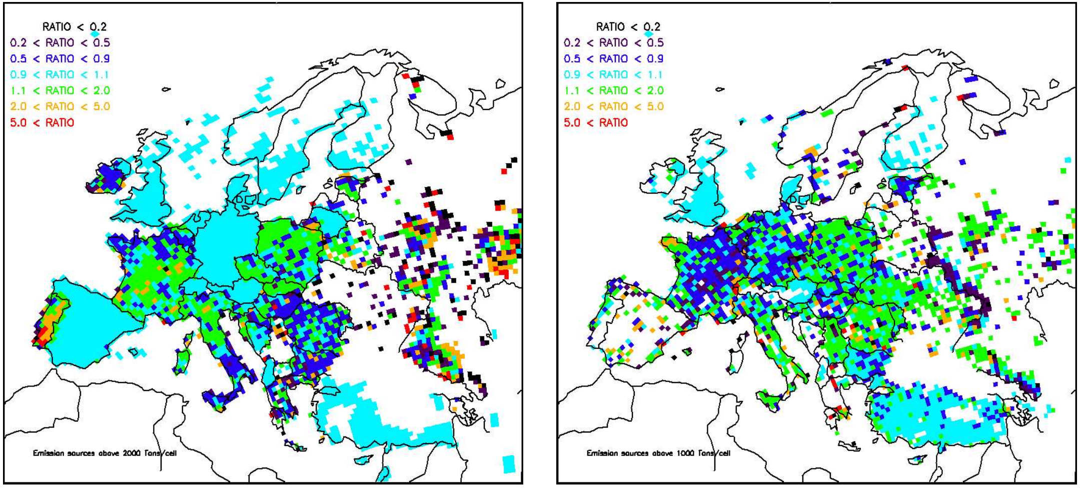
3.3.3 Significance of the new spatial distribution methods for the calculation of air concentrations and depositions

Initial tests with the Unified EMEP model have been carried out to determine the significance of the new spatial distribution of emissions in the model results. The model has been run twice, with the same sector and national emission totals but using the old spatial distribution in the first run and the new spatial distribution in the second run. Work is in progress to analyse the differences and initial results are presented in Chapter 5 in this status report (Fagerli, 2004). Here, only some preliminary conclusions are summarised.

The spatial differences in the sum of all sectors are driving the changes in the



SOx, all sectors NOx, all sectors



VOC, all sectors PM_{10}, all sectors

Figure 3.14: Differences in the distribution of SO_x, NO_x, VOC and PM_{10} emissions for all sectors with the new grid methodology. See text for further explanation.

model results. The individual sector distributions affect the model calculations mostly through the related height of emissions to the atmosphere. Figure 3.14 shows the spatial differences for all sectors for SO_x, NO_x, VOC and PM_{10}. Since 95% of ammonia sources originate from agriculture-related activities, the spatial differences for all sectors for NH_3 correspond well to the changes illustrated in Figure 3.13. These figures can be compared directly with the concentration changes in Chapter 5 (Figure 5.26). To facilitate the comparison, spatial distribution changes for ammonia emissions and ammonia and ammonium concentrations in air are depicted in Figure 3.15. As for all other studied primary pollutants, the concentration changes for NH_3+NH_4 spatially correspond with the emissions changes.

The largest differences in the spatial distribution of emissions are for primary PM.

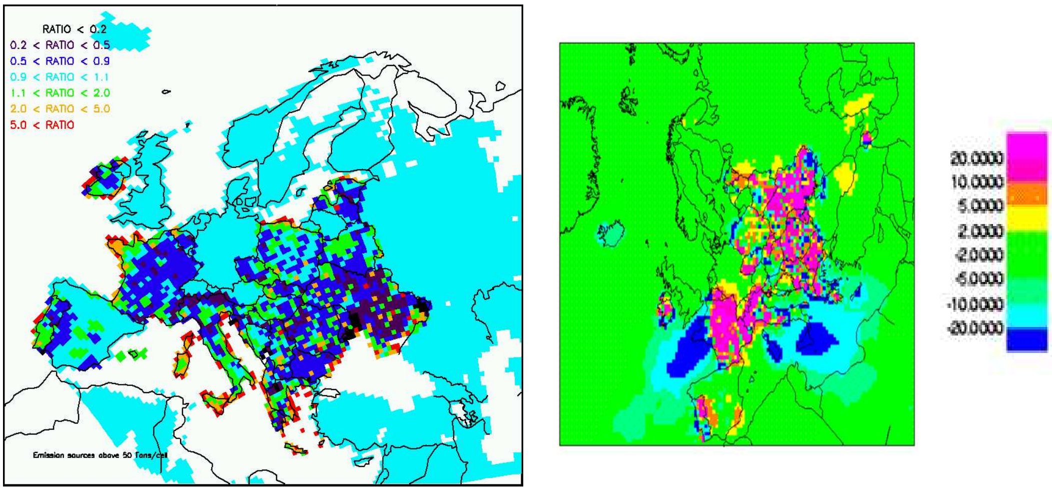


Figure 3.15: Comparison of emission distribution changes in ammonia emissions from S10 (left panel) and the corresponding changes in air concentrations of NH3+NH4 in air (right panel) from EMEP model calculations in Chapter 5 (Fagerli, 2004).

This is because there are considerable changes in the spatial distribution of emissions from source sectors that contribute significantly to the total emissions of PM mass, namely sectors 4, 7 and 10. For gaseous emissions, the contribution of sources from sectors 4 (production processes) and 10 (agriculture and forestry) are generally below 12-14%, for primary PM emissions the contribution of these two sectors to total emissions is 20% for PM$_{2.5}$and 30% for PM$_{10}$. While the changes in the distribution of sources from road traffic are an obvious improvement in the new methodology, there are recognised limitations in the new methodology results for sector 4 and sector 10. The implications of these are presently under evaluation.

The initial study is presented in Chapter 5 in this status report (Fagerli, 2004) for gaseous compounds indicates that the changes in the air concentrations of pollutants due to the proposed re-distribution of emissions can be significant. For annual averages of SO$_2$ and NH$_3$+NH4 concentrations in air changes can be over 20%. For nitrate and sulphate, the changes in some areas can be up to 10-20%. For ozone, changes become more significant in the vicinity of cities. For example, the changes in VOC and NO$_x$ emission distributions around Paris and Milan, imply changes in the mean ozone concentrations up to 20% (see Figure 3.16). This is most relevant for population impact studies.

It will be difficult to validate the changes in the emission distribution through comparison of model results with observations, because the most significant changes are in Eastern Europe, that is, in areas where there are few available monitoring results. However, the initial analysis of the derived concentrations indicates that the new spatial emission generally improves the spatial correlation of modelled results with observations. Further work will continue in this direction but the initial results are reassuring for the validity of the new emission gridding method.

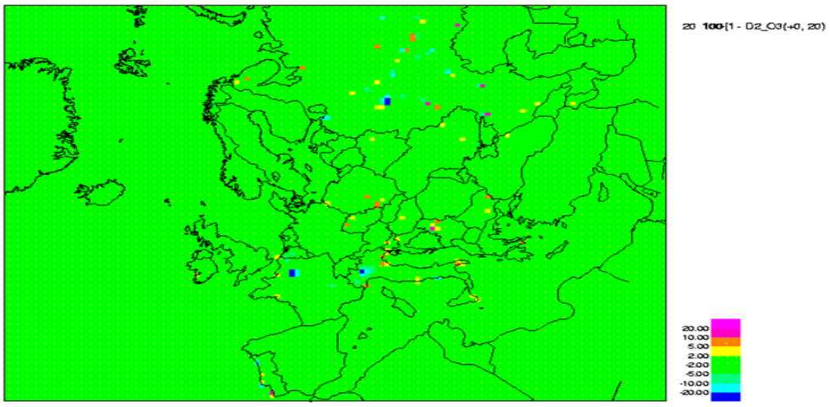


Figure 3.16: Percentage changes in yearly mean ozone concentrations due to changes in the distribution of emissions using the new gridding methodology. See text for further explanation.

3.4 Conclusions

All model calculations, source-receptor studies and scenario analysis carried out in 2004 with the Unified EMEP model use the same basic assumptions on the spatial distribution of emission sources. All calculations have also assumed the same basic sector distribution per pollutant and per country. The basic national sector distribution has been revised and updated by IIASA through bilateral discussions with the Parties. Scenario runs and source-receptor calculations have used the 2010 and 2020 national projections developed by IIASA under the EU CAFE_BASELINE project (see EMEP Status Report 4/2004, Amann et al., 2004). Status calculations for 2002 and model runs for previous years have used national emission totals as reported by the Parties and revised by MSC-W in co-operation with ETC/ACC (Vestreng et al., 2004).

Changes on national emission totals in 2002 are small with respect to 2001. For all main components and primary particle emissions, national emission changes in the EMEP domain are below 1%. For individual countries and components, changes in the national emissions are generally below 20%. For sector distributions, the adopted new distribution introduces the largest changes in ground-based sectors for primary PM emissions: residential combustion and traffic emissions, both in the vicinity of population centers. The new sector distribution resolves a series of identified inconsistencies in the sector allocation of emissions reported by the countries.

Significant changes with respect to previous calculations especially concern the spatial distribution of the emissions. A new methodology has been applied that ensures consistency in the location of sources for different pollutants across the whole EMEP domain. For the first time since CEPMEIP emissions where introduced in EMEP modelling, the distribution of primary PM emissions is now generally consistent with the emissions of PM gaseous precursors.

The new methodology relies on validated official gridded sector GS data reported from the Parties and on ancillary information on population, large point source (LPS) intensities and locations, traffic patterns, agricultural activities and land-use. Differences in the spatial distribution of emissions are considerable for all source sectors and can be well above a factor of 5 in single areas. Countries that have not reported consistent gridded sector GS information are those more affected by the changes in spatial distribution of emissions derived from the new methodology. Changes are most significant in France, Italy, Ireland, Portugal and the Eastern European countries.

The main reasons for the differences are that the new methodology: a) makes a consistent use of ancillary data on for all components and countries that have not reported GS data, b) it does no longer use gridded totals information and c) it explicitly uses information on LPS intensities instead of only their locations. The generalized use of official gridded total information in the old methodology introduced considerable inconsistencies among pollutants and had consequences for the model derived air concentrations and depositions. Recognised examples are the imbalances in the distribution of NO_x and VOC emissions in urban areas that area now corrected with the new methodology

To check the validity of the new methodology and the robustness of the results we have identified a series of relevant pollutant ratios and tested them against independent emission estimates. In particular, for traffic emissions, the comparison of the derived NO_x/VOC, NO_x/$PM_{10}$and NO_x/CO ratios with COPERT results has been reassuring for the gridding of traffic emissions.

Initial tests with the Unified EMEP model have been carried out to determine the significance of the new spatial distribution of emissions in the model results and its validity in comparison with observations. In general, the new spatial distribution can imply up to 20% changes in the modelled concentrations and depositions. Although it is difficult to validate the changes in the emission distribution since the most significant changes are in Eastern Europe or in areas where there are few available monitoring, the initial analysis of the derived concentrations indicates that the new distribution of emissions generally improves the spatial correlation of modelled results with observations.

The accuracy of the new methodology depends on the quality of the ancillary data used to distribute the emissions. It is intended to continue updating and improving such information in the future, especially concerning non-combustion sources in sector 4 and agricultural activities in sector 10, where the present methodology has recognised limitations. In the process of updating the ancillary information, co-operation with national experts will be essential and we hope that the presentation of this new methodology will also encourage the national elaboration of gridded sector data.

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