

A European aerosol phenomenology – 3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe

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ABSTRACT

This paper synthesizes data on aerosol (particulate matter, PM) physical and chemical characteristics, which were obtained over the past decade in aerosol research and monitoring activities at more than 60 natural background, rural, near-city, urban, and kerbside sites across Europe. The data include simultaneously measured PM₁₀ and/or PM_{2.5} mass on the one hand, and aerosol particle number concentrations or PM chemistry on the other hand. The aerosol data presented in our previous works (Van Dingenen et al., 2004; Putaud et al., 2004) were updated and merged to those collected in the framework of the EU supported European Cooperation in the field of Scientific and Technical action COST633 (*Particulate matter: Properties related to health effects*). A number of conclusions from our previous studies were confirmed. There is no single ratio between PM_{2.5} and PM₁₀ mass concentrations valid for all sites, although fairly constant ratios ranging from 0.5 to 0.9 are observed at most individual sites. There is no general correlation between PM mass and particle number concentrations, although particle number concentrations increase with PM_{2.5} levels at most sites. The main constituents of both PM₁₀ and PM_{2.5} are

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generally organic matter, sulfate and nitrate. Mineral dust can also be a major constituent of PM₁₀ at kerbside sites and in Southern Europe. There is a clear decreasing gradient in SO₄²⁻ and NO₃ contribution to PM₁₀ when moving from rural to urban to kerbside sites. In contrast, the total carbon/PM₁₀ ratio increases from rural to kerbside sites. Some new conclusions were also drawn from this work: the ratio between ultrafine particle and total particle number concentration decreases with PM_{2.5} concentration at all sites but one, and significant gradients in PM chemistry are observed when moving from North-western, to Southern to Central Europe. Compiling an even larger number of data sets would have further increased the significance of our conclusions, but collecting all the aerosol data sets obtained also through research projects remains a tedious task.

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1. Introduction

Because aerosols affect human health, ecosystems (acidification, eutrophication) and visibility, emission abatement measures have been implemented over the last decades to reduce particle matter (PM₁₀ and PM_{2.5}) concentrations. As the legislation on particulate emission and air pollution is based on PM mass all over the world and is becoming more and more stringent (WHO, 2005), PM₁₀ and PM_{2.5} concentrations are expected to further significantly decrease over the next decades. But can we assert that future PM concentration reductions will show positive effects on human health and ecosystems? Can we forecast the impact PM abatement measures on climate global and regional radiative forcing? Aerosol impacts are indeed linked to different characteristics that do not necessarily co-vary. Climate effects are related to its optical properties (scattering, absorption) throughout the atmospheric column, and cloud condensation formation potential, which in turn depends on particle size distribution and chemical composition. Impact on ecosystems depends on the deposition flux of specific species (acids, oxidized and reduced nitrogen, base cations, etc...). And we do not know yet which PM properties cause the health effects, although several epidemiological studies highlighted a link between short or long term exposure to PM mass concentration and human health (Zanobetti and Schwartz, 2005; Pope et al., 2008; Boldo et al., 2006; Pope, 2007; Miller et al., 2007). Therefore, abatement policies might be unnecessarily costly, inefficient, and even counter-productive if they address PM mass concentrations only. A thorough knowledge of the aerosol would help in designing a better legislation, using potential synergies and anticipating possible trade-offs between measures aiming at limiting air pollution and climate change. But PM₁₀ and more recently also PM_{2.5} mass concentrations are still the only aerosol metrics measured systematically in national and international air pollution monitoring networks.

Our goal is to show how different (or similar) other aerosol characteristics can be among various sites located in different regions across Europe, even where equal PM mass concentrations are observed. These characteristics are generally not available from regulatory monitoring networks (see e.g. EMEP and AirBase data banks), but rather from research projects. We have already pursued this process in two previous works (Van Dingenen et al., 2004; Putaud et al., 2004). The present study revises our previous conclusions in the light of data collected from more than 30 new sites. In view of the current European directive on particulate air pollution (2008/50/EC), we stratified the data according to PM mass concentration bins to address the following questions: (1) can PM_{2.5} and PM_{coarse} mass concentrations be inferred from PM₁₀ and PM_{2.5} mass concentrations, respectively, (2) which PM constituents are responsible for PM₁₀ high mass concentrations, and (3) can sub- μ m and ultrafine particle number concentrations be estimated based on PM_{2.5} mass concentration measurements. Answers are modulated according to the type of sites, from natural background to kerbside sites, and their location in Northwestern, Southern or Central Europe.

2. Compilation of European aerosol data

To achieve our goal, we selected data sets including PM mass concentrations on the one hand, and particle number and/or aerosol chemical composition data on the other hand, which were representative for a site during at least a season (i.e. minimum 6 weeks of continuous measurements). The selected sites and analytical methods employed are listed in Annexes 1 and 2.

Our previous articles (Van Dingenen et al., 2004; Putaud et al., 2004) were fruits of a voluntary collaboration among scientists for synthesizing the results they obtained from advanced monitoring or atmospheric research in single documents. These works compiled aerosol physical and/or chemical characteristics from 34 sites in 11 countries. Few data sets had been collected for Eastern Europe and the semi-arid Mediterranean area though. An attempt to fill these gaps was conducted in the framework of the COST (European Cooperation in the field of Scientific and Technical Research) Action 633 "Particulate matter: Properties related to health effects" (<http://cost633.dmu.dk>). Further observations of aerosol physical characteristics and aerosol chemical characteristics were collected from 28 and 29 additional sites, respectively, mostly located in Southern and Central Europe. New data sets were also provided by some of the co-authors of the first two "European aerosol phenomenologies". By merging these data, we can present and discuss here the physical and chemical characteristics observed over a network of more than 60 sites across Europe (Fig. 1). As previously, we have categorized the sampling sites using criteria proposed by the European Environment Agency (Larssen et al., 1999):

- Natural Background	- distance from large pollution sources > 50 km
- Rural Background	- distance from large pollution sources 10–50 km
- Near-City Background	- distance from large pollution sources 3–10 km
- Urban Background	- less than 2500 vehicles/day within a radius of 50 m
To which we have added the following	
- Industrial	- located within industrial areas
- Kerbside	- located by traffic lanes

For highlighting regional similarities and differences in aerosol characteristics, these sites have further been split in 3 large regions: Northwestern (NW), Southern and Central Europe (Fig. 1). Our scope is to describe intensive properties of the aerosol, i.e. not directly dependent on pollution dispersion. We therefore compile data sets in which particle number concentrations or size segregated (PM₁₀, PM_{2.5}, PM_{coarse} = PM₁₀ - PM_{2.5}) aerosol chemical composition can be related to PM₁₀ and/or PM_{2.5} mass concentrations.

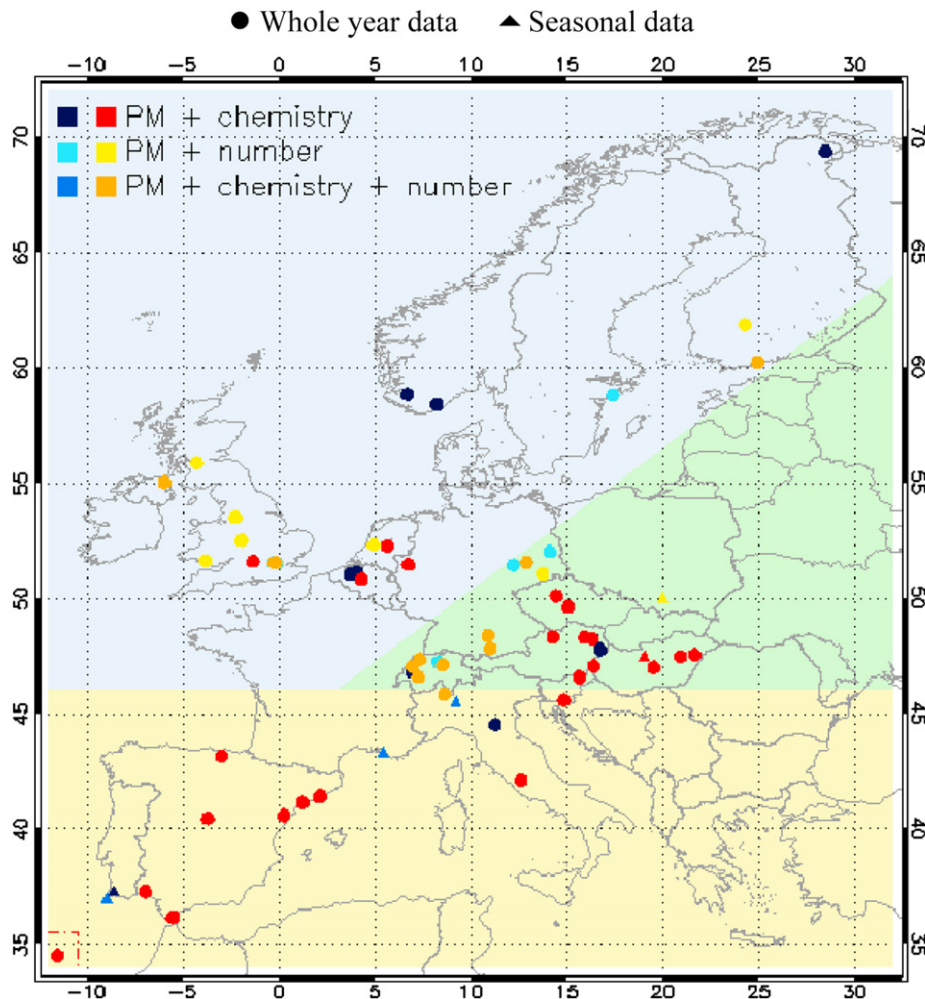


Fig. 1. Location of the sampling sites. The pastel background colors delimit the 3 geographical sectors Northwestern, Southern, and Central Europe.

3. PM₁₀ and PM_{2.5} mass concentrations

While PM₁₀ mass concentrations have been monitored by national and international air pollution monitoring networks for two decades (see e.g. EMEP and AirBase data banks), PM_{2.5} measurements are still being started up at many sites, because the new European Directive 2008/50/EC establishes target values for PM_{2.5} concentration and exposure to be met by 2010. Fig. 2a and b show 24-h averaged PM₁₀ and PM_{2.5} yearly statistics observed at 48 sites across Europe (see also Annexes 3 and 4). The methods used to measure PM mass concentrations (Annex 2) and the uncertainties arising from comparing data produced by different institutes using different techniques were described by Van Dingenen et al. (2004), and are further discussed in Annex 5. When another technique than the European reference method was used to obtain PM mass concentrations, correction were applied by the data providers to make them equivalent to the reference method when necessary. PM mass data are therefore comparable within $\pm 15\%$ (Annex 5). However, the data plotted in Fig. 2 were acquired over a decade (1996–2007), over which PM mass concentrations also decreased by up to probably 30% at some of the sites (Van Dingenen et al., 2004), which is nevertheless not sufficient to affect the spatial gradients in PM mass we describe here.

A large range of PM₁₀ concentrations (5–54 $\mu\text{g m}^{-3}$ annual average) is observed across the network (Fig. 2a). In each of the NW, Southern and Central sectors, an increasing gradient in PM₁₀

concentration is generally observed when moving from natural–rural background to kerbside sites. Exceptions are however observed in all three sectors (e.g. lower or similar PM₁₀ at urban or kerbside sites compared to rural sites). As there are no reasons why impact of local to urban sources should be significantly smaller at these specific urban or kerbside sites, we believe that these exceptions are due to differences in regional PM₁₀ background at a smaller scale. Annual and 24-h limit values are exceeded in all three sectors of Europe, generally (but not solely) at urban and kerbside sites. Urban background PM₁₀ annual mean and median values are significantly larger in southern Europe (median = 36 $\mu\text{g m}^{-3}$) compared to NW and Central Europe (medians = 24 and 26 $\mu\text{g m}^{-3}$, respectively).

The range in PM_{2.5} concentrations observed across the network (3–35 $\mu\text{g m}^{-3}$ annual average) is similar to that of PM₁₀ (Fig. 2b). An increasing gradient in PM_{2.5} is generally observed when moving from natural to urban background sites in Northwestern and Southern Europe. In Central Europe, PM_{2.5} can be as large at rural sites as at urban background sites, and concentrations at kerbside sites do not appear to be particularly high compared to urban background. The 2010 EU target value for annual PM_{2.5} mean concentration (25 $\mu\text{g m}^{-3}$) is exceeded at more urban background sites and less kerbside sites than the PM₁₀ annual limit value (40 $\mu\text{g m}^{-3}$) is. Therefore, the PM_{2.5} directive is more stringent for urban background sites, while PM₁₀ directives were more stringent for kerbside sites. The largest PM_{2.5} concentrations at near-city, urban background, and kerbside sites are observed in Southern Europe.

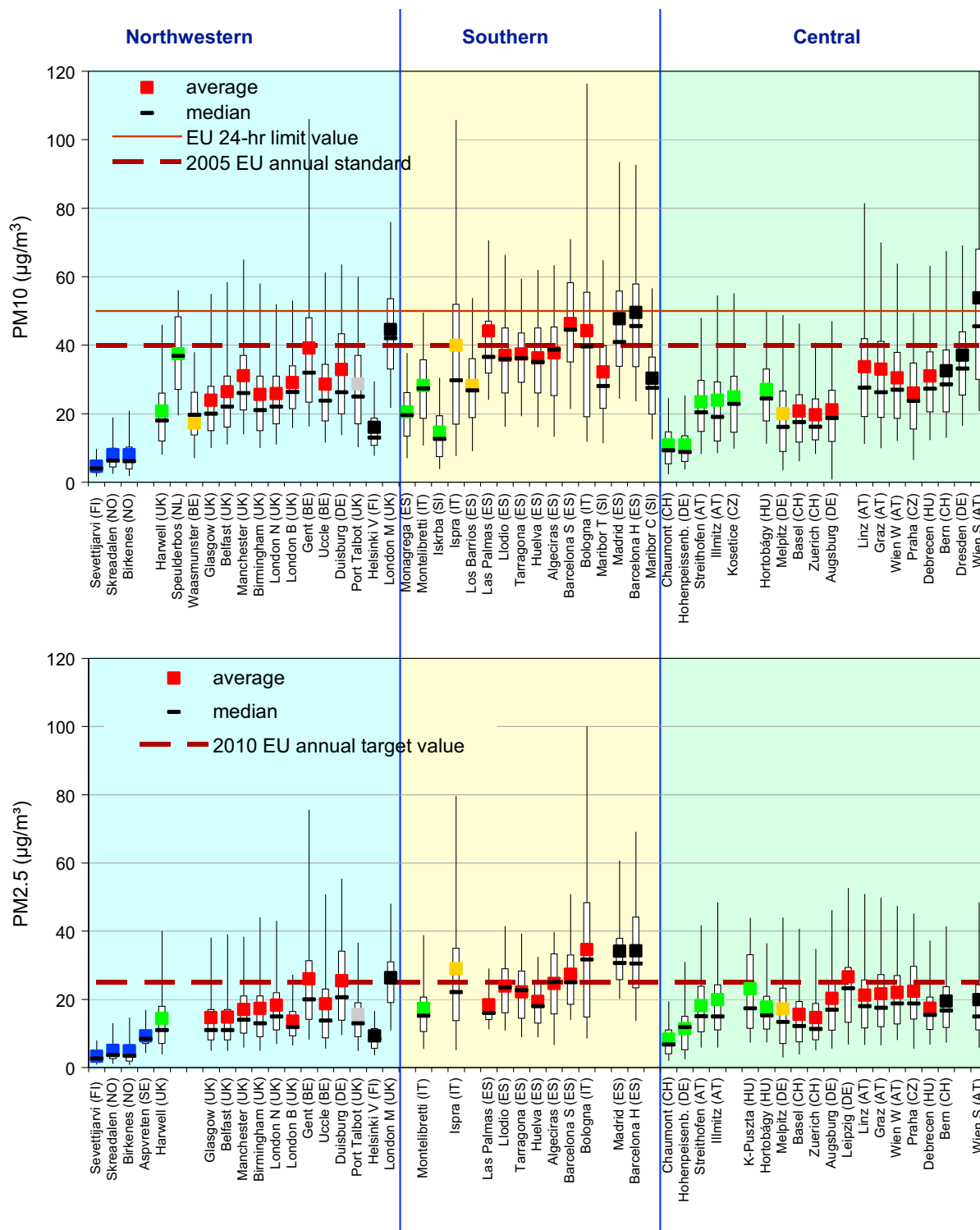


Fig. 2. Annual averages of PM₁₀ and PM_{2.5} mass concentrations, including the 5, 25, 50 (median), 75, and 95% percentiles of their 24-h integrated concentrations. Symbol colors indicate the type of site (blue: natural background, green: rural background, yellow: near-city, red: urban background, grey: industrial, black: kerbside).

Considering that the European directive 2008/50/EC sets limits for PM_{2.5} concentration and exposure, one might wish to estimate what levels of PM_{2.5} can be anticipated at sites where PM₁₀ measurements have been carried out. For each site, days were categorized according to the PM₁₀ daily mean mass concentrations in 4 bins: $0 \leq PM_{10} < 20$, $20 \leq PM_{10} < 50$, $50 \leq PM_{10} < 70$, and $70 \leq PM_{10} \leq 100 \mu\text{g m}^{-3}$. Fig. 3a shows averages of simultaneous

measured PM_{2.5} and PM₁₀ over these bins for 34 sites of the network. Although a significant correlation is observed ($R^2 = 0.89$, $n = 118$), PM_{2.5} can obviously not be predicted from PM₁₀ concentrations, since e.g. for $50 \leq PM_{10} < 70 \mu\text{g m}^{-3}$, PM_{2.5} mean concentrations range from 20 to 49 $\mu\text{g m}^{-3}$ across the network. However, PM₁₀ and PM_{2.5} concentrations are well correlated ($R^2 \geq 0.99$) at 26 sites. Site-specific PM_{2.5}/PM₁₀ ratios (slopes) range from 0.44 to 0.90, without

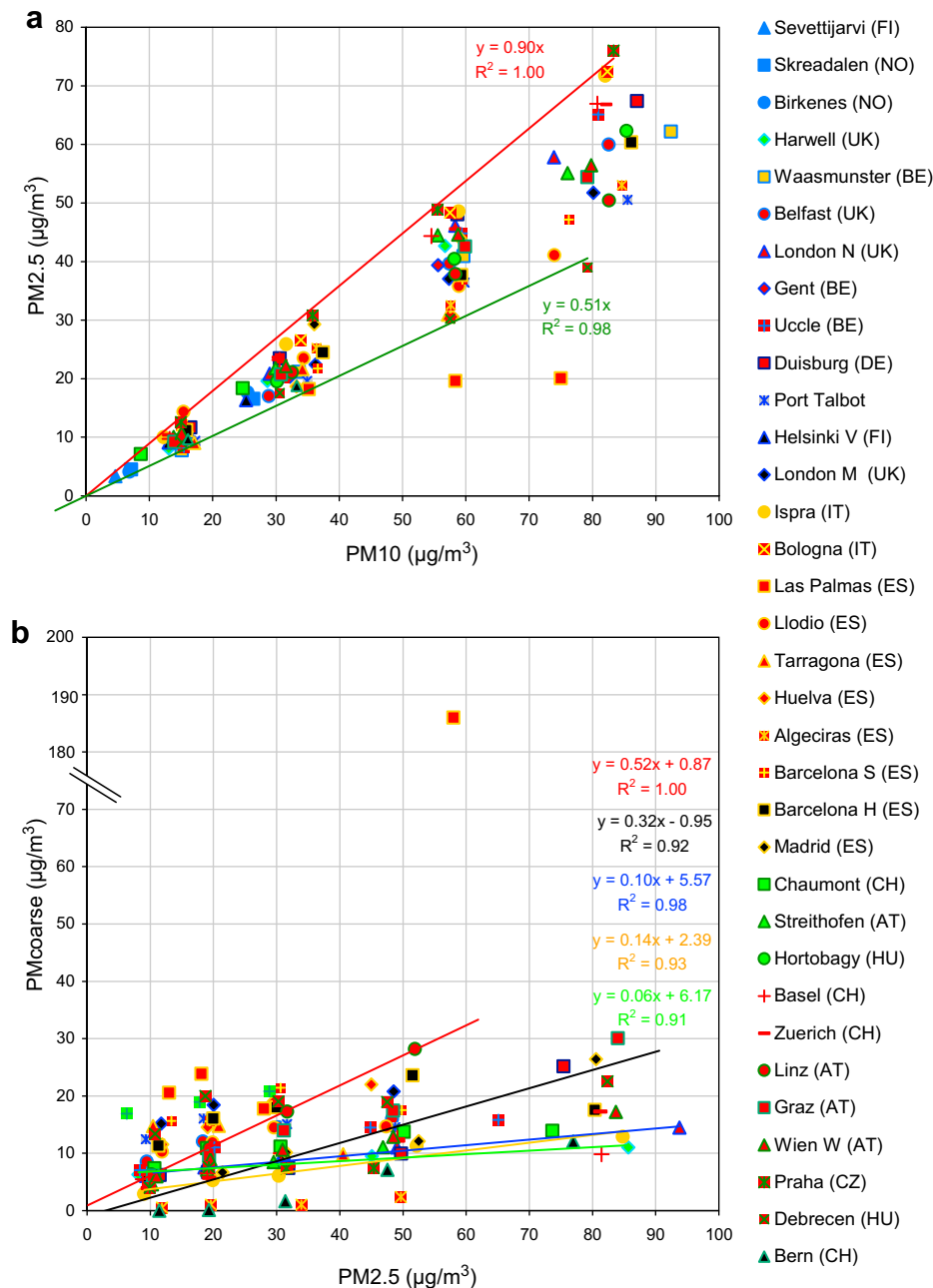


Fig. 3. Averaged (a) PM_{2.5} vs. PM₁₀ and (b) PM_{coarse} vs. PM_{2.5} mass concentrations. Mass concentrations were averaged over days on which daily mean (a) PM₁₀ were in the bins [0–20], [20–50], [50–70], [70–100] μg m⁻³ and (b) PM_{2.5} mass concentrations were in the bins [0–15], [15–25], [25–40], [40–70], and [70–100] μg m⁻³, respectively. Regression lines highlight (a) the lowest and highest site-specific regression slopes, and (b) examples of site-specific regressions between PM_{coarse} and PM_{2.5}.

any clear relationship between the value of the PM_{2.5}/PM₁₀ ratio and the type of site or its location in Europe. Correlations between PM_{2.5} and PM₁₀ concentrations indicate that PM₁₀ and PM_{2.5} main source strengths co-vary, and/or that both concentrations are driven by a common phenomenon: pollution dispersion, depending on meteorology. A closer examination reveals that PM_{2.5}/PM₁₀ ratios slightly increase with PM₁₀ levels at 12 sites, ranging from natural to urban background sites, which indicates that pollution periods are predominantly due to increases in PM_{2.5} mass concentration at these locations. Among noticeable exceptions, the kerbside site in Bern (CH), the urban background site in Debrecen (HU), and 2 urban background sites (Las Palmas and Llodio) in Spain (ES), where PM_{2.5}/PM₁₀ ratios slightly to sharply decrease with increasing PM₁₀ levels.

Significant contributions to high PM₁₀ concentrations of coarse mineral dust suspended from the ground or transported from African deserts could explain this observation (see Section 5).

Focusing PM monitoring on PM_{2.5} has sometimes been discussed in Europe. PM₁₀ Fig. 3b shows that there is obviously no relationship between PM_{2.5} and PM_{coarse} which is valid across the whole network. This means that PM_{coarse} concentrations can generally not be inferred from PM_{2.5} measurements, and that PM_{2.5} and PM_{coarse} can be controlled by different sources and/or processes. However, PM_{2.5} and PM_{coarse} are well correlated at several locations ranging from rural to kerbside sites, and from Northwestern to Southern to Central Europe (Fig. 3b). At those sites, it would be possible to estimate average PM_{coarse} concentrations from PM_{2.5} mass measurements,

but of course not to detect sporadic and seldom occurrence of large PM_{coarse} concentrations.

4. Particle number concentrations

We collected aerosol particle data from 12 more sites compared to our previous work (Van Dingenen et al., 2004), mostly located in the Northwestern sector of Europe. Total ($D_p > 10$ nm) and ultrafine ($10 < D_p < 100$ nm) particle concentrations rather than detailed size distributions were generally available from these new sites. The size distributions measured with Scanning or Differential Mobility Sizing systems (S/DMPSS) were integrated from 10 nm upwards to obtain comparable data. The intercomparability of particle number concentrations obtained from S/DMPSS and Condensation Particle Counters (CPCs) is estimated to $\pm 15\%$, as further discussed in Annex 5. Fig. 4a and b shows total and ultrafine particle number concentrations averaged over days on which daily mean $PM_{2.5}$ mass

concentrations were $PM_{2.5} < 10 \mu g m^{-3}$, $10 \leq PM_{2.5} < 20 \mu g m^{-3}$, etc.... All data sets cover 1 year or more except those from Helsinki V (no summer data), Milano B (winter and summer data only), and Sagres, Mt. Foia, and Marseille V (summer data only).

Total particle number concentrations as low as $2000 cm^{-3}$ can be observed in all 3 sectors of Europe, and not only at sites impacted by clean maritime air. In contrast, total particle concentrations can exceed $40\,000 cm^{-3}$ at urban and kerbside sites e.g. in Northwestern and Central Europe. No big difference in particle number appears at rural sites among the three large Northwestern, Central, and Southern sectors of Europe. Clear increasing gradients in particle number concentrations generally occur when moving from natural background or rural sites to urban background or kerbside sites in all 3 sectors, excluding Helsinki V (FI), a site located 50 m away from a traffic lane with 14 000 vehicles a day, where the total particle concentration is very low compared to other kerbside sites. An increase in total particle concentration with increasing $PM_{2.5}$ levels is clear at numerous

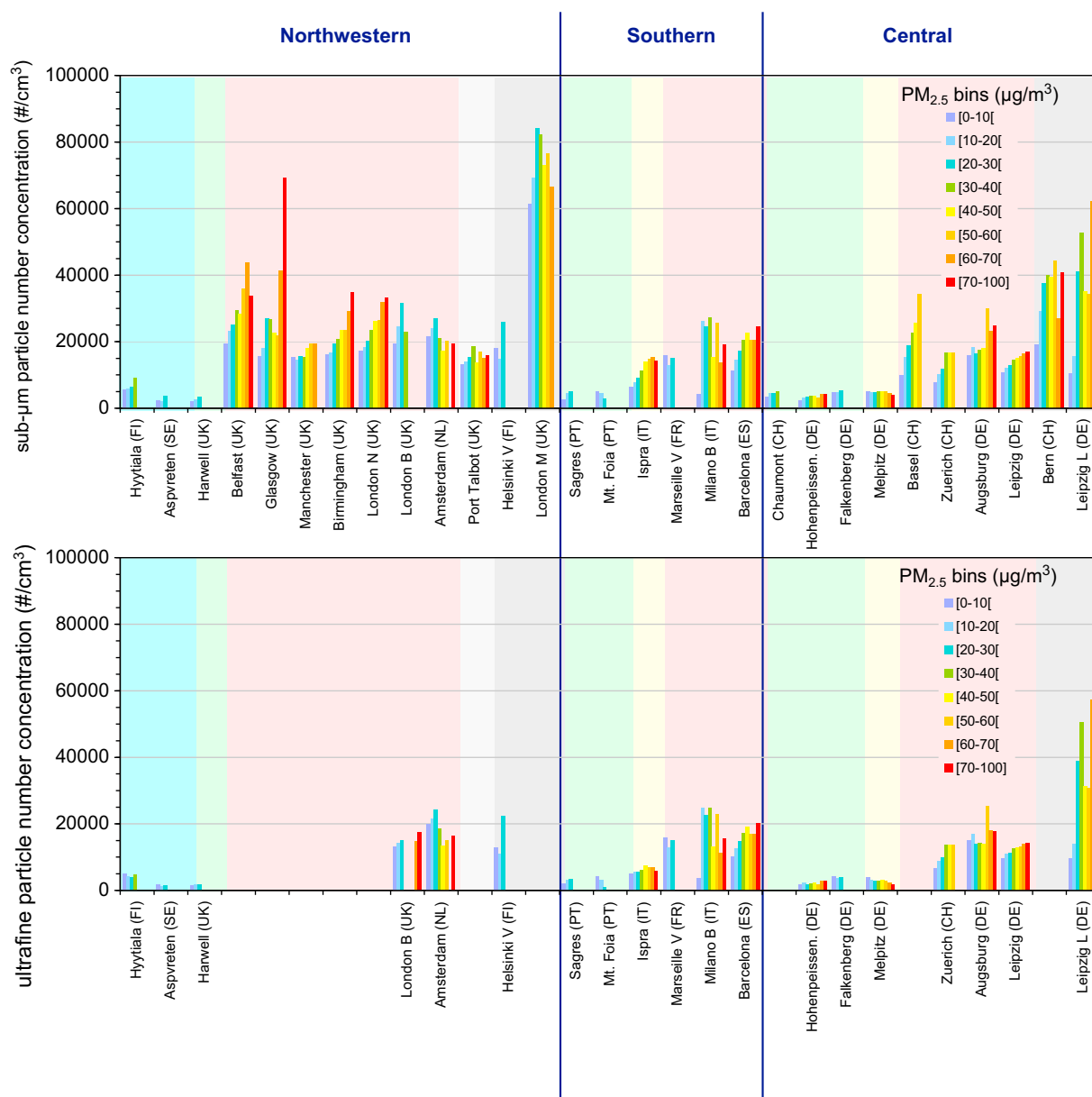


Fig. 4. Particle number concentration averaged over $10 \mu g m^{-3}$ wide $PM_{2.5}$ bins at the different sites of the network, split according to the sectors shown in Fig. 1. Pastel background colors indicate the site types (blue: natural background, green: rural background, yellow: near-city, rose: urban background, light grey: industrial, grey: kerbside).

rural, urban or kerbside sites, but nowhere does the particle number increase proportionally to $PM_{2.5}$: the ratio particle number/PM mass rather decreases with increasing $PM_{2.5}$. This results in a lack of correlation ($R^2 = 0.16$, $n = 132$) between particle number and $PM_{2.5}$ mass concentrations, already observed at the US-EPA PM supersites (Solomon et al., 2008). This is due to the facts that most particles are generally smaller than 100 nm (ultrafine/total particle median ratio equals 0.76, and ranges 0.4–0.9), and that the ultrafine particle number concentration does generally not increase with $PM_{2.5}$ levels (Fig. 4b). In contrast, increasing $PM_{2.5}$ mass are related to increasing number of particles larger than $D_p = 100$ nm. As a consequence, the ratio between ultrafine particle and total particle number concentrations decreases at all sites but one (Helsinki V, FI) when $PM_{2.5}$ mass concentration increases (Table 1). The explanation of these observations is that different mechanisms are responsible for the production of ultrafine particles, which generally control the total particle number, and for the production of larger particles, which control PM mass. The production rates of these mechanisms do not necessarily co-vary with space and time (see e.g. Harrison and Jones, 2005).

5. Particulate matter chemical composition

Fig. 5a–c shows annual average chemical composition of PM_{10} , $PM_{2.5}$ (the fine fraction) and PM_{coarse} ($= PM_{10} - PM_{2.5}$), respectively, at 39 sites, among which 23 are new compared to Putaud et al. (2004). Only sites for which at least one whole year of data was available are included in Fig. 5. A table with atmospheric concentrations of the main PM constituents is available in Annex 4.

Elemental carbon (EC), nitrate (NO_3^-), and ammonium (NH_4^+) concentrations derive directly from measurements. Organic matter (OM) and carbonaceous matter (CM) concentrations were calculated from organic carbon (OC) and total carbon data to account for the non-C atoms contained in particulate OM. Although sea-spray was clearly identified as a component of the aerosol at sites located <50 km from seashore only, sea salt concentrations were calculated (from a sea salt tracer, Cl^- concentrations, and a standard seawater composition) at all sites for consistency. Likewise, non-sea-salt sulfate ($nssSO_4^{2-}$) concentrations were calculated from measured SO_4^{2-} , a sea salt tracer, and a standard seawater composition. The total amount of mineral dust (min. dust), including silicates, carbonates, calcium sulfate and trace elements, was estimated based on measurements of selected elements. Further details are provided in Annex 5. The unaccounted mass (obtained as the difference between the gravimetrically measured aerosol mass concentration and the sum of the aerosol component concentrations) includes different species for the various sites of the network (Annex 6). Questions regarding the accuracy and comparability of PM chemistry data obtained by different laboratories over a decade were

discussed by Putaud et al. (2004), and are quantitatively assessed in Annex 5. Differences among laboratories in the determination of the main ionic species are not expected to exceed $\pm 15\%$. In contrast, EC concentrations obtained with various techniques can differ by a factor of 2, and OC by $\pm 30\%$ (Schmid et al., 2001). This is why we decided to rather focus on TC values, which are measured by a large range of European laboratories with discrepancies smaller than $\pm 25\%$. The uncertainty of mineral dust amounts, derived from measurements of tracers, can reach 150%. Sampling artefacts also affect the amount of semi-volatile species collected on filters or impactor substrates, but we discuss here the contribution of various constituents to PM mass concentrations as measured with reference or equivalent methods designed to judge compliance with European air pollution directives, which do not always accurately represent what is actually in the air.

PM constituents' relative contributions to PM mass are independent of dilution and reflect differences in the sources and processes controlling the aerosol composition. Depending on the relative contribution of the fine and coarse fractions, the chemical composition of PM_{10} and $PM_{2.5}$ (Fig. 5a,b) can be similar (e.g. Duisburg, DE) or significantly different (e.g. Las Palmas, ES). When all main aerosol components except water are quantified (i.e. at 21–23 sites), they account for 73–94% of the PM_{10} mass (average 81%) and for 67–97% of $PM_{2.5}$ mass (average 84%). The remaining unaccounted mass fraction (*unacc.*, Annex 6) can result from analytical errors, a possibly systematic underestimation of the PM constituents whose concentrations are calculated from measured data (e.g. OM, CM, and mineral dust), and aerosol-bound water (especially if mass concentrations are determined at $RH > 30\%$). All components are present in both the fine and the coarse fractions, but mineral dust and sea salt contribute more to the coarse fraction, whereas NH_4^+ , $nss-SO_4^{2-}$, EC, CM, and to a lesser extent OM, contribute more to the fine fraction. On average, NO_3^- partitioning over the fine and coarse fractions is 70:30, but it is quite variable (from 45:55 at natural background sites in Northwestern Europe and urban sites in Spain to 80:20 at urban sites in Central Europe), depending on the ability of the coarse particles to trap gaseous HNO_3 . Further discussion about coarse/fine partitioning can be found in Putaud et al. (2004).

It appears from Fig. 5 that OM (estimated as $1.4 * OC$) is the major single component of PM_{10} at almost every site where its contribution was determined. However, PM_{10} , $PM_{2.5}$ and PM_{coarse} chemical compositions show differences across Europe: on average there is more carbonaceous matter in PM_{10} in Central Europe, more nitrate in Northwestern Europe, and more mineral dust in all fractions in southern Europe. This is why we have calculated an average chemical composition for each type of site in each of the 3 geographical sectors (Table 2). According to the law of propagation of errors, the random uncertainties of the figures listed in Table 2

Table 1
Ultrafine/sub- μm particle number mean ratios for various $PM_{2.5}$ concentration bins ($\mu\text{g m}^{-3}$).

		$PM_{2.5}$	[0–10]	[10–20]	[20–30]	[30–40]	[40–50]	[50–60]	[60–70]	[70–+]
Northwestern	Hyytiälä (FI)	Rural	0.88	0.72	0.60	0.51				
	Aspvreten (SE)	Rural	0.76	0.61	0.41					
	Harwell (UK)	Rural	0.76	0.65	0.51					
	Amsterdam (NL)	Urban	0.92	0.89	0.89	0.88	0.78	0.75		
	Helsinki V (FI)	Kerbside	0.72	0.75	0.86					
	London M (UK)	Kerbside	0.90	0.87	0.85	0.82				
Southern	Ispira (IT)	Near-city	0.80	0.72	0.62	0.55	0.53	0.47	0.45	0.42
	Barcelona (ES)	Urban	0.90	0.87	0.86	0.84	0.84	0.83	0.82	0.81
Central	Hohenpeissen (DE)	Rural	0.81	0.71	0.51	0.57	0.64	0.51	0.70	0.68
	Melpitz (DE)	Near-city	0.76	0.67	0.60	0.59	0.60	0.59	0.50	0.47
	Zuerich (CH)	Urban	0.88	0.86	0.84	0.83	0.81	0.82		
	Augsburg (DE)	Urban	0.94	0.92	0.85	0.81	0.78	0.85	0.77	0.71
	Leipzig (DE)	Kerbside	0.91	0.90	0.88	0.87	0.86	0.85	0.84	0.84

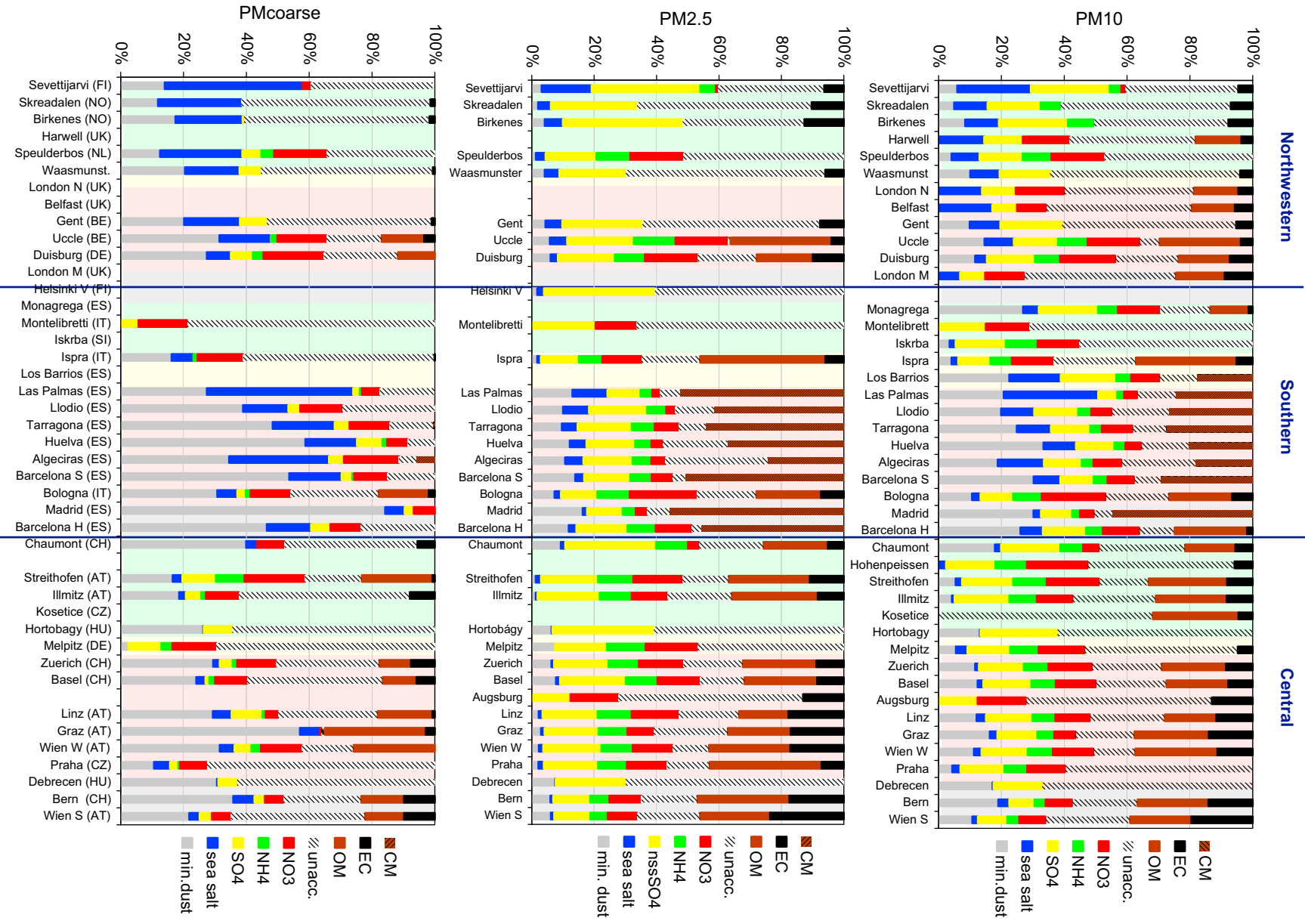


Fig. 5. PM₁₀, PM_{2.5} and PM_{coarse} annual mean chemical composition in the sites of the network, split according to the sectors shown in Fig. 1. Pastel background colors indicate the site types (green: rural background, yellow: near-city, rose: urban background, grey: kerbside). Note that not all the constituents were measured at each site (see Annex 6).

Table 2
Major constituent contributions to PM₁₀, PM_{2.5} and PM_{coarse}.

		PM ₁₀			PM _{2.5}			PM _{coarse}		
		Rural	Urban	Kerbside	Rural	Urban	Kerbside	Rural	Urban	Kerbside
N-western Europe	Min. dust	4%	12%			5%	1%		26%	
	Sea salt	12%	10%	7%		4%	1%		15%	
	SO ₄	13%	14%	8%		21%	18%		6%	
	NO ₃	16%	14%	12%		16%			20%	
	OM	15%	18%	16%		25%			14%	
	EC	4%	5%	9%		7%			1%	
	TC	14%	18%	20%		25%			12%	
Southern Europe	Min. dust	15%	21%	28%		11%	14%		42%	69%
	Sea salt	3%	12%	5%		6%	2%		22%	11%
	SO ₄	16%	12%	12%		15%	15%		4%	5%
	NO ₃	14%	9%	8%		7%	7%		11%	9%
	OM		26%			23%			13%	
	EC		6%			8%			2%	
	TC	13%	21%	28%		30%	38%		11%	
Central Europe	Min. dust	9%	12%	15%	3%	5%	6%	22%	25%	29%
	Sea salt	2%	2%	2%	1%	1%	1%	2%	3%	5%
	SO ₄	19%	15%	9%	17%	19%	12%	5%	4%	4%
	NO ₃	13%	12%	8%	6%	13%	10%	10%	7%	6%
	OM	23%	21%	21%	15%	22%	26%	5%	15%	13%
	EC	6%	10%	17%	5%	14%	21%	3%	3%	10%
	TC	32%	32%	38%	19%	31%	35%	6%	14%	19%

are up to 3 times less than the random uncertainty of the chemical composition at a single site. Only statistically significant differences in PM chemical composition are further discussed.

Whereas the contribution of mineral dust to PM₁₀ is significantly larger at all types of site in Southern Europe compared to the 2 other sectors, PM₁₀ chemical composition in central Europe is characterised by low sea salt and high EC and TC content with respect to Northwestern and Southern Europe. In contrast, SO₄²⁻ and NO₃⁻ contributions to PM₁₀ are more similar in all three sectors of Europe. For PM_{2.5}, the only possible comparison among sectors regards urban sites. Some conclusions made for PM₁₀ still apply (mineral dust contribution larger in Southern Europe, sea salt percentage smaller in Central Europe, and TC percentage smaller in Northwestern Europe), others do not: for instance, compared to, SO₄^{2-"/PM_{2.5} and NO₃^{-"/PM_{2.5} ratios in Southern Europe are not as large as in the other sectors but significantly smaller. Regarding PM_{coarse} chemical composition, the specificity of the geographical gradient is the larger contribution of both SO₄²⁻ and NO₃⁻ in Northwestern Europe compared to Southern and Central Europe.}}

Differences in PM chemistry among the 3 types of sites can also be observed within each sector of Europe. Both SO₄²⁻ and NO₃⁻ contributions to PM₁₀ generally decrease when moving from rural to kerbside sites. The same is observed for NO₃⁻ in PM_{coarse} and for SO₄²⁻ in both PM_{2.5} and PM_{coarse} in Central Europe. In contrast, the contribution of EC to PM₁₀ increases from rural to urban to kerbside sites in all sectors. The contribution of OM to PM₁₀, PM_{2.5} and even PM_{coarse} is remarkably similar at all types of sites where available. As a consequence, TC/PM₁₀ and TC/PM_{2.5} ratios increase when moving from rural to kerbside sites.

Fig. 6 shows mean ratios between mineral dust, sea salt, nss-SO₄²⁻, NO₃⁻, TC and (a) PM₁₀ or (b) PM_{2.5} on days when PM₁₀ daily average mass concentrations were in the intervals [0, 20], [20, 50], [50, 70], and [70, 100] µg m⁻³. This could reveal what components (and suggest which sources) are mainly responsible for particulate air pollution episodes, keeping in mind that meteorological conditions (controlling pollution dispersion) generally strongly influence PM mass concentrations. To avoid biases due to possible seasonal variations, only sites for which at least a whole year of data is available are included in Fig. 6 (except for Helsinki V, which does

not include summertime data). A few clear patterns can be observed. Exceptions are underlined when observed, but we did not try to explain the PM chemical composition observed at each single site. Everywhere in the network except at urban sites in Las Palmas (ES), Huelva (ES) and the kerbside site in Bern (CH), the contribution of sea salt to PM₁₀ decreases with increasing PM₁₀ levels. The percentage of SO₄²⁻ in PM₁₀ does not increase with PM₁₀ mass, except at background sites in Scandinavia (Skreådalen, NO; Birkenes, NO), and some sites located near to heavy industries like Llodio and Huelva (ES). The contribution of NO₃ to PM₁₀ increases with PM₁₀ at all of the urban background sites except Las Palmas (ES) and Praha (CZ), and at 3 rural, 2 near-city and 2 kerbside sites across Europe. This suggests that for reducing the number of PM₁₀ daily limit exceedances, reducing the emissions of NO_x and NH₃ (which reacts with NO_x oxidation products to form particulate NH₄NO₃) would be efficient. In contrast, the TC/PM₁₀ ratio does not increase significantly with PM₁₀ at any site of the network, but rather clearly decreases at a number of locations, including most urban background sites. It can therefore be stated that particulate air pollution events are not due to an enhanced contribution of carbonaceous matter, the overall major constituent of PM₁₀ across Europe (Fig. 5 and Table 2), which is also expected to comprise hazardous substances. At kerbside sites, no single PM₁₀ constituent (not even mineral dust) shows up as solely responsible for high PM₁₀ concentrations, indicating that meteorology is the major controlling factor upon pollution levels at kerbside sites too.

The same features can be observed when looking at the chemical composition of PM_{2.5} in Fig. 6b, since differences between PM_{2.5} and PM₁₀ compositions can be seen at a few sites only. In Huelva (ES), the increase in sea salt and NO₃⁻ contributions to PM₁₀ with increasing PM₁₀ levels is not observed in PM_{2.5} chemical composition, which shows that sea salt affects mainly the coarse PM fraction, but also that most NO₃⁻ resides in coarse particles (including sea-spray) during PM pollution episodes. In Las Palmas (ES), the contribution of mineral dust to PM_{2.5} strongly increases with PM₁₀ concentration (which was not observed in PM₁₀ composition): during high PM₁₀ episodes, PM_{2.5} (the concentration of which remains constant) is enriched in mineral dust and depleted in other constituents.

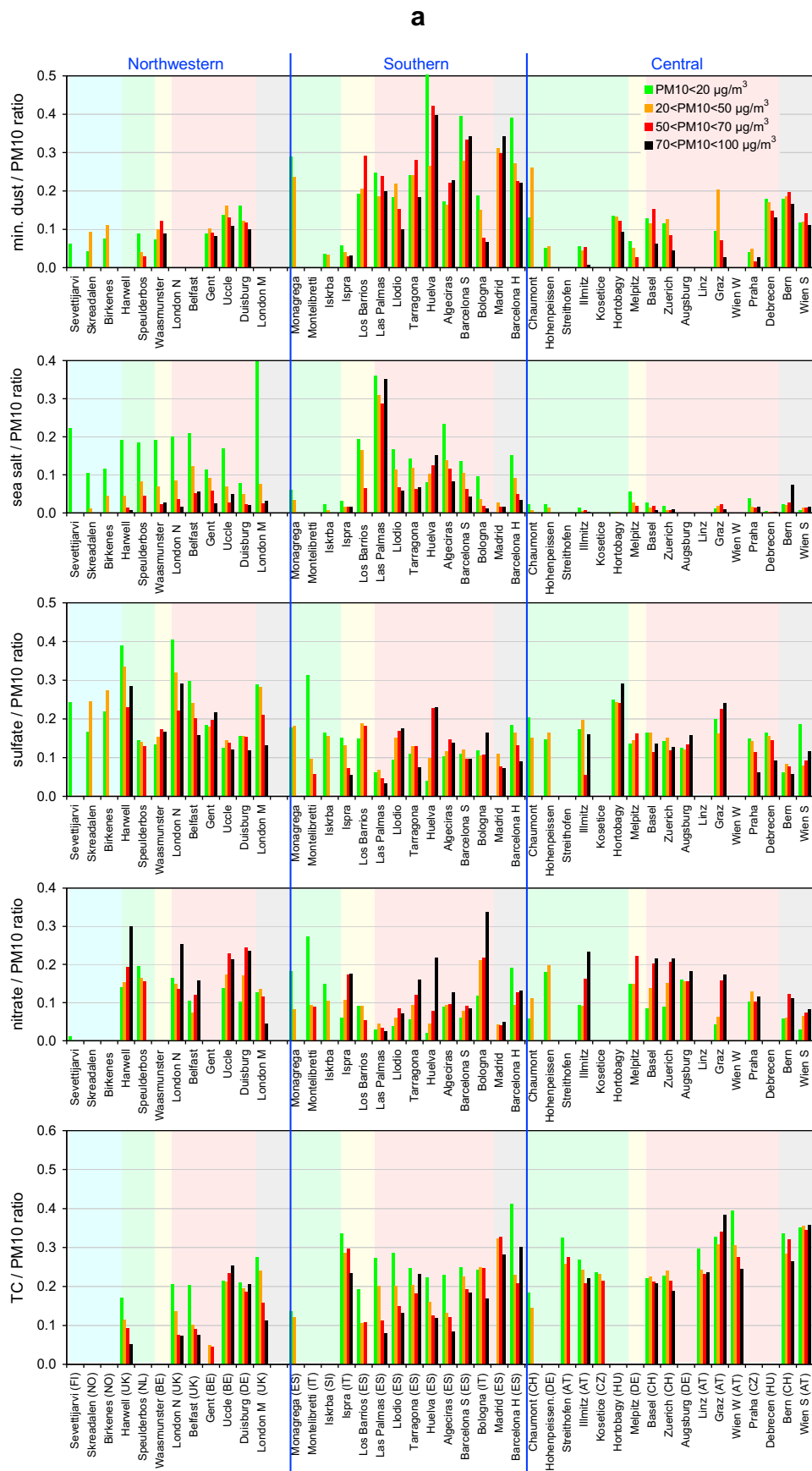


Fig. 6. a. Annual mean contributions to PM_{10} of major constituents calculated over four PM_{10} mass concentration bins ($[0-20]$, $[20-50]$, $[50-70]$, $[70-100]$ $\mu g m^{-3}$). Sites are categorized according to the sectors shown in Fig. 1. Pastel background colors indicate the site types as in Fig. 5. Note that not all the constituents were measured at each site (see Annex 6). b. Same as Fig. 6a for annual mean contributions to $PM_{2.5}$.

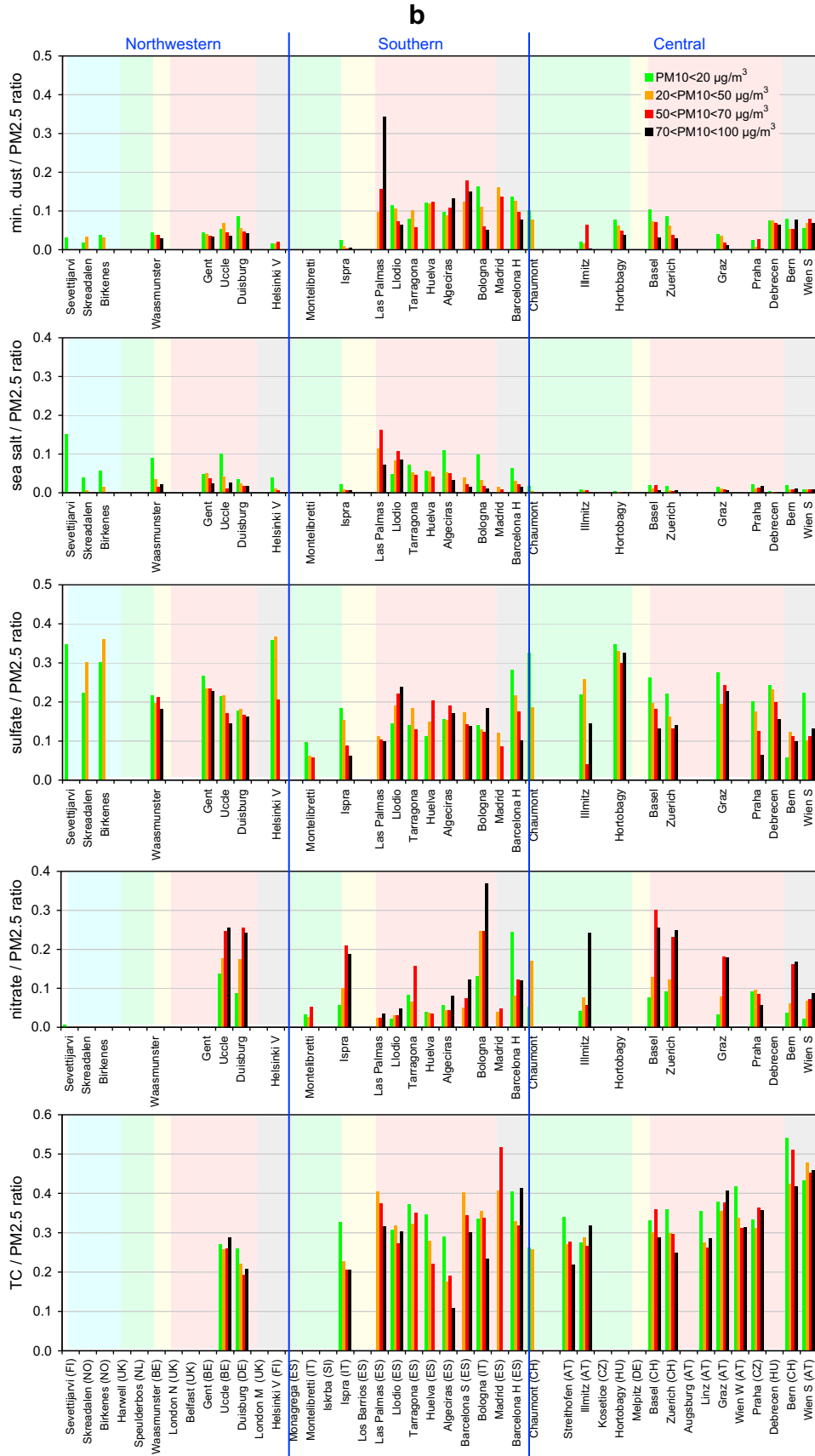


Fig. 6. (continued).

6. Conclusions

Datasets on aerosol physical and chemical characteristics from more than 60 European sites were compiled, among which more than 30 were not part of our previous studies (Van Dingenen et al., 2004; Putaud et al., 2004). This work covers also the Mediterranean area, as well as Central Europe. It appears that regional differences in PM characteristics (particle number abundance, chemical composition) are significant enough to justify a grouping of sites in three large geographical sectors (Northwestern, Central and Southern Europe), on top of the classification according to site types (rural, urban, kerbside) we used in our previous articles.

Considering 45 new data sets (15 on PM mass and particle number concentrations, and 30 on PM mass and chemistry), some of our former conclusions are confirmed and reinforced, among which:

1. There is a significant correlation ($R^2 = 0.89$, $n = 118$) between the PM_{10} and $PM_{2.5}$ average mass concentrations calculated at 34 sites over 4 PM_{10} concentration bins ($PM_{10} < 20$, $20 \leq PM_{10} < 50$, $50 \leq PM_{10} < 70$, $70 \leq PM_{10} \leq 100$). However, site-specific $PM_{2.5}/PM_{10}$ ratios differ very much from each other (range 0.44 and 0.90), and no general relationship can be established between the value of the $PM_{2.5}/PM_{10}$ ratio and the location (site type - rural, urban, kerbside, or region - Northwestern, Southern, Central Europe). There are only 3 sites among 34 where a poor correlation ($R^2 < 0.98$, $n = 4$) between PM_{10} and $PM_{2.5}$ is observed.
2. A poor correlation is observed ($R^2 = 0.16$, $n = 132$) between particle number and $PM_{2.5}$ mass concentrations averaged over $10 \mu\text{g m}^{-3}$ wide $PM_{2.5}$ concentrations bins. However, the particle number concentration increases with $PM_{2.5}$ at most sites, but less than proportionally, because the number of ultrafine particles does generally not increase with $PM_{2.5}$.
3. In each geographical sector in Europe, the particle number/ $PM_{2.5}$ mass ratio increases from rural to near-city, to urban, and to kerbside sites.
4. The main constituents of both PM_{10} and $PM_{2.5}$ all over Europe are generally organic matter (OM), SO_4^{2-} and NO_3^- . Mineral dust and sea salt are the main constituents of PM_{coarse} . Mineral dust can be a major constituent of PM_{10} as well at kerbside sites.
5. There is a clear decreasing gradient in SO_4^{2-} and NO_3^- contribution to PM_{10} when moving from rural to urban to kerbside sites. In contrast, the TC/ PM_{10} ratio increases from rural to kerbside sites.
6. There is no relationship between PM mass concentration and PM chemical composition valid for each type of site in all 3 sectors of Europe.

A few of the conclusions we drew from our previous studies should be revised when considering the larger set of data compiled in the present work:

1. The increase of the $PM_{2.5}/PM_{10}$ ratio with PM_{10} mass concentration is not wholly systematic. There are even a few sites where the opposite is observed, which highlights the predominant role of coarse particles in PM_{10} high concentration episodes at those sites.
2. The observation we made previously that the contribution of NO_3^- systematically increases with PM levels does not hold when considering the whole set of data we present here. However, NO_3^- contribution to PM_{10} and $PM_{2.5}$ does increase with PM mass concentrations at almost all urban background sites of the network, and at several other locations across Europe. NO_x emissions should therefore be particularly targeted to reduce the number of PM_{10} limit value daily exceedances.

Finally, a larger number of data sets allowed us to draw some new conclusions compared to our previous work:

1. The ratio between ultrafine particle and total particle number concentration decreases with increasing $PM_{2.5}$ concentration at all sites but one (Helsinki V, FI).
2. Comparing PM chemical composition in 3 large geographical sectors, significant differences appears: the contribution of mineral dust to all PM size fractions is larger in Southern Europe, that of sea salt to PM_{10} is larger in Northwestern Europe, and the ratio TC/ PM_{10} is generally larger in Central Europe. In contrast, the contributions of SO_4^{2-} and NO_3^- to PM_{10} do not vary much across these three sectors, compared to the gradients observed when moving from rural to kerbside sites within each sector. The variability in $PM_{2.5}$ chemistry may be a little larger, but the smaller number of sites for which such data are available limits the robustness of this observation.

Actually, there are still very few sites where all the major constituents of PM_{10} and $PM_{2.5}$ have been measured over time periods long enough to obtain representative averages. Even less is known about sub- μm particle and ultrafine particle number concentrations. Is this really due to a lack of measurements or to a narrow bottle neck in data flowing from “producers” to “consumers”? We know that we still missed several pertinent data sets in this new aerosol phenomenology. However, two different ways of collecting data were implemented for getting this work done (unofficial contacts among colleagues used to collaborate with each other, and official contacts through National Representatives in COST633), and both were only partially successful. It seems therefore that a cultural reluctance from European scientists to make data available for large scale assessments still persists. Another stumbling block remains the lack of control on data comparability. There are currently no reference methods for measuring the aerosol characteristics presented in this work, except for PM mass concentrations. Even for this metric, the reference methods (EN12341 and EN14907, for PM_{10} and $PM_{2.5}$, respectively) do not guarantee the comparability of measurements performed at various locations across Europe, since significant sampling and analytical artifacts remain, which are PM chemistry and meteorology dependent. Hopefully, the standardization of the measurements of particle number, ionic and carbonaceous species in the aerosol being developed in Europe will shortly solve some of these problems.

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Appendix. Supplementary material

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.atmosenv.2009.12.011.

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