



## A European aerosol phenomenology—1: physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe

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Received 16 June 2003; received in revised form 20 December 2003; accepted 19 January 2004

### Abstract

This paper synthesizes data on aerosol (particulate matter, PM) physical characteristics, which were obtained in European aerosol research activities at free-troposphere, natural, rural, near-city, urban, and kerbside sites over the past decade. It covers only two sites in the semi-arid Mediterranean area, and lacks data from Eastern Europe. The data include PM<sub>10</sub> and/or PM<sub>2.5</sub> mass concentrations, and aerosol particle size distributions. Such data sets are more comprehensive than those currently provided by air quality monitoring networks (e.g. EMEP, EUROAIRNET). Data available from 31 sites in Europe (called “The Network”) were reviewed. They were processed and plotted to allow comparisons in spite of differences in the sampling and analytical techniques used in various studies. A number of conclusions are drawn as follows:

Background annual average PM<sub>10</sub> and PM<sub>2.5</sub> mass concentrations for continental Europe are  $7.0 \pm 4.1$  and  $4.8 \pm 2.4 \mu\text{g m}^{-3}$ , respectively.

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The EU 2005 annual average PM<sub>10</sub> standard of  $40 \mu\text{g m}^{-3}$  is exceeded at a few sites in The Network. At all near city, urban and kerbside sites, the EU 2010 annual average PM<sub>10</sub> standard of  $20 \mu\text{g m}^{-3}$ , as well as the US-EPA annual average PM<sub>2.5</sub> standard of  $15 \mu\text{g m}^{-3}$  are exceeded. In certain regions, PM<sub>10</sub> and PM<sub>2.5</sub> in cities are strongly affected by the regional aerosol background.

There is no “universal” (i.e. valid for all sites) ratio between PM<sub>2.5</sub> and PM<sub>10</sub> mass concentrations, although fairly constant ratios do exist at individual sites. There is no universal correlation between PM mass concentration on the one hand, and total particle number concentration on the other hand, although a ‘baseline’ ratio between number and mass is found for sites not affected by local emissions. This paper is the first part of two companion papers of which the second part describes chemical characteristics.

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**Keywords:** Aerosol; Chemical composition; PM<sub>10</sub>; PM<sub>2.5</sub>; Compilation

## 1. Introduction

Today, the interest in aerosols is high mainly because of their effect on human health and their role in climate change. They have also a determining effect on visibility and contribute to the soiling of monuments. Compared to trace gases, aerosols are relatively complex to characterize because of their multi-component chemical composition, and because of the large range in particle size, ranging from nanometers to several micrometers. Furthermore, aerosol sampling is still a challenge due to the fact that a significant fraction of the mass is semi-volatile and can transfer between the gas and aerosol phase as a function of temperature, relative humidity, aerosol acidity, sampling and handling procedures, etc. (e.g. ammonium nitrate, semi-volatile organic compounds). Data on comprehensive physical–chemical aerosol characterization are needed for several reasons.

### 1.1. Aerosols and health

Epidemiological studies show that an increase in PM<sub>10</sub> mass concentration by  $10 \mu\text{g m}^{-3}$  results in an increase of 0.5–1.5% in premature total mortality in case of short term/episodic exposure, and in an increase up to 5% in premature total mortality in case of long-term/life-long exposure (Wilson and Spengler, 1996). As yet, there is no indication which physical or chemical PM characteristic is responsible for these effects. However, recent research seems to indicate that PM<sub>10</sub> is associated with respiratory responses and PM<sub>2.5</sub> with cardiovascular diseases (Wyzga, 2002). Legislation in the EU and US is therefore expressed in terms of target values for PM<sub>10</sub> and PM<sub>2.5</sub> (= the mass of particles with a diameter below 10 or  $2.5 \mu\text{m}$ , respectively). Because of the undifferentiated nature of the metric “PM mass” and the many sources contributing to it, implementing such legislation might be unnecessarily costly. Ideally, health effects of aerosol particles should be related to a well-defined set of physical or chemical aerosol char-

acteristics, which can be related to a well-defined set of sources. Major research programmes are devoted to understand the effects of aerosols on health and to the apportionment of their sources.

### 1.2. Aerosols and climate

Observations and model calculations show that the increase in the atmospheric aerosol burden is delaying the global warming expected from the increase in greenhouse gasses (GHG: CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, halocarbons). Whereas the increase in GHGs since pre-industrial times is producing a warming of  $2.4 \text{ W m}^{-2}$ , the overall cooling effect of aerosols might be up to  $-2.5 \text{ W m}^{-2}$  (IPCC, 2001). The latter value is composed of contributions by e.g. sulphate and organic particles, which have a cooling effect, and black carbon, which has a heating effect. International and EU climate change policies aim at reducing the emissions of GHGs by implementing the Kyoto Protocol. It is expected that negotiations of reductions beyond the Kyoto Protocol might consider also the role of aerosols (Hansen et al., 2000). The effect of policies on climate to reduce adverse effects of aerosols on health (see above) also needs to be addressed. As with the health issue, knowledge about physical and chemical aerosol characteristics and their relationship with sources is important to develop cost-effective mitigation policies.

### 1.3. Aerosol modelling

Numerical models describing aerosol particle emissions and production, transport, transformation and removal have become accepted tools to extrapolate monitoring data (Council Directive 96/62/EC of 27 September 1996 on ambient air quality). They are used in source apportionment studies and, obviously, they are the only tools to assess the effects of future changes in aerosol and aerosol precursor emissions. The calculation of PM<sub>10</sub> levels or radiative forcing must necessarily be based on a description of the emissions of the individual

chemical species and how they transform and mix in the atmosphere. Models with this capability are becoming available (Hass et al., 2002) but need testing and validation against fundamental data such as the aerosol particle size distribution and chemical composition.

## 2. Compilation of European data

### 2.1. The network

The present aerosol “phenomenology” synthesizes physical data that have been collected during the last 10 years. In this paper we present and discuss simultaneously measured PM<sub>10</sub> and PM<sub>2.5</sub> mass, and (sub-micrometer) number size distributions, from which integrated properties such as number concentration, (fine fraction) aerosol volume and surface area can be calculated. Such data are presently not measured in regulatory monitoring networks (such as EMEP and EUROAIRNET), but rather in research projects. Here we consider only projects providing data representative for a site during at least a season (i.e. minimum 6 weeks of continuous measurements). Sites and instrumentation are listed in Appendix A.

Fig. 1 shows the location of the 31 sites, operated by 12 institutes. We will refer to these sites as “The Network”. We have categorized the sampling sites using criteria proposed by the European Environment Agency (Larssen et al., 1999). Among those criteria are the distance of the station from large pollution sources such as cities, power plants and major motorways, and the traffic volume.

- 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—<2500 vehicles/day within a radius of 50 m

To which we have added the

- Free troposphere—above the mixed boundary layer
- Kerbside—within street canyons

In the text we will also refer to

- Clean sites—free troposphere, natural and rural background sites,

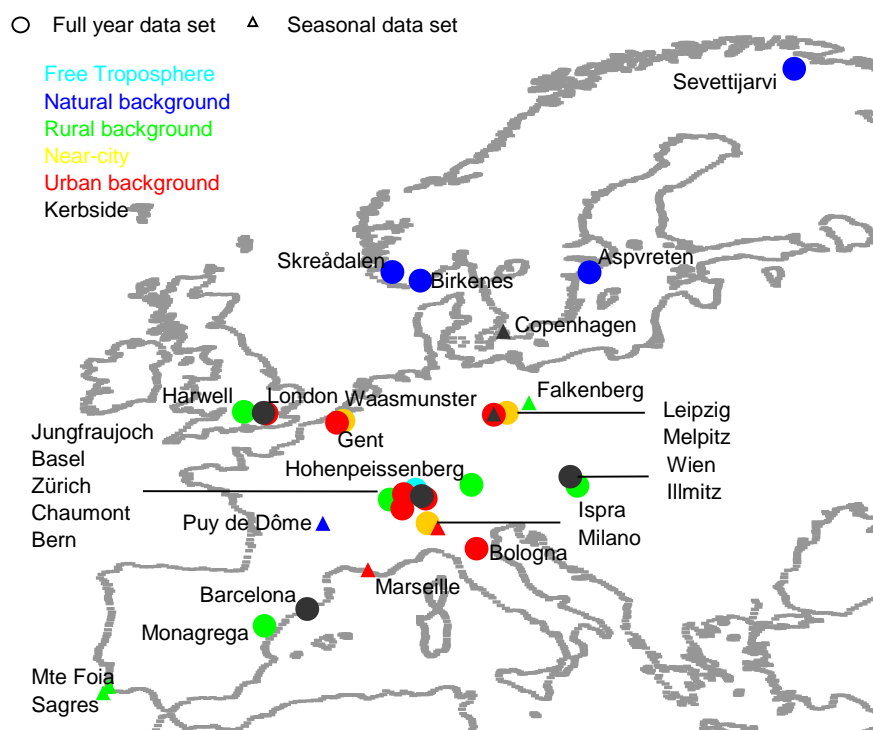


Fig. 1. Location of the sampling sites.

- Polluted sites—near-city, urban background and kerbside sites

## 2.2. Data consistency and representativeness

### 2.2.1. Systematic sampling and analysis errors

The data set was obtained from a number of measurement and monitoring campaigns, geographically and chronologically scattered, by different research groups, using different techniques, in particular concerning PM<sub>2.5</sub> and PM<sub>10</sub> mass determination. Comparing data from such a compilation requires that systematic errors and temporal variations that occurred over this period are carefully evaluated and considered in the interpretation.

Possible systematic errors in PM mass determination, resulting from the use of different samplers, sampling heads, substrates, gravimetric analysis and positive or negative sampling artefacts for the various chemical compounds in particulate matter, are discussed in the companion paper by Putaud et al. (2004). The major source of uncertainty in the mass determination originates from positive and negative artefacts in the capturing of ammonium nitrate and semi-volatile organic compounds on filters or other substrates. The magnitude of these artefacts depends strongly on the actual chemical composition of the aerosols, as well as on meteorological conditions.

Also the presence of particle-bound water during off-line gravimetric mass determination at 50% humidity can cause a positive artefact. This may be an important source of inconsistency between the PM mass concentrations determined according to the EN12341 norm and TEOMs. TEOMs indeed dry the sampled air stream to limit the quantity of water associated with aerosol particles. Routine TEOMs do this by heating the inlet at 50°C, whereas TEOMs equipped with a sample equilibration system (SES) achieve RH < 30% by sampling through dryers and heating at 30°C only.

Several studies, covering a range of ambient conditions have demonstrated that, because of these combined problems, routine TEOMs underestimate PM<sub>10</sub> measurements by up to 35%, when compared with the EN12341 reference method (Airborne Particles expert group, 1997; Allen and Reiss, 1997). This underestimation is more severe in winter than in summer, because in summer the ambient and instrument temperatures are more comparable. On an annual basis, and for the different conditions for this work, we estimate that a 35% difference between gravimetric PM<sub>10</sub> measurements carried out at 50% RH and on-line measurements using TEOMs seems therefore to be an upper limit, which we also extend to PM<sub>2.5</sub>, and the PM mass data presented here are comparable within 35%.

Particle size distributions were measured using differential mobility analysers (DMA) connected to a particle

counter (CPC). Some systems worked in scanning mode (SMPS) and others in step modes (DMPS). Standard deviations among 11 SMPS and DMPS instruments were shown to be <22% and <10% for sizing and counting, respectively (Dahmann et al., 2001).

The size measured by the DMA is affected by the RH at which the DMA is operating, and should therefore be specified. Most size distributions were obtained at RH < 20% (see Appendix A) and therefore comparable. Size distribution measurements at the sites Harwell, Bloomsbury and Marylebone were obtained at unspecified ambient relative humidity. Assuming a particle growth factor of the “more hygroscopic particles” between 1.2 and 1.5 at 90% RH (see e.g. Baltensperger et al., 2002), and an ambient RH between 50% and 75%, this may lead to an increase of the average particle size with 5–28% and integrated aerosol volume increase of 16–110% compared to the dry size and volume.

The particle concentration is obtained by integrating the number size distribution. As the range of particle diameters measured by various groups is different (Appendix A), we considered the number of particles with  $D_p > 10$  nm when comparing number concentrations between sites. Number concentrations obtained in this way are expected to be comparable within 10%.

### 2.2.2. Temporal representativeness

Most of the PM data are obtained in the period 1998–2000, and span from 1 to 3 years (except for the short campaign datasets which are not considered in the annual averages). We have, however, also included older PM data from Scandinavian natural stations, as well as for two Belgian near-city and urban sites (all obtained before 1996).

When comparing PM characteristics of the sites with “old” data to sites with recent data, one must consider a possible bias in the former, mainly due to a generally European-wide downward trend in SO<sub>2</sub> emissions during the last decade, which is being reflected in a downward trend in particulate sulphate (see Fig. 2a in Putaud et al., 2004). However, this trend is not as obvious to detect in the particulate mass measurements. For the Scandinavian background stations, EMEP data archives have been consulted. The EMEP network has recently started the conversion (in the period 1999–2001) from TSP to PM<sub>10</sub> sampling as required by European norm EN12341. Therefore, no contiguous sampling period with a single technique exists from which the trend in the period 1995–2001 can be evaluated from this data set. Still, 2000–2001 PM<sub>10</sub> data for the EMEP site Birkenes (Tørseth et al., 2003, Eurotrac AEROSOL final report) can be compared with the PM<sub>10</sub> data for that site (this work) collected by the Ghent University in the period 1991–1996. The average PM<sub>10</sub> concentration at

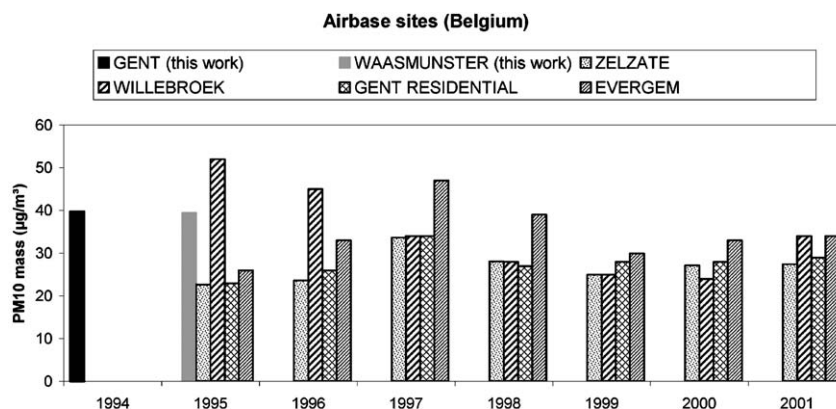


Fig. 2. Trend in PM10 mass for four representative sites in the region of the Waasmunster and Gent sites presented in this work (source: AirVIEW, Air quality Visualization Instrument for Europe on the Web, EEA European Topic Centre on Air and Climate Change).

Birkenes from November 2000 to October 2001 is  $6.3 \mu\text{g m}^{-3}$ , which is indeed 20% lower than the 1991–1996 average. On the other hand, this difference is within the range of the inter-annual variability observed in long-term trends of EMEP TSP since 1991 (e.g. <26% for German EMEP sites, < 20% for Swiss EMEP sites).

For an evaluation of the near-city and urban sites of Waasmunster and Gent, respectively, in Belgium (the sites being 16 km apart), we consulted the AIRBASE database of the “European Topic Centre on Air and Climate Change” (<http://bettie.rivm.nl/etc-acc/appletstart.html>). PM10 data since 1995 are available for four sites within a radius of 16 km from Waasmunster, including another site in Gent. Fig. 2 shows the trend in annual averages for PM10 mass, for the four sites. Also the PM10 levels for the 2 sites in this study for the years 1994 and 1995 are shown. The Waasmunster and Gent levels for 1994–1995 fall within the range of measured PM10 levels in the four other sites. From 1995 to 2001, absolute concentrations, as well as the difference in PM10 levels between the four sites become smaller. For the period 1998–2000, annual averages range between 24 and  $39 \mu\text{g m}^{-3}$ , with an average over that period for the four sites of  $29 \mu\text{g m}^{-3}$ . The downward trend in PM10, which occurred almost entirely in the period 1995–1998, indicates that the old data are probably overestimating the PM10 levels by about 30% when confronting them with more recent data. However, also here inter-annual variability can be as high as 35%.

The lack of trend observed in the Belgian data between 1998 and 2001 is also documented by Gehrig and Buchman (2003) for PM10 and PM2.5 annual averages for seven Swiss measurement stations (including the ones presented in this work). Together with similar observed features in EMEP data archives for

TSP in German sites (since 1977), this suggests that the more recent data from our compilation are probably not biased due to inter-annual trends.

Size distribution data in this study have been obtained throughout the period 1997–2000. Particle number shows in general a much larger variability than the mass concentration, in particular, in the size range below 100 nm which is highly dependent on local emissions and atmospheric processes such as nucleation, condensation and coagulation. As the distribution data for this study have been obtained in a relatively brief time span, we assume that the observed differences between various sites are in the first place due to the site characteristics, season and time of the day, and not due to inter-annual trends.

### 3. PM10 and PM2.5 mass concentrations

Fig. 3 illustrates which fractions of the total suspended particles (TSP) are accounted for by PM2.5 and PM10 measurements. In this example obtained at an urban site in Sweden PM10 comprises most of the TSP mass. It illustrates the general appearance in the particle population of “fine” and “coarse” particle modes, with respective geometric mean aerodynamic diameters  $<1 \mu\text{m}$  and  $>1 \mu\text{m}$ . In the following we will refer to “coarse particles” as the fraction of particles between PM10 and PM2.5, although from the shown example it is clear that, apart from the fine particle mode, PM2.5 also may include a significant fraction of the coarse particle mode(s).

PM10 mass concentrations are started to be monitored by national and international (EMEP, EURO-AIRNET) air quality networks, where they are replacing TSP measurements. Here we show yearly statistics



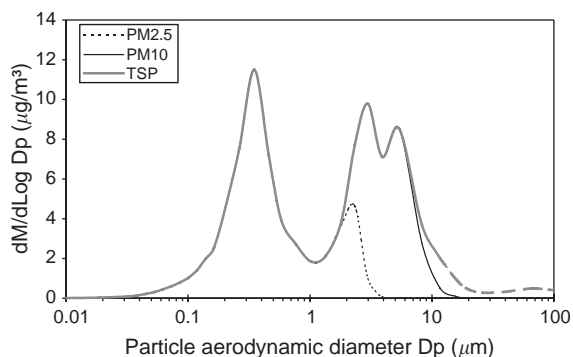


Fig. 3. Fractions of the aerosol that is accounted for by total suspended particles (TSP), PM10 and PM2.5 measurements, for an urban site in Sweden.

regarding 24-h averaged PM10 and PM2.5 concentrations observed at 23 sites in The Network. Figs. 4a and b show the 5%, 25%, 50% (median) 75% and 95% percentiles of 24-h integrated PM10 and PM2.5 mass concentrations, as well as their annual averages. PM10 and PM2.5 annual average concentrations both vary by more than a factor of 10 between their lowest (Sevettijarvi, Finland) and highest values (Wien, Austria; Bologna, Italy). This variation is larger than that for annually averaged ozone values, which in Europe range from about 35 to 105  $\mu\text{g m}^{-3}$ , hence a factor of 3 only (Hjellbrekke and Solberg, 2002). This shows that PM mass concentration is more sensitive to local sources than ozone.

Fig. 4 also shows that the 5% percentile values at rural and near city (and even in some of the urban) background sites are similar to the annual average concentrations observed at natural background sites. This means that the former sites encounter natural background conditions in about 5% of the days. From this observation, we derive a European continental background concentration by taking the average of the natural background annual mean concentrations and of the rural and near-city background 5% percentile values. We obtain values of 7.0  $\mu\text{g m}^{-3}$  ( $\pm 4.1$ ) and 4.8  $\mu\text{g m}^{-3}$  ( $\pm 2.4$ ) for PM10 and PM2.5, respectively. This latter value is about 5–10 times lower than the highest values observed in The Network. However, this background is not purely natural: the presence of black carbon (BC) in the aerosol at natural background sites (see the companion paper by Putaud et al., 2004) indicates that these sites are also affected by (long-range transport of) combustion sources.

When sites are ordered according to the European Environmental Agency (EEA) criteria, PM10 and PM2.5 annual average concentrations do not gradually (i.e. monotonically) increase when moving from

near-city background to urban background to kerbside sites. In fact, concentrations at kerbsides in one city might be lower than at urban background sites elsewhere (e.g., PM2.5 at Barcelona, E, and Bologna, I). PM levels at Waasmunster and Gent have to be considered as 20–30% too high as discussed before. This does, however, not affect this (lack of) trend, which has also been observed in a compilation of PM10 data in the EMEP/EUROAIRNET networks (Kahnert, 2002).

The similarity in PM level between near-city, urban and kerbside sites is even more striking in the PM2.5 data set (Fig. 4b). The production of secondary aerosol during the aging of pollution plumes (Wehner et al., 2002; Baltensperger et al., 2002), combined with sedimentation of re-suspended dust produced at the kerbside, could explain this picture.

Only a few sites exceed the EU annual PM10 standard of 40  $\mu\text{g m}^{-3}$  targeted by 2005, whereas all near-city, urban background and kerbside sites are above the EU annual PM10 standard of 20  $\mu\text{g m}^{-3}$  targeted for 2010. These data are consistent with those obtained from larger networks, which show that annual average PM10 concentrations reach 50  $\mu\text{g m}^{-3}$  in Western Europe and up to 60–70  $\mu\text{g m}^{-3}$  in Eastern Europe (Lazaridis et al., 2001).

The annual 95% and 75% PM10 percentiles obtained from 24-h averaged values, indicate that the EU 24-h PM10 limit value of 50  $\mu\text{g m}^{-3}$  is exceeded more than 18 and 90 times a year, respectively, at most near-city, urban background and kerbside sites. Targets are 35 exceedences a year by 2005 and 7 exceedences a year by 2010 (this is equivalent to the 90 and 98 percentiles, respectively, being lower than 50  $\mu\text{g m}^{-3}$ ).

As of today the EU has not put forward any PM2.5 limit values. However, the annual average PM2.5 mass concentration at all near-city and urban background sites and at kerbside sites exceeds the US-EPA standard of 15  $\mu\text{g m}^{-3}$ .

In the light of a possible conversion from PM10 to PM2.5 sampling for future air quality standards, it is of interest to investigate the relation between PM2.5 and PM10 for the various site types. The question to address is: what (averaged) PM2.5 value corresponds to a given (averaged) PM10 value at a given site, and is this dependent on the absolute PM10 concentration? For each site, all daily PM10 data (and their corresponding PM2.5 values) were grouped and averaged over 10  $\mu\text{g m}^{-3}$ -wide bins (e.g. 30 < PM10 < 40  $\mu\text{g m}^{-3}$ ), resulting in a single (PM10, PM2.5) pair within each PM10 concentration bin for each site, and maximum 8 pairs for each site.

Fig. 5 shows these averages of simultaneously measured PM2.5 vs. PM10 mass concentrations for 11 sites of The Network. PM2.5 and PM10 mass concentrations are clearly correlated with a mean PM2.5 vs.

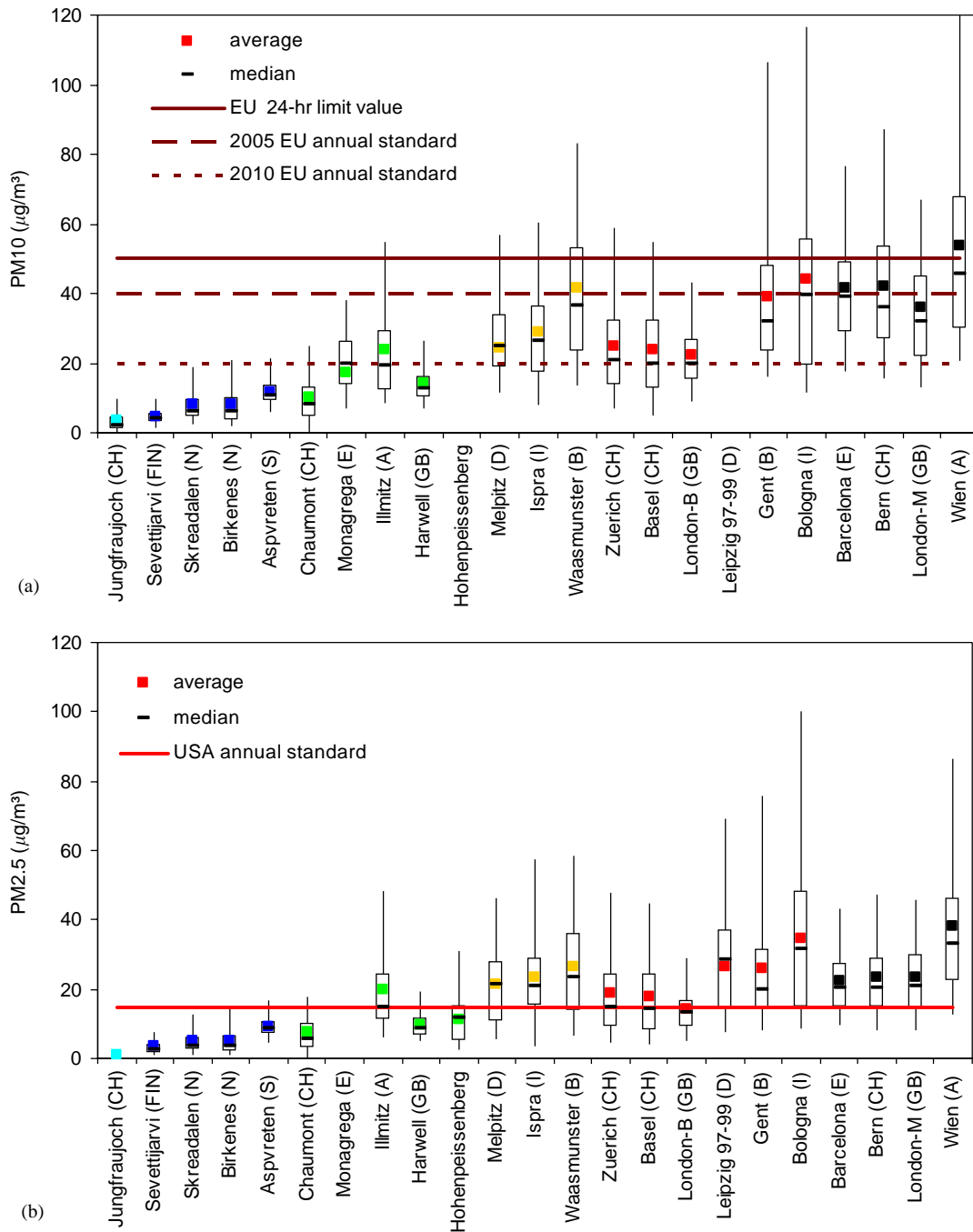


Fig. 4. (a, b) Annual averages of PM10 and PM2.5 mass concentrations, including the 5%, 25%, 50% (median) 75% and 95% percentiles of their 24-h integrated concentrations.

PM10 slope =  $0.73 \pm 0.02$  (95% confidence level). Individual PM2.5/PM10 ratios range from 0.5 to 0.9, but Fig. 5 also shows that a given site has its characteristic

PM2.5/PM10 slope. Slope values for each site are given in Table 1. The good correlation for single sites is due to the fact that meteorology (dispersion) is the main factor

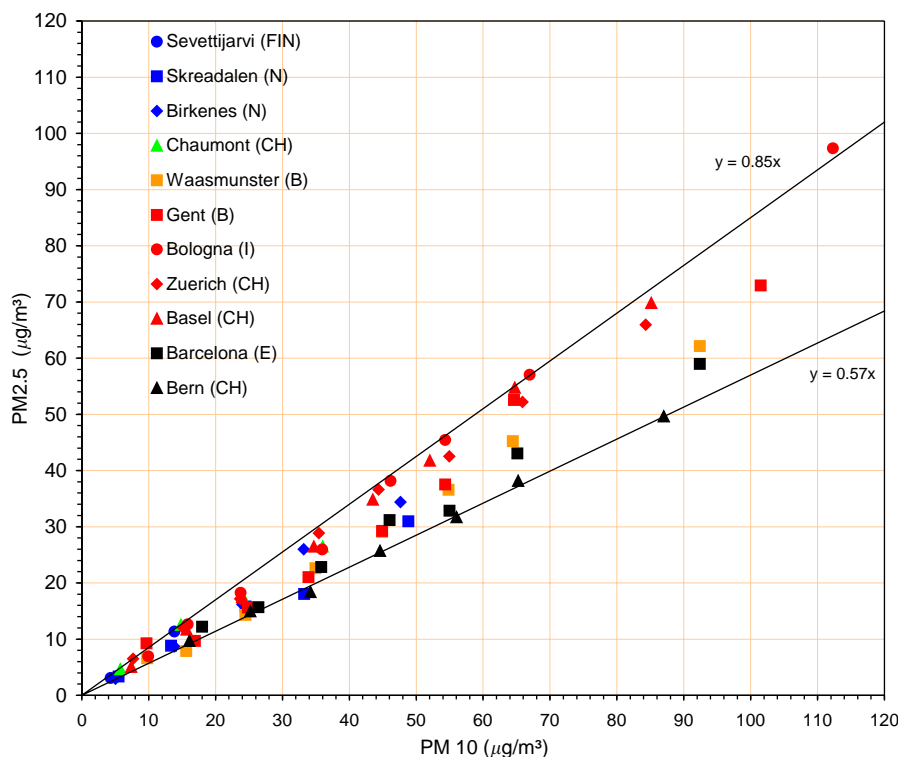


Fig. 5. Averages of simultaneous measured PM<sub>2.5</sub> vs. PM<sub>10</sub> mass concentrations.

Table 1

Site-specific slope and correlation coefficient between “binned” PM<sub>2.5</sub> and PM<sub>10</sub> 24-h values (see text and Fig. 5)

Site type	Site name	Slope	$R^2$	Number of values
Rural	Sevetijarvi (FIN)	0.87		2
Rural	Skreadalen (N)	0.61	0.983	5
Rural	Birkenes (N)	0.76	0.992	5
Rural	Chaumont (CH)	0.71	0.995	4
Rural	Waasmunster (B)	0.70	0.996	8
Urban	Zuerich (CH)	0.78	0.998	8
Urban	Basel (CH)	0.85	0.998	8
Urban	Gent (B)	0.75	0.980	8
Urban	Bologna (I)	0.89	0.998	8
Urban	Barcelona (E)	0.64	0.992	7
Kerbside	Bern (CH)	0.57	0.998	7

controlling PM mass concentrations. It also suggests that the intensities of fine (PM<sub>2.5</sub>) and coarse (PM<sub>10</sub>–PM<sub>2.5</sub>) aerosol sources co-vary.

Lower ratios are observed for kerbside sites (Barcelona, E, Bern, CH), suggesting a large contribution of

re-suspended road dust to the coarse fraction. Higher ratios are observed at natural, rural, near-city and urban background sites where secondary aerosol sources, which produce fine particles, are predominant.

Fig. 6 shows the PM<sub>2.5</sub>/PM<sub>10</sub> ratio for the different sites, further categorized according to the levels of PM<sub>10</sub> mass concentration. This figure refines the observations in Fig. 5, showing that the PM<sub>2.5</sub>/PM<sub>10</sub> ratio increases with PM<sub>10</sub> level. This indicates that pollution periods are predominantly due to increases in the PM<sub>2.5</sub> mass concentration. This is true except at the kerbside sites Bern (CH) and Barcelona (E), which again indicates the large contribution of re-suspended road dust to high PM<sub>10</sub> concentrations at such sites. Barcelona, in Eastern Spain, is furthermore influenced by coarse aerosol of marine and African dust origin. Recent and ongoing studies (Querol et al., 2003; Rodríguez et al., 2002, 2003) show indeed that around 10 African dust events/year (2–4 days duration each) take place, which are characterized by 24-h mean mineral dust concentrations usually  $>25 \mu\text{g m}^{-3}$  in PM<sub>10</sub> and  $10\text{--}15 \mu\text{g m}^{-3}$  in PM<sub>2.5</sub>. The annual mean mineral dust load in PM<sub>10</sub> ranges from 37% at kerbside and 30% at urban background to 13–20% at rural background sites.



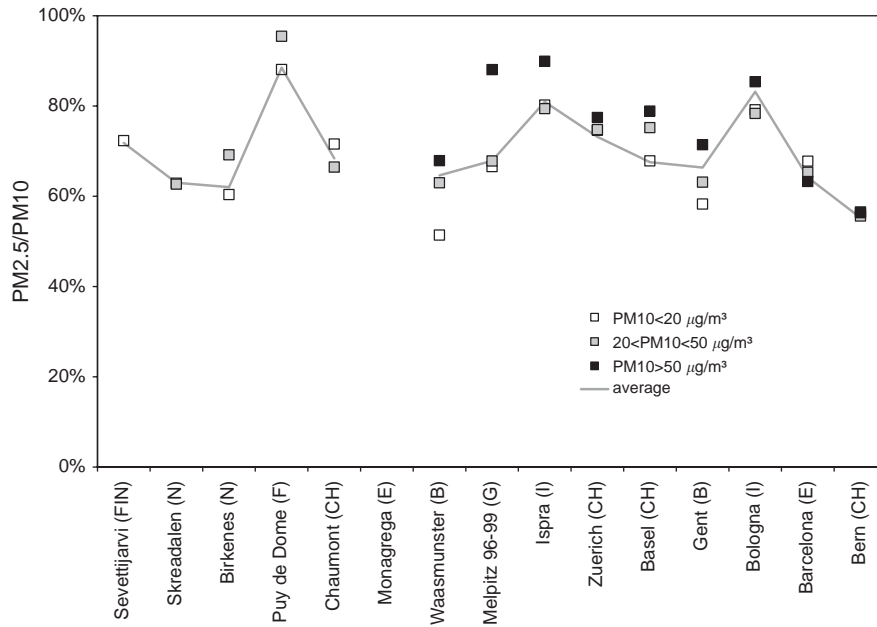


Fig. 6. PM<sub>2.5</sub>/PM<sub>10</sub> ratio for the different sites, further categorized according to the levels of PM<sub>10</sub>.

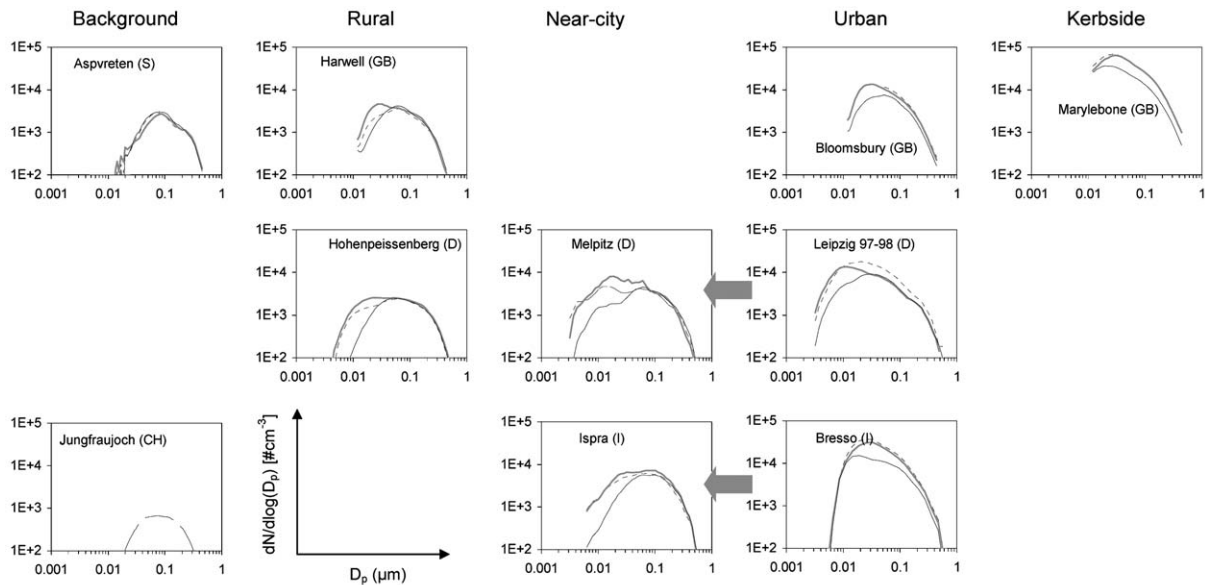


Fig. 7. Median particle number size distributions during summer, during morning hours (black dashed line), afternoon (grey full line) and night (black full line).

#### 4. Particle size distributions and derived properties

We collected aerosol particle distributions from 15 sites of The Network. Sites were selected where

measurements lasted long enough to calculate seasonal averages. From the size distributions, the aerosol number concentrations were derived by integrating the size distribution from  $D_p = 10$  nm upwards. The size

distributions were measured with Differential Mobility Analysers and typically do not include particles with  $D_p > 800$  nm. The collected size distributions were grouped into seasons and into 3 time periods of the day (morning hours, afternoon, night). Fig. 7 shows median particle number size distributions from 10 sites of The Network during summer (Fig. 7a) and winter (Fig. 7b) for each of the three periods of the day. Log-normal mode parameters for 3-mode fittings of the number size distributions can be found in Putaud et al. (2003).

Morning size distributions are expected to be largely influenced by traffic (rush hours in cities), and afternoons are the most intensive photochemistry periods. The aerosol observed during night time should indicate the local background, i.e. the prevailing size distribution when emissions and secondary aerosol formation are minimal (but not necessarily absent). The arrows indicate that Melpitz (Germany) and Ispra (Italy) are influenced by Leipzig (Germany) and Bresso-Milano (Italy), respectively.

At kerbside and urban background sites, particle number size distributions are dominated by a mode with a maximum at  $D_p = 20$ – $30$  nm, but also show a second mode with a maximum around 100 nm. Part of this mode is to be attributed to the regional background (see below). However, the particle number concentration in both modes increases during morning hours. This is most visible at the kerbside site, indicating that these two modes are related to/affected by traffic. Measurements in the urban background of Milano indicated that the smallest particles consist mostly of semi-volatile organic matter, whereas freshly emitted soot particles appeared with a diameter  $> 50$  nm (Baltensperger et al., 2002).

The summer time size distributions of the rural, near-city and urban background sites also show a maximum around  $D_p = 10$ – $30$  nm, during the afternoon, whereas this mode is mostly absent during winter. We conjecture that this is due to photochemically induced nucleation of new particles in the local atmosphere. Major research programmes have been undertaken recently to understand such nucleation events and their role in sustaining the atmospheric particle number concentration (e.g. Kulmala et al., 2001; Boy and Kulmala, 2002; Uhrner et al., 2003).

In moving from kerbside to natural background sites there is a gradual decrease in the number concentration and a gradually decreasing contribution of particles with  $D_p = 10$ – $30$  nm. This can generally be explained as follows: the clean sites are mainly affected by transport of aerosol particles from the polluted regions. During transport, dilution and dispersion reduce the number concentration, whereas coagulation not only reduces the number but also removes the smaller particles from the size distribution (small

particles collide with the bigger ones). Wehner et al. (2002) showed indeed that the transformation from kerbside to urban background size distributions happens by coagulation, condensation and dilution during dispersion of the urban source aerosols on a time scale of less than 1 h.

Winter time size distributions (not shown) show broadly the same features and diurnal trends as summer time distributions. The major difference is the lack of enhanced number concentrations during the afternoon (in fact afternoon distributions coincide practically with rush hour distributions). Further, in Ispra (Italy, near-city site), urban background and kerbside sites, winter time size distributions show a shift towards larger numbers and diameters compared with the summer time distributions, in particular during afternoons. Increased diameters can be explained by the condensation of semi-volatile species, which is favoured by cold temperatures in winter (see companion paper, Putaud et al., 2004).

Fig. 8 shows the seasonal average of (a) PM<sub>2.5</sub> mass and (b) particle number concentrations, for those sites where both measurements are available simultaneously. These sites are different from those plotted in Fig. 4, where we showed the sites where simultaneous annual averaged PM<sub>10</sub> and PM<sub>2.5</sub> data are available. Here, all data have been obtained within the period 1997–2000 (except Marseille: 2001) so biases due to long term trends are minimal. We also have included shorter data sets obtained during one season only.

PM<sub>2.5</sub> mass concentrations are highest during winter in the polluted sites. The large seasonal variations in PM<sub>2.5</sub> mass concentrations at those sites are generally not related to large seasonal variations in particle number concentrations. This indicates that processes other than meteorological ones are active. Large PM<sub>2.5</sub> mass concentrations during cold seasons are likely to be due to the fact that particles are on average larger (size distributions are shifted towards larger diameter) due to the condensation of semi-volatile species.

Among the sites in Fig. 8 and excluding the free tropospheric site (Jungfraujoch, Switzerland), the annual average PM<sub>2.5</sub> concentration varies by a factor of about 3 between its minimum (Aspvreten, Sweden) and maximum value (London-M, Great Britain). The annual average particle number concentration, however, varies by a factor of about 10 between the same sites. This is explained by the fact that the high number concentrations in urban and kerbside sites are due to particles with  $D_p < 100$  nm, which hardly contribute to the PM<sub>2.5</sub> mass concentration.

Fig. 9 shows the relationship between the seasonal average PM<sub>2.5</sub> mass concentration and (a) the corresponding number concentration of particles with

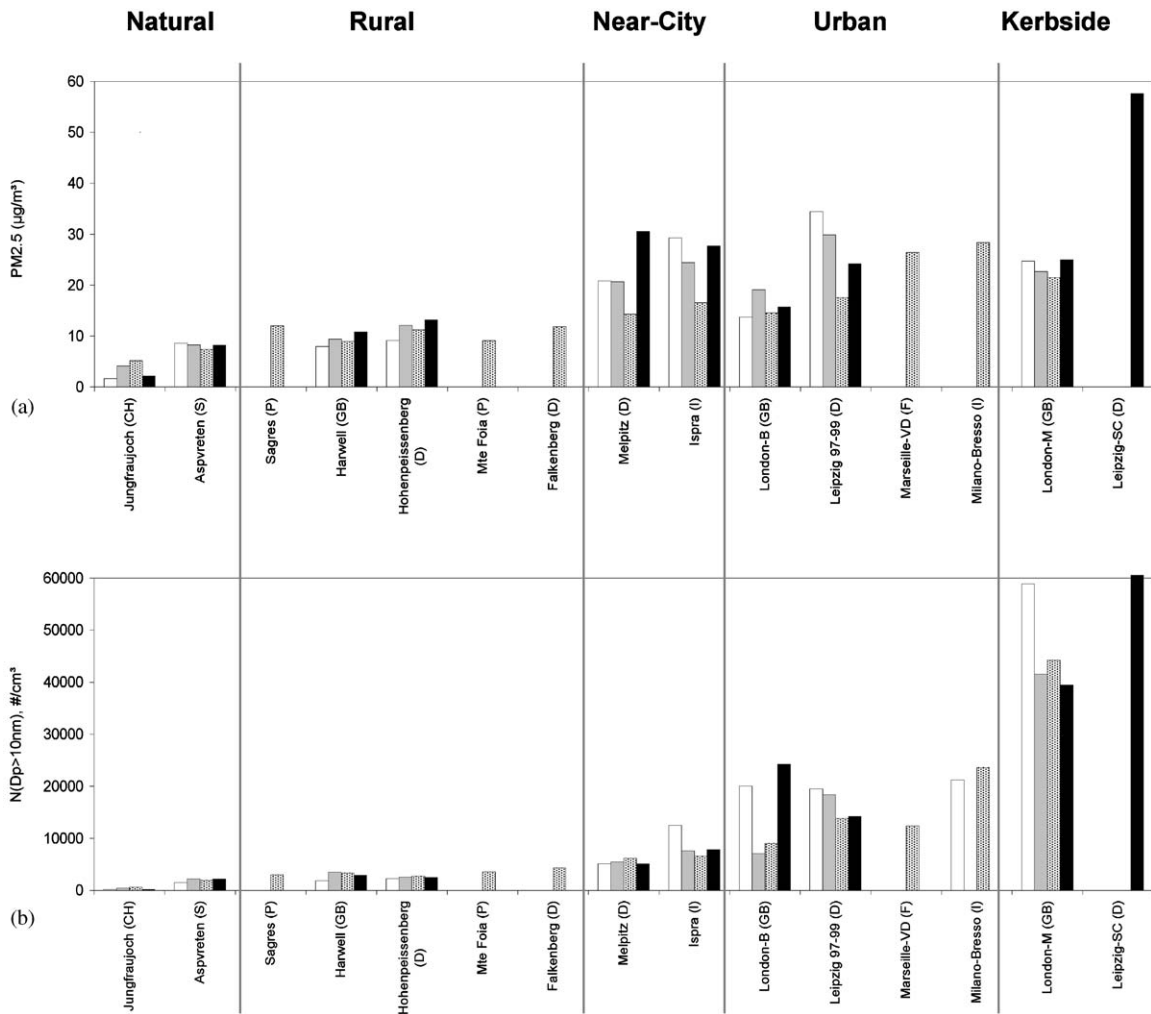


Fig. 8. The seasonal average of (a) PM<sub>2.5</sub> mass concentration and (b) particle number concentrations ( $D_p > 10 \text{ nm}$ ). Winter (DJF, white), Spring (MAM, grey), Summer (JJA, dashed), Fall (SON, black).

$D_p > 10 \text{ nm}$ , (b) particles with  $D_p > 100 \text{ nm}$  and (c) the particle volume concentration. The different colours refer to the different sites (natural, rural, near-city, urban, kerbside: see Fig. 4) and the different symbols refer to the different seasons (square = winter (DJF), triangle = spring (MAM), circle = summer (JJA), diamond = autumn (SON)).

At the clean and rural sites, there is a good linear correlation between PM<sub>2.5</sub> and the particle number concentrations. Fig. 9a shows indeed that for those sites the relation between PM<sub>2.5</sub> and  $N > 10 \text{ nm}$  can be described by  $N > 10 \text{ nm} (\text{cm}^{-3}) = 250 \text{ PM}_{2.5} (\mu\text{g m}^{-3})$ . This relation apparently represents the minimum particle number concentration associated with a given PM<sub>2.5</sub> loading. Increasingly higher number

concentrations are observed at urban background and kerbside sites respectively, which can be interpreted as due to local sources. The features shown in Fig. 9a are consistent with the notion that particle number decays rapidly when moving away from the kerbside (see Wehner et al., 2002), and that local sources of small particles do not contribute much to the seasonal average number concentration at clean sites.

A similar “minimum relation” between PM<sub>2.5</sub> and number can be obtained for particles larger than 100 nm  $N > 100 \text{ nm} (\text{cm}^{-3}) = 50 \text{ PM}_{2.5} (\mu\text{g m}^{-3})$ . Apart from 2 outliers, the over-all correlation between PM<sub>2.5</sub> and number is better than in Fig. 9a because these larger particles are the ones that contribute to the mass. The

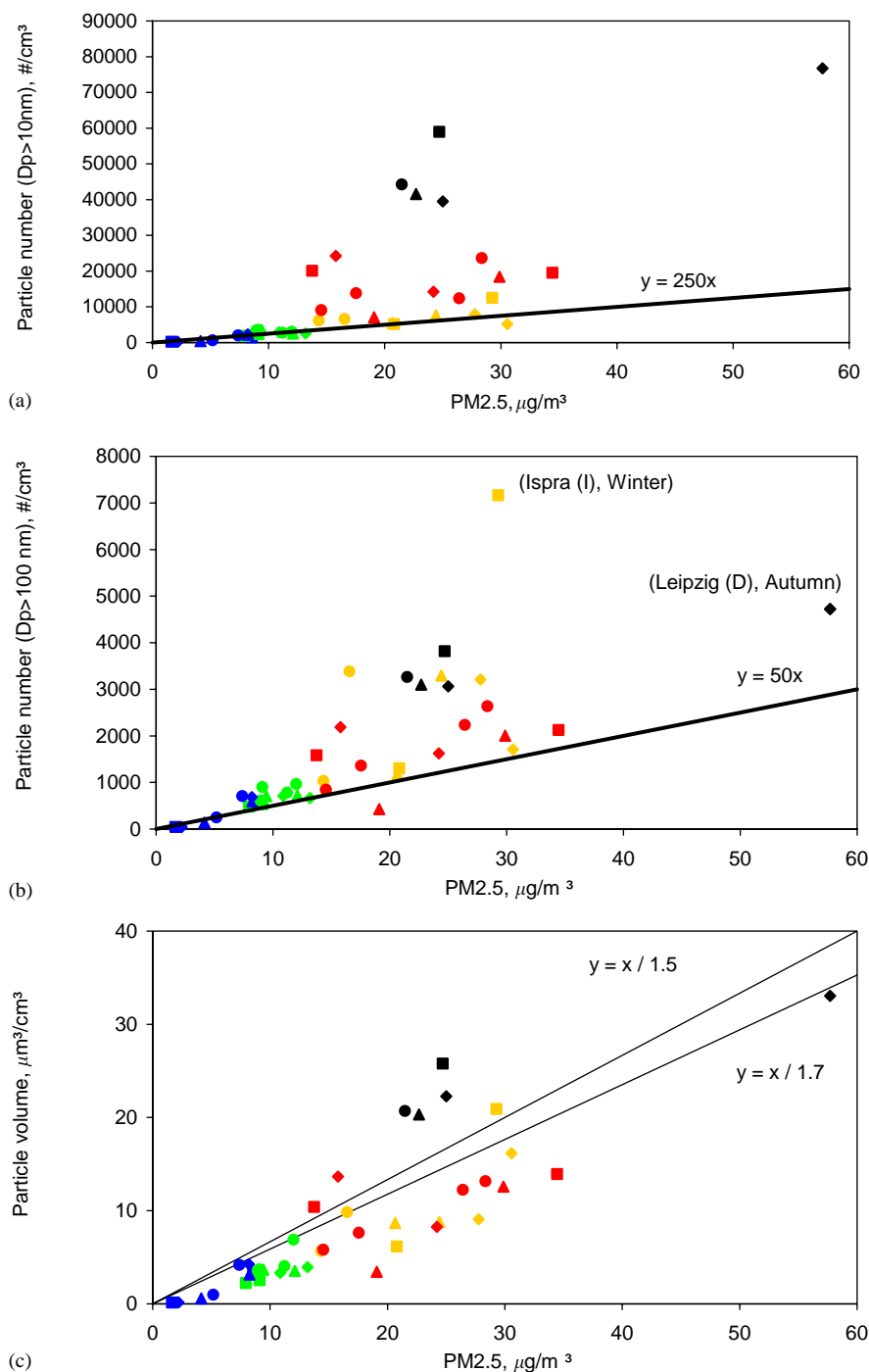


Fig. 9. Relationship between the seasonal average PM<sub>2.5</sub> mass concentration and (a) the corresponding number concentration of particles with  $D_p > 10$  nm, (b) particles with  $D_p > 100$  nm and (c) the particle volume concentration. Colours refer to different sites as in Fig. 4.

extremely high  $N_{>100}$  value for Ispra can be explained by the large shift in particle size during winter (see discussion on the size distributions).

Comparison of particle numbers in Figs. 9a and b shows that 70–80% of the particles have a  $D_p < 100$  nm. The size-segregated composition of the aerosol at two

polluted sites (see Putaud et al., 2004) shows that in this size range, particles mainly consist of carbonaceous material. Hence, this indicates that, in terms of particle number, most of the particles consist mainly of organic and elemental carbon. Sulphates and nitrates may contribute significantly to the mass, but this happens through a “minority” of larger particles.

PM<sub>2.5</sub> mass and sub-micron particle volume concentrations are fairly well correlated (Fig. 9c). For identical cut-off sizes for volume and mass measurements, the ratio PM<sub>2.5</sub>/volume would correspond to the aerosol density, which is, based on the chemical composition, expected to range between 1.5 and 1.7 (Mc Murry et al., 2002). In our data set, size distributions have been obtained below 800 nm. In fact, most points, corresponding to rural, near-city and urban background sites, are located below the lines. This indicates that the sub-micron aerosol mass accounts only for a part of the PM<sub>2.5</sub> mass and confirms what was shown as the example in Fig. 3, namely that PM<sub>2.5</sub> comprises a significant fraction of coarse-mode particles.

The data points falling above the lines correspond to size distributions obtained at ambient relative humidity. As explained earlier, the corresponding dry volume can be as low as 50–85% of the shown value which would bring the values in the line of the other (dry) volume data.

## 5. Summary and conclusions

In this study we have presented a compilation of aerosol physical characteristics from 31 European sites. PM<sub>10</sub> and PM<sub>2.5</sub> mass, as well as number size distributions have been presented as they have been measured with current sampling methods. In spite of the geographical, chronological and methodological heterogeneity in the presented dataset, we believe it contains relevant information which cannot be obtained from routine PM mass-only monitoring.

Background annual average PM<sub>10</sub> and PM<sub>2.5</sub> mass concentrations for continental Europe have been derived from 5 percentile values to be  $7.0 \pm 4.1 \mu\text{g m}^{-3}$  and  $4.8 \pm 2.4$ , respectively. PM concentrations in urban background sites in some areas in Europe can be as

high as PM concentrations at kerbside sites in other parts of Europe, which shows the importance of the regional aerosol background, in particular when considering PM<sub>2.5</sub>.

Considering all sites in The Network, PM<sub>2.5</sub> and PM<sub>10</sub> mass concentrations are correlated with a correlation coefficient  $R^2 = 0.95$ . The over-all PM<sub>2.5</sub>/PM<sub>10</sub> ratio ( $0.73 \pm 0.15$ ) is too variable to propose a European-wide valid PM<sub>2.5</sub> to PM<sub>10</sub> ratio. However, site-specific ratios can be obtained, ranging between 0.57 and 0.85.

An important observation is that pollution events (high PM<sub>10</sub>) are characterized by an increased contribution in fine aerosol, except at kerbside sites where coarse re-suspended or advected dust contributes more to PM<sub>10</sub>. In the latter sites, abatement strategies based on reduction of secondary aerosol, which is associated with the fine fraction, will be less effective.

Particle ( $D_p > 10 \text{ nm}$ ) number concentrations increase more than proportionally to PM mass. Number size distribution measurements explain this by a higher contribution of traffic- and photochemically generated small ( $D_p < 100 \text{ nm}$ ) particles when moving from clean to polluted sites. Comparison between number distributions in this work, and chemical size distributions in the companion paper, leads to the conclusion that the majority of the particles (70–80% in terms of number) consist mainly of carbonaceous material, which is highly relevant for health issues.

Considering data from all sites in The Network, no useful “universal” correlation is found between PM<sub>2.5</sub> or PM<sub>10</sub> on the one hand and the particle ( $D_p > 10 \text{ nm}$ ) number concentration on the other. The correlation is fairly good at clean sites but deteriorates in polluted sites. At polluted sites PM<sub>2.5</sub> and PM<sub>10</sub> mass concentrations, as well as the particle ( $D_p > 10 \text{ nm}$ ) number concentration are highest during winter.

More general comments regarding the status and future of aerosol monitoring in Europe are made in the accompanying paper (Putaud et al., 2004).

## Appendix A

Data availability, data sources, instruments and sampling devices are shown in Table 2.

Table 2

Institute	Site	Reference (see notes)	Lat	Long	Altitude (m)	Category	Period	Number size distribution, relative humidity	PM10 sampling	PM2.5 sampling	PM10 mass measurements	PM2.5 mass measurements
Ghent Univ.	Sevettijarvi (FIN)	1	69.35	28.50	130	Natural	Nov 93–Jan 96		Virtual impactor	Virtual impactor	Weighing (40% RH)	Weighing (40% RH)
Ghent Univ.	Skreådalen (N)	2, 3	58.82	6.72	465	Natural	Feb 91–Feb 96		Stacked Filter Unit	Stacked Filter Unit	Weighing (50% RH)	Weighing (50% RH)
ITM	Aspvreten (S)	Unpublished	58.80	17.40	20	Natural	Jan 00–Dec 00	DMPS (11–450 nm), dry				TEOM—50°C
Ghent Univ.	Birkenes (N)	2, 3	58.38	8.25	190	Natural	Feb 91–Feb 96		Stacked Filter Unit	Stacked Filter Unit	Weighing (50% RH)	Weighing (50% RH)
NERI	Copenhagen-Jagtvej (DK)	4	55.70	12.55	10	Kerbside	Feb 00–Mar 00	DMPS (6–700 nm), dry				
IfT	Falkenberg (D)	5	52.00	14.13	60	Rural	Jul 98–Aug 98	DMPS (3–800 nm), dry				Estimated from volume
U. Birmingham	Harwell (UK)	Unpublished	51.57	–1.32	125	Rural	May 98–Nov 00	SMPS (12–437 nm), amb.			TEOM—50°C	TEOM—50°C
IfT	Melpitz 97–99 (D)	6, 7	51.53	12.93	86	Near-city	Dec 96–Nov 97	DMPS (3–750 nm), dry	Hi Vol sampler		Weighing (50% RH)	
									Partisol (weekly)			
IfT	Melpitz 99–01 (D)	6, 7	51.53	12.93	86	Near-city	Dec 99–Nov 01		Hi Vol sampler		Weighing (50% RH)	Weighing (50% RH)
									Partisol (weekly)	Partisol (weekly)	Weighing (50% RH)	Weighing (50% RH)
U. Birmingham	London-Bloomsbury (UK)	Unpublished	51.52	–0.13	30	Urban	Mar 98–Nov 00	SMPS (12–437 nm), amb			TEOM—50°C	TEOM—50°C
U. Birmingham	London-Marylebone (UK)	Unpublished	51.52	–0.15	30	Kerbside	Mar 98–Nov 00	SMPS (12–437 nm), amb			TEOM—50°C	TEOM—50°C
IfT	Leipzig-SC (D)	6, 7	51.42	12.23	90	Kerbside	Oct 97–Nov 97	DMPS (3–750 nm), dry				Estimated from volume
IfT	Leipzig 97–99 (D)	6, 7	51.35	12.43	90	Urban	Aug 97–Feb 99	DMPS (3–750 nm), dry				Estimated from volume
IfT	Leipzig 99–01 (D)	6, 7	51.35	12.43	90	Urban	Feb 99–Apr 01	DMPS (3–800 nm), dry				Estimated from volume
Ghent Univ.	Waasmunster (B)	8	51.12	4.08	20	Near-city	Jul 94–Nov 95		Stacked Filter Unit	Stacked Filter Unit	Weighing (50% RH)	Weighing (50% RH)
Ghent Univ.	Gent (B)	8, 9	51.02	3.73	10	Urban	May 93–July 94 Sep 99–Oct 99		Stacked Filter Unit	Stacked Filter Unit	Weighing (50% RH)	Weighing (50% RH)
Umweltbundesamt	Illmitz (A)	10	48.23	16.36	117	Rural	Oct 99–Oct 00					
IfT	Hohenpeissenberg (D)	5	47.80	11.02	988	Rural	Apr 98–Aug 00	DMPS (3–677 nm), dry				Estimated from volume
Umweltbundesamt	Wien-Spittelauer Lände (A)	11	47.75	16.75	160	Kerbside	Oct 99–Oct 00					



PSI	Jungfraujoch (CH)	12	47.55	7.98	3580	Free troposph.	Jun 97–May 98	SMPS (19–788 nm), dry				Estimated from volume
EMPA	Basel (CH)	13, 22	47.53	7.58	316	Urban	Jan 98–Mar 99		Hi Vol sampler	Hi Vol sampler	Weighing (50% RH)	Weighing (50% RH)
EMPA	Zürich (CH)	13, 22	47.37	8.53	409	Urban	Jan 98–Mar 99		Hi Vol sampler	Hi Vol sampler	Weighing (50% RH)	Weighing (50% RH)
EMPA	Chaumont (CH)	13, 22	47.05	7.58	1136	Rural	Jan 98–Mar 99		Hi Vol sampler	Hi Vol sampler	Weighing (50% RH)	Weighing (50% RH)
EMPA	Bern (CH)	13, 22	46.95	7.43	545	Kerbside	Jan 98–Mar 99		Hi Vol sampler	Hi Vol sampler	Weighing (50% RH)	Weighing (50% RH)
EMPA	Payerne (CH)	13, 22	46.80	6.93	409	Rural	May 98–Oct 98		Hi Vol sampler		Weighing (50% RH)	Weighing (50% RH)
JRC	Ispra (I)	Unpublished	45.82	8.63	209	Near-city	Feb 00–Dec 00	DMPS (5–800 nm), dry	Klein Filter Gerät	Klein Filter Gerät	Weighing (30% RH)	Weighing (30% RH)
LaMP	Puy de Dome (F)	14	45.77	2.97	1465	Rural	Feb 00–Mar 01	CPC + PCASP, amb	ELPI and SDI Impactors	ELPI and SDI Impactors	Weighing (50% RH)	Weighing (50% RH)
JRC	Milano-Bresso (I)	15	45.53	9.20	130	Urban	May 98–Jun 98	DMPS (5–800 nm), dry	Virtual impactor	Virtual impactor	Weighing (20% RH)	Weighing (20% RH)
ISAC	Bologna (I)	16, 17	44.53	11.29	88	Urban	Jan 00–Dec 00		6-st. Berner impactor	6-st. Berner impactor	Weighing (20% RH)	Weighing (20% RH)
JRC	Marseille-Vallon Dol (F)	Unpublished	43.35	5.40	150	Urban	Jun 01–Jul 01	DMPS (7–566 nm), dry	9-st. Berner impactor	9-st. Berner impactor	TEOM-SES & weighing	Weighing (20% RH)
CSIC	Barcelona (E)	18, 19	41.37	2.12	30	Kerbside	Jun 99–Jun 00		Hi Vol sampler	Hi Vol sampler	Weighing (50% RH)	Weighing (50% RH)
CSIC	Monagrega (E)	19, 20	40.50	–0.20	598	Rural	Mar 99–Jul 00		Hi Vol sampler		Weighing (50% RH)	
IfT	Mt. Foia (P)	Unpublished	37.32	–8.62	902	Rural	Jun 97–Jul 97	DMPS (3–750 nm), dry			Weighing (60% RH)	Weighing (60% RH)
IfT	Sagres (P)	21	36.98	–8.95	50	Rural	Jun 97–Jul 97	DMPS (3–788 nm), dry	6-st. Berner impactor		Weighing (60% RH)	Weighing (60% RH)

DMPS: Differential mobility particle sizer; SMPS: Scanning mobility particle sizer; TEOM: Tapered element oscillating microbalance; CPC: Condensation particle counter; PCASP: Passive cavity aerosol spectrometer probe; SES: Sample equilibration system <sup>1</sup>Virkkula et al. (1999); <sup>2</sup>Maenhaut et al. (2000); <sup>3</sup>Tørseth et al. (1999); <sup>4</sup>Wählin et al. (2001); <sup>5</sup>Wiedensohler et al. (2002); <sup>6</sup>Wehner et al. (2002); <sup>7</sup>Wehner and Wiedensohler (2002); <sup>8</sup>Maenhaut and Cafmeyer (1998); <sup>9</sup>Maenhaut et al. (1996); <sup>10</sup>Maenhaut et al. (2002); <sup>11</sup>Schneider and Lorbeer (2002); <sup>12</sup>Weingartner et al. (1999); <sup>13</sup>Hüglin and Gehrig (2000); <sup>14</sup>Sellegrì et al. (2003); <sup>15</sup>Putaud et al. (2002a); <sup>16</sup>Decesari et al. (2001); <sup>17</sup>Matta et al. (2003); <sup>18</sup>Querol et al. (2001); <sup>19</sup>Rodríguez et al. (2002); <sup>20</sup>Rodríguez et al. (2003); <sup>21</sup>Neusüss et al. (2000); <sup>22</sup>Gehrig and Buchmann (2003).

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