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Umweltschutzes „Modellierung und Prüfung von Strategien
zur Verminderung der Belastung durch Ozon“

Aerosols over Europe.
Focus on Black Carbon

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16. Kurzfassung

Der Bericht beschreibt von der TNO, Niederlande, für das Vorhaben erstellten Arbeiten:
Es wird eine erste Abschätzung der europaweiten Emissionen von elementarem Kohlenstoff (EC oder black carbon, BC) erstellt. Diese Emissionen wurden für eine europaweite Ausbreitungsrechnung für BC benutzt. Der Vergleich mit Messungen zeigt, daß die Modellergebnisse eine klare Tendenz zur Unterschätzung haben.

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16. Abstract

The report describes work carried out by TNO, Netherlands:
Based on a first estimate of Black Carbon (BC) emissions over Europe model calculations have been performed to obtain BC-concentrations. The model results have a clear tendency to be lower than the observations.

17. Keywords
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Summary

Based on a first estimate of Black Carbon (BC) emissions over Europe model calculations have been performed to obtain BC-concentrations. The model results have a clear tendency to be lower than the observations.

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1. Introduction

Fine particulate matter is the most dangerous component of air pollution. Elevated levels of these pollutants are associated with thousands of preliminary deaths each year. In the near future European guidelines on particulate matter are to be evaluated. At this moment PM 10 is the regulated compound, however, replacement by PM 2.5 is under discussion. In climate research particles in suspension in air are denoted as aerosols. Aerosols influence the radiation balance of the earth directly via scattering and absorption of light and indirectly by influencing the properties of clouds. On a regional scale aerosols provide the means for long range transport of acidifying compounds and eutrophication. These effects depend largely on aerosol composition and size. Most important are the particles in the fine aerosol fraction (PM2.5), which have the longest life time in the atmosphere.

Aerosols are a mixture of species of anthropogenic, natural and biogenic origin. The majority of the mass in the PM2.5 size range is of anthropogenic origin, whereas the PM10-2.5 is at least partly of natural origin like sea-salt and wind-blown dust. The levels and composition of these aerosols over Europe are determined by meteorological conditions, hemi-spherical back ground levels, trace gas concentrations and the major anthropogenic sources and may therefore vary regionally. For both the health and the climate/acidification community including policy makers it is of interest to determine the aerosol composition and understand the sources, formation processes and sinks of particles.

Several studies have addressed this subject from the measurement point of view see for example Putaud (2001). The largest fraction of PM2.5 (and PM10) consists of the secondary inorganic ions sulphate, nitrate and ammonium and elementary and organic carbon. From these compounds only nitrate may be found at significant concentrations in the PM10-2.5 size range. The PM composition is illustrated in Figure 1. For three sites the composition of PM10 is shown. The sites represent an urban site (Barcelona) and two near city sites in the Po-valley, Italy, and the Ruhr area, Germany. At all sites the inorganic ions contribute half the total PM10 mass. Hence, their contribution to the PM2.5 mass is probably larger, since nitrate is thought to be in the PM2.5 size range, at least in the Po-Valley and Voerde. At these locations carbonaceous material is about 20-38 % of PM10 mass. The highest contribution is found at the urban site. This feature is shown for EC as well as OC but most pronounced for the EC concentration. This can be explained by the primary nature of these components. Other, mostly natural elements such as Ca, Na, Cl are found in PM10 but are less likely to contribute much to the fine aerosol mode.

From the measurement point of view only a limited amount of data is available at the moment and modelling may provide a tool to assess the mass closure in uncovered regions. In addition, models are very well suited to investigate the most im-

portant processes involved. Modelling studies addressing the total aerosol mass have been started only recently. Most of these models are not complete and/or have a very coarse resolution to address issues. Here we try to make a mass closure with the regional LOTOS model. In this report we will focus on primary particulate matter. We present a new emission database for BC, which is derived from the CEPMEIP database. The total primary emissions will be classified in black carbon and additional particulate matter (APPM). The first will receive most attention due to its role as a greenhouse gas. BC is also under discussion as an indicator for human health related particles. In section 3 we will describe the basic features of the LOTOS model and the results are discussed in section 4. Next, we will evaluate the mass concentration for the separate components and discuss the contribution of each component to the total modelled PM2.5 mass.

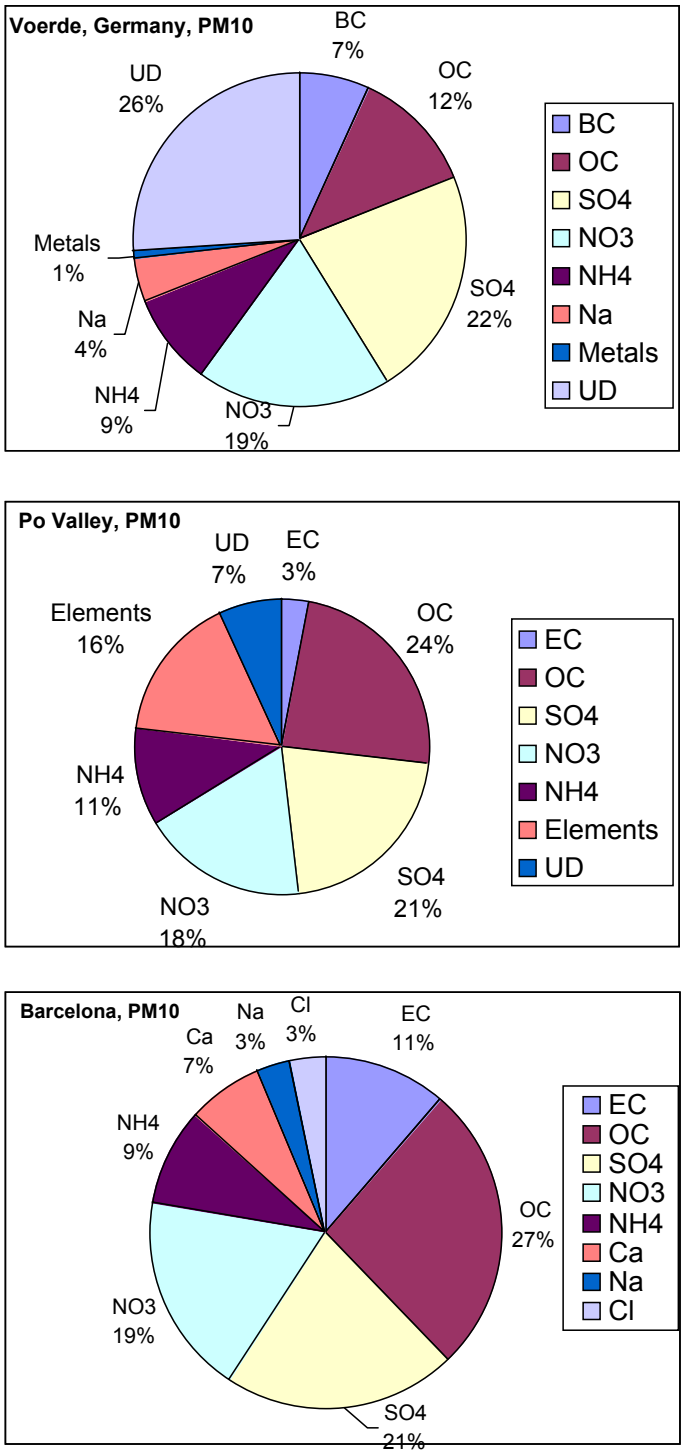

Figure 1 Composition of PM10 expressed in relative units at three sites in Europe.

2. Primary emissions

Over Europe anthropogenic burning of fossil fuels is thought to dominate the total BC emissions. On a global scale, however, (semi-)natural biomass burning is also very important. Streets et al (2001) present a review study for BC emission factors from anthropogenic sources, in which also earlier work by Penner et al. [1993], Cooke and Wilson [1996] en Cooke et al. [1999] is discussed. Streets et al (2001) describe a method to estimate the sub-micron fraction of the total particle emissions and, next, to estimate the BC emission from the sub-micron particle emissions. They applied this method to estimate the BC emissions in China. The most important emission sources for BC are similar for China and Europe and are given in Table 1. In this study we focussed on the emission by these sources. The BC fractions of the sub micron particles as presented by Streets et al (2001) are almost exclusively based on western technology, since specific Chinese data were not available. These data are thus applicable to Europe. The most important difference between China and western countries are the different burning technologies and removal efficiencies (Streets et al, 2001). The sub micron particle emissions, which are largely influenced by the state of the technology, are known for Europe and are part of the CEPMEIP database.

Table 1 Relevant emission sources for BC following [Streets et al., '01].

Sector	Fuel
Residential combustion	Bituminous Coal
	Anthracite or briquette
	Fuel oil
	Biomass
Industrial combustion (stokers)	Coal
	Fuel oil
	Biomass
Power generation (pulverised coal)	Coal
	Fuel oil
	Biomass
Road transport	Gasoline
	Diesel
Ship transport	Diesel
	Heavy fuel oil
Other transport	Gasoline
	Diesel
	Coal
Field combustion	Crop residue

A first estimate of the BC emission from the most relevant sources, listed in Table 1, can be made by combining the estimates of fine particulate matter with the black carbon fractions of fine particles as published by Streets et al (2001). In this way a top down approach was followed to come to an emission estimate. Combining these data on SNAP level 2, results in a BC fraction of the PM2.5 emission. The

European averaged values are given per SNAP level 1 category in Table 2. The European total emission is 800 Gg/yr, excluding sea ships. The largest contributions are from transport followed by wood burning in house holds. Other important sources are industrial combustion, energy transformation (especially with liquid fuels), flare-burning at oil platforms, agricultural waste burning and emissions from sea ships in international waters. The latter are not included in the LOTOS-model calculations presented here. The annual emissions as input for the LOTOS model are shown in Figure 2. The primary PM2.5 emissions are discussed in section 4.3.

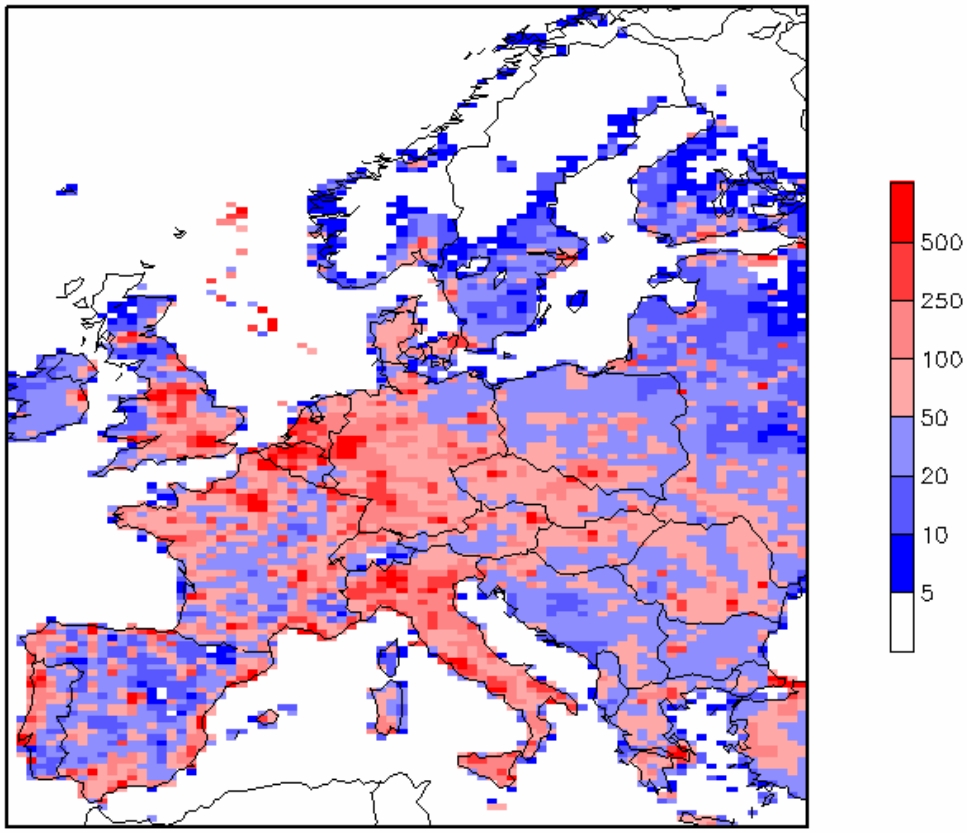

Figure 2 European BC emission (tonnes) derived in this study.

Table 2 Black carbon emissions and fraction of European PM2.5 emissions from CEPMEIP.

SNAP	Description	BC emission (tonnes)	Fraction BC of PM2.5
01	Energy transformation	56000	0.12
02	Small combustion sources	160000	0.20
03	Industrial combustion	62000	0.13
04	Industrial process emissions	0	0
05	Extraction of fossil fuels	37000	0.79
06	Solvent and product use	0	0
07	Road transport	180000	0.47
08	Non road transport	270000	0.49
09	Waste handling and dispo-sal	130	0.0037
10	Agriculture	27000	0.13
11	Nature	0	0
Seaships		23000	0.12
Total		820000	

3. Model description

A chemistry transport model was applied to calculate the fine aerosol concentration fields over Europe. For this purpose the LOTOS model (LOng Term Ozone Simulation) has been used. Calculations have been performed over the year 1995. The model is a 3-D Eulerian grid model with a horizontal grid resolution of 0.25 x 0.5 latitude-longitude. The LOTOS model is restricted to the lower troposphere and has 4 vertical layers up to about 3.5 km (depending on mixing layer height). More than 90% of the aerosol contributing to the AOD usually occurs up to this height.

The meteorological input is diagnostic, derived from ECMWF using procedures developed by the Free University of Berlin. The emission input is based on CORINAIR. The gas-phase chemistry used in LOTOS is a modified version of the Carbon Bond IV mechanism (CBM-IV). Biogenic VOC (Volatile Organic Compounds) emissions are also included. LOTOS has been thoroughly tested for ozone. In order to calculate (ammonium) sulphate and (ammonium) nitrate concentration fields over Europe, LOTOS has been extended with the Isorropia aerosol module.

In this paper we calculated the distribution of primary emitted particles, which are then combined with the inorganic species to obtain an estimate of the total PM2.5 concentration field. The emissions of the primary particles are taken from the CEPMEIP project . The BC fraction of the primary emissions was estimated as described below. In the model we have treated all primaries as of they act as sulfate. Hence, we assumed all BC and OC to be hydrophyllic direct after emission.

4. Results

4.1 Concentration fields

Primary emitted components
In Figure 4 the concentration fields of the total primary material and its black carbon content are shown. Annual averaged concentrations of total fine primary material range up to 5 μg/m^3 over Europe. Over the largest part of Europe the levels of primary components are between 1 and 2.5 μg/m^3. Areas with the highest concentrations are found in industrialised and/or densely populated regions, e.g. north western Europe, the Po valley and southern Poland. Secondary maxima can be identified over a number of large cities, such as London and the Manchester area (UK), Athens, Madrid and Oslo. In Scandinavia concentrations trail off from about 1 in the south to less then 0.25 μg/m^3 over mid and northern Scandinavia. Ship emissions are not included in the current model calculations yet and, hence, the concentrations of primary particles trail of from the edges of the continent to more marine locations due to dry and wet deposition. The local maxima over the North Sea can be explained by oil platforms.

The black carbon content of the primary aerosol shows a very similar distribution as the total primary components. This is explained by the fact that the source categories with the highest PM emissions also show the highest BC fractions. The black carbon levels are about one fifth of the total primary aerosol. Concentrations are lower than 50 ng/m^3 over remote regions such as the northern Scandinavia. In relatively clean areas over Spain and southern Scandinavia BC are about 250 ng/m^3 whereas BC concentrations exceed 1000 ng/m3 in the densely populated areas and 500 ng/m^3 over central Europe.

In the near future a compilation of mass closure studies will become available (Putaud et al, 2003) which contains data on BC. This compilation will be used to verify model results against. For now, we will limit the verification to data found for a number of sites in the literature ad, see Table 3. The data represent a period ranging from the end of the eighties to 2001 and are often obtained in campaigns. The model to measurement comparison is shown in Figure 3. Black carbon concentrations are mostly underestimated by the model by a bout a factor of 2-3. Several reasons exist for the underestimation of the measured data. First of all, the results from the model represent boundary layer averaged concentrations over a whole grid cell, whereas the measurements are performed at ground level, most often in cities or small towns. In addition, some of the data are obtained several years before the study period, when emission control was not as effective. Lastly, the campaign wise data are inevitably strongly influenced by the specific meteorology during those campaigns, whereas the model uses the meteorology from 1995.

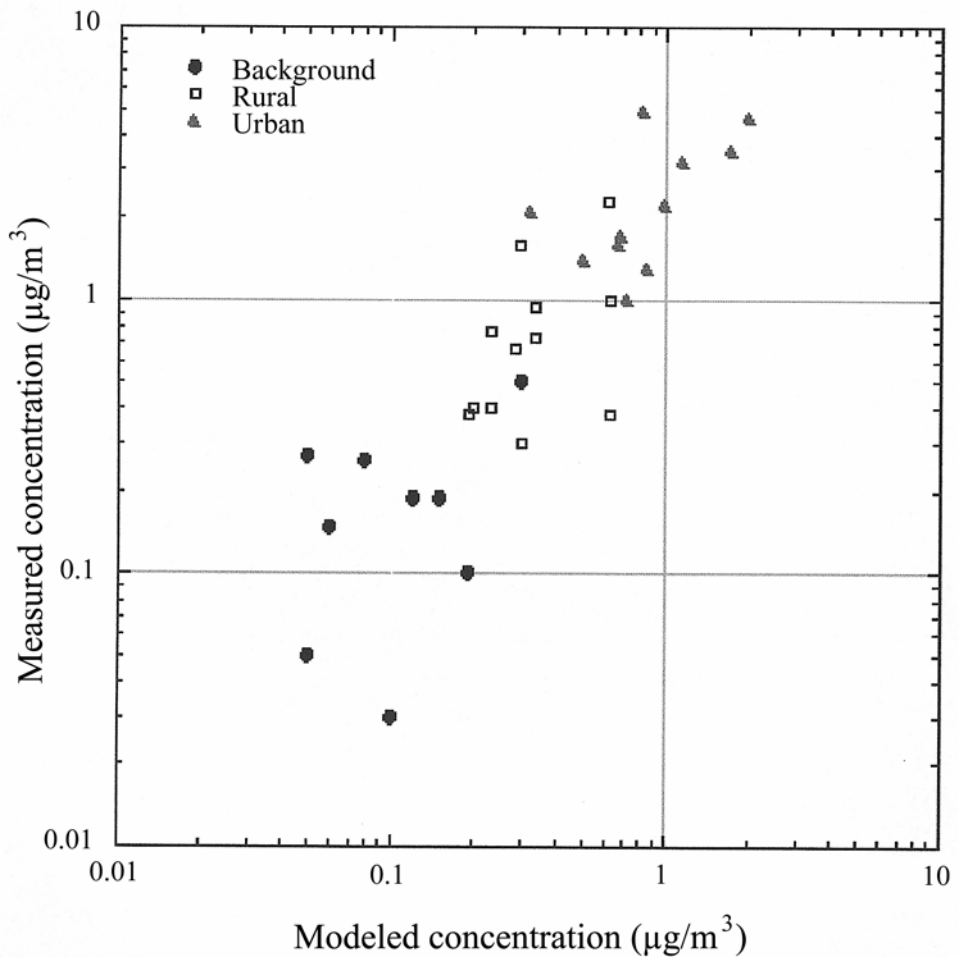

Figure 3 *Comparison between simulated and modelled BC concentrations through Europe.*

Secondary components

The calculated fields of the inorganic components are shown in figure 4. Sulphate contributes more than 3 µg/m^3 to PM 2.5 over a region that spans from England, over north western Europe to south eastern Europe. The same applies to the Po valley and the north west of Spain. Maximum contributions are calculated in the Ruhr area, southern Poland, Hungary and Rumania. To the north of 57 degrees latitude the annual averaged sulfate concentrations are lower than 2 µg/m^3, whereas those in more remote continental areas are between 2 and 3 µg/m^3. Comparison with measured data revealed that the annual averages were slightly underestimated (0.94) and correlated well with those measured (r = 0.58).

High concentrations of nitrate are confined to continental areas, with maxima in north western Europe, the UK and the Po valley. Averaged over the year concentrations of nitrate are as high as those of sulfate in western Europe, whereas those of sulphate are higher in (south) eastern Europe and Scandinavia. In contrast to

sulphate nitrate levels show a distinct seasonal variation. Maximum concentrations occur in winter, when ammonium nitrate is stabile. During summer ammonium nitrate formation is limited over the largest part of Europe due to the high ambient temperatures. In the model sulphate and nitrate are neutralised by ammonium. Therefore, ammonium concentrations are significant and only somewhat lower than those of its associated ions alone. Nitrate levels simulated by the model correlate somewhat less with observations (r = 0.52) and are on average 20 % too high. Largest overestimation was found in the centre of the Netherlands and at the Czech sites. Data from the latter may be influenced by measurement artefacts and their location, as they are sampling with Teflon filters and are located somewhat higher than the surrounding.

4.2 PM2.5 distribution

The calculated anthropogenic PM2.5 distribution is shown in figure 5. Maximum concentrations are found in the most industrialised and populated areas of Europe. Concentrations exceed 20 $\mu g/m^3$ over the south of the Netherlands, Belgium, the Ruhr area and southern Poland. Secondary maximums can be found in the Po valley, the Czech Republic and metropolitans of London, Manchester and Paris. Over central and south eastern Europe concentrations are calculated to be between 10 and 20 $\mu g/m^3$. Towards the north the anthropogenic concentration of PM2.5 trails of to about 4 in southern Scandinavia to less than 2.5 further north. Also Spain and southern France are relatively clean with respect to PM2.5. Further, a number of cities can be recognised, e.g. Madrid, Barcelona and Athens.

4.3 Composition of PM2.5

In figure 5 the contribution of all model components to the total fine aerosol mass is shown.

Primary emitted particles contribute 15 to 20 % of the total fine aerosol mass over continental Europe. This percentage is rather constant. Higher contributions above 30 % are found in densely populated regions. For BC the contribution to PM2.5 ranges between 3 and 10 %. Similar to the total primaries highest values occur over the most densely populated regions in Europe. The high values over the north sea are due to oil platforms there. Striking is the low relative contribution of BC over eastern Europe, less than 4 %. This region coincides with the band of high sulfate concentrations. This minimum is only slightly visible in the contribution of total primary material. This illustrates that the BC emissions are largely due to diffuse sources as traffic and off road transport, whereas the sulphur and to a lesser extent total primary emissions have a large stationary component.

Sulphate contributes the largest mass fraction to PM2.5 over Europe. Its contribution is about 30-35 % over most of continental Europe. The mass fraction increases towards more remote locations and is slightly lower in those areas where primaries and nitrate are important. The percentage of nitrate in PM25 over continental Europe is modelled to be 20 to 30 %. Contributions over 30 % are found in the regions with high ammonia emissions. The contribution trails off from the coast towards the open sea, since ammonia is rapidly deposited. In reality, the reaction of nitric acid with sea salt plays an important role transferring the nitrate from the fine to the coarse aerosol fraction, which is not represented in the model yet. Since ammonium neutralises both sulphate and nitrate in the model, ammonium contributes about 15 to 25 % to the fine aerosol mass. Maxima coincide with those of nitrate.

In section 4.1 we compared the modelled data for the inorganic ions and BC with measured data. For APPM, although probably largely composed of organic carbon, an opportunity to verify the performance of the model in an indirect way is via mass closure.

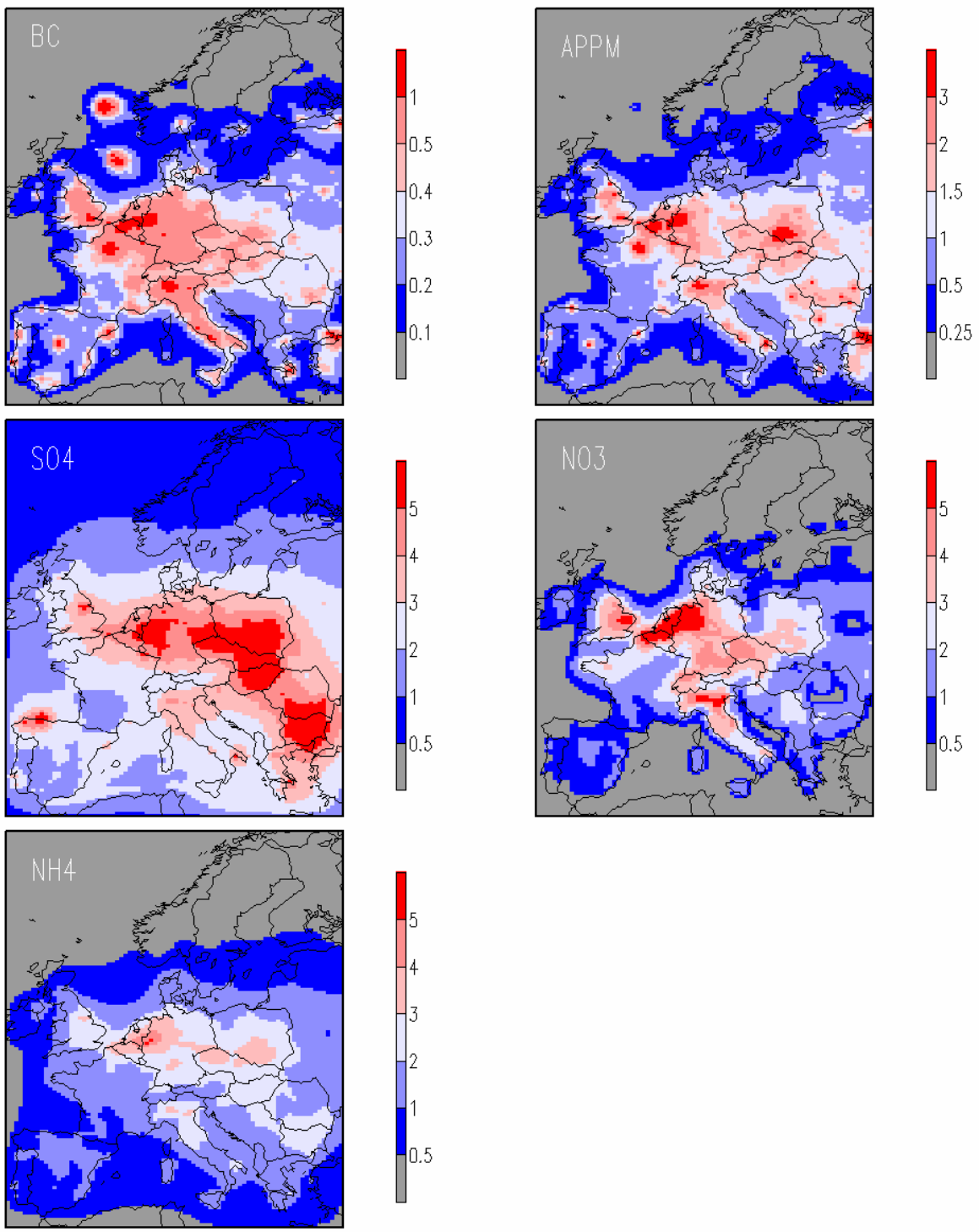

Figure 4 Annual average distribution of BC, APPM, SO4, NO3 and NH4 over Europe (µg/m^3).

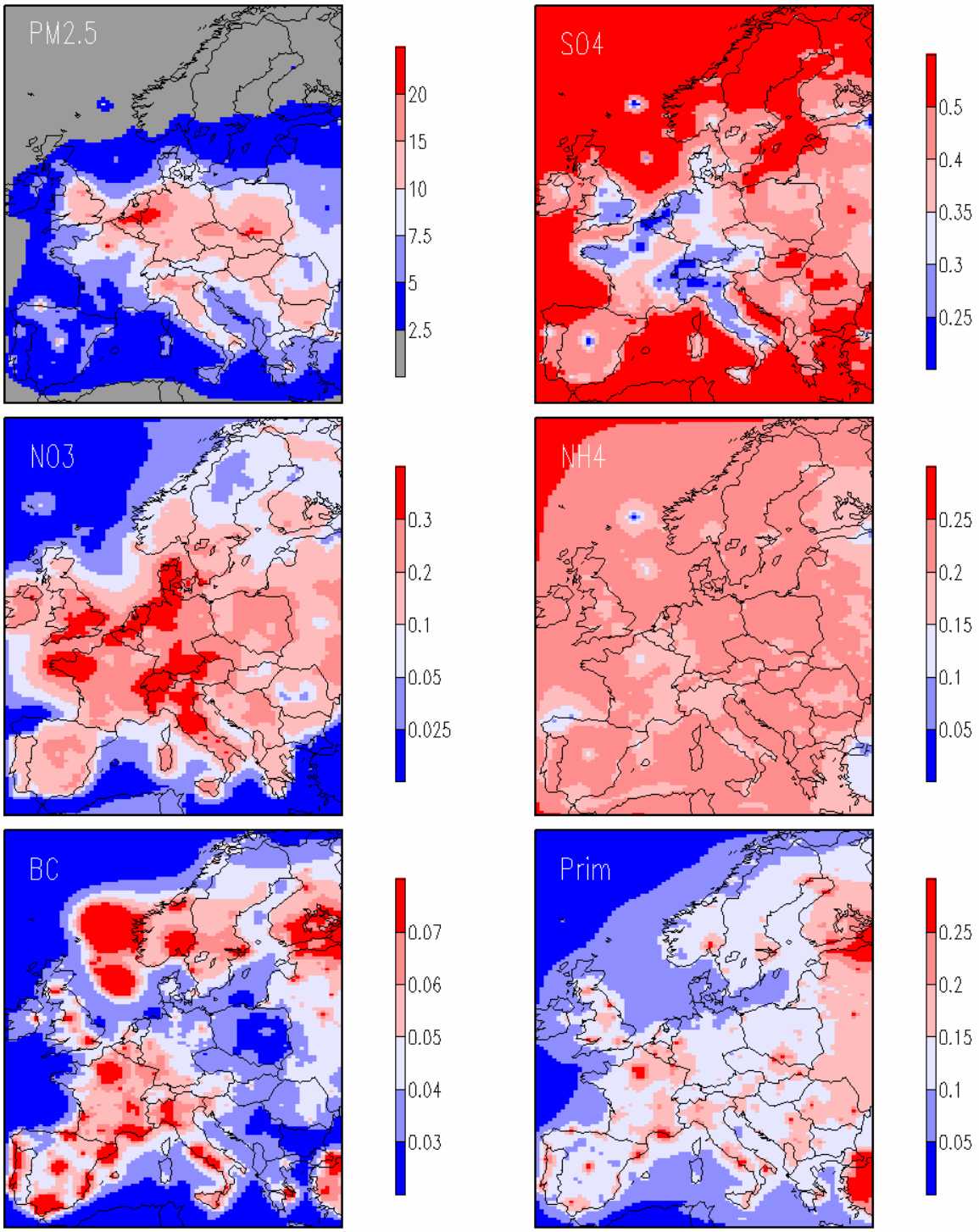

Figure 5 *Annual average distribution of PM2.5 (µg/m^3) over Europe in the upper left panel and the associated fractions of BC, APPM, SO4, NO3 and NH4 in the other panels.*

5. Discussion on BC estimates

We have presented a novel emission database on BC emissions in Europe, which was constructed by combining sub-micron particle emissions from the CEPMEIP database and BC fractions of sub-micron aerosol presented by Streets et al (2001). Using this database we calculated concentration fields of BC and in addition all primary particulate mass over Europe. These data were then combined with secondary inorganic aerosols to assess its mass fraction in PM2.5 over Europe, showing that BC is a minor aerosol component when regarding only its mass. However, BC plays a very potent role in climate change.

Here we will discuss the uncertainties in the BC estimates presented above. Black carbon concentrations are mostly underestimated by the model by a bout a factor of 2-3. Several reasons exist for the underestimation of the measured data. First of all, the concentrations from the model represent boundary layer averages over a whole grid cell, whereas the measurements are performed at ground level, most often in/near cities or small towns. In addition, the campaign wise data are inevitably strongly influenced by the specific meteorology during those campaigns, whereas the model uses the meteorology from 1995.

Another reason is that a part of the measured data was obtained several years before the study period. This is illustrated in Table 3 in which we show an overview of BC emission estimates for different reference years. It appears that the reference year is an important factor and that studies for different reference years can not be easily compared. This overview suggests a strong decrease in BC emissions since the eighties. A study searching for a trend in the BC emissions and connecting those with technical improvements and changing emission factors appears to be useful.

An important reason for discrepancies is the uncertainty in the emission estimates. The goal of the study presented here was to evaluate the BC concentration over Europe in a simple way. For this reason a top down approach was chosen to derive the BC emissions. A very detailed bottom up study to derive these estimates by improving our knowledge on emission factors is beyond the scope of the study. However such an exercise seems very meaningful at the moment since the emission factors are under constant discussion. In this respect one should mention that the original emission factor for diesel cars used in CEPMEIP has been lowered by a factor two, based on official Auto-oil data. Using the old emission factor would result in 30-50 % higher BC concentrations over western Europe.

Uncertainties are also present in the treatment of BC in the model. BC is treated in a very simplified manner in LOTOS. The simple treatment in models, not only LOTOS, reflects the low amount of knowledge on the behavior of BC in the atmosphere. Especially the aging of black carbon, e.g. the timescale at which BC

becomes hydrophylic , is under discussion. In this study we assumed BC to be hydro-phylic direct after emission. Assuming an aging time scale in the order of a day would result in a longer life time and may result in higher concentrations. The latter is most probable for remote locations where the concentration is not determined by the local source strength.

Finally, black carbon and elemental carbon data are associated with considerable uncertainty due to differences in measurement techniques. Different thermal and optical techniques for these measures have been compared at Melpitz showing a spread of (at least) a factor of 3 between the individual samplers. Optical instruments measure the absorption or "blackness" of a filter. However, soot originating from multiple sources may have different optical properties (Liousse et al, 1993), which gives rise to uncertainty in the data. A compilation of black carbon and elemental carbon data with appropriate uncertainty estimations is desperately needed to verify model results against. However, we are not sure if this is the right time for such an exercise, given the large uncertainty and discussion on this subject.

Table 3 Estimates of the European Black Carbon emissions for different base years.

Author	Year	Europe BC Tg/yr	Remark
Penner et al. '93	1980	4.07	
Cooke & Wilson '96	1984	4.32	
Cooke et al. '99, bulk	1984	2.81	Fossil fuel BC emission only
Cooke et al. '99, sub-micron	1984	1.95	Fossil fuel BC emission only
Bond et al. '02	1996	0.79	
This work, sub-micron	1995	0.80	Only anthropogenic sources

6. Conclusions

We have presented a novel emission database on BC emissions in Europe, which was constructed by combining sub-micron particle emissions from the CEPMEIP database and BC fractions of sub-micron aerosol. The total European BC emission was estimated to be 800 Gg/yr for 1995. Diffuse sources contribute most to the emission total. Most important sources categories are transport and wood combustion in house holds.

Using the LOTOS model we calculated the distribution of primary components over Europe. Combining these data with distributions of inorganic ions we were able to evaluate the relative importance of different components in fine particulate matter. The calculations show that sulphate is the most important aerosol component over Europe. Sulphate contributes 25 to 50 % of the total modelled aerosol mass. Nitrate contributes 15 to 30%, the highest values in those areas with high ammonia emissions. Ammonium neutralises both sulphate and nitrate and contributes 15-25 %. Black carbon fractions are in the order of 3-7 percent, with higher values in the most populated regions. A similar distribution of the additional primary material is calculated with typical mass fractions of 10-15%, with higher numbers in the most populated regions.

The concentration of inorganic aerosol components compares quite well with measured data. However, BC levels over central Europe seem to be underestimated by the model. The latter may be explained by uncertainties in the emission database and the simple treatment of BC in the model. In addition, the measured data are often obtained near sources, whereas the model represents boundary layer averages over grids of about 30 x 30 km2. In this respect, it should also be mentioned that the measurements were made in a period of two decades of data. Older data may not be representative for the situation in 1995. Overall, we conclude that the emission and modelling of BC is very uncertain due to large uncertainties in emission estimates and the lack off reliable data to verify the model results.

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8. Authentication

Name and address of the principal:
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Names and functions of the cooperators:
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