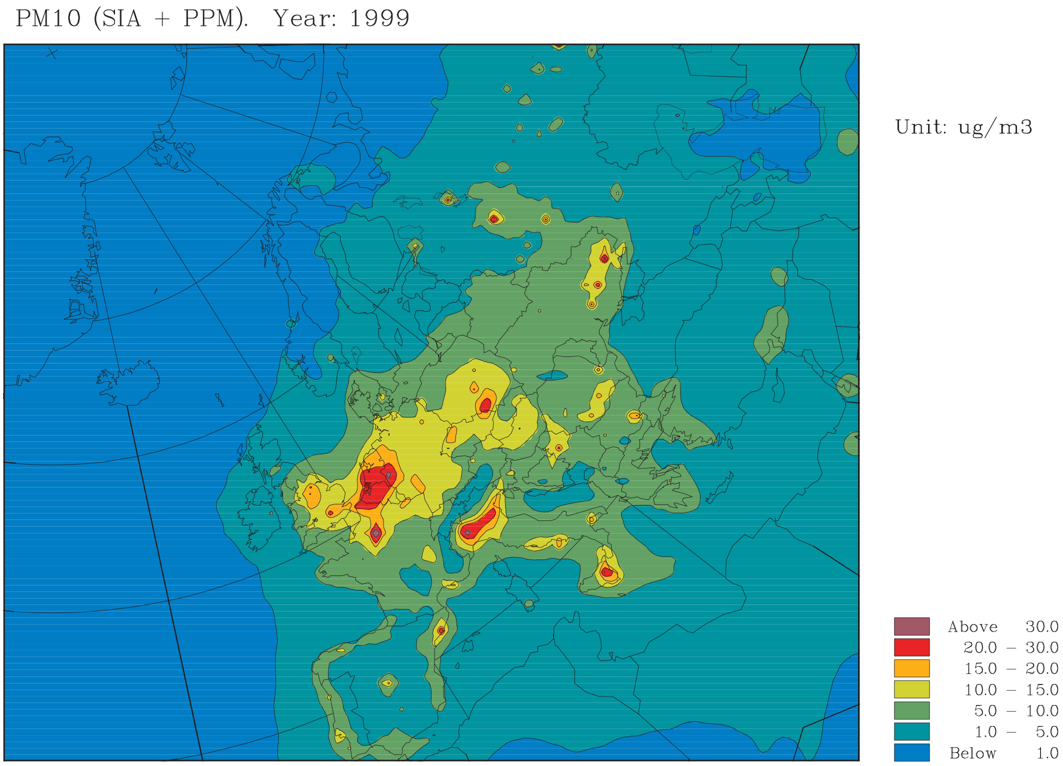
EMEP Report 4/2001

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

Transboundary Particulate Matter in Europe: Status Report 2001



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Report 2001

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Contents

1. Executive Summary

Long-range transboundary transport is responsible for a significant fraction of the particulate pollution in European cities as well as in rural areas (EMEP-WMO, 1999). A large part of this contribution is secondary particulate matter in the form of sulphate, nitrate, ammonium and organic aerosol particles formed by the oxidation of sulphur dioxide, nitrogen oxides, ammonia and organic gaseous species. It is apparent that particulate matter is not a single pollutant and its mass includes a mixture of many pollutants distributed differently at different sizes. The traditional EMEP approach is to find how the air quality and the deposition of pollutants are affected by emissions in the different countries in Europe and involves emission inventories, emission-based model simulation of atmospheric processes, transport and deposition, and verification and assessment by observations and measurements. These elements are to a large extent in place but many challenges remain. This report summarises the recent developments in the particulate matter work of EMEP.

The establishment of a relevant and adequate monitoring programme is a crucial step for the development of abatement strategies in Europe. The minutes of the first meeting of the Task Force on Measurements and modelling (TFMM) describes the development of the monitoring strategy for PM (available at http://www.ubavie.gv.at/tfmm/pages/meet.htm). The meeting also gave the recommendations for the PM monitoring programme to be established in EMEP. Minor alterations to this have later been introduced and the final recommendations will be included in the EMEP manual. The monitoring strategy for particulate matter can be found at www.nilu.no/projects/ccc/pm_strategy.html. The strategy puts requirements to the development of the monitoring programme and countries are strongly encouraged to expand their activities to meet the requirements given.

The EMEP monitoring programme currently provides insufficient data for model validation. This is particularly the case for information on the chemical composition of the aerosol. Although secondary inorganic aerosol components like nitrate and ammonium (partly also sulphate) have been part of the programme for many years, only few countries report the data separating the gas and the aerosol component. In addition to being essential for the improved description with respect to acidification and eutrophication, these compounds also are needed for the further development of the PM-modelling. For other parameters even less data are available and in particular for carbonaceous species.

Comparison of the EMEP model with available measurement data indicates a general underestimation of ambient PM_{10} concentration levels, and for some regions the discrepancies are large (e.g. the Mediterranean area). It should be noted however that natural and re-suspended anthropogenic mineral dust, sea-salts and biogenic aerosols are currently not incorporated in the model. The best agreements are found for German sites for which secondary inorganic aerosols are expected to account for a relatively large mass of the ambient PM_{10}. Verification of the model with observations of chemical composition in Switzerland indicates a fairly good correspondence for the secondary inorganic compounds, whereas for carbonaceous material the correspondence is poorer. However, the Swiss sites are to some extent influenced by more local emission sources. These inconsistencies between model estimates and measurements cannot be fully resolved at the moment. Important factors to consider are however inadequate emission data (particularly important for primary particulate matter from anthropogenic sources and for natural emission sources) as well as inadequate monitoring data for validation.

Further progress has been made in the implementation of physical aerosol dynamic processes in the Unified EMEP Eulerian model. A box model version of MULTIMONO is presently under testing, and initial results from studies on the choice of time integration schemes are presented in this report. The on-going testing of the performance of the box model for condensation, coagulation and nucleation is a necessary requisite prior to the implementation of the aerosol in the Unified EMEP Eulerian model.

Analysis of the non-linearities in the transport of primary particulate matter is also presented. It is shown that source-receptor matrices for primary particulate matter should differentiate between coarse and fine particles. This enhances the need for cost control estimates that are able to separate between fine and coarse particles, as those reported in Chapter 4. It also implies that PM emissions estimates and PM mass measurements should distinguish at least between $PM_{2.5}$ and PM_{10}.

Chapter 4 presents progress in the development of a model for estimating primary emissions of PM in Europe and the costs involved to reduce these emissions from the various sources is presented. Currently available estimates must be considered provisional, and the RAINS PM should therefore be seen as an initial step towards a harmonised approach to assess control costs on a European scale. It is shown that anticipated emission changes by 2010 will influence both the sectoral composition of emission sources and that the individual sectors show important differences depending on the particle size. Preliminary examples of so-called “national cost curves” for reduction in emissions are also presented.

Progress has also been made concerning the difficult task of quantifying primary particulate matter (PM) emissions in Europe. Through the Coordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP), TNO has recently completed an interim European emission inventory for primary particles. The purpose of this initiative is to provide guidance to the Parties to the LRTAP Convention in the compilation of their national emission inventories of PM. As an example, TNO emission inventories for Norway and Poland, year 1995, were compared to calculations by the RAINS PM module and national inventory estimates. The results point out how important it is to provide a detailed documentation of the methodology used to assess primary PM emissions (e.g., on sector aggregations, economic activities, derivation of emission factors). Results of total PM emissions, country totals or emissions aggregated according to the SNAP 1 codes, can only be compared and differences explained if this information is available. This supports the recommendations from the last Task Force on Emission Inventories and Projections (TFEIP) on the new draft Guidelines for Estimating and Reporting Emission data.

Although the basic approach in the RAINS, TNO and the Norwegian and Polish 1995 inventories is very similar, it could be shown that assumptions made on aggregations within the 10 SNAP 1 classes can differ considerably. This, among other things, results in different conclusions on the major PM emission sources (e.g. for Norway: residential wood burning according to RAINS and the national study, extraction of fossil fuels according to the TNO inventory). In many cases different units have been used for both activities and emission factors and these must be harmonised before direct comparison of basic input assumptions and thus calculation results is possible. An additional difficulty is the identification of the underlying assumptions concerning control options in the different economic sectors. The resulting emission factors can be very different whether they are abated or unabated.

In conclusion, a comprehensive description of the atmospheric particles requires the evaluation of particle number, surface and volume distributions in addition to the mass and chemical composition. Size distribution measurements combined with chemical speciation are necessary for identifying also the sources of atmospheric particulate matter. Measurements have to be combined with comprehensive modelling studies together with detailed size/chemical composition emission inventories for primary particles. The model validation requires sufficient measurement data both in terms of site density, data quality and chemical/physical parameters determined. Still major challenges remain ahead in all these respects.

2. Overview on particulate matter measurements in Europe

by Mihalis Lazaridis, Kjetil Tørseth, Arne Semb, Steinar Larssen, Øystein Hov

This chapter summarizes the present status of measurement program for particulate matter data available to EMEP. An overview of both particulate matter mass concentrations and of the chemical speciation of the aerosol is presented, while for a more comprehensive description of the monitoring data of particulate matter it is referred to EMEP/CCC-Report 5/2001.

It should be noted that the availability of data on particulate mass and of the chemical composition of particles over Europe is still very limited. Only four countries have reported PM mass data to EMEP while four additional countries have delivered PM mass data to the EIONET network. Also data on the chemical composition of PM is very limited in particular for the organic fraction but also for components which for long have been included in EMEP measurement programme (e.g. nitrate and ammonium). Recently a new monitoring strategy for PM has been developed within EMEP. The strategy recommends the measurement programme to be expanded to sufficiently describe the ambient concentrations of PM in Europe.

Switzerland, Germany, Spain and Italy performed particulate matter measurements during the 1999 using gravimetric methods. Germany reports PM_{10} data from 8 stations and Switzerland from 5 stations while Italy reports TSP data from one station (Ispra). Spain started in 1999 to report PM_{10} data from 10 stations across the country.

Annual average concentrations from the EMEP stations for the 1998 and 1999 are presented in Figure 2.1 and Figure 2.2 respectively. The annual average concentrations for both 1998 and 1999 are fairly similar. Concentrations in Germany for 1999 show an annual average of 19.5 $\mu g/m^3$ similar to Switzerland (20.5 $\mu g/m^3$). The station of Jungfraujoch at height of 3500 m has an annual average of 3.7 $\mu g/m^3$ for 1999 since it is located usually above the boundary layer height. Concentration levels are however higher during episodes when the boundary layer moves above the station.

In Switzerland, PM concentrations in 1999 are higher during winter than other seasons in agreement with observations from previous years. In Spain, higher PM concentrations are observed in summer but there is no consistent seasonal variability also in agreement with measurements from previous years. There is no consistent seasonal variability in Germany but a number of stations exhibit higher concentrations during the winter.

Annual averages of PM_{10} for 1999 at the EMEP-sites are compared with modelling results from the EMEP Eulerian model in Chapter 3.1.2. A comparison of the modelled chemical composition with observed concentrations is presented in chapter 3.1.3.

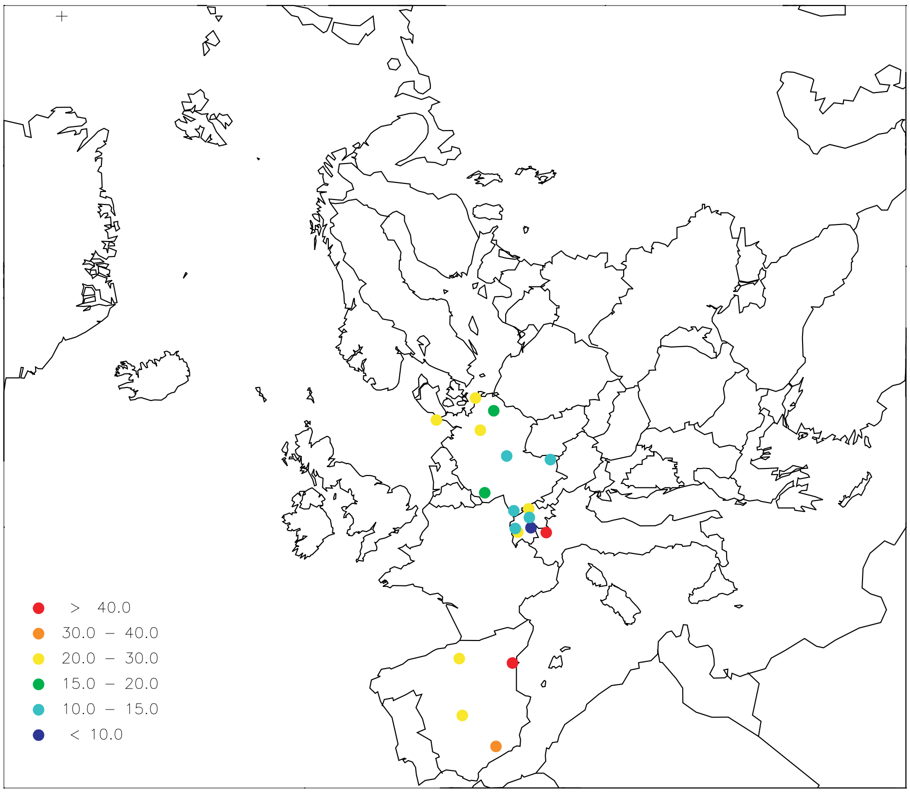

Figure 2.1: Annual averages of the PM_{10} concentrations from the EMEP monitoring Framework for 1998.

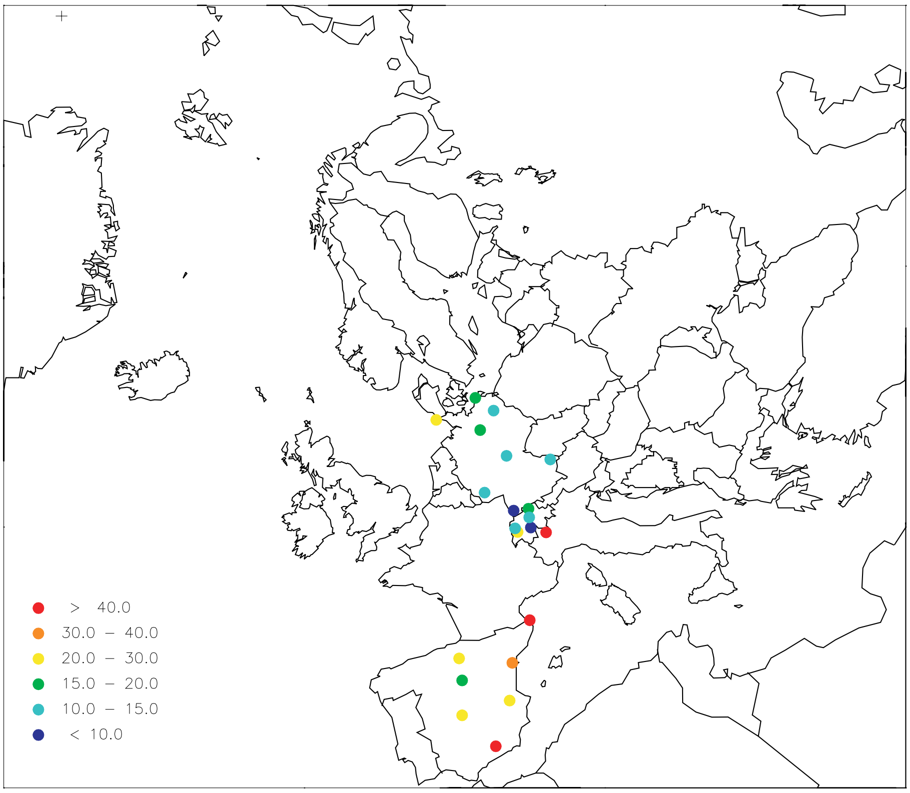

Figure 2.2: Annual averages of the PM_{10} concentrations from the EMEP monitoring Framework for 1999.

Daily measurements of particulate matter sulphate and total nitrate and total ammonium are also performed within the EMEP framework. Figure 2.3 and Figure 2.4 show annual average concentrations for sulphate and nitrate at the monitoring stations for 1999. Sulphate is determined at the majority of EMEP sites, showing higher concentrations in central and Southern Europe with annual average values close to 1 μg S/m^3 (3 μg/m^3) whereas in northern Europe and Scandinavia annual average values close to 0.4 μg S/m^3 (1.2 μg/m^3) are observed.

Nitrate is generally measured as total nitrate (sum of nitrate and nitric acid). The measurement of total nitrate ($HNO_3(g)+NO_3^-(p)$) and of ammonia and ammonium with filter packs may give biased results for the aerosol/gas partition due to chemical reactions and volatilisation of the filters. In this aspect a number of sites in Hungary, the Netherlands and Italy have started to use denuders to separate particle and gaseous components. In an EMEP pilot study held during 1992-1993, the separate gas and particle concentrations of nitrogen compounds was determined at 10 sites across Europe. Typically concentrations of the particulate phase was larger then the gaseous fraction with the exception of sites influenced by local ammonia emissions or in episodes during spring and summer. Concentration levels of total nitrate and total ammonium for 1999 are presented in EMEP/CCC 2/2001. Total nitrate annual average concentrations close to 0.4 μg N/m^3 (~1.8 μg/m^3) were measured in Scandinavia, U.K. and parts of Spain whereas in northern Germany and Denmark annual concentrations close to 1 μg N/m^3 (~ 4.4 μg /m^3) were measured.

In addition to sulphate and nitrate measurements heavy metals in aerosols are also determined in the EMEP programme. Annual average concentrations of lead and cadmium for 1998 in aerosols are shown in Figure 2.5 and Figure 2.6. The lowest concentrations of lead (below 1 ng Pb/m^3) have been observed at Svalbard and Iceland. Maximum concentrations have been measured at the Slovak stations with annual means close to 20 ng Pb/m^3. Cadmium shows a similar trend with higher concentrations at Slovak sites. A detailed description of heavy metal concentrations in Europe has recently been presented by EMEP (Berg et al., 2000).

To improve the availability of data, measurement data submitted by the European countries in the framework of the “Exchange of Information” Decision (97/101/EC) and from research programmes have been included below. Data from 1997 from both EMEP and ETC/ACC have already been reported elsewhere (Lazaridis et al., 2000) and only parts of the data are presented here.

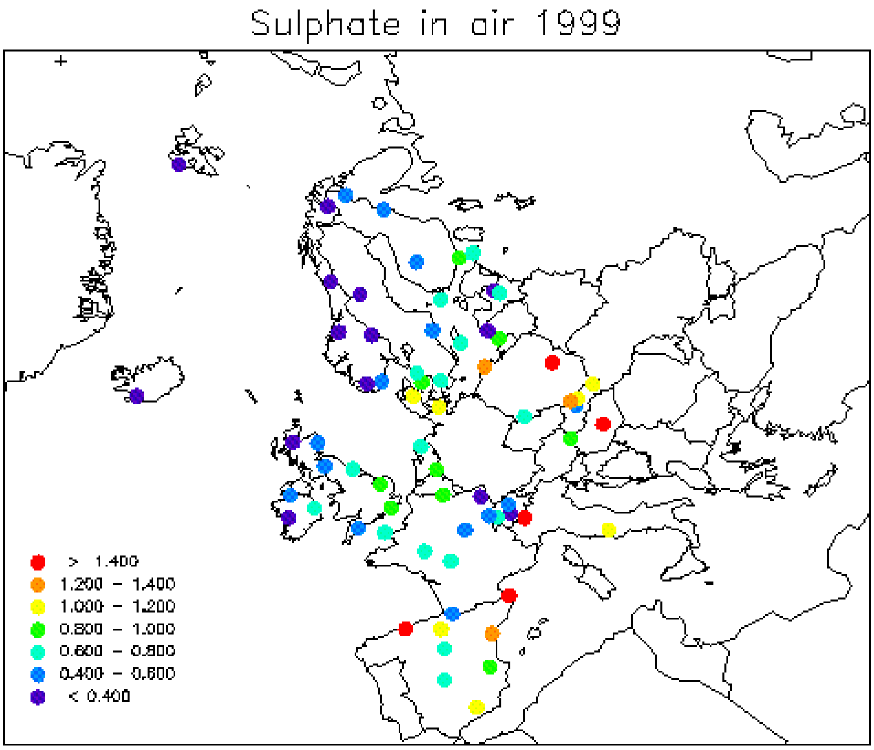

Figure 2.3: Annual averages of the sulphate concentrations (µg S/m^3) from the EMEP framework for 1999.

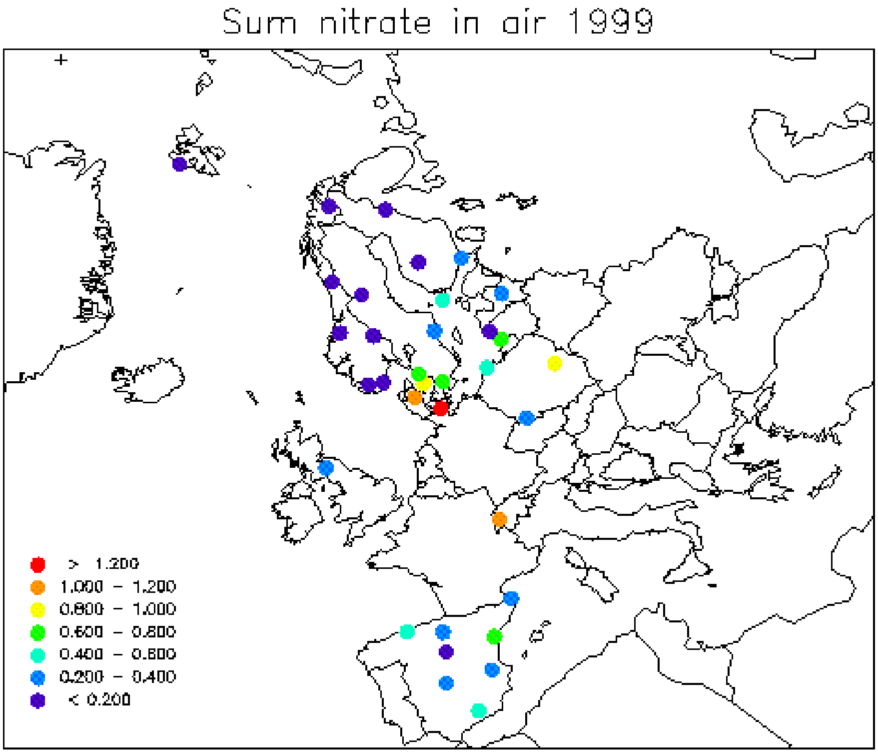

Figure 2.4: Annual averages of the total nitrate concentrations (µg N/m^3) from the EMEP framework for 1999.

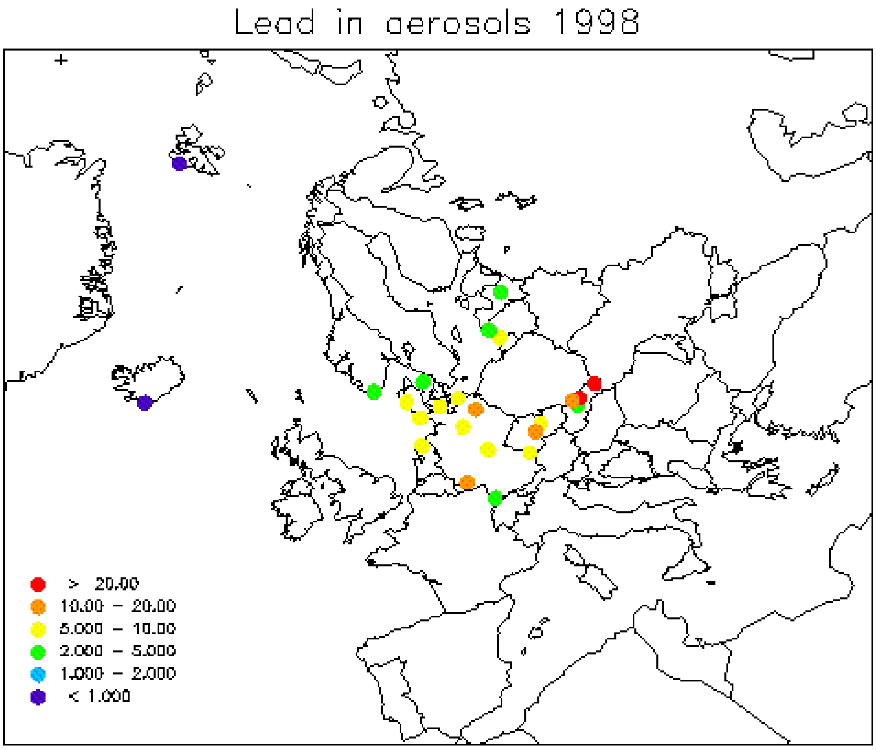

Figure 2.5: Annual average concentrations (ng/m^3) for lead in aerosols (1998).

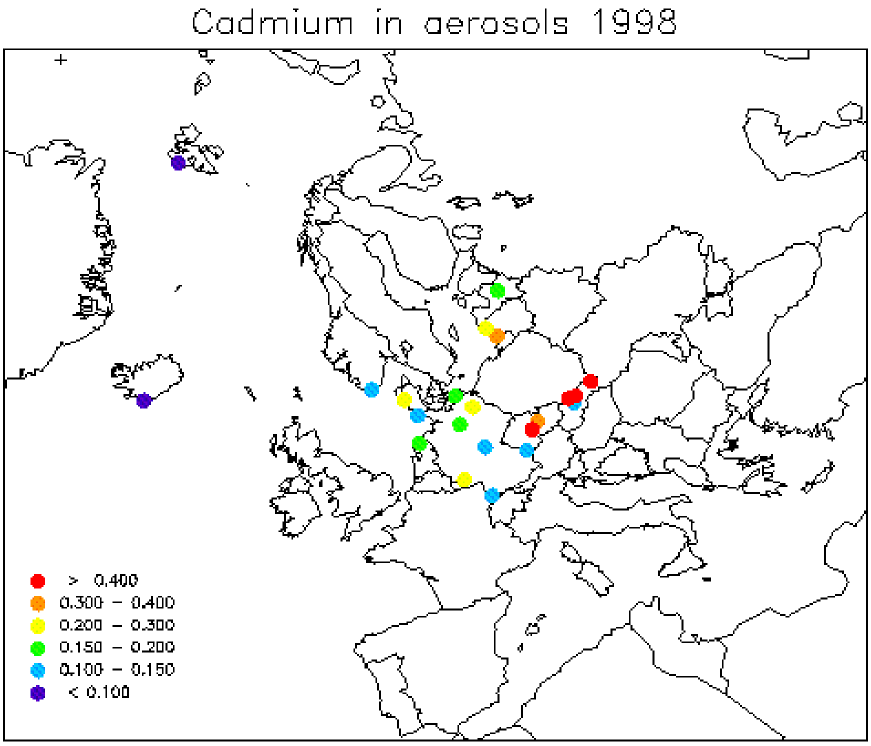

Figure 2.6: Annual average concentrations (ng/m^3) for cadmium in aerosols (1998).

Figure 2.7 and Figure 2.8 show the measured levels of PM_{10} at rural, urban and roadside sites for 1998 and 1999 respectively. Data outside the EMEP framework were obtained from the European Topic Centre under contract from the European Environmental Agency (EEA).

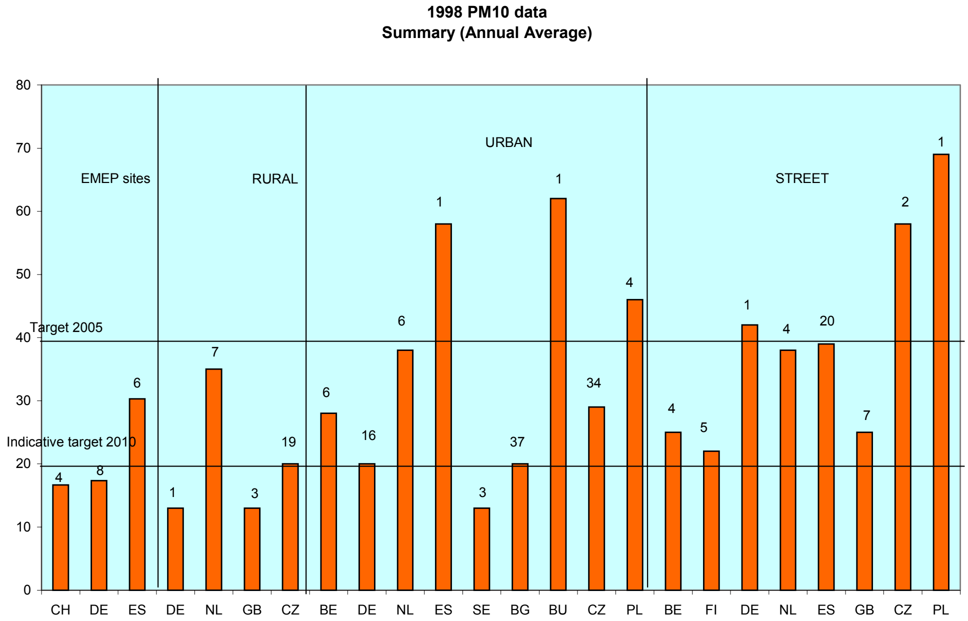

Figure 2.7: Annual averages of the PM_{10} concentrations in different European countries for various site types (units: $\mu g/m^3$) during 1998. Number of stations by country are indicated on top of each bar.

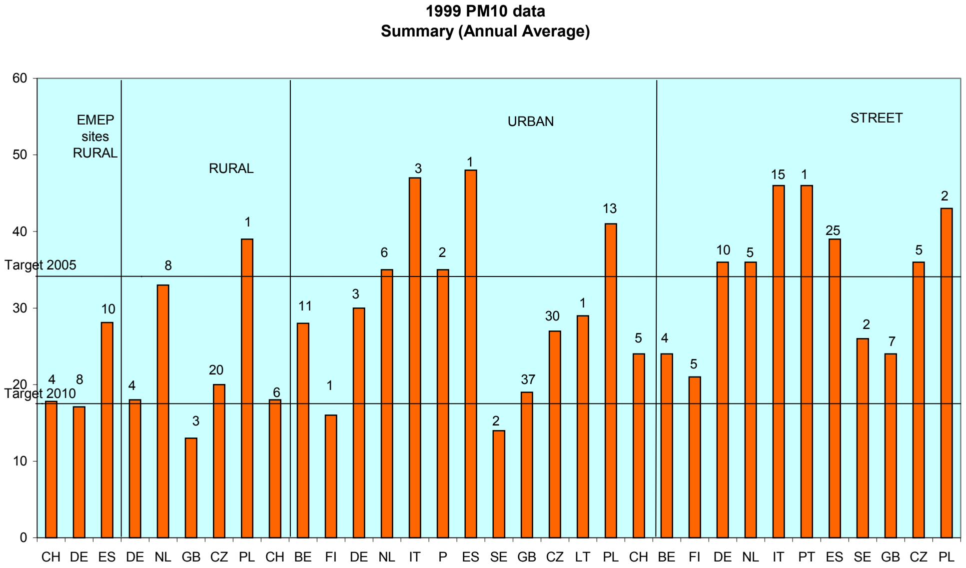

Figure 2.8: Annual averages of the PM_{10} concentrations in different European countries for various site types (units: $\mu g/m^3$) during 1999.

Different measurement methods have been used to quantify aerosol levels in different European countries and at different locations. Detailed information on the different methodologies used in the different countries has been presented by Lazaridis et al. (2000). Therefore, a comparison of data is not straightforward and only qualitative results can be obtained from this figure. In addition, for some countries the number of urban and street sites is very limited. As can be seen, the number of sites have increased from 1998 to 1999. The number of exceedances to target levels was also somewhat higher in 1999 compared to 1998.

Figure 2.9 illustrates the geographical distribution of measured particulate mass concentrations for 1998 from the ETC/ACC database system AIRBASE. Although measurements are not comparable between countries (see also Lazaridis et al., 2000) there are some interesting patterns with relatively high concentrations in the most populated areas of Europe in agreement with the data from 1997. Note that most data is TSP data and not PM_{10}. Concentrations in Scandinavia and northern Europe are lower but it is interesting to point out the higher TSP concentrations in southern countries such as Italy and Spain. It is expected that re-suspended dust contribute part of the TSP mass in these cases due to dry conditions as well as dust from Sahara.

A more detailed picture of the annual average concentration for PM_{10} measurements at rural sites in Europe from ETC/ACC[1] is shown in Figure 2.10 and Figure 2.11 for 1998 and 1999 respectively. Compared to the EMEP network, additional data from the United Kingdom, Poland, the Netherlands and the Czech Republic are included.

It is apparent from the above measurement data and previous studies (Lazaridis et al., 2000) that more precise, harmonised and quality controlled measurements are needed to obtain a comprehensive picture of geographical and seasonal characteristics of particulate matter in Europe. Additional data with more detailed physico-chemical characterization of ambient aerosols is a necessary aspect in understanding their dynamics at urban and rural locations in Europe.

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[1] Figures 10-11 were produced by Jaroslav Fiala and Hana Livorova (Czech Hydrometeorological Office) from data obtained by the ETC/ACC (European Environmental Agency).

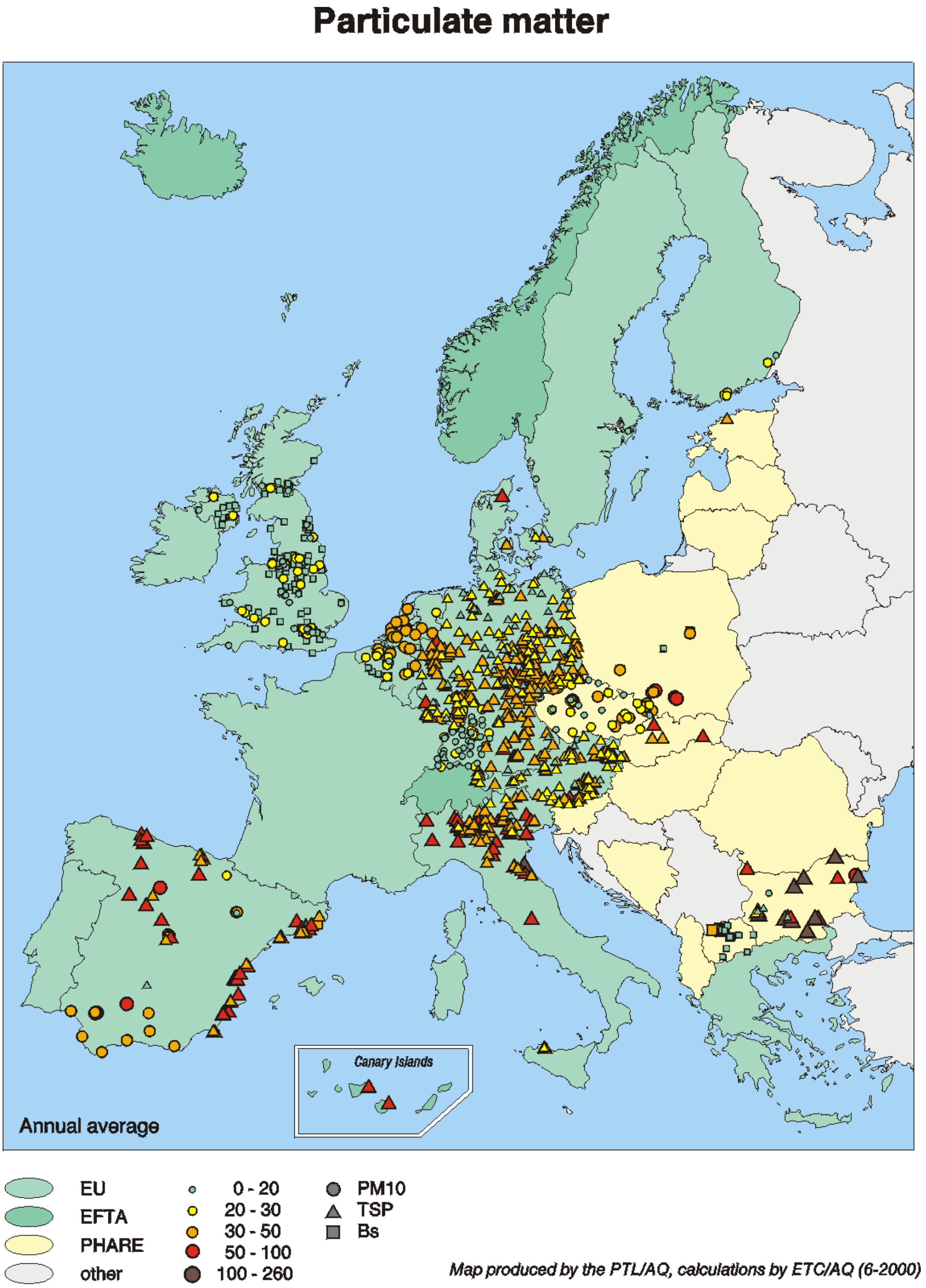


Figure 2.9: Particulate matter annual average of 24-hour concentrations (μg/m^3), 1998.

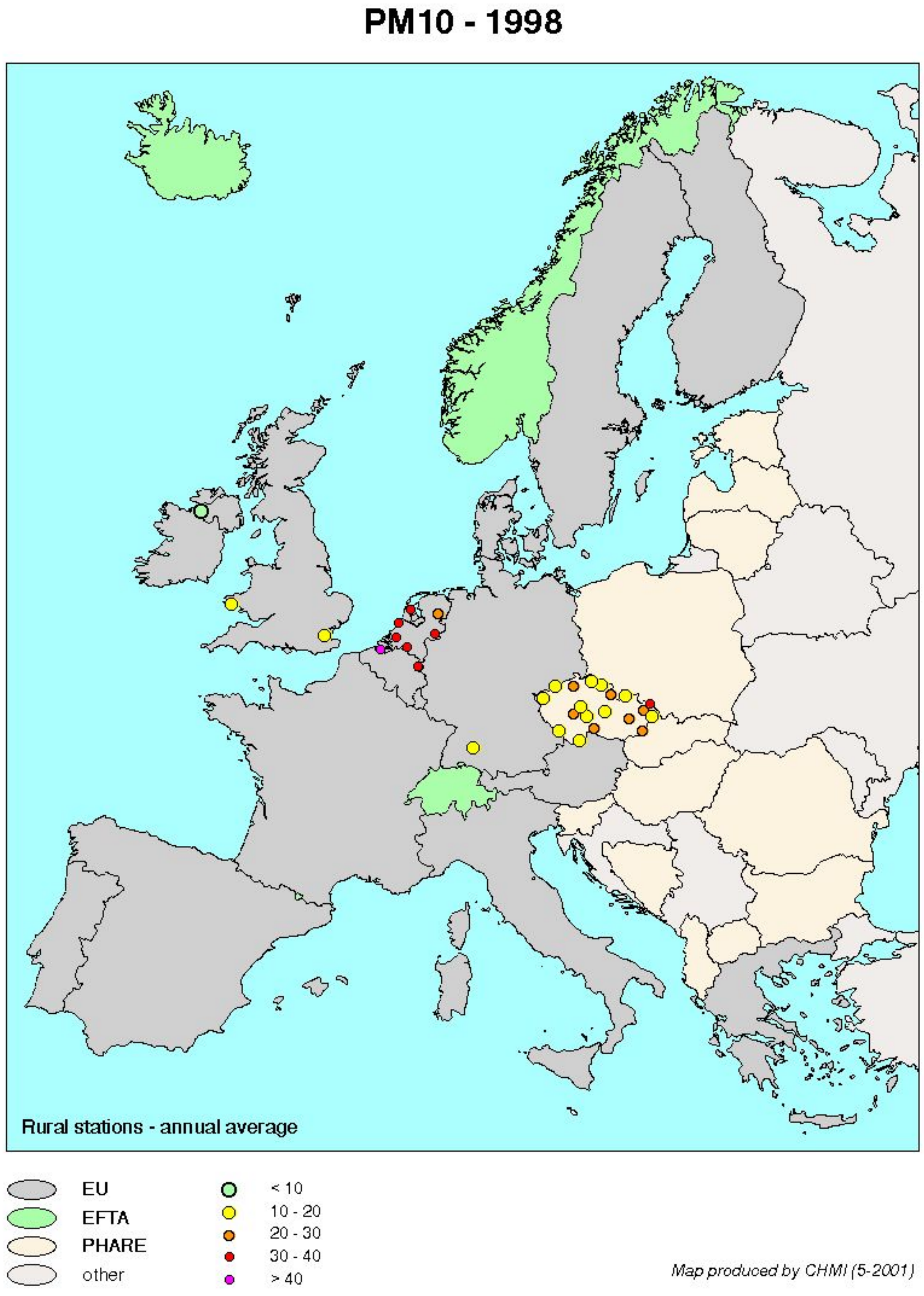


Figure 2.10: Particulate matter (PM_{10}) annual average (1998) of 24 hour concentrations from rural stations under the AIRBASE framework ($\mu g/m^3$).

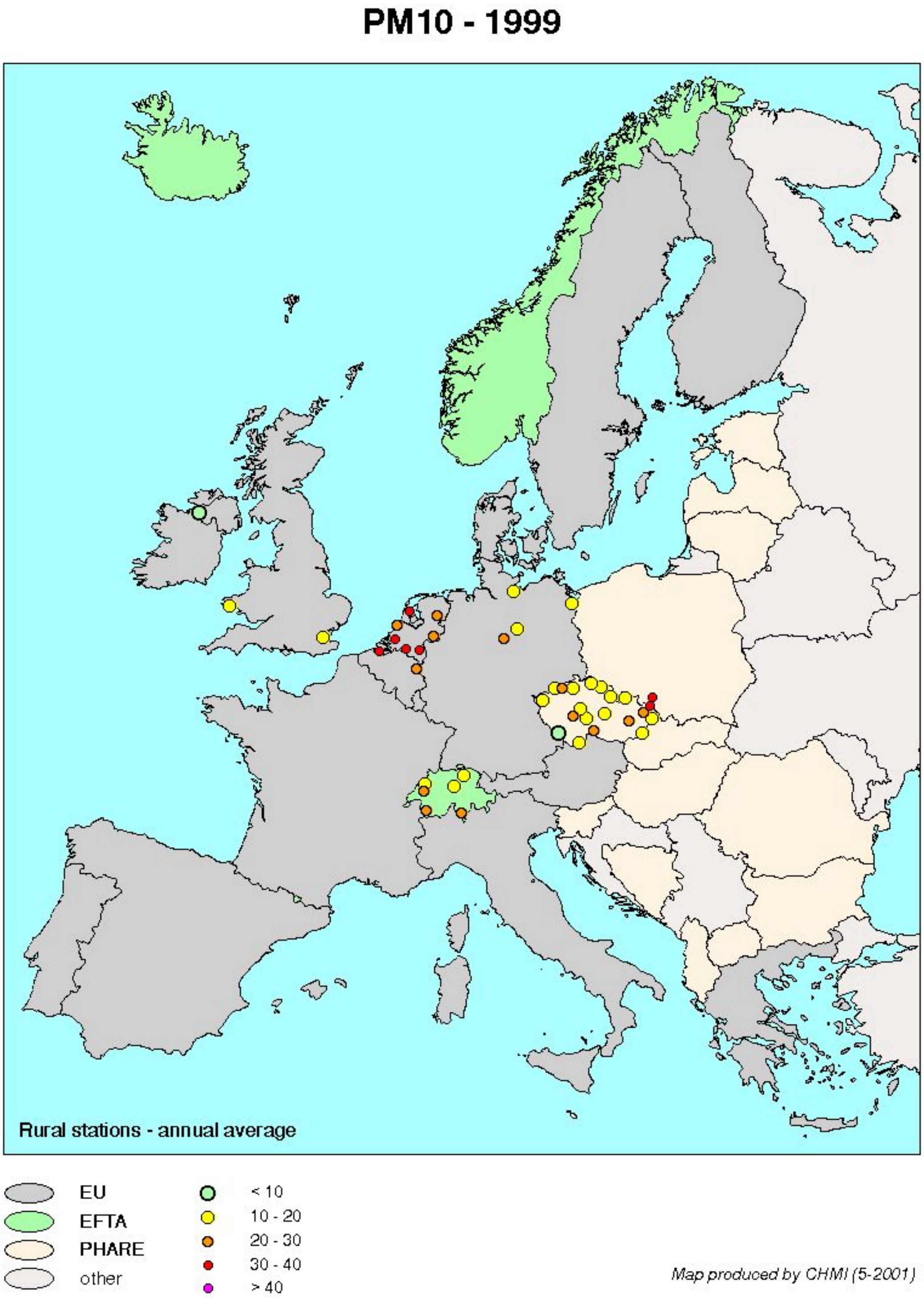


Figure 2.11: Particulate matter (PM$_{10}$) annual average (1999) of 24 hour concentrations from rural stations under the AIRBASE framework (µg/m^3).

3. Model estimation of particulate matter levels in Europe

by Svetlana Tsyro and Leonor Tarrasón

The origin of particulate matter is essential information in the design of emission reduction strategies. In particular, the natural and anthropogenic components to the anthropogenic aerosol need to be distinguished and the contribution of different anthropogenic sources needs to be identified. Model development at EMEP/MSC-W has up to now focused on the anthropogenic contribution to particulate matter concentrations and has been determined by the needs of source allocation. This chapter presents a separate analysis of the different physical and chemical properties of the aerosol relevant for policy development purposes. Three different topics are considered: 1) the need to determine the chemical composition of the aerosol, 2) the progress in inclusion of aerosol dynamic processes in the model and 3) the analysis of non-linearities in the transport of primary particular matter.

The first section of this chapter focuses on the chemical composition of the aerosol. Last year, preliminary results from the calculations of PM_{10} concentration levels were presented. These concentrations included the following components: 1) primary anthropogenic emissions of particulate matter (PPM), 2) secondary inorganic particulate material (SIA) and 3) secondary organic particulate material (SOA). Results showed that model generally underestimated the concentrations compared to measurements. This can be expected, because natural aerosols from soil erosion, re-suspension of dust from the surface, sea-spray is not included in the model. Ambient particulate mass is composed of many different components and, unless the model accounts for all aerosol components, comparison of estimated and measured concentrations of total PM mass will not be adequate to resolve inconsistencies between these. The section presents a first effort to validate the chemical composition of the aerosol and shows that the comparison of model results with observations is in this case is hampered by the limited amount of measured chemical information.

In order to validate the model, sufficient measurement data both in terms of site density, data quality and chemical/physical parameters determined, are needed. Even though a new monitoring strategy for particulate matter has recently been developed, the EMEP monitoring programme provides insufficient data. This is particularly the case for information on the chemical composition of the aerosol. Although secondary inorganic aerosol components like nitrate and ammonium (partly also sulphate) have been part of the programme for many years, only few countries report the data separating the gas and the aerosol component. In addition to being essential for the improved description with respect to acidification and eutrophication, these compounds also are needed for the further development of the PM modelling. The new monitoring strategy for PM puts requirements to the development of the monitoring programme and countries are strongly encouraged to expand their activities to meet the requirements given.

A summary of the progress in the implementation of physical aerosol dynamic processes in the Unified EMEP Eulerian model is given in the second section of this chapter. A box model version of MULTIMONO is presently under testing, and initial results from studies on the choice of time integration scheme are presented. The on going testing of the performance of the aerosol dynamics module for condensation, coagulation and nucleation is a necessary requisite prior to its implementation in the Unified EMEP Eulerian model.

In the last section, an analysis of the non-linearities in the transport of primary particulate matter is presented. It is shown that source-receptor matrices for primary particulate matter should differentiate between coarse and fine particles. This enhances the need for cost control estimates that are able to separate between fine and coarse particles, as those reported in Chapter 4. It also implies that PM emissions estimates and PM mass measurements should distinguish at least between $PM_{2.5}$ and PM_{10}.

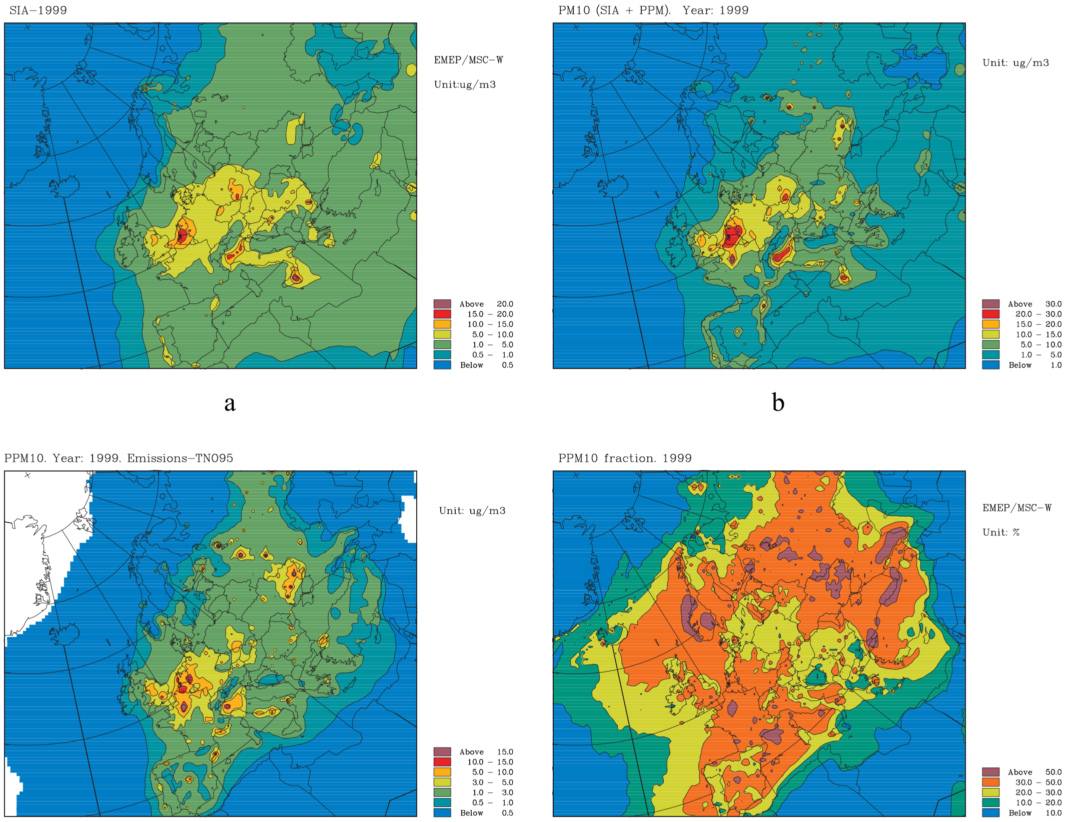
3.1 PM_{10} calculations for 1999 meteorological conditions

3.1.1 Geographical distribution of PM_{10} in Europe

Air concentrations of particulate matter PM_{10} for the meteorological conditions of 1999 have been estimated as the sum of primary particulate matter (PPM) and secondary inorganic aerosols (SIA). SIA, namely ammonium sulphate, ammonium nitrate and particulate sulphate, have been calculated with the EMEP Acid Deposition Model (EMEP/MSC-W Report 1/2001) using the latest emission update given in Vestreng (2001). The air concentrations of PPM have been calculated with the primary particulate matter version of the EMEP Eulerian model (Tsyro and Erdman, 2000, Tarrasón et al., 2000) using the 1995 emission inventory compiled by the Netherlands Organisation for applied Scientific Research (TNO). This new inventory of primary particulate matter emission in 1995 was compiled within the Co-ordinated European programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP) and has been used here only as a first estimate. Further work on the compilation of primary PM emissions is expected within the framework of the LRTAP Convention as commented further in Chapter 5.

Figure 3.1 shows the annually averaged concentrations of secondary inorganic aerosols and the concentration of PM_{10} (defined as the sum of PPM + SIA) in 1999. Compared with the year 1998, SIA concentration level in 1999 (Figure 3.1a) has slightly decreased in polluted areas of Central Europe, particularly in Germany and Poland. Annually averaged SIA concentrations are estimated to be 5-10 $\mu g/m^3$ in central and south-eastern Europe and 1-5 $\mu g/m^3$ elsewhere.

Total PM_{10} concentrations have decreased compared to the last year calculations in Central and East-European countries, the Russian Federation and Ukraine (Figure 3.1b). Maximum concentrations of 20 to 30 $\mu g/m^3$ are still found in Belgium, the Netherlands, northern Italy, and also in several smaller regions associated with large point PPM sources in e.g. Spain, Poland, Italy, Yugoslavia, Romania, Ukraine, and Russia.

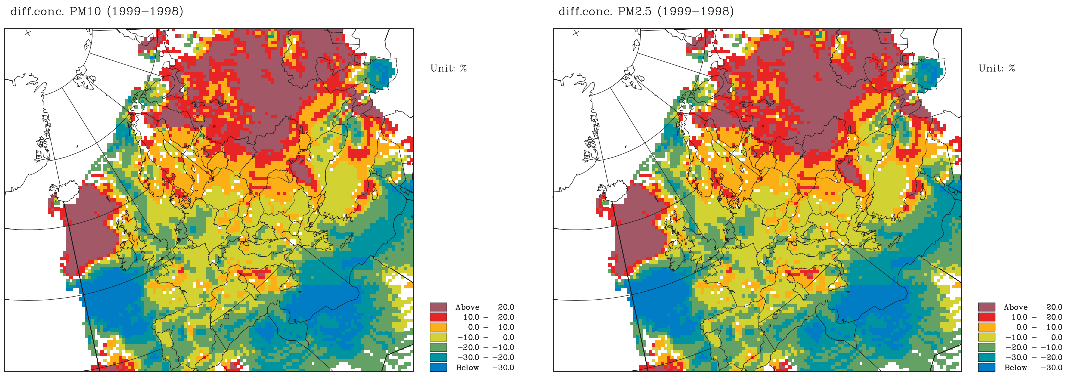


a b

c d

Figure 3.1: Modelled concentrations in 1999 of (a) secondary inorganic aerosols (particulate sulphate, ammonium sulphate and ammonium nitrate), (b) total PM_{10}, primary PM_{10} concentrations (c) and their relative contribution to the total PM_{10} (d)

Concentrations of PPM in 1999 and their relative contribution to the total PM_{10} mass has changed compared to 1998 PPM concentrations reported in 2000 (EMEP Report 5/2000) primarily due to updated emissions from the new TNO inventory (Figure 3.1c and Figure 3.1d, see also discussion in Section 3.3). The change of PPM concentrations due the meteorological variations has been studied based on comparison of calculations for the years 1998 and 1999. It should be pointed out that the differences in meteorological data for 1999 and 1998 are due to both inter-annual variability and improvement of the meteorological model. Figure 3.2 shows that fine and coarse particles respond differently to meteorological changes. As we could anticipate, coarse PPM due to its relatively local character is more sensitive to meteorological conditions close to the emission sources, while the sensitivity of $PPM_{2.5}$ concentrations to changes in meteorology increases with the distance to the sources. In general, changes in the air concentrations of both fine and coarse PPM due to meteorological inter-annual variations range within 25%.


PPM10

PPM2.5

coarse PPM

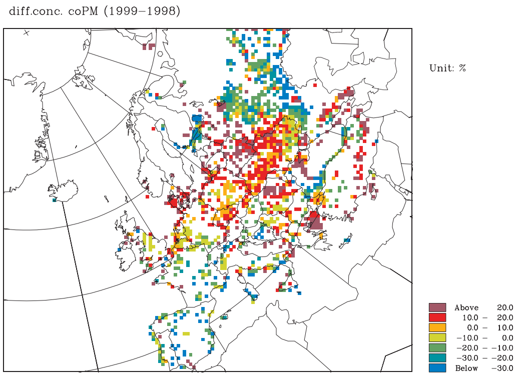


Figure 3.2: Relative difference in concentrations of PPM calculated with 1999 and 1998 meteorology.

3.1.2 Verification of modelled PM$_{10}$ with EMEP measurements

Model calculated PM$_{10}$ has been compared with concentrations of particulate matter measured in 1999 at EMEP stations. As it was implicitly mentioned above, modelled PM$_{10}$ does not include all elements of the ambient aerosol. Such aerosol components as secondary organic aerosols (SOA), natural and re-suspended anthropogenic mineral dust, sea salt and biogenic aerosols, have not been incorporated in the model. It should also be pointed out that liquid water associated with soluble fraction of aerosols has not been calculated in the present model version.

Thus, the present model results **should** necessarily **underestimate** ambient PM$_{10}$ concentrations. The relative contribution to PM mass from different sources and consequently the chemical composition of particles changes considerably at different locations across Europe. Therefore, the degree of model underestimation is expected to vary geographically depending on the relative importance of different components in the local aerosol.

Measurements of PM concentrations at 24 EMEP stations in Switzerland, Germany, Spain and Italy (EMEP/CCC Report 5/2001) have been used for evaluating model estimates of PM$_{10}$. As anticipated, calculated annual mean PM$_{10}$ concentrations are lower than measured PM$_{10}$ at all the stations (Figure 3.3) except for two German sites, Schauinsland (DE3) and Deuselbach (DE4). The comparison of measured and modelled data is also shown in Appendix 1.

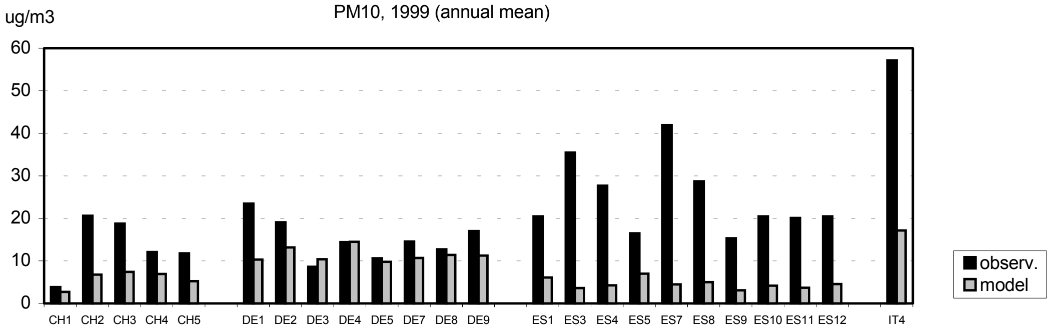


Figure 3.3: Model calculated and measured at EMEP stations annual mean concentrations of PM_{10} in 1999.

In general, the best agreement between model and monitored PM_{10} concentrations on the yearly basis is found for German stations. These stations are representative for rather polluted background air in Central Europe, where the secondary inorganic aerosols and anthropogenic PPM are expected to account for a relatively large mass of the ambient PM_{10}.

Rather large differences between measured and modelled PM_{10} are seen at Spanish stations and the Italian site IT4 (Ispra), which can partly be explained by the fact that total suspended matter (TSP), and not PM_{10}, concentrations have been reported for these sites.

A possible reason pointed out earlier for the disagreements between modelled and measured PM_{10} is that not all aerosol components are presently included in calculations. Some of the left out aerosols, e.g. coarse wind-blown dust, sea salt and biogenic aerosols, are believed to be coarse particles and therefore expected to contribute considerably to the total mass. Only a part of mineral dust, namely primarily emitted from anthropogenic sources, has been considered in the calculations. This is thought to be the main reason for large model underestimation in Spain, where Saharan dust is expected to be rather a large contributor to PM_{10} mass. Studies by Rodríguez et al. (2000) have shown that high PM_{10} and TSP episodes occur concurrently at sites up to 1000 km apart when high-dust Saharan air masses are transported over the Iberian Peninsula. During such events, mineral dust contribution to bulk PM_{10} levels in rural areas of Southern and Eastern Spain may reach up to 30 and 70%.

An evaluation of the performance of the model for different components of the aerosol is expected to shed some light on reasons for the general underestimation of modelled results against observations and broaden our understanding on the transport of atmospheric particles.

3.1.3 Verification of chemical composition

Atmospheric aerosol is a complex dynamic system, comprising particles with the whole spectrum of sizes and chemical composition. Different physical and chemical processes can be of particular relevance when behaviour of different particles is described in dispersion models. Therefore, verification against measurements of total PM_{10} concentrations alone cannot provide an explicit evaluation on how well various processes involved are represented in the model. Additional validation of individual aerosol chemical components is needed to evaluate the model performance.

Presently, the measurements compiled at the EMEP network do not provide satisfactory information on the chemical speciation of particulate matter.

As a part of the monitoring programme within EMEP for acidifying compounds, air concentrations of particulate sulphate, total nitrate and reduced nitrogen have been reported from a number of EMEP sites. As it is discussed the EMEP/MSC-W Report 1/2001, the Eulerian acid deposition model tends to underestimate sulphate, nitrate and ammonium concentrations in air. The need for a better spatial coverage of monitored data on the air concentrations of sulphate, nitrate and ammonium has been already expressed in previous reports (Tarrasón and Schaug, 1998; Schaug et al, 2001). For example, among the stations measuring $PM_{10,}$ concentrations of SO_4^{2-}, NO_3^- and NH_4^+ are monitored only at Spanish sites and one Swiss station (Payerne), but no SIA measurements have been reported for Germany. Better insight on the partitioning between gaseous and aerosol phases of nitrate, reduced nitrogen and secondary organic material is essential for an adequate characterisation of particles.

Measurements on the chemical composition of PM_{10} for 1998-99 in Switzerland have been performed within the framework of the Swiss National Air Monitoring Network (NABEL) and made available to EMEP. Figure 3.4 compares mean concentrations of PM_{10} at several cities (see Tables 4 and 7 in EMEP/CCC Report 5/2001) as well as concentrations of inorganic ions, namely SO_4^{2-}, NO_3^- and NH_4^+ with EMEP model estimates. Calculated PM_{10} concentrations are lower than measurements by approximately a factor of two at rural and city background sites, and by a factor of six at a kerbside site in the Bern city centre.

As long as possible, the comparison has been carried out also for different aerosol components averaged for the period 01.04.98-31.03.99. Figure 3.4 shows underestimation by the model of SO_4^{2-} concentrations at all sites and overestimation of NO_3^- at all but one city site, Bern, which is directly exposed to traffic emissions. Modelled and measured NH_4^+ concentrations are quite close and only overestimated by the model in Chaumont, which is a mountain site. It should be pointed out measurements tend to underestimate air concentrations of nitrate and ammonium aerosols due to the sampling losses of volatile components.

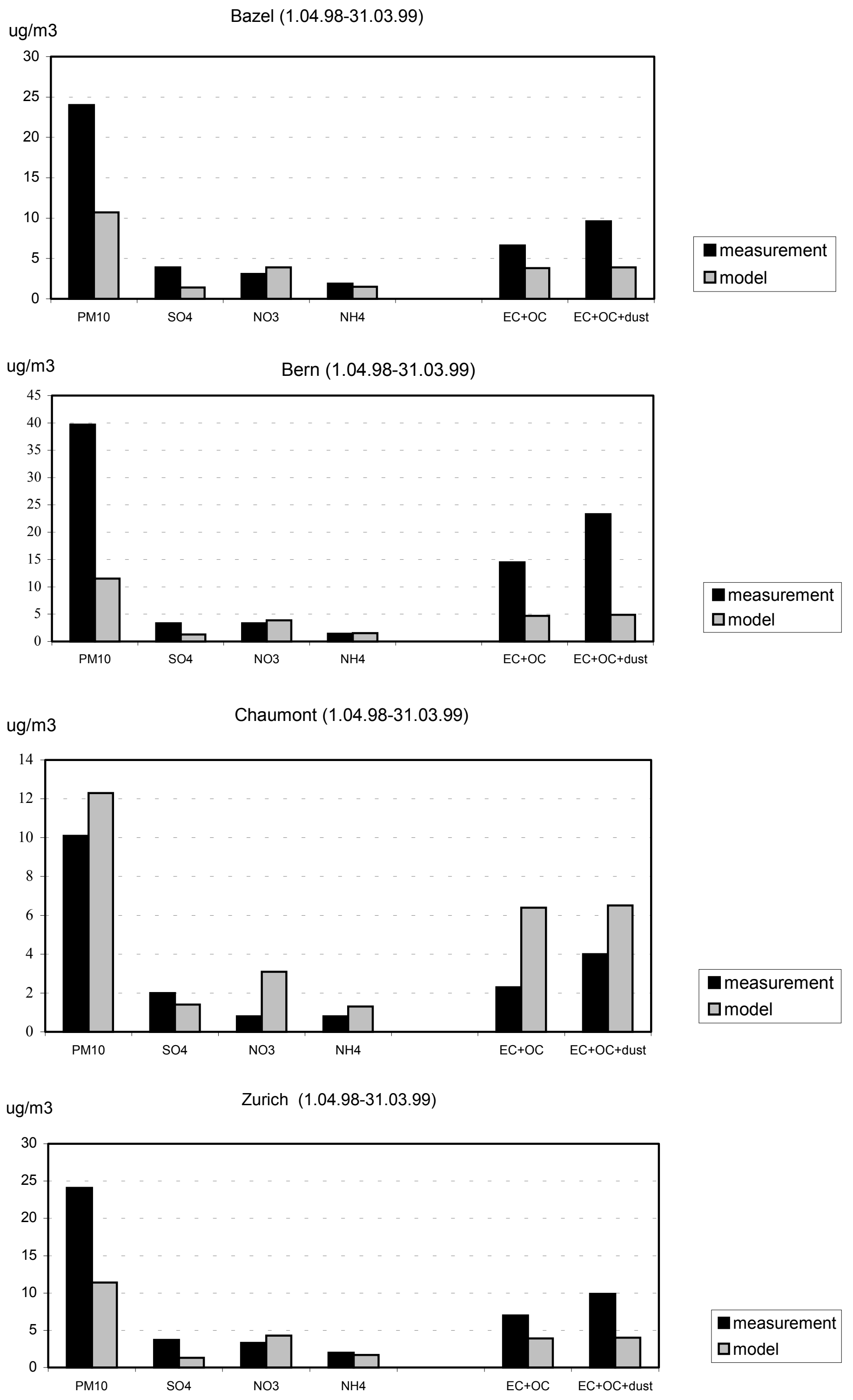
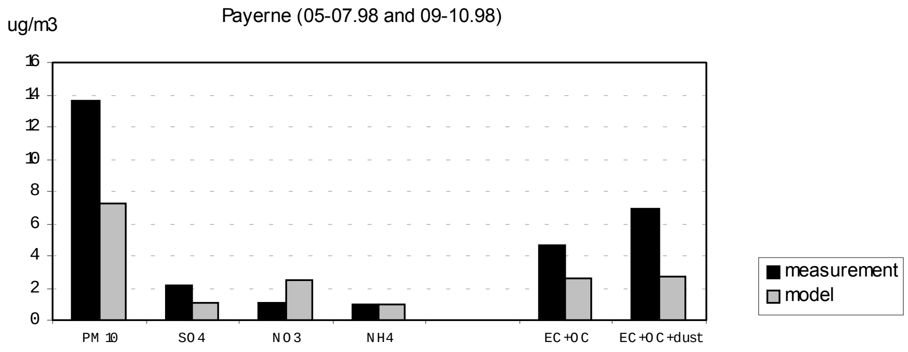


Figure 3.4: Comparison of PM_{10} concentrations and its mean chemical composition calculated with the model and measured at selected sites under 1998-99 campaign in Switzerland.
Note: *The sum of measured (EC+OC) is compared with model $PPM_{2.5}$ concentrations. Measured (EC+OM+dust), which includes both primary and secondary particles (also trace elements) is compared with calculated PPM_{10}.*



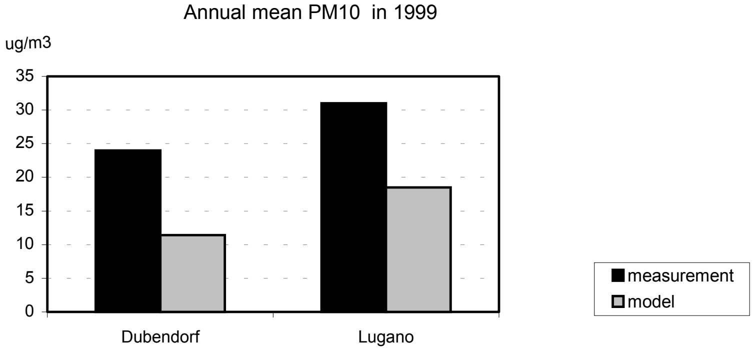


Figure 3.4, cont.

In the present calculations no chemical speciation of PPM emissions has been applied. Therefore, it is not possible to distinguish primary EC, OC, dust and inorganic aerosols in the model primary PM_{10} concentrations. However, a trivial comparison of calculated PPM_{10} with the sum of measured EC, organic material, mineral dust and trace elements (the last pair of bars in each diagram) gives an indication that primary anthropogenic aerosols are likely to be underestimated by the model. All these components, except for EC, can be both primary and secondary. To exclude the contribution in measurements from natural and re-suspended dust, which can be significant, we have also compared calculated primary $PM_{2.5}$ to the sum of measured OC and EC (next to the last pair of bars), which are fine particles. In this case, calculated $PM_{2.5}$ underestimates measurements within a factor of 2 (except at Bern kerbside station). The under-prediction by the model of PPM_{10} may partly be due to the uncertainties in PPM emissions used. Based on the comparison of PM_{10} emissions in Switzerland from the TNO inventory with the Swiss national emission inventory for the year 1995 (Kropf, 2001), TNO estimate for total PM_{10} emission is about 20% lower than the national estimate.

Finally, it should be noted that remaining discrepancies in PM_{10} concentrations between model and measurements can also be due to the fact that a number of components contributing to the ambient aerosol are unaccounted-for in the model. These are secondary organic aerosols, re-suspended mineral dust, biogenic particles, trace elements and liquid water. Mean water content of aerosols during the measurement campaign was estimated to be about 16% in the warm season and 6% in the cold season.

3.1.4 Conclusions on model validation

Despite the scarceness of measurement data, it is possible to indicate that the EMEP model calculations of particulate matter in Europe generally underestimate the observed PM_{10} concentrations. A separate analysis of the chemical composition of the aerosol has helped to shed more light on the reasons for this general underestimation. The following reasons have been discussed:

- General underestimation of secondary inorganic aerosols with the EMEP acid deposition model. This underestimation has been reported before and further measurement and modelling efforts are necessary in order to investigate the aqueous component of aerosol chemistry, the partitioning of gas and particulate nitrate and the speciation of reduced nitrogen.

- Non-accounting for several aerosol components contributing to the total PM mass, such as secondary organic aerosols, re-suspended anthropogenic and natural mineral dust, sea salt, biogenic aerosols, and liquid water.

- Uncertainties in the modelled concentrations of PPM arise to a large extent from the uncertainties in emission data. It is not presently feasible based on available measurements to carry out an appropriate validation of calculated primary PM_{10} concentrations because measurements do not provide necessary information on the origin of specific aerosol components (for example, primary vs. secondary OC, anthropogenic vs. natural dust etc.). An accurate evaluation of $PM_{2.5}$ concentrations has not been possible either because not all the measurements do presently distinguish these two size classes. Also, the acid deposition model SIA has not presently been distributed between fine and coarse modes. Integration of aerosol dynamics in the Eulerian model will facilitate calculating aerosol size distribution.

3.2 Progress with the aerosol dynamics module in EMEP

Size distribution and the number of particles are expected to affect the atmospheric transport of particulate matter (PM). Although the general patterns in the distribution of PM mass are not expected to change drastically with introduction of aerosol dynamics processes, source allocation of particles and the associated cost effectiveness results may be affected when particle size distribution is considered. Therefore, an aerosol dynamics module describing nucleation, condensation and coagulation of atmospheric particles is presently under implementation at MSC-W.

3.2.1 Brief description of MULTIMONO

MULTIMONO is the **multi**component, **mono**disperse aerosol dynamics module developed at the University of Helsinki in close co-operation with EMEP. Despite its simplicity the monodisperse approach (which assumes all particles within the same mode having the same diameter), the model has been proved to be physically sound compared with more sophisticated modal and sectional aerosol models (Pirjola et al., 1998). An essential advantage of the monodisperse approach is that it limits the number of prognostic variables to two (i.e. mass M and number N) per mode, and thus, allows for computationally efficient source allocation calculations.

MULTIMONO describes aerosol size distribution with four modes: nucleation (d_p< 0.02 μm), Aitken (0.02 <d_p< 0.1 μm), accumulation (0.1 <d_p< 2.5 μm), and coarse (d_p< 10.0 μm). Concentrations of $PM_{2.5}$ and PM_{10}, which the European air quality directives refer to, can be easily derived. The aerosol module distinguishes seven chemical components: sulphate, nitrate, ammonium, organic carbon, elemental carbon, mineral dust, and sea salt. All particles within each mode are assumed to have the same diameter and chemical composition (that is, they are assumed to be internally mixed). The aerosol water content is treated as a prognostic variable. It is calculated using empirical polynomials described in Tang and Munkewitz (1994), and a calculation scheme is adopted developed by Binkowski and Shankar (1995, see also http://www.epa.gov/asmdnerl/models3/). There are a total of 32 prognostic equations in MULTIMONO: 1 for particle number and 7 for masses of the aerosol chemical components in each of 4 modes. In fact, as indicated in Table 3.1, the number of prognostic equations can be reduced, as not all of those aerosols are present in all the modes. A mean diameter for each size bin is calculated at every time-step from the particles total mass and number.

Table 3.1: Prognostic variables in MULTIMONO: N-number and M-mass concentration.
(component present (✓) may be present (✧) in the mode)

	N	M SO4	M NO3	M NH4	M EC	M OC	M dust	M Sea salt	Water
Nucleation D< 0.02 μm	✓	✧							Diagnostic parameter
Aitken 0.02 <D< 0.1 μm	✓	✓	✓	✓	✓	✓	✧		
Accumulation 0.01 <D< 2.5 μm	✓	✓	✓	✓	✓	✓	✓	✓	
Coarse 2.5 <D< 10 μm	✓	✓	✓	✧	✧	✧	✓	✓	

A detailed description of the parameterisation of binary and ternary nucleation, condensation and coagulation processes in MULTIMONO can be found in Pirjola and Kulmala (2000).

3.2.2 Examples of test simulations with the aerosol box model

Prior to the implementation of MULTIMONO in the Unified EMEP Eulerian model, a box model version of MULTIMONO is presently tested at MSC-W. Presently, the main difference between MULTIMONO and the box model (henceforth referred to as MM32) is the time integration scheme. While most aerosol dynamic modules use analytical computationally expensive time integration routines, the implementation of aerosol dynamics in the EMEP Eulerian model needs to be computationally effective to allow for the necessary long-term dispersion calculations.

Initially, the MULTIMONO version developed at the University of Helsinki uses FORTRAN NAG libraries for the integration of the time evolution of the aerosol.

The MM32 model uses instead the two-step integration scheme (Verwer et al., 1994) to facilitate the integration to the EMEP Eulerian model. Figure 3.5 compares 24-hour simulations of Aitken and accumulation mode particle growth by coagulation alone, when using different time integration routines. The experiments have shown good agreement when comparing the integration of NAG routines with a time step of 600s (NAG600), a time step of 60s (NAG60) and when using the two-step numerical integration routine (two-step) with 60 to 600s time step. Reassuringly, the choice of the numerical integration scheme has little influence in the results. Somewhat larger differences are observed when comparing MULTIMONO and MM32 with the sectional model AEROFOR2 (Kulmala and Pirjola, 2000). These are due to the different resolution of size distribution used in the models.

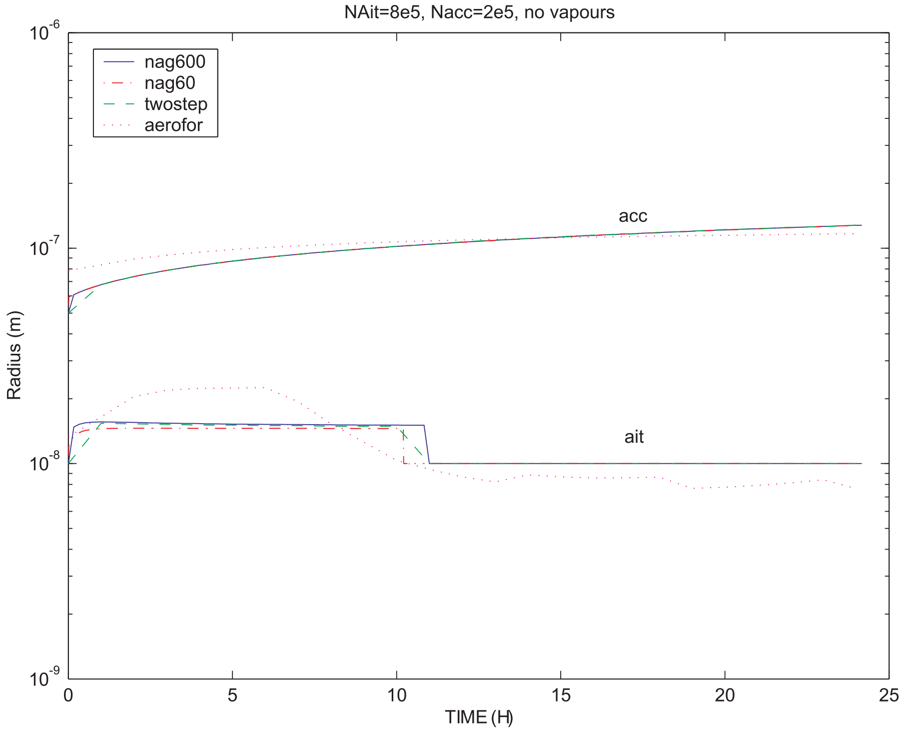


Figure 3.5: Simulation of coagulation processes using AEROFOR2 (initial Ntot=10^6 1/cm^3) and MULTIMONO with different time integration routines (see text).

Before introducing the aerosol dynamics module in the EMEP Eulerian model, a series of tests need to be carried out with MM32 to ensure that it represents correctly our present understanding of the individual aerosol dynamic processes. In the following, results from initial tests of the box model for sulphate aerosols are presented analysing its performance for the different aerosol dynamic processes.

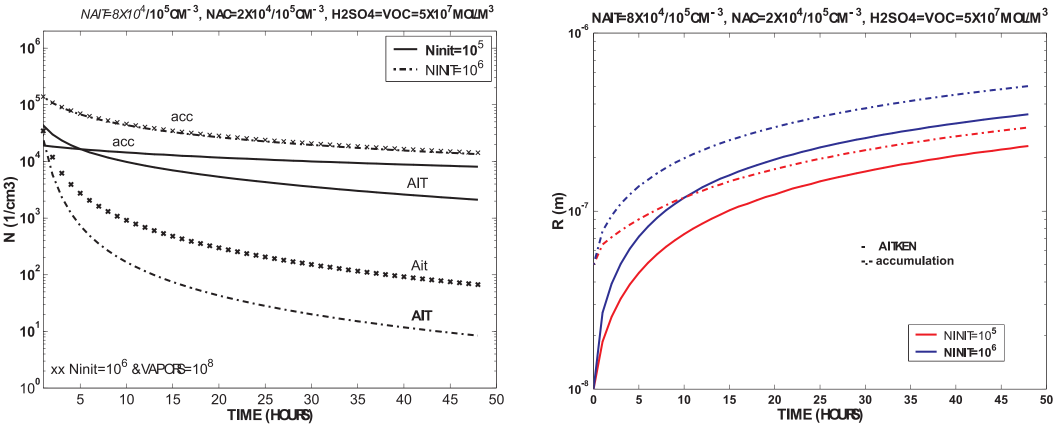
Coagulation and condensation

The box model simulation of coagulation and condensation processes for particles in the Aitken mode and in the accumulation mode is illustrated in Figure 3.6. The tests show the response of the box model to different initial number of particles

(N_{init}) and different concentrations of condensable vapours (both sulphuric acid and VOC).

As expected, an increase in the initial number particles from $N_{init}=10^5$ 1/cm^3 to $N_{init}=10^6$ 1/cm^3 results in a coagulation enhancement. This is seen from the greater decrease in particles number concentration (Figure 3.6a) as particles coagulate with each other. The process is especially pronounced for particles in the Aitken mode, which coagulate rapidly to particles in the accumulation mode. The enhanced coagulation results in particle growth and the increase in particle radius R as depicted in Figure 3.6b.

The increase in H_2SO_4 and VOC concentrations from 5·107 to 1·108 molec/cm^3 causes the faster condensational growth of particles especially in Aitken mode. This, in turn, slows down the coagulation of Aitken aerosols to accumulation mode (slower decrease in N_{ait} in Figure 3.6a).



a.

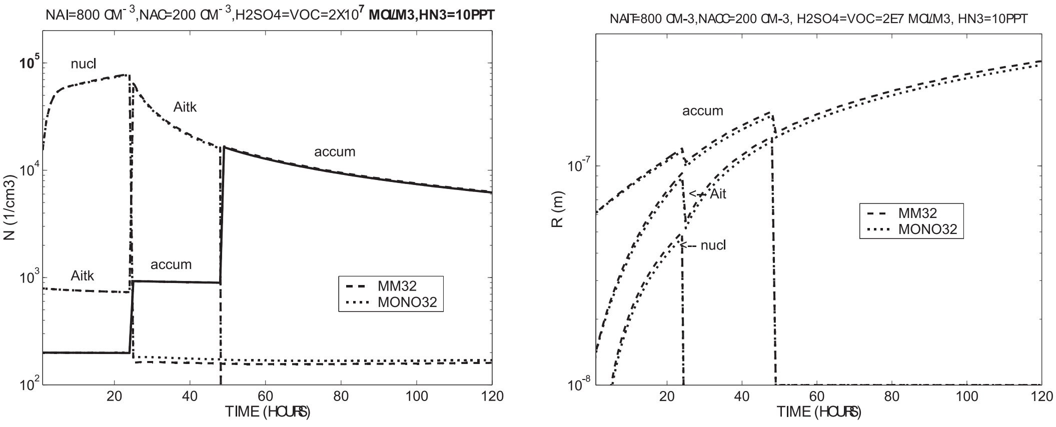
b.

Figure 3.6: Evolution of (a) particle number concentration and (b) particle radius depending on the initial number concentration and the concentration of condensable vapours.

Ternary nucleation

A preliminary parameterization of ternary nucleation processes developed by Korhonen et al. (1999) Helsinki University has been implemented in MULTIMONO. The 5-day simulation of aerosol size and number evolution presented in Figure 3.7 includes a ternary nucleation event. The calculations have been performed with both with MULTIMONO and with the MM32 model version and good agreement between the two model versions has been found.

A nucleation burst is clearly seen in a great increase of the number of particles in nucleation mode in the first hours of calculation (Figure 3.7a). The newly formed nucleation particles grow by condensation and intra-mode coagulation (Figure 3.7b) and they are moved to Aitken mode as they exceed an upper size limit. No more nucleation occurs the next 4 days because there are now too many particles (the total number has increased from 10^3 to well above 10^4 cm^{-3}) so that the most of H_2SO_4 is consumed by condensation onto existing aerosols, causing the particles growth in both Aitken and accumulation modes.



a. b.

Figure 3.7: The evolution of particles number (a) and radius (b) in case of ternary nucleation, simulated with the box model (MM32) and MULTIMONO (MONO32).
Note: *Initial aerosols number is 800 m^{-3} in Aitken and 200 cm^{-3} in accumulation mode; constant H_2SO_4 concentration of $2 \cdot 10^7$ molec·cm^{-3} and NH_3 concentration of 10 ppt.*

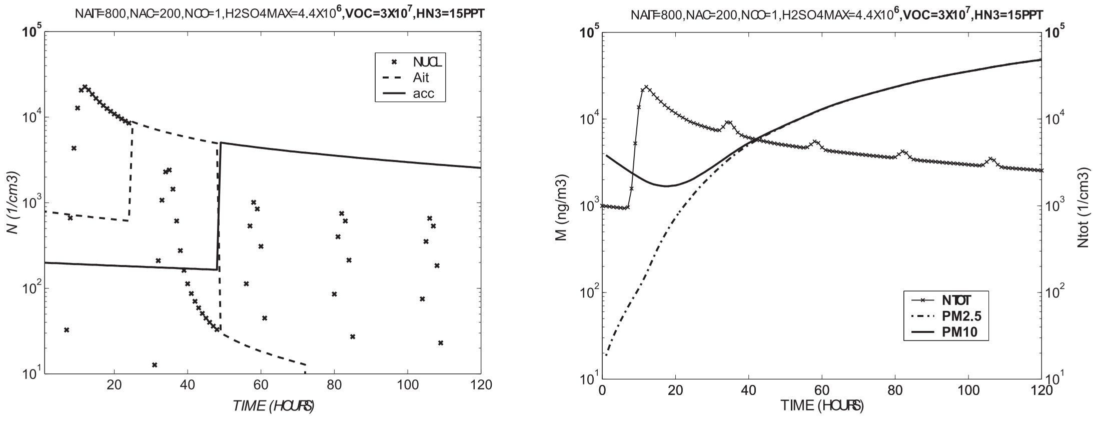
The original version of MULTIMONO has been further developed in co-operation with the University of Helsinki in order to allow for particle growth to larger modes. In an initial simple scheme tested here, all particles from a particular mode are moved to a larger one as the mean radius exceeds the threshold value. In the present simulations number and mass re-distribution has been performed once a day, at midnight, if the threshold value was exceeded. However, more frequent re-distribution of the particles may be needed in cases of their fast growth in polluted areas.

Simulation of an actual nucleation event

Figure 3.8 shows a 5-days evolution of particles number concentration calculated with MM32 using two-step integration routine. For this simulation, values for H_2SO_4, VOC and NH_3 concentrations are based on observations measured at Hyytiälä station in Finland (Kulmala et al. 2000).

An effective formation of new aerosols by nucleation is found to occur only on the first day of the simulation (Figure 3.8a). The next days, nucleation is inhibited by the smaller amount of available H_2SO_4 that then rather contributes to particle growth by condensation.

The total number of particles (Figure 3.8b) reaches the maximum at the nucleation burst and then decreases due to particle coagulation. Small mid-day increases in the total number are associated with minor nucleation events. The results obtained with the present box model version compares reasonably well with simulations and measurement data reported in Kulmala et al. (2000).



a. b.

Figure 3.8: The time evolution of (a) particles modal number and (b) total number and mass of $PM_{2.5}$ and PM_{10} in the case of a ternary nucleation event.

Note: *Initial numbers are 800 1/cm^3 in Aitken and 200 1/cm^3 in accumulation mode, sinusoidal pattern for H_2SO_4 with maximum of 4.4·106 molec/cm^3 and constant concentration of 10 ppt for NH, and 3·10^7 molec/cm^3 cm^{-3} for VOC).*

Figure 3.8b shows also the derived $PM_{2.5}$ and PM_{10} concentrations. $PM_{2.5}$ mass has been calculated as the sum of aerosol mass in the nucleation, Aitken and accumulation modes. PM_{10} is calculated as the sum of $PM_{2.5}$ and coarse aerosols mass. $PM_{2.5}$ increases mainly due to the gaining of aerosol mass through condensation of gases onto existing particles and to a minor extend, due to the production new particles. In the start of simulation, $PM_{2.5}$ concentration is relatively small and PM_{10} consists largely of coarse aerosols. PM_{10} mass is seen to decrease as the simulation evolves, most probably because of the effective removal of large particles by dry deposition. From the second day, PM_{10} mass starts to increase as the contribution of fine particles increases. By the end of simulation period, most of coarse particles have been removed by dry deposition, while fine particles have grown by condensation. Ultimately, PM_{10} mass consists mainly of $PM_{2.5}$ mass.

3.2.3 Conclusions on the implementation of aerosol dynamics

The evaluation of the aerosol dynamics box model (MM32) has primarily been carried out against the original version of MULTIMONO and the more comprehensive sectional model AEROFOR2. The main difference between MM32 and MULTIMONO is the choice of the time integration scheme. The box version of the model now successfully derives the time evolution of condensation and coagulation processes. However, there have been some disagreements in the results from MM32 and MULTIMONO compared to the full sectional model AEROFOR2. The differences, found mainly for particles in nucleation mode, concern particularly particles condensational growth and are due to the different description of particles size distribution in these models. However, because nucleation mode dominates the number concentration, but contributes negligibly to the aerosol mass, these models disagreements are not expected to affect significantly $PM_{2.5}$ and PM_{10} concentrations. Work at MSC-W is presently addressed to get a better understanding the causes of these disagreements and possibly improve the performance of MM32. This work also involves further verification of MM32 against other aerosol dynamics models and actual measurements from BIOFOR measurements campaign (Kulmala and Hämeri, 2000).

3.3 Analysis of non-linearities in the transport of Primary Particulate Matter (PPM): responses to emission changes

Fine particles ($PM_{2.5}$, or those smaller than 2.5 μm in diameter) and coarse particles (those with diameters between 2.5 and 10 μm) have very different dispersion patterns because of large differences in their wet and dry removal efficiencies. As a consequence, the analysis of the responses of PPM concentrations to emission changes should be carried out separately for $PM_{2.5}$ and coarse PM.

Primary aerosols are considered to be chemically inert in the EMEP Eulerian model and the only non-linearities that can be expected in the description of their transport are of numerical origin. As reported in Bartnicki (2000), numerical non-linearities are small in and can be neglected for source-allocation purposes. Therefore changes in the concentrations of fine and coarse particles are expected to be linear with respect to the changes in respective emissions. The response of primary PM_{10} concentrations to changes in PPM_{10} emissions will be the combined effect of the changes in emissions of $PM_{2.5}$ and coarse PM and thus NOT linear.

It should also be noted that the linear response of concentration fields for fine and coarse aerosols to emission changes holds only if the emission spatial distribution of the emissions is unchanged.

A sensitivity test to analyse the response of air concentrations of $PPM_{2.5}$ and coarse PPM to changes in PPM emissions has been carried out here. The EMEP Eulerian model has been run for the meteorological conditions of 1998 for two different emission estimates: the 1990 TNO emission estimate and the new 1995 update compiled within a framework of the Coordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP).

3.3.1 Primary PM emissions in 1990 and 1995

The 1995 distribution of the primary PM_{10} and $PM_{2.5}$ emissions in the EMEP grid as calculated by TNO is depicted in Figure 3.9.

Compared to the previous TNO estimate of primary PM emissions in 1990, the total European emissions of PM_{10} and $PM_{2.5}$ in 1995 have increased by 27% and 29% respectively (Figure 3.10). However, the changes in emissions estimated for 1990 and 1995 vary considerably between countries and source categories, and are not necessarily the same for $PM_{2.5}$ and PM_{10}. This alters the relative fractions of fine and coarse particles in the PM_{10} mass emitted and will consequently affect particle transport.

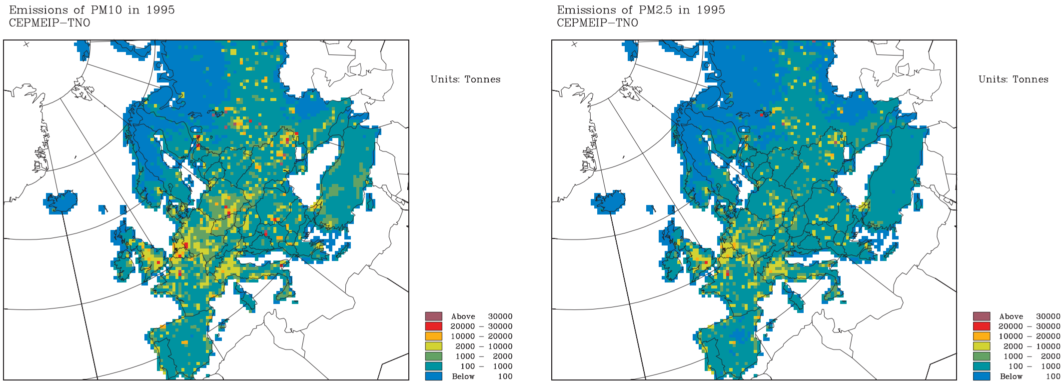


Figure 3.9: Emissions of PM_{10} and $PM_{2.5}$ in 1995. Source: TNO inventory.

The differences in 1990 and 1995 emissions of $PM_{2.5}$ and PM_{10} aggregated to SNAP1 sectors are summarised in Figure 3.10. New PM_{10} emission numbers are considerably larger in sectors 3 (Combustion in manufacturing industry), 4 (Production processes), 8 (Other mobile sources and machinery), and 9 (Waste treatment and disposal) due to more accurate and complete underlying data.

Categories 5 (Extraction & distribution of fossil fuel) and 6 (Solvent & other product use) appear for the first time in the 1995 TNO inventory and comprise mainly gas flaring and consumer-related activities. Other major updates concern sectors 2 (Non-industrial combustion plants), where more wood combustion has been accounted for, and sector 4, where new processes have been added. Furthermore, uncontrolled wood burning has been accounted for in sector 9 and also other activities besides animal farming have been included in sector 10 (Agriculture).

The most significant differences between the two PPM emission estimates are found for $PM_{2.5.}$ The two sectors with larger differences are combustion plants and transport. In the new 1995 estimates, a much larger fraction of emissions from power plants (high point sources) are considered to be coarse particles. By contrast, most PPM emitted from transport (sectors 7 and 8) is now characterised to be fine aerosols (e.g. almost 95% of PM_{10} emission from road transport are $PM_{2.5}$). Furthermore, the fraction of fine PM has increased in the emissions from Non-industrial combustion plants and Combustion in manufacturing industry (sectors 2 and 9), while emissions from Production processes (sector 4) have now a much larger fraction of coarse particles.

These 1995 updates have resulted in the changed spatial distribution of emissions with respect to particle size and therefore affect the actual transport patterns of both PM_{10} and $PM_{2.5}$. This is expected to have consequences for the geographical distribution of air concentrations of primary particles in Europe and their relative importance in the ambient aerosol concentrations.

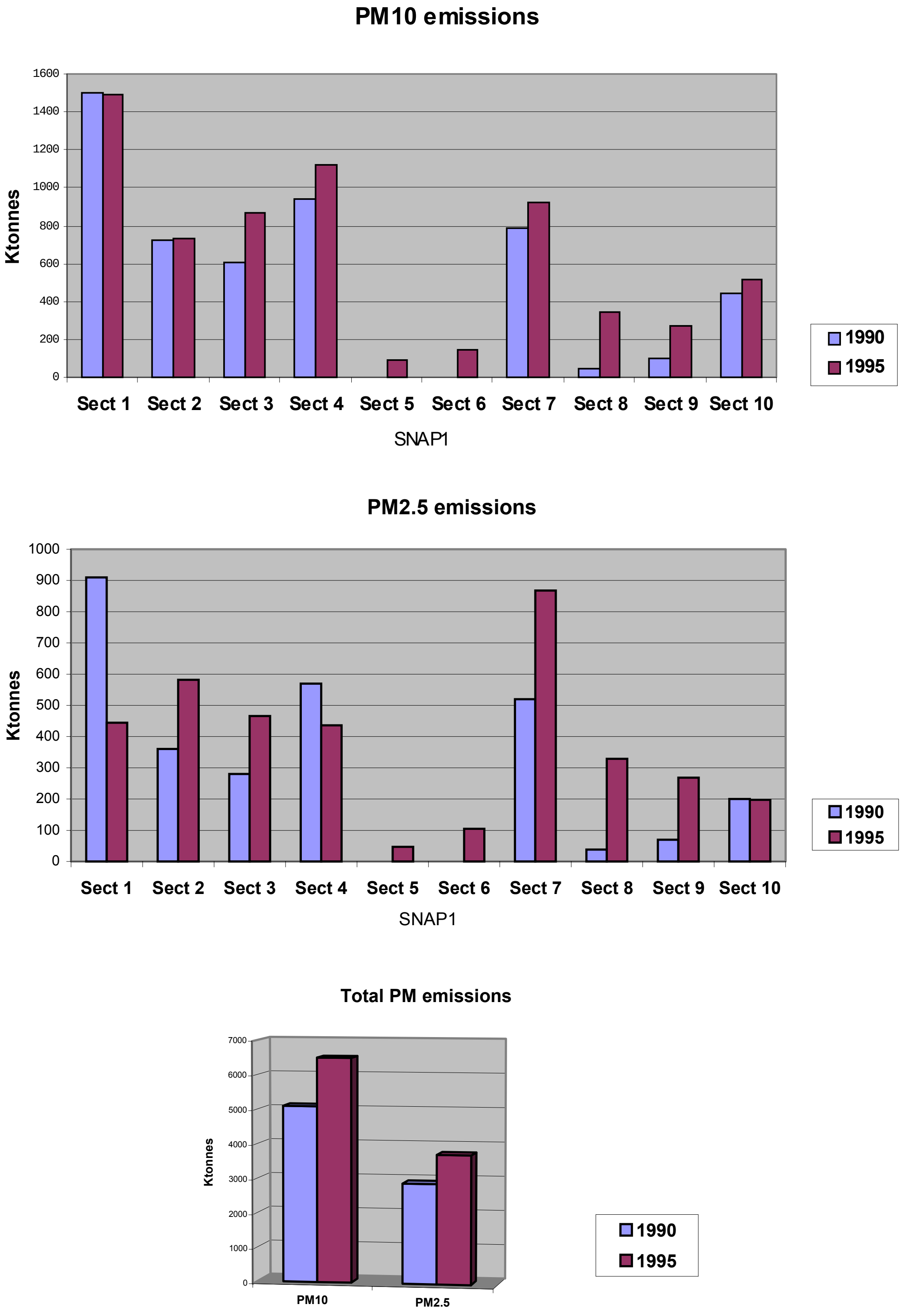


Figure 3.10: Differences in PM emissions in SNAP1 sectors in 1990 and 1995 by TNO emission inventories.

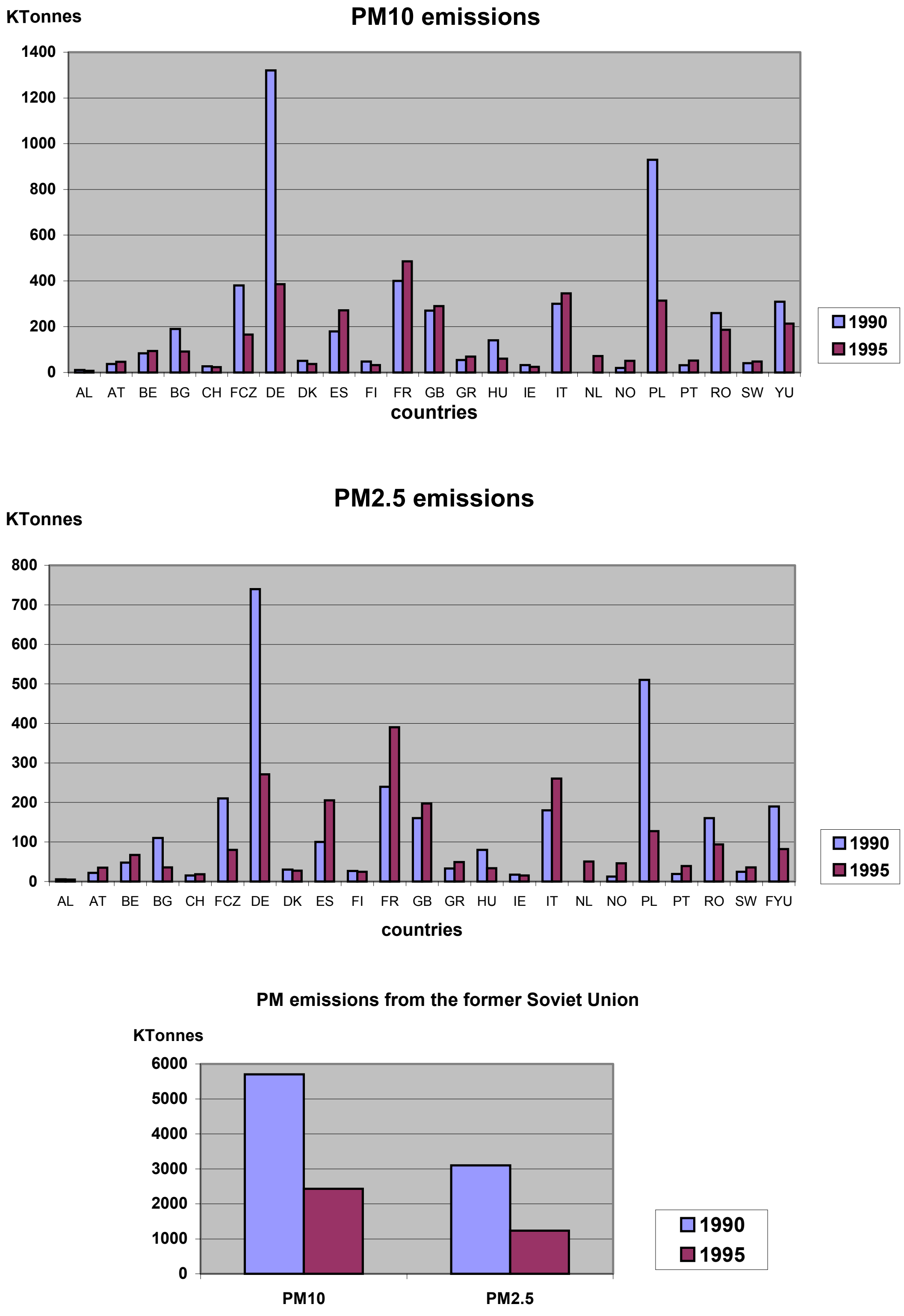


Figure 3.11: Differences in the national PM emissions in 1990 and 1995 by TNO emission inventories.

At national level (Figure 3.11), the TNO emissions estimates for 1995 are considerably smaller than the previous 1990 emissions in the most of east-European countries: Germany, Poland, former Soviet Union, former Czechoslovakia, Hungary, Bulgaria, former Yugoslavia, and Romania. This is due to both the changes in emission factors in 1995 compared to 1990 and to the decrease in activity levels. Changes in emission factors reflect actual decreases and are also a result of a more accurate methodology applied. Activity levels decrease follows lower fuel consumption in 1995. On the other hand, the 1995 total national emissions of PM_{10} and $PM_{2.5}$ for Norway, Spain, Great Britain, France, and Italy are greater than the former estimate for 1990. Figure 3.9a and Figure 3.9b showed also that a redistribution of the national emissions for some individual countries.

3.3.2 Responses to emission changes

Geographical distribution by TNO of PM_{10} and $PM_{2.5}$ emissions is different in 1990 and 1995. Therefore no strict linearity can be expected for concentration change of fine and coarse PPM calculated with 1990 and 1995 emissions. Still, a general consistency in the concentration responses to emission changes can be seen from Figure 3.12. These figures depict the relative differences between 1995 and 1990 emissions of PM_{10}, $PM_{2.5}$ and coarse PM emissions (Figure 3.12a) and relative changes in the correspondent PPM concentrations (Figure 3.12b).

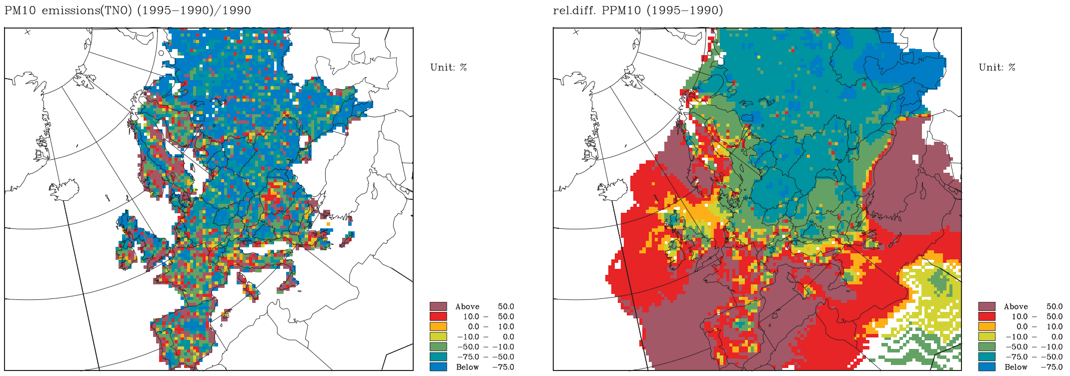
Better correspondence is found between the emission and concentration changes for coarse particles. This is because the rather short residence time of coarse PPM in the atmosphere and thus their rather local character. Changes in the emissions of coarse PPM have relatively local effect on their concentrations.

On the other hand, fine PPM can remain airborne for longer periods and be transported over longer distances away from their sources. Typically, the area of influence of changes in $PM_{2.5}$ emissions is considerably larger than for coarse PM. This is clearly seen from the Figure 3.12b, where "concentrations response" field for $PM_{2.5}$ is much smoother than the "emission changes" field.

Generally, $PPM_{2.5}$ emissions have increased in southern, western and northern Europe and decreased in Eastern Europe and Russia. Almost overall Europe a reduction of coarse PPM emissions has been estimated by TNO. As anticipated, a general increase in the air concentrations of $PPM_{2.5}$ is derived by the model in southern, western and northern Europe. $PPM_{2.5}$ air concentrations decrease in Eastern Europe and Russia in accordance with the emission changes. Overall air concentrations of coarse PPM have decreased consistently with the emissions reduction. The increase in PPM concentrations in Turkey is because PM emissions have been estimated for the first time for the country.

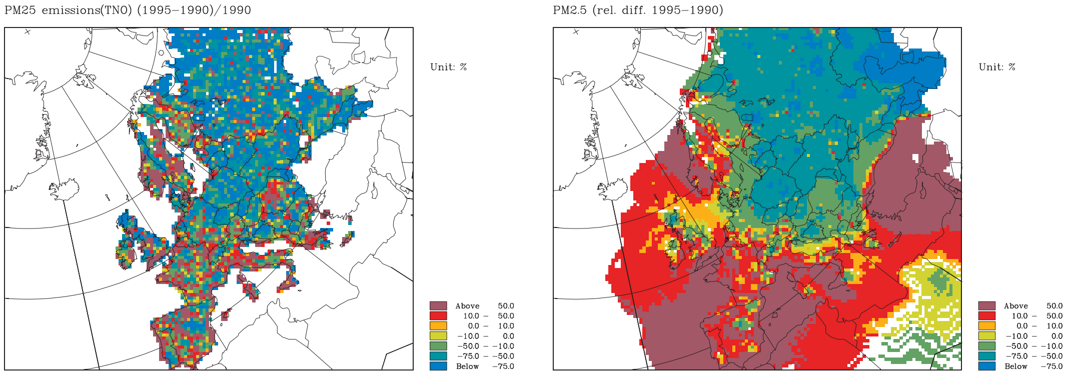
Fine aerosols have relatively long residence time, while coarse particles are very efficiently removed from the atmosphere by dry deposition. Therefore, model calculated air concentrations of coarse particles are much lower than those for fine particles. In the present calculation of primary particles, modelled $PPM_{2.5}$ concentrations have been found to be the dominating component in the total PPM10 concentrations. Therefore calculated PPM2.5 determines the response of primary PM10 concentrations to PPM10 emission changes (upper panels in Figure 3.12).

PM10


PM10 emissions(TNO) (1995–1990)/1990

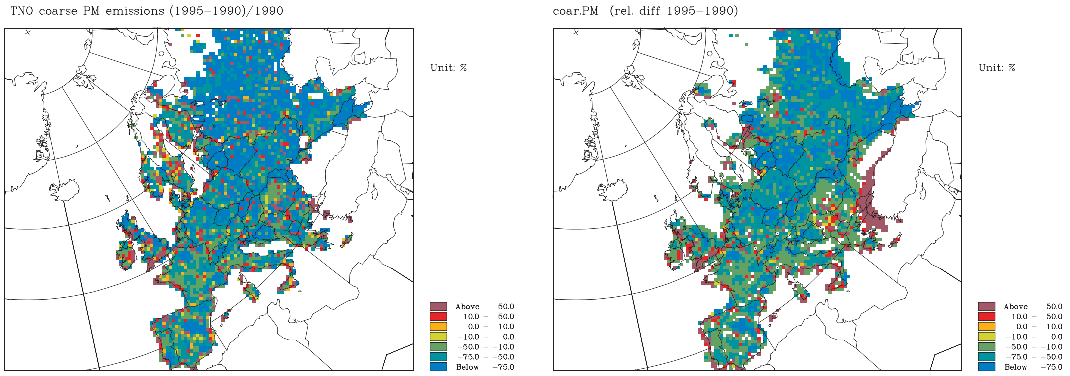
rel.diff. PPM10 (1995–1990)

PM2.5


PM25 emissions(TNO) (1995–1990)/1990

PM2.5 (rel. diff. 1995–1990)

coarse PM


TNO coarse PM emissions (1995–1990)/1990

coar.PM (rel. diff 1995–1990)

a. b.

Figure 3.12: Relative differences in: (a) 1995 and 1990 emissions by TNO and (b) concentrations calculated with TNO 1995 and 1990 emissions (meteorological conditions in 1998).

4. Potential and costs for the control of fine particulate emissions in Europe

by Janusz Cofala

This chapter reports on the progress made by IIASA in the development of a model for estimating primary emissions of particulate matter (PM) in Europe and the costs involved to reduce these emissions from the various sources in the European countries. The framework is compatible with existing approaches to estimate emissions and costs for SO_2, NO_x, NH_3 and VOC in the RAINS model. The emissions of PM are calculated for three different size classes (i) fine fraction ($PM_{2.5}$), (ii) coarse fraction (PM_{10} - $PM_{2.5}$) and (iii) large particles (PM>10 µm). Inclusion of different size classes is necessary because of different dispersion patterns and different health impacts for each type of particles.

4.1 Progress on estimating potential and cost for PM control in Europe

The model includes for each country six primary emission sectors, which are further divided into 26 secondary sectors. For every secondary sector more detailed categories are distinguished depending on fuel use, size or type of boiler etc. The model takes in to account several control options that are applicable for each emission category (e.g., electrostatic precipitators for stationary boilers, new types of engines with better emission characteristics for mobile sources). The estimates of emissions are done on a national level and include several country specific factors like structure of energy consumption and industrial output, structure of production capacities, implemented control technologies, fuel quality etc.

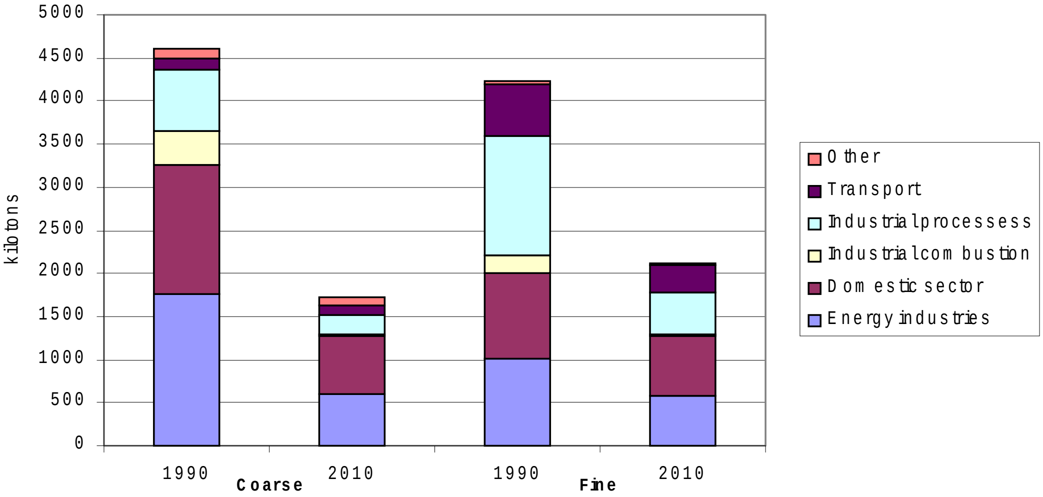


Figure 4.1: Emissions of fine and coarse particles in Europe in 1990 and in 2010.

Results of a first estimate of the PM emissions in Europe for 2010 are shown in Figure 4.1. The estimates take into account the legislation on controlling the emission sources that is currently in force in each country. Implementation of stricter emission standards and economic restructuring in Central and Eastern Europe causes that, compared with 1990, the emissions of coarse particles decrease by 63 percent. The emissions of fine particles decrease to 50 percent of the 1990 level. Also the sectoral composition of emission sources changes. For instance, the share of coarse particles emitted from the domestic sector increases from 33 percent to 40 percent. In turn, the contribution of emissions from industrial combustion decreases from 9 percent in 1990 to only 2 percent in 2010. There are important differences in the contribution of individual sectors depending on the particle size. For instance, the share of transport in total emissions of coarse fraction in 1990 was only 3 percent. This share was as high as 15 percent for the fine fraction.

The model also calculates the emission control costs for each PM size fraction. The annual costs are calculated by taking into account the technical lifetime of the abatement technologies in each emission sector. The expenditures are differentiated into: (i) investments, (ii) fixed operating costs, and (iii) variable operating costs. The calculations include common parameters for all countries (e.g., interest rate and technology-specific data such as removal efficiencies, basic investment and maintenance costs) as well as country-specific data such as average size of installations, prices for labour and electricity, and annual mileage for the various vehicle categories.

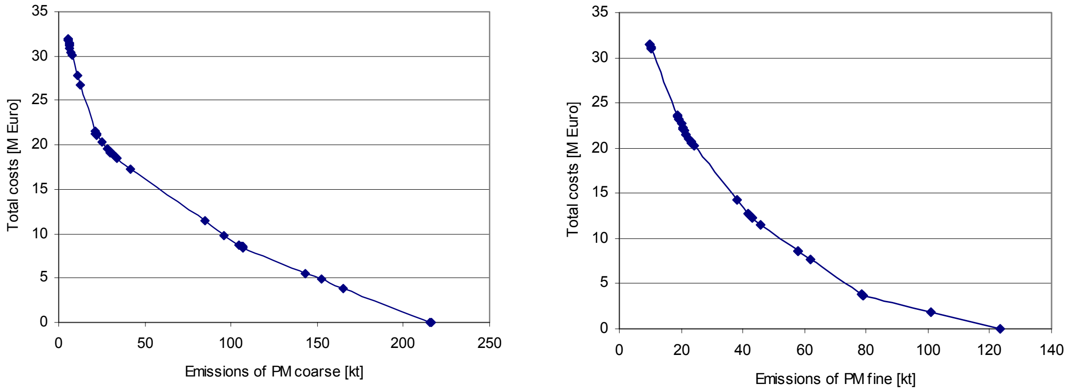


Figure 4.2: Examples of the cost curves for coarse and fine particles.

The information about costs and the reduction potential for each technology is combined into the so-called “national cost curves”. The cost curves rank all the emission control technologies in all economic sectors according to increasing marginal reduction costs. Marginal costs relate the extra costs for an additional measure to the marginal abatement of that measure (compared to the abatement of the less effective option). Figure 4.2 presents the example of such cost curves. The remaining emissions of PM are on the x-axis and the total cost on the y-axis. The cost curves are for the unabated case, i.e., they begin with “no control” emission levels. The lowest level is often referred to as maximum feasible reduction

(MFR)[2]. Comparing the example cost curves for different size fractions reveals differences, which stem from different unit reduction costs and different reduction potential for the same technology but different size fractions.

Currently available estimates must be considered provisional. For many sources of PM emissions work has started only recently and at the moment much of the available information is contradictory (e.g., for non-exhaust emissions from mobile sources, emissions from gasoline cars, industrial processes, etc.). Further work is required. RAINS PM should, therefore, be seen as an initial step towards a harmonized approach to assess PM emissions and control costs on a European scale; intense interaction with national experts will be essential to arrive at a verified database that could be used as a basis for international agreements to reduce PM pollution in Europe. To facilitate this interaction, the RAINS PM module has been implemented on the Internet (http://www.iiasa.ac.at/~rains/PM/pm-home.html). It provides free access to the input data and results.

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[2] For simplicity, the graphs show only the initial part of the cost curve with marginal abatement costs below two Euro per kg. The marginal costs for some abatement options can be two orders of magnitude higher. However, their reduction potential is rather low.

5. Assessment of primary particulate matter emissions in Europe: examples for Norway and Poland

by Anke Lükewille, Eilev Gjerald, Janina Fudala, Jozef M. Pacyna, Svetlana Tsyro and Vigdis Vestreng

By the beginning of next year, Parties to the LRTAP Convention are to provide with national emission estimates of primary particulate matter emissions for year 2000. Aware of the difficulties that such reporting requirement might impose for national experts, EMEP, in co-operation with the European Environment Agency initiated the Co-ordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP). The purpose of the project was to provide European wide documentation and guidance to the compilation of primary particulate matter emissions.

This chapter presents an example on how European wide estimates of particulate matter emissions can be useful to national experts when updating and compiling their national emission estimates. The chapter proposes a systematic comparison of the methodologies used in national inventories with the two European-wide inventories available through the co-operation in CEPMEIP: the TNO inventory and the RAINS inventory. It is important to point at the qualitative and quantitative differences between the data sets used and the basic assumptions made in the two inventories, and it is even more important to try to explain these differences.

The countries selected as examples are Norway and Poland. Norway was chosen because it has already made a national PM_{10} emission inventory for 1995, prepared by the Norwegian Pollution Control Authority (SFT). Polish emission experts contributed to the CEPMEIP project through providing the information on emission factors for source categories and industrial installations used in the country. Therefore, there is also a good basis for comparison of emission estimates in Poland.

5.1 Introduction

Fine particles, emitted directly to the air or formed from precursor substances in the air, play an important role in changing the atmosphere and affecting human health. Therefore, it is vital to obtain as complete and accurate as possible information on the sources and emissions of these pollutants to the atmosphere.

In Europe, countries have been encouraged to spend more effort on this subject, particularly in connection with international agreements on pollution reductions within the UN LRTAP and its EMEP program. Only a few European countries are now in the position to provide information on emissions of primary particulate matter (PM). Therefore, the European-wide emission inventories were prepared by groups of international experts. The aim was to provide a first insight into the possible magnitude of PM emissions in individual European countries and to encourage national experts to elaborate national emission inventories for fine particles in the countries where such data are not yet available.

A short description of emission inventories for fine particles in Europe was presented in a previous EMEP report (EMEP, 2000). The focus was placed on the 1990 emission inventory prepared by the TNO team of experts (Berdowski et al., 1996). During the recent years, research on formation, transport, behaviour and effects of fine particles has been in focus of major international and national organisations involved in the assessment of the role fine particles play in changing the atmosphere on local, regional, and global scales.

The 1990 emission inventory by TNO has been updated for 1995 within the Co-ordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP). The results of this project are now available at http://www.mep.tno.nl/emissions/, where also documentation on the emission factors and activity statistics used can be found.

Additionally, the International Institute for Applied Systems Analysis (IIASA, EMEP CIAM) has developed a tool to calculate European primary PM emissions for the reference year 1995 (http://www.iiasa.ac.at/~rains). A Particulate Matter Emissions and Costs module has been included in the RAINS model. It should be noted that the RAINS PM Module is part of an integrated assessment model, a tool to address cost-effective emission control strategies in a multi-pollutant / multi-effect framework (Amann et al., 2000). The development of the module has so far focused on the structure (e.g., consideration of all crucial input data) and the documentation of information and data used.

These two European wide inventories are well documented and can be used to support the compilation of PM emission data at national level. However, the source sector splits and the input data used in these two studies are somewhat different, making comparability of emission data difficult.

5.2 Primary particulate matter emissions in Norway in 1995

5.2.1 Comparison of the RAINS, TNO and SFT inventories

The comparison of the emission inventories is based on the information on methodologies summarized in Appendix 2. The basic approach to calculate PM emissions is the same in all three inventories:

$$\text{Emissions} = \text{Activities} * \text{Emission Factors}$$

However, the source sector aggregations, activity input data and the way of deriving emission factors might differ. This has also an influence on the assumptions made for aggregating the single sectors and activities according to the CORINAIR SNAP codes.

In the RAINS PM module first unabated emission factors are derived. Then, in a second step, control options and strategies can be chosen according to the most detailed information available in a region (country). The TNO and SFT inventories use mainly abated emission factors, partly based on measurements.

The objective of this chapter is to point at similarities, but also at differences between the results and methodologies used to estimate PM emissions in Norway in 1995. It is not the goal to try a detailed analysis of the methods applied within the RAINS, TNO and SFT studies. Such an effort can be based on the first comparison given in this report and will require more detailed documentation of the basic assumptions made in all three inventories.

5.2.2 *Methodology used for the comparison*

To compare the PM_{10}, $PM_{2.5}$ and TSP emissions calculated within the three inventories the different sources and activities were aggregated according to the CORINAIR SNAP codes (Table 5.1). Whereas the TNO inventory for the year 1995 covers all SNAP codes, some of them were not considered in the national SFT inventory and the RAINS calculation. A major source of PM emissions, non-combustion production processes, is missing in the SFT inventory. Both the SFT and the RAINS inventories do not cover emissions from SNAP05, "extraction and distribution of fossil fuels and geothermal energy".

Table 5.1: Aggregation of the RAINS, TNO and SFT results according to SNAP codes.

SNAP11		**Included in**		
	CEP MEIP source category name	**TNO**	**SFT**	**RAINS**
1 Total SNAP01	Combustion in energy & transformation industries	X	X	X
2 Total SNAP02	Non-industrial combustion plants	X	X	X
3 Total SNAP03	Combustion in manufacturing industry	X	X	X
4 Total SNAP04	Production processes	X	---	X
5 Total SNAP05	Extraction &distribution of fossil fuels and geothermal energy	X	---	---
6 Total SNAP06	Solvent and other product use	X	---	---
7 Total SNAP07	Road transport	X	X	X
8 Total SNAP08	Other mobile sources and machinery	X	X	X
9 Total SNAP09	Waste treatment and disposal	X	X	---
10 Total SNAP10	Agriculture	X		X
PM size classes	**PM_{10}**	**X**	**X**	**X**
	$PM_{2.5}$	**X**	**X** (*)	**X**
	TSP	**X**	**---**	**X**

(*) Only for combustion processes in road traffic.

Note that the national emission inventory from SFT does not include PM size distribution classes in the same level of detail as the European wide studies. The distinction of size classes (PM_{10}, $PM_{2.5}$, $PM_{1.0}$ etc.) is particularly important for the analysis of primary particulate matter transport in the air. It has also relevance concerning the evaluation of the potentials and costs of primary PM emissions control options (see Chapter 4).

5.2.3 Calculated Primary PM emissions in Norway in 1995

Comparing total PM emissions calculated by the three different inventories, SNAP05 excluded, the values are in the same order of magnitude (Table 5.2). However, looking at the single SNAP classes differences become more apparent. The first major difference is the high number given by TNO for SNAP05, “extraction and distribution of fossil fuels and geothermal energy”. This source is not considered in the RAINS and SFT calculations. According to TNO, SNAP05 increases the total Norwegian PM emissions in 1995 by 100 % (Table 5.2). 99.95% of those emissions are assumed to be in the size range < $PM_{2.5}$.

Table 5.2: Total TSP, PM_{10} and $PM_{2.5}$ Emissions in Norway according to the RAINS, TNO and SFT inventories.

Total emissions (kt) in 1995	***TSP***	***PM_{10}***	***$PM_{2.5}$***
RAINS, high emission factor for wood burning	33	27	22
TNO, excluding SNAP05	27	26	20
TNO, including SNAP05	*67*	*52*	*46*
SFT	---	24	---

The results (PM_{10} and $PM_{2.5}$) for the single SNAP codes and for the three inventories are shown in Figure 5.1 and Figure 5.2. The SFT study includes only $PM_{2.5}$ exhaust emissions for road traffic (Figure 5.2). SNAP codes 06 and 09 are not shown separately but lumped in “Other”. For the TNO inventory “Other” includes “solvent and other product use” (SNAP06) and “Waste treatment and disposal” (SNAP09). For the SFT inventory “Other” includes SNAP09 only and in RAINS “material handling” is lumped in this class.

SNAP01: Combustion in Energy and Transformation Industries (“Power Plants”)

According to all three inventories the PM_{10} emissions from power plants in Norway in 1995 were relatively low (Figure 5.1). This is due to low activities in this sector and because of the highly efficient removal technologies applied (e.g., electrostatic precipitators). While the emissions are assumed to be almost the same in the RAINS and TNO inventories (0.18 and 0.17 kt PM_{10}) they are about double as high in the national SFT study (0.36 kt). This may be due to the higher maximum emission factors used in the SFT study (Table 5.3) and/or differences in activity data. Activity data used in RAINS and by TNO are similar (see Appendix 2). For SFT detailed activities per sector were not available for this study.

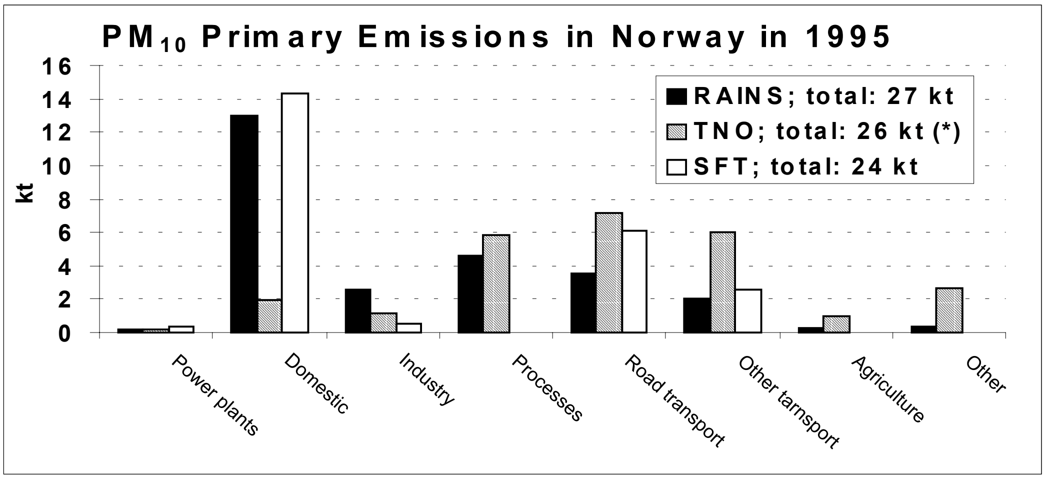

Figure 5.1: PM_{10} primary emissions in Norway in 1995: RAINS, SFT, TNO results according to SNAP codes. (*) SNAP05 excluded.

SNAP code:	**Abbreviations in figure:**
01 Combustion in energy and transformation industries	Power plants
02 Non-industrial combustion plants	Domestic
03 Combustion in manufacturing industry	Industry
04 Production processes	Processes
07 Road transport	Road transport
08 Other mobile sources and machinery	Other transport
10 Agriculture	Agriculture
---	Other

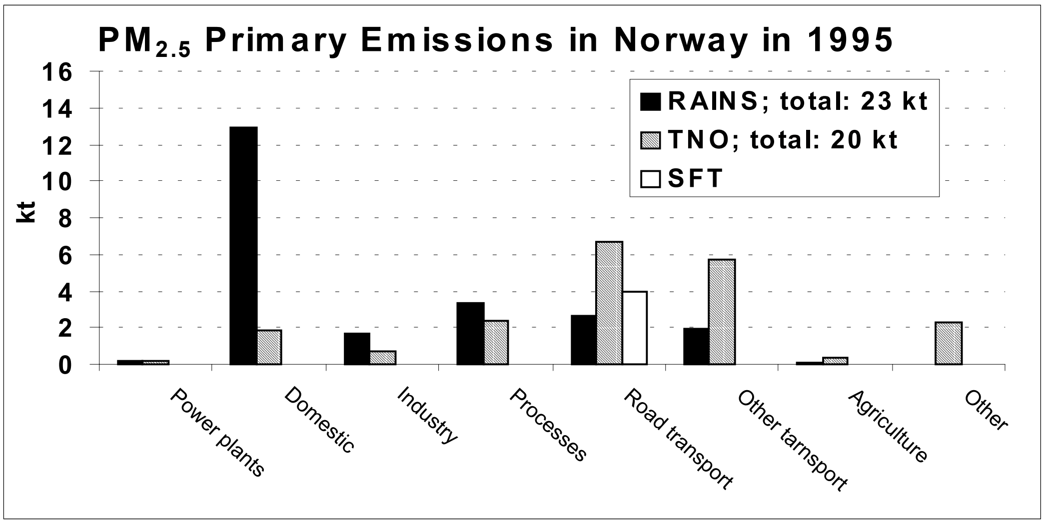

Figure 5.2: $PM_{2.5}$ primary emissions in Norway in 1995: RAINS, SFT, TNO results according to SNAP codes.

Table 5.3: Comparison of **abated** *emission factors (ranges) used for combustion in power plants and industrial boilers in Norway in 1995.*

Emission factors in kg / PJ *1000			
Inventory	**hard coal**	**biomass (waste) / wood**	**heavy fuel oil**
RAINS	8 – 130	6 – 41	14 – 19
SFT (*)	50 – 160	14	19 – 123
TNO	6 - 35	3 - 15	12 – 15

(*) Values transferred from kg / t to kg / PJ by using the fuel specific calorific values included in the RAINS database.

SNAP02: Non-industrial Combustion Plants ("Domestic")

The PM_{10} emissions in the domestic sector are almost exclusively caused by wood burning activities in ovens and fireplaces in domestic houses and cottages. According to the RAINS and SFT studies they were clearly the major source of PM_{10} (Figure 5.1) and $PM_{2.5}$ (Figure 5.2) emissions in Norway in 1995. The TNO inventory assumes much lower emission factors compared to the other two studies.

The emission factor for PM_{10} given by TNO is 95 kt/PJ * 1000, for $PM_{2.5}$ it is 95 kt/PJ * 1000. The factors are based on a report by Joaneum Research in Austria (Spitzer et al., 1998). In the European TNO inventory they were applied for sources in Norway, Sweden, Finland, Austria, Germany, Denmark and The Netherlands.

The PM_{10} and $PM_{2.5}$ emission factors used in RAINS for this study are 588 and 570 kt/PJ * 1000, respectively. They were based on Norwegian measurement data (Skreiberg et al., 1997). Even the RAINS scenario assuming that 100% of all domestic facilities are equipped with catalytic afterburners lead to higher emission factors for PM_{10} and $PM_{2.5}$ than those given by TNO (294 and 285 kt/PJ * 1000). The data for wood fuel consumption in the domestic sector in 1995 is also slightly different in the TNO and RAINS studies (22 PJ and 19 PJ, respectively).

The unit of wood burning emission factors given by SFT is kg/t. Thus, they are not directly comparable to those applied by RAINS and TNO. When using the calorific value for Norwegian wood fuel included in the RAINS database and transferring kg / t to the energy unit kt/PJ the RAINS and SFT values are nearly the same. However, according to SFT (personal communication, 2001) 39% (1381 kt) of the total wood consumed in Norway in 1995 was burnt in domestic stoves, 61% (2154 kt; wood and bark) were used in industrial boilers. In RAINS it was assumed that 46% (22 PJ) of the wood burnt in 1995 were used for domestic heating and 54% (26 PJ) were consumed by industry and power plants. According to TNO 50 % (19 PJ) of wood (waste) were burnt in industrial boilers and power plants and 50% in domestic ovens (19 PJ).

This example shows that not only emissions themselves but also basic assumptions on sectors, activities and emission factors have to be compared to allow explaining differences in results. Proper documentation of those basic assumptions is thus essential.

SNAP03: Combustion in Manufacturing Industry ("Industry")

Concerning combustion in industry the highest PM emissions were calculated by RAINS, the lowest by SFT (Figure 5.1 and Figure 5.2). The control strategy for 1995 in RAINS assumes less efficient control of PM emissions in industrial boilers compared to power plants. 40 % of all facilities in 1995 were equipped with electrostatic precipitators (ESP) having $PM_{2.5}$ removal efficiencies of 96%. 60% of all sources used more efficient ESPs (99% removal efficiency for $PM_{2.5}$), resulting in abated $PM_{2.5}$ emission factors for hard coal of 11 and 3 kt/PJ * 1000, respectively. TNO used an overall emission factor of 3 kt/PJ * 1000, i.e., assumed higher control of PM emissions in SNAP03 than RAINS.

Looking at the available documentation of methodologies used by SFT it could be possible that sector aggregations (SNAP01, SNAP03) are slightly different than those in the RAINS and TNO studies. Further, total PM_{10} emissions from major industrial sources such as petrol refineries are higher in the TNO inventory than in the SFT study (0.15 and 0.09 kt in 1995, respectively). The emissions due to coal burning assumed by TNO for different industrial sectors seem to be rather high and should be checked thoroughly.

SNAP04: Production Processes ("Processes")

Non-combustion emissions from industrial processing were not considered in the SFT inventory. The PM_{10} values calculated by RAINS are higher than those given by TNO (Figure 5.1), while the $PM_{2.5}$ emissions are lower (Figure 5.2). This is certainly due to differences in TSP to PM_{10} or $PM_{2.5}$ ratios, removal efficiencies assumed for different size classes or fractions, i.e., differences in abated emission factors.

Another reason is differences in sector aggregations in SNAP04. They are much more detailed within the TNO inventory than in RAINS. In the TNO study they include "storage and handling" of goods. Those activities are attributed to "Other" in RAINS (Figure 5.1 and Figure 5.2). PM_{10} emissions in RAINS, and almost all emissions from material handling were assumed to be in this size class in 1995, account for 0.35 kt.

As for boilers RAINS includes assumptions on control strategies and technologies applied in 1995. Thus, several abated emission factors per sector were calculated (see Appendix 2). Therefore a detailed comparison between TNO and RAINS results is difficult.

Cement and primary aluminium production were used as separate sectors in both the TNO and RAINS inventory. Activity data in Mt cement or aluminium produced in 1995 were not the same but similar for both inventories:

Cement production	TNO: 1.61 Mt	RAINS: 1.12 Mt
Primary aluminium production:	TNO: 0.85 Mt	RAINS: 0.94 Mt.

Comparing abated emission factors TNO obviously assumed relatively high removal efficiencies for cement production. The emission factors were 0.18 kg/t for PM_{10} and 0.08 kg/t for $PM_{2.5}$. They refer to those calculated in RAINS for most efficient removal technologies (ESP with three and more fields): 0.13 for PM_{10} and 0.12 for $PM_{2.5}$. TNO emission factors used for aluminium production were high, e.g., 2.9 kg/t for PM_{10} (1.6 kg/t in RAINS for an ESP with one field).

According to TNO aluminium production was the major source of PM emissions from processing in Norway in 1995 (2.41 kt PM_{10} and 1.09 kt $PM_{2.5}$). In RAINS PM_{10} and $PM_{2.5}$ emissions calculated for this sector were 1.59 and 0.91 kt, respectively, and pulp industry was assumed to be the major source (1.75 kt PM_{10} and 1.64 kt $PM_{2.5}$ in 1995). The TNO inventory includes a sector called "fugitive emissions from small industrial emitters" with relatively high emissions in 1995: 0.79 kt PM_{10} and 0.26 kt $PM_{2.5}$. RAINS does not consider such a source.

SNAP07: Road Transport ("Road Transport")

Exhaust and non-exhaust emissions from road traffic are most difficult to compare. The source sectors are much more aggregated in RAINS compared to the TNO and SFT studies. Further, the activity data are given in different units: kt/PJ fuel consumed in RAINS, km driven in the TNO inventory, and kg/t fuel consumed in the SFT study. Of course, the units can be converted. RAINS, e.g., includes assumptions on fuel consumption per kilometre driven to deal with the non-exhaust PM emissions (factors in g/km) or changes in fuel efficiency over time. However, to go in detail through all assumptions made in the three studies goes beyond the scope of this first comparison.

The revised Norwegian model for estimating 1995 emissions from road traffic (Flugsrud et al., 2000) seems to be the most reliable source of information and could be used as a basis for checking the data included in RAINS and the TNO study (see Table 5.4).

Table 5.4: Fuel consumption (t) and PM emissions (t) from road traffic in Norway in 1995 (Flugsrud et al., 2000).

Unit: t	Fuel consumption	PM_{10} emissions	$PM_{2.5}$ emissions
Total combustion	**2682**	**4163**	**3973**
Gasoline, total	***1603***	***539***	***539***
Passenger cars	1366	483	483
Vans and minibuses	206	53	53
Trucks	12	1	1
Buses	2	0	0
Diesel, total	***1080***	***3624***	***3434***
Passenger cars	100	382	365
Vans and minibuses	223	876	837
Trucks, light diesel	121	334	319
Trucks, medium diesel	53	175	165
Trucks, heavy diesel	392	1285	1203
Buses	191	571	543
Mopeds	7	1	1
Motorcycles	10	1	1
Road dust, total	**---**	**1943**	**---**

Using the SFT inventory as a basis, RAINS seems to underestimate total PM_{10} emissions from road traffic (Figure 5.1) while $PM_{2.5}$ emissions calculated by TNO seem to be far too high (Figure 5.2). Emission factors for non-exhaust (tyre, brake and road abrasion wear) are assumed to be much higher in the Norwegian study compared to RAINS and especially the TNO inventory.

SNAP08: Other Mobile Sources and Machinery (Other Transport)

The major PM source in this SNAP class is obviously transport by ships, contributing 1.00 kt (SFT) or 1.07 kt (RAINS) to total emissions in 1995. The TNO inventory seems to overestimate emissions in SNAP08 (again see Figure 5.1 and Figure 5.2). This could be due a mistake in fuel allocations introduced within the sector "internal navigation, inland ways" (brown coal as major fuel used). In contrary to RAINS, the TNO and the SFT inventories include air transport as a PM emission source, though PM_{10} emissions in 1995 were assumed to be relatively low: 0.03 kt according to TNO, and 0.06 kt in the SFT study.

Other (SNAP) Sources

The TNO inventory claims that gas flaring in the oil and gas industry was the major source of PM emissions in Norway in 1995, contributing with 25.82 kt to total PM_{10} emissions of 51.52 kt. Those emissions are included in SNAP05 "extraction and distribution of fossil fuels and geothermal energy" and not considered in the RAINS and SFT inventories. The TNO calculation is based on a study by Mulder et al. (1993).

Other sectors not included in the RAINS and SFT studies contribute according to TNO significantly to total PM_{10} emissions in 1995: "construction-related activities: dwellings" (0.30 kt), "construction-related activities: utilities" (0.16 kt), "commercial and residential meat frying" (0.39 kt), "commercial and residential barbecues, food heating" (0.02 kt), "tobacco smoking" (0.01 kt), and "use of fire works" (0.15 kt). Those emissions are summarized under SNAP06 "solvent and other product use".

Both TNO and SFT include information on SNAP09 "waste treatment and disposal". SFT accounts 0.01 kt for this SNAP class, TNO 1.69 kt (1.46 kt for incineration of municipal solid waste and 0.23 kt for open burning of waste). In RAINS "waste" is defined as a fuel used in the power plant sector (SNAP01). However, the activity for Norway in 1995 was assumed to be zero.

SNAP10: Agriculture

The RAINS and TNO studies give numbers for SNAP10 "agriculture". Emissions from this sector were assumed to be low in 1995 (Figure 5.1 and Figure 5.2). They are somewhat lower in the RAINS calculations. One reason is certainly that emissions from "agricultural waste burning" are not considered in RAINS but in the TNO inventory.

5.3 Comparison of the RAINS, TNO and national inventories for Poland

Polish national experts have compared their national total suspended particles (TSP) emissions estimate (1995) with the TNO and RAINS inventories for

Poland. The comparison, presented within the EMEP/ CORINAR source category split used by TNO, is shown in Table 5.5.

Table 5.5: Comparison of emission estimates for fine particulate matter, TSP, in Poland in 1995.

TSP (kt)	**TNO**	**RAINS**	**POLAND**
SNAP01 "Combustion in energy and transformation industries"	259.44	398.93	157. 00
SNAP02 "Non-industrial combustion plants"	85.00	630.08	625.00
SNAP03 "Combustion in manufacturing industry"	113.38	60.30	193.00
SNAP04 "Production processes"	98.79	103.22	490.00
SNAP05 "Extraction & distribution of fossil fuels and geothermal energy"	20.32		
SNAP06 "Solvent and other product use"	6.00		
SNAP07 "Road transport"	23.71	25.88	
SNAP08 "Other mobile sources and machinery"	2.73	11.84	
SNAP09 "Waste treatment & disposal"	2.81		
SNAP10 "Agriculture"	32.83	4.36	
Total all sectors	**645.00**	**1237.61**	**1308.00**

Total emissions estimated by national experts in Poland are close to the values given by RAINS and two times larger than the emissions calculated by TNO within the CEPMEIP project. It should be mentioned, however, that national estimates relate only to emissions from stationary sources. If the emission estimates for mobile sources are included, the national TSP emissions will be much higher than either of the numbers given by the two international emission inventories.

Major differences are noted for individual sectors, except for SNAP02 category on non-industrial combustion plants, where the RAINS and national emission estimates are quite similar.

National experts in Poland carried out no estimates for PM_{10} and $PM_{2.5}$ emissions so far. The TNO and RAINS estimates for emissions of these two fractions of particles in Poland show a tendency of RAINS data higher than the TNO data by a factor of 2 to 3, thus similar to the difference for TSP.

It is interesting to observe the differences in emission estimates for combustion of fuels in the electricity and heat production sector in Poland. This sector seems to be relatively well known with respect to the magnitude of fuels burnt, ash contents of fuels, combustion technologies and emission control installations. Thus, parameters affecting the PM emission factors and total PM emissions are well studied. Even so, emission factors given for this category vary significantly in the TNO and RAINS emission inventories. There is a need for national experts to review the emission factors for this important source category and provide more information on emission factors based on measurements. Such measurements have been carried out in major power plants in Poland, and this data should be used more extensively.

5.4 Conclusions and Recommendations

This first comparison at a national level has shown how important it is to provide a detailed documentation of the methodology used to assess primary PM emissions (e.g., on sector aggregations, economic activities, derivation of emission factors). Certainly, the information should be provided at a higher detail level than SNAP 1 to allow the Parties to the LRTAP Convention to report PM emission data by the end of 2001 in a consistent way. Results of total emissions, country totals or emissions aggregated according to SNAP codes, can only be compared and differences explained if this more detailed information is available.

For the RAINS, TNO and SFT 1995 inventories it could be shown that assumptions made on aggregations within the 10 SNAP 1 classes can differ considerably. Further, especially the TNO inventory used much more detailed sector aggregations, e.g., for road traffic and process emissions. In many cases different units have been used for both activities and emission factors. They can only be harmonized and then compared by introducing additional assumptions (e.g., on calorific values of fuels or fuel consumption per kilometre driven).

Activity data for the same sectors or sector aggregations were similar in most cases, but still different in RAINS and the TNO study. The methodology to derive emission factors is clearly documented for the RAINS PM Module. Mostly unabated emission factors are used and then certain control options and strategies are applied. The user of the module can change all those input data him-/herself, if better or more detailed information is available. The TNO inventory, and also in the SFT study, give abated emission factors. It is very difficult to figure out what kinds of assumptions were made concerning control options in the different economical sectors.

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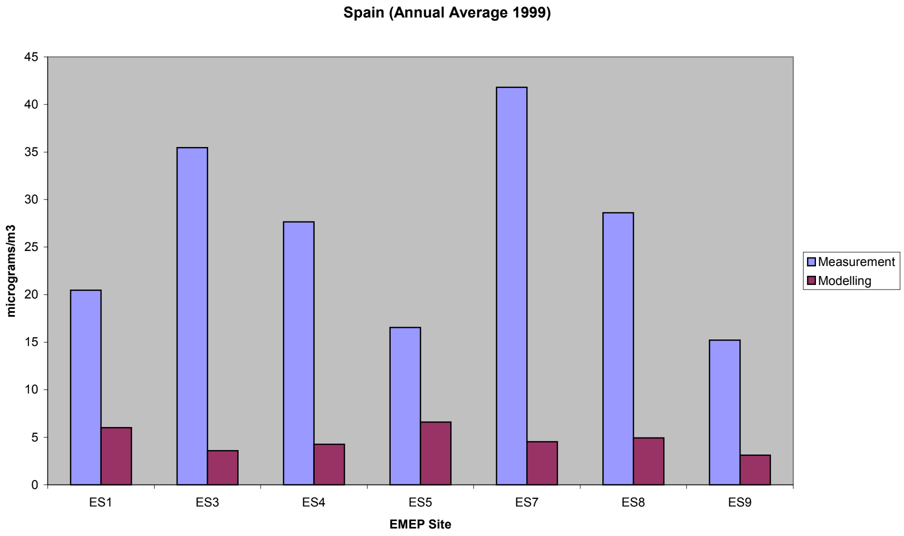
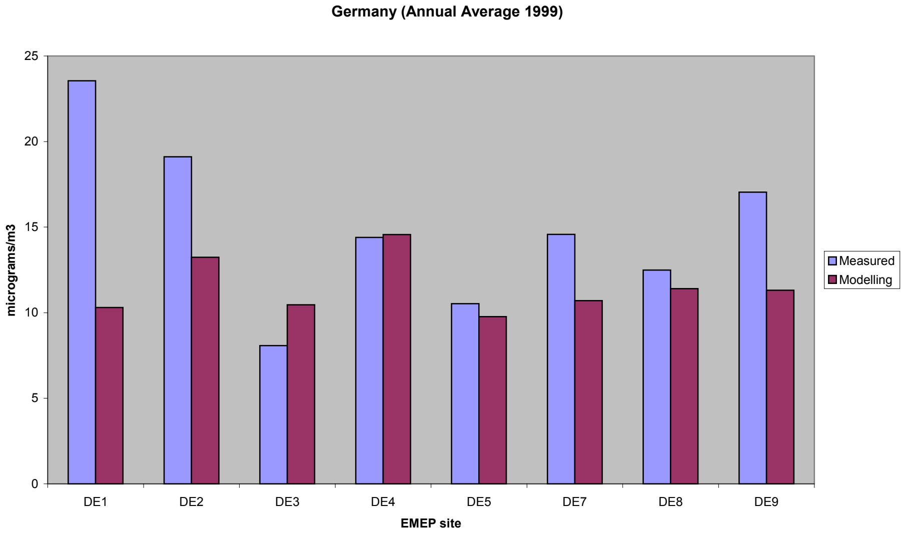
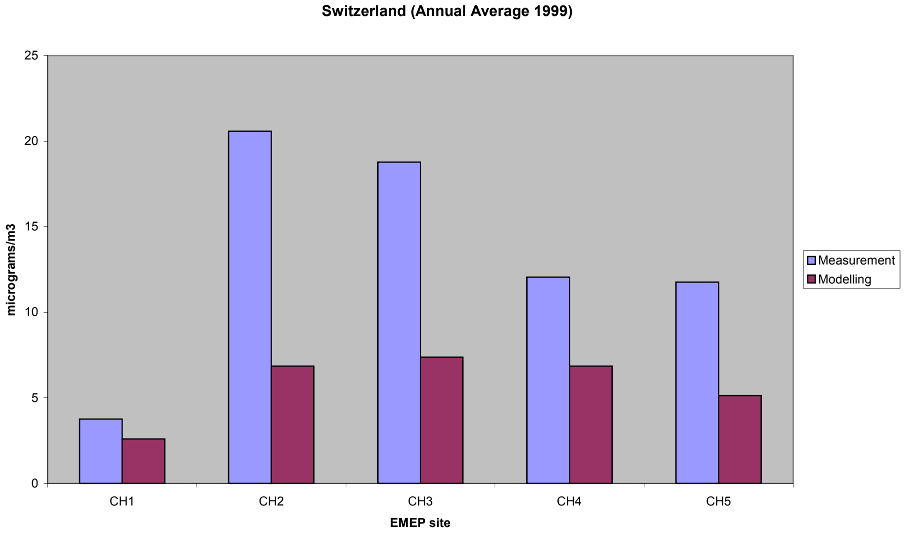
Appendix 1

Comparison between measured and modelled annual average PM values in EMEP sites (1999)


Spain (Annual Average 1999)
micrograms/m3
0
5
10
15
20
25
30
35
40
45
ES1
ES3
ES4
ES5
ES7
ES8
ES9
EMEP Site
Measurement
Modelling


Germany (Annual Average 1999)
micrograms/m3
0
5
10
15
20
25
DE1
DE2
DE3
DE4
DE5
DE7
DE8
DE9
EMEP site
Measured
Modelling


Switzerland (Annual Average 1999)
micrograms/m3
0
5
10
15
20
25
CH1
CH2
CH3
CH4
CH5
EMEP site
Measurement
Modelling

Appendix 2

Description of methodologies used for the calculation of Particulate Matter emissions in Norway

I. Methodology used in the RAINS PM emission module

The basic approach within the RAINS PM Module is to calculate ***unabated*** PM emission factors for TSP, PM_{10}, $PM_{2.5}$, and also for the fractions TSP minus PM_{10}, and PM_{10} minus $PM_{2.5}$ ("coarse"). Emission factors for the fractions are derived because control technologies such as electrostatic precipitators (ESP) have fraction specific removal efficiencies. Further, this allows looking at emissions of the coarse and fine (< 2.5 μm) PM fractions separately. For liquid fuels and non-combustion sources emission factors from the literature were applied.

This chapter gives only a brief description of the methodologies used within the RAINS PM module. For details we refer to the documentation by Lükewille et al. (2001) and updates on the IIASA-CIAM web page http://www.iiasa.ac.at/~rains. The April 2001 PC version of the RAINS PM module was taken as a basis to calculate Norwegian PM emissions for 1995.

Aggregated Sectors, Activities and Emission Factors

For all sectors and sub-sectors included in the RAINS PM module, TSP emission factors for solid fuels (e.g., hard coal, biomass) are computed within the PM module by applying a mass balance approach. For Norway in this study information on average ash contents and the fraction of ash retained in boilers from the literature were used. Heat (calorific) values were taken from the RAINS database:

$$ef_{TSP} = ac / hv * (1 - ar)$$

ef unabated emission factor,
ac ash content (%),
hv lower heat value,
ar ash retained in boiler (1/100).

In a second step, emissions for two size fractions, PM_{10} and $PM_{2.5}$, are calculated using the TSP estimates and typical size profiles available in the literature.

TSP, PM_{10} and $PM_{2.5}$ mass concentrations as well as PM size distribution profiles depend on the way of burning different fuels. Therefore an option to choose three boiler types was introduced for all RAINS power plant sectors, for industrial boilers and combustion conversion processes.

1. Grate combustion,
2. Fluidized bed combustion,
3. Pulverized fuel combustion.

Differences in emission factors for 1) to 3) can be addressed by changing the "retained ash" database, respectively. In RAINS pulverized fuel combustion was assumed to be the most common technology in 1995. However, in Norway combustion activities of solid fuels in power plants and industrial facilities are rather low. The highest energy production in 1995 was assigned to the hydropower sector (393 PJ), producing no PM emissions.

Three solid fuel categories were included in the RAINS database for Norway in 1995: biomass (wood; 48 PJ), derived coal (coke, briquettes; 14 PJ, and high quality hard coal (13 PJ). The use of gas was accounted to be 104 PJ in 1995 (power plants). The PM emission factors for gas combustion were assumed to be very low (Table 1).

Table 1: **Unabated** *emission factors in kt/PJ * 1000 for combustion in stationary sources in Norway in 1995. Activities in PJ (fuel consumption) are given in brackets.*

kt/PJ *1000 (PJ)	**Existing power plants**		**New power plants**		**Fuel conversion processes**	
	PM_{10}	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**
Hard coal	2359.40 (0.85)	589.90 (0.85)	2359.40 (0.15)	589.90 (0.15)		
Wood (biomass)	588.40 (3.40)	570.00 (3.40)	588.40 (3.20)	570.00 (3.20)		
Heavy fuel oil					19.30 (3.50)	18.90 (3.50)
Light fractions (*1)					1.00 (32.50)	1.00 (32.50)

kt/PJ *1000 (PJ)	**Combustion in industrial boilers**		**Other combustion, industry**		**Residential combustion**	
	PM_{10}	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**
Hard coal			2359.40 (12.35)	589.90 (12.35)		
Wood (biomass)			588.40 (19.00)	570.00 (19.00)	588.40 (22.00)	570.00 (22.00)
Heavy fuel oil			17.10 (15.58)	16.80 (15.58)		
Light fractions (*1)			1.00 (3.00)	1.00 (3.00)	1.00 (6.60)	1.00 (6.60)
Gas	0.10 (104.00)	0.10 (104.00)				
Medium distillates (*2)	2.10 (3.10)	1.90 (3.10)	2.10 (62.00)	(62.00)	1.90 (17.00)	1.60 (17.00)
Derived coal (*3)			23.10 (14.00)	(14.00)		

(*1) Light fraction: Unleaded gasoline, kerosene etc.
(*2) Medium distillates: diesel, light fuel oil.
(*3) Derived coal: coke, briquettes.

Concerning biomass burning, 22 PJ in 1995 were attributed to wood combustion in domestic ovens. 19 PJ were consumed in industrial boilers and 7 PJ in power plants. For this study, ash content for wood (biomass) burning in the domestic sector was adjusted to reach the TSP emission factors obtained in a round robin test of a Norwegian wood-burning stove (Skreiberg et al., 1997). Two scenarios were calculated, using a low and a higher emission factor (Table 2).

Table 2: *RAINS, biomass burning in the domestic sector: assumptions made for Norway, 1995.*

Parameter	**Value**	**Unit**
TSP emission factor – high, non-catalytic oven (*1)	610	kt / PJ *1000
TSP emission factor – low, catalytic oven (*1)	310	“
PM_{10} emission factor – high, non-catalytic oven (*2)	588	“
PM_{10} emission factor – low, catalytic oven (*2)	294	“
$PM_{2.5}$ emission factor – high, non-catalytic oven (*2)	570	“
$PM_{2.5}$ emission factor – low, catalytic oven (*2)	285	“
Heat (calorific) value	15.50	GJ / t
Ash content (high emission factor)	3.80	%
Ash content (low emission factor)	1.90	“
Ash retained in oven	0.75	“
Fuel consumption (activity) in 1995	0.22	PJ

(*1) Values based on measurements by Skreiberg et al. (1997).
(*2) Ratios from the literature (see Lükewille et al., 2001).

The second largest activity in the domestic sector in Norway in 1995 was the combustion of light fuel fractions such as kerosene or liquefied petrol gas (17 PJ). However, the emission factors assumed in the PM module are very low (Table 1).

Besides stationary combustion sources, industrial non-combustion processes are important PM emission sources in Norway. Table 3 summarizes the activities and emission factors used in this study. The factors usually merge emissions from different sub-sources in a certain sector (e.g., sinter production: wind box, sinter discharge; see detailed documentation by Lükewille et al., 2001).

***Table 3**: **Unabated** emission factors for primary PM emissions from industrial processes used for Norway in 1995.*

Industrial process emissions	*Activities*	*Emission factors (kg / t)*		
	(Mt)	**PM_{10}**	**$PM_{2.5}$**	**TSP**
Cement production	1.12	27.30	11.70	65.00
Sinter production	1.14	1.92	0.65	8.96
Petroleum refining	14.62	0.12	0.10	0.12
Aluminium smelters	0.94	27.26	13.16	47.00
Pulp industry	2.22	11.00	10.00	12.00
Fertilizer production	0.51	1.00	0.65	1.02

According to RAINS the fuels consumed by transport in Norway in 1995 were mainly unleaded gasoline, diesel and heavy fuel oil (Table 4). While the PM emission factors for gasoline and heavy fuel oil are average values from the literature, the diesel fuel factors were based on the Auto-Oil II study (country-specific).

Table 4: *Emission factors for primary PM emissions from traffic exhaust in Norway in 1995.*

	Activity (PJ)	Emission factor (kt / PJ *1000)	
		PM_{10}	$PM_{2.5}$
Exhaust, unleaded gasoline [(*)]			
Transport road			
Light duty cars and motorcycles, 2-stroke	0.50	111.00	94.00
Light duty cars and vans, 4-stroke	82.70	10.00	9.00
Heavy duty vehicles (trucks, buses and others)	0.08	26.00	22.00
Off-road transport			
Other mobile sources and machinery, 2-stroke	1.35	111.00	94.00
Other land-based mobile sources and machinery, 4-stroke	0.65	10.00	9.00
Exhaust, diesel	**Activity**	**PM_{10}**	**$PM_{2.5}$**
Transport road			
Light duty cars and vans, 4-stroke	8.00	116.00	104.00
Heavy duty vehicles (trucks, buses and others)	25.03	59.00	53.00
Off-road transport			
Other land-based mobile sources and machinery, 4-stroke	17.81	47.00	44.00
Maritime activities, ships			
Medium vessels	7.97	28.00	27.00
Large vessels	14.52	28.00	27.00
Exhaust, heavy fuel oil	**Activity**	**PM_{10}**	**$PM_{2.5}$**
Maritime activities, ships			
Large vessels	3.54	12.00	12.00

Another important source of PM emissions is tyre, brake and road abrasion wear in road transport. An example for emission factors used in RAINS is given in Table 5 for heavy duty vehicles. Emission factors for light duty vehicles were assumed to be ca. 50 % lower.

Table 5: *Emission factors for primary PM emissions from non-exhaust road traffic used in RAINS for Norway in 1995.*

Non-exhaust, heavy duty vehicles	Activity (PJ)	Emission factor (g / km)	
		PM_{10}	$PM_{2.5}$
Tire wear	25.30	0.400	0.020
Brake wear	25.30	0.228	0.071
Road abrasion wear	25.30	0.380	0.209

There are several other primary PM sources included in the RAINS PM module, such as agriculture and material handling. According to RAINS they are no major sources of PM emissions in Norway and are thus not further described here. They *are mainly included in the SNAP code aggregations used for comparing the RAINS, TNO and SFT inventories (see Chapter 5 in main text.).*

Control Options and Control Strategies in Norway in 1995

The choice of control options and control strategies used in this study for Norway are only first estimates (Table 6). They will have to be checked against more detailed national data, an important step before calculating reliable cost curves for PM emission abatement measures in Norway.

Table 6: *Control options included for Norway in 1995 and their efficiencies.*

Control technology / option	**Removal efficiency (%)**		
	PM$_{2.5}$	**PM$_{10}$ - PM$_{2.5}$**	**TSP - PM$_{10}$**
Electrostatic precipitator, 1 field (ESP1)	93.0	95.0	97.0
Electrostatic precipitator, 2 fields (ESP2)	96.0	99.0	99.9
Electrostatic precipitator, 3 fields (ESP_PLUS)	99.0	99.9	99.95
Cyclone (CYC)	50.0	70.0	90.0
Improved wood burning techn. (WOOD1)	50.0	50.0	50.0
EURO package 1, light duty veh. (MDEU1)	61.0	61.0	61.0
EURO package1, heavy duty, veh. (HDEU1)	77.0	77.0	77.0
Catalytic Converter (LFCC)	50.0	50.0	50.0
Good housekeeping, industry (GHIND)	30.0	30.0	30.0
Good housekeeping, domestic (GHDOM)	30.0	30.0	30.0

In the RAINS PM module emission control strategies for a certain year have to be defined. Table 7 shows the assumption made for Norway in 1995. The strategy describes how many percent of a certain sector are controlled by which of the different control technologies or options that are available in the module.

The percentages in Table 7 are estimates made during the first steps of the PM module development. They will have to be checked thoroughly by Norwegian experts.

For wood burning in the domestic sector two scenarios were introduced in this study, based on emission factors given by Skreiberg et al. (1997):

1) *All Norwegian domestic wood burning facilities are non-catalytic ovens,*
2) *All Norwegian domestic wood burning facilities are catalytic ovens (emissions are removed by 50%).*

The recent version of the PM module allows introducing three "new" types of wood burning stoves that reduce PM emissions with efficiencies of 50%, 60% and 76%.

Table 7: *RAINS control strategy for the year 1995 in Norway.*

Contr. techn. or option	Industrial combustion	Power plants	Domestic ovens	Industrial processes (*1)	Light duty vehicles	Heavy duty vehicles
ESP1	89	0	---	80	---	---
ESP2	10	40	---	19	---	---
ESP_PLUS	0	60	---	0	---	---
CYC	1	0	---	0	---	---
WOOD1(*2)	---	---	0 / 100	---	---	---
MDEU1	---	---	---	---	37	0
HDEU1	---	---	---	---	---	23
LFCC	---	---	---	---	70	0
GHIND (*3)	100	100	---	100	---	---
GHDOM (*3)	---	---	100	---	---	---
Not suitable for control	0	0	0	1	0	0

(*1) *Exception: Cement industry, 99% ESP2, 1% not suitable for control.*
(*2) *Two scenarios: 0 % control, 50 % control.*
(*3) *For liquid fuels only! Options are also fuel specific.*

Uncontrolled and Controlled PM Emissions

In Chapter 3 results of the PM inventory calculated with the RAINS PM module will be compared with the TNO and SFT studies. Table 8 gives an overview on emissions per aggregated sectors calculated by RAINS for Norway in 1995.

According to the assumptions made in RAINS 82% of the PM_{10} *emissions in Norway were controlled in 1995 (Table 8). The percentage of* $PM_{2.5}$ *emissions hold back by using different control options and strategies was slightly lower (not shown).*

The ratio of PM_{10} *emissions controlled is largest in those sectors where highly efficient removal technologies such as electrostatic precipitators (ESP) were assumed to be in place in 1995: combustion in industrial and power plants, emissions from industrial non-combustion processes.*

The major source of PM emissions in Norway in 1995 was, according to RAINS, wood burning in domestic ovens. This is even the case if it is assumed that 100% of all facilities are equipped with modern combustion technologies (here: catalytic ovens).

Process emissions are also high. However, according to the control strategy applied for 1995 it is assumed that 1% of all facilities are not suitable for control. This option was chosen because not all emission sources during processing in different industrial sectors might be covered by the emission factors used.

***Table 8**:* **Uncontrolled** *and* **controlled** *PM_{10} emissions by aggregated sectors in RAINS for Norway in 1995.*

Emissions in kt 1995	**PM_{10} uncontrolled**	**PM_{10} controlled**	***% PM_{10} controlled***	**$PM_{2.5}$ controlled**	**TSP controlled**
Conversion combustion	0.10	0.08	***20 %***	0.08	0.08
Domestic combustion (*)	12.97 [6.50]	12.97 [6.50]	***0 %*** ***[100%]***	12.57 [6.29]	13.51 [6.76]
Industrial combustion	36.69	2.58	***93 %***	1.53	3.41
Power plants	6.20	0.10	***98 %***	0.10	0.11
Road, heavy duty vehicles	1.50	1.34	***11 %***	1.21	1.40
Road, light duty vehicles	1.86	1.36	***27 %***	1.19	1.40
Off-road machinery	0.99	0.99	***0 %***	0.91	1.02
Shipping	1.07	1.07	***0 %***	1.05	1.09
Road non-exhaust emissions	0.81	0.81	***0 %***	0.29	4.02
Process emissions	85.1	4.63	***95 %***	3.38	6.08
Material handling	0.35	0.35	***0 %***	0.01	0.53
Agriculture	0.29	0.29	***0 %***	0.10	0.67
SUM	**148**	**27**	***82***	**22**	**33**

(*) In brackets [ ]: results based on low emission factor for wood burning, see Table 2.

According to the RAINS calculation PM emissions from ships, off-road machinery, material handling and agriculture were not controlled at all in 1995. The input parameters used so far have to be discussed and refined in co-operation with national experts.

II. Methodology used by TNO

The intention of the CEPMEIP project is to compile a general and consistent methodology to estimate the emissions of all known anthropogenic sources of PM in Europe and document those for further use by national emission experts. The CEPMEIP team send out a questionnaire to national experts asking them to submit any available underlying information to existing national PM emission inventories. Many countries responded by sending all kinds of useful information such as emission factors, concentration measurements in stacks and particulate size distribution information.

After reviewing all the submitted material, available TNO data and recent publications in the open literature a set of representative emission factors were derived for TSP, PM_{10} and $PM_{2.5}$ emissions. As much as possible it has been attempted to propose regional-specific emission factors. Most of the factors refer to *abated* emissions, i.e., they consider control options in place in different economic sectors.

The CEPMEIP emission factors were applied in a European PM inventory for the year 1995, using various national and international information sources for the activity data. A fully updated database of major point sources in Europe and high resolution maps of population density, land use etc. was used to prepare gridded files of the 1995 inventory. Detailed information has recently been established on the TNO web page: http://www.mep.tno.nl/emissions/.

Aggregated Sectors, Activities and Emission Factors

Source-sector aggregations within the TNO 1995 inventory are more detailed than those calculated in RAINS and the SFT study. For combustion in stationary sources activity data is given in PJ per year (consumption of different fuels; Table 9).

Activities of combustion processes related to road transport are expressed in million kilometres driven per year. Within the TNO 1995 inventory they are not only listed according to vehicle types (passenger cars, light duty vehicles, heavy duty vehicles etc.) but also split into highway, rural, urban and different EU abatement packages (pre ECE, ECE etc.). For every combination an emission factor for TSP, PM_{10} and $PM_{2.5}$ is given. Table 10 summarizes the range of emission factor of PM_{10} and $PM_{2.5}$ used per vehicle category. The tables for road traffic available while preparing this study did not include a split according to fuels (gasoline, diesel etc.).

Generally spoken emission factors in the single vehicle categories are assumed to be highest for urban areas and lowest for highways, respectively. The splits given by TNO are very detailed, and for further information we refer to the TNO web page: http://www.mep.tno.nl/emissions/.

***Table 9*:** **Abated** *emission factors in kt / PJ * 1000 for combustion in stationary sources, PM_{10} and $PM_{2.5}$. Activities in PJ (fuel consumption) are given in brackets.*

kt / PJ *1000 (PJ)	**Industrial comb. iron & steel**		**Industrial comb. non-ferrous**		**Industrial comb. chemical industry**	
	PM_{10}	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**
Hard coal	7.00	3.00			7.00	3.00
	(12.23)	(12.23)			(5.16)	(5.16)
Coke [*1]	7.00	3.00	7.00	3.00	7.00	3.00
	(8.78)	(8.78)	(0.37)	(0.37)	(1.65)	(1.65)
Wood [*2]						
Black liquor [*3]						
Waste fuels [*4]						
Diesel			2.00	2.00	2.00	2.00
			(0.76)	(0.76)	(1.43)	(1.43)
Gasoline					2.00	2.00
					(0.04)	(0.04)
Heavy oil					12.00	10.00
					(1.48)	(1.48)
Kerosene					2.00	2.00
					(0.04)	(0.04)
LPG [*5]			0.10	0.10	0.10	0.10
			(0.33)	(0.33)	(36.70)	(36.70)

kt/PJ *1000 (PJ)	**Public electricity and CHP [*7] plants**		**Autoproducer electricity [*8]**		**Residential combustion [*9]**	
	PM_{10}	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**
Hard coal	6.00	5.00			50.00	25.00
	(0.87)	(0.87)			(0.14)	(0.14)
Coke [*1]					60.00	30.00
					(0.06)	(0.06)
Wood [*2]					95.00	90.00
					(19.00)	(19.00)
Black liquor [*3]			3.00	2.50		
			(1.44)	(1.44)		
Waste fuels [*4]			13.00	10.00		
			(0.15)	(0.15)		
Diesel			2.00	2.00	5.00	5.00
			(0.30)	(0.30)	(18.56)	(18.56)
Heavy oil					50.00	40.00
					(0.12)	(0.12)
Kerosene					5.00	5.00
					(6.73)	(6.73)
LPG [*5]					0.20	0.20
					(0.14)	(0.14)
Natural gas [*6]			0.10	0.10		
			(1.13)	(1.13)		
Other gas			0.10	0.10	0.20	0.20
			(0.86)	(0.86)	(0.65)	(0.65)

kt/PJ *1000 (PJ)	**Industrial comb. paper and pulp**		**Industrial comb. other sectors**		**Petrol refineries [*10]**	
	PM_{10}	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**	**PM_{10}**	**$PM_{2.5}$**
Hard coal	7.00	3.00	35.00	20.00		
	(0.28)	(0.28)	(0.03)	(0.03)		
Black liquor [*3]	12.00	10.00	15.00	10.00		
	(12.29)	(12.29)	(5.20)	(5.20)		
Diesel	2.00	2.00	3.00	3.00		
	(0.26)	(0.26)	(7.40)	(7.40)		
Gasoline	2.00	2.00	3.00	3.00		
	(0.04)	(0.04)	(0.13)	(0.13)		
Heavy oil	12.00	10.00	15.00	10.00	0.06	0.03
	(6.30)	(6.30)	(0.36)	(0.36)	(535.40)	(535.40)
Kerosene			3.00	3.00		
			(0.22)	(0.22)		
LPG [*5]	0.10	0.10	0.20	0.20		
	(0.38)	(0.38)	(2.27)	(2.27)		

[*1] Hard coal, brown coal, and petroleum cokes.
[*2] Wood and wood waste.
[*3] Black liquor and other bio wastes.
[*4] Solid and liquid waste fuels, waste tyres, and sludge.
[*5] Liquefied petrol gas.
[*6] Natural gas and derivatives.
[*7] Central heating plants.
[*8] Autoproducer electricity, heat and CHP plants, and public heat plants.
[*9] Residential, commercial, institutional and other combustion.
[*10] Process emissions from FCC in petroleum refineries.

***Table 10**:* *Range of emission factors for PM emissions from* **exhaust** *road traffic used by TNO for Norway, 1995.*

Activity **(million km)** *Vehicle category*		**Emission factor (g / km)**	
		PM_{10}	**$PM_{2.5}$**
Passenger cars	27400	0.001 – 0.150	0.001 – 0.150
Light duty vehicles	3292	0.001 – 0.990	0.001 – 0.990
Heavy duty vehicles	2790	0.180 – 1.600	0.180 – 1.600
Motorcycles	776	0.040 – 0.120	0.040 – 0.120

The TNO study includes non-exhaust emission factors for tyre wear, brake wear and road abrasion wear in grams per kilometre driven in 1995 (road traffic; Table 11).

Table 11: *Emission factors for PM emissions from* **non-exhaust** *road traffic used by TNO for Norway, 1995.*

Activity **(million km)** *Vehicle category*		**Emission factor (g / km)**	
		PM_{10}	**$PM_{2.5}$**
Passenger cars, tyre wear	27400	0.003	0.000
Passenger cars, brake wear	27400	0.006	0.006
Passenger cars, road abrasion wear	27400	0.007	0.000
Light duty vehicles, tyre wear	3292	0.005	0.000
Light duty vehicles, brake wear	3292	0.008	0.008
Light duty vehicles, road abrasion wear	3292	0.010	0.000
Heavy duty vehicles, tyre wear	2790	0.019	0.000
Heavy duty vehicles, brake wear	2790	0.032	0.032
Heavy duty vehicles, road abrasion wear	2790	0.027	0.000
Motorcycles, tyre wear	776	0.003	0.003
Motorcycles, brake wear	776	0.004	0.000
Motorcycles, road abrasion wear	776	0.002	0.000

Industrial process emissions in Norway in 1995 were split into 16 economic sectors, for each sector abated emission factors are given in kilograms per ton material processed (Table 12).

***Table 12**:* **Abated** *emission factors for PM emissions from industrial processes used in the TNO inventory, Norway, 1995.*

Industrial process emissions	*Activities (Mt)*	*Emission factors (kg / t)*		
		PM_{10}	**$PM_{2.5}$**	**TSP**
Agglomeration plants, sinter	0.45	0.10	0.10	0.20
Agglomeration plants, pellets	1.50	0.03	0.03	0.03
Hot metal transport, iron/steel works	0.43	0.80	0.30	1.00
Electric arc furnaces	0.51	0.10	0.06	0.10
Grey iron foundries	0.06	0.60	0.09	2.00
Production of primary aluminium	0.85	2.85	1.28	3.00
Production of primary copper	0.03	0.48	0.40	0.50
Production of secondary aluminium	0.07	0.90	0.41	1.00
Production of cement	1.61	0.18	0.08	0.20
Production of glass	0.40	0.03	0.02	0.03
Production of lime	0.10	0.15	0.03	0.30
Production of nitrogen fertilizers	3.84	0.24	0.18	0.30
Hard coal mining	0.29	0.03	0.00	0.05
Iron ore mining	2.17	0.03	0.00	0.05
Copper ore mining	0.01	0.03	0.00	0.05
Zinc ore mining	0.02	0.03	0.00	0.05

The TNO inventory includes very detailed sector splits for material handling. However, those were not assumed to be major sources of PM emissions in Norway in 1995.

According to TNO gas flaring in the oil and gas industry was the major source of PM emissions in Norway in 1995, contributing ca. 50% to the total national PM emissions. The activity in 1995 was assumed to be 403 PJ with relatively high emission factors of 0.064 kt/PJ for both PM_{10} and $PM_{2.5}$.

A few other sources should be mentioned because the TNO calculations say that the sum of those activities contributes considerably to total PM emissions or single SNAP aggregations, especially SNAP06 (solvent and other product use). Most of them were not included in the RAINS and SFT calculations: “fugitive emissions from small industrial emitters”, “construction-related activities: dwellings, utilities, “commercial and residential meat frying, barbecues, food heating”, “tobacco smoking”, and “use of fire works”.

III. Methodology Used by SFT (Norwegian Pollution Control Authority)

The Norwegian 1995 PM emission inventory was organized around a general model called "Kuben" ("the Cube"). Several emission sources, e.g., road traffic and air traffic were covered by more detailed satellite models. Aggregated results of those models were used as input to the general one. The inventory addressed PM_{10} emissions, except for road traffic where $PM_{2.5}$ exhaust was also covered. The methodologies used are described in detail by Flugsrud et al. (2000), only a brief summary of the approach applied is given here.

For combustion-related PM emissions the emission factors depend on the fuels, sectors, and technical sources involved. Combustion of a fuel (the emission carrier) takes place in a particular type of equipment (the technical source) in a certain economic sector. Most of the emission factors refer to controlled emissions.

In order to fit non-combustion activities into the general model, emissions were assigned to an emission carrier and a technical source. The emission carrier is then the physical object (e.g., road asphalt) emitting primary particles, and the technical source is the process leading to emissions. In the 1995 inventory industrial processes were not included as PM emission sources.

Aggregated Sectors and Emission Factors

The calculations of emission factors followed three steps. The subscripts refer to the four axes of "the Cube": emission carrier (*i*), economic sector (*j*), technical source (*k*), and pollutant (*l*) (Table 13).

1. The combustion emission factors are multiplied by energy use figures from the national energy accounts resulting in PM emissions *E*:

$$E_{ijkl} = A_{ijk} \cdot EF_{ijkl}$$

2. PM emissions from major manufacturing plants (point sources) had been measured directly or determined from mass balances. If measurement data was available it was used to replace the estimated values:

$$E_{ijkl} = (A_{ijk} - APS_{ijk}) \cdot EF_{ijkl} + EPS_{ijkl}$$

APS and *EPS* are the activities and the measured emissions at the point sources, respectively.

3. Non-combustion emissions were calculated by combining appropriate activity data with emission factors or by more complicated methods (see Flugsrud et al., 2000). Some of the emission values were obtained from current reports and investigations, and some had been measured directly.

Table 13: *Emission carriers and sources of PM emissions in the Norwegian model.*

Emission carrier	**Sources** (non-combustion sources in *italic*)
Fuel wood, wood waste, black liquor	Heating, fire
Coal	Combustion
Coal coke	Combustion
Petrol coke	Combustion
Natural gas	Combustion, flaring
Other gases (*)	Combustion, flaring
LPG (liquefied petroleum gas)	Heating, flaring
Motor gasoline	Road traffic, boats, motor equipment
Aviation gasoline	Air traffic
Kerosene (heating)	Heating
Jet fuel (kerosene)	Air traffic
Auto diesel	Heating, road traffic, railways, boats, motor equipment
Marine fuel	Combustion, ships
Light fuel oils	Heating, ships, motor equipment
Special distillate	Combustion, ships
Heavy fuel oils	Combustion, ships
Waste oil	Combustion
Waste	Combustion
Crude oil	Flaring
Asphalt	*Road abrasion wear*

(*) *Other gases* include refinery gas, landfill gas and an excess gas (mainly methane and hydrogen) produced and consumed in chemical industry. A given sector consumes only one specific gas, so different emission factors were used for the different sectors.

The classification of economic sectors is almost identical to that used in the National Accounts (Statistics Norway, 1994). To make the standard sectors more appropriate for calculating emissions from major sources, a few changes were introduced, e.g. "private households" were defined as an extra sector. The technical sources used in the model are shown in Table 14. Only anthropogenic sources were considered.

A model for estimating 1995 emissions from road traffic was developed by SFT in 1993 and revised in 1999 (Flugsrud et al., 2000). The results (as average aggregated emission factors) of this model were used as input to the general emission model. When the model was revised in 1999, emissions of primary particles caused by the use of studded tyres were included for the first time.

Concerning combustion processes in road traffic PM emission factors depend on the kind of vehicle (type, weight, technology, age), fuel type, and driving mode. The total number of vehicle-kilometres did not enter the calculations directly. However, fractions of the total were estimated for each combination of vehicle category and driving mode. These fractions were used to allocate fuel *consumption to the various combinations. Emission factors can be expressed as emissions per vehicle-kilometre (in g / km) or per unit fuel consumed (kg/t).*

Table 14: *Sources of PM emissions in the Norwegian model.*

Source	Emission carrier
Stationary combustion	
Direct-fired furnaces	Coal, coke, gas, fuel oils
Boilers	Wood etc., coal, coke, gas, kerosene, fuel oils, waste
Small stoves	Wood , coal, coke, LPG (*), kerosene, fuel oils
Gas turbines	Gas, marine diesel
Flares	Gas, crude oil
Mobile combustion	
Road traffic (several categories)	Gasoline, auto diesel
Snow scooters	Gasoline
Two-stroke boats	Gasoline
Four-stroke boats	Gasoline, diesel
Ships	Marine diesel, heavy oil
Railway	Diesel
Air traffic (landing/take-off below 100 m)	Jet fuel (kerosene), aviation gasoline
Air traffic (landing/take-off 100-1000 m)	Jet fuel (kerosene), aviation gasoline
Air traffic (cruise)	Jet fuel (kerosene), aviation gasoline
Motorized tools (two-stroke)	Gasoline
Motorized tools (four-stroke)	Gasoline, auto diesel
Non-Combustion	
Road abrasion wear	Asphalt

(*) Liquefied petrol gas.

The fuel-based model calculates changes in emissions between years from changes in total fuel consumption. The number of vehicles in the various categories, technologies in use, annual average distance (km) driven per vehicle, driving patterns, vehicle age, and driving mode were considered (urban, rural, highway; for details see Flugsrud et al., 2000).

Road traffic exhaust emissions were calculated for each combination of PM_{10} or $PM_{2.5}$, and the vehicle categories. The combinations of vehicle type, weight, and fuel resulted in 13 different classes (Table 15).

Emissions from evaporation and cold starts were added to the tailpipe emissions from warm motors, they were calculated separately outside the general model (for details see Flugsrud et al., 2000).

The fractions of the vehicle-kilometre totals for each fuel were calculated using the number of vehicles (by category and age), the average annual mileage (by category), and the average annual mileage (by age and aggregated vehicle categories). Those fractions were used to allocate total fuel used by road traffic in 1995.

***Table 15**: Norwegian 1995 inventory, vehicle categories $^{(*1)}$ in the emission model for road traffic.*

Fuel	Type	Total weight
Gasoline	Passenger car	
"	Light duty	< 3.5 t
"	Heavy duty	> 3.5 t
"	Bus	> 3.5 t
Diesel	Passenger car	
"	Light duty	< 3.5 t
"	Light heavy duty	3.5 - 7.5 t
"	Medium heavy duty	7.5 - 16 t
"	Heavy duty	> 16 t
"	Bus	> 3.5 t
LPG	Passenger car	
"	Light duty	< 3.5 t
"	Bus	> 3.5 t

(*1) Emissions from motorcycles and mopeds were calculated outside the main model.

"Road dust" was included in the SFT inventory as non-combustion PM emission source. It was defined as asphalt abrasion wear plus powdered grit material and tyre wear. Re-suspension was not considered. However, the influence of spikes during wintertime as well as driving on humid and ice-covered streets was considered.

Unfortunately, the documentation of the Norwegian 1995 PM inventory available for this report did not include information on the activities per sectors related to the single emission factors. Factors for 1997 were available (Tables 16 to 17), but for combustion processes the unit of PM emission factors given in the SFT inventory is kg / t while it is an energy unit in the TNO and RAINS studies (e.g., kt/PJ). Thus, direct comparison of emission factors is difficult because calorific values of the different fuels are needed to translate the mass into energy units.

However, the information on Norwegian emission factors given by SFT is rather detailed and can be used for further in depth comparison of assumptions made within RAINS and the TNO inventories. The RAINS database includes for example Norwegian calorific values for several fuels, which can be discussed with national experts and then used for harmonizing emission factors.

The factors shown in Tables 16 to 18 are mostly addressing abated PM_{10} emissions. One important exception is the high emission factor for wood burning in small stoves (10 kg/t), certainly referring to more or less uncontrolled emissions (Table 16). Assuming the same "unabated" emission factor for boilers, the value of 0.22 kg/t points at a 98% control of PM_{10} emissions estimated for boilers in Norway in 1995.

Table 16: *Overview on PM_{10} emission factors combustion in boilers, stoves and furnaces, Norway 1997.*

kg/t	**Boilers**		**Small stoves**		**Direct fired furnace**		
	General	Others(*2)	General	Domestic	General	Structural clay	Refineries
Coal	4.50	1.40	4.50	8.50	---	---	---
Coal coke	3.00	1.40	3.00	3.00	---		
Petrol Coke	---	1.40	---	---	---	---	---
Wood etc.	0.22	0.22	10.00	10.00	---	---	---
Natural gas	0.00	0.00	0.00	0.00	---		
Other gas	0.00	0.00	---	---	---	---	---
LPG (*1)	0.12	---	---	0.12	---	---	---
Kerosene (heating)	0.16	0.16	0.16	0.10	---	---	---
Marine gas oil	---	---	---	---	1.20	---	---
Light fuel oil	1.55	1.43	1.55	1.55	---	---	---
Special distillate	1.48	1.36	1.48	1.48	1.30	5.00	1.30
Heavy fuel oil	0.75	1.03	0.75	---	1.30	5.00	1.30
Waste oil	0.75	1.03	---	---	1.30	---	---

(*1) Liquefied petrol gas.
(*2) Manufacturing, cellulose, refineries.

Finally, Tables 17 and 18 summarize 1997 emission factors for road traffic and off-road machinery. The factors consider control options in place in 1997.

Table 17: *Overview on PM_{10} emission factors for Norway (1997), combustion traffic.*

kg/t	**LDV <3500 kg**	**LDV >3500 kg**	**HDV**	**Railways**	**Air traffic LTO <100 m**	**Air traffic LTO 100-1000 m**	**Air traffic cruising**
Gasoline	0.28	0.21	0.10		0.42	0.40	0.40
Diesel	3.32	3.00	2.45	3.8			
Kerosene (general)	---	---	---	---	0.40	0.40	0.40
Kerosene (defense)	---	---	---	---	0.32	0.32	0.32

	Motor cycles	**Mopeds**	**Snow scooters**	**2-stroke boats**	**4-stroke boats**
Gasoline	0.15	0.14	0.14	8.00	1.00
Diesel	---	---	---	4.00	---

Table 18: *Overview on PM_{10} emission factors for Norway (1997), combustion off-road machinery.*

kg/t	**Ships** (*1)	**Motorized equipment** (*2)				
		Agriculture, Forestry	**Mining and metal ores, railways**	**Soil and stone**	**Construction**	**Defence**
Gasoline		1.00	1.00	1.00	1.00	1.00
Diesel, light fuel oil	0.50 - 0.87	7.10	3.80	4.20	5.30	5.40
Marine gas oil	0.50 - 0.87	---	---	---	---	---

(*1) Maximum value for extraction of oil and gas, minimum value for fishing, oil drilling and defence.
(*2) Emission factor foe 2-stroke machinery, gasoline is 8.00 kg/t.

The PM_{10} emission factors for non-exhaust road dust (excluding re-suspension) for 1997 were:

- *For light duty vehicles with spikes: 0.31 g/km,*
- *For heavy duty vehicles with spikes: 1.53 g/km.*