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Anthropogenic black carbon and fine aerosol distribution over Europe

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[1] We present a model simulation for the year 1995 accounting for primary particles, which are an important component of fine aerosols over Europe. A new emission inventory for black carbon (BC) was developed on the basis of the recent European emission inventory of anthropogenic primary particulate matter (Coordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP)). The annual BC emissions of Europe and the former Soviet Union for 1995 are estimated at 0.47 and 0.26 Tg C, respectively, with highest contributions from transport (off-road and on-road) and households. Modeled BC concentrations range from ≤0.05 μg/m^3 in remote regions to more than 1 μg/m^3 over densely populated areas. The modeled BC concentration is about 25% of the total primary aerosol concentration. The primary aerosol fields were combined with previously calculated secondary aerosol concentrations to obtain an estimate of the total anthropogenic fine aerosol distribution. Modeled BC levels contribute only 4–10% to fine aerosol mass, whereas sulphate and nitrate contribute 25–50 and 5–35%, respectively. Comparison with experimental data revealed that the model underestimates PM2.5 levels, mostly caused by the underprediction of total carbonaceous material (BC and OC) by a factor of ~2. The underestimation can partly be explained by the influence of local emissions, measurement uncertainties, natural sources, and representation of wet deposition. However, the uncertainties associated with the emission inventory for BC (and total PM) may be the most important cause for the discrepancy. In comparison with previous studies, our BC emission estimate is a factor of 2 lower, caused by the choice of more recent emission factors. Therefore a better knowledge of emission factors is urgently needed to estimate the BC (and PM) emissions reliably. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 9335 Information Related to Geographic Region: Europe; *KEYWORDS:* black carbon, PM2.5, particle emissions, aerosol composition, LOTOS, Europe

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1. Introduction

[2] Atmospheric particulate matter (PM) is a complex mixture of anthropogenic and natural airborne particles. Particulate matter in ambient air has been associated consistently with excess mortality and morbidity in human populations [e.g., *Dockery et al.*, 1993; *Pope et al.*, 1995; *Brunekreef*, 1997; *Hoek et al.*, 2002]. Respiration is the only route of exposure that is of concern in relation to the effects of PM on human health. Therefore the European air quality standards currently focus on all particles smaller than 10 μm in diameter (PM10), which covers the inhalable size fraction of PM. Mass and composition of PM10 tend to divide into two principal groups: coarse particles, mostly larger than 1 μm in aerodynamic diameter, and fine particles, mostly smaller than 1 μm in aerodynamic diameter. The fine

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particles contain secondary aerosols, combustion particles and condensed organic and metal vapors. The larger particles usually contain sea salt, earth crust materials and fugitive dust from roads and industries [*World Health Organization* (*WHO*), 2000]. Although adverse health effects are associated with elevated levels of both PM10 and PM2.5, these health effects were most strongly and consistently associated with particles derived from fossil fuel combustion [e.g., *Hoek et al.*, 2002], which mostly occur in the PM2.5 size range. Another reason to study PM2.5 rather than PM10 is that long range transport of aerosols also depends strongly on aerosol size.

[3] Moreover, various components of fine particulate matter (PM2.5) in the atmosphere also have climate-forcing impacts, either contributing to or offsetting the warming effects of greenhouse gases [*Kiehl and Briegleb*, 1993; *Hansen and Sato*, 2001; *Menon et al.*, 2002]. In particular, black carbon (BC) has recently been identified as an important contributor to radiative heating of the atmosphere [*Haywood et al.*, 1997; *Myhre et al.*, 1998; *Jacobson*, 2001, 2002]. Organic carbon (OC), which is often emitted along with BC, may act to offset some of the global warming impact of BC emissions [*Hansen and Sato*, 2001]. In case of biomass burning aerosol, OC is thought to completely compensate the warming potential of BC [*Penner et al.*, 1998; *Grant et al.*, 1999]. However, for fossil fuel derived emissions this is not the case and a net positive forcing remains [*Penner et al.*, 1998; *Cooke et al.*, 1999]. Control of fossil fuel black carbon may be a cost effective way to reduce global warming emissions [*Jacobson*, 2002] in conjunction with abatement of GHG emissions. So measures to abate climate forcing by reducing BC emissions often would have collateral benefits by reducing emissions of health-related pollutants.

[4] The main sources for carbonaceous aerosols are incomplete combustion of fossil fuel and biomass, and the oxidation of biogenic volatile organic compounds (VOC) [*Seinfeld and Pandis*, 1998]. Carbonaceous aerosol is predominantly present in the submicron size fraction [*Echalar et al.*, 1998; *Cooke et al.*, 1999]. The global emissions of black carbon and organic carbon from fossil fuel burning has been estimated at 6 to 8 Tg yr^{-1} and 10 to 30 Tg yr^{-1}, respectively [*Penner et al.*, 1993; *Cooke and Wilson*, 1996; *Liousse et al.*, 1996; *Cooke et al.*, 1999; *Scholes and Andreae*, 2000]. However, recent studies by *Bond et al.* [1998, 2004], in which a different technique for the determination of black carbon emissions was used, suggest significantly lower emissions. The differences indicate that there is still a large uncertainty associated with emission estimates of carbonaceous aerosols.

[5] Most of the studies on primary aerosols have been performed with global models. However, satellite retrieved aerosol optical depth (AOD), a measure for the aerosol burden, over Europe shows a large spatial variation and strong gradients around industrialized areas, indicating the influence on AOD of local emissions of primary aerosols and/or precursor gases [*Robles-Gonzalez et al.*, 2000]. Therefore we expect that a model with a higher horizontal resolution is more suitable to assess aerosol fields and especially those of primary particles than global models.

[6] In this paper we present a regional model study of the composition of anthropogenic-induced PM2.5 in Europe with special emphasis on the mass concentration of primary particles and more specifically BC. For this purpose we use a recent emission inventory of anthropogenic primary particulate matter in Europe (http://www.air.sk/tno/cepmeip/), which is based on the fuels used in different anthropogenic activities. As BC is formed by the incomplete combustion from, e.g., diesel engines, cooking fires and coal burning, a BC emission inventory can be derived from the PM inventory based on the fractions of BC in the respective (ultra) fine PM emissions [e.g., *Cooke et al.*, 1999; *Streets et al.*, 2001]. The burdens of primary emitted particles over Europe are calculated with the LOTOS model [*Builtjes*, 1992; *Schaap et al.*, 2004a]. By combining these results with earlier calculations for the secondary aerosol components sulphate, nitrate and ammonium [*Schaap et al.*, 2004a], estimates of PM2.5 levels over Europe are obtained. The consistency of the model calculations is checked with observations at a number of European locations.

2. Emission Data

[7] Black carbon (BC) is mostly released from incomplete combustion of carbonaceous fuels. Black carbon sources and their distributions over Europe are estimated using the results of a spatially distributed pan-European inventory of anthropogenic PM emissions for the year 1995 (CEPMEIP; see http://www.air.sk/tno/cepmeip/) CEPMEIP covers nearly all of the relevant sources of primary anthropogenic TSP, PM10, PM4, PM2.5 and PM0.95 emissions, however, without chemical speciation. An European BC emission inventory can be derived from the CEPMEIP data if the fraction of BC in PM2.5 for each of the source categories is known. A similar approach has recently been described by *Streets et al.* [2001] to obtain a BC inventory for China. *Streets et al.* [2001] present a review of BC fractions in submicron PM from anthropogenic sources, incorporating earlier work by, e.g., *Penner et al.* [1993], *Cooke and Wilson* [1996] and *Cooke et al.* [1999]. The BC fractions in fine PM presented by *Streets et al.* [2001] are used to estimate the BC emission from the submicron particle emissions derived from the CEPMEIP $PM_{2.5}$ inventory. Although the work by *Streets et al.* [2001] focused on China, the BC fractions of the sub micron particles for different fuels (Table 1), are almost exclusively based on western technology. Combining the fractions in Table 1 with emission data on a detailed source sector level (SNAP level 2) results in the BC fraction of the respective PM2.5 emissions. The BC fraction of PM2.5 was calculated per major source category (SNAP level 1) for each country separately. The European PM2.5 and BC emissions and averaged BC fractions of PM2.5 are given per major source category in Table 2. The European total PM2.5 emission, excluding the former Soviet Union (FSU) and international shipping amounts to 1880 Gg yr^{-1}. The corresponding BC emissions are 473 Gg yr^{-1}. The largest source contributions are from transport followed by wood burning in-house holds. Other important sources are industrial combustion, energy transformation (especially with liquid fuels) and gas flaring at oil platforms.

[8] The particle emissions have been gridded to the LOTOS grid, distinguishing area and point sources. For the majority of area sources the distribution of emissions

Table 1. Black Carbon Fractions of PM0.95 Emissions by Fuel Type for Different Source Sectors

SNAP[a]	Sector Description	Solid Fuels	Light Liquid Fuels	Medium/Heavy Liquid Fuels	Gaseous Fuels	Waste/Biomass
1	energy transformation	0.005	0.32	0.52	0.9	0.25
2	small combustion sources	0.5	0.32	0.55	0.9	0.25
3	industrial combustion	0.1	0.32		0.9	0.25
5	extraction of fossil fuels				0.9	
7	road transport	0.1	0.32	0.55	0.9	
8	nonroad transport	0.1	0.32	0.55	0.9	
9	waste handling and disposal					0.005
10	agriculture					0.25

[a] SNAP, selected nomenclature air pollution.

can be approximated by the spatial patterns of population density based on the CIESIN-NOAA high resolution gridded population data (http://sedac.ciesin.org/plue/gpw). Agricultural emissions are distributed according to animal populations by NUTS 3 regions (see http://europa.eu.int/comm/eurostat/) and the distribution of arable land and pastures. To include international shipping emissions in our emission grid the $PM_{2.5}$ and BC emissions from shipping were distributed following the EMEP SO_2 ship emissions (www.emep.int).

[9] Point source data have been updated to the reference year 1995 in the CEPMEIP inventory by means of a questionnaire to national experts. About 70% of all countries responded and for the remaining countries various literature sources have been used to update point source lists. Therefore large point sources (LPS) are distributed according to their exact location and characteristics (fuel types, capacities etc.). Point sources include large combustion plants, large industrial processes (steel, nonferrous, cement etc.), waste combustors, refineries and offshore oil production platforms. The gridded emissions (0.5–0.25 longitude-latitude) as input for the LOTOS model are shown in Figure 1.

[10] To assess the concentration of primary components it is necessary to describe the temporal variation of the emissions correctly. The temporal variation of the emissions is broken down for each source category using a monthly, daily and hourly factor. These factors were developed for ozone precursor emissions and are described by *Builtjes et al.* [2003].

3. Model Description

[11] A chemistry transport model was applied to calculate the distribution of fine primary aerosol over Europe. For this purpose the LOTOS model (Long Term Ozone Simulation) has been used. LOTOS has been applied for oxidant modeling [*Builtjes*, 1992; *Hass et al.*, 1997] and for the calculation of secondary aerosol fields over Europe [*Schaap et al.*, 2004a; *Robles-Gonzalez et al.*, 2003]. In this study we separately consider the tracers black carbon and non-BC primary PM2.5, further denoted as APPM. The geographical domain of LOTOS ranges from 10°W to 40°E and from 35°N to 70°N with a spatial resolution of 0.5 × 0.25 degrees longitude-latitude. The vertical domain is divided in three layers and extends to 3.5 km. The lowest layer

Table 2. Emissions of BC and Particulate Matter Smaller Than 2.5 μm From the CEPMEIP Database[a]

SNAP[b]	Description	PM2.5, kt	Black Carbon Emission, kt	Derived Fraction Black Carbon of PM2.5[c]
1	energy transformation	256	28	0.11
2	small combustion sources	460	96	0.21
3	industrial combustion	258	64	0.25
4	industrial process emissions	197	0	0.00
5	extraction of fossil fuels	36	30	0.85
6	solvent and product use	81	0	0.00
7	road transport	332	158	0.48
	gasoline	44	14	0.32
	diesel	262	144	0.55
	volatilization losses	27	0	0.00
8	nonroad transport	160	83	0.52
9	waste handling and disposal	25	0.1	0.004
10	agriculture	83	15	0.17
11	nature	NE[d]	NE	NE
	total	1889	473	
	international shipping[e]		23	0.12

[a] Estimated emissions and for anthropogenic activities in Europe excluding the former USSR.

[b] SNAP, selected nomenclature air pollution.

[c] These fractions are the net result of a mix of activities and fuels within each category. The original calculations are based on the fractions presented in Table 1.

[d] NE, not estimated. The CEPMEIP database only considers anthropogenic sources.

[e] International sea going ships are not included in the CEPMEIP inventory.

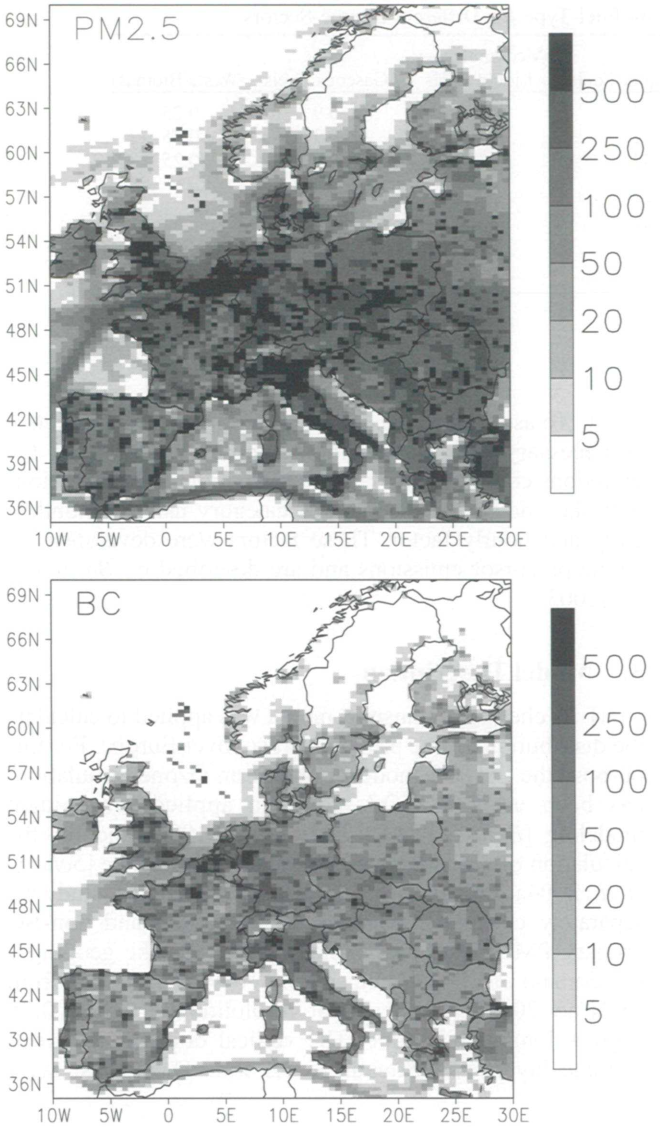

Figure 1. Emissions (tons gridbox^{-1} yr^{-1}) of PM2.5 and black carbon for 1995. A gridbox cover 0.5–0.25 degrees.

represents the variable mixing layer on top of which two layers with equal, but variable, depth are located. Hence, due to the continuously changing mixing height, the grid volumes vary in space and time. The mixing height is part of the meteorological input for 1995, which is diagnostic and derived from ECMWF using procedures developed by the Free University of Berlin [*Kerschbaumer and Reimer*, 2003]. Every hour, the mixing layer height is updated and the mass in the three layers is redistributed by mass conserving linear interpolation. The vertical extend of the model is considered sufficient since it has been shown that 80 to 90% of the aerosol burden is located below 3 km [*Banic et al.*, 1996; *ten Brink et al.*, 2001]. All boundary conditions (lateral and altitude) for BC and APPM were assumed to be zero. At the eastern boundary, however, the assumed boundary conditions and, hence, the model results are highly uncertain and hence we choose to present results only west of 30°E.

[12] In LOTOS a bulk approach is followed for the description of the aerosols, size distributions are not explicitly accounted for. Hence our model parameterizations are based upon accumulation mode aerosol since we are interested in mass concentrations. Recent modeling results using explicit aerosol dynamics show that the ageing time of BC is in the order of only a few hours [*Riemer et al.*, 2004]. We assume all BC and additional primary material (APPM) to be hydrophyllic direct after emission and thus to be present in an internal mixture with other (inorganic) aerosol components. Owing to the efficient transfer of small particles, i.e., BC, to the accumulation mode this assumption will have minor effects on our results. Further note that the assumption on hydrophilic or hydrophobic seems mainly important for the wet removal of BC/APPM aerosol, which is as we explain later, anyhow a main uncertainty in our model.

[13] Dry deposition of the accumulation mode aerosol is parameterized by the surface-atmosphere interaction module DEPAC [*Erisman et al.*, 1994]. In this module the commonly used resistance approach is followed to calculate deposition velocities. Within DEPAC the calculation of the deposition velocity for low vegetation and other areas with a roughness length below 0.5 m follows *Wesely et al.* [1985]. For areas with a roughness length above 0.5 m such as forests the parameterizations by *Erisman et al.* [1994] is used. The aerodynamic resistance, which is an input parameter of DEPAC, is calculated using the formula's given by *Seinfeld and Pandis* [1998]. From a 1.1 × 1.1 km^2 resolution land use database the fraction of surface in each grid cell covered by the land use classes used in DEPAC have been calculated [*Nijenhuis and Groten*, 1999]. For each cell the deposition velocity is calculated weighting the surface fractions of every land use class.

[14] Below cloud scavenging is represented as for the highly soluble sulphate using scavenging coefficients [*De Leeuw et al.*, 1988]. Precipitation data are obtained from our meteorological input [*Kerschbaumer and Reimer*, 2003]. Owing to our limited vertical domain and insufficient data on cloud occurrence, in-cloud scavenging is neglected. In addition, deep convection is not explicitly accounted for. Although deep convection occasionally empties the boundary layer, correct representation of this process is mainly important for simulation of aerosol levels in the free troposphere. Moreover, during conditions of deep convection (and convective rain) associated turbulence is high, which causes a higher outflow of material through the model top. Hence we assume that these processes do not strongly influence the simulation of ground-level concentrations and that these processes are (partly) compensated for by the associated higher atmospheric instability. However, further research is recommendable to better quantify the effects of these assumptions. For a more elaborate description of the model, including the secondary inorganic aerosol formation, we refer to *Schaap et al.* [2004a]. Shortly, the formation of sulphate and nitrate occurs via gas-phase and heterogeneous oxidation routes, which are included in the oxidant chemistry of the model. Ammonium nitrate formation is calculated using a thermodynamic equilibrium module.

[15] In this study we used LOTOS version 5.2 to calculate the distribution of primary emitted particles for the base year of 1995. The calculated distributions can be combined with previously calculated distributions of the inorganic

species [*Schaap et al.*, 2004a] to obtain an estimate for the total PM2.5 concentration field due to anthropogenic sources. Such a superposition is justified by the same model setup for both simulations. The contribution of anthropogenic secondary organic aerosol is neglected.

4. Results

4.1. Primary Aerosol Distribution

[16] The simulated annual averaged distribution of total primary material and its black carbon content for 1995 are presented in Figure 2. Annual averaged concentrations of primary material range up to 5 $\mu g/m^3$ over Europe. Areas with the highest concentrations are found in industrialized and/or densely populated regions, e.g., northwestern Europe, the Po valley and southern Poland. Secondary maxima can be identified over a number of large cities, such as London, Manchester, Athens, Madrid and Oslo. The local peaks over the North Sea can be explained by the emissions from oil platforms. Over more rural regions, which cover most of continental Europe, the levels of primary components are between 1 $\mu g/m^3$ and 2.5 $\mu g/m^3$. In Scandinavia concentrations trail off from about 1 $\mu g/m^3$ in the south to less then 0.25 $\mu g/m^3$ over mid and northern Scandinavia. Important ship routes are mainly located near the European continent and, hence, the concentrations of primary particles decline from the edges of the continent to the open sea due to dry and wet deposition.

[17] Modeled BC 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 the calculated BC concentrations exceed 500 ng/m^3 over central Europe and 1000 ng/m^3 in the densely populated areas (Figure 2). In western Europe the black carbon levels are about one fifth of the total primary aerosol. Over eastern Europe the BC content is calculated to be slightly less. The black carbon content of the primary aerosol shows a very similar distribution as the total primary components because the source categories with the highest PM emissions also have the highest BC fractions.

4.2. Comparison of Modeled BC Concentrations With Measured Data

[18] Monitoring data on BC (or EC) for 1995 are almost not available. Therefore we have compared our calculated BC concentrations with observations on a number of sites representing a period ranging from the end of the 1980s to 2001 and often obtained in campaigns. The measured concentrations are compared the modeled values for the corresponding months of 1995 in Table 3. The data are plotted as a function of the regional characteristic in Figure 3. The comparison shows that the simulated BC concentrations consistently underestimated those measured by about a factor of 2. The relative underestimation is found to be rather constant for remote to rural sites with measured concentrations above 0.1 $\mu g/m^3$ (Figure 3). This is an indication that the distribution of the concentrations and the main sources are well represented in the model, however, the absolute concentration is not. For urban sites the underestimation is larger, which can be explained by the high influence of local emissions, not represented in our model.

[19] For Belgium site Waasmunster data were available for 1995 (W. Maenhaut, personal communication, 2004). We compare the calculated BC concentrations to these measured values in Figure 4. The model reproduces the seasonal variation with higher concentrations in winter due to higher emissions and lower boundary layer depths in this season. Moreover, the model reproduces the measured temporal pattern based on daily values remarkably well (r = 0.79). This indicates that the most important processes governing the dynamics of the mixing layer BC concentrations are well represented in the model. In the case of Waasmunster the absolute concentrations measured are also well reproduced by the model. This agreement, however, is an exception since for all other sites a systematic underestimation is found (Table 3). A similar correlation between measured and modeled values is found for Melpitz (r = 0.7, data not shown), where the model underestimates the measured concentrations by a factor of 3–4.

5. Explaining the Total Fine Aerosol (PM2.5) Distribution Over Europe

5.1. Fine Aerosol Distribution

[20] To investigate the PM2.5 distribution over Europe we added fields of secondary inorganic components (Figure 2) calculated by *Schaap et al.* [2004a] to the primary particle simulations. Sulphate contributes more than 3 $\mu g/m^3$ to PM2.5 over a region that spans from England, over northwestern Europe to southeastern Europe. Similar contributions are found in the Po valley and the northwest of Spain. Maximum contributions are calculated in the Ruhr area, southern Poland, Hungary and Rumania. These regions are markedly different from those of the primary aerosols. North of 57°N the annual averaged sulphate concentrations are lower than 2 $\mu g/m^3$, whereas those in more remote continental areas are between 2 and 3 $\mu g/m^3$.

[21] High concentrations of nitrate are confined to continental areas with maxima in northwestern Europe, the UK and the Po valley. Annual averaged nitrate concentrations are similar to sulphate concentrations in western Europe, but sulphate concentrations are higher in (south) eastern Europe and Scandinavia. In contrast to sulphate, nitrate levels show a distinct seasonal variation. Ammonium nitrate formation is limited during summer over the largest part of Europe due to high ambient temperatures and maximum concentrations occur in winter, when ammonium nitrate is stable. Both sulphate and nitrate are neutralized by ammonium and ammonium concentrations are only somewhat lower than those of its associated ions alone.

[22] The sum of all modeled anthropogenic components of PM2.5 is shown in the lower right panel of Figure 2. Highest concentrations of PM2.5 are found in the most industrialized 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 maxima can be found in the Po valley, the Czech Republic and metropolitans of London, Manchester and Paris. Over central and southeastern Europe concentrations are calculated to range between 10 and 15 $\mu g/m^3$. Toward the north the anthropogenic induced concentration of PM2.5 trails off from about 4 in southern Scandinavia to less than 2.5 $\mu g/m^3$ further north. Also, the rural areas in Spain and southern

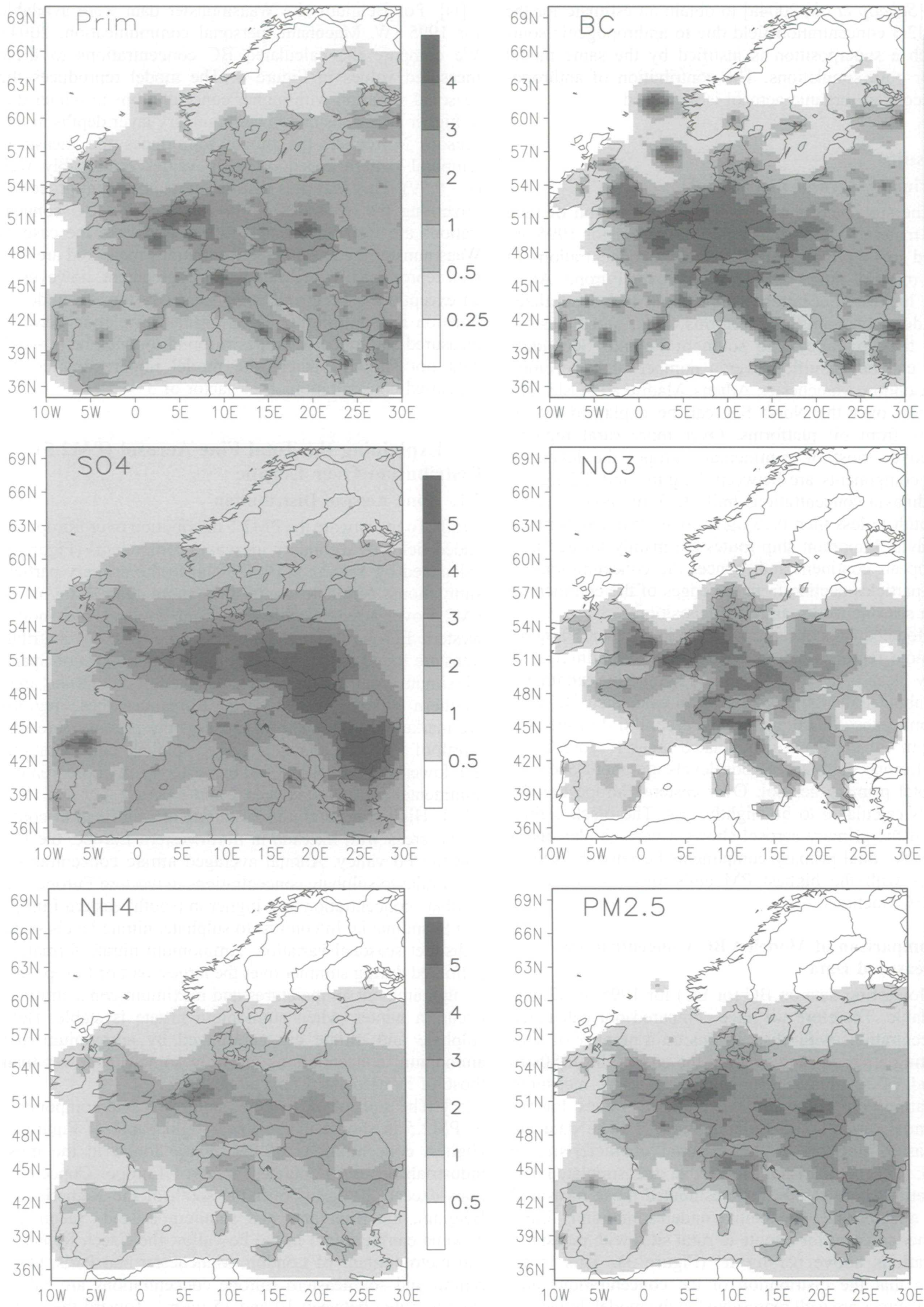

Figure 2. Annual average distribution of the indicated aerosol components (μg m^{-3}). Note that dust and sea salt are not taken into account.

Table 3. Comparison of Simulated and Observed BC Concentrations ($\mu g/m^3$)[a]

Site	Longitude	Latitude	Period	Simulated	Observed	Reference
			Background			
Mace Head	−9.5	53.2	July–August	0.10	0.03	*Krivacsy et al.* [2001]
Mace Head	−9.5	53.2	year	0.09	0.05–1.0	*Derwent et al.* [2001]
Hyytiala	24.3	61.9	June	0.15	0.19	*Alves et al.* [2002]
South Uist	−7	57	winter	0.06	0.02–0.33	*Loew et al.* [1996]
South Uist	−7	57	summer	0.05	0.03–0.08	*Loew et al.* [1996]
Finokalia	25.6	35.2	May	0.09	0.26	*Kouvarakis et al.* [2002]
Prasses	25.1	35.2	May	0.13	0.19	*Kouvarakis et al.* [2002]
Aspvreten	17.4	58.8	June–July	0.15	0.10	*Zappoli et al.* [1999]
NE Atlantic	−5.0	61.0	October–November	0.05	0.27	*O'Dowd et al.* [1993]
			Rural			
Kap Arkona	13.2	57.4	year	0.21	0.4	*Zier* [1991]
Melpitz	12	51	year	0.61	2.3	*Heintzenberg et al.* [1998]
Corsica	9	42	spring	0.23	0.38	*Cachier et al.* [1989]
Landes Forest	−1	44	autumn	0.30	0.3	*Cachier et al.* [1989]
Edgbaston	−1	51	year	0.65	0.38	*Smith et al.* [1996]
K-Puszta	19.5	46.9	July–August	0.28	0.60	*Molnar et al.* [1999]
K-Puszta	19.5	46.9	July–August	0.28	0.75	*Krivacsy et al.* [2001]
Po-Valley	11.4	44.4	September	0.63	1.0	*Zappoli et al.* [1999]
Anadia	−8.4	40.3	August	0.29	1.59	*Castro et al.* [1999]
Areao	−8.8	40.5	year	0.34	0.95	*Castro et al.* [1999]
Aveiro	−8.4	40.4	July	0.23	0.77	*Alves et al.* [2002]
Ebro	−0.2	40.5	year	0.24	0.4	*Rodríguez et al.* [2002]
Roervik	12.1	57.2	January–May	0.21	0.73	*Brostrom-Lunden et al.* [1994]
			Urban			
Vienna	16.2	48.1	year	0.83	4.9	*Hitzenberger and Tohno* [2001]
Gent	3.4	51	summer	0.77	1.7	*Kubatova et al.* [2002]
Gent	3.4	51	winter	1.24	3.2	*Kubatova et al.* [2002]
Gorlitz	14.6	41.1	year	0.87	1.3	*Zier* [1991]
Halle	12	51.3	year	0.68	1.6	*Zier* [1991]
Potsdam	13.4	52.3	year	0.73	1.0	*Zier* [1991]
Radebeul	13.6	51.6	year	0.51	1.4	*Zier* [1991]
Gif sur Yvette	2.8	48.4	year	1.03	1.6/2.7	*Cachier et al.* [1989]*Bremond et al.* [1989]
Paris	2.2	48.5	year	2.04	4.6	*Ruellan and Cachier* [2001]
Aveiro	−8.4	40.4	year	0.32	11.8	*Nunes and Pio* [1993]
Lisbon	−9.2	38.4	June	0.36	2.1	*Alves et al.* [2002]
Barcelona	2.0	41.2	year	1.76	3.0	*Rodríguez et al.* [2002]

[a] The modeled concentrations (for 1995) represent the period of the year in which the measurements were performed. For Mace Head and South Uist the range of measured data is tabulated.

France are relatively clean with respect to anthropogenic PM2.5. As mentioned before, our simulation does not consider mineral dust, which is known to contribute strongly to PM in Spain (Rodriguez et al., 2002). Further, a number of cities can be recognized by high concentrations, e.g., Madrid, Barcelona and Athens.

5.2. Composition of PM2.5 Distribution

[23] The relative contribution of the main components to the total fine aerosol mass is shown in Figure 5. Primary emitted particles (including BC) contribute 15 to 20% to the computed total fine aerosol mass over continental Europe. This fraction is rather constant but higher contributions (>30%) are found in densely populated regions. For BC the contribution to PM2.5 ranges between 3% and more than 10%. Similar to the total primary contribution, highest values occur over the most densely populated regions in Europe. The high values of BC over the North Sea are due to the presence of oil platforms there. The low (<4%) relative contribution of BC to PM2.5 over eastern Europe is striking; and coincides with the band of high sulphate concentrations. This relative BC contribution minimum is only slightly visible in the contribution of total primary material and illustrates that the BC emissions are largely due to diffuse sources such as traffic and off-road transport (e.g., river ships, trains) whereas the sulphur and, to a lesser extent, non-BC emissions have a large stationary point source component.

[24] In this simulation sulphate contributes the largest calculated mass fraction to PM2.5 over Europe. Its contribution is about 30–35% over most of continental Europe. The mass fraction increases toward more remote locations and is slightly lower in those areas where primary material and nitrate are important. The percentage of nitrate in PM2.5 over continental Europe is modeled to be 20 to 30%. Contributions over 30% are found in the regions with high ammonia emissions such as northwestern Europe and the Po valley. The nitrate contribution declines from the coast toward the open sea since in our model ammonia is rapidly deposited [*Schaap et al.*, 2004a]. In reality, the reaction of nitric acid with sea salt plays an important role by transferring nitrate from the fine to the coarse aerosol fraction [*Vignati et al.*, 1999]. Ammonium, which neutralizes both sulphate and nitrate, contributes about 15 to 25%

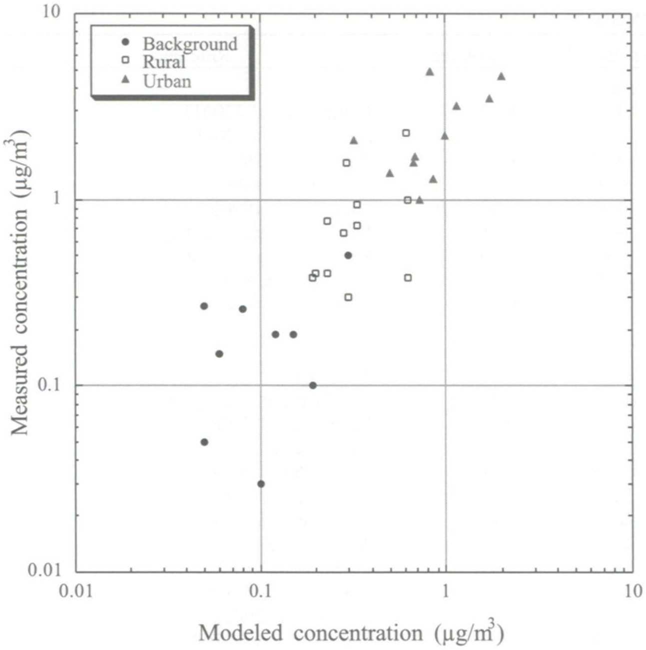

Figure 3. Comparison between modeled and measured BC concentrations μg m^{-3}) at background, rural, and urban locations.

to the fine aerosol mass; maxima in ammonium coincide with those of nitrate.

5.3. Comparison With Measured Data

5.3.1. Uncertainties in PM Sampling and Data Selection

[25] Before we compare our calculations to measured data we shortly discuss the uncertainties inherent to the sampling of PM and its composition. PM is mostly sampled using filters (Teflon, quartz, cellulose) or impactors. Several mechanisms may cause artefacts during sampling. Volatile compounds such as ammonium nitrate and a number of organic species may evaporate from inert filters [*Chow*, 1995]. On the other hand, nitric acid or organic vapors may be adsorbed to the filter or previously collected material [*Spicer and Schumacher*, 1979; *ten Brink et al.*, 2004]. In case of nitrate, artefacts are probably not significant during winter [*Schaap et al.*, 2002, 2004b]. However, evaporation from inert filters is thought to be significant at temperatures above 20 degrees Celsius [*Hering and Cass*, 1999; *Schaap et al.*, 2004b]. A field comparison for total carbon (the sum of OC and EC) at Melpitz, Germany showed differences between filter samplers of up to 20% for a 10 day average [*ten Brink et al.*, 2004]. A similar range was found for the impactors although they recovered only 75% of the TC measured on the filter samplers. The TC data agreed reasonably, however, its OC and EC content is difficult to determine. EC and BC are ideally both elemental carbon but are named according to their detection technique, thermal and optical respectively. Measured EC and BC concentrations showed differences up to a factor of three. As a consequence of these possible artefacts the determination of PM mass is associated with a rather large uncertainty. Numerous studies have described measurements of aerosol composition in Europe. Very recently, a number of studies has been compiled by *Putaud et al.* [2004]. The compilation contains data for sites at which the total particulate mass was available in combination with its chemical composition, including carbonaceous aerosols (Table 4). A part of the PM mass is unaccounted for, this is suspected to be due to water [*Putaud et al.*, 2004; *Pakkanen et al.*, 1999] and uncertainty in the conversion factor for the noncarbon atoms in organic carbon [*Putaud et al.*, 2004]. In addition, we have compared our inorganic aerosol simulations to the *European Monitoring and Evaluation Programme* (*EMEP*) [1997] and data compiled by *Schaap et al.* [2002] [*Schaap et al.*, 2004a]. Unfortunately, the EMEP data do not comprise

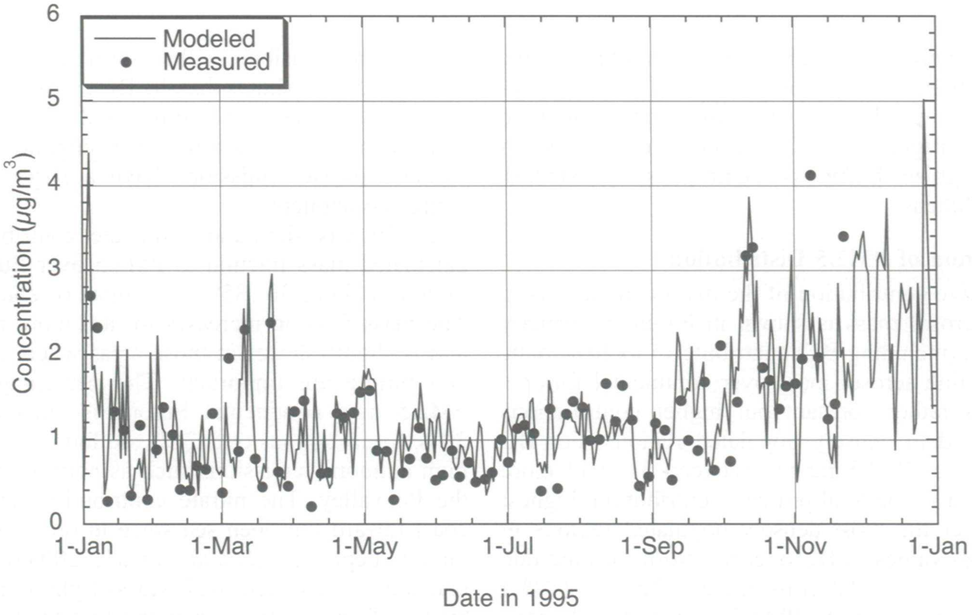

Figure 4. Comparison of measured and modeled BC concentrations (μg m^{-3}) at Waasmunster, Belgium.

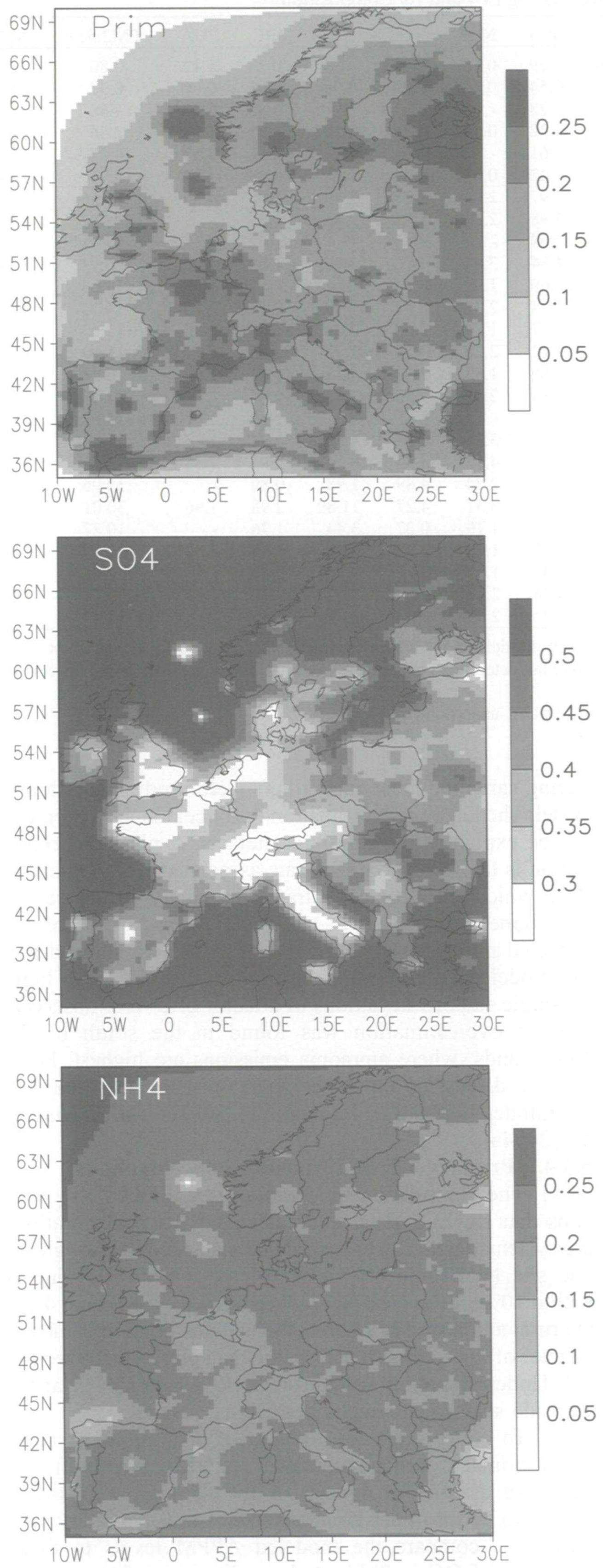

Figure 5. The relative contribution of different species to the modeled fine aerosol mass. Note that dust and sea salt are not taken into account.

Table 4. Comparison of Simulated and Measured PM2.5 Concentrations (μg/m^3) and Its Composition[a]

Category[b]	Site (Country)	Concentrations	$PM_{2.5}$	MD	SS	SO4	NH4	NO3	OC	BC	UA	PM2.5cor	NM
B	Sevettijarvi (F)	observed	3.41	0.15	0.46	1.29	0.15	0.02	-	0.23	1.12	2.80	
		simulated		-	-	0.54	0.22	0.09	0.09	0.03	0.09	0.97	1.83
B	Skreadalen (N)	observed	5.05	0.18	0.29	1.28	-	-	-	0.48	2.33	4.58	
		simulated		-	-	1.32	0.56	0.26	0.29	0.15	1.11	2.58	2.0
B	Birkenes (N)	observed	4.66	0.25	0.34	1.61	-	-	-	0.52	1.59	4.07	
		simulated		-	-	1.42	0.61	0.34	0.38	0.18	1.33	2.93	1.14
R	Illmitz (A)	observed	19.94	0.26	0.11	3.97	2.04	2.28	5.49	1.66	4.12	19.56	
		simulated		-	-	3.98	2.6	3.81	2.65	0.83	-	13.87	5.69
R	Waasmunster (B)	observed	25.58	0.98	1.13	5.04	-	-	-	1.42	15.12	23.48	
		simulated		-	-	4.84	3.84	6.96	3.92	1.38	14.72	20.94	2.54
R	Melpitz (D)	observed	16.85	1.80	-	2.69	1.99	3.43	-	-	10.83	15.04	
		simulated		-	-	5.34	2.99	3.64	1.59	0.58	2.15	14.14	0.90
U	Zuerich (CH)	observed	18.98	1.18	0.17	3.27	1.84	2.68	4.44	1.66	3.60	17.63	
		simulated		-	-	2.72	2.27	4.28	1.77	0.65	-	11.69	5.98
U	Basel (CH)	observed	17.85	1.40	0.25	3.80	1.83	2.47	4.21	1.54	2.57	16.21	
		simulated		-	-	2.84	2.41	4.58	1.87	0.7	-	12.40	3.81
U	Ghent (B)	observed	26.39	1.05	1.31	6.17	-	-	-	1.83	13.70	24.02	
		simulated		-	-	4.87	3.85	6.94	4.52	1.24	15.31	21.42	2.60
U	Bologna (I)	observed	35.82	2.89	0.84	4.62	4.12	8.54	8.24	2.91	7.55	32.10	
		simulated		-	-	3.55	2.74	4.89	1.98	0.80	-	13.96	18.1
K	Barcelona (E)	observed	34.61	3.79	0.81	5.60	3.31	4.27	11.89	2.98	1.96	30.01	
		simulated		-	-	3.61	1.19	0.27	3.44	1.76	-	10.27	19.7
K	Bern (CH)	observed	22.65	1.35	0.21	2.67	1.40	2.32	6.68	3.95	4.07	21.09	
		simulated		-	-	2.43	1.94	3.52	1.36	0.49	-	9.74	11.3
K	Vienna (A)	observed	38.51	2.37	0.41	4.44	2.18	3.57	8.62	9.10	7.84	35.74	
		simulated		-	-	4.2	2.53	3.34	1.72	0.53	-	12.32	23.4

[a] PM2.5cor gives the comparison of PM2.5 corrected for mineral dust and sea salt. Modeled non-BC primary material is assumed to consist of OC. The observed unaccounted mass (UA) is compared to those model components that were not determined in the measurements. NM is the difference between the PM2.5cor and modelled PM2.5. Observational data from *Putaud et al.* [2004].

[b] B, background; R, rural; U, urban; K, kerbside; MD, mineral dust; SS, sea salt; UA, unaccounted.

carbonaceous aerosol and PM2.5 data but do represent a larger number of sites and thus the data sets should be used complementary.

5.3.2. PM2.5

[26] The comparison of the modeled annual mean PM2.5 data against the compilation by *Putaud et al.* [2004] is shown in Table 4. Because sea salt and dust are not included in the model, this table also contains the measured PM2.5 concentration (PM2.5cor) corrected for sea salt and dust contributions. The model shows an increasing underestimation (listed as "not modeled" (NM)) with increasing PM levels. Hence underestimation is largest at the kerbside stations. The chemical data allow us to address this increasing gap in more detail.

5.3.3. Secondary Inorganic Components

[27] The secondary inorganic components compare rather well with observed data by *Putaud et al.* [2004]. Comparison is best for sulphate showing a slight underestimation of the averaged sulphate levels by 12%. Comparison with measured data from the EMEP network (www.emep.int) also revealed that the annual average modeled sulphate concentrations were slightly underestimated (6%). The correlation on a daily basis was on average reasonable ($r = 0.58$).

[28] For nitrate the picture is more complicated. Except for Barcelona the modeled values are within a factor of 2 of the measured data [*Putaud et al.*, 2004]. The model overestimates the annual average concentrations at the Swiss sites (Zurich and Basel) by 80%. However, independent data show annual average concentrations of 4.7 μg/m^3 for two sites in the Swiss valleys [*Schaap et al.*, 2002], which is close to the modeled concentrations. The difference between the measurements may be explained by losses during sampling since inert filters were used in Zurich and Basel. The large underestimation in Barcelona, however, can not be explained by sampling artefacts. A plausible explanation is the formation of nitrate associated with sodium or dust, which are the main nitrate compounds in this region in summer [*Rodríguez et al.*, 2002]. Comparison of the modeled nitrate concentrations to EMEP data showed that the modeled concentrations are on average 10% too high. At single stations deviations of a factor of 2 were observed. Largest overestimation was found in the south of the Netherlands, where ammonia emissions are highest. For a detailed discussion on the comparison with measured data for nitrate, sulphate and ammonium we refer to *Schaap et al.* [2004a].

5.3.4. Primary BC and Non-BC Concentrations

[29] The compilation by *Putaud et al.* [2004] also contains data on EC(=BC). The calculated BC concentrations are systematically lower than those measured. Although for one site (Waasmunster, Belgium) the calculated value is within 10% of the measured concentration (see Figure 4), at the rural and near city stations the underestimation is mostly a factor of 2–3, consistent with the data presented in section 4.2. Underestimation is highest, up to a factor of 9, at the kerbside stations of Vienna and Bern.

[30] To address the role of non-BC emissions or additional primary particulate matter (APPM), which in this work we assume to consist of mostly OC, we assume APPM to be an upper boundary estimate for primary OC. Hence we compare the modeled APPM levels to those measured of OC in Table 4. In reality, secondary organic aerosols (SOA, neglected in this study) also contribute to measured OC levels. However, in polluted areas such as continental Europe the contribution of biogenic SOA is

expected to be small and the formation of anthropogenic SOA is ineffective [*Schell et al.*, 2001]. The comparison of APPM and the OC concentrations compiled by *Putaud et al.* [2004] show an underestimation similar to BC.

[31] The measured ratio of OC to BC ranges from 0.95 to 4.0. Assuming the APPM fraction of PM2.5 to be OC, the OC to BC emission ratio for the European countries as a whole is on average 3. As this ratio for emissions resulting from the transport sector is about 1 (see Table 2), the concentration ratio in areas with large traffic emissions (e.g., cities) could be significantly lower than 3, approaching 1 at sites where traffic emissions dominate. Such a tendency is indeed observed with the exception of Barcelona. Modeled and measured ratios at urban locations are very similar and range between 2.5 and 2.8. At the kerbside stations (e.g., Vienna) similar ratios are simulated as for the urban background sites, which is to be expected for a regional model. Thus, whereas the modeled primary aerosol concentrations are underestimated, the relative amounts of modeled APPM and BC appear to be quite consistent with observations. As indicated above, assuming APPM to be OC induces uncertainty in this observation. However, we feel that the uncertainty present in the measured OC to BC ratios is much larger than that induced by the APPM = OC assumption.

[32] The availability of measured chemical speciation of PM is crucial to verify the modeled PM levels and its components. However, not always all components of PM2.5 are determined for all sites (Table 4). For instance for the Belgian sites, Waasmunster and Ghent, the nitrate, ammonium and OC contents of PM2.5 were not measured and the modeled concentrations of these unaccounted species can largely fill the gap and thus help interpreting the PM2.5 data at these sites.

6. Discussion

[33] The comparison between modeled and measured BC concentrations strongly suggests that the model underestimates BC concentrations. Since the inorganic ions are simulated reasonably well [see also *Schaap et al.* [2004a] and discussion therein), it could be deduced that the underestimation of PM2.5 levels is mainly caused by primary carbonaceous particles. As the model performance of the primary components is much less favorable compared to measurements than that of the secondary inorganic species, the discussion presented below focuses on aspects that are of specific interest to explain the systematic difference between modeled and measured concentrations of carbonaceous components.

6.1. How to Compare Modeled and Measured Concentrations

[34] The BC data compiled in this study cover a large time span (1980s–2001). Data from campaigns of only a few months may be influenced by the specific meteorological conditions prevailing during the campaign. In addition, inter annual variability and possible trends in BC emissions may introduce systematic errors and variability in the model to measurement comparison. In addition, recent intercomparison tests show that BC measurements with different techniques are associated with an uncertainty of about a factor of 3 [*ten Brink et al.*, 2004]. Moreover, EC/BC data may be systematically too high, e.g., due to the charring of organic material [*Schmid et al.*, 2001]. However, regardless of technique or observational period, the model results show quite consistently an underestimation of a factor of two or more. Although the model values are mostly just within the factor 3 uncertainty range of the observations, it is worrying that the differences appear to be systematic where a random difference is expected. Moreover, total carbon concentrations, which are measured with a much higher accuracy of about 30% [*ten Brink et al.*, 2004], are also underestimated in much the same way. Since the measured and modeled BC to TC ratios are very similar, we conclude that the modeled BC values are too low. This conclusion appears robust and not obscured by the measurement uncertainties.

[35] The modeled concentrations represent grid box average concentrations. Although the tendency for increasing deviation between the modeled and measured data going from rural to urban sites can be explained by the influence of local sources, the modeled BC concentrations for the available background sites and rural sites are also associated with a systematic underestimation of a factor of 2. The influence of local emissions is assumed to be small in these locations suggesting that the representativity of the stations also does not explain our finding that the modeled values are systematically too low (Figure 3).

[36] Uncertainties in the removal of BC may also affect our results. Coating or oxidation of a BC particle, i.e., aging, changes its physical and chemical properties, most notably its solubility in water and its ability to form cloud droplets. The timescale at which BC becomes hydrophilic determines its lifetime in the atmosphere against wet deposition. The ageing timescale is under scientific debate and ranges from an hour [*Köhler et al.*, 2001; *Riemer et al.*, 2004] to 1.5 day [*Cooke and Wilson*, 1996; *Wilson et al.*, 2001]. In this work we assume BC as well as the non-BC primary PM2.5 (APPM) to be hydrophilic instantaneously. Since we neglect in-cloud scavenging in our model wet deposition is not very effective [*Schaap et al.*, 2004a] and the wet deposition rates of BC/APPM will most probably be underestimated, regardless of the aging timescale. On the other hand, the influence of the mixing state on the efficiency of below cloud scavenging is not known, but we feel this is a secondary effect. Nevertheless, the influence of the ageing process on the lifetime of BC with respect to (in-cloud) scavenging is an important issue and needs to be resolved. Unfortunately, our model configuration is not suitable for this purpose. Therefore a more detailed study with a state of the art aerosol model using explicit aerosol dynamics is needed to assess the ageing timescale and its effects under ambient conditions prevailing in Europe.

6.2. Comparison With Previous and Alternative Emission Inventories

[37] The black carbon emission data used (Table 2) can be compared with emission estimates presented in previous studies (Table 5). The year for which the emissions have been estimated is an important variable in such a comparison because PM2.5 and associated black carbon emission are expected to change considerably over time due to

Table 5. Overview of Black Carbon Emissions Estimates for Europe and the Former USSR

	Black Carbon, Tg yr^{-1}				
Year	Europe	Former USSR	Aerosol	Remark	Reference
1980	3.80	5.60	bulk	based on BC/SO$_2$ ratio's	*Penner et al.* [1993]
1980	2.14	1.93	bulk	based on emission factors and fuel use	*Penner et al.* [1993]
1984	2.65	1.55	bulk		*Cooke and Wilson* [1996]
1984	1.74	1.07	bulk	fossil fuel BC emission only	*Cooke et al.* [1999]
1984	1.26	0.69	submicron	fossil fuel BC emission only	*Cooke et al.* [1999]
1996	0.44 (0.33–1.20)	0.36 (0.22–1.24)	submicron		*Bond et al.* [2004]
1995	0.47 (0.23–0.70)	0.26 (0.12–0.40)	submicron	only anthropogenic sources	this work

economical and technological changes. Furthermore, it is important to consider the aerosol size, e.g., bulk versus submicron aerosol, which is covered by each emission inventory. This can be illustrated with the data from *Cooke et al.* [1999] for 1984 that indicate that about 70% of the bulk aerosol black carbon is in the submicron range. The contribution of the submicron range to the bulk aerosol has probably increased over time in Europe due to a further increase in air pollution control measures, which tend to capture coarse particles more efficiently.

[38] The emission estimates in Table 5 suggest a strong decline of black carbon emissions between 1980 and 1995 for both Europe and the former Soviet Union (FSU). Emission estimates for Europe range from 2.1–3.8 Tg C in 1980 [*Penner et al.*, 1993] to 0.47 Tg C in 1995 (this study). This decline maybe partly real due to technological improvements, air quality control measures and economical and political changes, most notably those in Eastern Europe and the USSR, which resulted in a decline of emissions after 1989 (see below). However, the lower emission estimates may also largely be caused by a more accurate assessment of the PM emitting activities in relation to their technological level, the resulting PM emissions and their fractional black carbon content. In the past such improvements resulted in revisions of inventories. For example, *Cooke et al.* [1999] revised the 1984 inventory of *Cooke and Wilson* [1996] resulting in a 30% lower emission estimate.

[39] Recently, *Bond et al.* [2004] presented a new bottom up BC emission inventory using the latest available emission factors. Their European emission estimate (0.44 Tg C) is very similar to our estimate. The range reported here is narrower than reported by *Bond et al.* [2004] most likely because (1) the CEPMEIP database provides a relatively good insight in the technological level, which has a large impact on the resulting emissions, of different European countries and facilities within a country and (2) *Bond et al.* [2004] use a different procedure for quantifying the uncertainty range (adding up the standard deviations for each source instead of the variances). The uncertainty range in our estimates of total BC emissions from Europe and FSU has been estimated from two components: the uncertainty in submicron particle emissions which has been assessed at ±20% [*Visschedijk*, 2002] and the uncertainties in the BC fractions for the different fuel categories as reported in Table 1.

[40] The more recent estimates such as *Bond et al.* [2004] and this study compare very well, but are more than a factor of 2 lower than the previous estimates. The recent and previous estimates are not necessarily in contradiction with each other since the uncertainty of the previously reported data is at least a factor of 2 [e.g., *Liousse et al.*, 1996; *Cooke et al.*, 1999]. However, our model results under-predict the observed BC concentrations (see also Table 3), whereas the results from studies using the higher emission estimates agreed better with observations, especially at rural and background locations [*Cooke et al.*, 1999; *Jacobson*, 2001; *Chung and Seinfeld*, 2002]. This poses an intriguing question: Are the previous (higher) emission estimates better than our inventory and if not, why do these previous emission estimates than result in more realistic simulation results?

[41] The current emission inventories are bottom-up inventories using the relatively good insight in PM2.5 emissions to derive BC emissions. The BC emissions can be derived using the available data on the BC fraction of combustion-derived PM because we have knowledge on fuel types used in different sectors and activities. It is important to realize that this is the proper way to derive the BC emissions because the fuel type and the way it is consumed are causal factors in the process of BC production and subsequent emission. Hence we are confident that the approach taken in the current emission inventories is an improvement. However, still, why do previous inventories produce a better match with observational data? A part of the discrepancy might be explained by the year of study. C. Liousse et al. (manuscript in preparation, 2004) revisited the data of *Cooke et al.* [1999] for 1984 and extended the emission inventory to 1997 by calculating the BC emission with the annual fuel consumption and activity data in combination with the original emission factors by *Cooke et al.* [1999] (Table 4). The revisited emission inventory for 1984 (1.22 Tg yr^{-1}) is somewhat lower than the original figure (1.26 Tg yr^{-1}) by *Cooke et al.* [1999] but these differences can be attributed to correction of algorithm errors. The extension to 1997 showed that BC emissions increased from 1984 to 1992 (Table 6), but decreased afterward due to the political and economical changes in eastern Europe. More important, by changing only the fuel use and activity data the calculated BC emission are very similar for 1984 and 1995. The emissions from domestic sources decrease significantly from 1984–1995 but this is mostly compensated by the steady increase of the combined traffic section (Table 6). Industrial emissions are very similar in both years. Since, the emission changes between 1984 and 1995 are due to changing activities and fuel use only, the choice of emission factors appears to be crucial to explain the difference between the reanalysis value (1.22 Tg) and our estimate (0.47 Tg). Currently, evaluation of the emission factors used in both inventories is ongoing and will be reported separately.

6.3. Traffic Emissions

[42] *Bond et al.* [2004] suggested two major reasons for possible underprediction of BC emissions: fuel use and

Table 6. European BC Emission (Tg C yr^{-1}) Based on a Re-Evaluation of the 1984 Data From *Cooke et al.* [1999] and an Extension of the Fuel Consumption and Activity Data to the Year 1992 and 1995[a]

Sector/Year	1984	1992	1995	This Study
Combined traffic	0.264	0.340	0.457	0.241
Domestic	0.537	0.507	0.361	0.096
Industrial	0.421	0.546	0.465	0.137
Total	1.222	1.393	1.283	0.474

[a] From C. Liousse et al. (manuscript in preparation, 2004).

emission factors. Underreporting of fuel use may occur when a portion of the fuel supply (e.g., wood or coal) does not pass through official channels, or when some "fuels" are not considered at all (e.g., house fires, waste paper). Emission factors may be underestimated if measurements pertain to better technology or more careful practice than the average. Increased emission factors might be associated with transient operation, poor quality or adulterated fuels, or badly maintained units. The latter circumstances also pertain to traffic emissions. Earlier studies [*Köhler et al.*, 2001; *Cooke et al.*, 1999] calculated the BC emissions for road traffic from the fuel consumption data and BC emission indices (EI_{BC}). Two EI_{BC} are used: 2 en 10 g C (kg fuel)$^{-1}$ for OECD countries and "Rest of the world", respectively by *Köhler et al.* [2001] and *Cooke et al.* [1999]. The uncertainty in the emissions from road traffic caused by the use of these two distinctly different EI will be high. As indicated by *Köhler et al.* [2001], the fuel consumption is reasonably well known but BC emission indices depend on vehicle type, maintenance, age, driving conditions etc., and therefore the EI_{BC} are highly variable. The use of the BC emission indices of *Köhler et al.* [2001] and *Cooke et al.* [1999] is transparent and may give a reasonable result on a global scale where the largest gradient will be between OECD and non-OECD countries. However, in a regional (i.e., European) study more differentiation according to technology level, such as vehicle fleet age is likely to give a better representation of actual emissions and gradients within the region of study. Instead of directly applying BC emission indices, the BC emission can be derived from the PM traffic emissions as is done in the present study (see Table 1). For PM better traffic emission data are available, derived from fuel consumption data and PM emission factors.

[43] There are presently various sets of PM emission factors for mobile sources e.g., for the Netherlands [*Netherlands Ministry of Housing, Spatial Planning and the Environment*, 2002], Germany [*INFRAS*, 1999] or Europe [*Ntziachristos and Samaras*, 1999]. The proposed emission factors are influenced by, e.g., sampling strategies, definitions of standard driving cycles, assumed effect of motor aging and may differ substantially for some source categories such as diesel-fuelled vehicles. In the CEPMEIP inventory (http://www.air.sk/tno/cepmeip/) emission factors for diesel-fuelled vehicles were taken from the German inventory [*INFRAS*, 1999], whereas the Dutch emission factors [*Netherlands Ministry of Housing, Spatial Planning and the Environment*, 2002] were taken for all other vehicles. These results were found to be in line with the COPERT estimates for country totals [*Ntziachristos and Samaras*, 1999] and IIASA model results (T. Pulles, 2003, personal communication). However, the selected emission factors are not necessarily the most representative. By applying different emission factors from the same sources the uncertainty surrounding the estimate for total traffic-related PM emissions was found to amount to a factor of ~2. Furthermore, the contribution of gasoline vehicles to daily PM levels may be substantial especially due to cold starts [*Watson et al.*, 1998], suggesting that PM emissions due to gasoline vehicles may be underestimated. In general, it can be concluded that the emission factor methodologies for traffic need to be harmonized and further validated to come to an accurate and consistent European set of traffic emission factors, which is however outside the scope of the present paper.

6.4. Unaccounted Sources

[44] A possible underestimation of the traffic BC emissions is caused by the PM2.5 particle emissions and not their BC fraction. Although traffic is a major source of primary particles, there are more uncertainties in the PM2.5 emissions. In Table 7 we present emission estimates for the United States [*Battye et al.*, 2002] from sources that are not present in the CEPMEIP database. The most important

Table 7. Relative Contribution of Sources Not Included in Table 2 to Reported U.S. PM2.5 Emissions, BC to PM2.5 Ratio for These Sources and Derived Relative Contribution to Total Estimated U.S. Black Carbon Emissions

Category	Fraction of Reported U.S. PM2.5 Emissions,[a] %	BC to PM2.5 Ratio		Fraction of Total Estimated BC Emissions	
		Best Estimate	High Estimate	Best Estimate,[b] %	High Estimate,[b] %
Industrial-metals processing	1.6	0.1	0.3	2.1	3.9
Industrial-asphalt manufacture	0.1			0.0	0.6
Rubber and plastics products	0.0			0.0	0.3
Fugitive dust-unpaved roads	21.8	0.01	0.019	2.8	3.3
Fugitive dust-paved roads	10.5	0.017	0.028	2.5	2.3
Fugitive dust-construction	6.0		0.005	0.0	0.3
Fugitive dust-other	2.3	0.006	0.013	0.2	0.3
Subtotal contribution of the above sources to reported U.S. emissions	42.3			7.6	10.9
Wildfires	3.6	0.072	0.12	3.5	3.4
Prescribed forest burning	8.1	0.072	0.12	7.9	7.7

[a] Total reported U.S. PM2.5 emissions is 5894 Gg yr^{-1} [*Battye et al.*, 2002].

[b] Best estimate BC emissions 433 Gg yr^{-1}, high estimate BC emissions 727 Gg yr^{-1} [*Battye et al.*, 2002].

source missing in CEPMEIP is fugitive dust accounting ~40% of the total PM2.5 emissions in the United States. Although these emissions are difficult to extrapolate to Europe due to different road and climatic conditions, it indicates that the European PM2.5 emission may be underestimated significantly, especially in southern Europe where climatic conditions compare best with the United States. The most important missing industrial source is metal processing which contributes only 2% of the total fine particle emissions in the United States. For BC, these missing sources contribute only 7–11% of the total U.S. emission and are, therefore, not likely to explain a large underestimation of the European BC emissions.

[45] In addition, the emissions from (controlled) vegetation fires are unaccounted for. Prescribed and/or intentional burning of grasslands and forest does not or hardly occur in Europe and is not considered in our black carbon emission inventory. Burning of agricultural residue has been considered although its importance has rapidly decreased in recent years due to policies to ban residue burning. On the other hand, accidental or lightning-ignited fires may cause an underestimation of the BC emissions in Europe. In the United States these emissions are thought to be about 8% of the total emission [*Battye et al.*, 2002]. *Lavoue et al.* [2000] estimated the European emissions from wildfires to be 0.01 Tg in 1995. The annual and seasonal variation in wild fire emissions is large [*Schultz*, 2002; *Lavoue et al.*, 2000] and the contribution may be 2 to 3 times higher in other years. The majority of the emissions occur in southern Europe, where the influence may be significant during specific episodes with forest fires. However, for Europe as a whole the contribution of wild fires to the BC emissions is expected to be small (2–5%).

[46] The unaccounted sources may contribute largely to PM2.5 emissions but probably less than 15% to the European BC emissions. We therefore conclude that the emission factors in general and not only for traffic are associated with the highest uncertainty and need to be further addressed in the future.

7. Conclusions and Recommendations

[47] We presented a model study dedicated to European BC and PM2.5 levels. We used the PM emissions from CEPMEIP (http://www.air.sk/tno/cepmeip/) and combined those with estimated BC fractions to derive a European BC emission inventory on 0.5–0.25° longitude-latitude for the year 1995. The BC emissions were estimated to be 0.47 Tg in Europe and 0.26 Tg in the former Soviet Union (FSU). Transport and small combustion sources (households) are the major sources of BC in Europe and cause these emissions to have a very diffuse nature.

[48] Calculated distributions of primary particles were combined with those previously calculated for secondary inorganic components. Sulphate was found to contribute most to the total PM2.5 mass, followed by nitrate and total primary material. The BC content of PM2.5 was estimated to range between 4 and 10%. The modeled PM2.5 concentrations underestimate measured concentrations, especially as compared with urban stations. The gap between modeled and measured PM2.5 concentrations appears to be strongly correlated with the primary carbonaceous (BC and OC) particles. BC concentrations on background and rural locations are often underestimated by a factor of 2. Since in-cloud scavenging is neglected in the model the assumption that BC is hydrophyllic direct after emission can not explain the discrepancies, although it may cause some uncertainties in our below cloud aerosol scavenging efficiencies.

[49] Local emissions cause an underestimation of primary pollutants in the source regions but not in remote locations. More importantly, the estimated emissions of BC and total particulate matter may be too low as indicated by the similar underestimation of BC and TC (BC + OC). Unaccounted sources are estimated to cause a maximum underestimation of the BC emissions of 15%. In comparison with some other studies our estimate is a factor of two lower, which is most likely caused by the choice of emission factors.

[50] At this moment we cannot conclude with certainty that we underestimate the emissions of BC in Europe, because the underestimation may be explained by a number of other factors, such as local emissions and large measurement uncertainties. However, the most important uncertainty is associated with the choice of emission factors, most notably for traffic. Therefore a better knowledge of emission factors including accessible documentation describing the conditions for which the emission factors are valid is urgently needed to estimate the BC (and PM) emissions. This may be a major prerequisite to understand the differences between the various inventories and possibly close the gap between modeled and observed concentrations of carbonaceous aerosols.

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References

Alves, C., A. Carvalho, and C. Pio (2002), Mass balance of organic carbon fractions in atmospheric aerosols, *J. Geophys. Res.*, *107*(D21), 8345, doi:10.1029/2001JD000616.

Banic, C. M., W. R. Leaitch, G. A. Isaac, M. D. Couture, L. I. Kleinman, S. R. Springston, and J. I. MacPherson (1996), Transport of ozone and sulfur to the North Atlantic atmosphere during the North Atlantic Regional Experiment, *J. Geophys. Res.*, *101*(D22), 29,091–29,104.

Battye, W., K. Boyer, and T. G. Pace (2002), Methods for improving global inventories of black carbon and organic carbon particulates, paper presented at 11th International Emission Inventory Conference: Emission Inventories—Partnering for the Future, Environ. Protect. Ag., Atlanta, Ga., 15–18 Apr.

Bond, T. C., R. J. Charlson, and J. Heintzenberg (1998), Quantifying the emission of light-absorbing particles: Measurements tailored to climate studies, *Geophys. Res. Lett.*, *25*, 337–340.

Bond, T. C., D. G. Streets, K. F. Yarber, S. M. Nelson, J. Woo, and Z. Klimont (2004), A technology-based global inventory of black and organic carbon emissions from combustion, *J. Geophys. Res.*, *109*, D14203, doi:10.1029/2003JD003697.

Bremond, M. P., H. Cachier, and P. Buat-Menard (1989), Particulate carbon in the Paris region atmosphere, *Environ. Technol. Lett.*, *10*, 339–346.

Brostrom-Lunden, E., A. Lindskog, and J. Mowrer (1994), Concentrations and fluxes of organic compounds in the atmosphere of the Swedish west coast, *Atmos. Environ.*, *28*, 3605–3615.

Brunekreef, B. (1997), Air pollution and life expectancy: Is there a relation?, *Occup. Environ. Med.*, *54*, 781–784.

Builtjes, P. J. H. (1992), The LOTOS (Long Term Ozone Simulation) project: Summary report, *Rep. TNO-MW-R92/240*, Neth. Org. for Appl. Sci. Res., The Hague.

Builtjes, P. J. H., M. van Loon, M. Schaap, S. Teeuwisse, A. J. H. Visschedijk, and J. P. Bloos (2003), Project on the modelling and verification of ozone reduction strategies: Contribution of TNO-MEP, *Rep. TNO-MEP-R2003/166*, Neth. Org. for Appl. Sci. Res., Apeldoorn.

Cachier, H., M. P. Bremond, and P. Buat-Menard (1989), Determination of atmospheric soot carbon with a simple thermal method, *Tellus, Ser. B*, *41*, 379–390.

Castro, L. M., C. A. Pio, R. M. Harrison, and D. J. T. Smith (1999), Carbonaceous aerosol in urban and rural European atmospheres: Estimation of secondary organic carbon concentrations, *Atmos. Environ.*, *33*, 2771–2781.

Chow, J. C. (1995), Measurement methods to determine compliance with ambient air quality standards for suspended particles, *J. Air Waste Manage. Assoc.*, *45*, 320–382.

Chung, S. H., and J. H. Seinfeld (2002), Global distribution and climate forcing of carbonaceous aerosols, *J. Geophys. Res.*, *107*(D19), 4407, doi:10.1029/2001JD001397.

Cooke, W. F., and J. J. N. Wilson (1996), A global black carbon aerosol model, *J. Geophys. Res.*, *101*, 19,395–19,409.

Cooke, W. F., C. Liousse, H. Cachier, and J. Feichter (1999), Construction of a $1' \times 1'$ fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model, *J. Geophys. Res.*, *104*, 2137–2162.

De Leeuw, F. A. A. M., H. J. van Rheineck Leyssius, K. D. van den Hout, H. S. M. A. Diederen, M. Berens, and W. A. H. Asman (1988), *RIVM Rep. 228471004*, Natl. Inst. of Public Health and the Environ., Bilthoven.

Derwent, R. G., D. B. Ryall, S. G. Jennings, T. G. Spain, and P. G. Simmonds (2001), Black carbon aerosol and carbon monoxide in European regionally polluted air masses at Mace Head, Ireland during 1995–1998, *Atmos. Environ.*, *35*, 6371–6378.

Dockery, D. W., C. A. Pope, X. Xu, J. D. Spengler, J. H. Ware, M. E. Fay, B. G. Ferris, and F. E. Speizer (1993), An association between air pollution and mortality in six US cities, *N. Engl. J. Med.*, *329*, 1753–1759.

Echalar, F., P. Artaxo, J. V. Martins, M. Yamasoe, F. Gerab, W. Maenhaut, and B. Holben (1998), Long-term monitoring of atmospheric aerosols in the Amazon Basin: Source identification and apportionment, *J. Geophys. Res.*, *103*, 31,849–31,864.

Erisman, J. W., A. van Pul, and P. Wyers (1994), Parameterization of surface-resistance for the quantification of atmospheric deposition of acidifying pollutants and ozone, *Atmos. Environ.*, *28*, 2595–2607.

European Monitoring and Evaluation Programme (EMEP) (1997), Data report 1995, part 1: Annual summaries, *EMEP/CCC Rep. 4/97*, Nor. Inst. for Air Res., Oslo.

Grant, K. E., C. C. Chuang, A. S. Grossman, and J. E. Penner (1999), Modeling the spectral optical properties of ammonium sulfate and biomass aerosols: Parameterization of relative humidity effects and model results, *Atmos. Environ.*, *33*, 2603–2620.

Hansen, J. E., and M. Sato (2001), Trends of measured climate forcing agents, *Proc. Natl. Acad. Sci. USA*, *98*(26), 14,778–14,783.

Hass, H., P. J. H. Builtjes, D. Simpson, and R. Stern (1997), Comparison of model results obtained with several European air quality models, *Atmos. Environ.*, *31*, 3259–3297.

Haywood, J. M., D. L. Roberts, A. Slingo, J. M. Edwards, and K. P. Shine (1997), General circulation model calculations of the direct radiative forcing by anthropogenic sulfate and fossil-fuel soot aerosol, *J. Clim.*, *10*, 1562–1577.

Heintzenberg, J., K. Muller, W. Birmili, G. Spindler, and A. Wiedensohler (1998), Mass-related aerosol properties over the Leipzig basin, *J. Geophys. Res.*, *103*, 13,125–13,135.

Hering, S., and G. Cass (1999), The magnitude of bias in the measurement of PM2.5 arising from volatilisation of particulate nitrate from Teflon filters, *J. Air Waste Manage. Assoc.*, *49*, 725–733.

Hitzenberger, R., and S. Tohno (2001), Comparison of black carbon (BC) aerosols in two urban areas: Concentrations and size distributions, *Atmos. Environ.*, *35*, 2153–2167.

Hoek, G., B. Brunekreef, S. Goldbohm, P. Fischer, and P. A. van den Brandt (2002), Association between mortality and indicators of traffic-related air pollution in the Netherlands: A cohort study, *The Lancet*, *360*, 1203–1209.

INFRAS (1999), *Handbuch Emissionsfaktoren des Straßenverkehrs, Version 1.2* [CD-ROM], Zurich.

Jacobson, M. Z. (2001), Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, *Nature*, *409*(6821), 695–697.

Jacobson, M. Z. (2002), Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming, *J. Geophys. Res.*, *107*(D19), 4410, doi:10.1029/2001JD001376.

Kerschbaumer, A., and E. Reimer (2003), Meteorological data preparation for the REM/CALGRID-model (in German), *UBA Proj. 29943246*, Freie Univ. Berlin, Berlin.

Kiehl, J. T., and B. P. Briegleb (1993), The relative roles of sulfate aerosols and greenhouse gases in climate forcing, *Science*, *260*, 311–314.

Köhler, I., M. Dameris, I. Ackermann, and H. Hass (2001), Contribution of road traffic emissions to the atmospheric black carbon burden in the mid-1990s, *J. Geophys. Res.*, *106*, 17,997–18,014.

Kouvarakis, G., Y. Doukelis, N. Mihalopoulos, S. Rapsomanikis, J. Sciare, and M. Blumthaler (2002), Chemical, physical, and optical characterization of aerosols during PAUR II experiment, *J. Geophys. Res.*, *107*(D18), 8141, doi:10.1029/2000JD000291.

Krivacsy, Z., A. Hoffer, Z. Sarvari, D. Temesi, U. Baltensperger, S. Nyeki, E. Weingartner, S. Kleefeld, and S. G. Jennings (2001), Role of organic and black carbon in the chemical composition of atmospheric aerosol at European background sites, *Atmos. Environ.*, *35*, 6231–6244.

Kubatova, A., R. Vermeylen, M. Claes, J. Cafmeyer, and W. Maenhaut (2002), Organic compounds in urban aerosols from Gent, Belgium: Characterization, sources, and seasonal differences, *J. Geophys. Res.*, *107*(D21), 8343, doi:10.1029/2001JD000556.

Lavoue, D., C. Liousse, H. Cachier, B. J. Stocks, and J. G. Goldammer (2000), Modeling of carbonaceous particles emitted by boreal and temperate wildfires at northern latitudes, *J. Geophys. Res.*, *105*, 26,871–26,890.

Liousse, C., J. E. Penner, C. Chuang, J. J. Walton, H. Eddleman, and H. Cachier (1996), A global three-dimensional model study of carbonaceous aerosols, *J. Geophys. Res.*, *101*, 19,411–19,432.

Loew, J. A., M. H. Smith, B. M. Davison, S. E. Benson, M. K. Hill, C. D. O'Dowd, R. M. Harrison, and C. N. Hewitt (1996), Physicochemical properties of atmospheric aerosol at South Uist, *Atmos. Environ.*, *30*, 3765–3776.

Menon, S., J. Hansen, L. Nazarenko, and Y. Luo (2002), Climate effects of black carbon aerosols in China and India, *Science*, *297*, 2250–2253.

Molnar, A., E. Meszaros, H. C. Hansson, H. Karlsson, A. Gelencser, G. Y. Kiss, and Z. Krivacsy (1999), The importance of organic and elemental carbon in the fine atmospheric aerosol particles, *Atmos. Environ.*, *33*, 2745–2750.

Myhre, G., F. Stordal, K. Restad, and I. S. A. Isaksen (1998), Estimation of the direct radiative forcing due to sulfate and soot aerosols, *Tellus, Ser.*, *50*, 463–477.

Netherlands Ministry of Housing, Spatial Planning and the Environment (2002), Methods to calculate the emissions of mobile sources in the Netherlands, *Rapp. MilieuMonitor 4*, The Hague.

Nijenhuis, W. A. S., and S. M. E. Groten (1999), Surface cover related input data for 3-D atmospheric-chemistry models, *NRSP-2 Proj. 4.1/DE-01*, Umweltbundesamt (UBA), Berlin.

Ntziachristos, L., and Z. Samaras (1999), COPERT III computer programme to calculate emissions from road transport—Methodology and emission factors, Final draft report, Eur. Environ. Ag., Copenhagen.

Nunes, T. V., and C. A. Pio (1993), Carbonaceous aerosols in industrial and coastal atmospheres, *Atmos. Environ. Part A*, *27*, 1339–1346.

O'Dowd, C. D., M. H. Smith, and S. G. Jennings (1993), Submicron particle, radon, and soot carbon characteristics over the northeast Atlantic, *J. Geophys. Res.*, *98*, 1123–1135.

Pakkanen, T. A., et al. (1999), Nordic intercomparison for measurement of major atmospheric nitrogen species, *J. Aerosol Sci.*, *30*, 247–263.

Penner, J. E., H. Eddleman, and T. Novakov (1993), Towards the development of a global inventory for black carbon emissions, *Atmos. Environ.*, *27*, 1277–1296.

Penner, J. E., C. C. Chuang, and K. Grant (1998), Climate forcing by carbonaceous and sulfate aerosols, *Clim. Dyn.*, *14*, 839–851.

Pope, C. A., M. J. Thun, M. M. Namboodiri, D. W. Dockery, J. S. Evans, F. E. Speizer, and C. W. Heath (1995), Particulate air pollution as a predictor of mortality in a prospective study of U.S. adults, *Am. J. Respir. Crit. Care Med.*, *151*, 669–674.

Putaud, J.-P., et al. (2004), A European aerosol phenomenology part 2: Chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, *Atmos. Environ.*, *38*, 2579–2595.

Riemer, N., H. Vogel, and N. Vogel (2004), A parameterisation of the soot aging for global climate models, *Atmos. Chem. Phys. Discuss.*, *4*, 2089–2115.

Robles-Gonzalez, C., J. P. Veefkind, and G. de Leeuw (2000), Aerosol optical depth over Europe in August 1997 derived from ATSR-2 data, *Geophys. Res. Lett.*, *27*, 955–958.

Robles-Gonzalez, C., M. Schaap, G. de Leeuw, P. J. H. Builtjes, and M. van Loon (2003), Spatial variation of aerosol properties derived from satellite observations, *Atmos. Chem. Phys.*, *3*, 521–533.

Rodríguez, S., X. Querol, A. Alastuey, and F. Plana (2002), Sources and processes affecting levels and composition of atmospheric aerosol in the western Mediterranean, *J. Geophys. Res.*, *107*(D24), 4777, doi:10.1029/2001JD001488.

Ruellan, S., and H. Cachier (2001), Characterization of fresh particulate vehicular exhausts near a Paris high flow road, *Atmos. Environ.*, *35*, 453–468.

Schaap, M., K. Muller, and H. M. ten Brink (2002), Constructing the European aerosol nitrate concentration field from quality analysed data, *Atmos. Environ*, *36*, 1323–1335.

Schaap, M., M. van Loon, H. M. ten Brink, F. J. Dentener, and P. J. H. Builtjes (2004a), Secondary inorganic aerosol simulations for Europe with special attention to nitrate, *Atmos. Chem. Phys.*, *4*, 857–874.

Schaap, M., et al. (2004b), Artefacts in the sampling of nitrate studied in the "INTERCOMP" campaigns of EUROTRAC-AEROSOL, *Atmos. Environ.*, in press.

Schell, B., I. J. Ackermann, H. Hass, F. S. Binkowski, and A. Ebel (2001), Modeling the formation of secondary organic aerosol within a comprehensive air quality system, *J. Geophys. Res.*, *106*, 28,275–28,293.

Schmid, H., et al. (2001), Results of the "carbon conference" international aerosol carbon round robin test stage 1, *Atmos. Environ.*, *35*, 2111–2121.

Scholes, M., and M. O. Andreae (2000), Biogenic and pyrogenic emissions from Africa and their impact on the global atmosphere, *Ambio*, *29*, 23–29.

Schultz, M. G. (2002), On the use of ATSR fire count data to estimate the seasonal and interannual variability of vegetation fire emissions, *Atmos. Chem. Phys.*, *2*, 387–395.

Seinfeld, J. H., and S. N. Pandis (1998), Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, John Wiley, Hoboken, N. J.

Smith, D. J. T., R. M. Harrison, L. Luhana, C. A. Pio, L. M. Castro, M. N. Tariq, S. Hayat, and T. Quraishi (1996), Concentrations of particulate airborne polycyclic aromatic hydrocarbons and metals collected in Lahore, Paki-stan, *Atmos. Environ.*, *30*, 4031–4040.

Spicer, C. W., and P. M. Schumacher (1979), Particulate nitrate: Laboratory and field studies of major sampling interferences, *Atmos. Environ.*, *13*, 543–552.

Streets, D. G., S. Gupta, S. T. Waldhoff, M. Q. Wang, T. C. Bond, and B. Yiyun (2001), Black carbon emissions in China, *Atmos. Environ.*, *35*, 4281–4296.

ten Brink, H. M., et al. (2001), Aerosol cycle and influence on the radiation balance: Cluster-project in the National Research Program on global air pollution and climate change, *Rep. 410200064*, Energy Res. Cent. of the Netherlands, Petten.

ten Brink, H. M., et al. (2004), Comparability of methods in use in Europe for measuring the carbon content of aerosol, *Atmos. Environ.*, in press.

Vignati, E., G. de Leeuw, M. Schulz, and E. Plate (1999), Characterization of aerosols at a coastal site near Vindeby (Denmark), *J. Geophys. Res.*, *104*, 3277–3287.

Visschedijk, A. J. H. (2002), Onzekerheid en deeltjesgrootte-specificatie van de CEPMEIP emissie-bestanden (in Dutch), *TNO-MEP Rapp. R 2002/162*, Neth. Org. for Appl. Sci Res., The Hague.

Watson, J. G., E. Fujita, J. C. Chow, B. Zielinska, L. W. Richards, W. Neff, and D. Dietrich (1998), Northern Front Range Air Quality Study (NFRAQS) Final Report, *DRI Doc. 6580–685–8750.2F2*, Desert Res. Inst., Reno, Nev.

Wesely, M. L., D. R. Cook, R. L. Hart, and R. E. Speer (1985), Measurements and parameterization of particulate sulphate dry deposition over grass, *J. Geophys. Res.*, *90*, 2131–2143.

Wilson, J., C. Cuvelier, and F. Raes (2001), A modeling study of global mixed aerosol fields, *J. Geophys. Res.*, *106*, 34,081–34,108.

World Health Organization (WHO) (2000), Air Quality Guidelines, 2nd ed., *WHO Reg. Publ.*, *Eur. Ser.*, vol. 91, Copenhagen.

Zappoli, S., et al. (1999), Inorganic, organic and macromolecular components of fine aerosol in different areas of Europe in relation to their water solubility, *Atmos. Environ.*, *33*, 2733–2743.

Zier, M. (1991), Mass concentrations of total dust and soot in the near surface air in east Germany, *J. Aerosol Sci.*, *22*, S597–S600.

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