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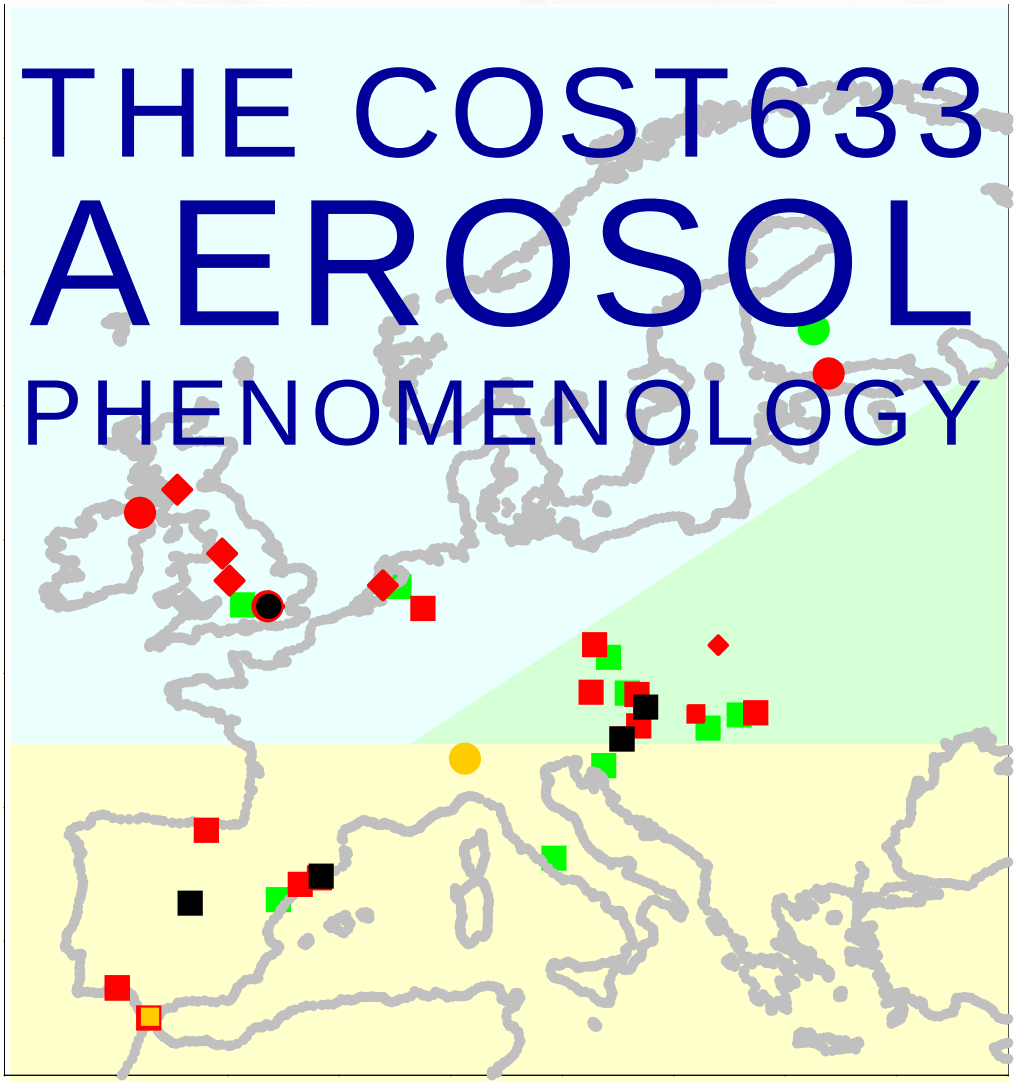
THE COST633 AEROSOL PHENOMENOLOGY



physical and chemical characteristics of particulate matter at rural, urban and kerbside sites compiled by the COST Action 633 “Particulate Matter: properties related to health effects”

EUR 23534 EN - 2008

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Summary

Atmospheric particulate pollution monitoring currently focuses almost exclusively on the mass concentrations of particles smaller than 10 µm (PM_{10}) and 2.5 µm ($PM_{2.5}$). However, particulate matter (PM) mass *per se* may not be the most relevant metric to assess the health risk related to atmospheric particles.

This report synthesizes the aerosol (=PM) physical and chemical data collected in the frame of the **COST Action 633** (**"Particulate Matter: Properties Related to Health Effects"**) for rural, near-city, urban, and kerbside sites. It covers a large portion of Europe, divided in Northwestern, Southern and Central sectors. It is complementing a European aerosol phenomenology already published several years ago (Van Dingenen et al. 2004; Putaud et al., 2004). The data sets we collected include PM_{10} and/or $PM_{2.5}$ mass concentrations, **and** aerosol particle number, **or** PM chemistry data. This compilation aims at highlighting similarities and differences in aerosol characteristics across the COST633 network, beyond PM mass concentrations that mainly reflect pollution dilution. It enables to work on the question "why do equal PM_{10} mass concentrations lead to different health effects across Europe", and contributes to identifying synergies and trade-offs between policies related to climate change mitigation and air pollution abatement.

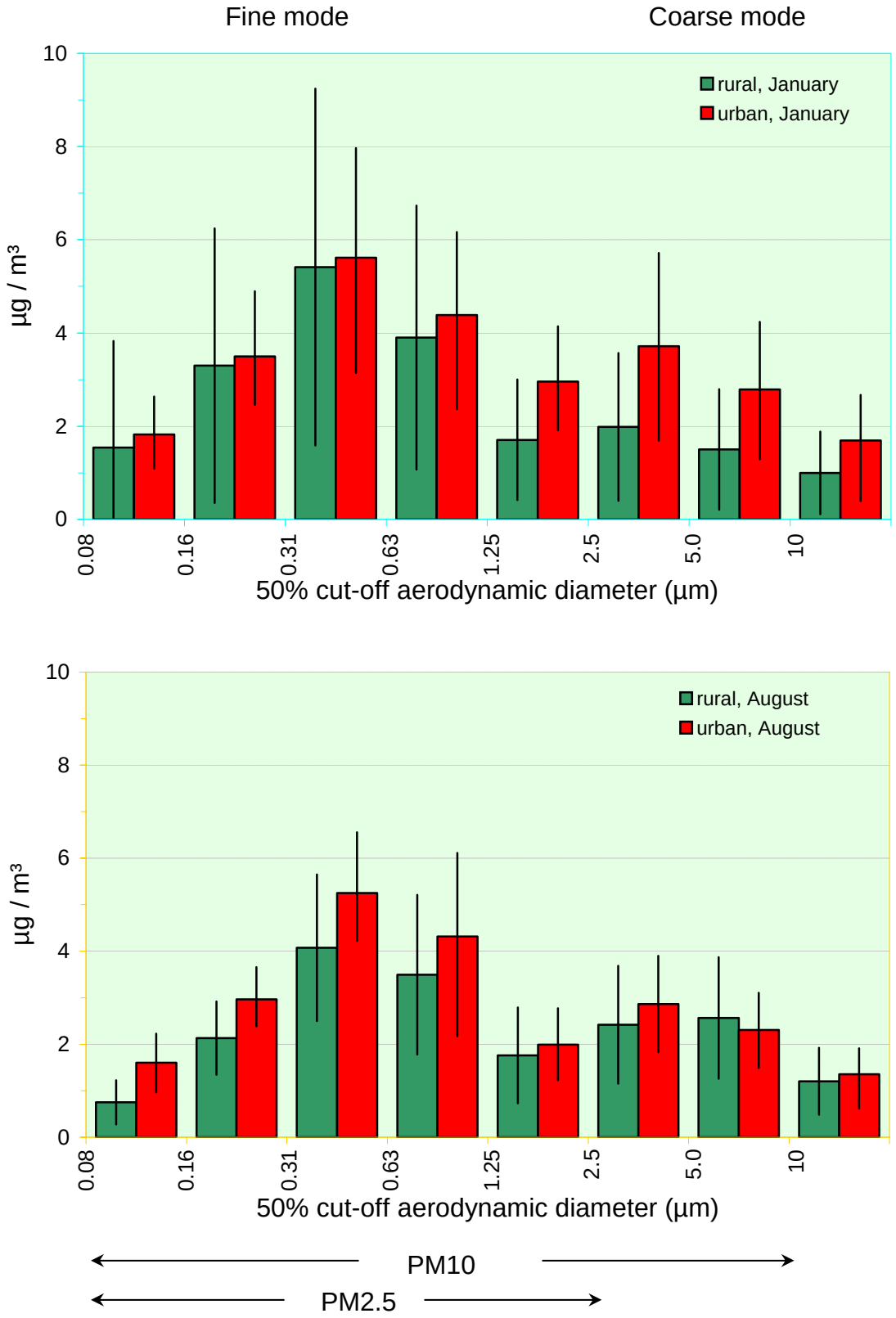
Data from 40 sites in Europe (called "the COST633 Network") were processed and sorted to allow comparisons. Absolute concentrations in PM_{10}, $PM_{2.5}$, particle number and major PM components are presented, but we focused on relative concentrations, to make data comparable in spite of differences in PM mass concentration influenced by pollution dispersion.

A number of conclusions are drawn among which the following:

- The COST633 network covers a wide range of pollution conditions, with sites where annual PM_{10} concentrations range from ≤ 20 µg m^{-3} to > 40 µg m^{-3}, the EU 2005 annual average PM_{10} standard.
- $PM_{2.5}$ and PM_{10} are well correlated at most (but not all) sites of the COST633 network. However, the $PM_{2.5}/PM_{10}$ ratio is site dependent (range 0.43 – 0.85).
- There is no simple correlation between PM mass and particle number concentrations: at a given site, particle number concentration does not increase monotonically with PM mass. Also, for a given PM mass range, particle number can vary by a factor of 2 across similar sites, within a factor of > 10 when all sites are considered.
- PM_{10} and $PM_{2.5}$ chemical compositions are season dependent at many sites. However, organic matter is on average the major constituent of PM_{10} and $PM_{2.5}$ at most site types, although nitrate, mineral dust and even sea salt can slightly take over in a few site categories. Clear gradients in PM_{10} chemical composition can be experienced when moving from Northwestern to Southern to Central Europe, and from rural to kerbside sites.
- There is no evidence that a specific aerosol constituent is systematically responsible for high PM concentrations. The fraction of PM that is not accounted for by the major PM components increases with PM mass.
- The largest Cu / PM_{10} and Pb / PM_{10} ratios are observed in Southern Europe, and Cu / PM_{10} ratios increase when moving from rural to kerbside sites.

Although more relevant data sets do certainly exist, that we did not manage to get into this compilation, the number of sites at which a complete characterization of the aerosol is available remains low. The development of a new approach, which would bridge the gap between atmospheric research and air pollution monitoring, is needed.

Fig 1


Fine mode
Coarse mode
rural, January
urban, January
µg / m³
0
2
4
6
8
10
0.08
0.16
0.31
0.63
1.25
2.5
5.0
10
50% cut-off aerodynamic diameter (µm)
rural, August
urban, August
µg / m³
0
2
4
6
8
10
0.08
0.16
0.31
0.63
1.25
2.5
5.0
10
50% cut-off aerodynamic diameter (µm)
PM10
PM2.5

1. Atmospheric Particles / Particulate Matter /Aerosol

Emitted directly as particles (primary aerosol) or formed in the atmosphere by (often photochemical) conversion of gaseous precursors (secondary aerosol), **atmospheric particles** range in size from a few nanometers (nm) to tens of micrometers (µm). A detailed description of the atmospheric processes leading to the formation of aerosols can be found in Raes et al., 2000. Particles smaller than 1 µm diameter generally have atmospheric concentrations in the range of 100 to 100.000's cm^{-3}, those exceeding 1 µm diameter typically exhibit concentrations smaller than 10 cm^{-3}. Atmospheric particles consist of natural constituents like soil dust, sea salt, molecules of biologic origin, and of anthropogenic species, mainly related to fossil fuel combustion and industrial emissions. Except in quite specific conditions (for instance vicinity of a source), an atmospheric particle contains constituents from various origins (internal mixing). It is therefore generally improper to talk about e.g. organic carbon (OC) particles, since particles containing OC do most often contain also other species, like e.g. elemental carbon (EC).
On the other hand, we use "particulate OC" to indicate the fraction of OC that is part of the atmospheric **particulate matter**, in contrast with the fraction of OC present in the gas phase. The suspension of particles in the air constitutes the atmospheric **aerosol**. The consideration of both phases is important because many of the relevant components are semi-volatile and exchange mass between the gas phase and the particle phase, like e.g. water, nitrate and a range of (often unspecified) organic compounds. However, aerosol is often used to indicate aerosol particles only.
PM*x* stands for the fraction of atmospheric particulate matter that consists of particles with an aerodynamic diameter lower than *x* µm. It is also used to indicate the mass concentration (µg / m³) of theses particles in the atmosphere. PM_{10} and $PM_{2.5}$ are the metrics currently used to quantify the level of particulate pollution in the air (see **BOX 1**).

Figure 1 illustrates which fractions of the suspended particulate matter (SPM) are accounted for by $PM_{2.5}$ and PM_{10} measurements. In this example derived from measurements performed at an urban site and a rural site in Austria (1999 – 2000), PM_{10} comprises most of the SPM mass. $PM_{2.5}$ includes the fine particle mode, but also a significant fraction of the coarse particle mode. In contrast, PM_1 contains the whole of the particles arising from combustion processes, but is negligibly influenced by natural sources as e.g. Saharan dust storms and sea spray.

European PM Directives

DIRECTIVE 2008/50/EC

EU standards for PM10

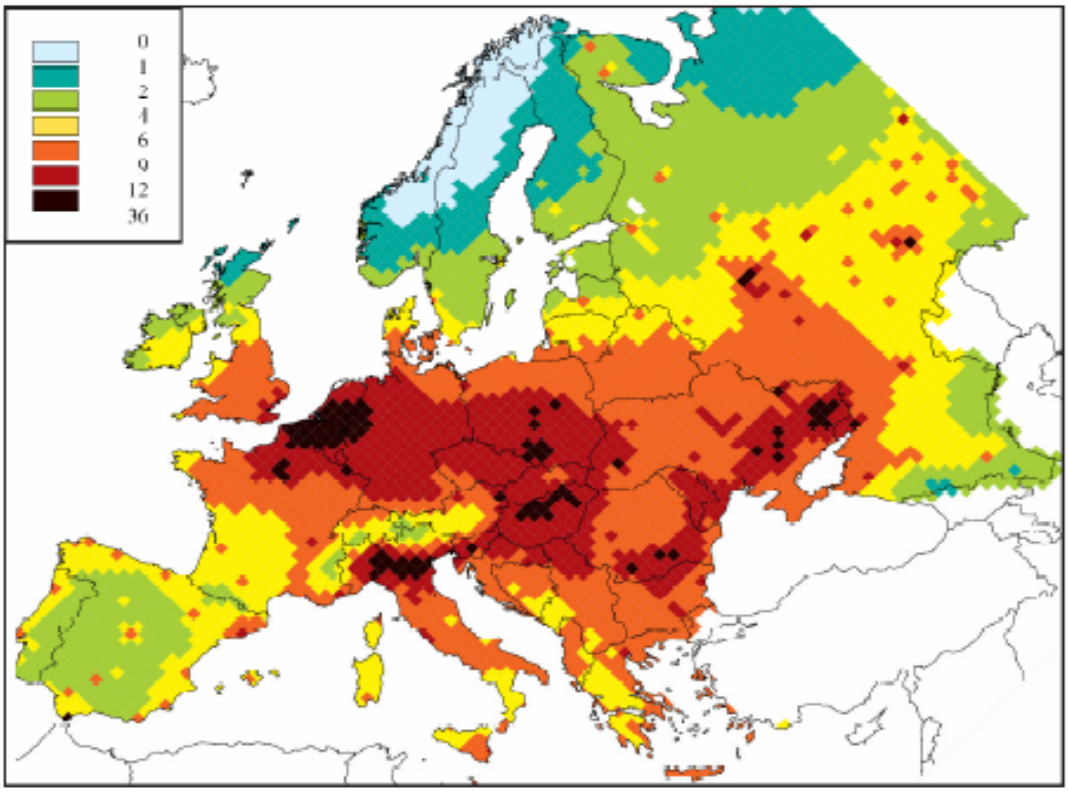
24-hour limit value	50 µg/m³ PM10 not to be exceeded > 35 times/year.	to be met 1 Jan 2005
annual standard	40 µg/m³ PM10	to be met 1 Jan 2005

EU standards for PM2.5

annual standard	25 µg/m³ PM2.5	to be met 1 Jan 2015
	20 µg/m³ PM2.5	to be met 1 Jan 2020
exposure	20 µg/m³ PM2.5 average exposure indicator	to be met in 2015 (3-yr average)

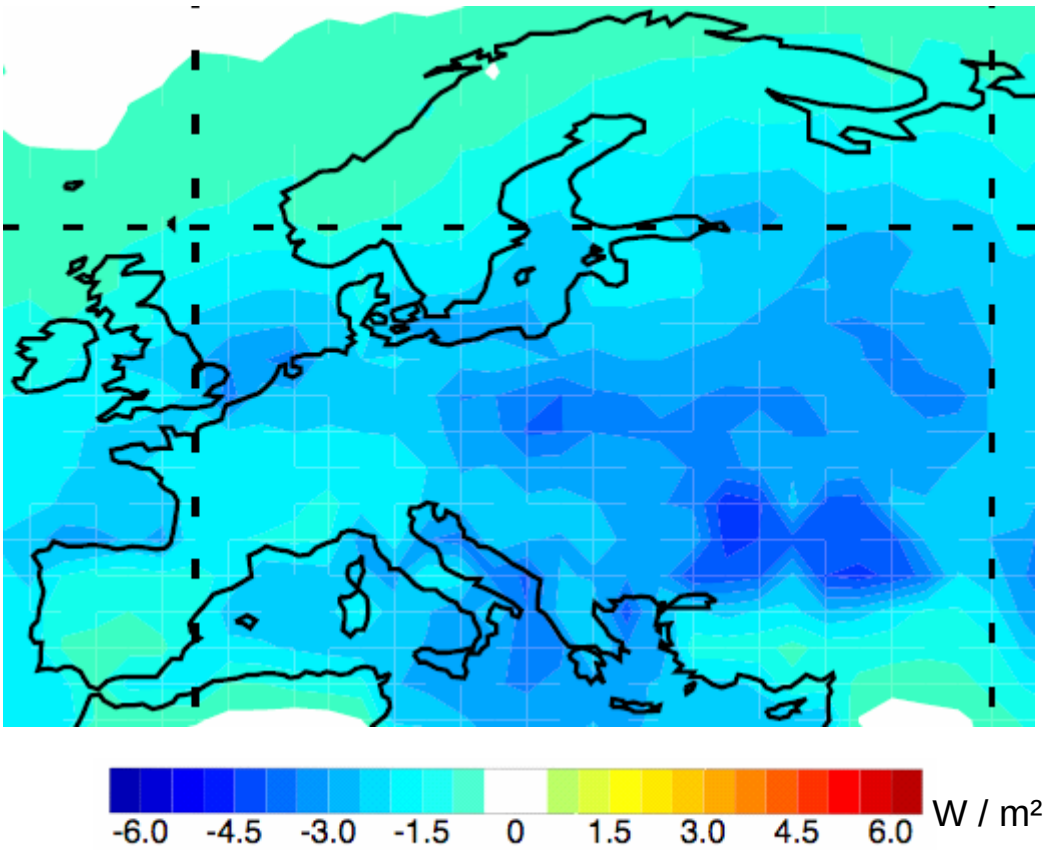
BOX 1

Fig 2



Loss in life expectancy (months) attributable to exposure to $PM_{2.5}$ – 2000 (Source: IIASA)

Fig 3



Direct radiative forcing (2000 -1750) due to tropospheric aerosols (Kloster et al., 2008)

2. Policy context

Currently, the interest in atmospheric particles 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 ecosystems, and contribute to the soiling of monuments.

PM and Health:

Many epidemiological studies showed positive correlations between PM mass concentrations and population health indicators, like the famous 6-city study by Dockery et al. (1993). Quantitatively, an increase in PM_{10} mass concentration by 10 µg m^{-3} would cause an increase of 0.5 -1.5% in premature total mortality in case of short term/episodic exposure, and an increase up to 5 % in premature total mortality in case of long term/life long exposure (Wilson and Spengler, 1996).

Based on such findings, Figure 2 maps the loss in statistical life expectancy that can be attributed to the identified anthropogenic contribution to $PM_{2.5}$. To reduce pollution to levels which minimize harmful effects on human health, EU Directives have also expressed limit values in terms of PM_{10} and $PM_{2.5}$ (BOX 1). As a consequence, monitoring efforts and pollution abatement measures mostly aim at controlling PM mass concentrations in the EU. Such abatement measures might be inefficient in reducing the health risks associated with atmospheric particles if they are linked to aerosol variables that do not correlate with PM mass. A monitoring strategy exclusively focused on PM mass would also prevent us from finding (a) better indicator(s) for measuring the health risk associated with PM. A series of other aerosol chemical (main constituents, tracers, toxic species) and physical (particle number and size distribution, hygroscopicity, volatility) characteristics have however been measured at a limited number of sites and / or for limited periods of time.

Aerosol and Climate:

Figure 3 shows the direct radiative forcing by aerosol particles in 2000. Although atmospheric particles may locally warm up the atmosphere, they globally lead to a cooling that may have partially balanced the warming due to the increase in greenhouse gases (GHG: CO_2, CH_4, N_2O, halocarbons) concentrations since the pre-industrial times (IPCC, 2007). Climate change mitigation policies like the promotion of wood burning and Diesel cars may have negative effects on particulate air pollution. On the other hand, PM abatement measures may have a negative effect on climate change if the dimming effect of aerosol particles decreases. However, synergies can be found between climate change mitigation and air pollution abatement policies. The simplest examples include battling against tropospheric ozone and EC aerosol, which are both warming agents and suspected to be related to health effects.

Aerosol Modelling is an indispensable tool for assessing the effect of PM on health and climate. Although different outputs, resolutions and scales are needed for addressing these 2 questions, numerical models will generally describe aerosol particle emissions and production, transport, transformation and removal to produce concentration fields that need testing and validation against fundamental data such as the aerosol particle size distribution and / or chemical composition.

Fig 4

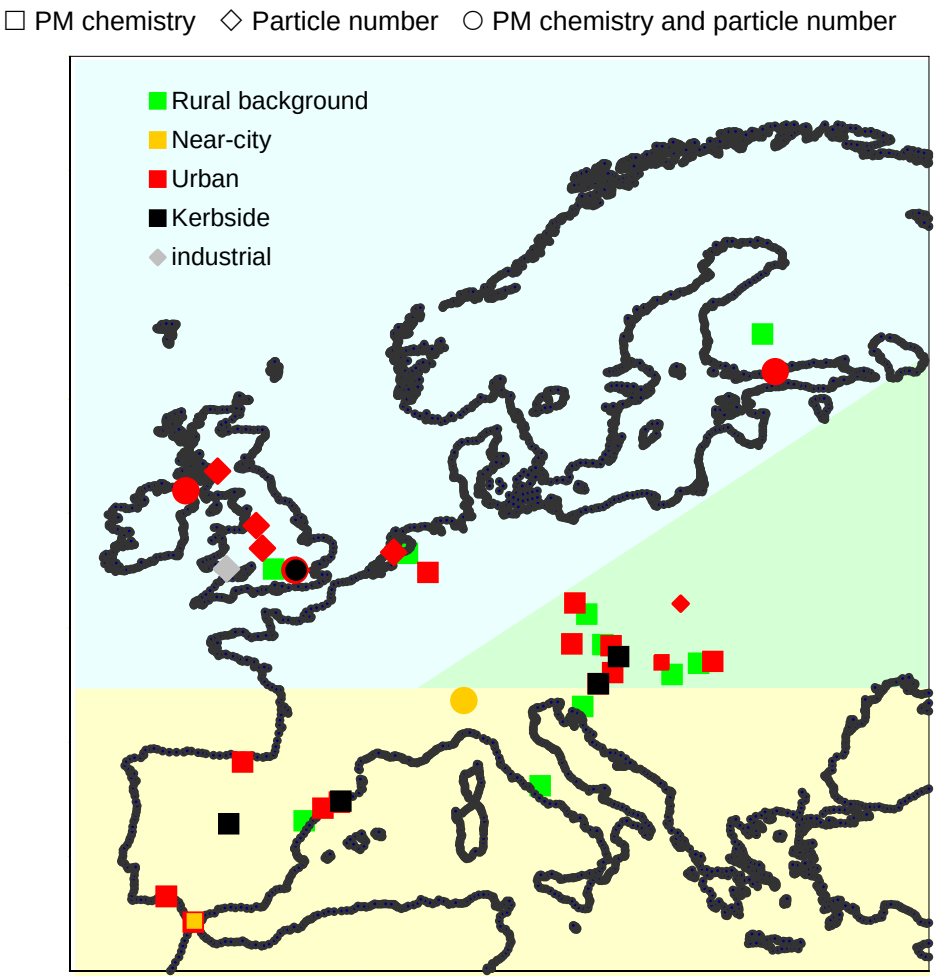
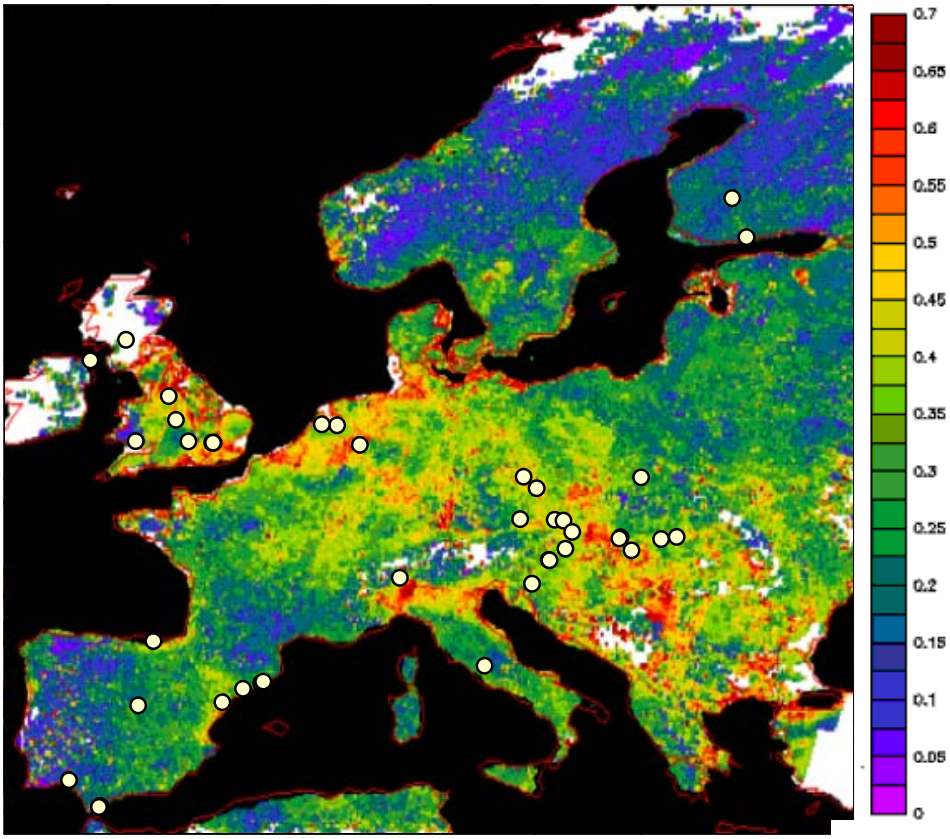


Fig 5



Aerosol Optical Depth at 555 nm, August 1997 (Source: TNO FEL)

3. Presentation of the results

The present aerosol phenomenology[1] synthesizes physical and size-resolved aerosol chemical data collected in the framework of the **COST Action 633** "Particulate Matter: Properties Related to Health Effects". Such variables are presently not often measured in regulatory monitoring networks (such as EMEP and AIRNET), but rather in research projects.
The goal of this work was to highlight similarities and differences in PM characteristics across Europe, beyond PM mass concentrations only. Therefore, we decided to select data sets including:

- PM_{10} and/or $PM_{2.5}$ mass concentrations
  and
- Size-segregated chemical composition.
  or
- Aerosol particle number concentrations or size distributions

This provides a minimum of consistency check and permits to normalize particle number or PM constituent concentrations to PM mass, thus highlighting characteristics that are independent of pollution dispersion, mainly controlled by meteorology and topography. We considered only measurements providing data representative for a site during at least 3 weeks during the same season. The methods used to measure these parameters and the uncertainties arising from comparing data produced by different institutes using different techniques are described in Annexes 1 and 2.

Figure 4 shows the location of the 40 sites, operated by 12 institutes, and classified in 3 large sectors: Northwestern, Southern and Central Europe. Their coordinates are given in Annex 3. We will refer to these sites as **"the COST633 Network".**

We also have categorised 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:

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	

We have added two categories:

Industrial	in the vicinity of large industrial sites
Kerbside	within street canyons

Figure 5 shows the sites projected on a satellite retrieved aerosol optical depth (AOD) picture for the month of August 1997 (Robles-Gonzales et al., 2000). Background AOD values are about a factor of 5 to 10 lower than in the most polluted areas. Fig. 5 shows that the COST633 Network covers a wide range of aerosol pollution conditions. However it lacks sites from France, and Germany (1 site only). Also, most particle number concentration data sets come from sites located in Northwestern Europe.

[1] Phenomenology: the branch of philosophy that deals with what you see, hear, feel, etc., in contrast to what may actually be real or true about the world

Fig 6a

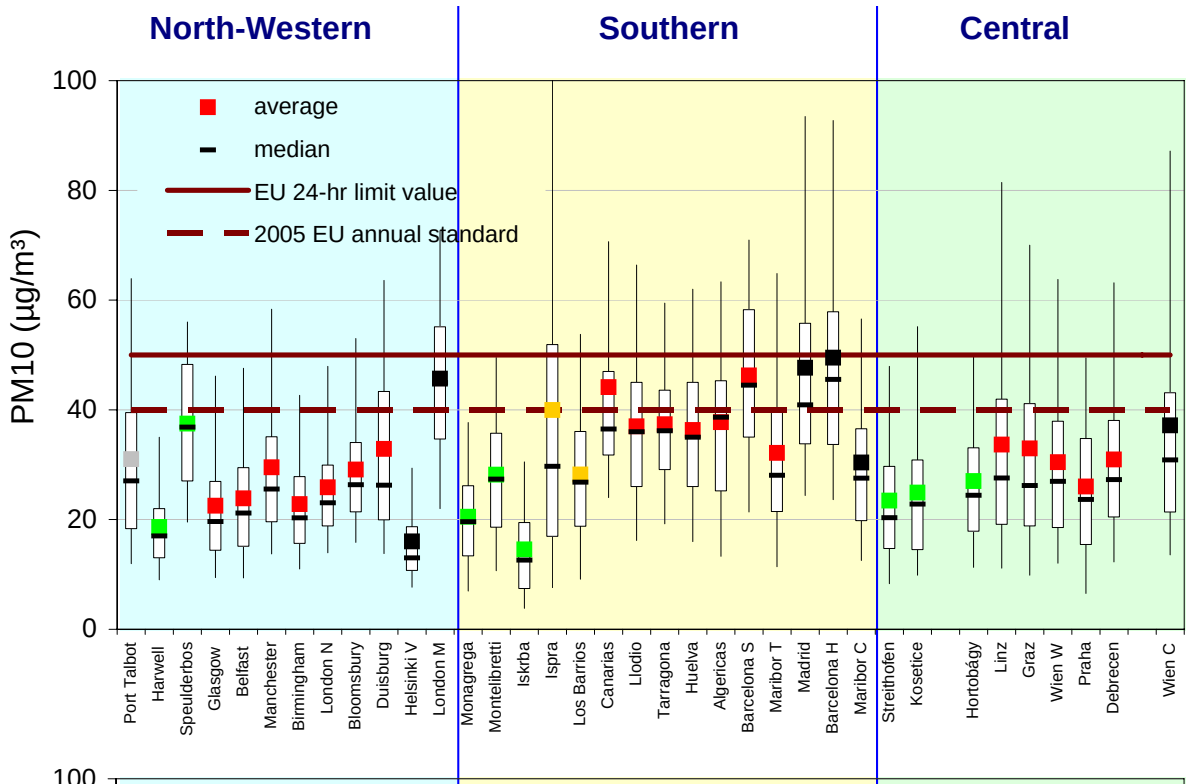
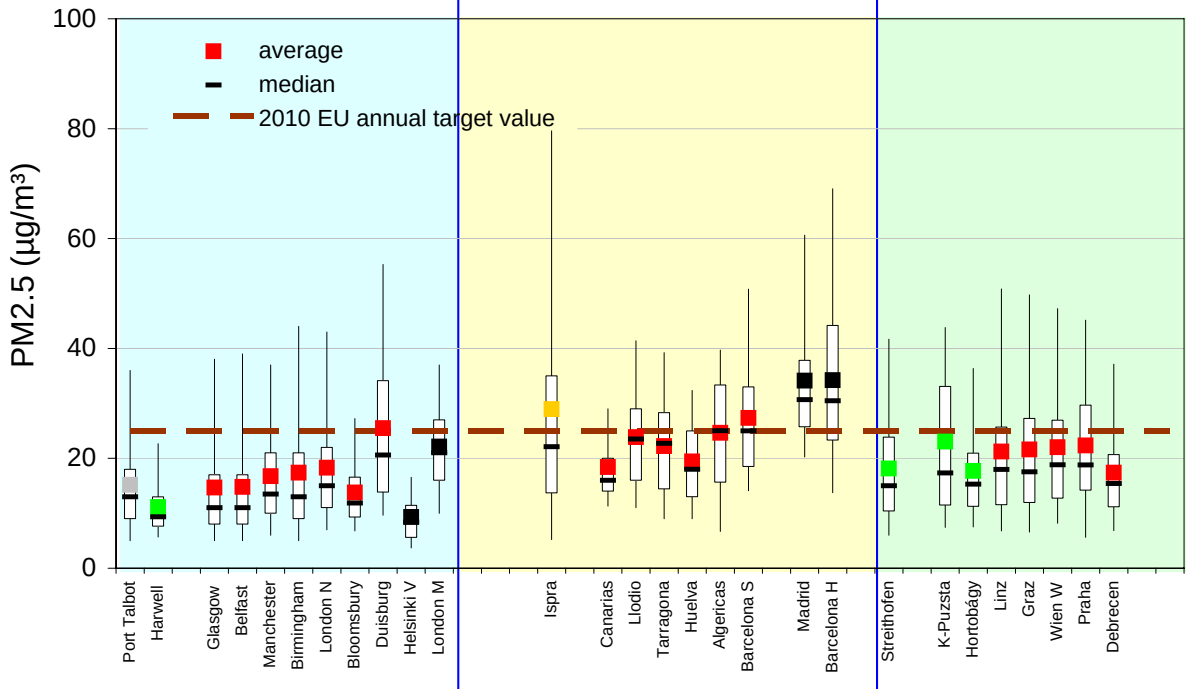


Fig 6b



industrial

rural background

near-city

urban background

kerbside

3.1 PM10 and PM2.5 mass concentrations

PM_{10} mass concentrations have been reported by national and international (EMEP, AIRNET) air monitoring networks for several years. $PM_{2.5}$ mass concentrations are still more rarely measured. Here we show yearly statistics regarding 24-hr averaged PM_{10} and $PM_{2.5}$ concentrations observed at 37 sites of the COST633 Network to put these data in a corrigendum broader context.

Figures 6a and 6b show the 5, 25, 50 (median) 75 and 95 % percentiles of 24-hr integrated PM_{10} and $PM_{2.5}$ mass concentrations, as well as their annual averages (colored symbol).

The EU annual PM_{10} standard of 40 µg m^{-3} (in force since 2005) is exceeded at 6 sites, and the EU annual $PM_{2.5}$ standard of 25 µg m^{-3} targeted for 2010 (to be met by 2015) at 5 sites. The 95 and 75 % PM_{10} percentiles indicate that the EU 24-hour PM_{10} limit value of 50 µg m^{-3} is exceeded more than 18 and 90 times a year, respectively, at most near-city, urban background and kerbside sites.

PM_{10} and $PM_{2.5}$ annual average concentrations both vary by a factor of 3-4 between their lowest (Iskrba, SI; Helsinki, FI) and highest values (Barcelona, ES). This variability is larger than that of annually averaged ozone concentrations, which in Europe ranged from about 40 to 100 µg m^{-3} in 2006 (EMEP/CCC 2/2008). This shows that within the COST633 Network, which comprises sites located in inhabited regions only, PM mass concentration is more sensitive to local sources than ozone. PM_{10} mean concentrations at urban sites are generally larger in Southern Europe compared to the other 2 regions. On the other hand, $PM_{2.5}$ average concentrations are generally smaller in Northwestern Europe. At rural sites, PM mass concentrations are on average quite similar in the 3 sectors. PM concentrations at kerbsides can be highly variable (compare e.g. Helsinki and London).

When sites are ordered according to the EEA criteria, PM_{10} and $PM_{2.5}$ annual average concentrations do not gradually (i.e. monotonically) increase in each region when moving from near-city background to urban background to kerbside sites. In fact, concentrations at kerbsides or urban sites in one city might be lower than at near–city or rural sites elsewhere in the same region. This shows that the regional aerosol background level can strongly influence PM concentrations in cities. However, the kerbside site in London (London M) shows higher PM than the urban background site (London B), which again shows higher PM concentration than a rural site upwind of London (Harwell). Same is observed in Spain (Barcelona H, Barcelona S and Monagrega), and Austria (Wien C, Wien W and Streithofen).

Fig 7

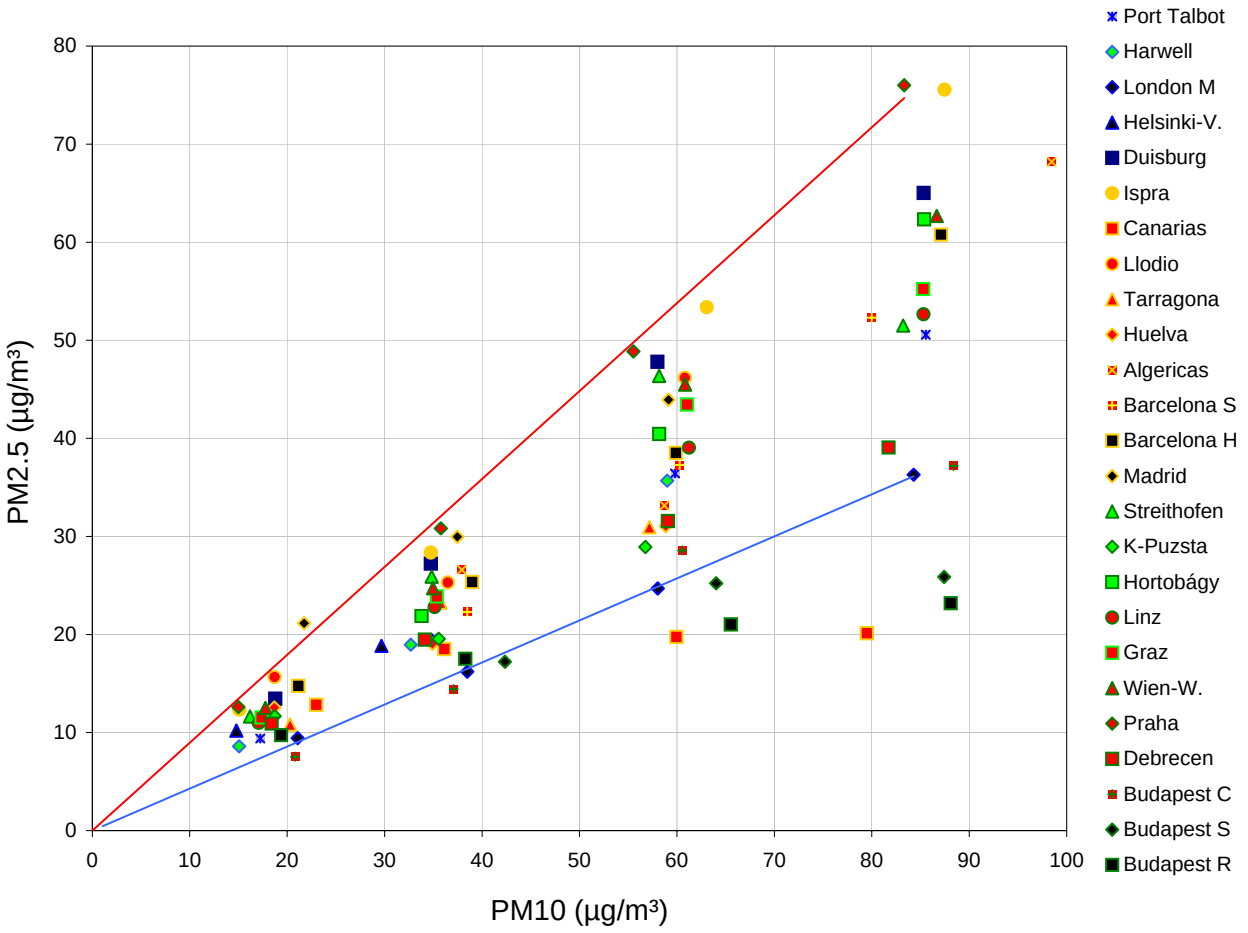

PM2.5 (µg/m³)
PM10 (µg/m³)
0
10
20
30
40
50
60
70
80
90
100
Port Talbot
Harwell
London M
Helsinki-V.
Duisburg
Ispra
Canarias
Llodio
Tarragona
Huelva
Algericas
Barcelona S
Barcelona H
Madrid
Streithofen
K-Puzsta
Hortobágy
Linz
Graz
Wien-W.
Praha
Debrecen
Budapest C
Budapest S
Budapest R

Fig 8

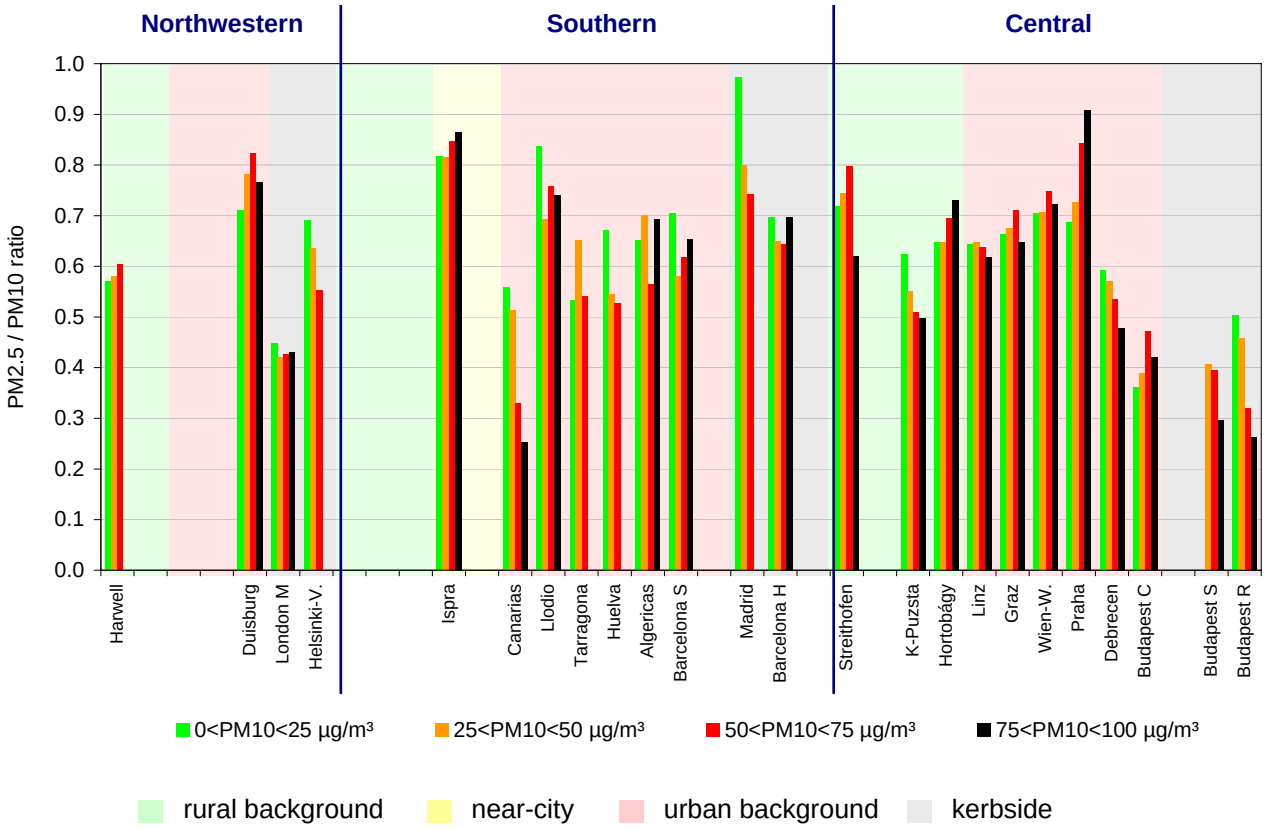

Northwestern
Southern
Central
PM2.5 / PM10 ratio
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
Harwell
Duisburg
London M
Helsinki-V.
Ispra
Canarias
Llodio
Tarragona
Huelva
Algericas
Barcelona S
Madrid
Barcelona H
Streithofen
K-Puzsta
Hortobágy
Linz
Graz
Wien-W.
Praha
Debrecen
Budapest C
Budapest S
Budapest R
0<PM10<25 µg/m³
25<PM10<50 µg/m³
50<PM10<75 µg/m³
75<PM10<100 µg/m³
rural background
near-city
urban background
kerbside

Figure 7 shows averages of simultaneously measured $PM_{2.5}$ vs. PM_{10} mass concentrations for 23 sites of the COST633 Network. For each site, data were averaged over 25 µg m^{-3}-wide PM_{10} bins (e.g. $25<PM_{10}<50$ µg m^{-3}). Foreground symbol colors refer to the 3 geographical sectors Northwestern, Southern and Central Europe. Symbol background colors refer to the different sites (■ rural, ■ industrial, ■ near-city, ■ urban, ■ kerbside). Fig. 7 illustrates the answer to the question: for a given value of PM_{10}, how much $PM_{2.5}$ should I expect ?

$PM_{2.5}$ and PM_{10} mass concentrations are clearly correlated with a mean $PM_{2.5}/PM_{10}$ ratio = 0.61±0.03 (95% confidence level). For a given site the $PM_{2.5}/PM_{10}$ ratio can be fairly constant. This is due to the fact that pollution dispersion (driven by meteorology) is the main factor controlling PM mass concentrations. It also suggests that the intensities of fine ($PM_{2.5}$) and coarse (PM_{10}-$PM_{2.5}$) aerosol sources covary. However, a closer look shows that $PM_{2.5}/PM_{10}$ ratios range from 0.43 to 0.90 (blue and red lines in Fig. 7). For a number of sites, $PM_{2.5}$ even levels off when PM_{10} increases above 50 µg m^{-3} (e.g. Canarias, Budapest S and Budapest R). At these sites, there is clearly a source contributing to high PM_{10} concentrations that does not contribute as much to $PM_{2.5}$ concentrations.

Thus, for a given PM_{10} concentration, a large range of $PM_{2.5}$ (factor ≥ 2) may be observed. This means that the different sources contributing to $PM_{2.5}$ and PM_{10}-$PM_{2.5}$ do not have the same strength everywhere. Lower ratios are observed in Canarias, frequently impacted by transport of Saharan dust, and at kerbside sites in Budapest, which suggests a large contribution of re-suspended road dust to the PM_{10}-$PM_{2.5}$ fraction. Higher ratios are observed at some rural (Streithofen, Hortobagy), near-city (Ispra), urban background (Duisburg, Llodio, Wien, Praha) and kerbside (Madrid) sites, where secondary aerosol production or car exhausts, which both produce fine particles, may be predominant.

The correlation between PM_{10} and PM_{coarse} (= $PM_{10} - PM_{2.5}$) is not as good (R^2 = 0.40). However, a significant correlation can be observed at a large number of sites (not shown).

Figure 8 shows $PM_{2.5}/PM_{10}$ ratios for the different sites, averaged over various bins of PM_{10} mass concentrations. At 3 sites (Harwell, Ispra, Hortobagy) located each in a different region of Europe, in non-urban areas, the higher the PM_{10} concentration, the higher the $PM_{2.5}/PM_{10}$ ratio, which indicates that PM pollution periods are predominantly due to increases in the $PM_{2.5}$ mass concentration. In contrast, $PM_{2.5}/PM_{10}$ decreases with increasing PM_{10} levels at 8 sites (Helsinki, Canarias, Huelva, Madrid, K-Puzsta, Debrecen, Budapest S and Budapest R) ranging from rural to kerbside sites, from all the 3 geographical sectors. At these sites, the highest PM_{10} concentrations are mainly due to large concentrations of coarse PM.

PM mass measurements

PM mass measurements carried out according to reference method EN 12341 suffer two main artifacts that prevent comparability of the values obtained at various sites:

1- Sampling artifacts that affect semi-volatile PM components are not addressed. As a consequence, up to 100% of the atmospheric particulate ammonium nitrate (NH_4NO_3) can be lost from filters during sampling. The collection of particulate organic carbon (OC) may also be affected by possibly large positive and negative artifacts. As NH_4NO_3 and OC contribute a large fraction of PM (Fig. 14), especially when PM gets close to limit values (Fig. 15), errors in sampling these two components may lead to large biases in PM measurements, depending on the concentrations of NH_4NO_3 and OC really occurring in the atmosphere.

2- At 50% relative humidity (recommendation for gravimetric measurements), the amount of water associated with the PM collected on a filter is still significant, specially if RH is *reduced* from ambient to 50% (upper branch of the hysteresis curve). The amount of water bound to PM depends on its hygroscopicity, resulting from its chemical composition. Sea salt, ammonium nitrate and sulfate are among the most hydrophilic PM components. Other species like mineral dust, OC and elemental carbon (EC) are less hydrophilic. Therefore, for a same amount of dry PM collected on a filter, gravimetric measurements at 50% RH will lead to larger values at sites where the fraction of sea salt, ammonium nitrate and sulfate (all PM components that do not look like particularly harmful) is large, compared to sites where PM is dominated by more hydrophobic species like dust, OC, and EC.

BOX 2

Fig 9a

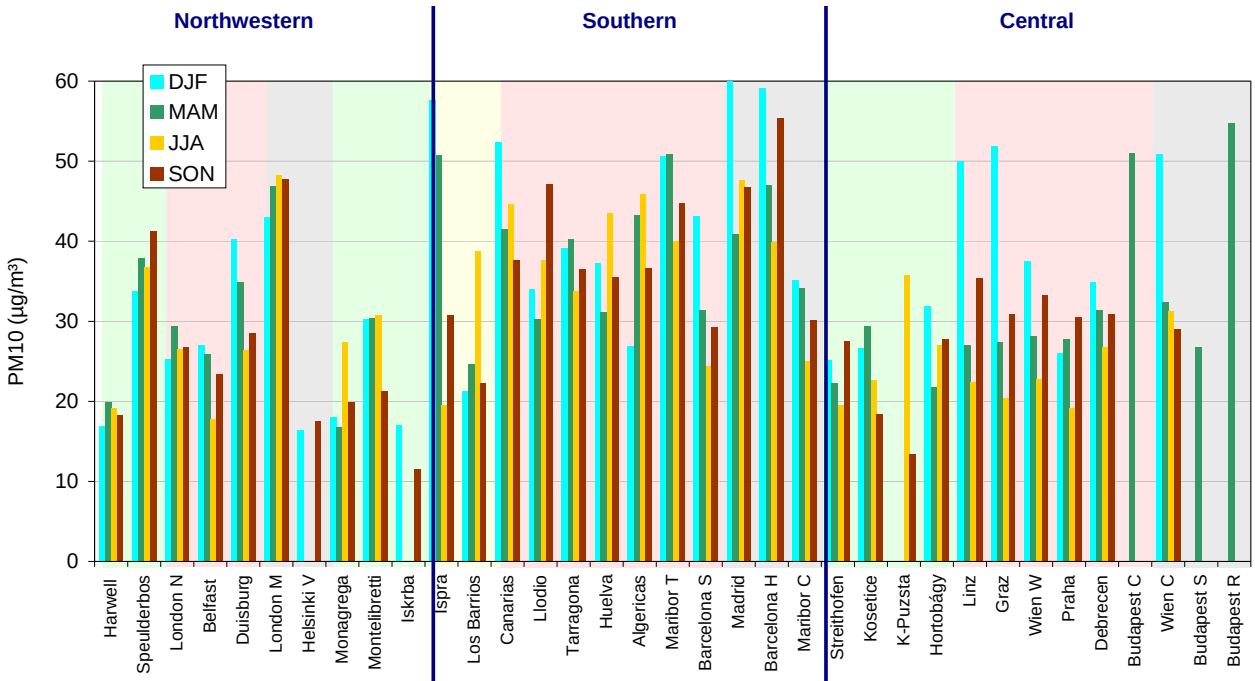
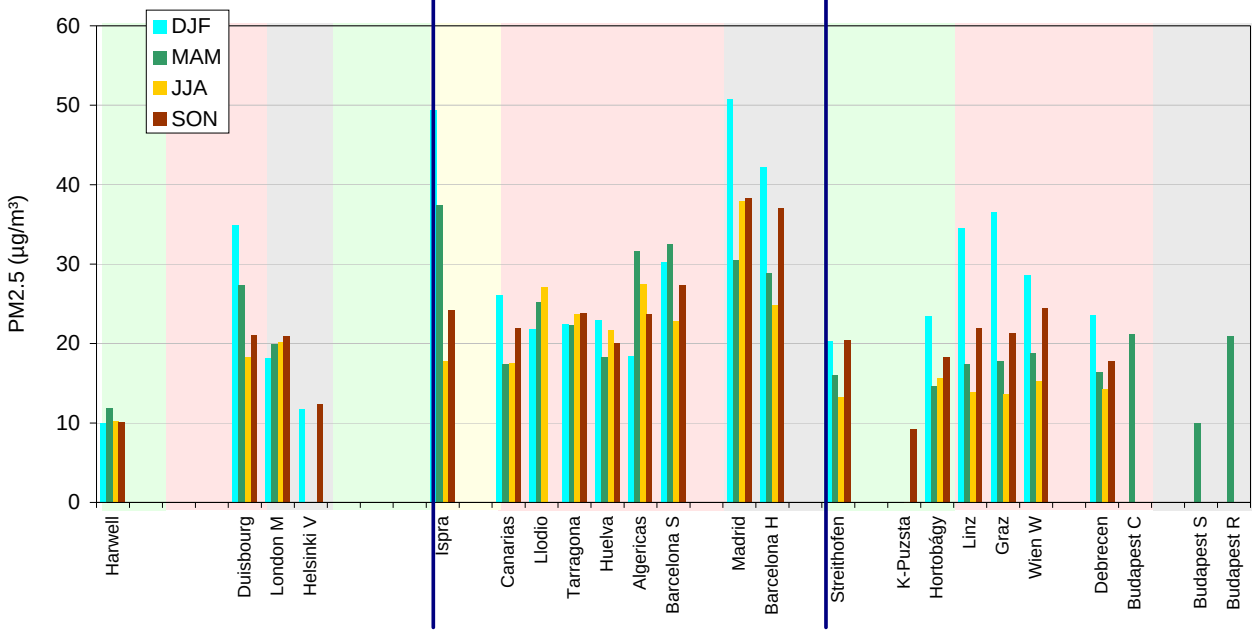


Fig 9b



rural background

near-city

urban background

kerbside

Figures 9a and **9b** show the seasonal averages of PM_{10} and $PM_{2.5}$ mass concentrations (■ December - January - February, ■ March - April - May, ■ June - July - August, ■ September - October - November).

PM_{10} mass concentrations present a seasonal cycle with a minimum in summer and/or a maximum in winter at 19 out of the 29 sites where this can be evaluated. When such a seasonal cycle is observed for PM_{10}, it is observed for $PM_{2.5}$ too. Sites where PM concentrations are minimum in summer range from near-city to kerbside sites, in all 3 geographical sectors. They also include 2 sites in central Europe. On the other hand, PM_{10} concentrations are maximum in summer at 4 sites, located in Southern Europe. These summertime maxima are not observed for $PM_{2.5}$, which suggests they are mainly due to coarse PM. There are also 4 sites were PM_{10} and $PM_{2.5}$ concentrations are rather constant along the year, all located in the Northwestern sector of Europe.

The occurrence and type of seasonal variations are generally related to the location rather than to the type of site. When similar seasonal variations are observed for PM_{10} and $PM_{2.5}$, this indicates that they are both driven by meteorology or that the variations in $PM_{2.5}$ control the variations in PM_{10} (in case $PM_{2.5}$ represents a large fraction of the PM_{10} mass). The large PM seasonal variations in e.g. Ispra (IT), Linz, Graz, and Wien (AT) are at least partly due to the larger stratification of the atmosphere (lower mixing heights) in more continental regions during winter. However, where seasonal variations are larger in urban sites compared to nearby rural sites (see e.g. Wien vs. Streithofen, AT), this indicates that additional PM sources, like residential heating, may contribute significantly to PM mass concentrations in cities in winter. Further comparisons of urban and rural sites in Austria are given in Puxbaum et al. (2004) and Gomiscek et al. (2004).

PM mass concentrations as indicators

Although it is quite unlikely that the *mass* of PM inhaled *directly* leads to health effects, ambient PM mass concentrations is currently the only metric subjected to European directives. In a next future, exposure to (still) PM mass concentrations will also be considered.

- Are PM_{10} and $PM_{2.5}$ good indicators ?

A good indicator has a direct link to a PM characteristic that leads to health effects. PM_1 would be better than $PM_{2.5}$ since it represents more accurately the fine PM mode. Super-µm particles could also be linked to different types of health hazards. However, ultra-fine particles (<100 nm) are very numerous and do not contribute much to PM mass, although they are suspected to have a stronger and/or other health impacts. Focusing monitoring and abatement strategies on PM mass only might be inefficient for tackling the issue of PM health effects.

- Are there better indicators ?

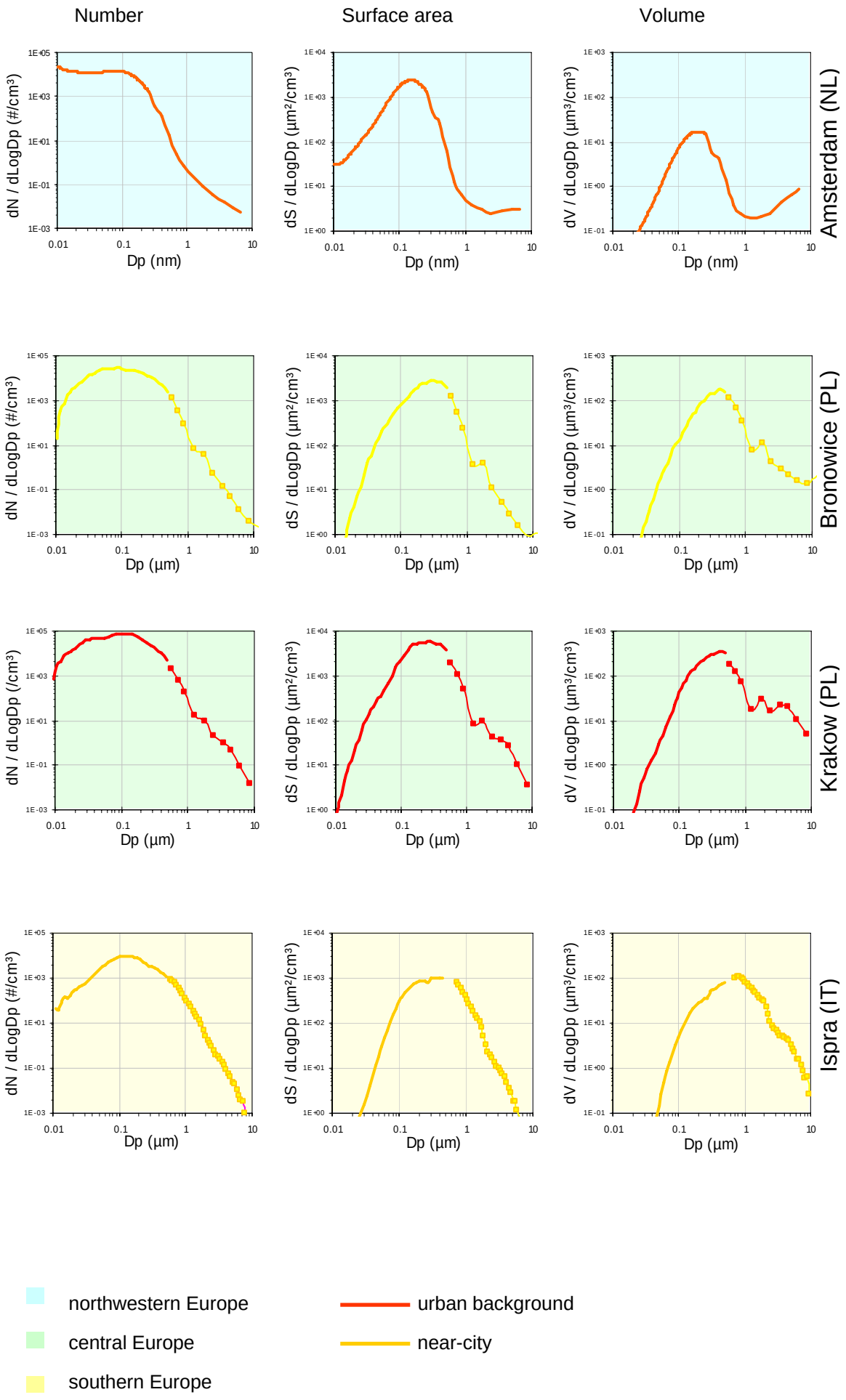
Black Smoke or elemental carbon, total or refractive particle number concentrations, and a surface reactivity or an oxidative stress index are valuable candidates because they are more specific than PM mass. These variables may be related to specific types of health impacts, but there is probably single indicator that can account for all types of health impacts (short term, long term effects, ...) caused by PM. It is indeed expected that a set of several variables are needed to describe the health hazard of PM. The scientific community lacks data regarding these alternative variables to confront population health status data with.

- Are we able to monitor these alternative indicators?

Robust methods have been developed in related fields like aerosol climate impact and toxicology research. They are currently adapted to cope with monitoring needs and might be proposed as European standardized methods in a near future.

BOX 3

Fig 10


Number
Surface area
Volume
Amsterdam (NL)
Bronowice (PL)
Krakow (PL)
Ispra (IT)
dN / dLogDp (#/cm³)
dN / dLogDp (/cm³)
dS / dLogDp (µm²/cm³)
dV / dLogDp (µm³/cm³)
Dp (nm)
Dp (µm)
northwestern Europe
central Europe
southern Europe
urban background
near-city

3.2 Particle size distributions and concentrations

We collected well resolved aerosol particle number size distributions from 4 sites of the COST633 Network only. This is too little to draw any conclusion regarding possible differences in particle size distribution in different regions of Europe, but enough to illustrate how particles of various sizes contribute to different metrics.

Figure 10 shows examples of particle size distributions obtained at 4 sites of the COST633 Network for wintertime conditions (end of January – beginning of February). Particle number size distributions were measured in dry conditions (RH <20%), using a combination of 2 instruments (see Annex 2). Particle surface area and volume (≈ proportional to mass) size distributions are calculated from number size distributions assuming spherical particles.

In these 4 examples, ultrafine particles (Dp < 100 nm) account for 30 – 80 % of the total particle number, but for only ≤ 2 to 10 % (upper limit in Amsterdam) of the particle volume below 10 µm. Assuming that ultrafine particles are spherical, they would account for 3 – 30 % of the total particle surface area only. The meaning of this metric is however questionable (Box 4). The contribution of super-µm particles to particle volume (below 10 µm) is generally low (8 – 19 %), and their contribution to particle number negligible (< 0.1 %).

As particle number and mass concentrations are mainly due to particles around Dp = 0.1 µm and 0.3 - 0.6 µm, respectively, these variables are governed by different processes, with different time scales. This implies that in general no straight correlation can be expected between particle number and volume (or $PM_{2.5}$, PM_{10}) concentrations, unless both are controlled by strong direct emission or determining meteorological processes (e.g. dilution, wet scavenging).

Number or surface area as indicators ?

The surface toxicity is currently thought to be a major cause of the health effects specifically induced by ultrafine particles. Although surface toxicity can only be determined by in vitro and in vivo studies, the aerosol surface area can be derived from a range of atmospheric measurements.

First, from the particle number size distribution measurements, particle surface area size distributions can be calculated assuming spherical particles. This may be adequate for addressing the effect of aerosol on visibility or radiative forcing. Indeed, most long-lived atmospheric particles result from an internal mixing and contain hydrophilic species that may eventually cover their possibly not spherical core with a spherical shell. However, this soluble shell will probably dissolve in lungs, to leave the bare core. It is possible to measure the number and size of the refractory cores, using the fact that the soluble shell can be volatilized. Still, the dimension measured with a differential mobility analyser (DMA) is the mobility diameter. Calculating the surface area of the refractory core assuming a spherical shape will lead to a value that can well be much different from its real surface area, if its actual structure is much more complex.

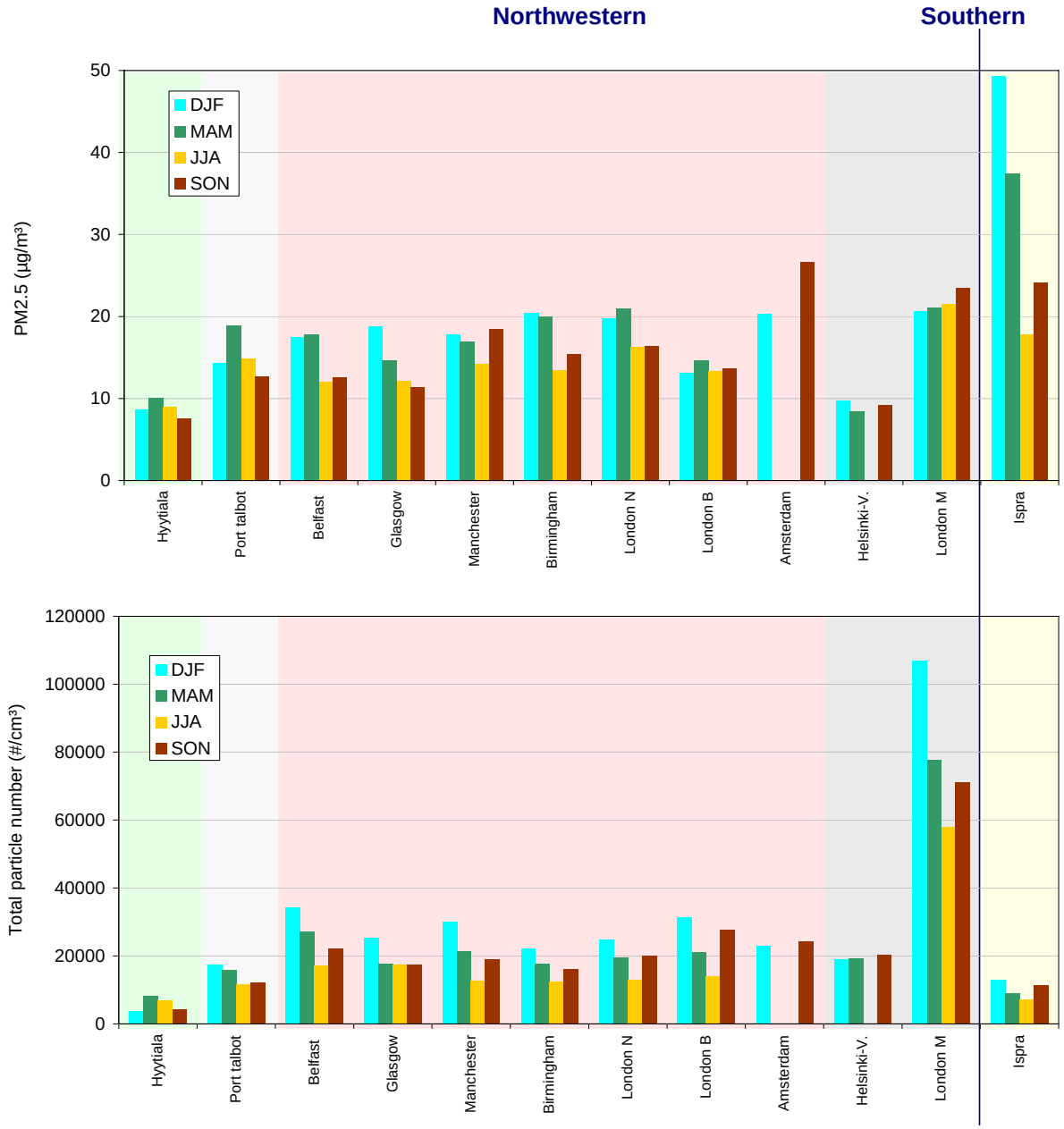
Almost 20 years ago, the Epiphaniometer was proposed as a device for measuring the aerosol surface area based on its gas adsorption capacity. It could have been used to determine the surface area of the refractory cores of the particles, after submitting the aerosol to an appropriate statement. However, the Epiphaniometer was never widely used by the community. It is perhaps because until now, the aerosol active surface area has never been demonstrated to be a proper metric for atmospheric particle risk assessment.

It remains that monitoring the number size distribution or concentration of refractory particles or refractory particle cores may be more relevant than measuring the size distribution or concentration of the whole particle population, that can include, specially during nucleation events, a huge number of completely soluble particles, of which health risk potential may be different.

BOX 4

Fig 11a

Fig 11b



Rural background

Industrial

Urban background

Kerbside

Near-city

Figure 11a shows the seasonal average of $PM_{2.5}$ mass concentrations, for the 12 sites of the COST633 network where both $PM_{2.5}$ and particle number concentrations measurements were available simultaneously. These sites are different from those plotted in Fig. 9b, where we showed $PM_{2.5}$ where PM_{10} data are available too. They are all located in the Northwestern region of Europe but one (Ispra). $PM_{2.5}$ concentrations are very low at the kerbside of Helsinki and very large in winter and spring at the near-city site of Ispra, located in Southern Europe though. In contrast, we can observe that there are no large differences in $PM_{2.5}$ mass concentrations between the industrial site, urban background sites, and the kerbside site in London. Among the 12 sites in Fig. 11, the annual average $PM_{2.5}$ concentration varies by a factor of 3.5 between its minimum (Hyytiälä, FI) and maximum value (Ispra, IT). No large seasonal variations in $PM_{2.5}$ can be observed, except in Ispra.

Figure 11b shows the seasonal average of particle (or condensation nucleus = CN) number concentrations, for the same 12 sites as in Fig. 11a. The annual average particle number concentration varies much more than $PM_{2.5}$ among the same sites (factor of 14 vs. 3.5). This is explained by the fact that a large number of very small particles do not necessarily lead to a large mass concentration (compare e.g. Hyytiälä, rural site, and London, kerbside site). It may be noticed that the near-city site of Ispra (IT) rather resembles an urban or even a kerbside site from the PM mass point of view, but rather a rural site in Finland from the particle number point of view.

The seasonal variations in $PM_{2.5}$ mass and particle number concentrations are generally not correlated to each other, which means that particle sizes do change across the year. In Hyytiälä (FI), particles are obviously bigger in winter and autumn compared to spring and summer. Roughly the same applies to Ispra. The fact that particle size distributions are shifted towards larger diameter during cold seasons may be due to the enhanced condensation of semi-volatile species. In contrast, the large seasonal variations in particle number observed in London M are not reflected in $PM_{2.5}$ mass concentrations, which means particles are relatively smaller in winter and spring compared to summer and autumn. This might be explained by a massive formation of secondary aerosol during summer, which would condense onto pre-existing particles. The different seasonal changes in particle number / $PM_{2.5}$ mass ratios observed at various urban sites may result from a combination of both processes.

Fig 12a

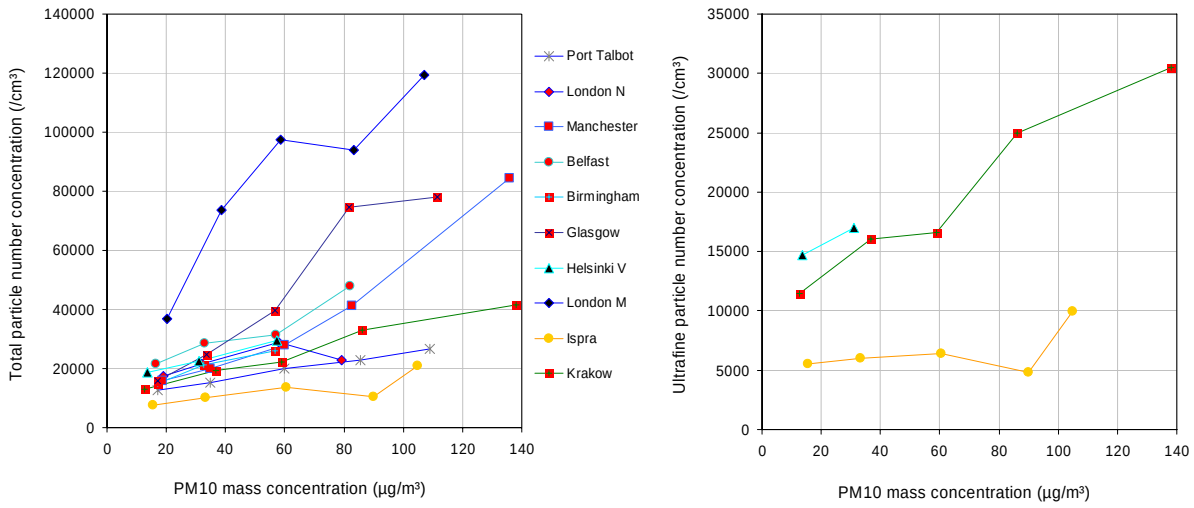


Fig 12b

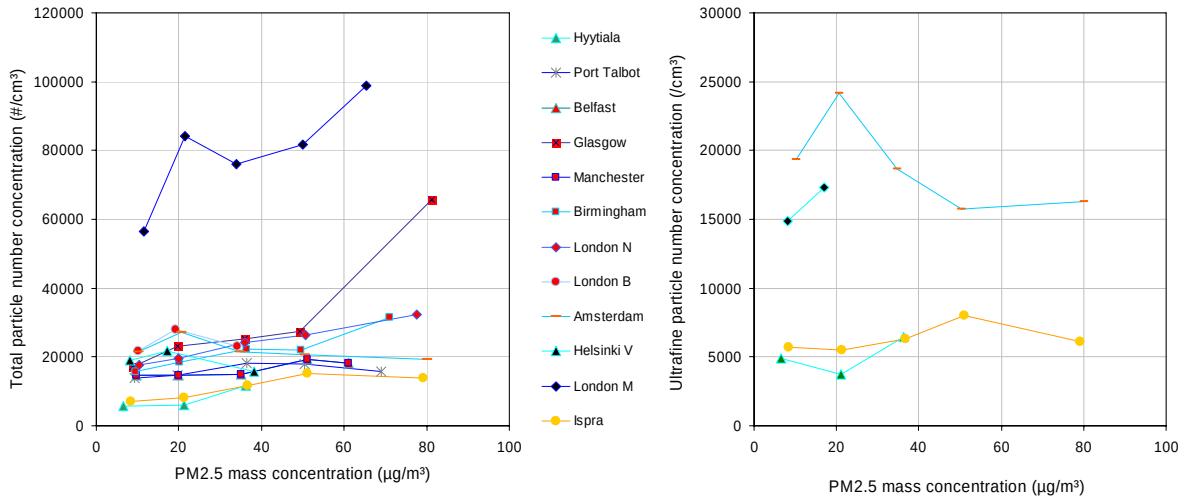


Table 1

Ultrafine / total particle number ratios for various ranges of **PM_{10}** concentrations (µg m^{-3}).

	PM10	[0-25]	[25-50]	[50-75]	[75-100]	[100-+]
Helsinki V	urban	0.78	0.75			
Ispra	near city	0.72	0.59	0.46	0.45	0.47
Krakow	urban	0.88	0.83	0.75	0.76	0.74

Table 2

Ultrafine / total particle number ratios for various ranges of **$PM_{2.5}$** concentrations (µg m^{-3}).

	PM2.5	[0-15]	[15-30]	[30-45]	[45-60]	[60-+]
Hyytiala	rural	0.85	0.62			
Amsterdam	urban	0.90	0.89	0.87	0.76	0.84
Helsinki V	kerbside	0.78	0.80			
Ispra	near city	0.80	0.67	0.54	0.52	0.44

Figure 12a and 12b aim at illustrating the answer to the question: how variable can particle number concentrations be for given PM_{10} and/or $PM_{2.5}$ mass concentrations? Line and foreground symbol colors refer to the 3 geographical sectors Northwestern, Southern and Central Europe. Symbol background colors refer to the different sites (■ rural, ■ industrial, ■ near-city, ■ urban, ■ kerbside).

Numerical values are provides in Annex 4.

At most but not all sites of the COST633 Network, particle number concentration increases with PM_{10} concentration, which suggests that pollution dispersion, resulting mainly from meteorology, control both variables. As already pointed out previously, the difference in particle number concentrations between a near-city and a kerbside site can be huge, for instance a factor of 7 for PM_{10} concentrations ranging between 50 and 75 µg m^{-3}. But the two sites considered here (Ispra, IT, and London, UK) are not located in the same region. More rigorously, among a range of urban sites all located in Northwestern Europe, particle number concentrations are still highly variable for a given PM_{10} bin, ranging for example from 20000 cm^{-3} to 40000 cm^{-3} (factor of 2) for 50 <PM_{10}< 75 µg m^{-3}. Particle numbers at both the industrial site in Northern Europe and the rural site in Central Europe are close to the lower values observed for each PM_{10} bin.

The number of sites where both PM_{10} and ultrafine particle number concentrations were measured is too limited to draw any robust conclusions, but it seems that for a given PM_{10} concentration, the difference in ultrafine particle number concentration can be even larger than for the total particle number concentration. Table 1 indicates that the ratio ultrafine / total particle number decreases with increasing PM_{10} at the 3 sites of the COST633 Network for which this could be looked at.

Particle number increases monotonically with $PM_{2.5}$ concentration at even fewer sites than with PM_{10}, which we could not explain so far. The difference in particle number for a given $PM_{2.5}$ mass range can be as large as a factor of 14 between Hyttiälä (rural site, FI) and London (kerbside site, UK) for 15 <$PM_{2.5}$< 30 µg m^{-3}. Among the urban sites located in Northwestern Europe, particle number concentrations generally vary within a factor of 1.4 – 1.9, and even more for $PM_{2.5}$ > 60 µg m^{-3}.

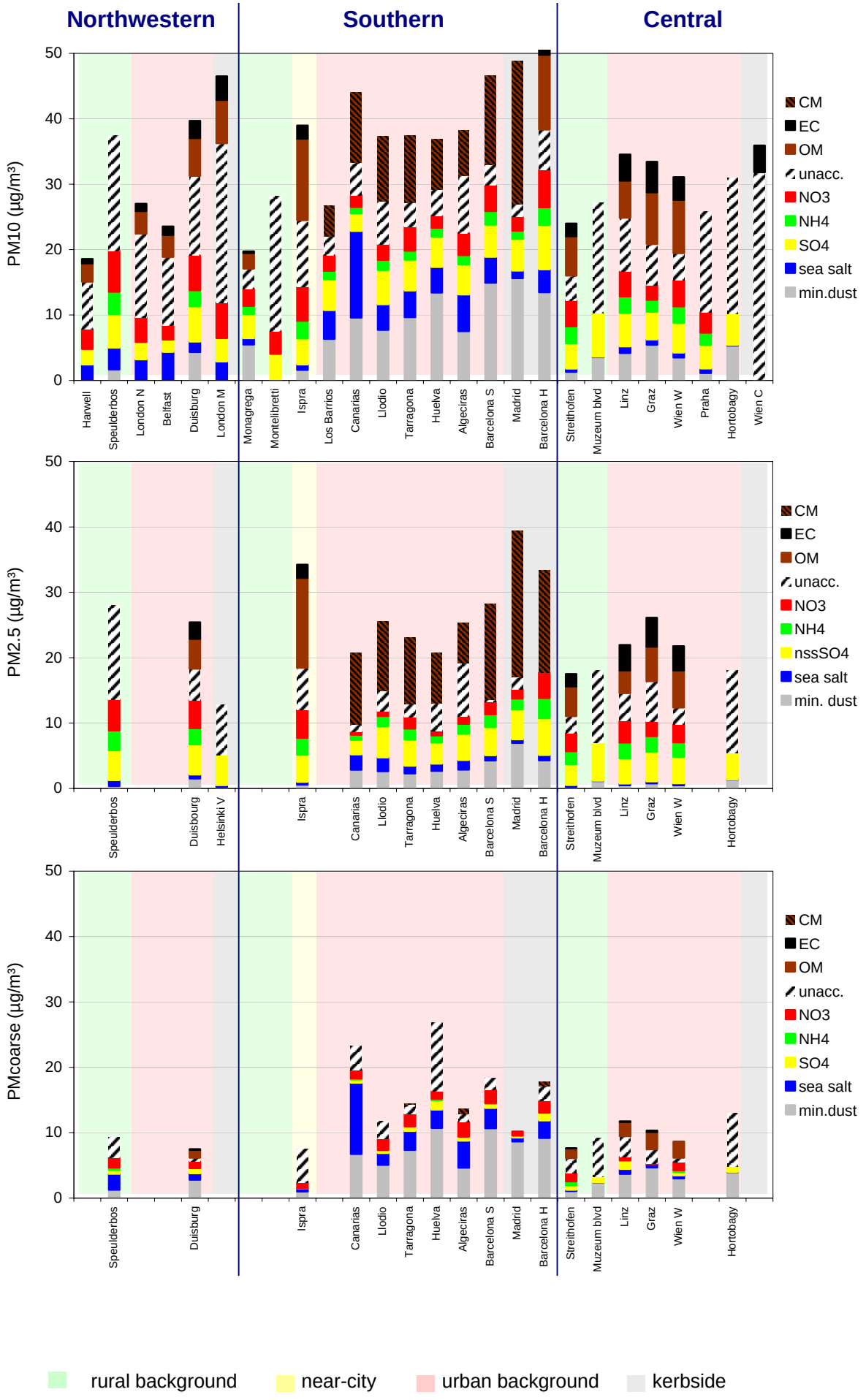
Also for a given $PM_{2.5}$ concentration, the difference in ultrafine particle number can be even larger than the difference in total particle number. Table 2 indicates that the ratio ultrafine / total particle number also decreases with increasing $PM_{2.5}$ at 3 sites of the COST633 Network. Nothing can be said about the kerbside site in Helsinki, because the range of $PM_{2.5}$ observed is too narrow.

At the polluted sites the relationship between $PM_{2.5}$ and the number of particles with Dp > 10 nm, seems to differ for near-city background, urban background and kerbside sites, and no single relationship between $PM_{2.5}$ and number (Dp > 10nm) can be proposed. In general the particle number (Dp > 10 nm) increases more than proportionally to $PM_{2.5}$ mass.

Fig 13a

Fig 13b

Fig 13c



3.3 PM_{10} and $PM_{2.5}$ chemical composition

We report on aerosol chemistry data that were obtained simultaneously with PM_{10} or $PM_{2.5}$ mass concentrations. This allows us to derive the PM chemical composition at the various sites of the COST633 Network, independently of PM concentrations that are mainly governed by pollution dilution.

Figure 13 shows absolute annual average concentrations of the main aerosol components in (a) PM_{10}, (b) $PM_{2.5}$, the "fine" fraction, and (c) PM_{10}-$PM_{2.5}$, the "coarse" fraction. Numerical values are given in Annex 5 and 6.

Elemental carbon (EC), nitrate (NO_3^-), sulfate (SO_4^{2-}) and ammonium (NH_4^+) concentrations result directly from measurements. Organic matter concentrations (OM) were calculated by multiplying organic carbon (OC) concentrations by a factor of 1.4 (e.g. Eatough et al., 1996) to account for the non-C atoms contained in particulate OM. Carbonaceous matter (CM) was calculated for sites where only total carbon (TC) measurements were available, assuming a constant EC/TC ratio = 0.1, and a conversion factor from OC to OM = 1.4. Sea-spray was clearly identified as a component of the aerosol at sites located < 50 km from seashore. For consistency, sea salt concentrations were calculated from Na^+, Cl^- concentrations and a standard sea water composition at other sites too, even when the purely marine origin of Na^+ was not obvious. The total amount of mineral dust (min. dust), which includes insoluble and refractory species (e.g. silicates, carbonates, calcium sulfate and trace elements,…) was estimated based on measurements of tracers such as Al or non-sea-salt Ca^{2+} (see Annex 1).

Full chemical characterisations of atmospheric PM are still rarely achieved due to the large range of methods needed to quantify its various components. Consequently, the unaccounted mass (obtained as the difference between the gravimetrically measured aerosol mass concentration and the sum of the aerosol component concentrations) can include different species for the various sites of the COST633 network. Table 3 specifies what components make up the unaccounted mass in PM_{10} for each site. When all main aerosol components are analyzed (i.e. at 8 sites only), the remaining unaccounted mass fraction (unknown) is thought to result from residual water associated with the aerosol at 50% RH (see Box 3), or non-C atoms associated with particulate organic matter which are not adequately accounted for by using a molecular mass - to - carbon mass ratio of 1.4 (see Annex 1), or analytical errors.

unk = unknowm, ss = sea salt, md = mineral dust

Sites	Components making up the unaccounted mass	Sites	Components making up the unaccounted mass
		Steithofen (AT)	unk
Harwell (UK)	unk NH_4^+ md	Hortobagy (HU)	EC OM unk NO_3^- NH_4^+
Speulderbos (NL)	EC OM unk	Linz (AT)	unk
London N (UK)	unk NH_4^+ md	Graz (AT)	unk
Belfast (UK)	unk NH_4^+ md	Wien W (AT)	unk
Duisburg (DE)	unk	Praha (CZ)	EC OM unk
London M (UK)	unk NH_4^+ md	Debrecen (HU)	EC OM unk NO_3^- NH_4^+
		Wien C (AT)	OM unk NO_3^- NH_4^+ SO_4^{2-} ss md
Monagrega (ES)	unk	Tarragona (ES)	unk
Montelibretti (IT)	EC OM unk SO_4^{2-} ss md	Huelva (ES)	unk
Ispra (IT)	unk	Algeciras (ES)	unk
Los Barrios (ES)	unk	Barcelona S (ES)	unk
Canarias (ES)	unk	Madrid (ES)	unk
Llodio (ES)	unk	Barcelona H (ES)	unk

Table 3

Fig 14a

Fig 14b

Fig 14c

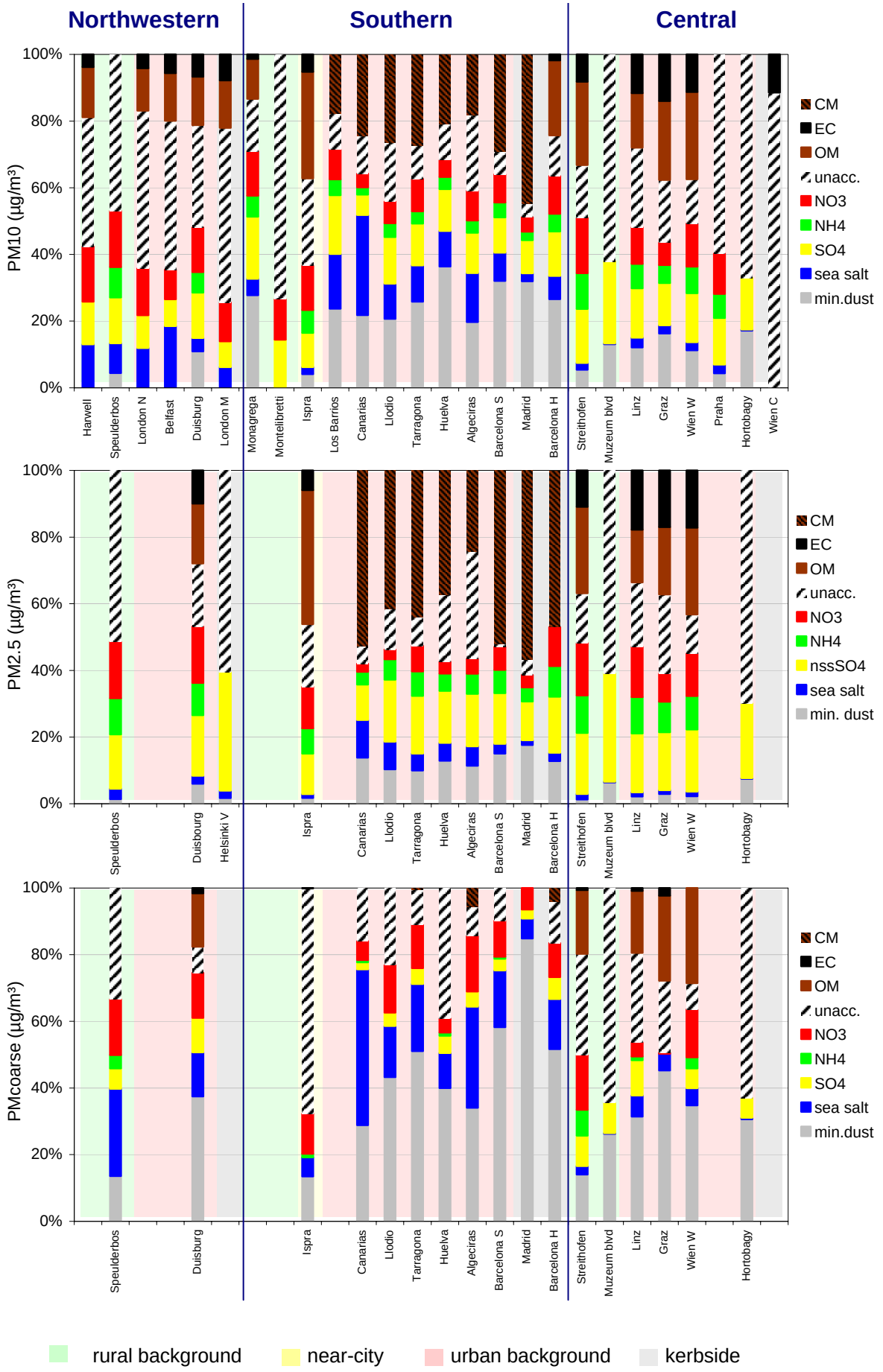


Figure 14 shows the annual averaged chemical composition of (a) PM_{10}, (b) $PM_{2.5}$, the "fine" fraction, and (c) PM_{10}-$PM_{2.5}$, the "coarse" fraction.

Where all main aerosol components are measured they account for close to 70% or more of the PM_{10} and $PM_{2.5}$ mass at the COST633 sites. The unaccounted mass is much larger in PM_{coarse} (average = 34%), calculated as $PM_{10} - PM_{2.5}$.

All aerosol components are present in both the fine and the coarse fraction. However, mineral dust and sea salt contribute more to the coarse fraction, whereas ammonium (NH_4^+), sulfate (SO_4^{2-}) and elemental carbon (EC) or total carbonaceous matter (CM) contribute more to the fine fraction. Organic matter (OM) and nitrate (NO_3^-) can contribute comparably to both fractions. OM in the fine fraction may result from primary emissions from combustion processes or from the oxidation of volatile organic compounds (VOC), whereas OM in the coarse fraction may also include biological debris (Bauer et al., 2008). Nitrate is bound to ammonium in the fine fraction, to sea-salt and/or mineral dust particles in the coarse fraction.

Table 4 presents mean PM_{10} chemical compositions for different site types in 3 geographical sectors of Europe. We included the total carbon variable TC =OC + EC, because it is a much more robust metric than OC and EC, and it is available at a larger number of sites of the COST633 Network. Clear gradients in PM_{10} chemistry can be observed:

- the contribution of mineral dust is much higher in Southern Europe, for any kind of site.
- the contribution of sea salt is minimum in Central Europe, and can be as high in Southern Europe (even excluding data from Canarias) as in Northwestern Europe.
- the contribution of SO_4^{2-} increases from NWestern to Southern to Central Europe for all site types.
- TC / PM_{10} ratios are much larger in Central Europe than in Southern or NWestern Europe
- the contributions of SO_4^{2-} and NO_3^- decrease when moving from rural to kerbside sites in all 3 sectors of Europe.
- the TC / PM_{10} ratio increases from rural to kerbside sites in all 3 sectors of Europe, less obviously in Central Europe though.

PM10		rural	near-city	urban	kerbside
N-Western Europe	min. dust	4%			
	sea salt	11%		12%	6%
	SO4	13%		11%	8%
	NO3	17%		13%	12%
	OM	15%		15%	14%
	EC	4%		6%	8%
	TC	*14%*		*16%*	*18%*
Southern Europe	min. dust	27%	13%	24%	30%
	sea salt	5%	10%	1*1%	5%
	SO4	16%	14%	11%	12%
	NO3	13%	10%	7%	8%
	OM	12%	32%		
	EC	1%	5%		
	TC	*13%*	*21%*	*18%*	*28%*
Central Europe	min. dust	9%		15%	
	sea salt	1%		2%	
	SO4	20%		15%	
	NO3	16%		7%	
	OM	26%		22%	
	EC	8%		13%	11%
	TC	*27%*		*29%*	

Table 4

* Canarias excluded

Fig 15a

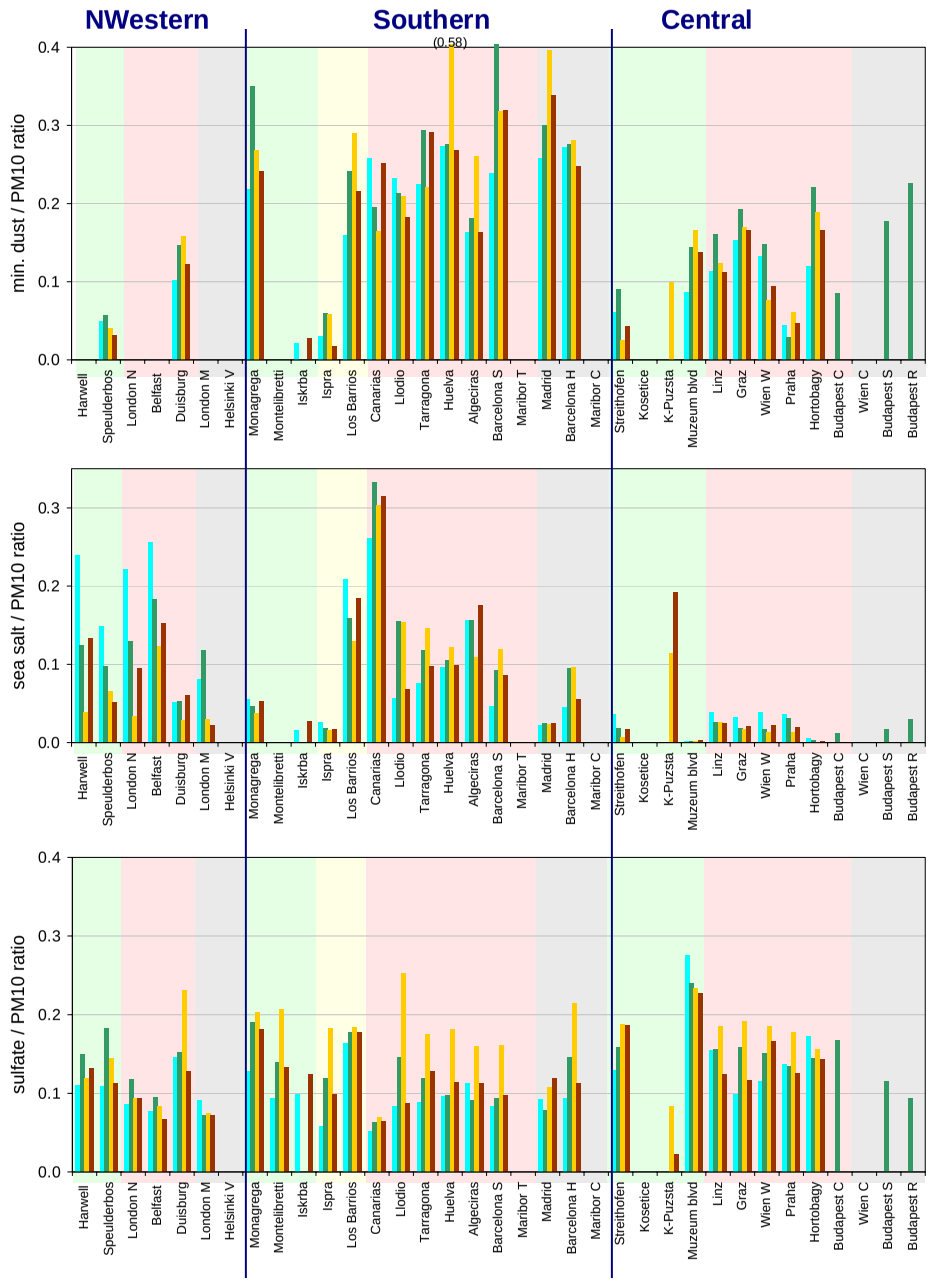
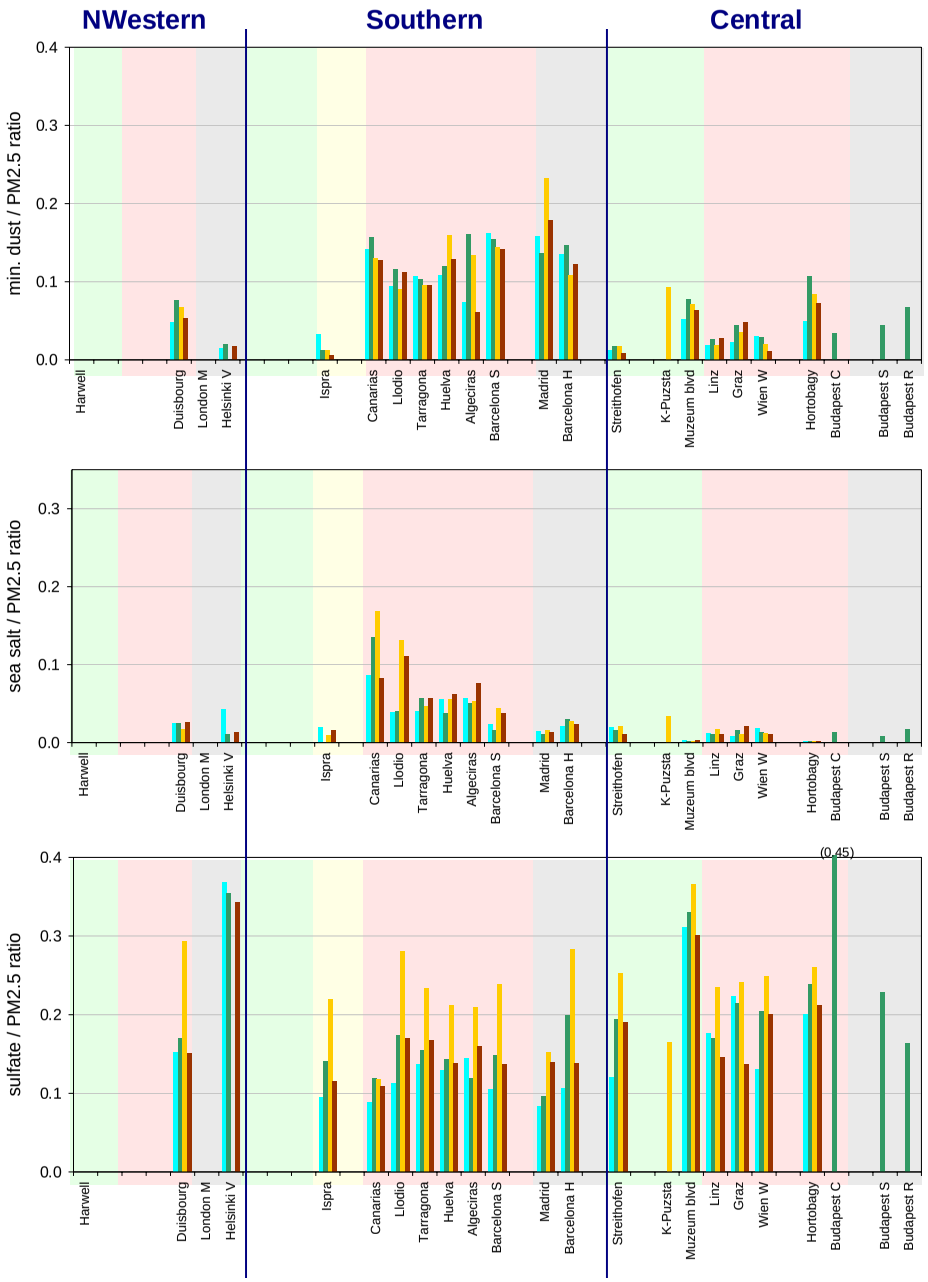
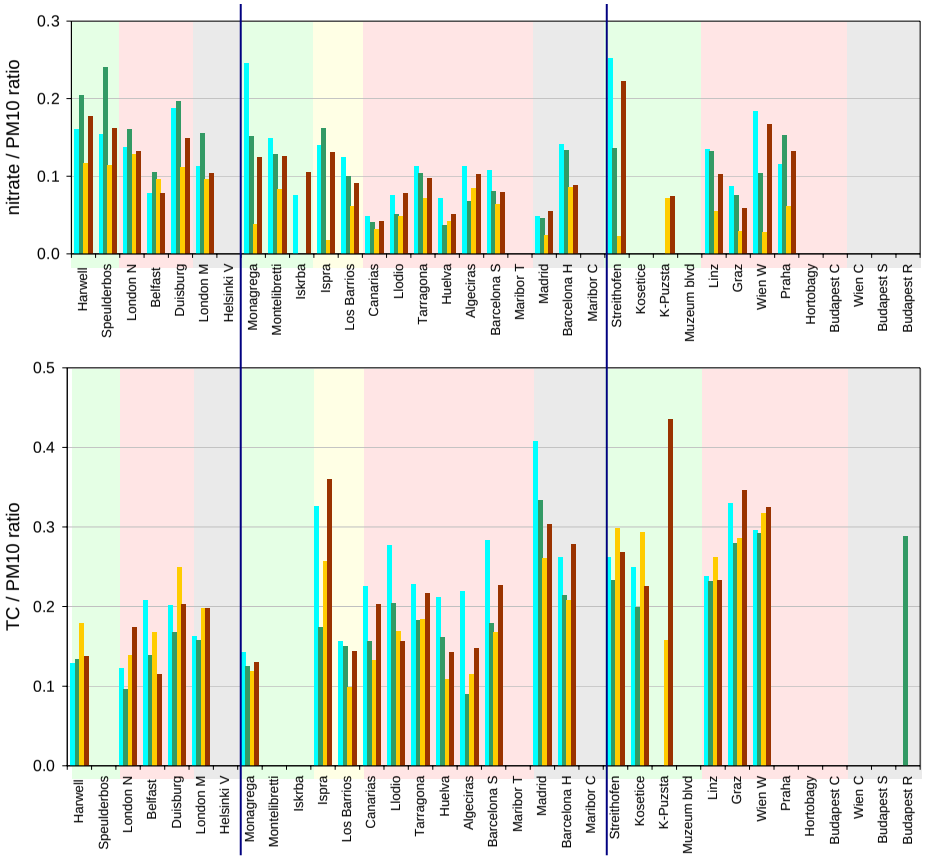
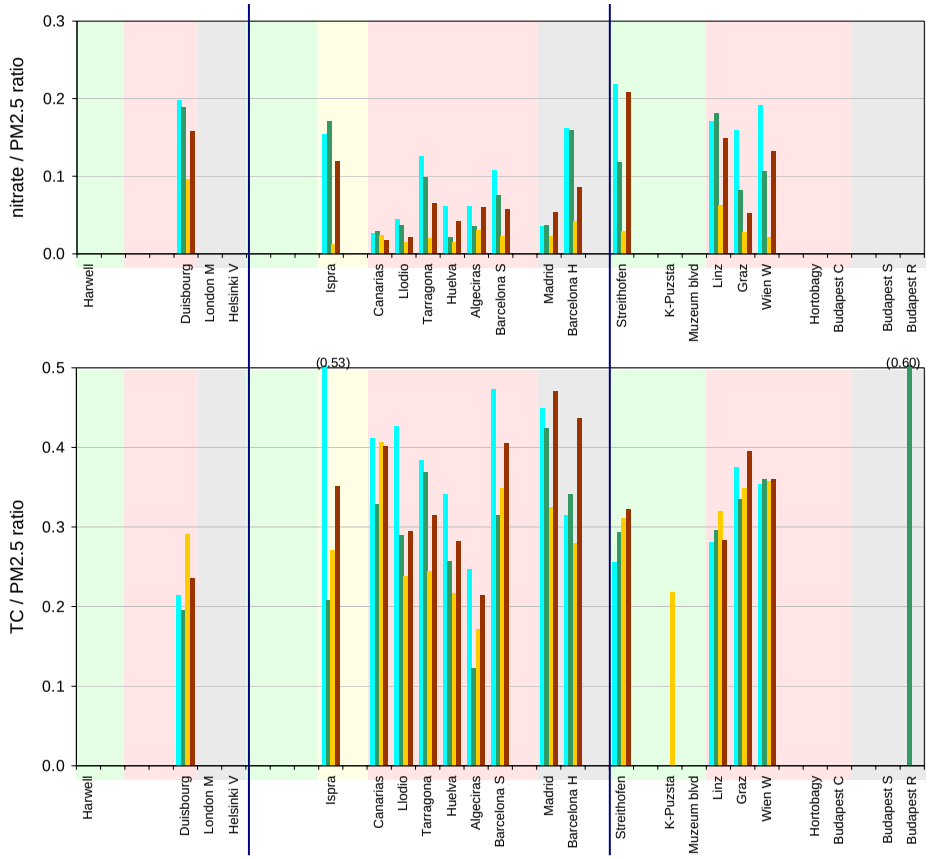


Fig 15b



DJF winter MAM spring JJA summer SON autumn





Figures 15a and 15b show the seasonal average contributions (DJF, MAM, JJA, SON) of major species to PM_{10} and $PM_{2.5}$, respectively, at the COST633 sites sorted according to 3 large geographical sectors and further categorized in 4 site types (rural, near-city, urban, and kerbside). These figures make it possible to assess at one glance the major components of PM_{10} and $PM_{2.5}$ at each site, and differences in PM_{10} and $PM_{2.5}$ chemical composition, even for sites at which PM chemistry data do not cover a whole year period.

It can be observed that the chemical composition of PM_{10} and $PM_{2.5}$ can be rather similar (e.g. Ispra, Linz, Graz, Wien W, etc…) or significantly different (e.g. Barcelona S), actually depending on the fraction of PM_{10} present in the coarse fraction. Let us notice that the contribution to $PM_{2.5}$ of sea salt and mineral dust is significant at several sites in Southern Europe.

The most striking observation related to seasonality regards the large changes in nitrate and TC contributions to PM_{10} and $PM_{2.5}$ observed at many sites Contributions of nitrate are up to > 10 times higher, those of TC > 2 times during winter compared to summer. Enhanced condensation of nitrate and organic matter during cold periods could explain these observations. However, they might also be at least partially due to sampling artifacts.

Fig 16a

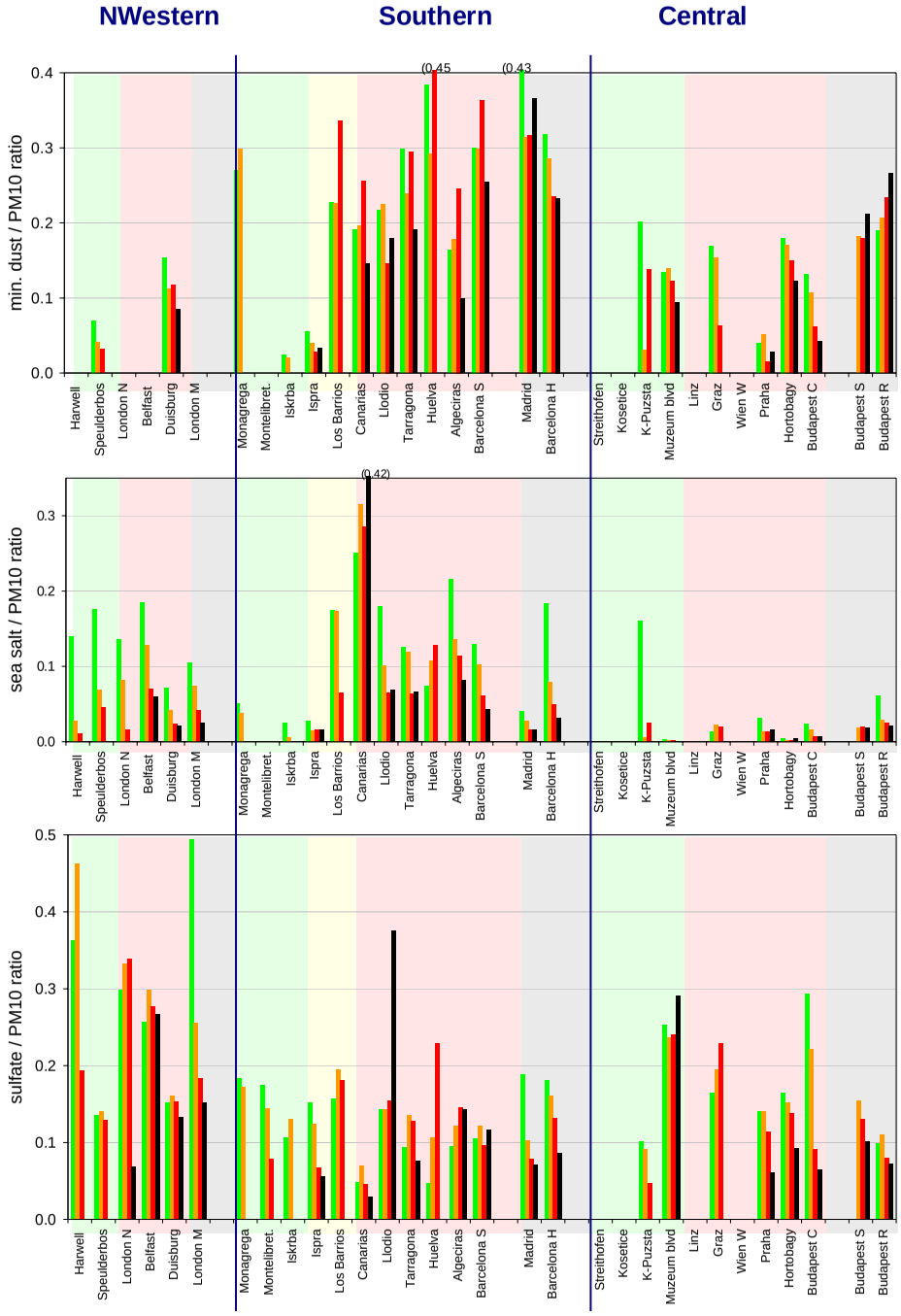
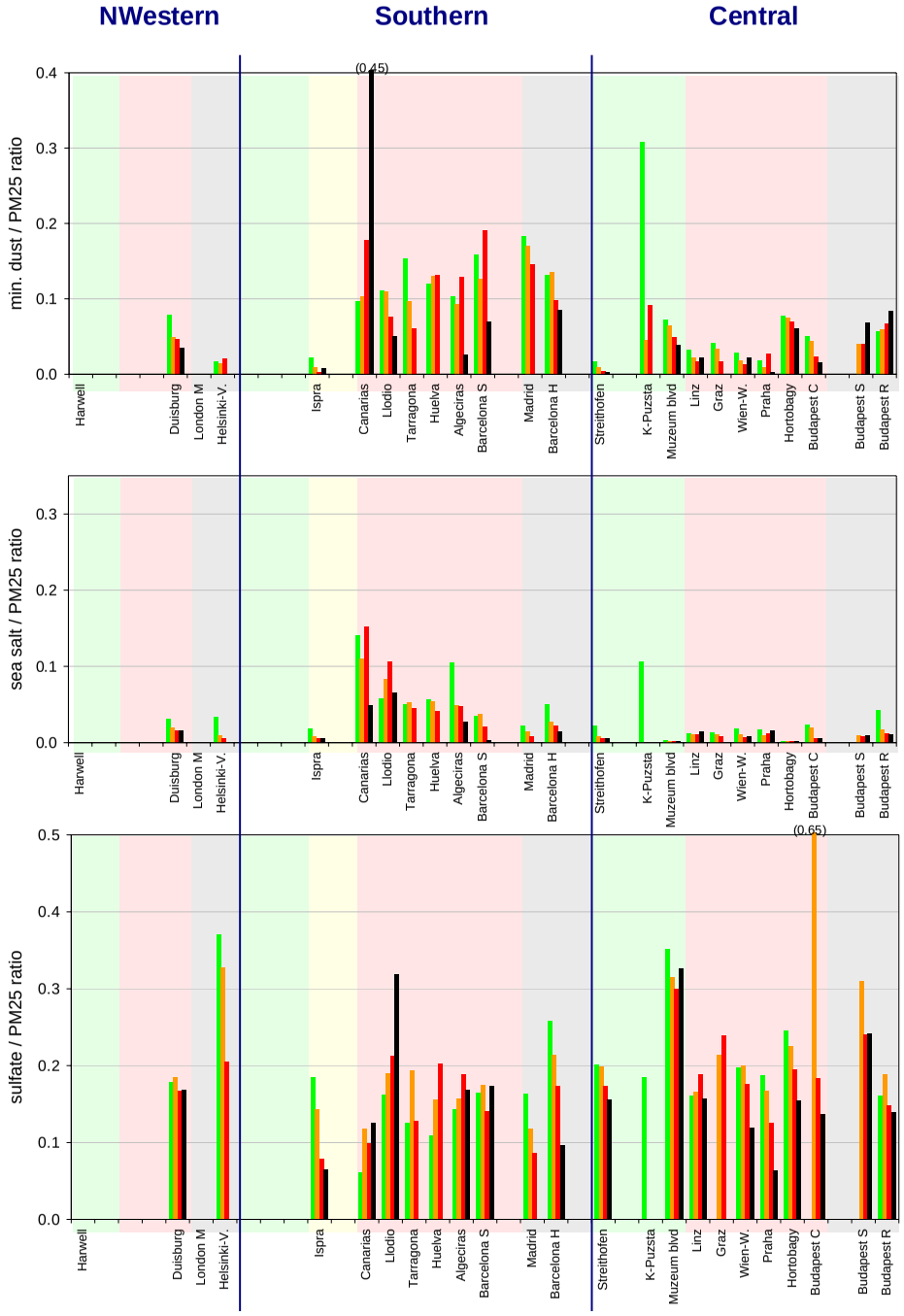
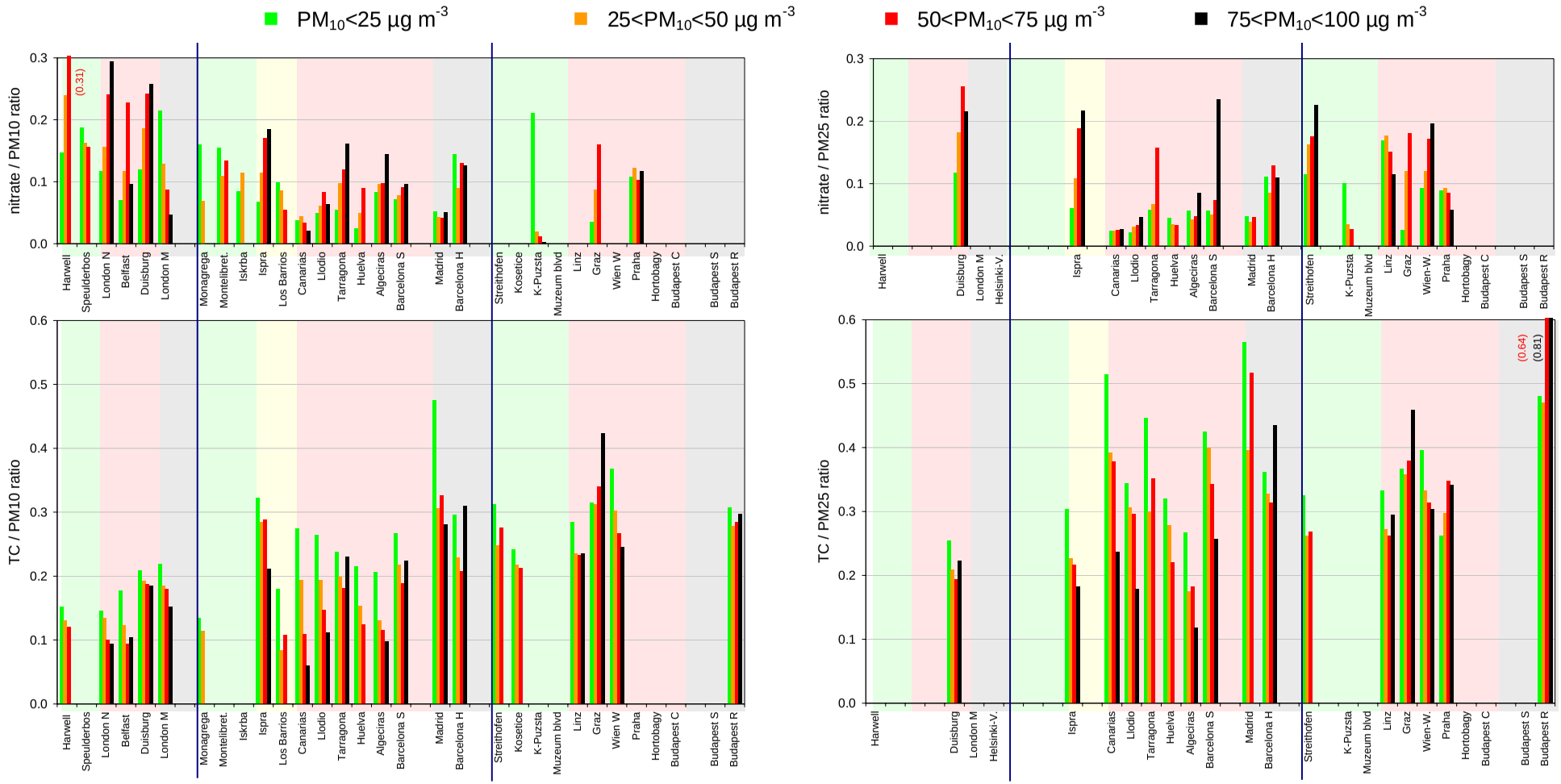


Fig 16b





Figures 15a and 15b show relative contributions of major PM constituents to PM_{10} and $PM_{2.5}$, respectively, averaged over four 25 µg m^{-3} wide PM_{10} size bins (■ PM_{10}<25 µg m^{-3}, ■ 25<PM_{10}<50 µg m^{-3}, ■ 50<PM_{10}<75 µg m^{-3}, ■ 75<PM_{10}<100 µg m^{-3}). Sites are categorized as in Fig. 15. Figs. 16 illustrate the answer to the question: Does PM_{10} and $PM_{2.5}$ chemistry change with increasing PM pollution levels ? At most sites, it does indeed. Mineral dust contribution to both PM_{10} and $PM_{2.5}$ decreases with PM_{10} levels except at kerbsides in Central Europe. As expected, the percentage of sea salt, a natural aerosol, decreases with increasing PM_{10} everywhere but in Canarias. The contribution of SO_4^{2-} to PM_{10} and $PM_{2.5}$ does not increase with PM_{10} concentrations, except at urban sites in Llodio, Huelva (ES), both affected by industrial emissions, and Graz (AT). Contributions of NO_3^- generally increase with PM_{10} levels, but clearly decrease at a few sites (London M, UK; Monagrega, Los Barrios, Canarias, ES; K-Puzsta, HU). The TC/PM_{10} ratio decreases with PM_{10} levels at all sites, except in Graz (increase) and Budapest R (constant). It peaks for PM_{10}> 75 µg m^{-3} at 3 sites in Southern Europe though. These exceptions are also observed in TC/$PM_{2.5}$ ratios. The "unknown" fraction (not shown) increases with PM_{10} at most sites of the COST633 network where it can be calculated.

Fig 17

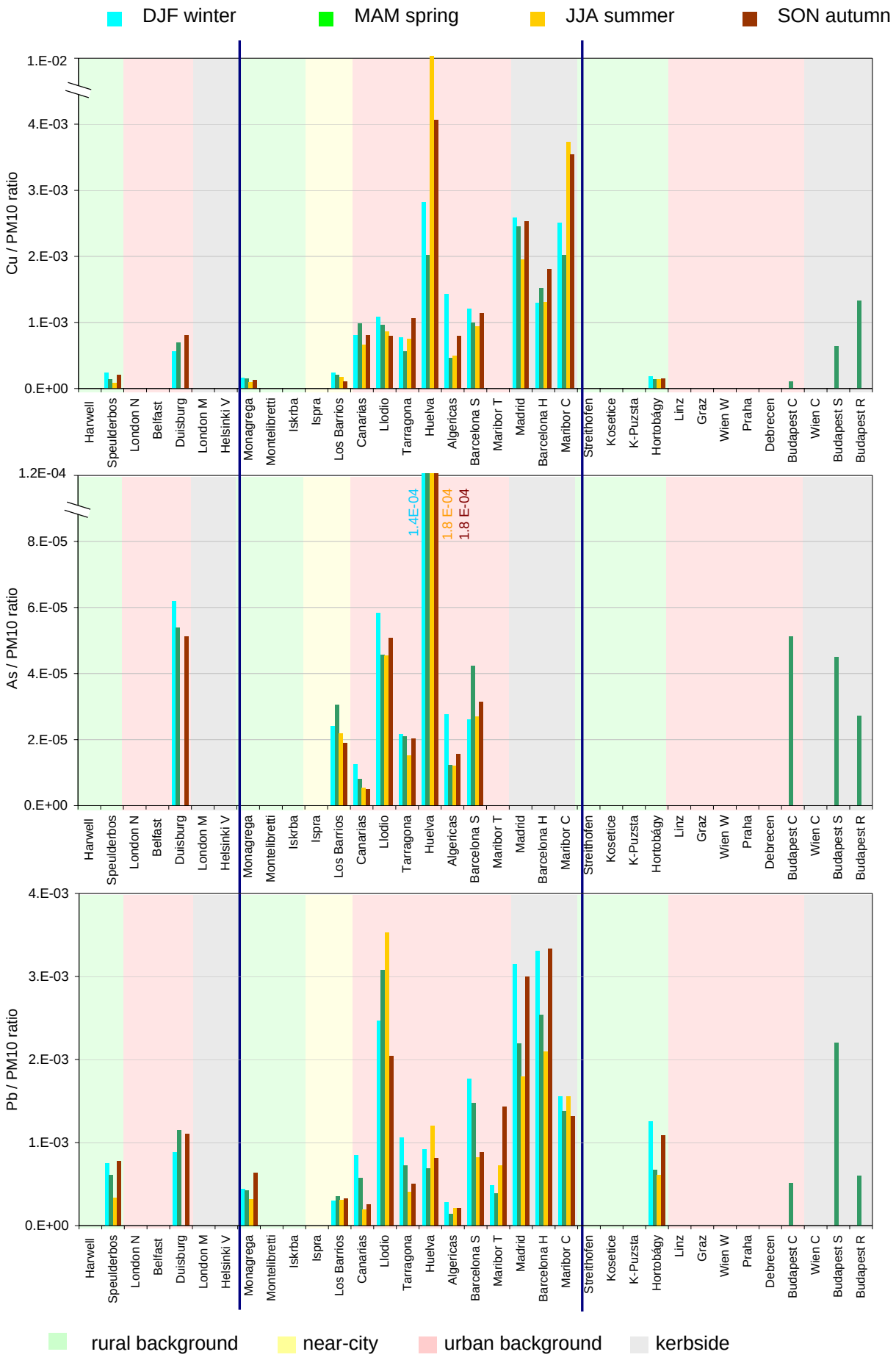

DJF winter
MAM spring
JJA summer
SON autumn
Cu / PM10 ratio
1.E-02
4.E-03
3.E-03
2.E-03
1.E-03
0.E+00
As / PM10 ratio
1.2E-04
8.E-05
6.E-05
4.E-05
2.E-05
0.E+00
1.4E-04
1.8 E-04
1.8 E-04
Pb / PM10 ratio
4.E-03
3.E-03
2.E-03
1.E-03
0.E+00
Harwell
Speulderbos
London N
Belfast
Duisburg
London M
Helsinki V
Monagrega
Montelibretti
Iskrba
Ispra
Los Barrios
Canarias
Llodio
Tarragona
Huelva
Algericas
Barcelona S
Maribor T
Madrid
Barcelona H
Maribor C
Streithofen
Kosetice
K-Puzsta
Hortobágy
Linz
Graz
Wien W
Praha
Debrecen
Budapest C
Wien C
Budapest S
Budapest R
rural background
near-city
urban background
kerbside

Figure 17 shows seasonal average (■ DJF, ■ MAM, ■ JJA, ■ SON) concentration ratios with respect to PM_{10} for 3 metals, selected because they were measured at a significant number of sites of the COST633 network. These sites are sorted according to 3 large sectors (Northwestern, Southern and Central Europe) and further categorized in 4 site types (rural, near-city, urban, and kerbside).

Although metals account for a very small fraction of PM mass concentrations, they are important elements because they can be used as tracers for specific sources, and they may have a role in determining the oxidative potential of the aerosol.

The limited number of data collected within COST633 prevents us from drawing definitive conclusions. However, we can observe the following features:

- Concentrations of As in PM_{10} are comparable in all 3 sectors. In contrast, the largest concentrations of Cu and Pb relative PM_{10} mass are observed in Southern Europe. The data from Speulderbos, Monagrega, Madrid, Barcelona H, Budapest S and Budapest R were collected in 1999-2000, before leaded gasoline was banned.
- Cu / PM_{10} ratios increase when moving from rural to kerbside sites, but As / PM_{10} and Pb / PM_{10} do not.
- Significant seasonal variations are observed for all 3 Cu / PM_{10}, As / PM_{10}, and Pb / PM_{10} ratios at some of the sites, which suggest seasonal variations in source strength. In most cases, these ratios are larger in autumn / winter than in spring / summer. A noticeable exception can be observed in Llodio (ES), where Pb / PM_{10} clearly peaks up in summer.

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Conclusions

1. The COST Action 633 compiled aerosol datasets where PM mass concentrations **and** PM chemistry **or** particle number data were available. The COST633 Network covers a wide variety of environmental conditions but still lacks sites from large regions in Europe. Most particle number data sets were collected from sites located in Northwestern Europe. (Fig. 4 & 5)

2. The EU 2005 annual PM_{10} standard of 40 µg m^{-3} and the EU 2010 annual $PM_{2.5}$ target of 25 µg m^{-3} may be exceeded at a few sites in the COST633 Network. PM concentrations in background urban sites in Southern 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. (Fig. 6)

3. Most PM_{10} mass is present in the $PM_{2.5}$ fraction, but $PM_{2.5}$ also contains a significant part of the coarse particle mode. (Fig. 1)

4. Considering all sites in the COST633 Network, $PM_{2.5}$ and PM_{10} mass concentrations are correlated with a correlation coefficient $R^2 = 0.69$. However the $PM_{2.5}/PM_{10}$ ratio (0.43 - 0.90) is too variable to propose a European-wide valid value. At each single site, the correlation between $PM_{2.5}$ and PM_{10} is generally much better, except at 3 sites where $PM_{2.5}$ levels off for $PM_{10} > 50$ µg m^{-3}. (Fig. 7)

5. $PM_{2.5}/PM_{10}$ ratios do not systematically increase or decrease with PM_{10} levels. Hence, PM pollution events (high PM_{10}) can be due to enhanced $PM_{2.5}$, or to enhanced coarse PM fraction, for instance where long range transported or re-suspended dust contribute significantly to PM_{10}. (Fig. 8)

6. Particle number size distribution measurements show that ultrafine particles (Dp<100 nm) contribute much to the total particle number, but little to PM mass. In contrast, super-µm particles (Dp>1 µm) are rare (<1% of the total particle number).(Fig. 10)

7. Particle (Dp>10 nm) number concentrations are rather similar at the urban background sites of the COST633 network, and somewhat smaller at the near-city site, while PM mass concentrations are significantly larger at this near-city site. In general, particle number does not increase monotonically with PM_{10} or PM2.5. For given range of PM_{10} or $PM_{2.5}$ mass concentration, particle number can vary by a factor of 2 among similar sites like city background in Northwestern Europe, and by more than a factor of 10 if all kind of sites are included (Fig. 11 & 12)

8. The ratio ultrafine / total particle number ratio generally decreases with increasing PM_{10} and $PM_{2.5}$ concentration levels (Table 1 & 2)

9. When all main chemical components of the aerosol are measured, they account for close to 70% or more of the PM_{10} and $PM_{2.5}$ mass. The rest may be due to the presence of water, to the underestimation of the molecular mass – to – carbon mass ratio when calculating organic matter concentrations, or to the underestimation of mineral dust when calculated based on tracers.

Determining the chemical composition of PMcoarse = PM_{10} – $PM_{2.5}$ leads to large uncertainties. (Fig. 13)

10. Organic matter is the main contributor to the annual average PM_{10} mass concentration for most site categories (type x region), except at rural sites in Northwestern Europe (nitrate), rural and kerbside sites in Southern Europe (mineral dust), and kerbside sites in Northwestern Europe (sea salt). Total carbon (TC) was considered instead of OM because it is a much more robust variable, and it was available at many more sites. There are clear gradients in mineral dust / PM_{10}, sea salt / PM_{10}, sulfate / $PM_{10,}$ and TC / PM_{10} ratios when moving from Northwestern to Southern to Central Europe, and in sulfate / $PM_{10,}$ nitrate / $PM_{10,}$ and TC / PM_{10} ratios when moving from rural to kerbside sites. (Table 3, Fig. 14)

11. The contributions of nitrate and TC to PM_{10} and $PM_{2.5}$ show large seasonal variations at many sites of the COST633 network. In winter, contributions of nitrate are up to > 10 times higher, those of TC > 2 times during winter compared to summer. Enhanced condensation of nitrate and organic matter during cold periods could explain these observations. However, they might also be at least partially due to sampling artifacts. (Fig. 15)

12. PM_{10} and $PM_{2.5}$ chemical composition significantly change at most sites, although no clear correlation exists between PM mass concentrations on the one hand, and chemical composition on the other hand. The TC/PM_{10} ratio generally decreases with PM_{10} levels at all sites, except in Graz (increase) and Budapest R (constant), but peaks for PM_{10} > 75 µg m^{-3} at 3 sites in Southern Europe. The "unknown" fraction (not shown) increases with PM_{10} at most sites of the COST633 network where it can be calculated. (Fig. 16)

13. Across the 16 COST633 sites for which Cu, As, and Pb concentrations in PM_{10} were reported, the largest Cu / PM_{10} and Pb / PM_{10} ratios are observed in Southern Europe. However some data from Spain were collected before leaded gasoline was banned. As / PM_{10} concentrations are quite comparable in Northwestern, Southern and Central Europe. In all 3 sectors Cu / PM_{10} ratios increase when moving from rural to kerbside sites, but As / PM_{10} and Pb /PM_{10} do not. Significant seasonal variations are observed at some of the sites (Fig. 17)

Concluding comments

The present document focuses on similarities and differences in aerosol properties which can be hidden behind similar or different PM_{10} and $PM_{2.5}$ mass concentrations, the only PM related variables measured by most air pollution monitoring networks. It highlights aerosol characteristics that may be considered when trying to explain why similar PM concentrations lead to different health effects in various locations in Europe, which is one of the main questions addressed by the COST Action 633. They might also be relevant for anticipating the effect of future climate change mitigation policies on air pollution and vice versa.

All data were collected within the frame of COST633 by the National Representatives and Working Group members. We do not pretend to be exhaustive, neither to explain the peculiarities of each site, but rather to underline the occurrence or lack of general features *within* the COST633 network. This was not always possible simply because the number of data available was too small. Indeed, besides the huge effort deployed in Europe for monitoring PM mass concentrations, there are very few sites where a full characterization of the aerosol was carried out, even for a limited period of time. The Environment Protection Agency in the USA established a number of super-sites to tackle this problem (http://www.epa.gov/ttn/amtic/supsites.html). In Europe, the development of a similar network of urban stations, measuring well selected relevant variables with a high level of quality control, would be helpful for developing efficient particulate pollution abatement strategies,

However, apart from an investment in new measurement/monitoring approaches, efforts should also be deployed for making available and analyzing existing data. Too many published aerosol data do not reach open access data bases, and get lost for further integrated assessments.

References to general publications.

Bauer, H., E. Schueller, G. Weinke, A. Berger, R. Hitzenberger, I. L. Marr and H. Puxbaum, Significant contributions of fungal spores to the organic carbon and to the aerosol mass balance of the urban atmospheric aerosol, Atmos. Environ.,42, 5542-5549, 2008.

Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe

http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:152:0001:0044:EN:PDF

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

Eatough, D.J., Eatough, D.A., Lewis, L., and Lewis, A.D., Fine particulate chemical composition and light extinction at Canyonlands National Park, , *J. Geophys. Res.*, *101,* 19515-19531, 1996.

Fjaeraa A.M. and Hjellbrekke A-G., Ozone measurements 2006, EMEP/CCC 2/2008, NILU, Norway, 2008.

Gomiscek B., Frank A., Puxbaum, H., Stopper, S., Preinig, O., Hauck, H., (2004) Case study analysis of PM burden at an urban and a rural site during the AUPHEP project. Atmospheric Environment 38 (24), 3935-3948

IPCC 2007, Intergovernmental Panel on Climate Change, Fourth Assessment Report, Cambridge University Press, 2007.

Kloster S., F. Dentener, J. Feichter, F. Raes, J. van Aardenne, E. Roeckner, U. Lohmann, P. Stier, and R. Swart, Influence of future air pollution mitigation strategies on total aerosol radiative forcing, ACPD 8 (2), 5563-5627. 2008

Larssen, S., Sluyter, R., and Helmis, C., Criteria for EUROAIRNET, theEEA air quality monitoring and information network, 1999, http://reports.eea.eu.int/TEC12/en.

Putaud, J.P., Raes , F, Van Dingenen, R, Brüggemann, E, Facchini, M.C., Decesari, S., Fuzzi, S., Gehrig, R., Hüglin, C., Laj, P., Lorbeer, G., Maenhaut, W., Mihalopoulos, N., Müller, K., Querol, X., Rodriguez, S., Schneider, J., Spindler, G., Ten Brink, H., Tørseth, K., and Alfred Wiedensohler, A., A European aerosol phenomenology–2: chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, *Atmos. Environ.*, 38, 2579-2595, 2004.

Puxbaum, H., Gomiscek, B., Kalina, M., Bauer, H., Salam, A., Stopper, S., Preinig, O., Hauck, H., A dual site study of PM2.5 and PM10 aerosol chemistry in the larger region of Vienna, Austria. Atmos. Environ. 38 (24), 3949-3958, 2004.

Van Dingenen, R., Putaud, J.P., Raes , F, etc..., A European aerosol phenomenology–1: physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, *Atmos. Environ.*, 38, 2561-2577, 2004.

Raes F., Van Dingenen R., Vignati E., Wilson J., Seinfeld J.H., and Adams P., Formation and cycling of aerosols in the global atmosphere, *Atmos. Environ*., 34, 4215-4240, 2000.

Robles-Gonzales, C.R., Veefkind, J.P., and de Leeuw, G., Aerosol optical depth over Europe in August 1997 derived from ATSR-2 data, *Geophys. Res. Let*, 27, 955958, 2000.

Wilson R. and Spengler J.D., Particles in Our Air: Concentrations and Health Effects. Harvard University Press, 1996

Wyzga E. R. Air Pollution and Health; Are Particulates the Answer? Proceedings of the NETL Conference "PM2.5 and Electric Power Generation: Recent Findings and Implications," Pittsburgh, PA, April 9-10, 2002.

http://www.netl.doe.gov/publications/proceedings/02/PM25/

References to sources of data.

Alastuey A., Sánchez-de-la-Campa A., Querol X., Rosa J.d.l., Plana F., Mantilla E., Viana M., Ruiz C.R. and Santos S.G.d. (2006) Identification and chemical characterisation of industrial PM sources in SW Spain. Journal of the Air and Waste Management Association, 56, 993-1006.

Aalto, P., Hämeri, K., Becker, E., Weber, R., Salm. J., Mäkelä, J. M., Hoell, C., O'Dowd, C., Karlsson, H., Hansson, H-C., Väkevä, M., Koponen, I., Buzorius, G. & Kulmala, M. Physical characterization of aerosol particles during nucleation events. Tellus 53B: 344-358, 2001.

Abdalmogith S. S., Harrison R. M. "The use of trajectory cluster analysis to examine the long-range transport of secondary inorganic aerosol in the UK" Atmospheric Environment 39, 6686-6695, 2005.

Berner A., Galambos Z., Ctyroky P., Frühauf P., Hitzenberger R., Gomiscek B., Hauck H., Preinig O., Puxbaum H. On the correlation of atmospheric aerosol components of mass size distributions in the larger region of a central European city. Atmospheric Environment, 38 (24), 3959-3979, 2004.

Dal Maso, M., M. Kulmala, I. Riipinen, R. Wagner, T. Hussein, P. P. Aalto and K. E. J. Lehtinen: "Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEARII, Hyytiälä, Finland", Boreal Environment Research 10 (5): 323-336, 2005.

Dal Maso, M., L. Sogacheva, P. P. Aalto, I. Riipinen, M. Komppula, P. Tunved, L. Korhonen, V. Suur-Uski, A. Hirsikko, T. Kurten, V.-M. Kerminen, H. Lihavainen, Y. Viisanen, H.-C. Hansson, and M. Kulmala: "Aerosol size distribution measurements at four Nordic field stations: identification, analysis and trajectory analysis of new particle formation bursts", Tellus B, 60, 350 – 361, 2007.

Erisman J.W., G. Draaijers, J. Duyzer, P. Hofschreuder, N. Van Leeuwen, F. Römer, Walter Ruijgrok, Paul. Wyers[‖] and M. Gallagher, Particle deposition to forests—Summary of results and application, Atmos. Environ. 31, 321-332, 1997.

Hari, P. & Kulmala, M. Station for Measuring Ecosystem-Atmosphere Relations (SMEAR II). Boreal environment research 10, 315-322, 2005.

Harrison R. M., Jones A. M. "Multisite Study of Particle Number Concentrations in Urban Air" Environ. Sci. Technol. **39**, 6063-6070, 2005.

Hauck H., Berner A., Frischer T., Gomiscek B., Kundi M., Neuberger M., Puxbaum H., Preining O. and AUPHEP Team. AUPHEP Austrian Project on Health Effects of Particulates General Overview. Atmospheric Environment 38 (24), 3905-3915, 2004.

Hauck, H., Berner A., Gomiscek, B., Stopper, S., Puxbaum, H., Kundi, M., Preinig, O. On the equivalence of gravimetric PM data with TEOM and beta-attenuation measurements, Journal of Aerosol Science 35, 1135-1149, 2004.

Hitzenberger, R. Absorption measurements with an integrating plate photometer - calibration and error analysis. Aerosol Sci. Technol. 18, 70-84, 1993.

Hitzenberger, R., Tohno, S. Comparison of black carbon (BC) aerosols in two urban areas (Uji, Japan and Vienna, Austria) - concentrations and size distributions Atmos. Environ. 35, 2153-2167, 2001.

J. de Hartog, J., G. Hoek, A. Mirme, Thomas. Tuch , G. P.A. Kos, H.. ten Brink, B. Brunekreef, J. Cyrys, J. Heinrich, M. Pitz, T. Lanki, M. Vallius, J. Pekkanen and W. G. Kreyling, Relationship between different size classes of particulate matter and meteorology in three European cities, *J. Environ. Monit.*, 7, 302 – 310, 2005.

Janssen NA, de Hartog JJ, Hoek G, Brunekreef B, Lanki T, Timonen KL, Pekkanen J. Personal exposure to fine particulate matter in elderly subjects: relation between personal, indoor and outdoor concentrations. J Air Waste Manag Assoc 2000;50:1133-1143.

Jones A. M., Harrison R. M., "Interpretation of particulate elemental and organic carbon concentrations at rural, urban and kerbside sites". Atmospheric Environment **39** 7114–7126, 2005.

Khlystov A, Kos GPA, ten Brink HM, Mirme A, Tuch Th, Roth Ch, Kreyling WG. Comparability of three spectrometers for monitoring urban aerosol. Atmos Environ 2001; 35: 2045-2051.

Maenhaut, W., N. Raes, X. Chi, J. Cafmeyer, W. Wang, I. Salma: Chemical composition and mass closure for fine and coarse aerosols at a kerbside in Budapest, Hungary, in spring 2002, X-ray Spectrometry 34 (2005) 290–296.

Mirme A, Kreyling WG, Khlystov A, ten Brink H, Ruuskanen J, Tuch T, Pekkanen J. Intercomparison of Aerosol Spectrometers for Ambient Air Monitoring. Aerosol Science and Technology 2002; 36: 866-876.

Putaud, JP, Dell'Acqua, A., Martins Dos Santos, S., and Huber, JRC Ispra EMEP – GAW regional station for atmospheric research 2003–2004 report, EUR22557, ISSN 1018-5593, 2006.

Quass, U., Kuhlbusch T.A.J., Koch M. (2004) Identification of Source Groups for Fine Dust (English summary), Public report to the Environment Ministry of North Rhine Westphalia, Germany, IUTA-Report LP15/2004, download:

http://www.lanuv.nrw.de/luft/berichte/FeinstaubNRW_2004_Summary.pdf.

Querol X., Alastuey A., Moreno T., Viana M., Castillo S., Pey J., Rodriguez S., Artiñano B., Salvador P., Sanchez M., Garcia Dos Santos S., Herce Garraleta M.D., Fernandez-Patier R., Moreno-Grau S., Minguillon.C., Monfort E., Sanchez M.J., Palomo-Marin R., Pinilla-Gil E. and Cuevas E. (2008) Spatial and temporal variations in airborne particulate matter (PM10 and PM2.5) across Spain 1999-2005. Atmospheric Environment 42, 39643979.

Querol X., Alastuey A., Viana M.M., Rodriguez S., Artiñano B., Salvador P., Santos S.G.D., Patier R.F., Ruiz C.R., Rosa J.D.L., Campa A.S.D.L., Menedez M. and Gil J.I. (2004) Speciation and origin of PM10 and PM2.5 in Spain. Journal of Aerosol Science 35(9), 1151-1172.
Rodriguez S., Querol X., Alastuey A. and Plana F. (2002). Sources and processes afecting levels and composition of atmospheric aerosol in the Western Mediterranean. Journal of Geophysical Research Atmospheres, 107 D24 doi:10.1029/2001JD001488

Ruuskanen J, Tuch Th, Ten Brink H, Peters A, Khlystov A, Mirme A, Kos GPA, Brunekreef B, Wichmann HE, Buzorius G, Vallius M, Kreyling W, Pekkanen J. Concentrations of ultrafine, fine and PM2.5 particles in three European cities. Atmos Environ; 35: 3729-3738, 2001.

Salma, W. Maenhaut, É. Zemplén-Papp, Gy. Záray: Comprehensive characterisation of atmospheric aerosols in Budapest, Hungary: physicochemical properties of inorganic species, Atmospheric Environment 35 (2001) 4367–4378.

Salma, I., W. Maenhaut, S. Dubtsov, É. Zemplén-Papp, G. Záray: Impact of phase out of leaded gasoline on the air quality in Budapest, Microchemical Journal 67 (2000) 127–133.

Salma, I., W. Maenhaut: Changes in chemical composition and mass of atmospheric aerosol pollution between 1996 and 2002 in a Central European city, Environmental Pollution 143 479–488, 2006.

Salma, I., X. Chi, W. Maenhaut: Elemental and organic carbon in urban canyon and background environments in Budapest, Hungary, Atmos. Environ. 38, 27-36, 2004.

UK Air Quality Archive, www.airquality.co.uk/archive.

Vallius M, Lanki T, Tiittanen P, Koistinen K, Ruuskanen J, Pekkanen J. Source apportionment of urban ambient $PM_{2.5}$ in two successive measurement campaigns in Helsinki, Finland. Atmospheric Environment; 37: 615-623, 2003.

Vallius M, Ruuskanen J, Pekkanen J. Comparison of multivariate source apportionment of urban PM2.5 with chemical mass closure. Boreal Environment Res; 13:347-358, 2008.

Viana M.M., Querol X., Alastuey A., Ibarguchi J.I. and Menendez M. (2006) Identification of PM sources by Principal Component Analysis (PCA) coupled with wind direction data. Chemosphere 65(12), 2411-2418.

Annex 1. Data consistency

It is of course expected that data quality wais assured by data providers. However, different types of techniques or instruments were used to measure some of the aerosol characteristics listed in this report. A limited amount of data processing was also necessary to make the data collected from all sites comparable.

A2.1 PM mass concentrations

PM_{10} and $PM_{2.5}$ were mostly measured by gravimetry. A reference method (EN 12341) exists for this technique. However, the substrates used for collecting atmospheric PM may lead to various (and not completely understood yet) positive and negative sampling artifacts.

At a few sites (see Annex 2), PM mass concentrations were obtained with on-line monitors (TEOM™, Beta gauge). The data obtained with these instruments were corrected (or not) by the data providers to make them equivalent to the reference method, according to inter-comparison results.

For Hyytiälä, $PM_{2.5}$ was derived from the particle volume, assuming an aerosol density of 1.5.

A2.2 Particle number size distributions

are usually measured with DMPS/SMPS. Significant differences in sizing and counting can be observed among instruments with different settings and from different origins. Most (but not all) measurements were performed in dry conditions (RH < 20%). Submicron particle counting is performed using condensation particle counters (CPC) that usually are consistent within ±10 %. Larger particles are sized and counted with APSs or OPCs. APS diameters were converted to mobility-like diameters assuming an aerosol density of 1.5.

A2.3 Main ions

Ion chromatography and capillary electrophoresis are robust techniques that can be easily calibrated and inter-compared using reference solutions. The uncertainty in ions concentration measurements is therefore expected to be <15%. However, larger systematic errors may arise from sampling artifacts affecting semi-volatile species like e.g. NH_4NO_3.

The sea salt component was calculated in the same way for all sites, based on measurements of the best sea salt tracer available (Na^+ or Mg^{2+}), assuming a standard sea water composition, but taking into account the possible loss of Cl^- when Cl^- was measured.

A2.4 Carbonaceous component

OC and EC can be measured with different techniques that may lead to very different results, mainly for EC. No standard method was available when the measurements we compiled were performed. Comparison between EC concentrations obtained at various sites could therefore lead to erroneous conclusions. TC concentrations are much more robust and are not expected to be affected by uncertainties larger than ±10%, sampling artifacts excluded.

Reported OC concentrations were converted to OM concentrations using 1.4 as a factor of conversion, although we are aware that this factor might be higher at sites located far away from pollution sources compared to sites located closer. Reported TC concentrations were converted to CM (carbonaceous matter) concentrations assuming a constant EC/TC ratio = 0.1, and a constant OM/OC ratio = 1.4. The overall uncertainty of CM is much more affected by the uncertainty associated with the OC→OM conversion factor than by the uncertainty of the EC/TC ratio.

A2.5 Mineral dust

This component is expected to include PM components that are nor soluble in water neither carbonaceous. Mineral dust was calculated from various tracers according to their availability and to the experience of the data providers according to one of the equations below, leading to similar results:

- min. dust = 2.1 Si +1.9 Al +2.5 Ca + 3.5 Mg + Fe + K

- min. dust = 7.6 Al +2.5 Ca + 3.5 Mg + Fe + K

- min. dust = 4.7 Ca + 3.5 Mg + Fe + K

- min. dust = 4.5 Ca

Annex 2. Methods and data sets

Country	Location	Size fraction	Sampling	Frequency/resolution
Austria	Wien C	TSP	LVS	Daily
	Wien W	PM10, PM2.5	HVS Digitel	Daily
	Graz	PM10, PM2.5	HVS Digitel	Daily
	Streithofen	PM10, PM2.5	HVS Digitel	Daily
	Linz	PM10, PM2.5, PM1	HVS Digitel	Daily
Czech Republic	Kosetice	PM10	LVS	weekly, 24 hr
	Praha	PM10, PM2.5, PM1, SD	LVS and LPBI	6th day, 24 hr
Finland	Hyytiälä	SD, PM10, PM2.5, PM1	Dekati impactor	Number: 10 min, mass:2-3 days
	Helsinki V	PM2.5, NSD (10nm-10um)	Harvard impactor	Impact. 24 hr, CPC & EAS < 1 hr
Germany	Duisburg	PM10, PM2.5, PM1	Digitel	24 hr
Hungary	Debrecen	PM10; PM2.5	Gent SFU sampler,	24 hours twice/week
	Budapest C	PM10-2.0, PM2.0	SFU	24 hours, daily
	Budapest S	PM10-2.0, PM2.0	SFU	24 hours, daily
	Budapest R	PM10-2.0, PM2.0	SFU	12 hours, daily
	Hortobágy	PM10; PM2.5	Gent SFU sampler	24 hours twice/week
	K-puszta	PM1.5	HVS	24-48 hours*
	K-puszta	size distribution	ELPI	12 hours, twice per day
Italy	Montelibretti	PM10	LVS	daily
	Ispra	PM10, PM2.5	LVS	daily
Netherlands	Amsterdam	PM2.5	Harvard LVS	24hr
	Speulderbos	PM10	LVS	day/night filter weekly
Poland	Krakow	PM10		1h
Slovenia	Maribor T	PM10		1hr, 1 day/week
	Maribor C	PM10		30 min and daily
	Iskrba	PM10 and PM2	SFU	daily and 2-3 days/week
	Iskrba	PM10	LVS	daily and 1d/week
Spain	Barcelona H	PM10, PM2.5	MCV (PM10 & PM2.5) HVS	24 hr
	Barcelona S	PM10, PM2.5	Andersen (PM10) & MCV (PM2.5) HVS	24 hr
	Algeciras	PM10, PM2.5	Andersen (PM10) & MCV (PM2.5) HVS	24 hr
	Los Barrios	PM10	Digitel (PM10) HVS	24 hr
	Llodio	PM10, PM2.5	Andersen (PM10) & MCV (PM2.5) HVS	24 hr
	Tarragona	PM10, PM2.5	Andersen (PM10) & MCV (PM2.5) HVS	24 hr
	Madrid	PM10, PM2.5	Andersen (PM10) & MCV (PM2.5) HVS	24 hr
	Huelva	PM10, PM2.5	Andersen (PM10) & MCV (PM2.5) HVS	24 hr
	Canarias	PM10, PM2.5	Andersen (PM10) & MCV (PM2.5) HVS	24 hr
	Monagrega	PM10	Andersen (PM10)	24 hr
United Kingdom	London M	PM10, PM2.5, SD	R&P5400 + Partisol/PTFE	1 hr
	London B	PM10, PM2.5, SD		1 hr
	Harwell	PM10, PM2.5, SD		1 hr
	Glasgow	10 nm -10 µm		15 min
	Belfast	10 nm -10 µm	R&P5400 + Partisol/PTFE	15 min
	Birmingham	10 nm -10 µm		15 min
	Harwell	PM10	R&P5400 + Partisol/PTFE	1d
	Manchester	10 nm -10 µm		15 min
	London N	PM10	R&P5400 + Partisol/PTFE	1d
	Port Talbot	10 nm -10 µm		15 min

LVS: low volume sampler

HVS: high volume sampler

LPBI: low pressure Berner impactor

SFU: stacked filter unit

ELPI: electrical low pressure impactor

SJAC: steam jet aerosol collector

PTFE: poly tetra fluoro ethylene

Annex 2. Methods and data sets (cont'd)

variable / method	variable / method	variable / method	variable / method	variable / method	time periods
mass / gravimetry			EC / integrated sphere		2001-2002
mass / gravimetry		ions / IC	OC, EC / TOA	trace metals / ICP	June 1999- May 2000
mass / gravimetry		ions / IC	OC, EC / TOA	trace metals / ICP	Oct. 2000 – Sept. 2001
mass / gravimetry		ions / IC	OC, EC / TOA	trace metals / ICP	June 1999- May 2000
mass / gravimetry		ions / IC	OC, EC / TOA	trace metals / ICP	Oct. 2000 – Sept. 2001
mass / gravimetry		ions / IC	OC, EC / TOA		04/07/05 - 03/07/06
mass / gravimetry		ions / IC	OC, EC / TOA	trace elem. / INAA, AAS	23/02/2006 - 13/01/2007
mass / volume size dist.	particle SD / DMPS, APS				2004 - 2006
mass / gravimetry	particle number / CPC	ions / IC	EC / SSR	trace elem. / AES, ED-XRF	Nov. 1998 - Apr. 1999
mass / gravimetry		ions / IC	OC, EC / TOA	trace elem. / ICPMS & TXRF	28/02/02 - 01/04/2003
mass / gravimetry		inorganic ions / CE	EC / SSR	trace elem. / PIXE	1996 - 2005
mass / gravimetry		inorganic ions / CE	EC / SSR	trace elem. / INAA, PIXE	Apr 10, 1996- May 17, 1996
mass / gravimetry		inorganic ions / CE	EC / SSR	trace elem. / INAA, PIXE	Apr 10, 1996- May 17, 1996
mass / gravimetry		inorganic ions / CE	OC, EC / TOT	trace elem. / INAA, PIXE	Apr 23, 2002 - May 05, 2002
mass / gravimetry		inorganic ions / CE	EC / SSR	trace elem. / PIXE	09-Feb-1995 - 22-Sept-1998
mass / TEOM		inorganic ions / CE	OC, EC / TOT		1996, 1999 - 2002
mass / TEOM		inorganic ions / CE	OC / TOC		1999 - 2001
mass / BAM		NO3- + SO42- / IC			whole 2003 + 2004
mass / gravimetry	particle SD / DMPS, APS, OI	inorganic ions / IC	OC, EC / EGA		whole 2003 + 2005
mass /BS	particle SD / SMPS				1/11/98 - 19/6/99
mass / BAM		inorganic ions / IC		trace elements/ICP	12/01/95 01/01/96
mass / FDMS-TEOM	particle SD / SMPS + OPC				Jan - Feb 2005
mass / TEOM				heavy metals / ICPMS	2002 - 2004
mass / TEOM + gravim.			PAHs / HPLC	heavy metals / ICPMS	2001 - 2002
mass, HM				heavy metals / ICPMS	Nov. 1995 to Oct. 1996
mass, HM				heavy metals / AAS	whole 2004
mass / gravimetry		inorganic ions / IC	OC, EC / TOA	trace elem. / ICP-AES & -MS	Jun. 1999 - Jun. 2000
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	08/01/2001-27/12/2001
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	2003 - 2004
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	2003 - 2004
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	08/01/2001-27/12/2001
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	08/01/2001-27/12/2001
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	08/01/2001-27/12/2001
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	08/01/2001-27/12/2001
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	08/01/2001-27/12/2001
mass / gravimetry		inorganic ions / IC	TC / CHN analyser	trace elem. / ICP-AES & -MS	Mar. 1999 - Jul. 2000
mass / TEOM	particle number / DMPS	inorganic ions / IC	OC, EC / R&P5400		2000 - 2005, 2001 -2005
mass / TEOM	particle number / DMPS				2002 - 2004
mass / TEOM	particle number / DMPS				2002 - 2005
mass / TEOM	particle number / CPC				2000 - 2005
mass / TEOM	particle number / CPC	inorganic ions / IC	OC, EC / R&P5400		2000 - 2005, 2002 - 2005
mass / TEOM	particle number / CPC				2000 - 2005
mass / gravimetry		inorganic ions / IC	OC, EC / R&P5400		2002 - 2005
mass / TEOM	particle number / CPC				2000 - 2005
mass / gravimetry		inorganic ions / IC	OC, EC / R&P5400		2000 - 2005
mass / TEOM	particle number / CPC				2000 - 2005

TEOM ™: tampered element oscilating microbalance
FDMS ™: filter dynamics measurement system
BAM: Beta Att. Monitor
BS: Black Smoke

SD: size distribution
DMPS: differential mobility particle sizer
APS: aerodynamic particle sizer
CPC: condensation particle counter
SMPS: scasnning mobility particle sizer
OPC: optical particle counter

IC: ion chromatography
CE: capillary electrophore

TOA: themal-optical analysis
SSR: smoke stain reflectometer
TOT: themal-optical, tranmittance
TOC: total organic carbon analyser
EGA: evolved gas analysis
CHN: elemental analyser

ICP:Inductively Coupled Plasma
INAA: Instrumental Neutron Activation Analysis
AAS: Atomic Absorption Spectrophotometry
ED-XRF: Energy-Dispersive X-ray Diffraction
PIXE: Particle-Induced X-Ray Emission
MS: Mass Spectroscopy
AES: Atomic Emission Spectrophotometry
TXRF: Total X-RayFluorescence Analysis

Annex 3. Sites and data providers

Country	Location	Site	Type	lat	long	alt	Contact	
Austria	Wien C	RHV	kerbside	48.20	16.37	185	R. Hitzenberger	Univ. Vienna
	Wien W	Währinger Str.	urban	48.22	16.35	185	H. Puxbaum	TU Vienna
	Graz	Hergottwiesgasse	urban	47.03	15.43	345	H. Puxbaum	TU Vienna
	Streithofen	Michelhausen	rural	48.27	15.93	220	H. Puxbaum	TU Vienna
	Linz	Blumauer Str.	urban	48.30	14.30	263	H. Puxbaum	TU Vienna
Czech Republic	Kosetice	EMEP station	rural	49.60	15.08	534	J. Santroch	CHMI
	Praha	ICPF campus	urban background	50.08	14.43	285	J. Smolik	ICPF, CHMI
Finland	Hyytiälä	SMEAR II	rural	61.85	24.28	180	Pasi Aalto	Univ. Helsinki
	Helsinki V	Vallila	urban traffic	60.19	24.97	3	Juha Pekkanen	National Public Health Institu
Germany	Duisburg	University	urban background	51.43	6.75	31	Thomas Kuhlbusch	IUTA
Hungary	Debrecen		city	47.54	21.68	121	Borbély-Kiss, I.	Hung. Ac. of Sciences
	Budapest C	KFKI campus	urban background	47.5087	19.0278	114	Salma, I.	Eotvos University
	Budapest S	Széna square	downtown	47.5087	19.0278	114	Salma, I.	Eotvos University
	Budapest R	Rákóczi street	downtown	47.5087	19.0278	114	Salma, I.	Eotvos University
	Hortobágy		rural	47.45	20.9333	83	Borbély-Kiss, Ildikó	Hung. Ac. of Sciences
	K-puszta	EMEP station	rural	46.97	19.55	125	Kiss, G.	Pannon University
	K-puszta	EMEP station	rural	46.97	19.55	125	Molnár, A.	Hung. Ac. of Sciences
Italy	Montelibretti	EMEP station	rural	42.10	12.63	48	C. Perrino	CNR - IIA
	Ispra	EMEP station	near city	45.82	8.63	209	JP Putaud	JRC - IES
Netherlands	Amsterdam		urban background	52.30	4.96	2	Harry ten Brink	ECN
	Speulderbos		rural	52.25	5.68	50	Harry ten Brink	ECN
Poland	Krakow	Nowa Huta	urban background	50.07	20.05	219	JP Putaud	JRC IES
Slovenia	Maribor T	Tabor	urban background	46.56	15.66	275	tanja.bolte(a)gov.si	Environmental Agency
	Maribor C	Centre	urban-traffic	46.55	15.68	275	benjamin.lukan(a)zzv-mb.si	Institute of Public Health
	Iskrba	EMEP/GAW st.	rural background	45.56	14.86	520	radojko.jacimovic(a)ijs.si	Josef Stephan Inst.
	Iskrba	EMEP/GAW st.	rural background	45.56	14.86	520	tanja.bolte(a)gov.si	Environmental Agency
Spain	Barcelona H	L'Hospitalet (18)	kerbside	41.37	2.12	12	X. Querol	IJA
	Barcelona S	Sagrera (3)	urban	41.4133	2.19	12	X. Querol	IJA
	Algeciras		urban background*	36.13	-5.54	1	X. Querol	IJA-UHU
	Los Barrios		near city*	36.18	-5.48	47	X. Querol	IJA-UHU
	Llodio		urban background*	43.14	-2.96	130	X. Querol	IJA
	Tarragona		urban background*	41.12	1.25	100	X. Querol	IJA
	Madrid		traffic	40.43	-3.68	667	X. Querol	IJA-CIEMAT
	Huelva		urban background*	37.26	-6.94	10	X. Querol	IJA-UHU
	Canarias	Las Palmas	urban background*	28.13	-15.41	30	X. Querol	IJA
	Monagrega		rural background	40.56	0.28	600	X. Querol	IJA
United Kingdom	London M	Marylbone	kerbside	51.52	-0.16	24	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	London B	Bloomsbury	urban	51.52	-0.13	24	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	Harwell		rural	51.57	-1.32	137	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	Glasgow		urban centre	55.86	-4.26	59	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	Belfast		urban centre	55.00	-5.93	81	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	Birmingham		urban centre	52.48	-1.91	134	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	Harwell		rural	51.57	-1.32	137	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	Manchester		urban centre	53.48	-2.24	89	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	London	Kensington	urban background	51.52	-0.21	24	r.m.harrison(a)bham.ac.uk	Univ. Birmingham
	Port Talbot		industrial	51.58	-3.76	1	r.m.harrison(a)bham.ac.uk	Univ. Birmingham

* with industrial influence

Annex 4. CN and PM annual average concentrations

Category	Site	Country	CN total #/cm³	CN ultrafine #/cm³	PM10 µg/m³	PM2.5 µg/m³
rural	Hyytiälä	(FI)	5766	4676		8.83
industrial	Port talbot	(UK)	14255		29.85	15.21
urban centre	Belfast	(UK)	25224		22.50	14.98
urban	Glasgow	(UK)	19438		21.67	14.26
urban	Manchester	(UK)	20761		28.30	16.89
urban	Birmingham	(UK)	17191		22.55	17.32
urban backgnd	London N	(UK)	19333		25.35	18.37
urban	London B	(UK)	23528		28.84	13.71
urban backgnd	Amsterdam	(NL)	23599	21076		23.51
kerbside	Helsinki V	(FI)	19576	14586	16.64	9.15
kerbside	London M	(UK)	78401		45.27	21.70
near-city	Ispra	(IT)	10079	5971	39.66	32.19

Annex 5. PM_{10} annual average chemical composition

Category	Site	Country	min.dust	sea salt	SO4	NH4	NO3	unacc.	OC	EC	TC	PM10
rural	Harwell	(UK)	NM	2.4	2.4	NM	3.1	7.2	2.0	0.7	2.7	18.6
rural	Speulderbos	(NL)	1.7	3.3	5.1	3.4	6.3	17.6	NM	NM	NM	37.4
urban backgnd	London N	(UK)	NM	3.2	2.7	NM	3.8	12.8	2.5	1.1	3.6	27.0
urban centre	Belfast	(UK)	NM	4.4	1.9	NM	2.1	10.5	2.4	1.3	3.7	23.6
urban	Duisburg	(DE)	4.3	1.6	5.4	2.4	5.3	12.2	4.1	2.6	6.8	32.9
kerbside	London M	(UK)	NM	2.9	3.6	NM	5.4	24.3	4.7	3.6	8.3	46.5
rural	Monagrega	(ES)	5.5	1.0	3.7	1.2	2.6	3.1	1.7	0.3	2.0	20.5
rural	Montelibretti	(IT)	NM	NM	4.1	NM	3.4	20.7	NM	NM	NM	28.2
near-city	Ispra	(IT)	1.6	0.8	4.0	2.7	5.2	10.2	8.9	2.0	10.9	39.7
suburban	Los Barrios	(ES)	6.4	4.4	4.7	1.3	2.4	2.9	NM	NM	3.5	26.8
urban backgnd	Canarias	(ES)	9.6	13.2	2.7	1.0	1.8	5.0	NM	NM	8.0	44.1
urban backgnd	Llodio	(ES)	7.7	3.9	5.2	1.5	2.4	6.6	NM	NM	7.3	37.3
urban backgnd	Tarragona	(ES)	9.7	4.1	4.7	1.4	3.6	3.7	NM	NM	7.6	37.4
urban backgnd	Huelva	(ES)	13.4	3.9	4.6	1.3	1.9	4.0	NM	NM	5.7	36.9
urban backgnd	Algericas	(ES)	7.5	5.6	4.6	1.4	3.4	8.7	NM	NM	5.1	38.2
urban backgnd	Barcelona S	(ES)	14.9	3.9	4.9	2.1	3.9	3.2	NM	NM	10.1	46.6
kerbside	Madrid	(ES)	15.6	1.2	4.9	1.2	2.1	2.0	NM	NM	16.2	48.8
kerbside	Barcelona H	(ES)	13.5	3.5	6.8	2.7	5.7	6.2	8.2	0.9	0.0	50.3
rural	Streithofen	(AT)	1.3	0.5	3.9	2.6	4.0	3.8	4.3	2.0	6.2	23.6
rural	Hortobágy	(HU)	3.6	0.1	6.7	NM	NM	16.9	NM	NM	NM	27.2
urban backgnd	Linz	(AT)	4.2	1.0	5.1	2.5	3.8	8.3	4.0	4.0	8.0	33.7
urban backgnd	Graz	(AT)	5.4	0.8	4.2	1.8	2.2	6.3	5.7	4.7	10.3	32.6
urban backgnd	Wien W	(AT)	3.5	0.8	4.6	2.5	4.0	4.1	5.8	3.5	9.3	30.4
urban backgnd	Praha	(CZ)	1.1	0.7	3.6	1.9	3.1	15.5	NM	NM	NM	25.9
urban backgnd	Debrecen	(HU)	5.3	0.1	4.8	NM	NM	20.8	NM	NM	NM	31.0
kerbside	Wien C	(AT)	NM	NM	NM	NM	NM	31.8	NM	4.1	NM	35.9

Concentrations in µg/m³
Unacc.: difference between PM mass and the sum of the quantified PM components (unaccounted mass)
NM: not measured

Annex 6. $PM_{2.5}$ annual average chemical composition

Category	Site	Country	min.dust	sea salt	SO4	NH4	NO3	unacc.	OC	EC	TC	PM2.5
rural	Harwell	(UK)	NM	NM	NM	NM	NM	NM	NM	NM	NM	10.5
rural	Speulderbos	(NL)	0.4	0.9	4.6	3.0	4.7	14.4	NM	NM	NM	NM
urban backgnd	London N	(UK)	NM	NM	NM	NM	NM	NM	NM	NM	NM	NM
urban centre	Belfast	(UK)	NM	NM	NM	NM	NM	NM	NM	NM	NM	NM
urban	Duisburg	(DE)	1.5	0.6	4.6	2.5	4.3	4.8	3.3	2.5	5.8	25.4
kerbside	Helsinki V*	(FI)	NM	0.3	4.6	NM	NM	7.8	NM	NM	NM	12.9
rural	Monagrega	(ES)	NM	NM	NM	NM	NM	NM	NM	NM	NM	NM
rural	Montelibretti	(IT)	NM	NM	NM	NM	NM	NM	NM	NM	NM	NM
near-city	Ispra	(IT)	0.6	0.4	4.2	2.6	4.3	6.4	9.8	2.0	11.8	32.2
suburban	Los Barrios	(ES)	NM	NM	NM	NM	NM	NM	NM	NM	NM	NM
urban backgnd	Canarias	(ES)	2.9	2.3	2.2	0.8	0.5	1.3	NM	NM	8.1	20.7
urban backgnd	Llodio	(ES)	2.6	2.1	4.8	1.6	0.7	3.1	NM	NM	7.9	25.5
urban backgnd	Tarragona	(ES)	2.3	1.2	4.0	1.7	1.8	2.0	NM	NM	7.5	23.1
urban backgnd	Huelva	(ES)	2.7	1.1	3.2	1.1	0.7	4.2	NM	NM	5.7	20.8
urban backgnd	Algericas	(ES)	2.9	1.5	4.0	1.5	1.1	8.2	NM	NM	4.5	25.3
urban backgnd	Barcelona S	(ES)	4.3	0.8	4.3	2.0	1.9	1.0	NM	NM	10.9	28.2
kerbside	Madrid	(ES)	7.0	0.6	4.6	1.7	1.5	2.6	NM	NM	16.5	39.4
kerbside	Barcelona H	(ES)	4.3	0.8	5.6	3.1	3.9	0.0	NM	NM	11.6	33.3
rural	Streithofen	(AT)	0.2	0.3	3.2	2.0	2.7	2.6	3.2	1.9	5.2	17.5
rural	Hortobágy	(HU)	1.2	0.0	5.8	NM	NM	11.0	NM	NM	NM	18.0
urban backgnd	Linz	(AT)	0.5	0.3	3.9	2.4	3.3	4.3	2.5	3.9	6.4	21.9
urban backgnd	Graz	(AT)	0.8	0.3	4.5	2.4	2.2	6.2	3.8	4.4	8.2	22.3
urban backgnd	Wien W	(AT)	0.5	0.3	4.1	2.2	2.8	2.6	4.0	3.7	7.8	21.8
urban backgnd	Praha	(CZ)	NM	NM	NM	NM	NM	NM	NM	NM	NM	NM
urban backgnd	Debrecen	(HU)	1.3	0.0	4.0	NM	NM	12.6	NM	NM	NM	18.0
kerbside	Wien C	(AT)	NM	NM	NM	NM	NM	NM	NM	NM	NM	NM

Concentrations in µg/m³
Unacc.: difference between PM mass and the sum of the quantified PM components (unaccounted mass)
NM: not measured

* no summer values for Helsinki

European Commission

EUR 23534 EN – Joint Research Centre – Institute for Environment and Sustainability

Title: The COST633 Aerosol Phenomenology - Physical and Chemical Characteristics of Particulate Matter at Rural, Urban and Kerbside sites compiled by the COST Action 633 "Particulate Matter: Properties Related to Health Effects"

Author(s): **J.P. Putaud** et al.

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Abstract

This report synthesizes the aerosol (=PM) physical and chemical data collected in the frame of the COST Action 633 (Particulate Matter: Properties Related to Health Effects) for rural, near-city, urban, and kerbside sites. It covers a large portion of Europe, divided in Northwestern, Southern and Central sectors. It is complementing a European aerosol phenomenology already published several years ago (Van Dingenen et al. 2004; Putaud et al., 2004). The data sets we collected include PM10 and/or PM2.5 mass concentrations, and aerosol particle number or PM chemistry data. This compilation aims at highlighting similarities and differences in aerosol characteristics across the COST633 network, beyond PM mass concentrations that mainly reflect pollution dilution. It enables to work on the question “why do equal PM10 mass concentrations lead to different health effects across Europe?”, and contributes to identifying synergies and trade-offs between policies related to climate change mitigation and air pollution abatement.

The mission of the JRC is to provide customer-driven scientific and technical support for the conception, development, implementation and monitoring of EU policies. As a service of the European Commission, the JRC functions as a reference centre of science and technology for the Union. Close to the policy-making process, it serves the common interest of the Member States, while being independent of special interests, whether private or national.

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