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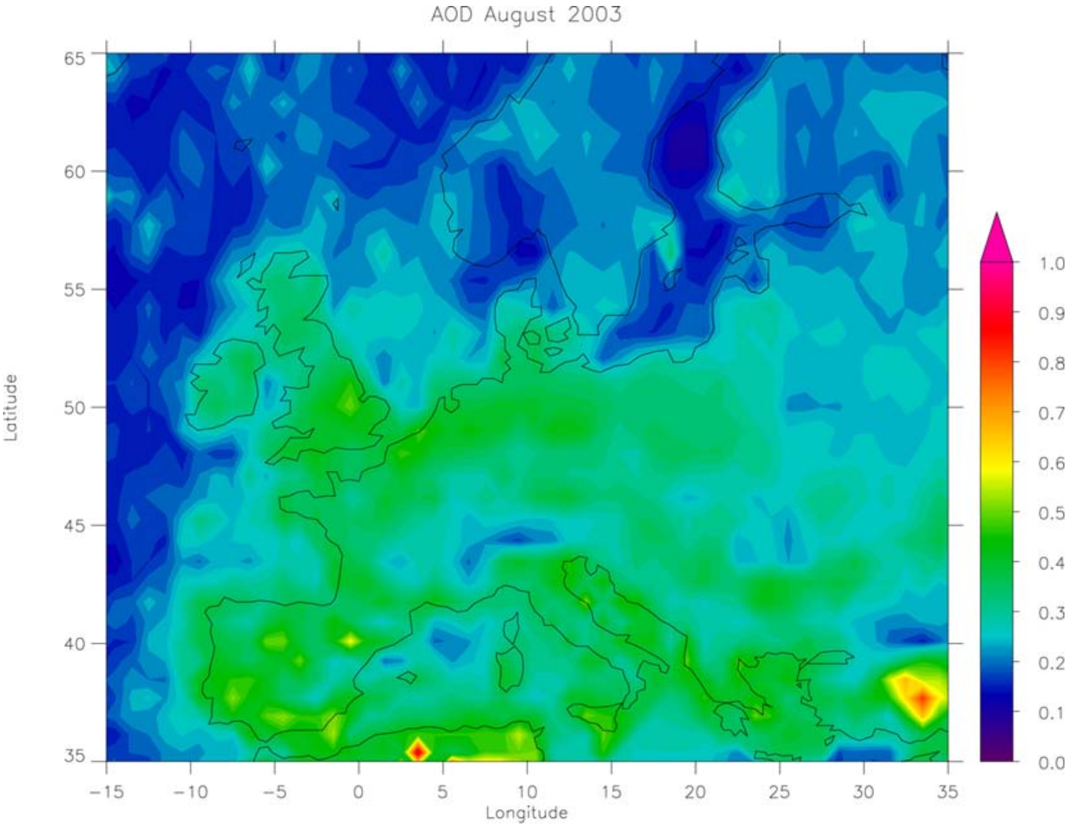
EMEP Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe

Measurements of Particulate Matter: Status Report 2006



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Executive summary

The present report provides an updated assessment of the particulate matter concentrations in 2004, and EMEP-related activities concerning particulate matter.

In 2004, 12 and 8 countries measured PM_{10} and $PM_{2.5}$ at their EMEP sites, which is two and one more country respectively than in 2003. In total there are 39 PM_{10} sites, 23 of these measure $PM_{2.5}$ as well. Furthermore, three of the sites reported concentrations of PM_1, which is one more than for 2003. Although the number of countries and sites that reports concentrations of particulate matter increase year by year, there are still large gaps in the geographical coverage. The EU annual limit value of 40 $\mu g\ m^{-3}$ of PM_{10} was not exceeded in any of the stations. The limit value for daily averages of PM_{10} was exceeded for more than 35 days at the Ispra station in Italy. The concentration level of PM was in general lower in 2004 compared to 2003, but there is no significant long-term trends over the period 1999-2004 for most of the EMEP stations, though the inter-annual variability of PM_{10} and $PM_{2.5}$ is large for this period. Different seasonal patterns for PM were observed at the individual stations. In Italy and Central Europe, the PM levels are generally higher during winter than in the summer, while in Spain the highest PM concentrations are during summer and spring. In Scandinavia, the PM concentrations are highest during spring, August, January and December. The sites in central and northern Europe generally experience their highest concentrations associated with air masses from south-eastern and southern directions, representing significant source areas of atmospheric emissions.

Furthermore, data from the EMEP network and the AIRBASE database have been examined with the objective to compare urban and rural PM concentration levels in Europe. As expected, higher PM concentrations were observed at urban sites compared to rural sites. It is interesting to note that annual average concentrations of PM_{10} at several countries exhibit the same temporal variation for the period 1997-2004 at urban, rural and traffic stations. A more detailed investigation regarding this issue has been undertaken for the Swiss network (NABEL) where extensive long-term parallel measurements of PM_{10}, $PM_{2.5}$ and PM_1 from sites of various categories have been conducted. At all sites, the daily concentrations of the different size fractions were highly correlated. Unless strong and variable local sources of coarse particles are present, parallel measurements of PM_1, $PM_{2.5}$ and PM_{10} seem to provide only limited additional information. Therefore, parallel measurements can be restricted to a few carefully selected sites in a monitoring network. Also the correlation of daily PM values from sites at some distance often shows quite high correlation. The analysis indicates that this primarily has meteorological reasons. Even distant sites show good correlations if they are situated in an area with similar meteorological conditions.

Chemical speciation of the particulate matter shows that the major compounds contributing to the mass concentrations are sulphate, nitrate and ammonium. Sulphate contributes between 9-23% of the PM_{10} while nitrate and ammonium between 3-16% and 3-8% respectively. Base cations (K, Ca) are measured at three sites only but the contribution to the PM_{10} is small, below 2%. But these results are not representative for southern Europe. Sea salts (mainly Na, Cl, Mg) may in coastal areas contribute significantly to the particulate mass concentrations. Most EMEP sites are however located at some distance from the coast to avoid direct contributions from sea-spray. Still only few sites report sea salt concentrations; on the regional scale this contribution is typically lower than 2% for inland site. In more costal areas, the contribution is higher, i.e. 15% at the Birkenes site (NO0001). In total, the inorganic fraction contributes between 20–60% of the PM_{10} mass across Europe.

The remaining aerosol mass mainly consists of carbonaceous compounds and water. The measurements of carbonaceous matter are still very scarce, and a complete chemical speciation is therefore only possible at a couple of sites. In 2004, the carbonaceous fraction was a major contributor to PM_{10} both at Birkenes (28%) and at Ispra (47%).

Different sampling approaches and analytical methods are used for measuring carbonaceous matter, and this hampers the comparison of EC/OC data. Due to artefacts related to sampling and analysis of elemental carbon (EC) and organic carbon (OC), it is difficult to use the monitoring data to establish a reliable picture of the air pollution situation in Europe. This is a challenge not only troubling EMEP but the entire scientific community, thus development of standardised protocols for sampling of OC and analysing EC and OC is of a high priority. Developing, validating and establishing a standardized protocol for sampling of OC and analysis of EC and OC is one of the main focus areas within the EU funded EUSAAR (*European Supersites for Atmospheric Aerosol Research*) project. Recent results from this project shows that the positive artifact may account for 10 – 50% of the total carbon (TC) at the EMEP site Ispra during winter, and that the positive artifact is most severe during less polluted periods. Testing of a sampling prototype has provided positive results, reducing the positive artifact down to 2 – 10%. If correspondingly positive results are obtained for all seasons, the sampling train will be proposed for validation by the end of 2006. Further, the analytical method is also being improved to achieve a high quality split between OC and EC in particulate matter samples. Interim recommendations for EC/OC sampling and analysis is given while awaiting the final protocol from EUSAAR.

Data from the EMEP EC/OC campaign reported a North-to-South gradient for EC, OC and TC in PM_{10} for Europe, with the higher concentrations reported in the central eastern and southern regions of Europe compared to the northern and western regions. On an annual basis, the EC fraction by mass of PM_{10} was 3.6 ± 1.2%, whereas the corresponding percentage for the OM (Organic Matter) fraction was 26 ± 8%. Without exception, levels of EC were higher in winter than in summer. A similar seasonal cycle was observed for OC, with the exception of Scandinavia, where concentrations were 1.5 times higher in summer compared to winter.

Birkenes is the only EMEP site with a time series of five years for EC, OC and TC. From 2001 to 2005, the concentrations of OC at Birkenes dropped by 36% for $PM_{2.5}$ and 10% for PM_{10}. For the same period, a substantial increase of 164% was reported for coarse OC. Unlike OC, the time series for EC was less clear. At Birkenes, the time series of TC and OC in PM_{10} and $PM_{2.5}$ resembles that of PM_{10}, $PM_{2.5}$ and SIA. At Ispra, the level of TC in PM_{10} and $PM_{2.5}$ during 2003-2004 was approximately nine times higher than for Birkenes during 2001-2005. The two sites are in the opposite ends of the concentration range with respect to TC (and EC and OC) in Europe.

Two chapters of this report are dedicated to PM measurements that are regarded as level three activities according to the EMEP-monitoring strategy. One chapter focuses on optical properties of aerosols in the European Arctic (Ny-Ålesund at Svalbard) and sub-Arctic region (ALOMAR research station situated at the northern Norwegian coast), and in particular on ground-based measurements of aerosol optical depth (AOD). The AOD measurements are discussed with respect to seasonal variation, the inorganic content of the aerosols, and transport of air masses into the region. The results presented for 2005 show elevated AOD levels both at Ny-Ålesund and at ALOMAR during the Arctic haze period in the spring. Further, three episodes with elevated levels of AOD during the summer months were investigated. The chemical content of the aerosols and the transport pattern of the air masses entering the region during these episodes, showed that the increased AOD values during one of the episodes most likely are explained by transport of black carbon emitted from boreal fires in North America. The other episodes seem to be explained by a variety of emissions in central Europe and there also seemed to be a small contribution from Asia.

The other level 3 activity chapter addresses measurements of particulate matter from satellite borne instruments. Monthly maps of Aerosol Optical Depth (AOD) on a 10 km x 10 km grid, retrieved from the AATSR (ESA) instrument by TNO, showed that elevated AOD values can be observed both above Western and Eastern Europe, although typically not in same months. The dataset is able to capture large-scale spatial and temporal variations in aerosol load, while cities and industrial areas only occasionally turn up in the maps. By the combined effort of the radiometer AATSR and the spectrometer SCIAMACHY, both onboard ENVISAT, ambient air concentrations of PM_{10}, $PM_{2.5}$ and PM_1 were calculated by the DLR, using the SYNAER algorithm, for five selected EMEP stations (Birkenes (NO0001), Illmitz (AT0002), Ispra (IT0004), Payerne (CH0002) and Zarra (ES0012)] for four months during 2003. Taking into account that a snapshot from the satellite is compared with a daily average from the station, the methods compare relatively well. It was found that SYNAER overestimates PM_{10} and $PM_{2.5}$ on certain occasions, especially at Zarra, whereas it seems to underestimate PM_{10} and $PM_{2.5}$ for locations surrounded by high mountains, such as Payerne.

Data from the current space-borne satellites are not alone able to monitor levels of air pollution by aerosols over Europe. The accuracy of the data are probably sufficient or are within reach to become so by ongoing algorithm developments, but temporal data coverage is not sufficient to properly monitor the regions of interest for EMEP. The satellite retrievals are however, complementary to groundbased measurements due to its ability to measure over oceans and remote areas. This monitoring is essential for the Task Force on Hemispheric Transport of Air Pollutants and satellite data such as AOD from AATSR and PM data from SYNAER are needed to realise the goals of this activity.

Contents

Measurements of Particulate Matter: Status Report 2006

1. Measurements of particulate matter (PM_{10}, $PM_{2.5}$ and PM_1) in 2004

Mihalis Lazaridis, Ilias Kopanakis, Victoria E. Cachorro, Karl Espen Yttri and Wenche Aas

1.1 Introduction

Airborne particulate matter (PM) is a complex mixture of many different chemical species originating from a variety of sources. Composition, morphology, physical and thermodynamic properties of PM vary with respect to time and location and quite typically have a seasonal variability (IPCC, 2001; Finlayson-Pitts and Pitts, 1986; Seinfeld and Pandis, 1998; EPA, 2003; CAFE, 2004; Yttri, 2005).

There is a considerable literature concerning patterns and trends of ambient aerosols and their concentration characteristics. The interested reader is referred to the following scientific books, papers, and reports (Seinfeld and Pandis, 1998; CAFE, 2004; Putaud et al., 2004; Van Dingenen et al., 2004; Yttri, 2005; EPA, 2003). In Europe, PM measurements are performed mainly in urban areas for the purpose of monitoring human PM exposure. Monitoring of PM in rural areas is not as extensive as seen for urban areas. Despite this, results presented in the recent CAFE (2004) for the period 1997 to 2001 show that the gap between concentrations of PM_{10} at rural and urban sites in Europe can be quite narrow. In addition, the annual variation of the PM concentrations for these two site categories is similar for the period 1997 to 2001. Hence, the regional contribution seems to be highly important for the concentrations of PM_{10} observed in urban areas. Also the level of PM_{10} at traffic-influenced sites in Europe follows the annual variation observed for urban and rural areas, although its concentration level is considerably higher.

The Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) is a regional policy-driven monitoring programme under the Convention on Long-Range Transboundary Air Pollution. The objectives of the EMEP monitoring programme are to provide data on pollutant concentrations over Europe, their deposition, emission, composition and transport (transboundary fluxes). Another objective is to provide a better understanding of the processes that affect the concentration of pollutants in relation to their effects on ecosystems and human health in order to support the development of cost effective abatement strategies.

Monitoring of aerosol particles within the framework of the EMEP Programme was included in 1999. The main goal of this activity within EMEP is to monitor long-range transport of aerosols in Europe and its long-term trends. The motivation for including long term particulate matter measurements in the EMEP programme was the need of adoption and validation of abatement strategies due to their effects on the environment (Andreae and Crutzen, 1997; Charlson et al., 1992; Tegen et al., 1997) and the association of exposure to particles through inhalation to adverse effects on human health by many epidemiological and toxicological studies (e.g. Pope et al., 1995; Schlesinger, 1995; Neuberger et al., 2004). More specifically, health effects caused by inhaled particles are pulmonary function decrements, respiratory symptoms, neurological dysfunction, cardiovascular dysfunction, morbidity and mortality (Rombout et al., 2000; EPA, 2003; Pope et al., 1995; Dockery et al., 1993; Schwartz, 2000; Pope and Dockery, 2006). Therefore, measurements of PM_{10}, $PM_{2.5}$, and recently PM_1 are conducted at selected EMEP stations, the number of which increases every year. Nevertheless, at present, the number of measurement sites reporting PM concentrations in Europe does not allow a comprehensive geographical overview of the rural PM levels. Especially in eastern Mediterranean region the site distribution is scarce. This part of Europe is situated on the outskirts of the domain area of the EMEP model, and receives a significant part of its particulate matter loading from outside Europe.

In this chapter, monitoring data of ambient particulate matter from the available measurements during the year 2004 are presented. An interpretation of the annual and seasonal trends of particulate matter concentrations has been performed and the effect of the air mass trajectories on the concentrations measured at each station is examined. Specific examples are provided from the current (measurements by the end of the year 2004) status of the EMEP monitoring work. Finally, the relative contribution of inorganic constituents to the aerosol mass is presented in brief as well as possible correlations with the particulate mass. The carbonaceous content of the aerosols is addressed in a separate chapter (Chapter 3).

1.2 Data availability and methods

In addition to the data reported to EMEP, data from the AIRBASE database for the year 2004 are presented mainly for the purpose of comparing urban/street PM concentrations with rural PM data. The European Air Quality monitoring Network (EuroAirnet) has been developed in close cooperation between the European Environmental Agency (EEA) and the European countries. Data from EuroAirnet are reported to AIRBASE (http://etc-acc.eionet.eu.int/databases/airbase.html), a database managed by the European Topic Centre on Air Quality and climate change under contract to EEA. The AIRBASE includes data on gaseous pollutants and particulate matter concentrations from stations in Europe reported by countries to EEA (European Environmental Agency) (Larssen and Lazaridis, 1998; Larssen and Hagen, 1999). The stations included in the AIRBASE database are classified into three main station types, traffic, background and industrial, and according to the type of the area where they are situated, into urban, suburban and rural (Larssen et al., 1999). For the rural background sites reported to AIRBASE there are some overlaps with the EMEP sites. The EMEP siting criteria (EMEP/CCC, 1996) is stricter than the definition of rural background sites, which might be situated relatively close to local emission sources. These groups of sites are therefore not directly comparable to the EMEP site. However if one select out those sites defined as EMEP, also for other components than PM, it should in principle be comparable. A few countries report PM data from some EMEP sites only to AIRBASE. It might be a good reason for that, since a site can be rural for some components but not for others. It may also be that the methodology used is not recommended by EMEP. This is probably the case for several Austrian and German EMEP sites where only a subset of their PM measurements in rural background are reported to EMEP. For other sites, like in GB, FI and CZ it can be lack of reporting routines for PM measurements to EMEP. We have assigned those sites defined as EMEP sites for other components (e.g. ozone or inorganic components) and report PM data to AIRBASE but not to CCC as EMEP2.

Different sampling methods for measuring mass concentrations are used. Most of the EMEP sites are using gravimetric methods, but e few is also using automatic systems like TEOM (tapered element oscillating microbalance), i.e. in SE and CY, on b-attenuation in GR. The measurements reported to AIRBASE are usually automatic methods. The different methods can give substantial systematic biases do to both positive and negative artefacts. However, each country has to provide equivalent measurements to gravimetric methodology and standards. Details on methods and equipment for particulate matter measurements used at the EMEP stations for 2005 can be found in Fjæraa (2006), as well as on the web pages.

The number of countries that reported PM_{10} mass concentrations directly to EMEP increased to 12 in 2004. Cyprus (CY0002) and Greece (GR0002) are the new countries reporting PM_{10} concentrations in addition to the 10 countries reporting the preceding years. PM_{10} data was reported directly to EMEP for 39 stations, whereas data for 23 more EMEP stations in five countries was retrieved from the AIRBASE database, these are defined as the EMEP2 dataset mentioned above. For 2004, there were eight countries reporting levels of $PM_{2.5}$ for 23 sites. There were two new stations reporting $PM_{2.5}$ in 2004, namely the Iskrba station in Slovenia (SI0008R) and the Zoebelboden in Austria (AT0048R). For 2004, PM_1 data was added for one more site, namely Langenbrügge (DE0002R) in Germany.

A significant fraction of the ambient particulate matter is secondary particulate matter in the form of sulphate, nitrate, ammonium and organic aerosol particles formed by the oxidation of sulphur dioxide, nitrogen oxides and organic gaseous species. Daily measurements of inorganic constituents in aerosols are performed within the EMEP framework but mainly sulphuric and nitrogen species, very little measurements of base cations and sea salts in air are measured in Europe. This is also the case for carbonaceous material that is only reported for a very few sites in EMEP. For 2004, only IT0004R and NO0001R reported levels of EC and OC, Chapter 3.

1.3 PM mass concentrations and exceedances

The annual mean concentrations of PM_{10}, $PM_{2.5}$ and PM_1 for 2004 are presented in Table 1.1, whereas the spatial coverages of PM_{10} and $PM_{2.5}$ are presented in Figure 1.1. Sites with poor data capture are presented in the table with a note of the time period they are valid. These data are not included in the maps.

The EMEP sites reporting concentrations of ambient air particulate matter to EMEP and/or AIRBASE are not uniformly distributed across Europe. Of the 62 stations reporting concentrations of PM_{10} in 2004, 32 were concentrated in Central Europe, 16 in Southern Europe, 7 in Northern Europe and 7 in Western

Europe. No PM_{10} measurements are reported for EMEP stations in Eastern Europe for 2004, although it can be argued that Slovakia is part of Eastern Europe as well as Central Europe.

Table 1.1: Annual mean concentrations of PM_{10}, $PM_{2.5}$ and PM_1 at EMEP sites for 2004 (concentrations in $\mu g/m^3$).

Code	PM_{10}	$PM_{2.5}$	PM_1	Code	PM_{10}	$PM_{2.5}$	PM_1
AT0002R	24.5	19.1	14.0	DK0005R	20.3		
AT0004R	*12.6^*	*Jan - March*		DK0041R*	22.0		
AT0005R	10.4			ES0007R	24.4	11.1	
AT0030R*	23.0			ES0008R	16.4	9.6	
AT0040R*	13.9			ES0009R	13.3	8.4	
AT0042R*	20.9			ES0010R	21.1	12.8	
AT0043R*	16.3			ES0011R	18.6	10.7	
AT0047R*	20.6			ES0012R	17.2	8.3	
AT0048R	10.9	9.2		ES0013R	13.3	8.5	
BE0033R*	32.1			ES0014R	22.3	12.6	
CH0002R	20.0	14.9		ES0015R	15.9	8.2	
CH0003R	19.4			ES0016R	13.3	9.1	
CH0004R	11.2	8.1	6.5	FI0007R*	13.2		
CH0005R	11.9			GB0006R*	10.4		
CY0002R	29.9			GB0036R*	18.6		
CZ0001R*	22.0			GB0043R*	13.0		
CZ0003R*	26.1			*GR0002R*	*23.2^*	*Sept - Dec*	
DE0002R	17.5	13.3	7.5	IT0001R	29.0		
DE0003R	10.4	7.2		IT0004R	34.7	28.3	
DE0004R	*18.1^*	*14.0^*	*Jan -July*	MK0007R*	15.8		
DE0005R	*12.3^*	*Jan -July*		NL0007R*	25.6		
DE0007R	13.8				24.6		
DE0008R	10.3			NL0010R*	24.3		
DE0009R	15.2			NO0001R	5.3	3.3	
DE0012R*	17.7			SE0011R	13.7	9.8	
DE0016R*	22.3			SE0012R	10.5	7.0	
DE0017R*	31.4			SE0035R	7.8	*4.4^*	*Jan -April*
DE0026R*	13.5			*SI0008R*	*14.4^*	*12.8^*	*Nov/Sept -Dec*
DE0035R*	14.4			SK0004R	13.7		
DE0039R*	20.1			SK0005R	17.9		
DE0041R	19.1			SK0006R	16.4		

*: Data obtained from the AIRBASE database, but the stations are defines as EMEP for other components, EMEP2

^: Data capture < 50%; data given in italic

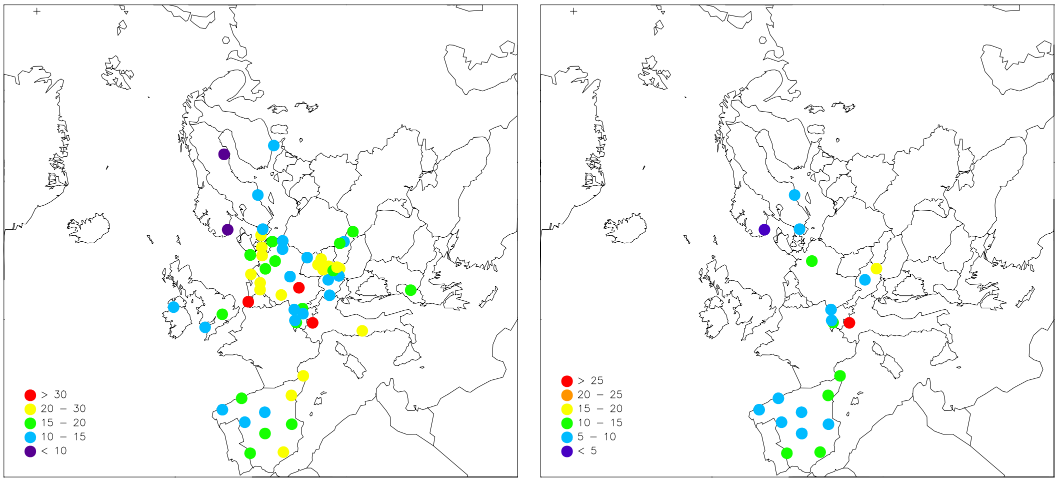

Figure 1.1: Annual mean concentrations of PM_{10} (left) and $PM_{2.5}$ for 2004 ($\mu g/m^3$).

From Figure 1.1 it can be seen that stations in the Central and the continental part of Western Europe have higher annual mean concentrations of PM_{10} than stations in Great Britain and in the Scandinavian countries during 2004. Country annual mean PM_{10} concentrations ranged from 5.3 $\mu g/m^3$ for Norway to 32.1 $\mu g/m^3$ for Belgium. The high levels can be attributed to the local anthropogenic emissions (traffic, industries) in Western and Central Europe whereas for Southern Europe this can rather attributed to biogenic emissions, resuspended dust from local sources and also Saharan dust events. A similar spatial distribution of PM_{10} concentrations for Europe was observed during 2003. During 2004, the annual limit value of 40 $\mu g/m^3$ of PM_{10} has not been exceeded in any of the EMEP stations.

The majority of the sites reporting $PM_{2.5}$ concentrations during 2004 were concentrated in Spain, Central and Western Europe. From Figure 1.1 it can be seen that the annual mean concentrations of $PM_{2.5}$ increased from northern to southern parts of Europe, and that the highest concentrations were reported for the two central European sites Ispra (IT0004R) (28.3 $\mu g\ m^{-3}$) and Illmitz (AT0002R) (19.1 $\mu g\ m^{-3}$). Hence, the US EPA annual limit value of 15 $\mu g\ m^{-3}$ was exceeded at both these sites. During 2003, four stations exceeded the US EPA annual limit value, and the maximum concentration of 28.6 $\mu g\ m^{-3}$ was reported for Ispra, as for 2004. The annual mean concentration of PM_1 did not exceed 15 $\mu g\ m^{-3}$ at any of the sites stations reporting this parameter in 2004.

The number of days for which the daily PM_{10} limit value was exceeded during 2004 is presented in Table 1.2, showing that most exceedances are observed at stations in Austria, Belgium, the Czech Republic, Cyprus, Italy, the Netherlands and Spain. The Italian site Ispra was the only site violating the daily limit value for PM_{10} (50 $\mu g\ m^{-3}$ > 35 days) for 2004. However, a decline in the number of exceedances was observed for 2004 (71 days) compared to 2003 (87 days) for this site. In fact the daily limit value was exceeded on fewer days in 2004 than in 2003 for the majority of the stations. This decline was particularly obvious for the Austrian site Illmitz (AT0002R), where the number of exceedances was reduced

Table 1.2: Number of exceedances of the daily PM_{10} limit value (50 µg/m^3) during 2004.

	Code	Number of exceedances of the daily limit value for PM_{10}				
		Total	*Winter*	*Spring*	*Summer*	*Autumn*
Central Europe	AT0002R	28	17	4	1	6
	AT0004R (Jan – 20. Mar)	*1*		*1*		
	AT0005R	1		1		
	AT0030R*	22	14	5		3
	AT0040R*	2		1		1
	AT0042R*	2	1	1		
	AT0043R*	0				
	AT0047R*	2	2			
	AT0048R	1		1		
	CH0002R	9	7	2		
	CH0003R	7	3	4		
	CH0004R	0				
	CH0005R	0				
	CZ0001R*	10	1	5		4
	CZ0003R*	20	8	8		4
	DE0002R	5	3	2		
	DE0003R	2	1	1		
	DE0004R (Jan – 1 July)	*2*		*2*		
	DE0005R (Jan –1 July)	*0*				
	DE0007R	2	2			
	DE0008R	0				
	DE0009R	5	3	1		1
	DE0012R*	6		6		
	DE0016R*	12				
	DE0026R*	5	5			
	DE0035R*	3	2		1	
	DE0039R*	6	5	1		
	DE0041R	6	1	5		
Southern Europe	CY0002R	28	7	8	2	11
	ES0007R	25		3	14	8
	ES0008R	1			1	
	ES0009R	10		4	1	5
	ES0010R	1	1			
	ES0011R	8		3	5	
	ES0012R	9		2	5	2
	ES0013R	4		2	2	
	ES0014R	15	5	2	2	6
	ES0015R	10		3	4	3
	ES0016R	2		1	1	
	GR0002R (Sept – 31 Dec)	*4*				*4*
	IT0001R	22	10	4	7	1
	IT0004R	71	43	13	1	14
	SI0008R (Nov – 31 Dec)	*0*				
	MK0007R*	1				1
Western Europe	GB0006R*	0				
	GB0036R*	0				
	GB0043R*	0				
	BE0033R*	46	21	15		10
	NL0007R*	19	8	9	2	
	NL0009R*	15	4	11		
	NL0010R*	25	14	10		1
Northern Europe	DK0005R	3		3		
	DK0041R*	10	4	4	1	1
	NO0001R	0				
	SE0011R	0				
	SE0012R	0				
	SE0035R	0				
	FI0007R*	0				

from 49 days in 2003 to 28 days in 2004. The sites experiencing an increased number of exceedances in 2004 compared to 2003 were mainly situated in Spain. Especially, the exceedances experienced at the Viznar station (ES0007R) during 2004 are associated with southern air masses, which suggest frequent intrusions of

Saharan dust. Table 1.2 also shows the number of exceedances for each station with respect to season. Although a common seasonal pattern cannot be associated to all the EMEP stations in each part of Europe, some common features are observed for stations with more than 15 exceedances. In particular, there is a prevalence of exceedances during winter in Central Europe and Italy. In Netherlands exceedances are observed during winter and spring. In addition, examination of PM_{10} data for the period 1997-2004 for the stations AT0002R, AT0047R, CH0002R, CH0003R, DE0007R, IT0001R and IT0004R, depicted the prevalence of exceedances during winter and/or spring. The above remarks indicate that exceedances at stations in Central Europe and Italy are possibly associated with enhanced emissions from anthropogenic sources during winter. On the other hand, exceedances of the daily limit value at the Spanish sites occur mainly during the summer and autumn during the period 2001-2004 due to dry conditions, which lead to resuspension of dust.

Annual mean concentration ratios of $PM_{2.5}$-to-PM_{10}, PM_1-to-PM_{10} and PM_1-to-$PM_{2.5}$ during 2004 are summarized in Table 1.3, showing that stations in central Europe and Italy have a rather high $PM_{2.5}/PM_{10}$ ratios compared to stations in Spain and Northern Europe. This difference is probably associated with the high contribution of anthropogenic emissions to PM_{10} concentrations in central Europe. In addition, the PM_{10} concentration in Spain is (considerably) influenced by resuspended dust (and biomass burning emissions), whereas for certain Scandinavian sites the influence by marine aerosols (sea-salt) can be important. Hence, this could be possible explanation for why a larger fraction of the particles in Spain and in the Scandinavian countries is found in the $PM_{2.5-10}$ fraction. However, there are exceptions to this general spatial pattern as observed for the Vavihill (SE0011R) station in Sweden and the O Savinao (ES0016R) station in Spain where the $PM_{2.5}$-to-PM_{10} ratios were 0.72 and 0.68, respectively.

For the three sites measuring PM_{10}, $PM_{2.5}$ and PM_1 (AT0002, CH0004R and DE0002R), there are only small variations in the $PM_{2.5}$-to-PM_{10} ratio reported, whereas a considerably lower PM_1-to-$PM_{2.5}$ ratio, and hence PM_1-to-PM_{10} ratio, is observed for the DE0002R site, compared to the two other sites. A detailed discussion on the PM mass concentration data in the EMEP stations is addressed in chapter 1.6

Table 1.3: PM concentration ratios during 2004.

		Code	$PM_{2.5}/PM_{10}$	PM_1/PM_{10}	$PM_1/PM_{2.5}$
Central Europe	Austria	AT0002R	0.77	0.57	0.74
		AT0048R	0.84		
	Switzerland	CH0002R	0.75		
		CH0004R	0.72	0.58	0.80
	Germany	DE0002R	0.76	0.43	0.56
		DE0003R	0.69		
Southern Europe	Spain	ES0007R	0.45		
		ES0008R	0.59		
		ES0009R	0.63		
		ES0010R	0.61		
		ES0011R	0.58		
		ES0012R	0.48		
		ES0013R	0.64		
		ES0014R	0.57		
		ES0015R	0.52		
		ES0016R	0.68		
	Italy	IT0004R	0.82		
Northern Europe	Norway	NO0001R	0.62		
	Sweden	SE0011R	0.72		
		SE0012R	0.42		

1.4 Comparing the EMEP and the AIRBASE networks

PM_{10} concentrations reported for the EMEP rural background stations were compared with concentrations of PM_{10} measured at other stations in Europe retrieved from the AIRBASE database. The data presented in Figure 1.2 to Figure 1.3 were obtained by averaging concentrations of PM_{10} for all stations of each station category, or area category, for the countries that reported PM_{10} data either to the AIRBASE or the EMEP database.

From Figure 1.2 it can be seen that the PM_{10} concentration was higher at traffic stations and at stations situated in urban areas compared to rural sites and the EMEP1 and EMEP2 sites in all countries where this comparison were possible. The annual limit value of 40 µg m^{-3} was exceeded at 208 of 1801 stations included during 2004. Most of these were background (52) and traffic (92) sites situated in urban areas, particularly in Spain (51) and Italy (49). The annual limit value for PM_{10} was also violated at seven rural background stations. Concentrations of PM_{10} measured at EMEP sites were typically lower than concentrations reported for background, industrial and traffic sites situated in rural areas, for all countries assessed (Figure 1.3). The only exceptions were seen for Spain and Italy where the annual mean concentrations of PM_{10} at the EMEP stations were higher than those at the traffic influenced rural sites and the background sites in rural areas, respectively. For Spain this is probably associated with Saharan dust intrusions, whereas in Italy it is due to the high concentrations measured at the Ispra station located at the northern part of the country.

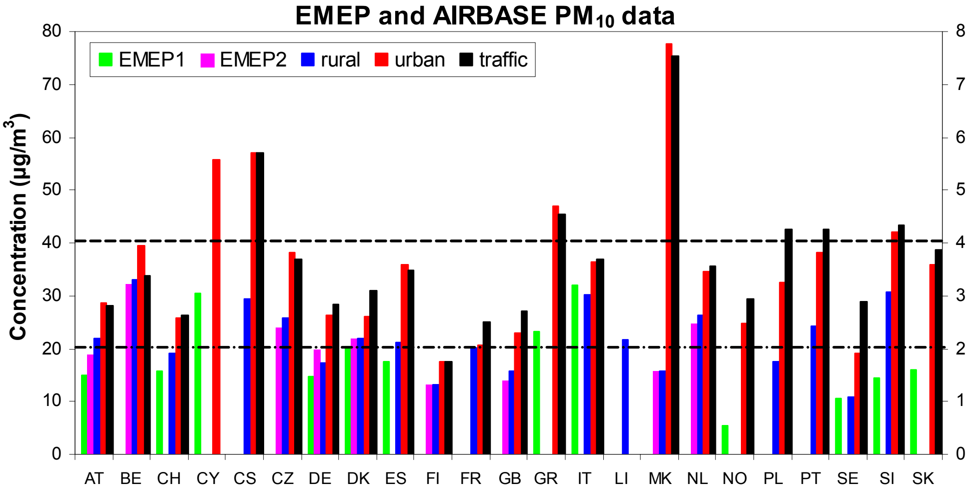

Figure 1.2: Annual mean concentrations of PM_{10} in European countries for 2004, based on data from EMEP and AIRBASE.
EMEP1: Data from EMEP sites reported to EMEP database.
EMEP2: Data from EMEP sites, but only reported to AIRBASE.

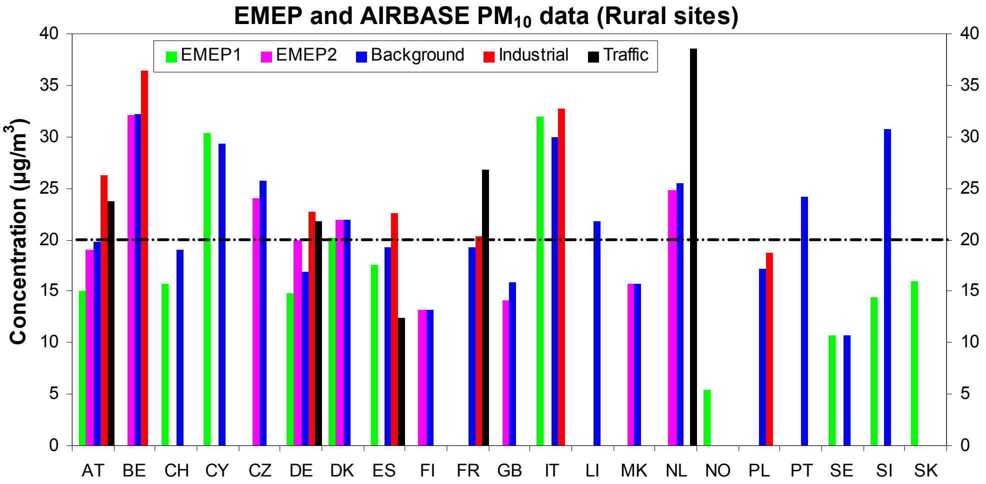

Figure 1.3: Annual mean concentrations of PM_{10} at rural areas of European countries for 2004, based on data from EMEP and AIRBASE.
EMEP1: Data from EMEP sites reported to EMEP database.
EMEP2: Data from EMEP sites, but only reported to AIRBASE.

It is also observed that EMEP2 stations experience higher concentrations than stations reporting PM_{10} data directly to EMEP and rural stations in each country. This is because some of these background stations are situated in urban and suburban areas (e.g. stations in Belgium) rather than rural. The concentrations at these stations were in fact higher than the annual mean concentration at industrial and background suburban stations in Finland and the Netherlands.

Further, the temporal variation of the annual mean concentration of PM_{10} at the EMEP stations were compared with that reported for traffic stations and stations situated in urban and rural areas for each country. It is observed that the annual mean concentrations of PM_{10} at EMEP stations exhibit the same variability over time as traffic sites and sites situated in urban and rural areas for several countries. This finding is exemplified in Chapter 2 for Switzerland, but it was also obvious for the countries Austria, Germany, the Czech Republic and the United Kingdom.

1.5 Annual Trends for Particulate Matter Mass

For 2004 the concentration of PM_{10} was found to be higher at only 11 sites compared to 2003. Measurements for more than four years have been conducted at 43 of the 62 EMEP sites listed in Table 1.1. Annual trends have been examined for the 26 sites, which reported PM mass concentrations directly to EMEP for four full years. Statistical significant trends were observed for four of stations, namely DE0001R, DE0004R, ES0016R and IT0004R. The trends have been calculated using the MAKESENS tool developed by FMI (Salmi et al., 2002).

At the Deuselbach (DE0004R) station in Germany the annual mean concentration of PM_{10} exhibited an increasing trend for the period 1999 – 2003, whereas for $PM_{2.5}$ an increasing trend was observed for the period 2000 - 2003 (Figure 1.4). The annual mean concentrations for 2004 have been excluded from the analysis because the data capture for this year was less than 40%. Nevertheless, the annual mean concentration of $PM_{2.5}$ slightly increased for 2004 compared to 2003, whereas the PM_{10} concentration for 2004 was lower than for 2003. For the period 1999–2003 PM_{10} concentrations at background and industrial sites in rural and suburban areas in Germany (data retrieved from AIRBASE) exhibited an increasing trend and were correlated with the values measured at Deuselbach. This might indicate that the variations in PM_{10} concentration at Deuselbach are attributed to regionally transported PM.

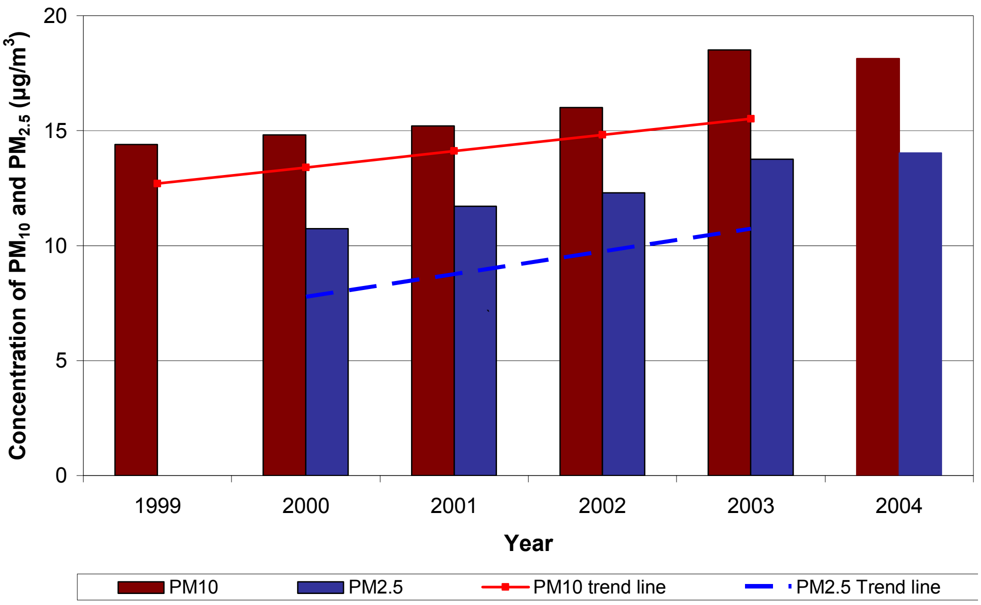

Figure 1.4: Time trends for annual mean concentrations of PM_{10} and $PM_{2.5}$ at the Deuselbach (DE0004R) station, in Germany, for the period 1999-2004. Trends calculated for 1999-2003.

Negative trends were observed for the $PM_{2.5}$ concentrations at the O Savinao station in Spain (Figure 1.5) and the Ispra (IT0004R) station in Italy (data not shown) for the period 2001–2004.

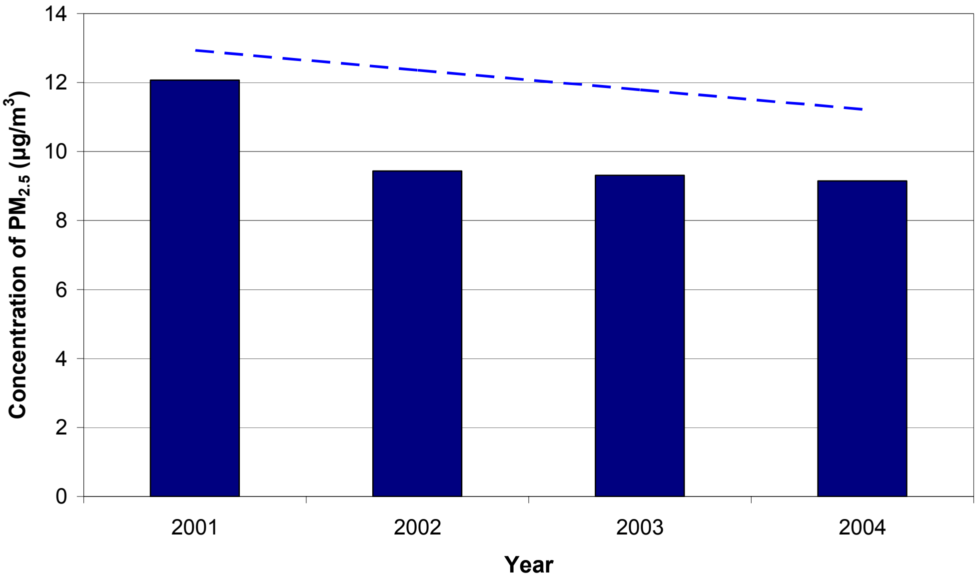

Figure 1.5: Time trends for annual mean concentrations of $PM_{2.5}$ at the O Savinao (ES0016R) station in Spain for the period 2001–2004.

At the other EMEP sites the annual average concentrations of PM_{10} and $PM_{2.5}$ typically had a large variability and no significant trends have been observed.

1.6 Analysis of time series for PM mass concentrations at EMEP stations

In this section time series of PM concentrations are explored with respect to common temporal patterns in countries or regions of Europe. For those sites that reported directly to EMEP, timeseries of daily and monthly mean concentrations of PM_{10} and $PM_{2.5}$ are reported in Appendix 1 and Appendix 3, respectively. In addition, the effect of the air mass trajectories reaching each station on the measured PM concentrations has been examined. The 2D Trajectories from the EMEP/MSC-W available at http://www.emep.int/Traj_data/traj2D.html have been used in the analysis. Available data include calculated 96h trajectories, daily sector values and trajectory crossings for all EMEP stations from 1985 to 2004. The aim of this analysis was to explain the observed variations of PM mass concentrations and PM concentration ratios, presented in section 1. Thus, results for selected stations, representative for each part of Europe, are presented. The influence of specific PM chemical components on the spatial and temporal patterns of PM concentrations is discussed in chapter 1.7.

1.6.1 Temporal patterns of PM concentrations at EMEP stations

A seasonal pattern has been observed for a number the EMEP stations where PM concentrations were increased during the summer and were lower during the winter period or the opposite. More specifically, PM concentrations have been higher during the winter period at stations in Central Europe and Italy. There is a tendency that sites reporting elevated PM levels in these regions experience elevated PM concentrations during winter. Figure 1.6 presents as an example data of mean monthly PM_{10} concentrations for the Tänikon station (CH0003R) in Switzerland. The seasonal pattern for PM_{10} concentrations is similar for the year 2004 and the period 1997–2004. This is comparable with the other Swiss sites discusses in chapter 2.

In Southern Europe, there is no characteristic seasonal variation for the PM concentrations common for the entire region. A seasonal pattern is observed in Spain where PM concentrations were higher during summer and spring. In Italia the concentrations are generally higher during the summer at the Montelibretti, whereas for the Ispra station in Northern Italy the concentrations have the same variation as for stations in Central Europe. In Cyprus PM_{10} concentrations ranged from 28–45 µg/m^3 during the warm period, which is extended for a longer period than in Central Europe, while during the winter the mean value was approximately 15 µg/m^3. A seasonal pattern was also evident in Northern Europe and specifically in Scandinavian countries where PM concentrations are generally higher during spring, August, January and December.

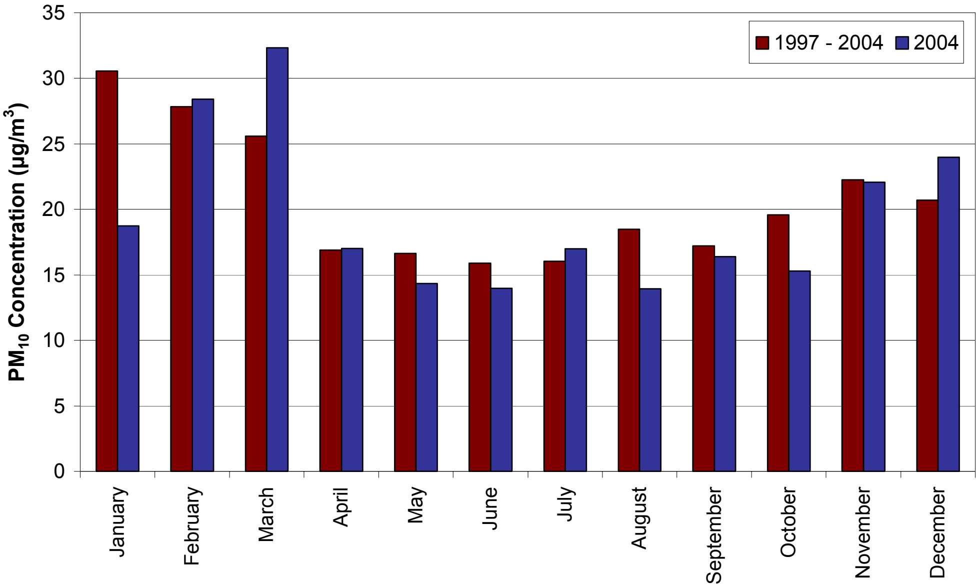

Figure 1.6: Mean monthly concentrations for PM_{10} at the Tänikon (CH0003R) station in Switzerland for 2004 and during the period 1997-2004.

The daily variation of particulate matter concentrations has also been examined. At each station the concentrations were averaged with respect to the days of the week. The analysis did not reveal any characteristic weekly pattern common for the majority of the stations. In Figure 1.8 and Figure 1.7, the daily variation of the PM_{10} concentrations for 2004 and for the period 1999–2004 are given for the sites Deuselbach (DE0004R) in Germany and Birkenes (NO0001R) in Norway to provide examples. While a clear weekly pattern was observed for Deuselbach in 2004, experiencing peak concentrations in the middle of the week, the concentrations observed at Birkenes showed very little variation throughout the week. High concentrations in the middle of the week are probably associated with car traffic and anthropogenic activities. For the period 1999-2004, no weekly pattern was observed for neither of the sites.

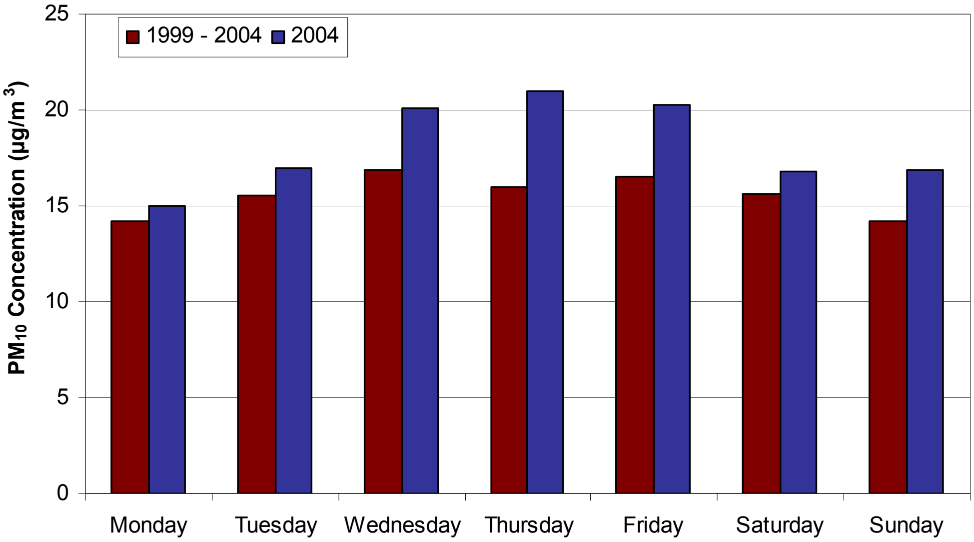

Figure 1.7: Mean weekly profile for PM_{10} concentrations at the Deuselbach (DE0004R) station in Germany for 2004 and for the period 1999–2004.

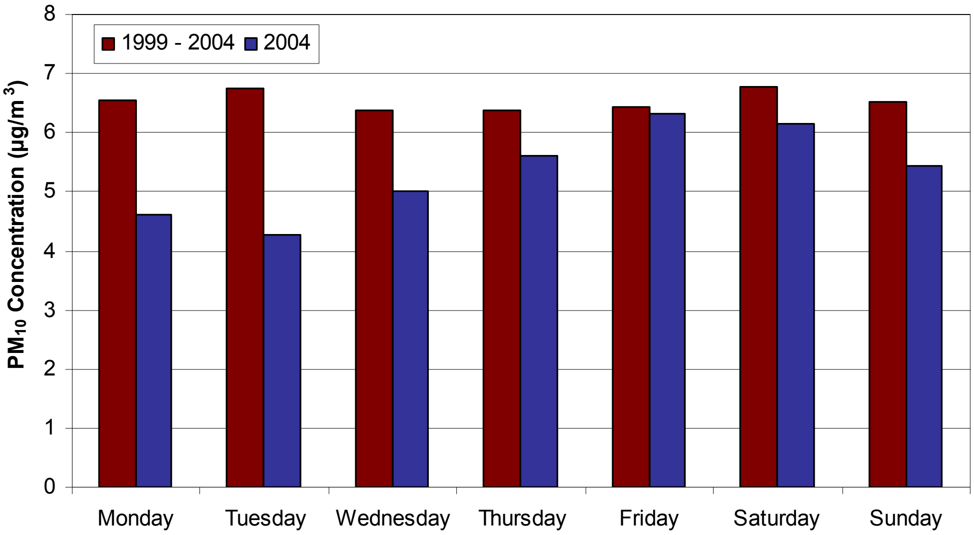

Figure 1.8: Mean weekly profile for PM_{10} concentrations at the Birkenes (NO0001R) station in Norway for 2004 and for the period 1999–2004.

1.6.2 Analysis of air mass trajectories

The PM_{10} and $PM_{2.5}$ (where available) concentrations have been averaged by daily sector values, day, year and month for each station during 2004 and are presented in Annex 4. An overall conclusion regarding air mass trajectories during 2004 is that particulate mass concentrations (for PM_{10}, $PM_{2.5}$ and PM_1) at the EMEP sites are increased when the air masses arrive at the stations from south-eastern locations. This has been exemplified in Figure 1.9 for concentrations of PM_{10} at the Aspvreten (SE0012R) station in Sweden in 2004. The figure clearly shows that the daily PM_{10} concentrations are enhanced when the air masses arrived to the station from south-eastern and southern directions after passing over continental and industrial areas and the generation of SIA (Secondary Inorganic Aerosol). The minimum concentrations were observed when air masses originated from the north-west (mainly marine areas).

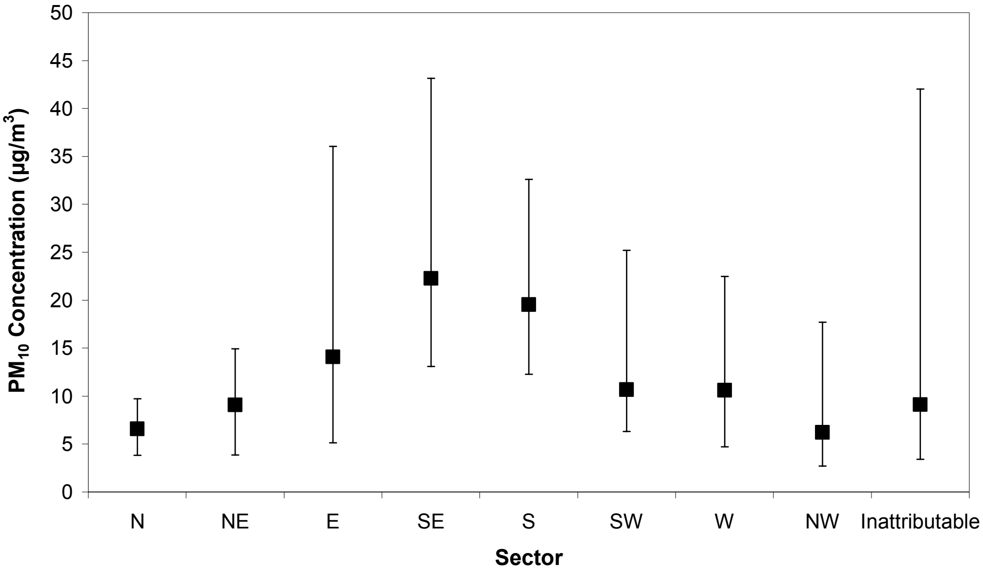

Figure 1.9: Concentrations of PM_{10} averaged by sector of air mass origin for 2004 at the Aspvreten (SE0012R) station in Sweden.

1.6.3 Analysis of time series of PM mass concentrations for representative EMEP stations

Time series for stations in each part of Europe were examined in order to explain the observed variations of PM mass concentrations. The stations have been selected based on the representation of stations for each part of Europe (Payerne, CH0002R; Langenbrügge, DE0002R; Zarra, ES0012R; Birkenes, NO0001R) or deviation from a general pattern valid for stations in a region (Ispra, IT0004R; Ayia Marina, CY0002R), as well as for high and low PM values (Illmitz, AT0002R; Birkenes, NO0001R). In addition data availability specifications were taken into account in order to determine the influence of specific PM chemical components on the observed concentration patterns (AT0002R, IT0004R, NO0001R).

EMEP stations in Central Europe

Figure 1.10 presents the time series of daily and monthly mean concentrations of PM_{10}, $PM_{2.5}$ and PM_1 at the Illmitz station in Austria, PM_{10} and $PM_{2.5}$ at the Payerne (CH0002R) station in Switzerland and PM_{10}, $PM_{2.5}$ and PM_1 at the German site Langenbrügge (DE0002R).

At the Austrian site the value of the daily limit value for PM_{10} of 50 µg/m^3 is also indicated, which may not be exceeded by more than 35 days a year. The annual mean concentration of PM_{10} for 2004 at the Illmitz station (24.4 µg m^{-3}) was relatively high compared to the other stations in Central Europe (See Table 1.1). In addition, the US EPA annual limit value of 15 µg m^{-3} for $PM_{2.5}$ was exceeded. From Figure 1.10 it can be seen that the concentrations of $PM_{2.5}$ and PM_1 correlate with the PM_{10} concentration (Pearson R = 0.96 for $PM_{2.5}$ vs. PM_{10} and R = 0.88 for PM_1 vs. PM_{10}). PM concentrations were increased during the winter compared to the summer period, and it should be noted that 26 of the 28 exceedances of the daily limit value were reported during the period October–March. Most exceedances were associated with air masses arriving at the stations from an inattributable direction (Appendix 4). In general, high PM_{10} concentrations are observed when air masses arrive at the station from south-eastern directions (Balkan area). The maximum concentrations observed during exceedances are observed with the arrival of air masses from SW (industrialized northern Italy and Czech Republic), W (traffic and industrial areas in Austria, central and western European countries) and NW (Germany) directions. In addition, the concentration of K^+ was found to be increased during winter (probably due to biomass burning for heating) compared to summer, whereas the concentration of SO_4^{2-} was approximately the same for both periods. However there is no experimental evidence that the relative contribution of $PM_{2.5}$, PM_1 mass and SIA (Secondary Inorganic Aerosol) increased during elevated PM_{10} concentrations. This indicated that the variations of PM concentrations measured at the Illmitz station might be attributed to local anthropogenic sources.

As seen for Illmitz, the PM_{10} and $PM_{2.5}$ concentrations at the Payerne station (CH0002R) are elevated during the winter. Exceedences of the daily limit value for PM_{10} concentrations did mainly occur during winter and were associated with air masses originating from the western or inattributable directions. The highest PM_{10} concentrations were observed when air masses arrived at the station from the eastern directions (see appendix 4 for details). It should be noted that although the variation of monthly mean concentrations at this station is quite typical for this part of Europe, the concentrations are rather high, especially during the winter period, compared to other stations in this region. The monthly variation of PM concentrations is, however, representative for stations with high annual concentrations in central Europe.

The Langenbrügge (DE0002R) station in Germany is maybe a more characteristic example of a station in Central Europe with respect to monthly mean concentrations and variation, Figure 1.10. At the Langenbrügge station exceedances of the daily limit value for PM_{10} in 2004 occurred during winter and spring. Air masses arrive at the station mainly from western (W, NW) or non-specific directions and increase ambient concentrations especially during the winter. Air masses from E and SE directions have also been associated with high PM concentrations (e.g. 52.23 µg/m^3 on 11/3/06). This can be explained by the presence of urban areas close to the station, specifically Langenbrügge at the west, Ützen at northwest, Wittingen in the south and Salzwedel east of the station It is also depicted in Figure 1.10 that concentrations of the different PM fractions have the same daily variation. Characteristic for Langenbrügge is that PM_1 accounts for a rather low fraction of the PM_{10} concentrations compared to that observed at Illmitz in Austria (AT0002R) and Chaumont (CH0004R) in Switzerland.

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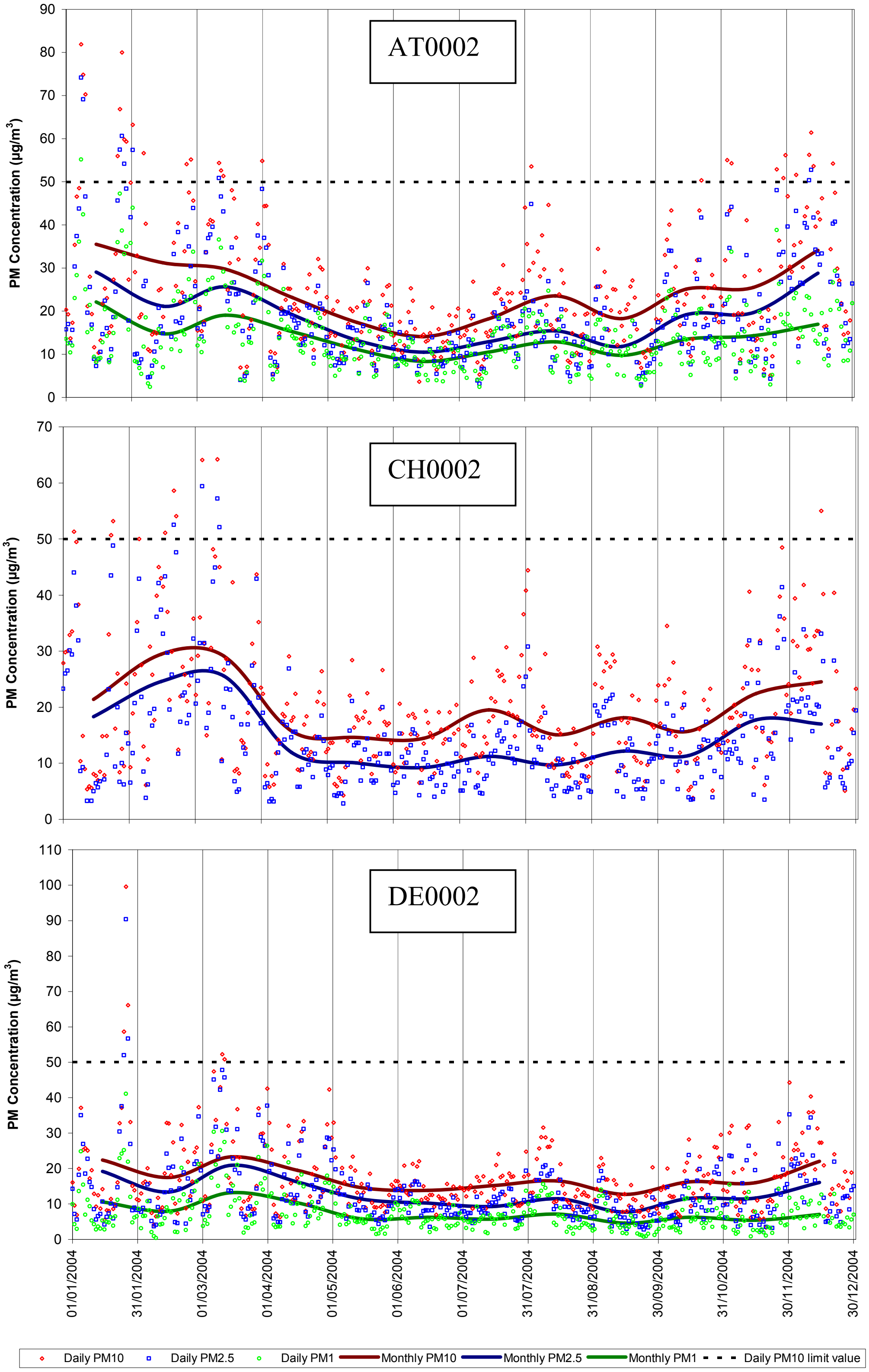


Figure 1.10: Time series of daily and monthly mean concentrations for PM_{10}, $PM_{2.5}$ and PM_1 at the Illmitz (AT0002R), Payerne (CH0002R) Langenbrügge (DE0002R) stations.

EMEP stations in Northern Europe

The Norwegian station Birkenes (NO0001R) reported the lowest annual mean concentrations of PM_{10} (5.3 µg m^{-3}) and $PM_{2.5}$ (3.3 µg m^{-3}) of all the EMEP stations for 2004, hence it should be representative for the low PM levels typically encountered in Scandinavia (see Table 1.1). The seasonal variation of PM_{10} and $PM_{2.5}$ (Figure 1.11) at Birkenes, differs from that typically seen for central Europe, experiencing the highest concentration during late winter and spring. This has been a characteristic feature for this site since the PM measurements started, and can be attributed to an increased frequency of episodes of long-range transport from the European continent for this time of the year. This is also consistent with the fact that air masses arriving Birkenes originating from south and southeast, are associated with increased concentrations of PM. The peak concentrations experienced during spring at Birkenes is also observed for the Swedish stations SE0011R, SE0012R and SE0035R (Annex 1).

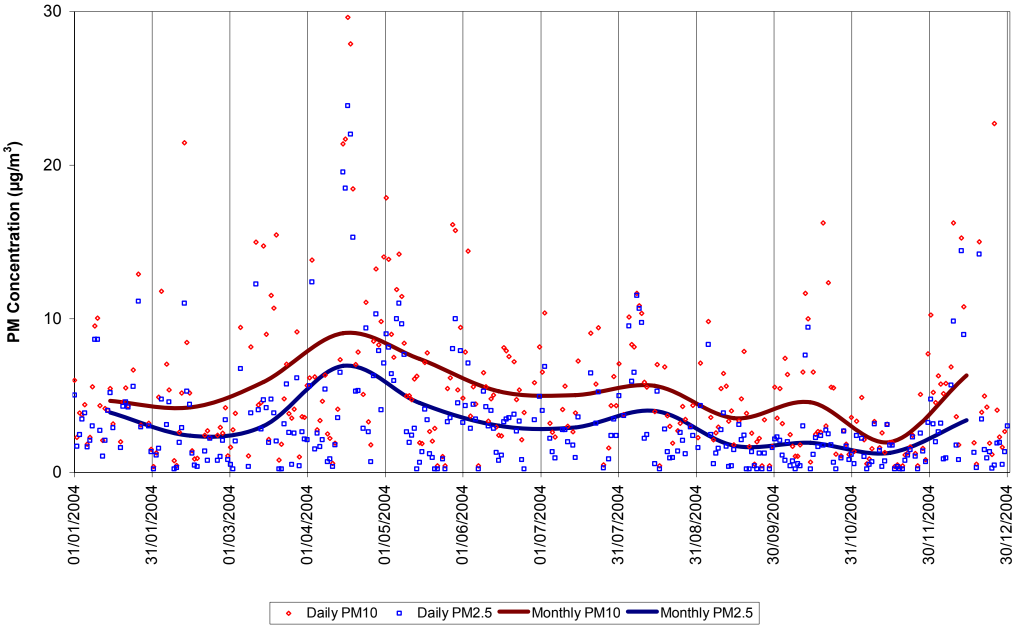


Figure 1.11: Time series of daily and monthly mean concentrations for PM_{10} and $PM_{2.5}$ at the Birkenes (NO0001R) station in Norway.

EMEP stations in Southern Europe

In Southern Europe, 16 stations reported PM concentrations directly to EMEP for 2004. However, only the stations in Spain, Italy and Cyprus reported a dataset for the entire year. There is no characteristic seasonal variation for the PM concentrations common for the entire region. In Spain and at the Montelibretti station in Italy, concentrations are generally higher during the summer, whereas for the Ispra station in Northern Italy the concentrations have the same variation as for stations in Central Europe. In Cyprus PM_{10} concentrations ranged from 28–45 µg/m^3 during the warm period, which is extended for a longer period than in Central Europe, while during the winter the mean value was approximately 15 µg/m^3

For the Zarra (ES0012R) station in Spain it is observed that the relative contribution of $PM_{10-2.5}$ particles is increased during the period July–October. This is also the period experiencing the highest number of exceedances of the daily limit value for PM_{10}. Especially the exceedances experienced during July–August are associated with southern air masses, which suggest frequent intrusions of Saharan dust (Figure 1.12). The episode on the 25 July is however experienced when continental air masses arrived from the north. This might be associated with dust resuspension, which occurs in Spain during the summer due to the dry conditions.

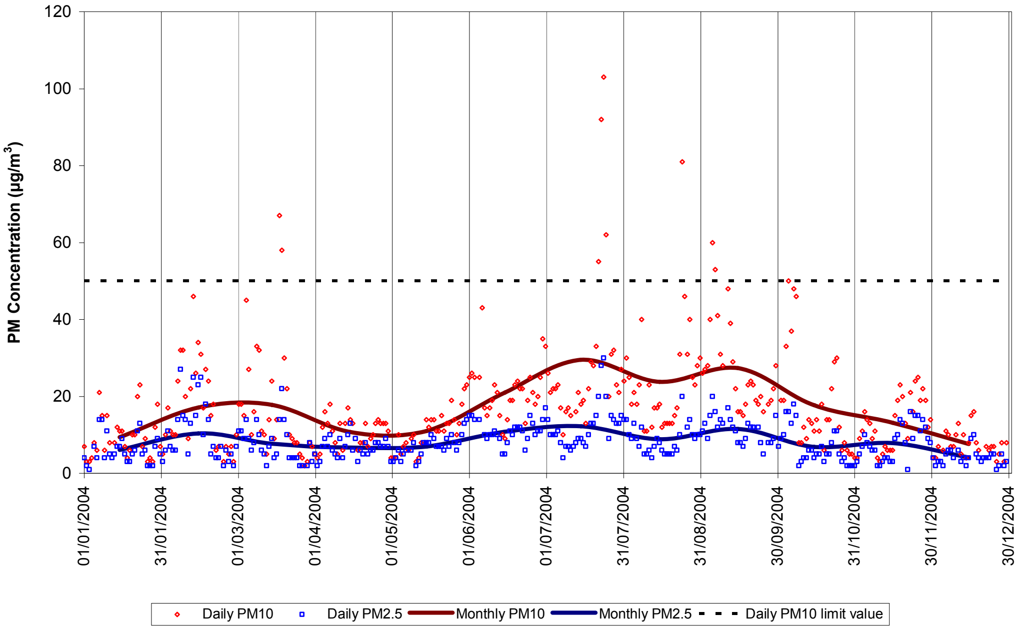


Figure 1.12: Time series of daily and monthly mean concentrations for PM_{10} and $PM_{2.5}$ at the Zarra station in Spain.

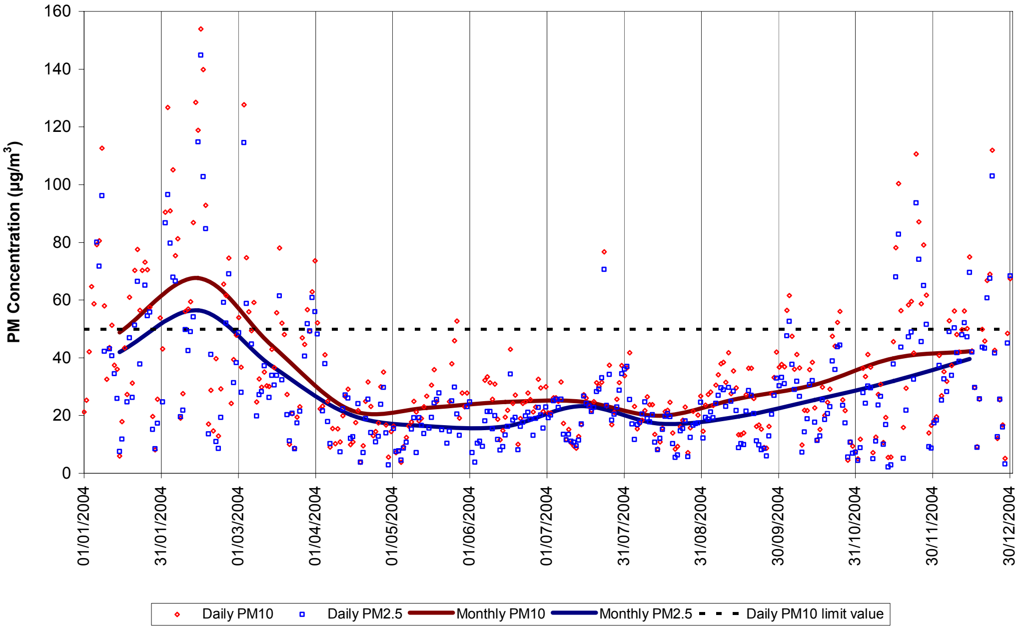


Figure 1.13: Time series of mean daily and monthly concentrations for PM_{10} and $PM_{2.5}$ at the Ispra station in Italy.

In Figure 1.13 the mean daily and monthly concentrations for PM_{10} and $PM_{2.5}$ at the Ispra station in Italy are presented. At the Ispra station the higher annual mean concentration for 2004 was measured among the other EMEP stations. The PM concentrations at Ispra were higher during the winter and lower in the summer, which is a typical situation for a station in central Europe. The elevated PM concentrations compared to other stations in the region can be partially explained by frequent arrival of air masses at the station from northern and north-western direction (Central Europe). However, PM concentrations are more probably associated with local anthropogenic sources and specifically industrial and traffic emissions at northern Italy. This is also supported by the fact that the annual mean concentration for 2004 in Ispra is higher than that at other rural stations in Italy and that the annual variation of PM_{10} mean concentrations during the period 2000–2004 is the same as for traffic sites in the region

Finally, the PM_{10} concentrations at the Ayia Marina station in Cyprus are presented in Figure 1.14. Cyprus is located in the eastern part of the Mediterranean Sea south of the Anatolian peninsula and therefore PM concentrations at the station are not influenced considerably by emissions in central Europe. The daily concentrations for 2004 at the station were high during the period February–October. Exceedances of the daily limit value for PM_{10} occurring during October are associated with air masses originating from the eastern Mediterranean Sea (ship emissions and marine aerosols) whereas the concentration peaks occurring during February are attributed to northern/north-eastern air masses arriving from Turkey (mainly anthropogenic activities).

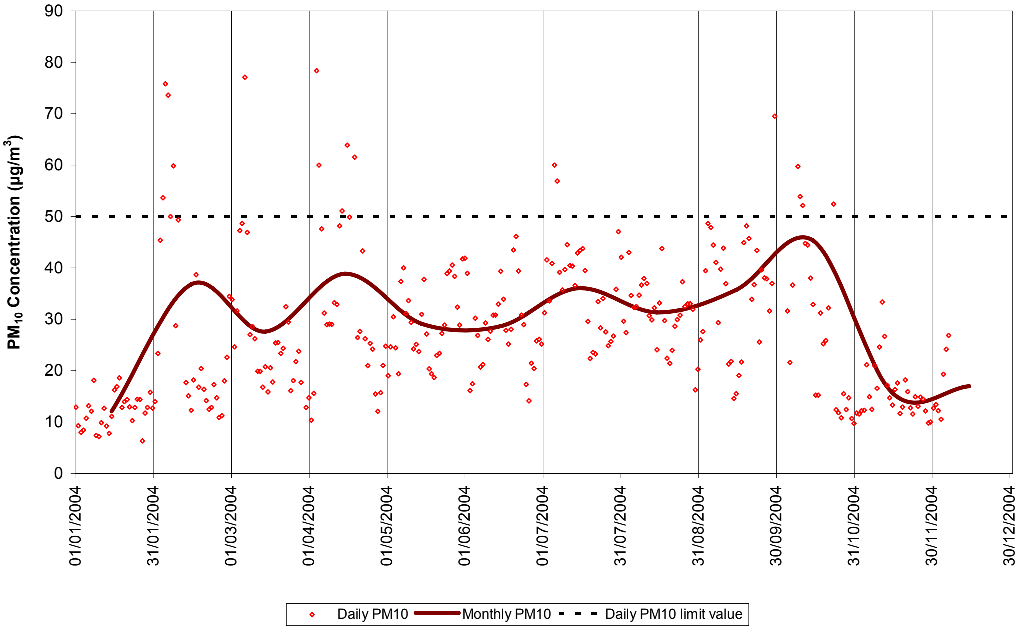


Figure 1.14: Time series of mean daily and monthly concentrations for PM_{10} at the Ayia Marina station in Cyprus.

1.7 The inorganic fraction of the particulate matter

Airborne particulate matter is a complex mixture of many different chemical species originating from a variety of sources. At EMEP stations, speciation of the ambient particulate matter is mainly focused on determination of the secondary inorganic aerosol constituents, sulphate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+). Sulphate is determined at the majority of the EMEP sites, ammonium and nitrate are reported as the sum of ammonia and ammonium, and the sum of nitrate and nitric acid, respectively, but occasionally also separately. However, these measurements are mainly done using the filter pack method, which is a biased method to separate gaseous and particulate nitrogen species. Another uncertainty is the undefined cut off for the filter pack measurements, but since most of ammonium sulphate and ammonium nitrate is found in the fine fraction it is possible to use these measurements as an estimate of the content in the PM_{10} fraction. The carbonaceous content of the aerosols are only measured at a very few stations, but has been extensively addressed during measurements campaign, such as the EMEP EC/OC campaign. In this chapter, we have focussed on the available measurement data of PM and SIA obtained during 2004. Carbonaceous matter is discussed in Chapter 3.

Previous studies have shown that total particulate sulphate and nitrate are dominant constituents of the PM mass, with ammonium nitrate prevailing in Western Europe and sulphate (mainly as ammonium sulphate) prevailing in Eastern Europe (Lazaridis et al., 2002; Van der Zee et al., 1998; Zappoli et al., 1999).

Inside the EMEP monitoring network extensive studies of chemical speciation of ambient particulate matter have been performed during 2004. The measurements are described in EMEP/CCC data report (Fjæraa, 2006). An overview of which sites are measuring what is given in Annex 5. The stations measuring sulphate during 2004 are concentrated in the Central, Western and Northern Europe. In Southern and Eastern Europe measurements of sulphate ions are less widespread. Country averaged concentrations ranged from 0.74 μg/m^3 for Norway to 3.62 μg/m^3 for Hungary. The lowest concentrations are reported for the Scandinavian countries, whereas the highest concentrations are recorded in the central and southern part of Europe (this difference can be attributed to the local anthropogenic emissions). The highest nitrate concentrations are observed in the central and southern parts of continental Europe. Higher concentrations of ammonium were measured at stations in Italy, Netherlands, Poland and Hungary, probably due to agricultural inputs, whereas lower concentrations were found for stations in Norway. There are also limited number of data for soluble base cations and sea salt ions making it difficult to draw conclusions regarding their spatial distribution in Europe.

The chemical speciated aerosol data reported from the EMEP stations during 2004 include only inorganic components, with the exception of two sites (NO0001R and IT0004R). Therefore, it was not possible to perform a full mass closure for the majority of the sites. A complete aerosol mass closure requires measurement of the PM carbonaceous content, and this remains a high priority objective in the EMEP Framework. Previous studies have shown that there is relatively good correlation between observed concentrations of particulate matter and concentrations of (aerosol) sulphate (and nitrate) at many of the EMEP sites, even if the sulphate and nitrate concentrations account for 20%-40% of the aerosol mass (Lazaridis et al., 2002). The current measurements Table 1.4 support the results from these previous studies and also show that ammonium sulphate and ammonium nitrate are important components of the atmospheric particulate matter. Sulphate contributes between 9–23% of the PM_{10} while nitrate and ammonium between 3–16% and 3–8% respectively. Base cations (K, Ca) are measured at three sites only but the contribution to the PM_{10} is small, below 2%. But these results are not representative for southern Europe. Sea-salts (mainly Na, Cl, Mg) may in coastal areas contribute significantly to the particulate mass concentrations. Most EMEP sites are however located at some distance from the coast to avoid direct contributions from sea-spray. Still only few sites report sea-salt concentrations; on the regional scale this contribution is typically lower than 2% for inland site. In more costal areas, the contribution is higher, i.e. 15% at the Birkenes site (NO0001). In total, the inorganic fraction contributes between 20–60% of the PM_{10} mass across Europe.

Pearson correlation coefficients obtained from correlating the inorganic aerosol constituents (NO_3^-, NH_4^+ and SO_4^{2-}) with particulate matter mass (PM_{10} and $PM_{2.5}$) are presented in Table 1.5. As PM measurements are available at a limited number of EMEP sites only, it is difficult to obtain a complete picture for all geographical regions. The results showed that there was a rather high correlation both between ammonium and $PM_{2.5}$ and ammonium and PM_{10} at four of the sites (see Table 1.5), but at the Italian site Montelibretti (IT0001R) it was much lower ($R = 0.39$) than for the other stations.

Correlation coefficient between sulphate aerosols and PM_{10} ranged from 0.34–0.91. Low correlations were observed for the Italian and Slovakian stations. Correlation coefficients between nitrate and PM_{10} ranged from 0.28–0.86. Correlations were generally better between the concentration of the inorganic species and the $PM_{2.5}$ mass.

Table 1.4: Contribution in per cent of nitrate, ammonium, sulphate, marine (Na^+, Cl^- and Mg^{2+}), base cations (K, Ca) and total inorganic aerosols to PM_{10} concentrations during 2004.

Station	Nitrate (NO_3^-)	Ammonium (NH_4^+)	Sulphate (SO_4^{2-})	Marine aerosols	Base cations (K, Ca)	Inorganic aerosols
AT0002R	2.8	3.4	10.0	0.5	1.2	17.9
CH0002R			10.5			
CH0005R			12.6			
CZ0001R*			11.2			
CZ0003R*			10.4			
DE0003R			16.8			
DE0004R[1]			16.3			
DE0007R			18.9			
DE0009R			17.3			
DE0041R			12.7			
ES0007R	9.0		9.9			
ES0008R	9.9		21.7			
ES0009R	8.7		12.7			
ES0010R	11.5		18.6			
ES0011R	6.8		13.4			
ES0012R	11.6		17.5			
ES0013R	9.9		16.2			
ES0014R	12.3		17.7			
ES0015R	9.7		11.9			
ES0016R	7.9		23.0			
GB0006R*	0.4	3.1	8.5			
IT0001R	10.4	7.2	11.4			
IT0004R	15.9	8.3	9.8			
NL0009R*		6.5				
NO0001R	15.0	7.1	19.6	14.6	1.8	58.0
SE0011R			11.9			
SI0008R[1]			(14.3)	(1.2)	(1.5)	
SK0005R	11.4		16.8			
SK0006R	7.4		17.2			

1 Data capture less than 50%.

* EMEP2 (PM_{10} data from AIRBASE).

Table 1.5: Correlation between nitrate, ammonium and sulphate aerosol concentrations and PM_{10}, $PM_{2.5}$ during 2004.

Station	Nitrate (NO_3^-)		Ammonium (NH_4^+)		Sulphate (SO_4^{2-})	
	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$
AT0002R	0.56	0.59	0.73	0.77	0.58	0.59
CH0002R					0.76	0.69
CH0005R					0.83	
CZ0001R*					0.45	
CZ0003R*					0.76	
DE0003R					0.50	0.84
DE0004R[1]					0.74	0.71
DE0007R					0.91	
DE0009R					0.73	
DE0041R					0.48	
ES0007R	0.39	0.55			0.51	0.63
ES0008R	0.48	0.47			0.82	0.79
ES0009R	0.57	0.60			0.52	0.70
ES0010R	0.37	0.48			0.65	0.69
ES0011R	0.36	0.59			0.47	0.66
ES0012R	0.51	0.62			0.67	0.73
ES0013R	0.28	0.38			0.46	0.68
ES0014R	0.55	0.70			0.65	0.66
ES0015R	0.40	0.47			0.64	0.71
ES0016R	0.42	0.35			0.71	0.81
GB0006R*					0.62	
IT0001R	0.50		0.39		0.39	
IT0004R	0.86	0.85	0.77	0.73	0.45	0.42
NL0009R*			0.78			
NO0001R	0.44	0.47	0.64	0.77	0.68	0.76
SE0011R					0.64	0.67
SI0008R[1]					0.83	0.45
SK0005R	0.69				0.49	
SK0006R	0.43				0.34	

1 Data capture less than 50%.

* EMEP2 (PM_{10} data obtained by AIRBASE).

1.8 Analysis of SIA for EMEP stations in each region of Europe

EMEP stations in Northern Europe

Concentrations of sulphate, ammonium and nitrate are reported for several stations in Northern Europe. However, only the Vavihill (SE0011R) station in Sweden and the Birkenes (NO0001R) station in Norway provide concurrent measurements of PM. In Denmark, measurements of sum $NH_3 + NH_4^+$, sum $HNO_3 + NO_3^-$, NO_3^- and SO_4^{2-} (µg/m^3) were performed. Sulphate and ammonium ions exhibited a good correlation in agreement with previous measurements (Lazaridis et al., 2002). Moreover, in Finland the concentrations of SO_4^{2-} correlate with the sum $NH_3 + NH_4^+$ concentrations indicating that sulphate is present as ammonium sulphate. Ammonium nitrate and gaseous ammonia can only occur when sulphate particles are fully neutralized (as $(NH_4)_2SO_4$). This indicates that the aerosol particle mass concentrations are part of regional air pollution components.

At the Birkenes (NO0001R) station (Birkenes), the sum of the inorganic ions SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, Na^+, Mg^{2+}, K^+ og Ca^{2+} comprises the 58% of PM_{10} mass in yearly basis. From the inorganic mass the 15% arises from marine sources (Cl^-, Mg^{2+}, Na^+). Their contribution is higher during autumn and winter. During February and December the marine aerosols contribute more to the PM_{10} mass more than the secondary inorganic components. The concentration of SO_4^{2-} exhibits a good correlation with the PM_{10} and $PM_{2.5}$ values ($R = 0.68$ and $R = 0.76$, respectively), Figure 1.15.

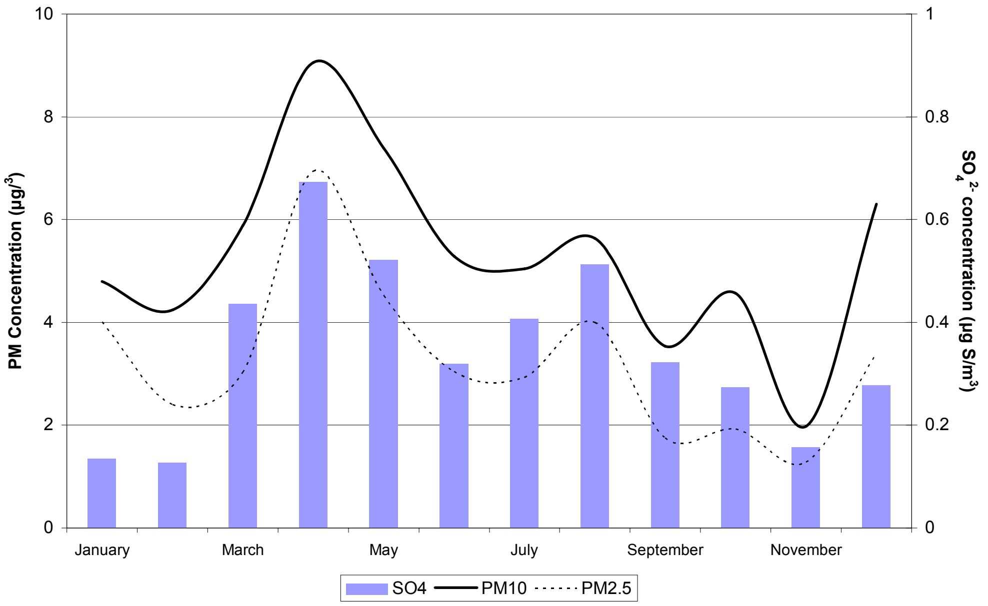


Figure 1.15: Mean monthly concentrations for SO_4-S (µg S/m^3), PM_{10} and $PM_{2.5}$ (µg/m^3) at the Birkenes (NO0001R) station in Norway during 2004.

EMEP stations in Western Europe

Although concentrations of PM inorganic constituents have been reported for many stations in Western Europe, data on PM mass concentrations are not available except for the Kollumerwaard (NL0009R) station in the Netherlands and the Lough Navar station in Great Britain. In the Netherlands the reported chemical speciated data include only NH_4^+ concentrations (Figure 1.16). The NH_4^+ concentrations are relatively high compared to other stations in Europe (mean value close to 1.5 µg N/m^3). Their daily concentration varies similarly to the concentration of PM_{10}.

In Great Britain, analysis of the SO_4^{2-} concentrations at the sites GB0002R–GB0006R and GB0007R-GB0013R have shown that they inherit no seasonal variability and that the concentration at several of the stations is highly/ moderately correlated. Higher SO_4^{2-} concentrations during 2004 were observed at the GB0007R–GB0013R stations. At the GB0002R–GB0014R stations, elevated SO_4^{2-} concentrations were correlated with air masses transported from SE and to some extent from E and NE, whereas in the stations GB0007R and GB0013R higher SO_4^{2-} concentrations were correlated with air masses transported from E (SE) (Central Europe).

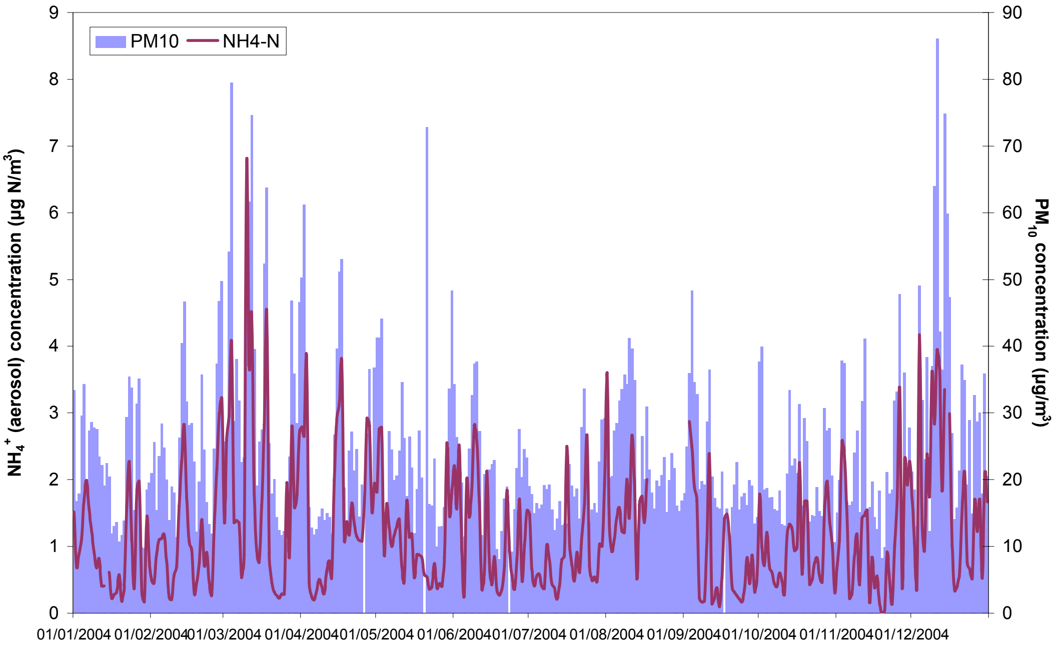


Figure 1.16: Concentration of NH_4-N (µg N/m^3) and PM_{10} (µg/m^3) during measurements in 2004 at the Kollumerwaard station in the Netherlands.

EMEP stations in Central Europe

Chemical speciated data from the EMEP stations in Germany are scarce to perform a mass closure. However, SO_4^{2-} concentrations were reported for five of the German stations. The contribution of SO_4^{2-} to the PM_{10} mass ranged from 13% to 19% (Table 1.4).

At the Payerne station in Switzerland there is a high correlation both between concentrations of SO_4^{2-} and PM_{10} ($R = 0.76$) and between SO_4^{2-} and $PM_{2.5}$ ($R = 0.69$). (Figure 1.17). One may use the sum of gases and particulate nitrogen species as an indicative of the nitrogen contribution to PM. The sum NO_3+HNO_3 and sum NH_3+NH_4 mass concentrations were higher during winter and the maximum concentrations of SO_4^{2-}, PM_{10}, $PM_{2.5}$, sum NO_3+HNO_3 and sum NH_3+NH_4 were observed when the air masses arrive from eastern directions.

On the other hand, the sum NH_3+NH_4 concentrations at the Rigi station are higher in the period March–September. There is high correlation between the SO_4^{2-} and PM_{10} data. Trajectory analysis showed that the concentrations of PM_{10}, NO_3+HNO_3 and NH_3+NH_4 have the same variability when the air masses come from NE and SE directions (from Germany and Italy respectively). However, when the air masses arrive from western directions low concentrations of PM_{10}, NO_3+HNO_3 and NH_3+NH_4 were observed. Maximum concentrations of SO_4^{2-} exist when the air masses arrive from NE and S directions.

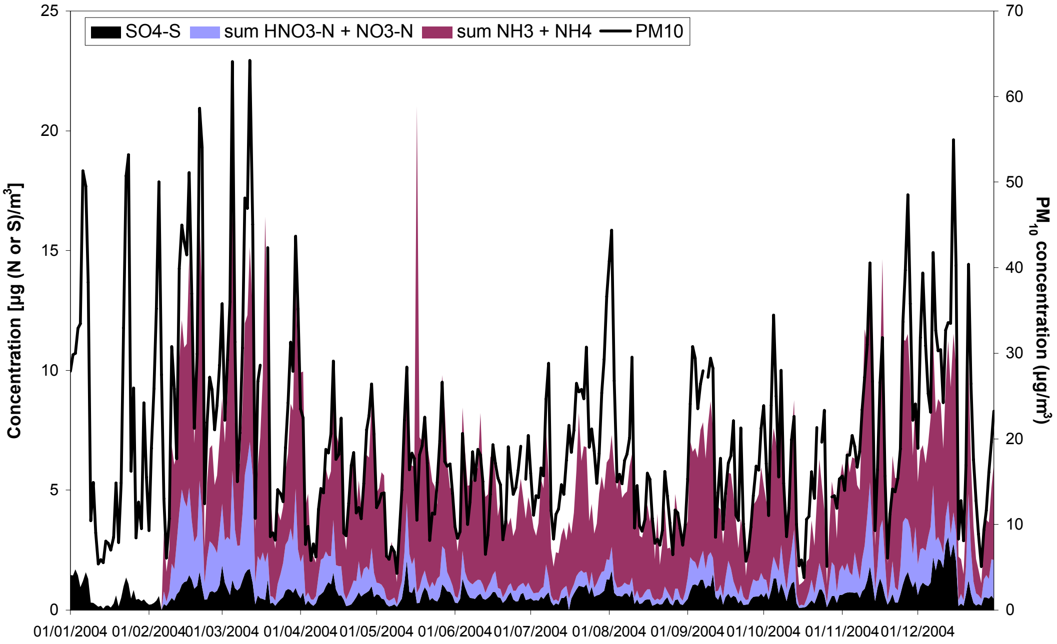


Figure 1.17: Concentration of sum HNO_3*-N +* NO_3*-N, sum* NH_3*-N +* NH_4*-N and* SO_4*-S (µg N or S/m*^{3}*) during measurements in 2004 at the Payerne station in Swizerland.*

Figure 1.18 shows the concentration of NO_3-N, NH_4-N and SO_4^{2-} (µg/m^3) at the Illmitz station (AT0002). High NH_4 concentrations were observed during the period April–August. The SO_4^{2-} and NH_4^+ mass concentrations had a high correlation (R = 0.80) which suggests that $(NH_4)_2SO_4$ was formed and the remaining ammonia formed NH_4NO_3. The correlations between several chemical components present in the particulate matter phase and PM_{10}/$PM_{2.5}$/PM_1 concentrations have been estimated. The higher correlation between NH_4^+, SO_4^{2-} and NO_3^- with particulate mass exists for PM_1 (SO_4^{2-}+NO_3^- corresponds to ~25.8% of PM_1 mass). In addition the annual variation of PM concentrations is the same as for NH_4^+ concentrations (Figure 1.19).

Further analysis of the chemical components present in aerosols has been made for the AT0002R station. Higher concentration of K^+ is observed during the winter period (wood burning during winter). The Mg^+ and Ca^{2+} components have high correlation (R = 0.78) with increased concentrations when the air masses arise from the NE. The Na^+ concentration had higher values when air masses come from W, NW, NE and E directions. Finally, the concentrations of K^+, NO_3^-, NH_4^+, SO_4^{2-} and PM_1/PM_{10}/$PM_{2.5}$ had higher values when the air masses arrive from the SE (Balkans).

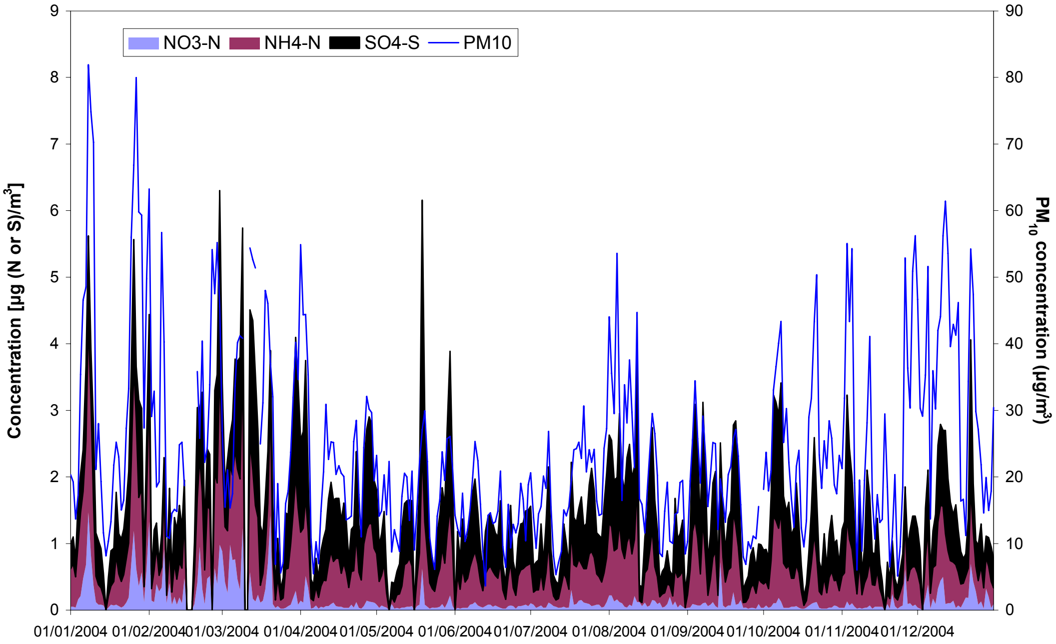


Figure 1.18: Concentration of NO_3-N, NH_4-N, SO_4-S (µg N or S/m^3) and PM_{10} (µg/m^3) during measurements in 2004 at the Illmitz station in Austria.

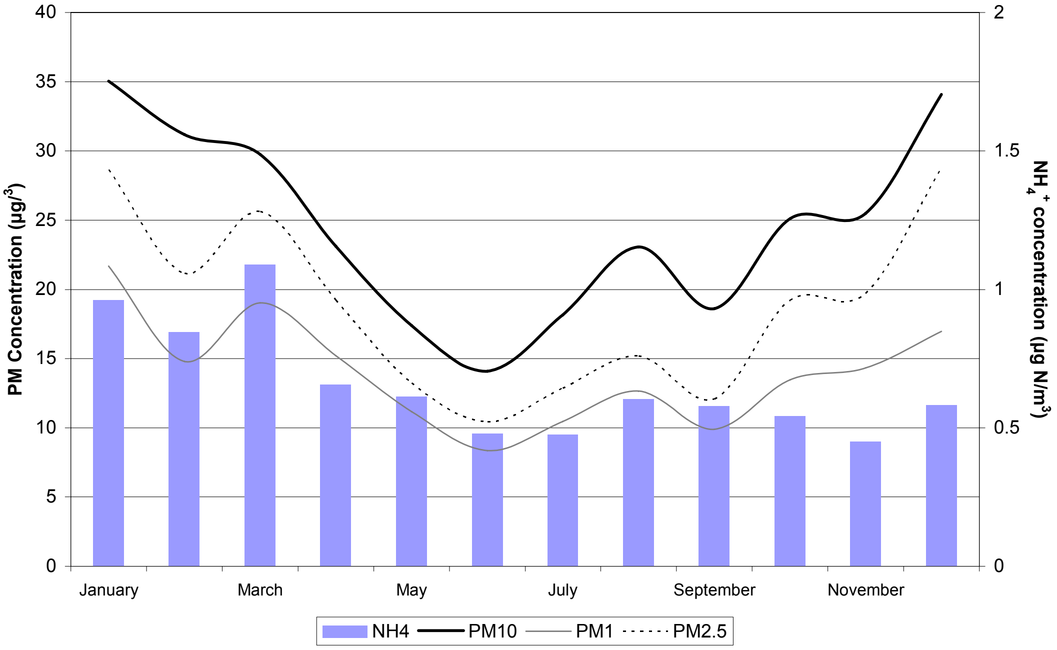


Figure 1.19: Mean monthly concentrations for NH_4^+, PM_{10}, $PM_{2.5}$ and PM_1 at the Illmitz station in Austria during 2004.

EMEP station in Eastern Europe

In Eastern Europe concentrations of sulphate and ammonium were reported during 2004. Nevertheless no station reports PM mass concentrations. The concentrations of sulphate and ammonium at stations in Latvia and Russia were correlated. Therefore ammonium and sulphate are probably in the form of ammonium sulphate.

EMEP stations in Southern Europe

Spain reported limited chemical speciated data and no certain conclusions can be drawn. At the Campisabalos station the NH_3 concentration is for the most of the cases small compared to the NH_4 concentration indicating the importance of NH_3 to the aerosol neutralization. Chemical components of the ambient aerosols measured in the EMEP stations in Italy include NH_3, NH_4^+, SO_4^{2-}, HNO_3 and NO_3^-. The measurements conclude that sulphate mass is correlated with ammonium and the ratio HNO_3/NO_3 is in general smaller than that provided in previous studies (Lazaridis et al., 2002). At the Cubuk II station in Turkey concentrations of NH_4^+ and NO_3^- had higher values during the period October–March. Higher concentrations of the aerosol chemical components were observed when the air masses originate from the south (Turkish mainland).

Slovenia has one EMEP station (SI0008) and the data during 2004 refer to inorganic ions. Figure 1.20 shows the concentration of NH_3- N, NH_4-N, sum NH_3-N + NH_4-N and SO_4^{2-} (µg/m^3). Sulphate mass concentration correlates well with the ammonium concentration indicating the presence of sulphate in the form of ammonium sulphate. There is no seasonal trend in the data and the concentrations of SO_4^-, K^+ and NH_3+NH_4^+ are higher when air masses arrive from the eastern directions (Balkans). However, higher concentrations of Ca^{2+}, and NO_3^-+HNO_3 correlate with air masses arrive from south (Italy) and higher concentrations of Na^+, Mg^+, Cl^- correlate with air masses arrive from SW and S (Adriatic Sea).

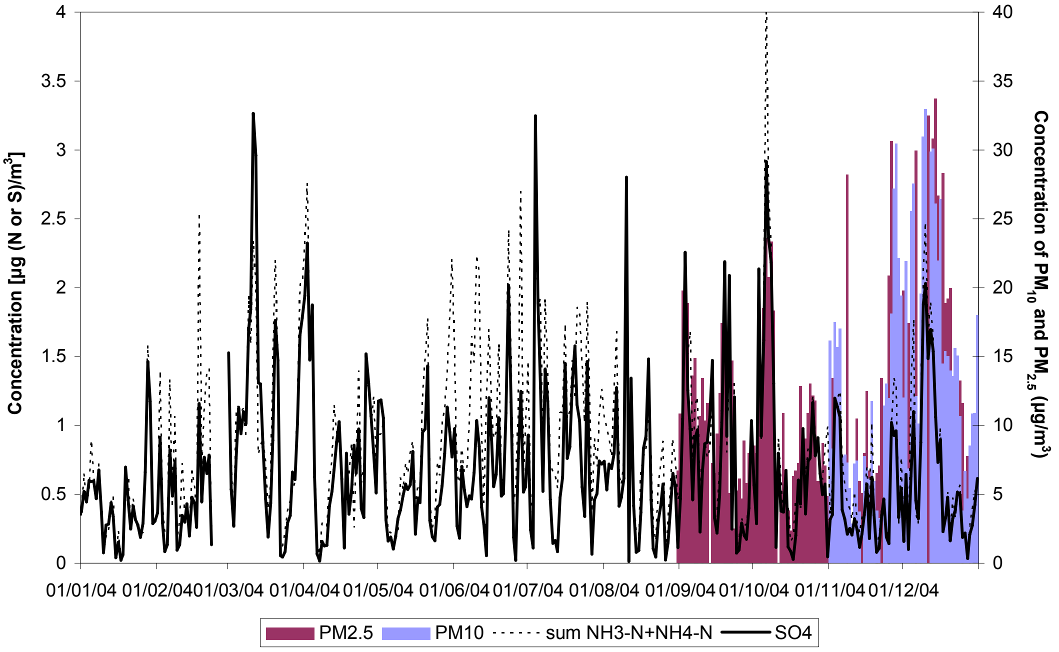


Figure 1.20: Concentration of PM_{10}, $PM_{2.5}$, sum NH_3-N + NH_4-N and SO_4^{2-} (µg/m^3) during measurements in 2004 at the Iskrba station in Slovenia.

1.9 Conclusions

The European Monitoring and Evaluation Programme (EMEP) aims to provide a basis for a quantitative assessment of the long-range transported aerosol component, and their concentration to the rural levels. During 2004 39 stations have reported PM_{10}, $PM_{2.5}$ and PM_1 data to the EMEP database. The data were mainly for concentrations of PM_{10} and $PM_{2.5}$.

During 2004, the annual limit value of 40 µg/m^3 of PM_{10} has not been exceeded in any of the stations. Furthermore, the annual limit value of 15 µg/m^3 of $PM_{2.5}$ was exceeded only at two stations during 2004. In addition, the limit value for daily averages of PM_{10} has been exceeded for more than 35 days only at the Ispra station in Italy. There were a general decrease in number of days with exceedances for most of Europe compared to 2003, but one should notice that 2003 had special meteorological conditions with very warm summer in most of Europe.

Trend analysis has been performed for the particulate matter concentrations in the EMEP stations for the period 1999-2003. No significant trends were observed and in most of the EMEP stations the variability of PM_{10} and $PM_{2.5}$ was large in the above period. However, during 2003 higher PM values were observed in most of the stations compared to the 2004 values. A seasonal pattern has been observed for some of the stations where concentrations of particulate matter were increased during the summer and were lower during the winter period or the opposite. Furthermore, the analysis did not reveal any weekly pattern for any of the stations but daily changes have been observed with lower PM values during the weekend.

In addition, the effect of the air mass trajectories reaching each station on the measured PM concentrations has been examined for the period 1997 to 2004. The majority of the EMEP stations are in central and northern Europe and for these stations south-eastern and southern directions are correlated with elevated PM levels since the air masses pass over continental and industrial areas, this includes primary PM as well as secondary aerosols. The minimum concentrations have been measured when air masses reaching the stations originated from north-west locations (mainly marine and pollution free areas).

Furthermore, data from the EMEP network and the AIRBASE database have been retrieved for the year 2004 with the objective to compare urban/rural PM concentration levels in Europe. As expected, higher PM concentrations were observed at urban sites compared to rural site. It is interesting to note that annual average concentrations of PM_{10} in several countries (such as Austria, Belgium, Switzerland, Germany and United Kingdom) exhibit the same time variation for the period 1997–2004 at urban, rural and traffic stations.

Particulate matter chemical speciation for inorganic ions has been performed in several EMEP stations, however, mainly nitrogen and sulphur species only. The major inorganic components are ammonium sulphate and ammonium nitrate. These chemical components correlate in several cases with the particulate matter mass. Finally, it is recognised that the organic speciation of ambient aerosols is additional information, which is needed for performing chemical mass closure studies in the EMEP stations.

2. Concentrations of particulate matter (PM_{10}, $PM_{2.5}$, PM_1) in Switzerland – Annual and seasonal trends and spatial variability

Robert Gehrig

2.1 Introduction

Measurements of PM_{10}, which is considered to represent the thoracic fraction of the ambient particles (ISO, 1995), have been performed within the Swiss National Monitoring Network (NABEL) already since 1997. Due to the increasing public interest for the finer alveolar fraction ($PM_{2.5}$), the measurement programme of the network has been extended to include $PM_{2.5}$ measurements at seven sites in 1998. A preliminary comparison of these parallel measurements has already been published earlier (Gehrig and Buchmann, 2003). From 2003 also PM_1 measurements were added to the measurement programme. This fraction provides, better than $PM_{2.5}$, information on the particle sources, as PM_1 represents in reasonable approximation combustion particles and secondary aerosol, while PM_{10-1} can be attributed to mechanically produced and geogenic particles. Long-term data sets of parallel measurements of the different PM fractions are still scarce if not lacking for Europe. Therefore, the Swiss data set forms a unique data basis for investigating the temporal and spatial behaviour of PM_1 and $PM_{2.5}$ compared to PM_{10}. It includes meanwhile eight years of parallel $PM_{2.5}$ and PM_{10} data and three years of parallel PM_1 and PM_{10} data at various sites and site categories important with respect to human PM exposure. This paper gives an overview of the measured concentrations and compares the temporal and spatial behaviour of the different size fractions.

2.2 Measurement programme and methods

Table 2.1 lists the seven sites where the parallel PM measurements have been performed, including their site category. The PM concentration recorded at the two rural sites, Chaumont and Payerne, are reported to EMEP. The map in Figure 2.1 shows the geographical position of the sites within Switzerland. All particle samplings were conducted with high-volume-samplers Digitel DA 80 (VDI, 1996). The sampling inlet is operated at a flow of 30 m^3/h and meets the requirements of EN 12341 for reference equivalency (UMEG, 1999). For $PM_{2.5}$ and PM_1 the same kind of instrument was used, but with correspondingly adapted sampling heads. Glass fibre filters of the type Ederol 227/1/60 were used for particle collection. The measurement uncertainty for the PM_{10} measurements has been quantified from collocated parallel measurements to be 10% (95% confidence interval for single daily values) in the concentration range 10–30 $\mu g/m^3$. The detection limit was determined from the standard deviation of field blanks to be 1 $\mu g/m^3$. Because the only difference between the applied method for PM_{10} and $PM_{2.5}$ or PM_1 is the number and diameter of the nozzles in the sampling heads, the same measurement uncertainty can be assumed for the $PM_{2.5}$ and PM_1 measurements.

Table 2.1: Characterisation of the measurement sites.

Basel	Suburban
Bern	Kerbside, street canyon
Chaumont	Rural, 1140 m a.s.l.
Härkingen	20 m from motorway
Lugano	Urban background
Payerne	Rural, 490 m a.s.l.
Zürich	Urban background

Figure 2.1: Geographical position of the seven investigated sites of the Swiss National Monitoring Network (NABEL).

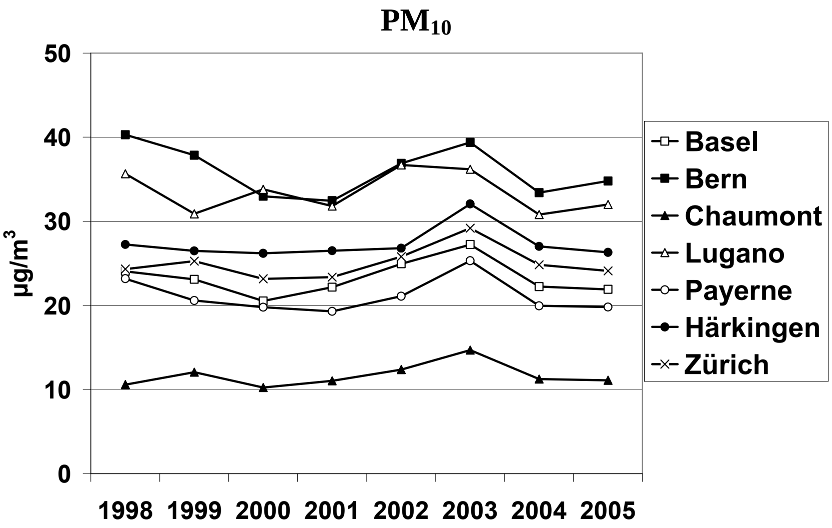
2.3 Results and discussion

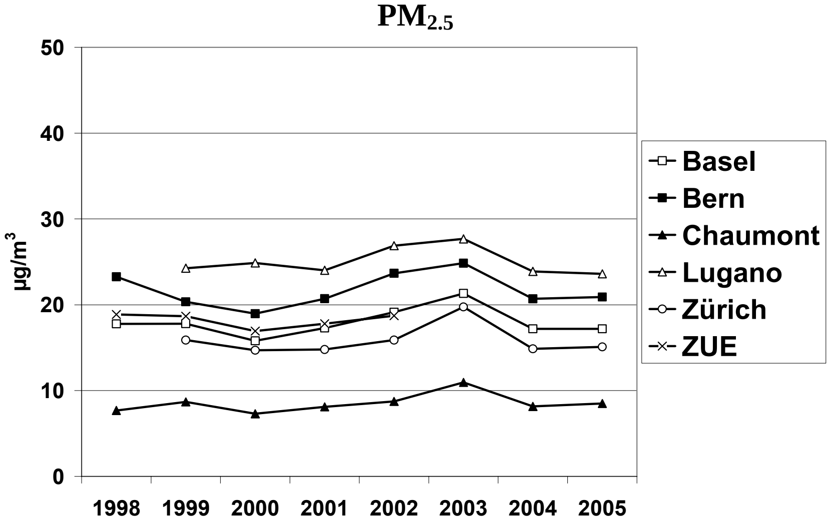
2.3.1 Comparison of PM_{10}, $PM_{2.5}$ and PM_1 concentrations

Figure 2.2 shows at a glance the annual mean concentrations of the measured size fractions. From Table 2.2 it can be seen that the variability of the long-term $PM_{2.5}/PM_{10}$ as well as the PM_1/PM_{10} ratios is very low. The only exception is the street canyon site Bern, where the local traffic forms a considerable source of coarse dust, which results in clearly lower ratios. Figure 2.3 shows the seasonal variations of the mass concentrations for the period 2003-2005. It can be seen that for all sites, with the exception of the elevated site of Chaumont, a characteristic seasonal variation can be observed for all mass fractions with elevated concentrations during the cold season. The reasons for this are not primarily caused by seasonal fluctuations of the emissions, but rather by meteorological effects. This is already well known from similar variations of other parameters such as sulphur dioxide and nitrogen oxide (frequent inversions during winter and good vertical mixing during summer). In contrast, Chaumont shows the lowest values in winter. This also shows the dominating influence of the meteorology. The site is situated on an altitude of 1140 m a.s.l. and, therefore, during wintertime most of the time above the inversion layer, thus protected from the emissions of the lowlands of the Swiss basin. From April to September the variations at Chaumont follow that of the other sites, though on a lower concentration level, due to the better vertical mixing of the lower atmosphere during the warmer season.

Table 2.3 shows, that there is a high correlation of PM_{10} with $PM_{2.5}$ at all sites. With the exceptions of the still high correlations at Bern (r=0.92) and Chaumont (r=0.91), all correlations were 0.95 or higher. The lower correlation at Bern reveals that the traffic induced coarse particles from abrasion and resuspension contained in PM_{10} follow different temporal emission patterns than $PM_{2.5}$ and PM_1, which are dominated by exhaust pipe emissions. This is plausible because mechanically produced particles, and in particular resuspension, depend not only on the vehicle frequency but also on the condition of the road surface (e.g. clean/dirty, wet/dry). At the site of Chaumont, the lower correlation can be explained with the generally lower concentrations and the correspondingly higher relative measurement uncertainties.

The correlation coefficients between PM_1 and PM_{10} are similarly high. For the reasons already explained, also here with somewhat lower values at Bern and Chaumont.





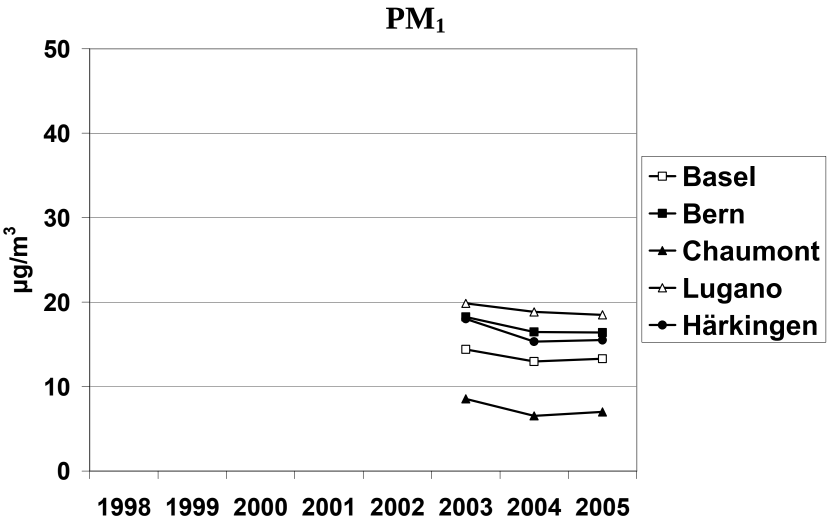


Figure 2.2: Annual mean concentrations of PM$_{10}$, PM$_{2.5}$ and PM$_1$.

Table 2.2: Mean $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} ratios of the daily values.

	$PM_{2.5}/PM_{10}$	PM_1/PM_{10}
Basel (Suburban)	0.76	0.60
Lugano (Urban background)	0.74	0.59
Zürich (Urban background)	0.75	
Payerne (Rural, 490 m a.s.l.)	0.75	
Chaumont (Rural, 1140 m a.s.l.)	0.75	0.63
Härkingen (Motorway)		0.59
Bern (Kerbside, street canyon)	0.61	0.49

Table 2.3: Pearson correlation coefficient (r) for PM_{10} vs. $PM_{2.5}$ and PM_{10} vs. PM_1 (daily values).

	PM2.5	PM1
Basel (Suburban)	0.95	0.92
Lugano (Urban background)	0.95	0.92
Zürich (Urban background)	0.97	
Payerne (Rural, 490 m a.s.l.)	0.97	
Chaumont (Rural, 1140 m a.s.l.)	0.91	0.89
Härkingen (Motorway)		0.92
Bern (Kerbside, street canyon)	0.92	0.88

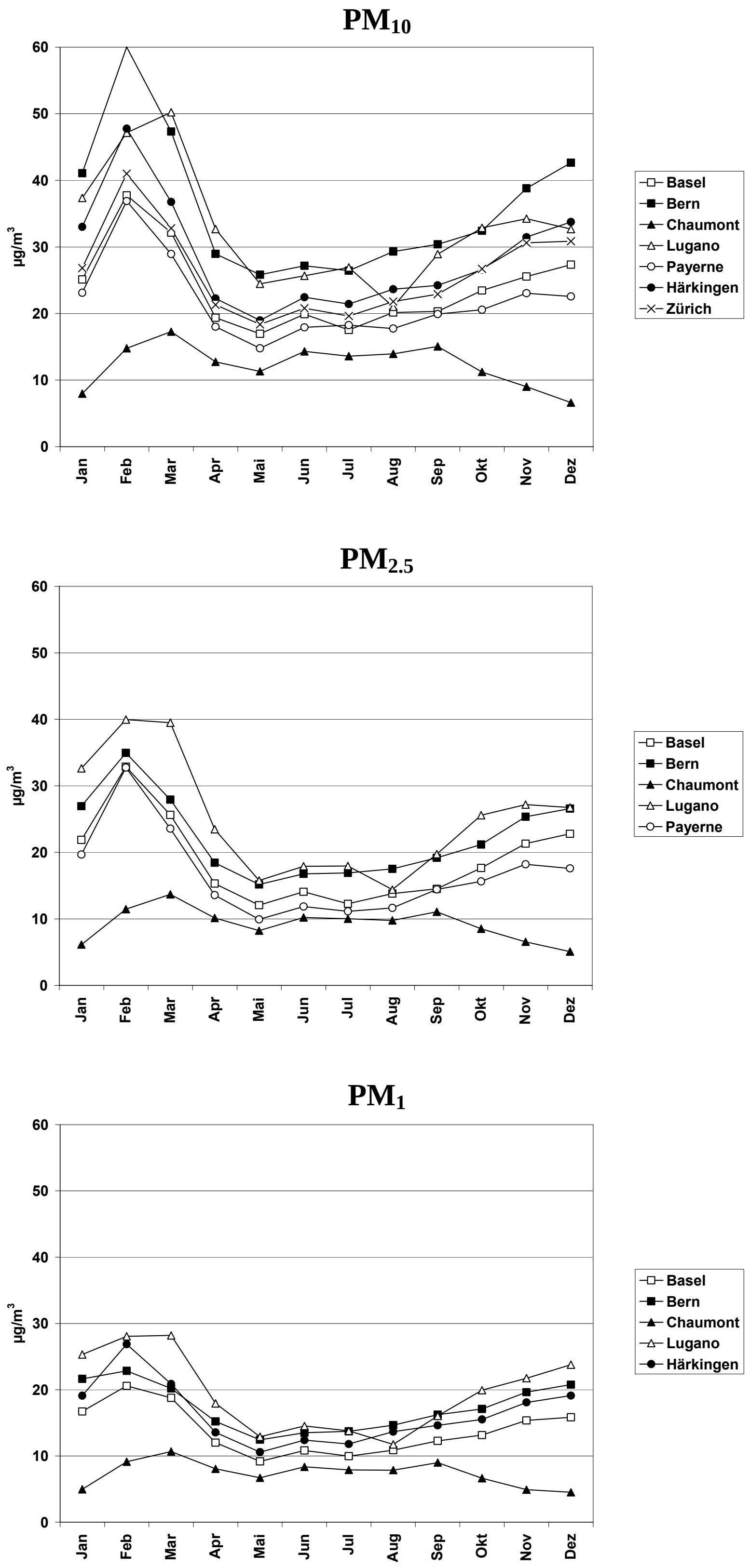


Figure 2.3: Seasonal variations of monthly means for PM_{10}, $PM_{2.5}$ and PM_1 for the period 2003-2005 .

2.3.2 Spatial variability of PM_{10}, $PM_{2.5}$ and PM_1 concentrations

Interesting information about the spatial variability of PM concentrations can be obtained from an analysis of the correlations of the PM concentration data between the different sites of the NABEL network. Table 2.4 shows the Pearson correlation coefficients r for the comparison of the daily values of different sites. It can be seen that the correlation coefficients for all mass fractions (PM_{10}, $PM_{2.5}$ and PM_1) between the sites situated north of the Alps at moderate altitude (Basel, Bern, Payerne and Härkingen) are quite high, i.e. in the range of 0.84-0.92. This indicates that the variability of the daily concentrations is not primarily influenced by local events and sources, but is rather determined by meteorological conditions. Chaumont, which is often situated above the inversion layer and Lugano, which is separated by the Alps show (not surprisingly) much lower correlations. This is elaborated in some more detail with the following three examples, which differentiate between summer (June-August) and winter (December-February). Figure 2.4 shows the correlation for PM_{10} between the sites Basel and Payerne. Though the distance between these two sites is 95 km and they are separated by the 600-800m high Jura Mountains, good correlation can be observed in particular in winter, but also during summer.

Figure 2.5 compares Basel, north of the Alps with Lugano, south of the Alps. As expected, no correlation can be observed during wintertime. During summertime the correlation is somewhat higher, but still very low. This shows clearly that the high mountains of the Alps form an efficient obstacle for the distribution and homogenisation of fine particles.

Figure 2.6 shows a comparison of the two sites Payerne and Chaumont for $PM_{2.5}$. The sites are located quite close together (distance 24 km) but on different altitudes. Chaumont is situated 650 m higher than Payerne. A high correlation can be observed during summertime when the vertical mixing of the lower atmosphere is generally good and the absolute concentration level of the mountain site is only about 20% lower than at Payerne, which is situated within the Swiss basin in a rural environment. However, during wintertime, when the meteorology is characterised by frequent inversions, the observed $PM_{2.5}$ levels are largely decoupled. The correlation is very low and the absolute concentration level at the mountain site Chaumont reaches only about 25% of that of Payerne.

Table 2.4: Pearson correlation coefficients (r) of the daily values of PM_{10}, $PM_{2.5}$ and PM_1 measured simultaneously at different sites (2003-2005).

PM_{10}

	Basel	Bern	Chaumont	Lugano	Payerne
Basel	1.00				
Bern	0.84	1.00			
Chaumont	0.60	0.45	1.00		
Lugano	0.48	0.49	0.31	1.00	
Payerne	0.89	0.88	0.60	0.51	1.00
Härkingen	0.88	0.89	0.47	0.50	0.89

$PM_{2.5}$

	Basel	Bern	Chaumont	Lugano
Basel	1.00			
Bern	0.89	1.00		
Chaumont	0.55	0.52	1.00	
Lugano	0.51	0.52	0.27	1.00
Payerne	0.90	0.92	0.58	0.52

PM_1

	Basel	Bern	Chaumont	Lugano
Basel	1.00			
Bern	0.85	1.00		
Chaumont	0.55	0.46	1.00	
Lugano	0.52	0.55	0.22	1.00
Härkingen	0.89	0.90	0.48	0.53

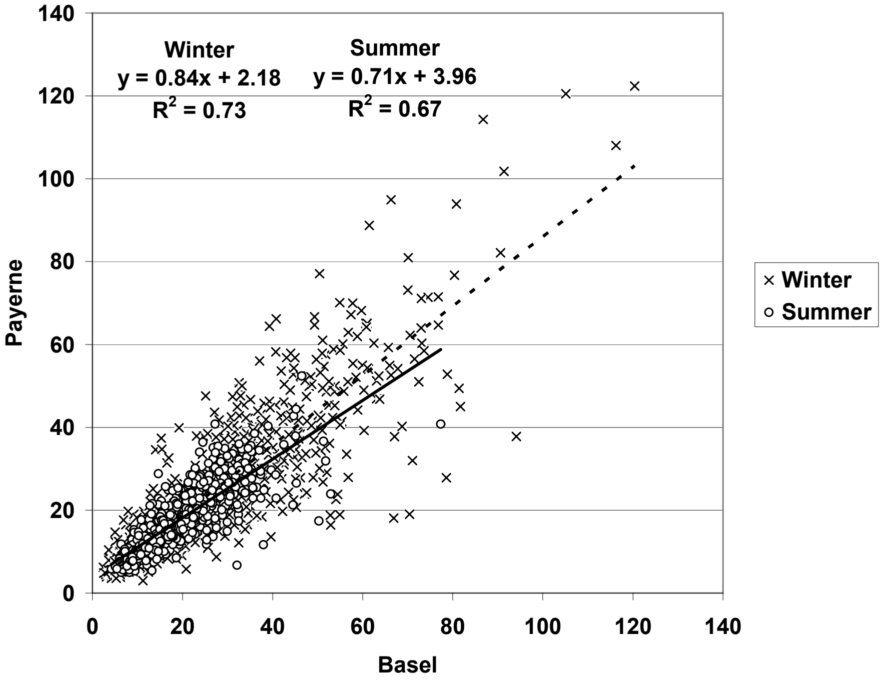


Figure 2.4: Scatterplot and linear regression of the daily values (1998-2005) of PM_{10} (µg/m3) at Basel and Payerne during summer (June to August) and during winter (December–February). Dotted line: winter; solid line: summer.

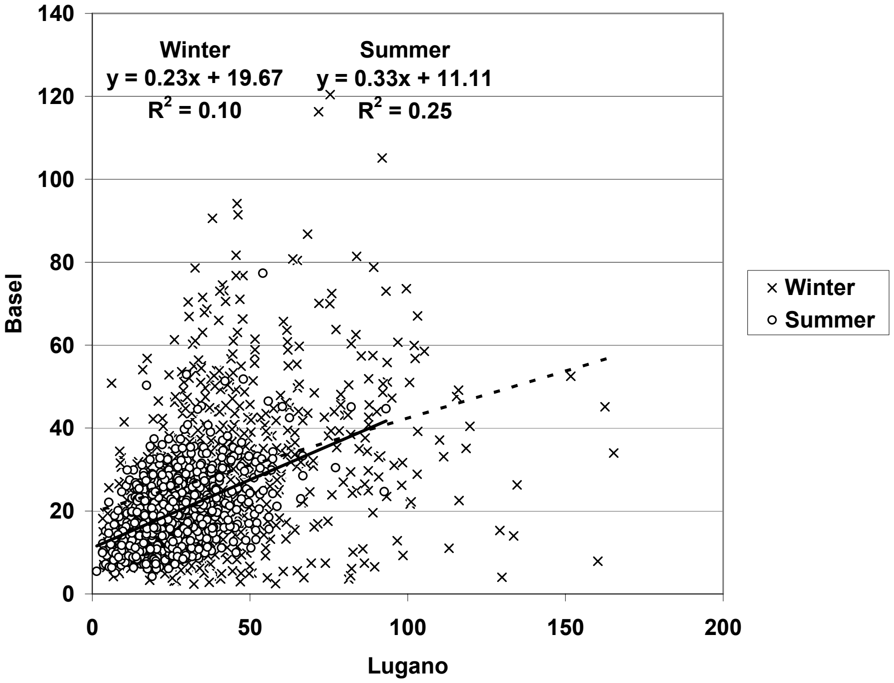


Figure 2.5: Scatterplot and linear regression of the daily values (1998-2005) of PM_{10} (µg/m3) at Basel and Lugano during summer (June to August) and during winter (December–February). Dotted line: winter; solid line: summer.

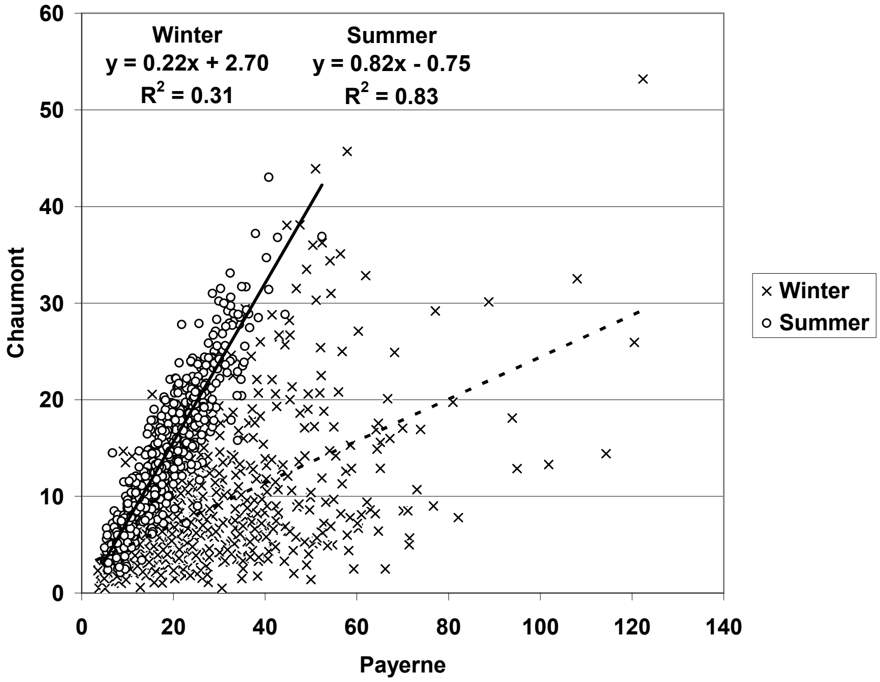


Figure 2.6: Scatterplot and linear regression of the daily values (1998-2005) of PM_{10} (µg/m3) at Chaumont and Payerne during summer (June to August) and during winter (December–February). Dotted line: winter; solid line: summer.

2.4 Conclusions

From the presented analysis of long-term parallel measurements of PM_1, $PM_{2.5}$ and PM_{10} concentrations in Switzerland the following conclusions can be drawn:

- At all sites the concentrations of the different size fractions were highly correlated on the level of daily values. Unless strong and variable local sources of coarse particles are present, parallel measurements of PM_1, $PM_{2.5}$ and PM_{10} provide only limited additional information. In order to make efficient use of the financial and personal resources, such parallel measurements can be restricted to a few carefully selected sites in a monitoring network.

- Also the comparison of daily PM values from different sites often show quite high correlation. The analysis of the Swiss data indicates that this has primarily meteorological reasons. Even distant sites show good correlations if they are situated in an area with similar meteorological conditions. However, the correlations drop drastically if this is not the case i.e. if the sites are divided by high topographical obstacles (Alps) or by an inversion layer. Particle transport modelling shows clearly the relevance of long-range transport of fine particles though robust quantification still seems to be difficult (EMEP, 2006). Therefore, high correlations of PM at even distant sites could be expected due to a relatively homogenous distribution of long-range transported PM over large areas. However, the high daily variability of the PM concentrations can not be explained with long-range transport because on a regional scale the daily variability of the PM emissions is quite small. In fact, no matter whether the particles are locally emitted or long-range transported, it is mainly the meteorology (wind speeds, turbulence, vertical mixing, inversions etc.), which causes more or less effective dilution of the emitted pollutants and thus the daily fluctuation of the concentrations.

3. Monitoring of EC and OC within EMEP – An overview

by K.E. Yttri

3.1 Introduction

The environmental relevance of the carbonaceous aerosol comprises a number of important topics, such as human health, direct and indirect climate forcing, and air-quality. The high number of organic molecules reported to be associated with ambient fine aerosols have a wide range of different physical and chemical properties, of which impact on human health and cloud formation largely remains unknown. Furthermore, black carbon is the principal light absorbing species in the atmosphere, significantly affecting the Earth's radiative balance (Penner et al., 1998).

Attempting to quantify the carbonaceous content of the ambient aerosol on the basis of single molecules is, however, an insurmountable task due to their sheer number, their various chemical and physical properties and the complex aerosol matrix. Thus, operational definitions of bulk carbonaceous material, such as elemental carbon (EC) and organic carbon (OC), have been established (Birch and Cary, 1996).

In Europe, long-term monitoring data of EC and OC is not yet available on the regional scale, although the importance of such data has been emphasized by e.g. Kahnert et al. (2004). Monitoring of EC and OC needs to rely on both robust and cost-efficient techniques, but at the same time a satisfactory quality of the data must be maintained. It has long been recognized that significant artefacts can be introduced during filter sampling of particulate matter for subsequent analysis of OC (McDow and Huntzicker, 1990), which can both grossly over and underestimate the samples content of OC. Furthermore, great analytical challenges are associated with splitting the aerosols content of EC from OC (Schmid et al., 2001). The challenges associated with sampling of OC and the separation of EC and OC is more thoroughly discussed in Chapter 4.

3.1.1 Status of sampling and measurement, and quality of data

According to the EMEP monitoring strategy, quantifying the aerosols content of EC and OC is regarded as a level 2 activity. In Table 3.1, the only three countries that have ever reported these parameters pr 2004 are listed; not accounting for the EMEP EC/OC campaign conducted during the period 2002 - 2003. These sites are Birkenes (NO0001R) in Norway, Illmitz (AT0002R) in Austria, and Ispra (IT0004R) in Italy. The longest data record is seen for Birkenes (NO0001R), which has reported concentrations of EC and OC in PM_{10} and $PM_{2.5}$ since 2001. At Ispra (IT0004R), measurements of EC and OC in PM_{10} and $PM_{2.5}$ were started in 2002, whereas for Illmitz (AT0002R) measurements of EC and OC were reported for a period of one year starting in October 1999 and lasting until October 2000. As for Birkenes and Ispra, EC and OC was quantified in both PM_{10} and $PM_{2.5}$.

Table 3.1: Sites reporting EC and OC to the EMEP database, including size fractions and sampling period.

Site (Country)	EC	OC	PM_1	$PM_{2.5}$	PM_{10}	Period
Birkenes (Norway)	x	x		x	x	2001, 2002, 2003, 2004
Illmitz (Austria)	x	x		x	x	1999, 2000, 2002(1)
Ispra (Italy)	x	x		x	x	2002(1), 2003, 2004

1) EMEP EC/OC campaign.

Table 3.2 shows the sampling time and frequency, the filter face velocity, the sampling technique, and the analytical instrumentation used at the three sites listed in Table 3.1. These parameters are the most crucial concerning the magnitude of the sampling artefact of OC and the split between EC and OC.

The two sites reporting concentrations of EC and OC in 2004 used samplers that operated according to different sampling time and sampling frequency. The sampler at Birkenes (NO0001R) operated at a filter face velocity of 54 cm s^{-1}, which was substantially higher than for the sampler used at Ispra (IT0004R) (20 cm s^{-1}). Neither of the samplers operated according to a sampling technique that corrects for, or quantifies, *both* the negative and the positive artefacts. At Ispra (IT0004R), a denuder was applied to account for the positive artefact, whereas at Birkenes (NO0001R) only a single filter was applied, accounting for neither the positive nor the negative artefact.

Table 3.2: Sampling equipment and analytical approach used at the various sites reporting EC and OC to the EMEP database.

Site (Country)	Sampling time/frequency	Filter face velocity	Sampling equipment	Analytical approach
Birkenes (Norway)	(6+1) days, weekly	54 cm s^{-1}	Single filter (no correction)	Sunset TOT (quartz. par)
Illmitz (Austria)	Every 6th day	54 cm s^{-1}	Single filter (no correction)	VDI 2465 part 1
Ispra (Italy)	24 hr, daily	20 cm s^{-1}	Denuder (pos. artifact)	Multi-step flash heating(1)

1) Two aliquots were analysed: one from the plain filter, the other one after baking for 2 hours in He/O2 carrier gas at 340°C. Charring-free EC determined from the latter.

Different analytical approaches were used to quantify the samples content of EC and OC at the two sites in question. At Birkenes (NO0001R) optical correction was used to correct for charring during analysis, whereas the approach used at Ispra baked one out of two aliquots of the sample for two hours in order to prevent charring and to provide charring free EC. According to Schmid et al. (2001) only methods that correct for charring during analysis, or that prevent charring to take place, should be recommended when it comes to splitting TC into EC and OC. Hence, both the approach used at Birkenes (NO0001R) and at Ispra (IT0004R) meet this requirement.

The differences between the two sites regarding sampling and analytical approach, pinpoints the great challenges associated with monitoring of EC and OC with respect to get comparable results. This lack of comparability make the data less suited for model validation, for validation of the effectiveness of implementation of current air pollution legislations, and for the work to revise and improve current policy. Still, the scientific shortcoming of separating the anthropogenic and natural contribution to the aerosol content of EC and OC is probably more important in this aspect. Despite that the results are not likely comparable, they still provide valuable information concerning seasonal variation, mass closure of PM, and time-trends at the respective sites.

3.2 EC and OC levels in Europe

The lack of comparable data make it necessary to turn to the EMEP EC/OC field campaign in 2002 – 2003 to address the spatial and temporal variation of EC, OC, and TC at European background sites (Figure 3.1) (Yttri et al., in prep). This dataset is quite comprehensive, and has the great benefit that all samples were analysed using the same instrument and the same temperature program, making it somewhat easier to compare the results obtained at the various sites.

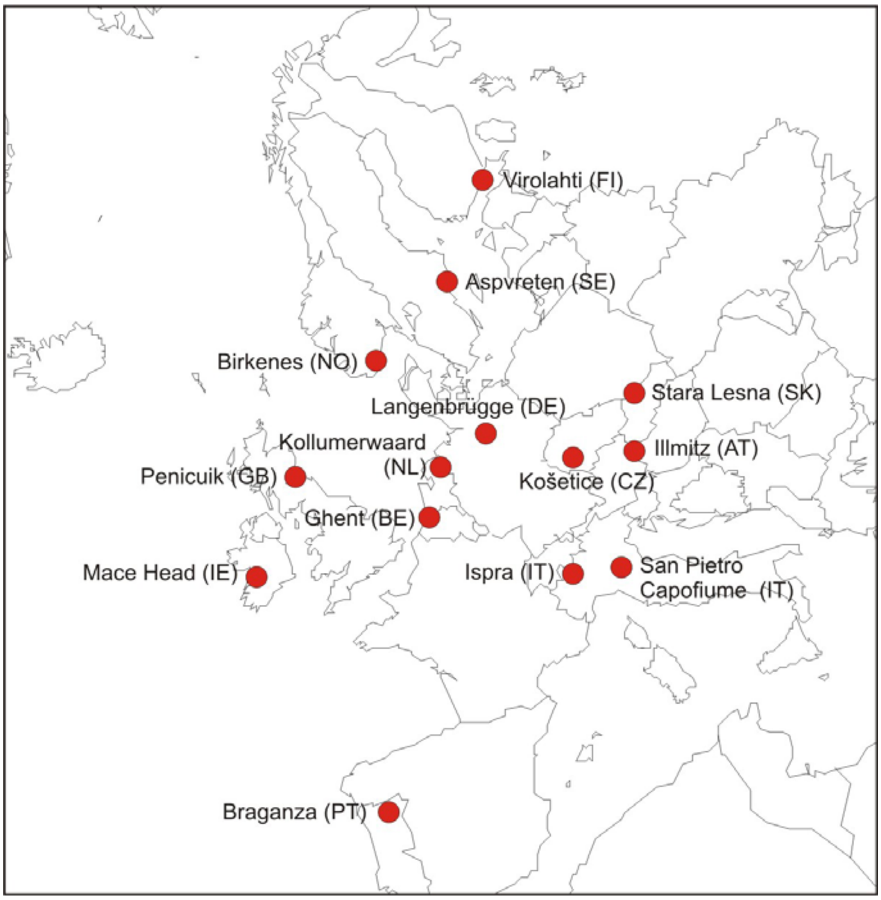


Figure 3.1: Sites participating in the EMEP EC/OC campaign conducted during the period 1 July 2002 – 1 July 2003.

The EMEP EC/OC campaign reported a North-to-South gradient for EC, OC, and TC in Europe for PM_{10}. Higher concentrations were reported for EC, OC and TC in the central, eastern and southern regions of Europe compared to the northern and western parts. This finding seems reasonable, taken into account the population density and number of sources in central Europe compared to e.g. Scandinavia. On an annual basis, the EC fraction by mass of PM_{10} appeared to be 3.6 ± 1.2%, whereas the corresponding percentage for the OM (Organic Matter) fraction was 26 ± 8% (Table 3.3). Without exception, levels of EC were higher in winter than in summer. A similar seasonal cycle was observed for OC, with the exception of Scandinavia, where concentrations were 1.5 times higher in summer compared to winter. The study nicely demonstrated how the influence of massive wildfires could explain parts of the observed seasonal variation, although their sources were located more than 1000 kilometres away. Non-published results indicate that a substantial part of the summertime increase of OC in PM_{10}, at least for parts of Scandinavia, could be attributed to primary biological aerosol particles that most likely have a local origin, and that mainly reside in the coarse fraction (Yttri et al., 2006).

Table 3.3: Relative contribution of EM (EC x 1.1)[1], OM (OC x 1.4 – 1.7)[2], and TCM (Total Carbonaceous Matter = EM + OM) to PM_{10} on annual basis (1 July 2002 – 1 July 2003).

Site	EM/PM_{10} (%)	OM/PM_{10} (%)	$EM+OM/PM_{10}$ (%)
Aspvreten (SE012)	3.0	32	35
Birkenes (NO01)	1.9	26	28
Braganza (PT01)	4.4	37	42
Ghent (BE02)	5.4	20	25
Illmitz (AT02)	3.6	31	34
Ispra (IT04)	5.0	31	36
Kollumerward (NL09)	2.7	17	20
Košetice (CZ03)	4.6	25	30
Langenbrügge (DE02)	2.7	28	31
Mace Head (IE31)	1.1	8.9	10
Penicuik (GB46)	3.8	16	19
San Pietro Capofiume (IT08)	3.9	24	28
Stara Lesna (SK04)	4.3	32	36
Virolahti (FI017)	3.5	31	35
Mean ± SD	3.6 ± 1.2	26 ± 8	29 ± 8

1. A conversion factor of 1.1 is used to account for hydrogen associated with EC.
2. The conversion factors are based on the relative contribution of WSOC and WINSOC at the respective sites.

3.2.1 EC and OC levels at the Norwegian site Birkenes (NO0001R)

The Birkenes atmospheric research station (58° 23'N, 8° 15'E, 190 m asl) is a joint supersite for EMEP and GAW and situated approximately 20 km from the Skagerrak coast in the southern part of Norway (see Figure 3.1). The site is often influenced by episodes of transboundary air pollution from continental Europe and has frequently been used to study long-range air pollution. The station is located in a boreal forest with mixed conifer and deciduous trees. The station has been operational since 1971.

Figure 3.2 a-c shows the annual mean concentrations of EC, OC, and TC in PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ at Birkenes for the period 2001 – 2005. For this period, OC in PM_{10} ranged from 0.8 µg m^{-3} to 1.1 µg m^{-3}, whereas the corresponding range for OC in $PM_{2.5}$ was 0.6 – 1.0 µg m^{-3}. For $PM_{10-2.5}$ the annual mean concentration of OC ranged from 0.1 – 0.3 µg m^{-3}. For PM_{10} and $PM_{2.5}$, the annual mean concentrations of EC ranged between 0.1 – 0.2 µg m^{-3} for the period in question. For $PM_{10-2.5}$ the annual mean concentration of EC did not exceed 0.03 µg m^{-3}.

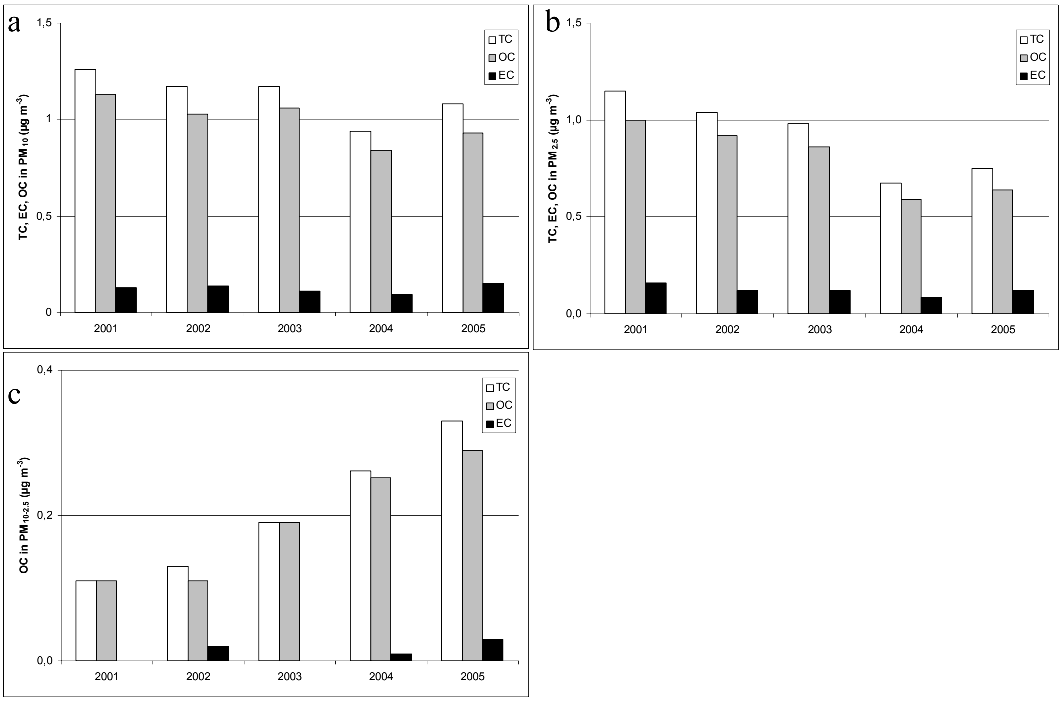


Figure 3.2: Annual mean concentrations of EC, OC and TC in PM_{10} (a), $PM_{2.5}$ (b) and $PM_{10-2.5}$ (c) at the Norwegian site Birkenes (NO0001R.)

The concentration of OC in PM_{10} is always higher during summer compared to winter for the period 2001 – 2005 at Birkenes. This can be attributed to increased levels of $OC_{PM10-2.5}$ during summer. For $PM_{2.5}$, there is no consistency whether concentrations of OC are higher in summer or in winter, and the difference is typically very small when comparing the seasonal mean concentrations. For EC, the concentration tends to be higher in winter compared to summer for both PM_{10} and $PM_{2.5}$, but this is not a consistent pattern.

From Table 3.4 is can be seen that OC always is the dominant sub fraction of TC at Birkenes, regardless of size fraction. For the period 2001 - 2005, OC accounted for 89 ± 2% of the TC fraction in PM_{10}, whereas the corresponding range for EC was 11 ± 2%. Only minor differences were observed for $PM_{2.5}$ with respect to the relative contribution of EC and OC to TC.

Table 3.4: Relative contributions of EC-to-TC and OC-to-TC for PM_{10} and $PM_{2.5}$ at the site Norwegian site Birkenes (NO0001R) and at the Italian site Ispra (IT0004R).

Site	PM_{10}		$PM_{2.5}$	
	EC/TC (%)	OC/TC (%)	EC/TC (%)	OC/TC (%)
Birkenes (NO01) (2001 - 2005)	11 ± 2	89 ± 2	13 ± 2	87 ± 1
Ispra (IT04) (2003, 2004)	17 ± 0.5	83 ± 0.6	16 ± 0.3	84

Fine ($PM_{2.5}$) OC is the dominant carbonaceous sub fraction, accounting for 67% (2005) to 88% (2001) of OC in PM_{10}. The relative contribution of coarse ($PM_{10-2.5}$) OC to OC in PM_{10} ranges from 11% (2001) to 30% (2005). Coarse OC typically makes a larger contribution during summer and fall at Birkenes, and there are strong indications based on analysis of tracer compounds that it can be attributed to primary biological aerosol particles (Yttri et al., 2006). The finding that as much as 30% of OC in PM_{10} (2005) might be attributed to natural primary sources is substantial and should be explored further, including at other EMEP sites. It also emphasizes the importance of monitoring OC in both $PM_{2.5}$ and PM_{10}, as well as performing additional analysis to improve current understanding of which sources are contributing.

Birkenes (NO0001R) is the only EMEP site that has a time series of EC, OC, and TC, for five years. The time series presented in Figure 3.2 (a) (PM_{10}) and Figure 3.2 (b) ($PM_{2.5}$) show that the concentrations of OC and TC have been decreasing for both PM_{10} and $PM_{2.5}$ for the period 2001 to 2005. The decrease has been most substantial for $PM_{2.5}$. From 2001 to 2005 the concentration of TC in $PM_{2.5}$ dropped by 35%, whereas the corresponding reduction for OC was 36%. For PM_{10}, TC fell by 14% and OC by 18%. The less reduction for PM_{10} can be attributed to the fact that OC in $PM_{10-2.5}$ has increased by a substantial *164%* from 2001 to 2005 (Figure 3.2 c). It could be speculated that the increased concentrations of $OC_{PM10-2.5}$ could follow from climatic changes, such as dry and windy conditions and reduced snow cover, which would promote resuspension of coarse OC from the ground.

Unlike TC and OC, the trends for EC are somewhat less clear. While experiencing reduced concentrations of EC in PM_{10} from 2001 to 2004, the highest annual mean so far was reported for 2005. An increased annual mean concentration of EC was also reported for $PM_{2.5}$ in 2005 compared to 2004.

Characteristic for EC, OC, and TC in PM_{10} and $PM_{2.5}$ is the significant drop in concentration observed for 2004. This reflects what has been observed for PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$, as well as for the major inorganic constituents at Birkenes (Figure 3.3), And also for several other EMEP sites measuring PM.

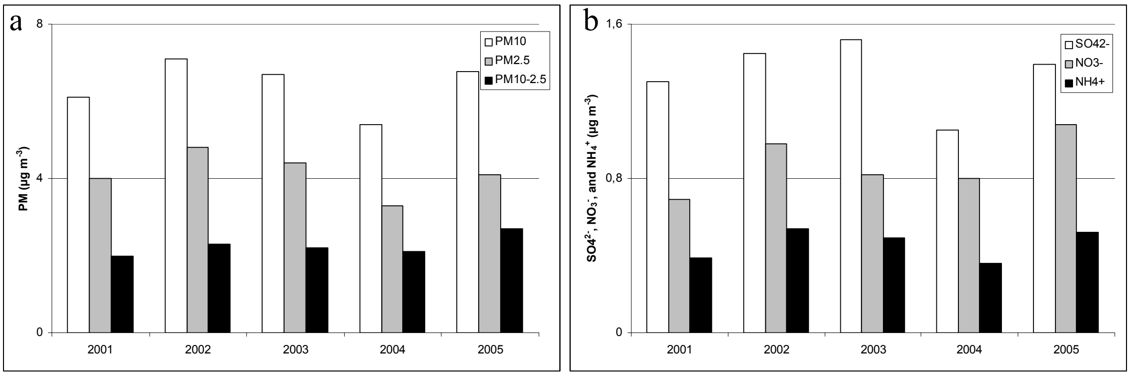


Figure 3.3: Annual mean concentration of PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ (a), and SO_4^{2-}, NO_3^- and NH_4^+ at the Norwegian site Birkenes (NO0001R).

For the period 2001 – 2005, the relative contribution of TCM-to-PM_{10} at Birkenes has decreased steadily from 34% in 2001 to 26% in 2005 (Figure 3.4 a). The decline is in accordance with the reduced concentrations of OC and TC observed in the same period. The relative contribution of TCM to $PM_{2.5}$ follows the same pattern as for TCM-to-PM_{10}, accounting for 47% in 2001 and 30 % in 2005. The relative contribution of TCM to $PM_{2.5}$ is somewhat higher than for PM_{10}, as the carbonaceous material mainly is associated with the fine aerosol. The relative contribution of TCM to $PM_{10-2.5}$ ranges from 9 – 21% for the period in question, and there is an increasing trend corresponding to the major increase in $OC_{PM10-2.5}$ shown in Figure 3.2 c.

Compared to SO_4^{2-}, NO_3^- and NH_4^+, TCM accounts for the largest contribution of mass to PM_{10} at Birkenes (Figure 3.4 b).

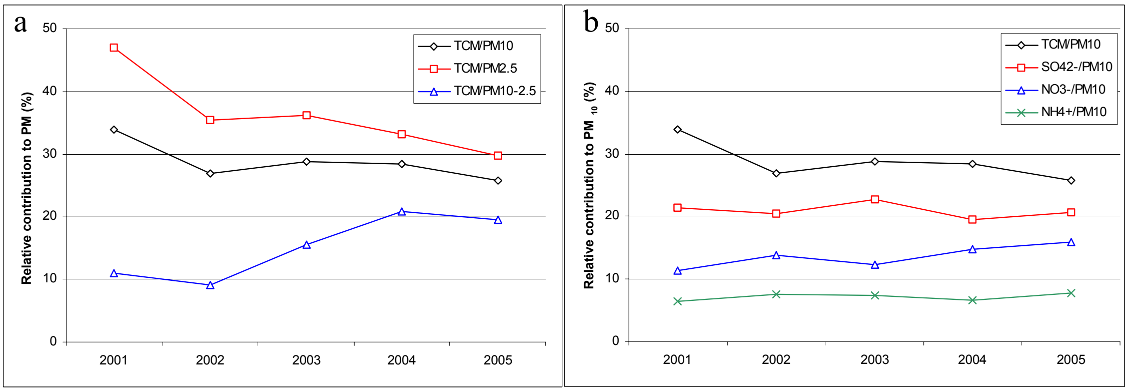


Figure 3.4: Relative contribution of TCM (Total Carbonaceous Matter) to PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ (a) and relative contribution of TCM (Total Carbonaceous Matter), SO_4^{2-}, NO_3^- and NH_4^+ to PM_{10} (b).

3.2.2 EC and OC levels at the Italian site Ispra (IT0004R)

The other site reporting levels of EC, OC and TC for 2004 was the Italian site Ispra (IT0004R). The Ispra atmospheric research station (45° 49'N, 8° 38'E, 209 m asl) is an EMEP station situated in the Po Valley in the northwestern part of Italy. The site is representative for the rural parts of the densely populated central Europe and has been operational since 1985.

For 2004 the annual mean concentration of OC at Ispra was 9.0 μg m^{-3} for PM_{10} and 8.6 μg m^{-3} for $PM_{2.5}$, whereas the corresponding levels of EC were 1.8 μg m^{-3} (PM_{10}) and 1.6 μg m^{-3} ($PM_{2.5}$) (Table 3.5). The annual mean concentration of total carbon (TC) was 10.8 μg m^{-3} for PM_{10} and 10.2 μg m^{-3} for $PM_{2.5}$. For $PM_{10-2.5}$ the annual mean concentration of EC, OC and TC was 0.14 μg m^{-3}, 0.42 m^{-3} and 0.56 μg m^{-3}, respectively.

Table 3.5: Annual mean concentrations of EC, OC, and TC in PM_{10} and $PM_{2.5}$ at the Italian site Ispra (IT0004R) for the years 2003 and 2004 (μg m^{-3}).

Year	PM_{10}			$PM_{2,5}$			$PM_{10-2.5}$		
	EC	OC	TC	EC	OC	TC	EC	OC	TC
2003	1.7	8.3	10.1	1.3	6.6	7.8	0.46	1.8	2.3
2004	1.8	9.0	10.8	1.6	8.6	10.2	0.14	0.42	0.56

The concentrations of EC, OC and TC, observed for PM_{10} and $PM_{2.5}$ in 2004, were all higher than for 2003. For PM_{10} the increase was less than 10% for the three fractions, whereas it was between 20% and 30% for $PM_{2.5}$. This finding contradicts that observed for PM_{10}, $PM_{2.5}$, and the secondary inorganic constituents SO_4^{2-}, NO_3^- and NH_4^+ at Ispra, for which the concentrations decreased going from 2003 to 2004. This might indicate some kind of systematic error in the dataset. For $PM_{10-2.5}$ the concentrations of EC, OC and TC decreased by a factor 3 - 4 from 2003 – 2004. This is in agreement with the reduced concentration observed for $PM_{10-2.5}$.

For 2004 the levels of EC and OC had an obvious seasonal variation with higher concentrations during winter compared to summer, both for PM_{10} and $PM_{2.5}$ (Figure 3.5). This is in agreement with what was observed at Ispra during the previous year (2003). There are indications of a similar seasonal variation for OC in $PM_{10-2.5}$, in particular during 2004, but not that pronounced for 2003 (Figure 3.5).

From Table 3.4 is can be seen that OC was the dominant sub fraction of TC at Ispra for the period 2003 - 2004, accounting for 83 ± 0.6% of TC in PM_{10} and for 84% of TC in $PM_{2.5}$. The corresponding percentages for EC were 17 ± 0.5 % (PM_{10}) and 16 ± 0.3 % ($PM_{2.5}$). The low standard deviations, although for two years only, indicate that there is a very low inter annual variation for the EC-to-TC and OC-to-TC ratios.

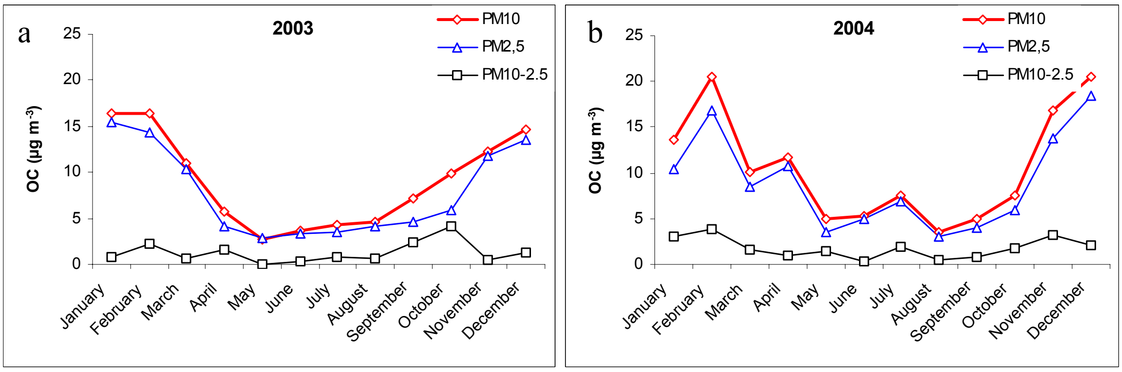


Figure 3.5: Monthly mean concentrations of OC in PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ at the Italian site Ispra (IT0004R) during 2003 (a) and 2004 (b).

The majority of the carbonaceous material was associated with fine aerosols (Table 3.6). However, a surprisingly low $PM_{2.5}/PM_{10}$ ratio was observed for EC (68 ± 0.1%) compared to OC (84 ± 0.9%) and TC (84 ± 0.3%), of which there is no obvious explanation. It should be mentioned though, that only days where data for both EC and OC existed, were included in the annual mean ratio. In addition, days with higher levels of EC, OC, and TC in the fine fraction compared to PM_{10} were excluded, which were quite a few.

Table 3.6: Annual mean $EC_{PM2.5}/EC_{PM10}$, $OC_{PM2.5}/OC_{PM10}$, and $TC_{PM2.5}/TC_{PM10}$ ratios at the Italian site Ispra for the years 2003 and 2004 (%).

Year	$EC_{PM2.5}/EC_{PM10}$	$OC_{PM2.5}/OC_{PM10}$	$TC_{PM2.5}/TC_{PM10}$
2003	68	84	84
2004	68	83	84
Mean ± SD	68 ± 0.1	84 ± 0.9	84 ± 0.3

For 2004 the annual mean concentration of TCM accounted for 47% of PM_{10} at Ispra (Figure 3.6). For $PM_{2.5}$ the contribution was somewhat higher (55%), as the carbonaceous material mainly is associated with fine aerosols at this site. These estimates are rather high in general, and considerably higher than for 2003 and than the estimate reported for TCM-to-PM_{10} for Ispra (36%) during the EMEP EC/OC campaign conducted in 2002–2003. TCM made only a small contribution to $PM_{10-2.5}$ during 2004, accounting for 14%. While the relative contribution of TCM-to-PM_{10} and $PM_{2.5}$ increased from 2003 to 2004, TCM-to-$PM_{10-2.5}$ decreased from 29% to 14%. A conversion factor of 1.6 was used to convert OC to OM for all size fractions at Ispra, whereas a factor of 1.1 was used to account for hydrogen associated with EC (Kiss et al., 2002). The conversion factors for OC reported in literature range from 1.2–2.6, depending on the origin of the aerosols and to what extent they have been aged in the atmosphere (Turpin and Lim, 2001). For the EMEP EC/OC campaign, conversion factors for OC ranging from 1.4–1.8 was calculated for the sites participating based on the relative contribution of WSOC and WINSOC to OC at the various sites (Yttri et al., in prep.). Without doubt, the use of such wide range conversion factors might introduce a significant level of uncertainty to the TCM-to-PM estimates.

TCM accounted for the majority of the mass concentration of PM_{10} at Ispra in 2004, followed by NO_3^- (16%), SO_4^{2-} (10%) and NH_4^+ (7%). Whereas the relative contribution of TCM to PM_{10} increased significantly from 2003–2004, only minor changes were observed for the inorganic aerosol constituents. This collaborates to the indication that the levels of carbonaceous material might have been erroneously high for 2004.

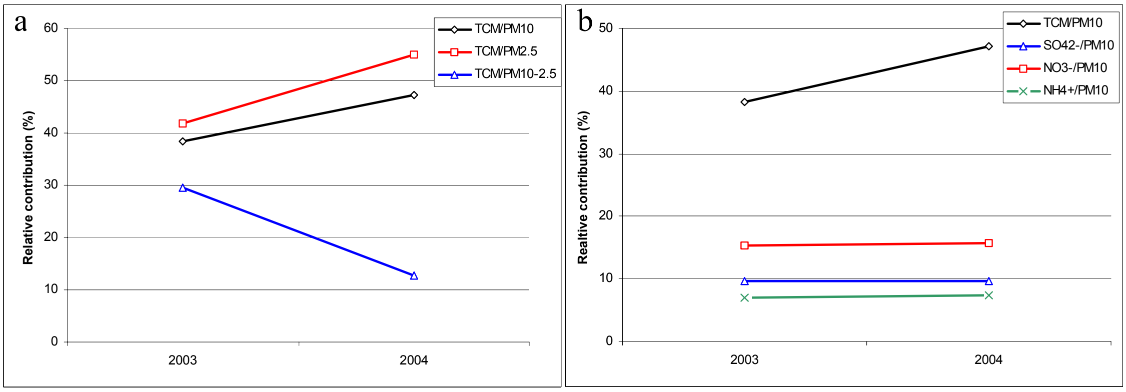


Figure 3.6: Relative contribution of TCM (Total Carbonaceous Matter) to PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ (a) and relative contribution of TCM (Total Carbonaceous Matter), SO_4^{2-}, NO_3^- and NH_4^+ to PM_{10} (b).

3.3 Summary

Using different sampling approaches and different analytical methodology for quantification, it is hard to compare the levels of EC and OC observed at the Norwegian site Birkenes (NO0001R) with those at Ispra (IT0004R). Hence, a comparison on the basis of the more robust parameter TC, avoiding the problems related to the split between EC and OC, is more suitable.

At Birkenes, the annual mean concentration of TC in PM_{10} ranged from 0.9 – 1.3 µg m^{-3} for the period 2001 – 2005. The corresponding range for TC in $PM_{2.5}$ was 0.7 – 1.2 µg m^{-3}. At Ispra, the annual mean concentration of TC in PM_{10} was 10.1 µg m^{-3} in 2003 and 10.8 µg m^{-3} in 2004. For $PM_{2.5}$ the annual mean concentration of TC was 7.8 µg m^{-3} in 2003 and 10.2 µg m^{-3} in 2004. The annual mean concentration of TC in PM_{10} and $PM_{2.5}$ reported for Ispra during 2003 – 2004, was approximately nine times higher than that reported for Birkenes for the period 2001 – 2005. This finding confirms that of the EMEP EC/OC campaign, showing that these two sites are in the opposite ends of the concentration range with respect to TC (and for EC and OC) in Europe.

Seasonal variations were observed at both sites. At Ispra, levels of EC and OC were increased during winter for both PM_{10} and $PM_{2.5}$, and there were indications of a similar seasonal variation for OC in $PM_{10-2.5}$. At Birkenes, concentrations of OC in PM_{10} were always higher during summer compared to winter. This was attributed to increased levels of $OC_{PM10-2.5}$ during summer. For EC, the concentrations tended to be higher in winter compared to summer for both PM_{10} and $PM_{2.5}$, but there was no consistent pattern.

OC was the dominant sub fraction of TC at both Birkenes and Ispra, regardless of size fraction. For the period 2001 – 2005, OC accounted for 89 ± 2% of the TC fraction in PM_{10}, whereas the corresponding range for EC was 11 ± 2%. Only minor differences were observed for $PM_{2.5}$ with respect to the relative contribution of EC and OC to TC.

At Ispra, OC accounted for 83 ± 0.6% of TC in PM_{10} for the period 2003 – 2004, whereas 17 ± 0.5% could be attributed to EC. As for Birkenes, there were only minor differences between PM_{10} and $PM_{2.5}$ with respect to the relative contribution of EC and OC to TC at Ispra.

As expected, carbonaceous material was mainly associated with the fine aerosol at both sites. On an annual basis, 67% to 89% of OC in PM_{10} was associated with fine aerosols at Birkenes for the period 2001 – 2005, whereas the corresponding percentage for Ispra was 83% for 2003 and 84% for 2004. At Birkenes, the relative contribution of coarse OC to OC in PM_{10} has increased steadily from 11% in 2001 to 33% in 2005. There are strong indications based on analysis of tracer compounds that this can be attributed to primary biological aerosol particles. The finding that 30% of OC in PM_{10} (2005) might be attributed to natural primary sources is substantial. Fine mode EC accounted for more than 80% of EC in PM_{10} at Birkenes on an annual basis for the five years of measurements reported. Only 68% of EC in PM_{10} could be attributed to fine EC for the two years of measurements (2003 – 2004) reported for Ispra.

The five-year time series at Birkenes show that the concentrations of OC and TC have been decreasing for both PM_{10} and $PM_{2.5}$ for the period 2001 to 2005. The decrease has been most substantial for $PM_{2.5}$. From 2001 to 2005 the concentration of TC in $PM_{2.5}$ dropped by 35%, whereas the corresponding reduction for OC was 36%. For PM_{10}, TC fell by 14% and OC by 18%. The less reduction for PM_{10} can be attributed to the fact that OC in $PM_{10-2.5}$ has increased by a substantial 164% from 2001 to 2005

TCM was the major contributor to the PM mass for the periods investigated at Birkenes and Ispra. At Birkenes, the relative contribution of TCM to PM_{10} has decreased steadily from 34 % in 2001 to 26 % in 2005. The relative contribution of TCM to $PM_{2.5}$ follows the same pattern as for TCM-to-PM_{10}, accounting for 47 % in 2001 and 30 % in 2005. For the period in question, the relative contribution of TCM to $PM_{10-2.5}$ has increased from 9% in 2001 to 21% in 2005.

For 2004 the annual mean concentration of TCM accounted for 47% of PM_{10} at Ispra. For $PM_{2.5}$ the contribution was somewhat higher (55%). These estimates should be regarded as rather high in general, and are considerably higher than for 2003 (TCM-to-PM_{10} = 38% and TCM-to-$PM_{2.5}$ = 42%). TCM made only a small contribution to $PM_{10-2.5}$ during 2004, accounting for 14%, while it accounted for 29% in 2003.

4. Development and validation of standardised protocols for sampling OC and analysing OC+EC for the EMEP network

by J.P. Putaud and F. Cavalli

4.1 Objectives

Collecting ambient aerosols for the purpose of quantifying OC is challenging because of the need to separate particulate OC from vaporous OC without influencing the gas/particle distribution for each of the compounds. During sampling the organic vapour phase components may adsorb onto the filter or on the deposited particulate matter impacted on the filter. This is commonly known as a positive artefact as the amount of carbonaceous material on the filter increases although no particulate matter has been added. Semi-volatile OC that is condensed onto particles trapped by the filter may evaporate during continued sampling, leading to a loss of carbonaceous material that was initially associated with the particles. This loss of carbonaceous material is denoted as a negative artefact. It should be added that particulate matter may react (mainly through oxidation) with vaporous compounds that pass through the filter during sampling and that this might change the chemical composition of the deposited material. Such reactions may result in both positive and negative artifacts, and care should be taken especially when measuring individual compounds.

Estimates of artefact contribution to measured particulate organic carbon concentrations range from -80 to +100%. There is, however, no consensus, whether it is the positive or the negative artefact that prevails. Various sampling trains exist that are supposed to account for the sampling artefacts of OC (Mader et al., 2003), but the performance of these approaches are disputed. “Artefact-free” sampling trains including denuder(s), filter(s), and sorbent(s) have been developed and used in the USA for ca. 20 years. However, these sampling trains are not well characterised with respect to their efficiency, lifetime, etc., hence, they are not widely used at the US-EPA supersites for instance. Furthermore, it might be risky to extrapolate the results obtained at a limited number of sites in the USA to the various climatic and pollution environments encountered in Europe, simply because the carbonaceous aerosol mix is likely to differ substantially. In Europe, various sampling techniques with insufficiently defined efficiency and/or artefacts are currently operated. Indeed, there are currently no standardized procedures for sampling and analysing the carbonaceous aerosol fraction, which implies that data obtained by different laboratories at various sites are affected by unknown errors. This status arises from the fact that carbonaceous particulate matter consists of thousands of different molecules, ranging from semi-volatile to refractory species, which makes it impossible to define and produce suitable standards for atmospheric organic and elemental carbon. Therefore, sampling and analytical techniques cannot be validated through laboratory tests.

In order to provide EC/OC data of high quality, a standardized protocol needs to be established. The development and validation of such a protocol is currently taken place within the frame of EUSAAR (*European Super-sites for Atmospheric Aerosol Research*), an EC-funded Integrated Infrastructure Initiative, which gathers 20 stations, of which nine are EMEP aerosol supersites. The participation of EMEP sites is particularly important with respect to subsequent implementation of the protocol to the EMEP monitoring network, and because sampling of particulate OC seems to pose particular challenges in areas with low aerosol loadings. The objectives of EUSAAR relative to the carbonaceous aerosol issues were listed as follows:

1. Deliver a sampling train for particulate carbonaceous matter, well characterised with respect to particle trapping efficiency, positive and negative artefact assessment, lifetime, running cost, maintenance, etc…
2. Define a standardised thermal programme for the thermal-optical analysis of EC and OC
3. Assess the comparability of this standard analytical method with the other methods used within the network and periodically check their consistency (intercomparisons)
4. Design a standardised method for analysing back up sorbents.
5. Get the overall standardised procedure for EC+OC determination adopted by the EMEP task force on measurements and modelling and the WMO Global Atmospheric Watch scientific advisory group.

4.2 Workplan

The workplan involves of course primarily the EUSAAR partner participating to the “carbonaceous aerosol” activity. However, additional EMEP stations might be included to the testing and inter comparison activities at an early stage if they wish so.

1. Based on the experience gained in the USA over the two last decades and in Europe over the last few years, a sampling train prototype is to be tested at JRC-Ispra. When satisfactory results are achieved, JRC will propose the first sampling train (denuder – filter – sorbent) to be tested at various sites of the EUSAAR network.
2. Tests including simultaneous sampling with various denuder – filter – sorbent combinations will be performed over different seasons at the various sites of the network participating to this activity. Feedbacks from the test stations will lead to the definition of the best affordable sampling train, which will be fully characterised in term of efficiency, positive and negative artefacts, lifetime, running cost, maintenance, etc.
3. Standard Operational Procedures for the validated sampling train will be edited and disseminated within the EMEP community, beyond the EUSAAR network. Technical training sessions for using this sampling train will be organised.
4. A harmonised procedure for the thermo-optical analysis of particulate elemental carbon (EC) and organic carbon (OC) will be defined, based on the experience of the EUSAAR partners participating to this activity, and on crossed analysis of aerosol standards.

5. Periodic intercomparisons of all the instruments used for EC and OC analyses within the EUSAAR network, will be carried out by circulating selected aerosol filters and standards among EUSAAR partners. This activity might also include other EMEP stations where EC+OC measurements are performed.
6. A standardised analytical procedure for analysing volatilised OC from the back-up sorbent will be decided upon, based on the results of concerted investigations performed by the EUSAAR partners participating to this activity.

4.3 Status

Recommendations for sampling and analysing EC+OC have been put forward during the TFMM meeting in Helsinki (May 2006). This set of recommendation should be regarded as a first step towards the definition of a standardised method for the measurement of carbonaceous aerosol within the EMEP network, and has not yet been approved by the EMEP steering body. These recommendations are based on a literature survey and on the experience we gained during the last years. They aim at getting a better grip on the quality of the carbonaceous aerosol data collected within EMEP, and to ensure a certain level of comparability among sites. These recommendations include:

1. Sampling time: not more than 24 hrs.
   To avoid as far as possible drastic changes in weather and air chemistry conditions and to be consistent with other EMEP filter sampling activity to allow for comparisons (mass closure)

2. Sampling substrate: back-to-back pre-fired quartz fibre filters (except if denuder is available).
   The back-to-back filter sampling technique has proved to lead to comparable results as sampling techniques including denuders. This is the only simple technique to address sampling artefacts currently available.

3. Daily calibration of TC with external (traceable) standards.
   Checking the instrument calibration with external standards is a common practise, which should be applied to the measurements of carbonaceous aerosols within EMEP.

4. EC/OC split quality assurance using standards (pure organics and organic mixtures should be detected as 100% OC, pure EC should be detected as > 95% EC).
   This is an essential requirement for defining an acceptable method for EC and OC analysis. However, there is no guarantee that methods fulfilling these requirements would lead to comparable results when applied to atmospheric samples.

5. Every year comparison of the EC/OC split among laboratories reporting EC and OC data to EMEP, based on test filter samples distributed by NILU.
   This would simply be an extension of the standard exchange practise-taking place within EMEP to the analysis of carbonaceous aerosols.

These recommendations will be tested and refined in the framework of EUSAAR.

EUSAAR started in Apr. 2006 and will last for five years. A sampling train prototype has already been tested during winter conditions at the Italian site IT04 (Ispra). Preliminary results are presented in the next section. Summertime tests have been delayed due to the technical problems with the EC/OC analyser. Indeed, sampling tests cannot be efficiently carried out if collected samples cannot be analysed immediately, so that the results of each test are known before starting the next one. If the performance of the currently tested sampling train is satisfactory, it is expected that it will be presented and proposed for validation to the EUSAAR partners participating to this activity by the end of 2006.

The work dedicated to the design of a standard method for OC and EC analysis has been going on since October 2005, but has temporarily been stopped due to instrumental problems. A method recently designed at JRC-Ispra can achieve recommendation # 4 (pure organics (or organic mixtures) detected as 100% organics and pure elemental carbon detected as > 95% EC), but different temperature programs might do it as well.

Preliminary results on carbonaceous sampling and analyses are presented below. They will be discussed at the next EUSAAR technical workshop in autumn 2006.

4.4 Preliminary results

4.4.1 Sampling train

Based on previous experience, it was decided to first test a sampling train (Figure 4.1) made up of:

1. a Carbon honey comb monolith denuder (commercially available), included to remove the volatile organic compounds (VOCs) from the air stream, which otherwise would condense on the quartz fibre filter and on the Carbon-impregnated glass fibre back up filter
2. a quartz fibre filter, which is currently the substrate universally used to collect aerosols for OC+EC analyses (impactor foil excepted)
3. a C-impregnated glass fibre filter (formerly commercially available) to trap the particulate OC volatised from the quartz fibre filter

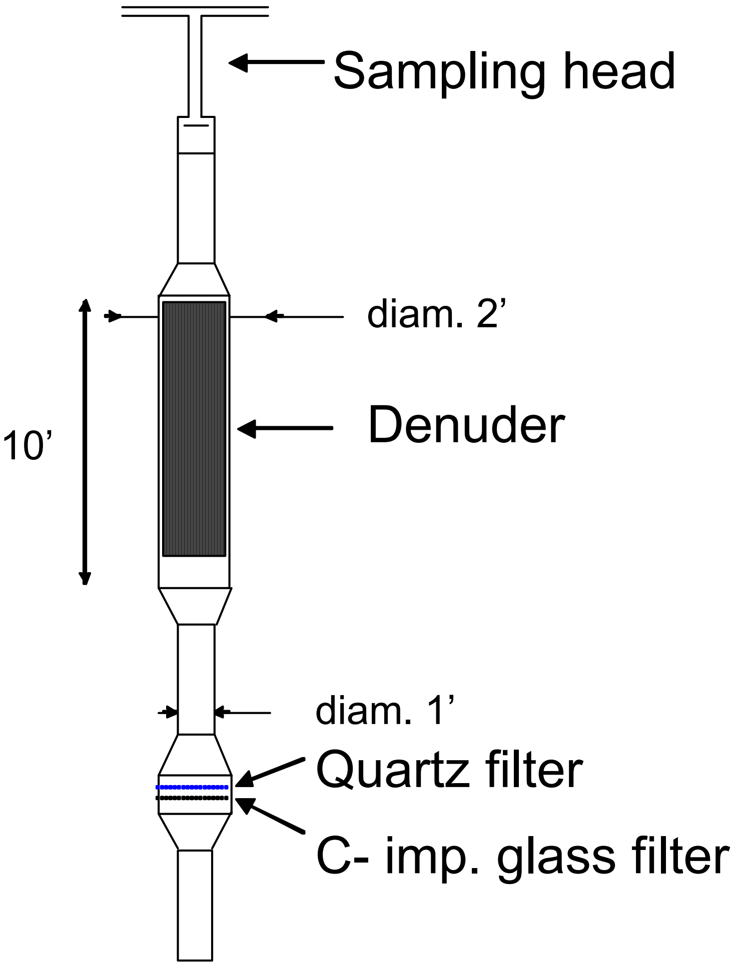


Figure 4.1: Sketch of the sampling train tested at IT04.

The denuder efficiency was assessed operating several samplers with different configurations in parallel (simultaneous sampling). The testing took place during winter-like conditions in March 2006 at the Italian site IT04 (Ispra). Samples were collected for a 24 hr sampling period (from 08:00 to 08:00 UTC), with a 24 hrs averaged temperature ranging from 5°C (sample 4) to 9°C (sample 11). Figure 4.2 shows that during this period, positive artefacts accounted for 10% to 50% of the total carbon collected on a plain quartz fibre filter, with a maximum contribution during the less polluted periods. The denuder reduced the amount of VOC collected by the quartz fibre filter by 65 to 85% (median = 78%). The worst performance was observed when the TC concentration was low (below 3 µg/m³). However, the use of this denuder would have reduced the contribution of the adsorbed VOC (positive artefact) to the amount of C collected by the quartz fibre filter to 2-10 % for the whole range of concentrations observed, which is a very promising achievement This result is to be confirmed in summertime conditions.

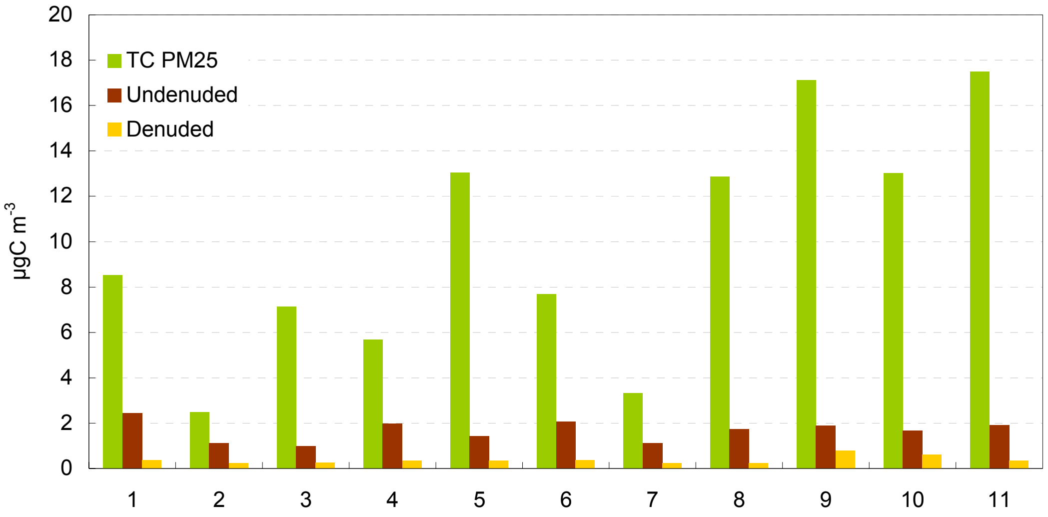


Figure 4.2: Concentrations of carbon collected with a PM2.5 inlet on quartz fibre filters (green), and by quartz fibre filters sampling particle-free air without (brown) and with (gold) a C-monolith denuder.

It was shown that the positive artefact mainly was due to the most volatile OC fractions, which evolved at T < 300°C, and that these were the fractions most efficiently removed by the Carbon-honey comb denuder.

The use of a denuder is expected to increase the negative artefacts, as air depleted in VOC passes through the filter where particulate organic matter is collected. Assessing the amount of carbon lost from this filter by volatilisation is therefore even more essential when a using a denuder. Carbon-impregnated glass fibre filters (CIG) were tested as sorbents during March 2006 at Ispra. For such filters, the analysis temperature program cannot go beyond 400°C, as it would destroy the filter (and provide artificially high levels of carbonaceous matter due to the thermal destruction of the carbon impregnation). Still, 400°C should be enough to volatilise semi-volatile compounds. Figure 4.3 shows that the Carbon-honey comb monolith denuder removed 55% to 75% of the VOCs, which was trapped by the CIG filters. The denuder was most efficient in removing compounds evolving above 300°C. It was found that the efficiency of the CIG filters to collect the VOCs going through the denuder was quite low, only 11 % on average. This was elucidated by comparing the amount of C found on two back-to-back CIG filters. Experiments have been carried out to determine the efficiency of CIG filters in trapping C volatilised from a Quartz filter, but the samples have not yet been analysed.

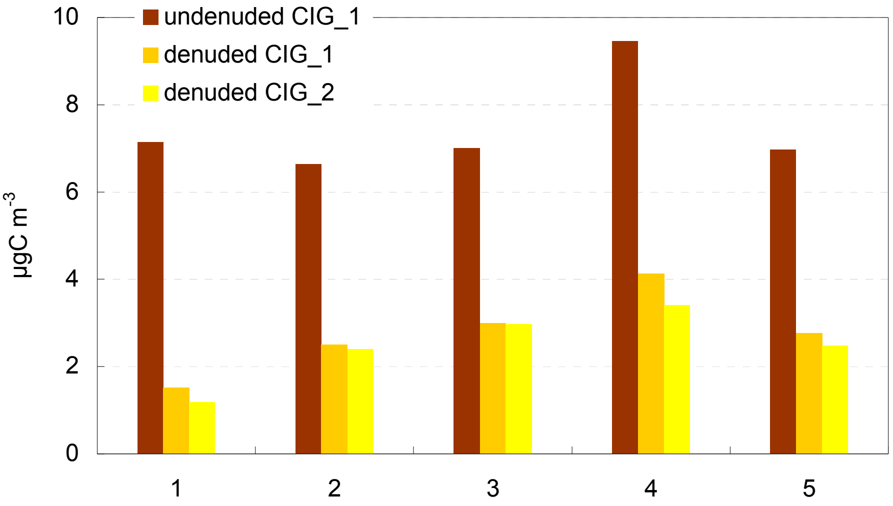


Figure 4.3: Concentrations of carbon measured from C-impregnated glass fibre filters sampling particle-free ambient air (brown) and particle free ambient air through the C-monolith denuder (gold and yellow for the fist and second CIG filter, respectively).

4.4.2 Analytical method

There have also been carried out tests using the thermal-optical EC/OC analyser from Sunset Laboratory in order to develop an artefact free analytical methodology for analysis of EC and OC. This instrument offers the possibility of monitoring variations in the sample reflectance and transmittance simultaneously during the analysis. This feature is used to correct for generation of artificial EC generated due to charring of OC during the analysis.

We observed that both the reflectance and transmittance of samples collected at Ispra generally increased when the oven temperature overpassed 550°C while the career gas was still pure He. This can be interpreted as combustion of photon absorbing material (EC), perhaps catalysed by other constituents of the particulate matter collected on the filter. It was shown by analysing pure EC synthesised in the instrument itself that 20% of the EC could evolve in He already at 850°C (Figure 4.4). This fraction dropped to 2.5% at 650°C, and to 0% (below detection limit) at 550°C.

It was also observed that humic and fulvic acids, which are among the less volatile organic molecules, completely evolved at 550°C in He. However, other molecules such as poly-acids might not evolve at 550°C. Indeed, we observed that a larger amount of OC was measured in samples collected at Ispra when shifting the last temperature plateau from 550°C to 650°C. The best compromise for volatilizing as much OC as possible without burning EC during the He phase seems therefore to be 650°C for the time being.

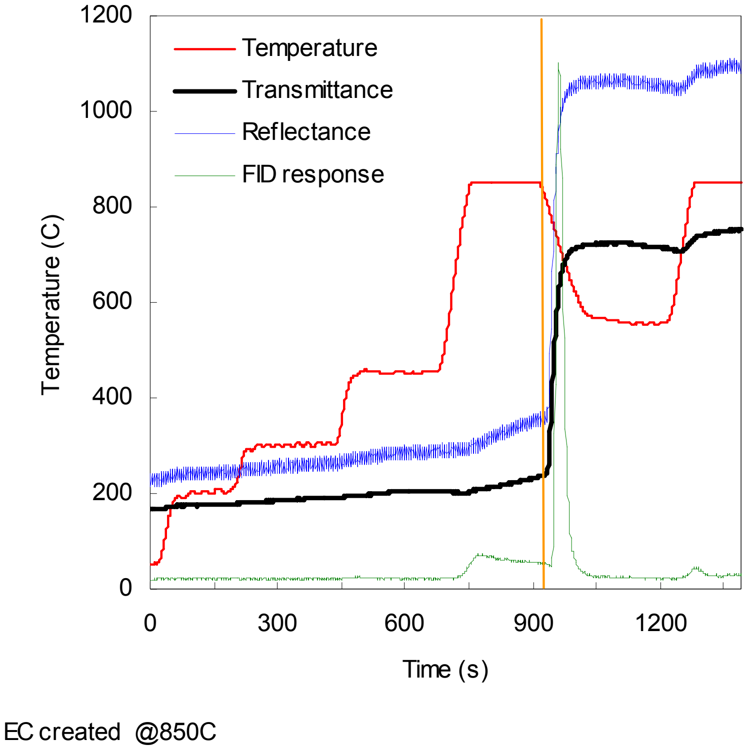


Figure 4.4: Thermogram of synthetic EC showing evolution of EC in He at 850°C.

Also the split between EC and OC in thermal-optical instruments depends strongly on the charring correction. Attention should be paid to the way the software calculates the split point between pyrolised carbon and elemental carbon, as it depends a lot on the laser signal value taken as the initial value (which is actually defined as the maximum value recorded during the first 70s of the analysis). Indeed, for determining this split point, the software looks backward from the end of the thermogram for a laser signal value (transmittance or reflectance) equal to the initial value. If the laser signal drifts e.g. upwards during the initial 70s, the split point will be shifted towards the end of the thermogram (and therefore EC will be underestimated) compared to a case in which the laser signal would be constant during the first 70s. On the other hand, manual definition of the split point is not recommended, as it would not take into account the changes in laser emission with temperature. Regarding the choice between the use of transmittance or reflectance for charring correction, there is currently no reason to believe that either of the two provides a more correct estimate of the split point than the other. We have observed though that EC determined using reflectance is in general better correlated with the equivalent Black Carbon determined by an Aethalometer™ than EC determined using transmittance. Charring correction using reflectance generally leads to higher EC values than charring correction using transmittance.

5. Aerosol optical depth in the European Arctic Region

by C.L. Myhre, K. Stebel, C. Toledano, V.E. Cachorro, Á.M. de Frutos, C. Forster, J. Schaug

5.1 Introduction

In recent years there has been an increased focus on climate change in the Arctic region. In particular, the extensive ACIA-report (ACIA, 2005) pointed to many challenging topics. Key findings are that the Arctic climate is warming rapidly and larger changes are projected. Further, the warming is faster than previously estimated and it will have global implications. Arctic vegetation zones are expected to shift, bringing wide-ranging impacts on animal, plants and humans. The reductions of sea ice will very likely increase marine transport and access to resources in the region.

In the investigations of climate change, aerosols are of vital interest as they have a direct impact on the radiative balance by scattering of solar radiation and absorption of solar and thermal radiation. The dominating process depends on the absorption and scattering characteristics of aerosols defined by their composition, shape, and phase. In the Arctic, knowledge about the optical properties of aerosols is of particular importance, due to the special surface conditions in this region. Ice and snow give rise to very high albedos and water to very low albedo dominating the surface albedo in the region. Together with the albedo and clouds, aerosols are an important factor in controlling the UV radiation as well.

At present, local and regional anthropogenic sources are almost absent in the Arctic region. Arctic haze commonly present in springtime is a well-known result of long-range transport into the region from mid-latitude sources in Russia, Europe and North America. In combination with transport there are favorable meteorological conditions with strong inversion in late winter and spring resulting in the high aerosol levels.

Recent studies indicate that boreal forest fires might be an important source of light absorbing aerosols containing black carbon (BC) in the Arctic region during summer (Stohl et al., 2006). In the Arctic, the importance of black carbon aerosols is even larger than elsewhere because atmospheric absorption is enhanced by the high surface albedo of snow and ice. Furthermore, the albedo of snow and ice can be reduced by the deposition of BC (Hansen and Nazarenko, 2004).

This chapter focuses on optical properties of aerosols in the European Arctic, particularly aerosol optical depth (AOD) measurements, which is primary to the information for the direct effect of aerosols. The measurements are discussed in relation to observations of chemical constituents and transport into the region.

5.2 Observations of aerosol optical properties in the European Arctic sector

In a global perspective, satellites are becoming increasingly important for measuring total columns and vertical profiles of aerosols (e.g. MODIS, MISR, CALIPSO). However, satellite measurements of aerosol properties in Polar Regions are very difficult due to the special conditions with high surface albedo, large solar zenith angle, long path through the atmosphere, and low background aerosol concentrations. Consequently, ground-based networks are of particular importance in these regions.

A large number of ground-based sites around the world measure aerosols optical properties. AERONET[1] (Aerosol Robotic Network, Holben et al., 1998), aims at the assessment of aerosol properties and the validation of satellite retrieval of aerosols optical properties. The network compiles data from a large number of sites around the globe, including about 60 sites in Europe, and one station, Hornsund, (77°N, 15°E), in the European Arctic.

The World Meteorological Organization, Global Atmospheric Watch (WMO GAW) programme runs a small trial network of 13 background stations operating sunphotometers (Precision-filter-radiometer, PFR) around the world (see Wherli, 2005). Six sites are or will be operated in Europe. Data are available through a web-site[2]. Two sites are located in the Arctic sector, the site in Ny-Ålesund and one in Sodankylä in Northern Finland.

AOD measurements have been included in EMEP since 2004 at the level 3 of monitoring. Level 3 is voluntary, but considered important for the understanding of processes and for controlling the transboundary air pollution. Level 3 activities will typically be undertaken by research groups, it may also include campaign data, and will provide additional data that are not part of the networks above.

The present report presents AOD measurements from the Sverdrup station in Ny-Ålesund (78.9°N, 11.9°E) located close to the EMEP-station at the Zeppelin Mountain. Additionally, measurements are compared to campaign observations from the sub-Arctic observatory at ALOMAR (69.2°N, 16.0°E) during summer 2005.

5.3 Measurements of optical properties in Ny-Ålesund during the period 2002-2005 with the WMO-GAW instrument

5.3.1 Location and experimental details

The PFR measurements in Ny-Ålesund are part of the global network of aerosol optical depth (AOD) observations, which started in 1999 on behalf of the WMO GAW program. The instrument is located at the roof of the Sverdrup station, Ny-Ålesund, close to the EMEP station at the Zeppelin Mountain. The PFR has been in operation since May 2002. In Ny-Ålesund the polar night lasts from 26th October to 16th February, leading to short observational seasons. However, during summer it is possible to measure day and night if the weather conditions are satisfactory. The instrument measures direct solar radiation in four narrow spectral bands centred at 862, 501, 411, and 368 nm. Data quality control includes instrumental control such as detector temperature and solar pointing control as well as objective cloud screening. The signals are recorded every 1.25 seconds and are given as one minute averages. In the calculations of the AOD values it is necessary to correct for the absorption of UV by ozone. For this we have used

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[1] http://aeronet.gsfc.nasa.gov
[2] http://wdca.jrc.it/

daily ozone values from TOMS[3] in the calculations. AOD measurements were obtained only at 38 % of the days in 2005 due to bad weather conditions. The number of days where measurements can be performed is reduced due to foggy weather conditions. Moving the instrument to the EMEP station on the Zeppelin Mountain can increase the number of observations during clear sky conditions.

5.3.2 AOD Measurements in 2005 in Ny-Ålesund

Hourly AOD values measured in Ny-Ålesund by the PFR-instrument are presented in Figure 5.1 for three different wavelengths. The observations show increased aerosol levels during the Arctic haze period during spring. However, there are also short episodes later in the year with elevated levels of AOD. These episodes are indicated in Figure 5.2 together with the Ångstrøm exponents and daily filter analysis of SO_4^{2-}, NO_3^-, and Cl^- from the EMEP-filterpack measurements at the Zeppelin observatory. The Ångstrøm exponent, α, provides information about the size of the aerosols. Larger values of α imply a relatively high ratio of small particles.

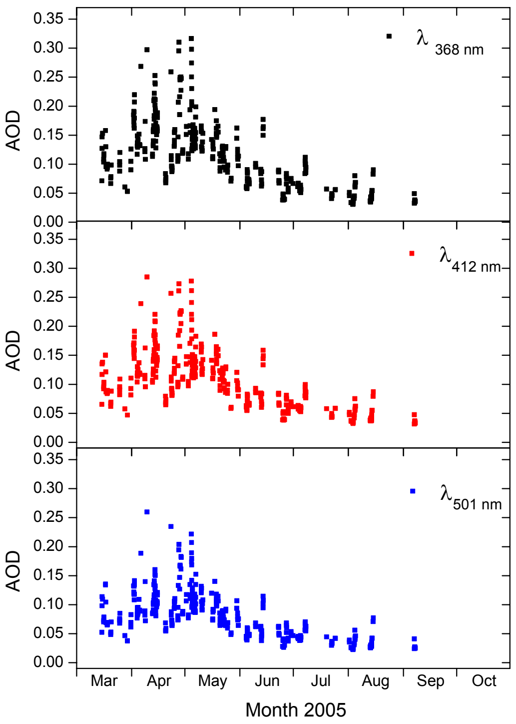

Figure 5.1: Hourly AOD values measured in Ny-Ålesund during 2005.

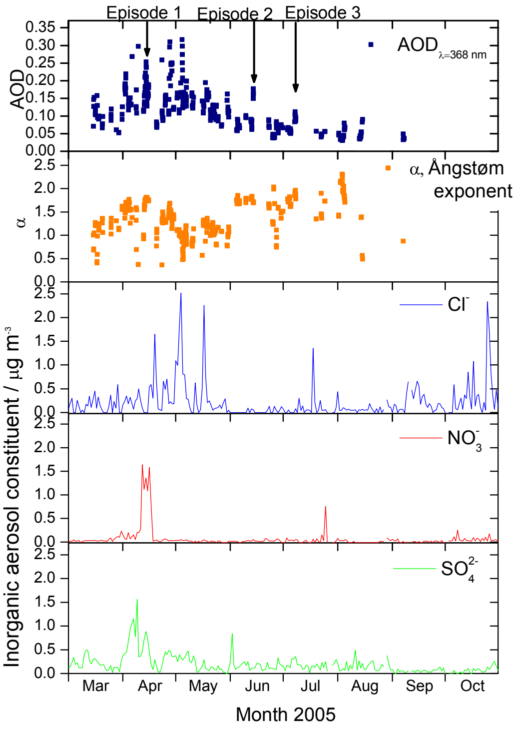

Figure 5.2: AOD measurements from Ny-Ålesund in 2005 together with Ångstrøm exponents and inorganic aerosol constituents from the Zeppelin observatory.

Figure 5.2 shows that the episodes do not necessarily coincide with increased levels of inorganic aerosol constituents measured at the Zeppelin station. Table 5.1 summarises the characteristics of the episodes.

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[3] http://toms.gsfc.nasa.gov/ozone/ozone.html

Table 5.1: Characteristics of selected episodes in Ny-Ålesund 2005.

	Date	**Max. $AOD_{\lambda=501\ nm}$**	**Ångstrøm exponent, α mean values**	**Inorganic constituents**
Arctic haze	March - April	0.25 (average 0.12)	1.18 σ = 0.27	Medium
Episode 1	13 -14 April	0.16	1.73 σ = 0.08	High
Episode 2	12 - 14 June	0.11	1.77 σ = 0.06	Low
Episode 3	7 - 8 July	0.08	1.88 σ = 0.06	Low

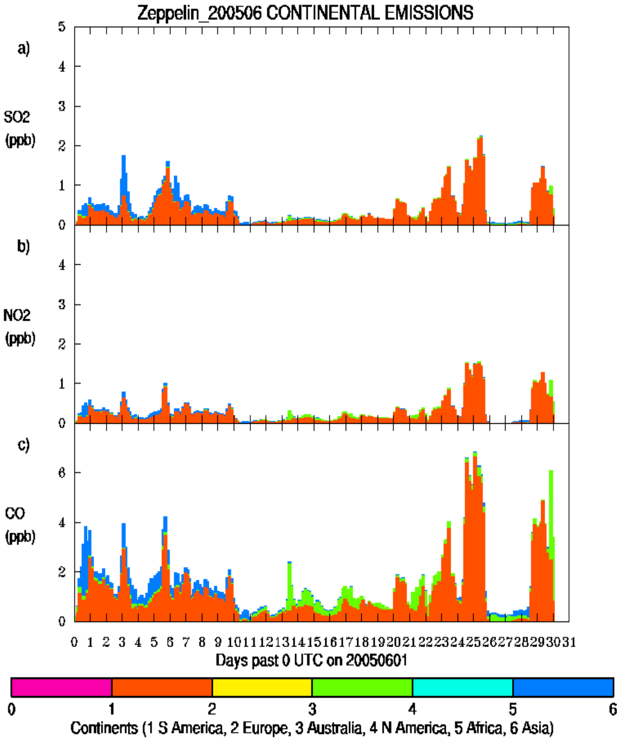

Figure 5.3: The continental emissions influencing the air masses arriving daily at Zeppelin in June 2005 (Episode 2).

The Ångstrøm exponents were relatively high during the episodes indicating small aerosols typical for long-range transport. During episode 2 and 3 very low concentrations of inorganic aerosols constituents were detected at the Zeppelin observatory. To interpret the episodes and the influence of transport on the measurements taken at the EMEP station at the Zeppelin Mountain we have performed backward simulations with the Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005). The model results and a description of the simulations are available at the web page http://zardoz.nilu.no/~andreas/STATIONS/ZEPPELIN/index.html. Figure 5.3 shows the anthropogenic emission contribution for SO_2, NO_2 and CO from the different continents in ppb arriving at Zeppelin in June 2005. SO_2 and NO_2 are tracers for inorganic aerosol constituents while CO is a good tracer for absorbing aerosols containing BC. The Figure shows that during episode 2, 12th – 14th June, there were almost no transport of SO_2 and NO_2 to Zeppelin. This is consistent with the low measured values of inorganic aerosol compounds. However, at the same time there was a contribution of CO mainly from North America, suggesting that the elevated AOD measurements at the nearby station Ny-Ålesund were due to North American anthropogenic emissions The analysis of episode 1, 13th – 14th April (see web page), indicates that the dominating anthropogenic source for both inorganic compounds and CO was Europe and that there was only a small contribution from Asia. For episode 3, 7th – 8th July (see web page), the main source of all compounds considered here seemed to be Europe with an additional small contribution of CO from North America.

5.3.3 AOD measurements during the period 2002-2005

Figure 5.4 presents the AOD measurements at 501 nm in Ny-Ålesund for the years 2002-2005.

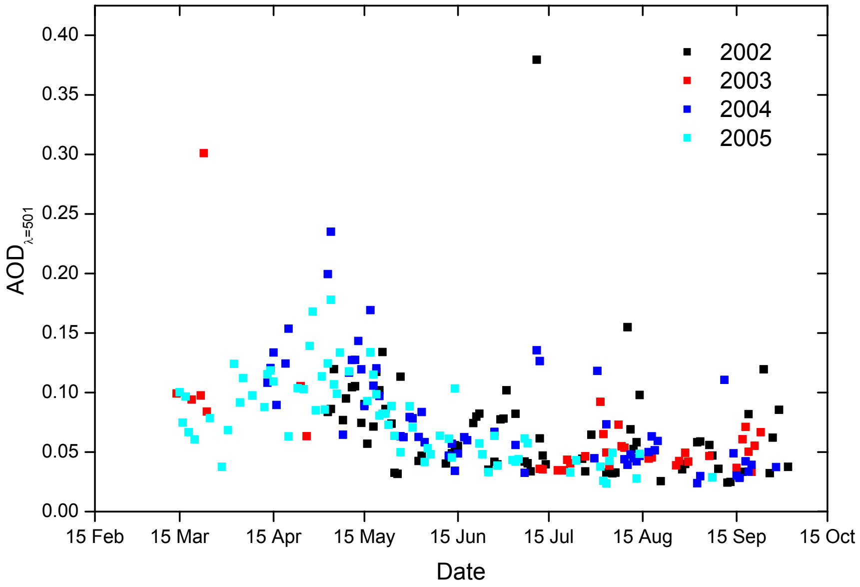

Figure 5.4: Daily average aerosol optical depth (AOD) measured in Ny-Ålesund during 2002-2005.

As expected the AOD values are considerable higher during the Arctic haze period. Yet,

Figure 5.4 illustrates that there are several episodes during these years with short-term elevated AOD values in the summer and autumn as well. Analyses of such episodes are important to understand the effect of pollution transported into the region. Stohl and co-workers (Stohl et al., 2006) analysed the observed episode in the end of July 2004. They showed that emissions from boreal forest fires in North America, with light absorbing aerosols containing BC, was transported into the region and very likely explain the elevated AOD levels.

The time series of four years is too short for trend analysis. We have calculated seasonal and annual mean AOD values to compare the years and the seasonal variations. Annual mean values and mean values for the Arctic haze period and the summer months are presented in Table 5.2.

Table 5.2: Annual mean values and mean values for the period March – May and June – August. The numbers in brackets are the number of days with measurements.

Year	Mean March-May (No. of days)	Mean June-Aug (No. of days)	Annual mean (No. of days)
2002	0.09 (19) $\sigma = 0.027$	0.06 (30) $\sigma = 0.058$	0.07 (72) $\sigma = 0.047$
2003	0.12 (7) $\sigma = 0.080$	0.04 (20) $\sigma = 0.014$	0.06 (35) $\sigma = 0.047$
2004	0.12 (23) $\sigma = 0.042$	0.06 (27) $\sigma = 0.026$	0.08 (60) $\sigma = 0.045$
2005	0.10 (43) $\sigma = 0.029$	0.04 (26) $\sigma = 0.016$	0.08 (70) $\sigma = 0.035$

5.4 AOD measurements at ALOMAR

Figure 5.5: *The location of ALOMAR (blue star) and the EMEP observatory in Ny-Ålesund (red star).*

The ALOMAR observatory managed by Andøya Rocket Range[4] is located at the Atlantic coast of Norway at 69.2°N, 16.0° E, 380 m. a.s.l., 300 km north of the Arctic Circle. Figure 5.5 shows the location of ALOMAR and the EMEP station in Ny-Ålesund. From the end of May to the end of July the sun is 24 hours above the horizon, with a maximum elevation during the solstice of 42° at noon and 2° at midnight. The climate is strongly influenced by the Gulf Stream, which provides mild temperatures during the entire year, with average temperature of – 2°C in January and 11°C in July.

The instrument used is a Cimel Electronique CE-318 sun photometer with spectral interference filters centred at 340, 380, 440, 501, 675, 870 and 1020 nm for aerosol measurements (Toledano et al., 2006). The measurements were carried out during summer campaigns in 2002, 2003 and 2005. In summer 2005, the campaign lasted from 1st of June until 26th August, with 24h measurement schedule during the midnight sun period. The AOD observations during the summer 2005 confirmed the results from previous years, showing higher levels of AOD during June (0.12 in 440 nm) and concentrations that slightly decreases throughout the summer. The associated Ångstrøm exponent (mean 1.67) was rather high indicating the predominance of fine particles. The Cimel sun photometer is deployed on a permanent basis at ALOMAR from July 2006 for continuous monitoring of aerosol optical properties.

5.5 Aerosols optical properties in Ny-Ålesund compared to ALOMAR

Comparison of AOD data from Ny-Ålesund and ALOMAR is interesting with respect to transport and aerosol sources in the European Arctic region. However, it is difficult due to few overlapping dates and the variability of the weather

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[4] http://alomar.rocketrange.no/

conditions in these geographical areas. A comparison of AOD measurements from summer 2005 at ALOMAR and Ny-Ålesund is presented in Figure 5.6 together with the Ångstrøm coefficients.

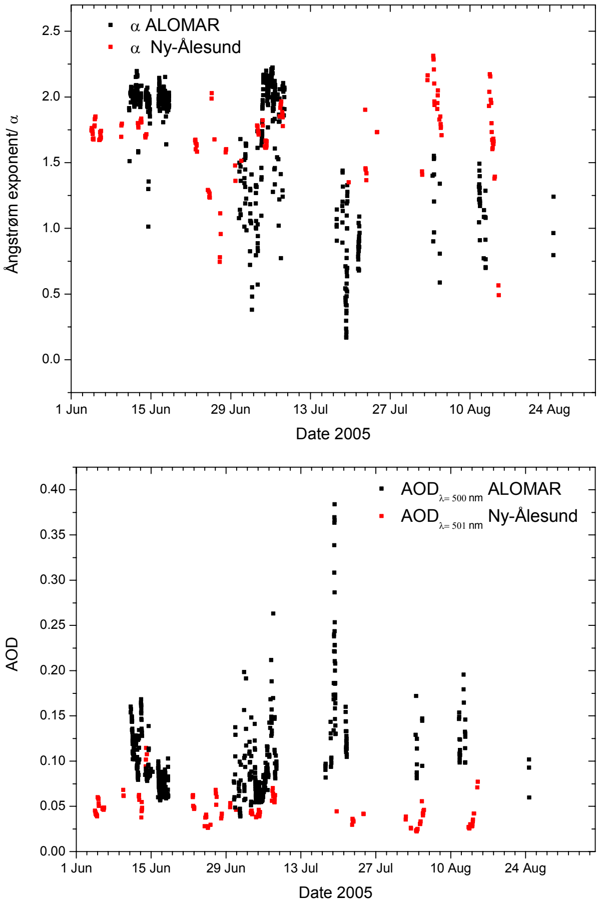

Figure 5.6: Lower panel: Hourly AOD values at ALOMAR and Ny-Ålesund for the period June – August 2005. Upper panel: Hourly Ångstrøm exponents, α, values at ALOMAR and Ny-Ålesund for the period June – August 2005

As expected, we observe much higher aerosol load at the Sub-Arctic site ALOMAR. Few periods with high AOD at ALOMAR agree with high AOD in Ny-Ålesund and we will continue the study with transport modelling to explore this further.

From the comparison of the Ångstrøm exponents, α, it seem that for both sites there is a predominance of large α indicating small aerosols. However, during peak episodes with elevated AOD values the α decreases indicating larger aerosols during the episodes.

The upper panel of Figure 5.7 show the Ångstrøm exponents for both sites and how it relates to the measured AOD values at 501 nm. The figure demonstrates that there is a clear tendency for both sites that low AOD values are connected with high Ångstrøm exponents and higher AOD values are connected with low Ångstrøm exponents. This suggests that episodes with high AOD values are connected with larger aerosols. The explanation to this needs further evaluation, but the lowest α values may be due to thin cirrus clouds, because of the difficulty of the automatic cloud-screening algorithm to detect them. The data will be further analysed and the time periods need to be expanded.

The hourly relative frequencies of different Ångstrøm exponents, α, during the summer 2005 are displayed in the lower panel of Figure 5.7. The results must be interpreted with caution, as they represent only a few days of measurements, particularly in Ny-Ålesund, and few overlapping periods. The α values in Ny-Ålesund are relatively widely distributed with signs of two peaks centred at α = 1.70 and α = 1.20. 30% of the α is in the range from 1.65 – 1.75, and as much as 25% in the range from 1.05 – 1.25 as well. 21 % of the Ångstrøm exponents are below 1.0 in Ny-Ålesund, i.e. the representative threshold for maritime aerosol type according to Smirnov et al. (2003). At ALOMAR the situation is different from that of Ny-Ålesund. As much as 51% of the α values are within the range from 1.95 – 2.05, and only 14 % is below 1.0. The low fraction of aerosols with α values below 1 are surprising as maritime aerosol characteristics were expected at ALOMAR (α typically below 1).

The high α values imply large loading of fine aerosols at both sites. The observed α values are not trivial to explain, and further studies and observations are necessary to confirm the origin of these fine particles. (Myhre et al., 2006). The differences between ALOMAR and Ny-Ålesund might be explained by the differences in the air masses arriving at each site, as well as the different importance of atmospheric processes for the lifetime of the aerosols (stratification, precipitation, etc.). However, there is reasonable agreement when the same air masses is present at both sites, which was the case during the haze event in April-May 2006, following from transport of smoke from fires in Russia and the Baltic states into the European Arctic.

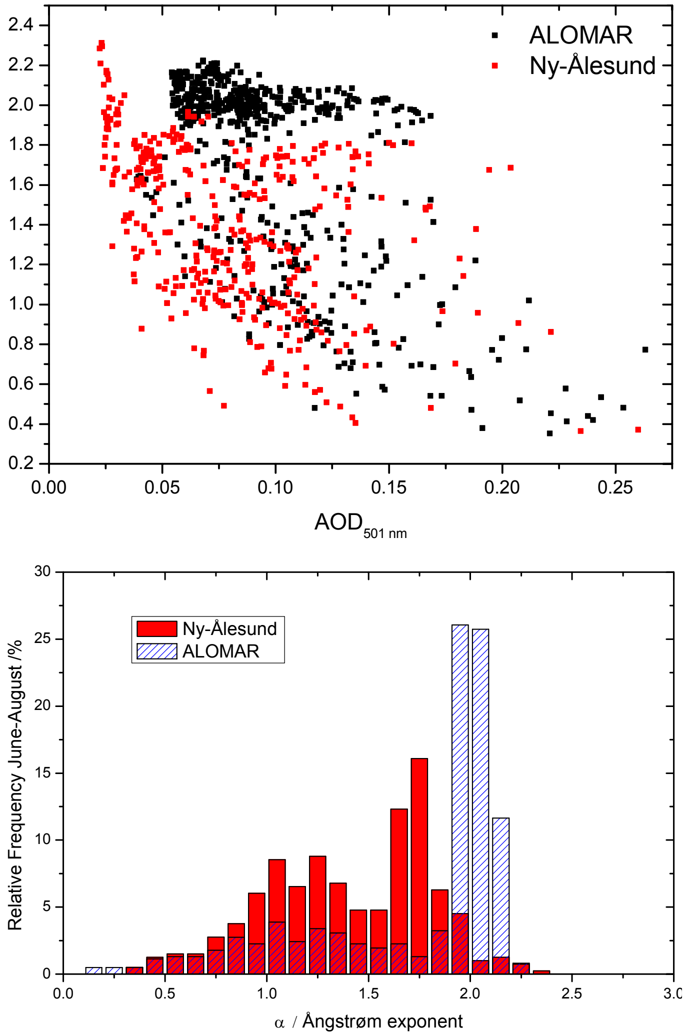

Figure 5.7: Lower panel: The relative frequency of hourly averaged Ångstrøm exponents, α, during summer 2005. Upper panel: A comparison of α based on hourly averaged data from ALOMAR and Ny-Ålesund during summer 2005.

6. European aerosol measurements from space

By Aasmund Fahre Vik, Ann Mari Fjæraa, Kerstin Stebel, Karl Espen Yttri,
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Air pollution by aerosols has become a global issue with the recognition of inter-continental transport as a significant source to the background levels of polluted air, especially in Europe and North America. While current regional monitoring networks are suitable for monitoring air pollution from regional sources, new measurement techniques and platforms are required to quantify the hemispheric transport and its contribution to regional background levels of aerosols. Satellite observations of the aerosol load is an interesting approach, as they offer data coverage over ocean and remote areas, and is able to provide “images” of the spatial air pollution distribution. In the context of the EMEP “Task Force on Hemispheric Transport of Air Pollution”, EMEP-CCC is currently seeking to incorporate space borne Earth Observation data in operational routines for assessment of air quality levels in Europe. An assessment of the accuracy and applicability of these data sources are therefore needed in order to make this possible.

Satellite measurements are by nature different from the traditional EMEP measurements, since aerosol properties are derived from observation of the atmospheric reflection from space, rather than by analyzing air samples collected at a station. A variety of instruments flying on different types of satellites are used to retrieve atmospheric aerosol properties and they generally provide information on the spatial distribution of particles. Most available data are from polar orbiting satellites that pass regularly over a certain location at a nearly fixed time. These instruments provide a snapshot of the aerosol distribution at the time of overpass (for European satellites measuring over Europe, this is commonly between 10 and 11 am local time) and measurement pixels are on the size of 1 km x 1 km or larger. In the presence of clouds it is normally not possible to retrieve aerosol properties for that pixel. The satellite therefore provides information on the spatial variation of aerosols over a larger area, but only for a certain time, while an EMEP monitoring station commonly provides daily averaged data on PM_{10} and $PM_{2.5}$, but for that site only.

Last years report on aerosol concentrations in Europe (EMEP/CCC, 2005) focused on the description of two new techniques for retrieving AOD (Aerosol Optical Depth) from the ATSR-2 instrument and AOD and aerosol type from the ATSR-2 and GOME instruments. The latter is done by a synergistic retrieval method, named SYNAER in brief. Both instruments were launched onboard the ESA (European Space Agency) spacecraft ESR-2 in 1995, which is still operational. ATSR-2 is designed for accurate measurements of sea surface temperature while GOME is designed mainly for stratospheric ozone measurements. The aerosol retrievals are additional bi-products. The methods for retrieving the data were described and examples of data were given for both cases. Both retrieval algorithms have undergone revisions since 2005 and much more data have become available, thus making it possible to assess the products both quantitatively and qualitatively. Both services are currently being continued and now use the next generation of European satellite instruments; the AATSR, which is an improved version of ATSR-2 and SCIAMACHY, which is a more advanced version of GOME.

Through the ESA-GSE project PROMOTE, a complete dataset of monthly means of AOD from AATSR over Europe in 2003 has been produced and made available through the project web pages (www.gse-promote.org). AOD is an important climate parameter as it describes the column integrated atmospheric optical extinction, i.e. the attenuation of the intensity of incoming solar radiation due to scattering and/or absorption of aerosols, and is important for assessment of radiative forcing. AOD varies with wavelength, aerosol composition, aerosol size distribution, height distribution and total concentration. If all except the latter is constant, the AOD values will be proportional to the aerosol concentration at ground levels. The satellite AOD data may in these cases provide an estimate of parameters such as PM_{10} and $PM_{2.5}$, which are important air quality. Such relations have been derived for certain cases and their generalisation for Europe is under investigation. Details of the AATSR dataset for 2003 are described in the next section.

6.1 Aerosol Optical Depth over Europe in 2003 as observed from AATSR

As described in EMEP/CCC (2005), methods to retrieve AOD from the Advanced Along Track Scanning Radiometer (AATSR) have been developed at TNO in The Netherlands. The AOD has been calculated for the year 2003. Below are maps of monthly mean AOD at 550 nm over land surfaces in Europe. Retrieval is done over cloud-free scenes for the 1 km x 1 km instrument resolution and binned in pixels of 10 km x 10 km by means of an automated post-processing step. The data over oceans are produced with a different algorithm and are not shown here. For September 2003, no data are available. The AOD scale in these figures runs from 0.0 to 1.0. AOD values of 0.7 are observed over highly polluted areas (Robles-Gonzalez et al., 2000).

A few artefacts (values >0.7) are present in the data in Figure 6.1–6.11. The AOD values above the mountain areas in Norway, the Alps and some other high-altitude areas, are unnaturally high in the spring months March, April and May (and June for Norway). This could be due to high albedo (high reflectivity) of snow-covered areas and the data values are not correct for these special cases. The validity and subsequent removal of such pixels is further investigated.

The general trend of the dataset is that AOD values are high in the summer months (e.g. August) and low in the winter (e.g. December). There is no clear distinction between Western and Eastern Europe, although in Eastern Europe the values are sometimes significantly higher. These high values are most probably not artefacts. Elevated AOD values, and thereby also pollution levels, are observed to be significant above Western and Eastern Europe, although often not in the same months. The dataset is typically able to capture large-scale spatial and temporal variations in aerosol load, but cities and industrial areas are only occasionally seen in the maps.

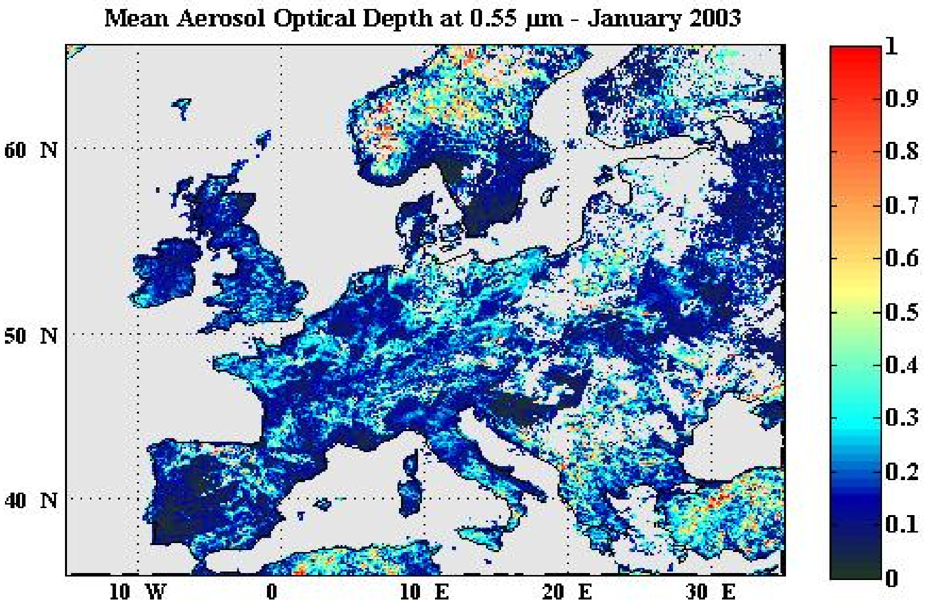


Figure 6.1: Monthly mean AOD at 550 nm over Europe in January 2003 from AATSR onboard ENVISAT.

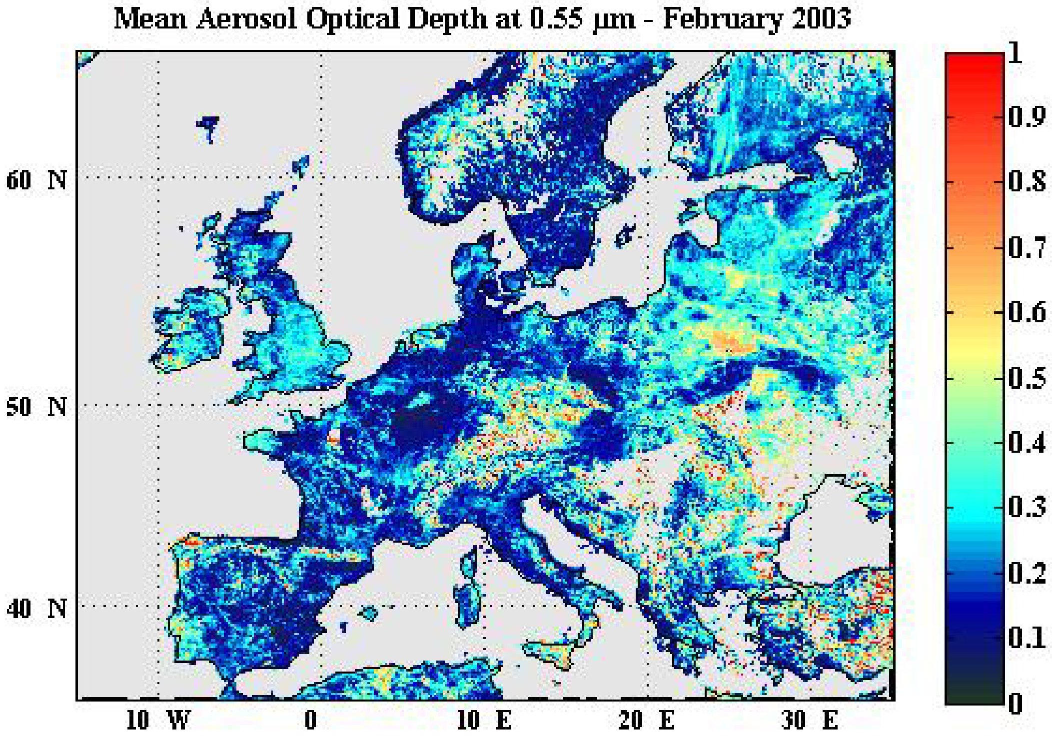


Figure 6.2: Monthly mean AOD at 550 nm over Europe in February 2003 from AATSR onboard ENVISAT.

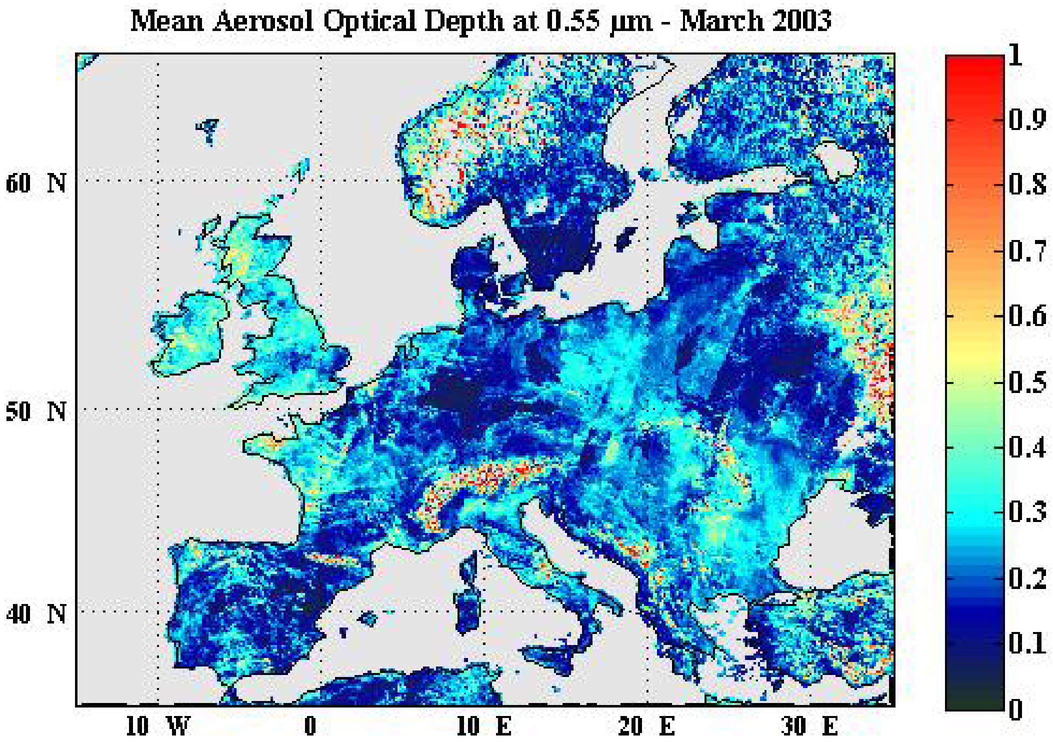


Figure 6.3: Monthly mean AOD at 550 nm over Europe in March 2003 from AATSR onboard ENVISAT.

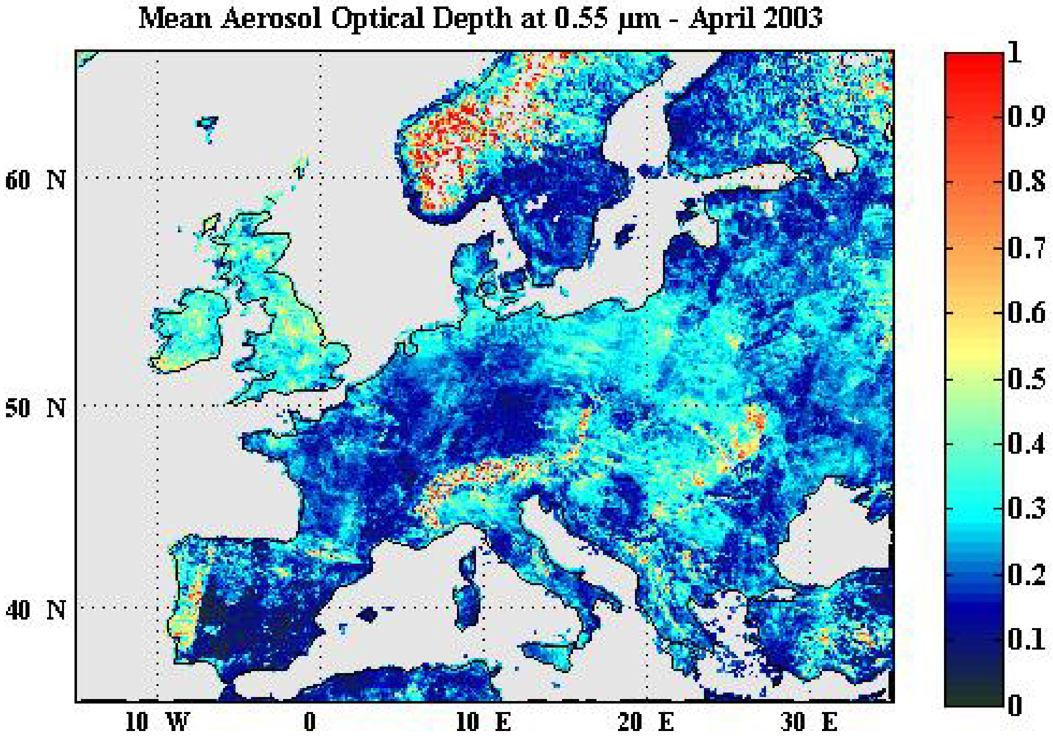


Figure 6.4: Monthly mean AOD at 550 nm over Europe in April 2003 from AATSR onboard ENVISAT.

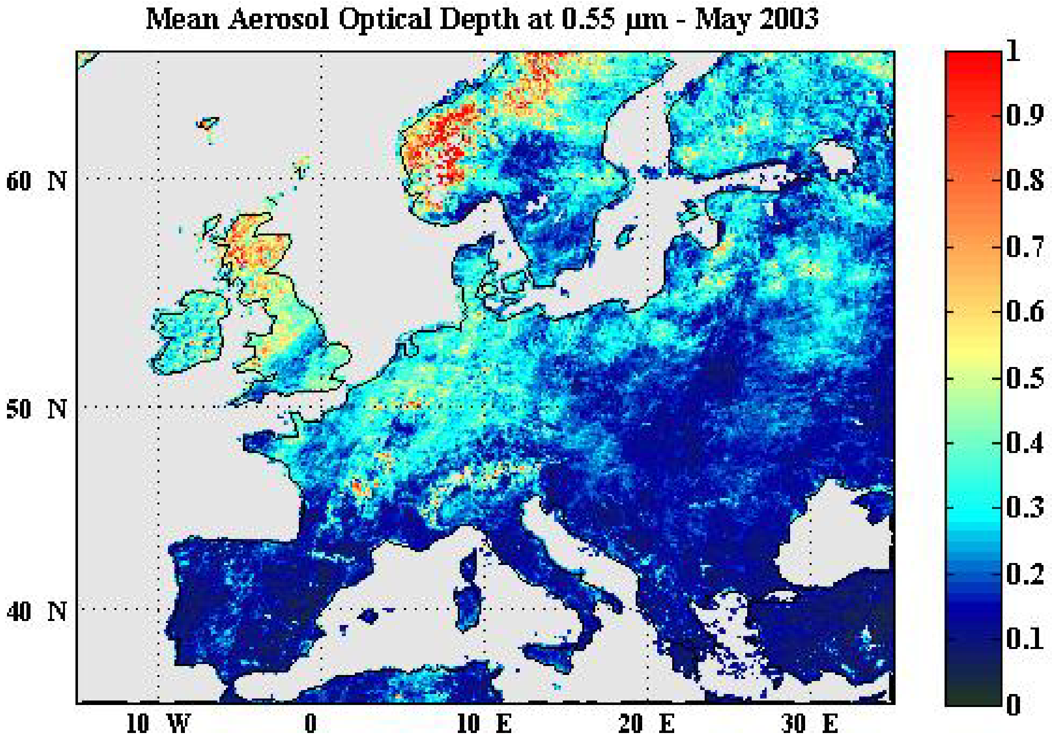


Figure 6.5: Monthly mean AOD at 550 nm over Europe in May 2003 from AATSR onboard ENVISAT.

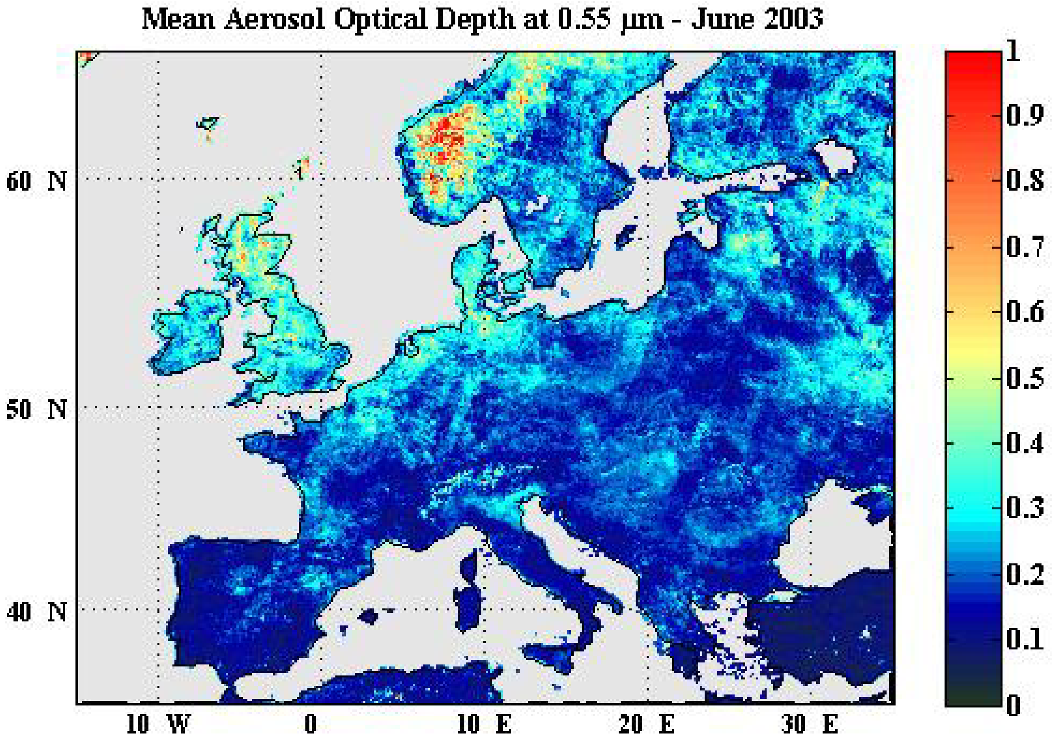


Figure 6.6: Monthly mean AOD at 550 nm over Europe in June 2003 from AATSR onboard ENVISAT.

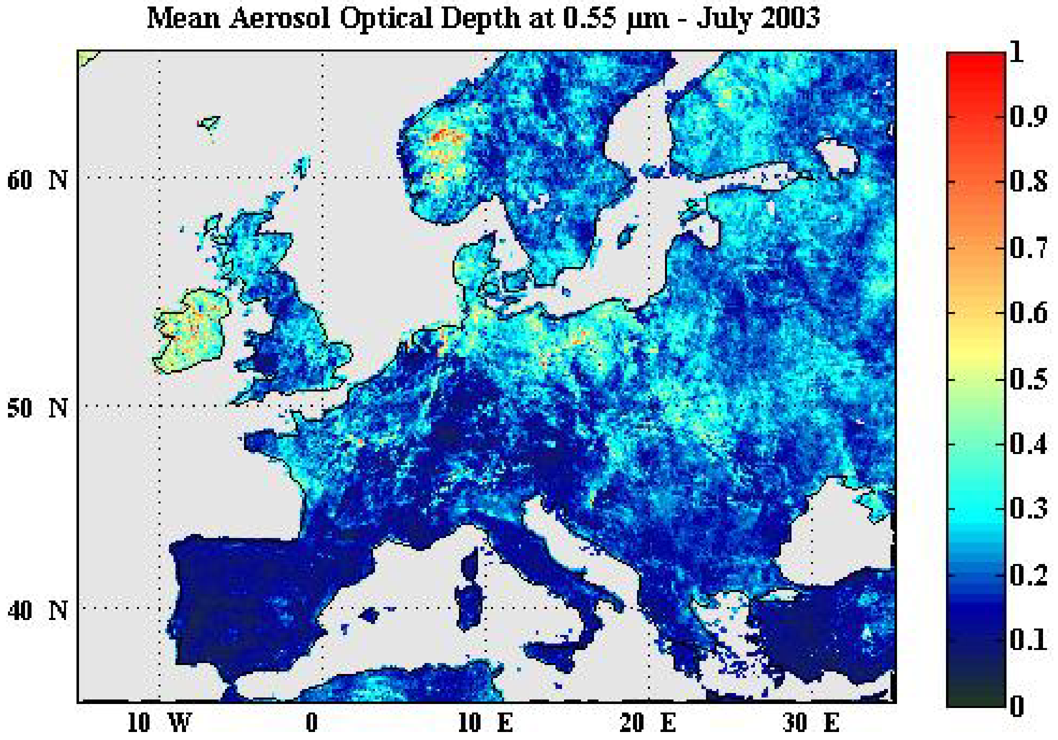


Figure 6.7: Monthly mean AOD at 550 nm over Europe in July 2003 from AATSR onboard ENVISAT.

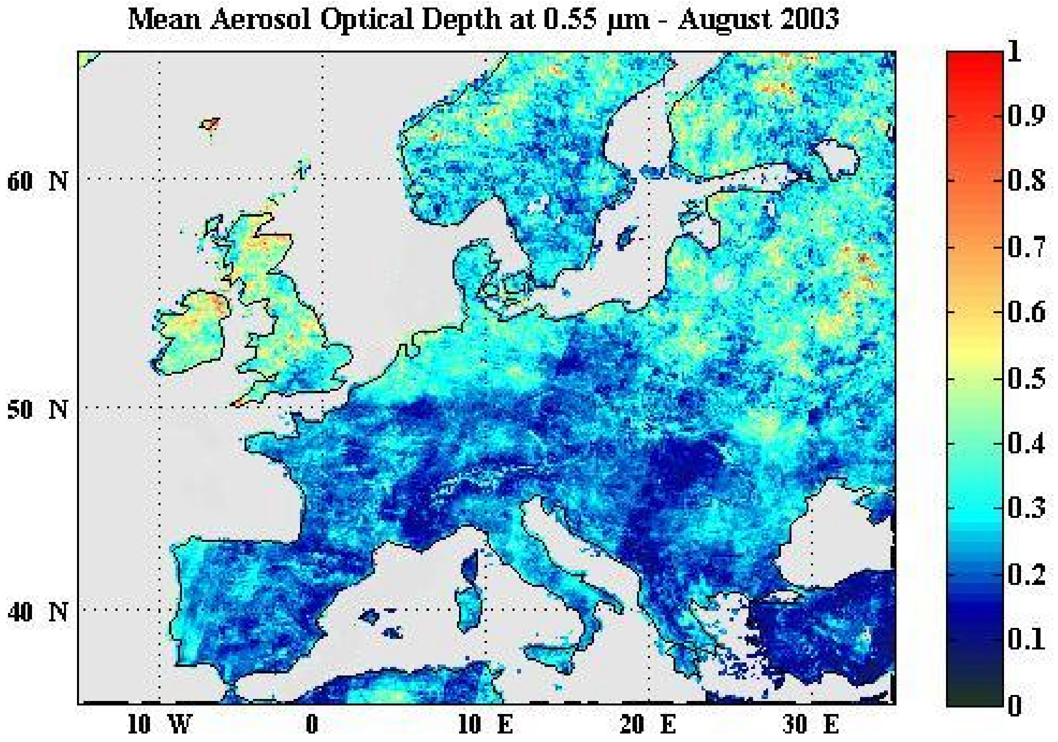


Figure 6.8: Monthly mean AOD at 550 nm over Europe in August 2003 from AATSR onboard ENVISAT.

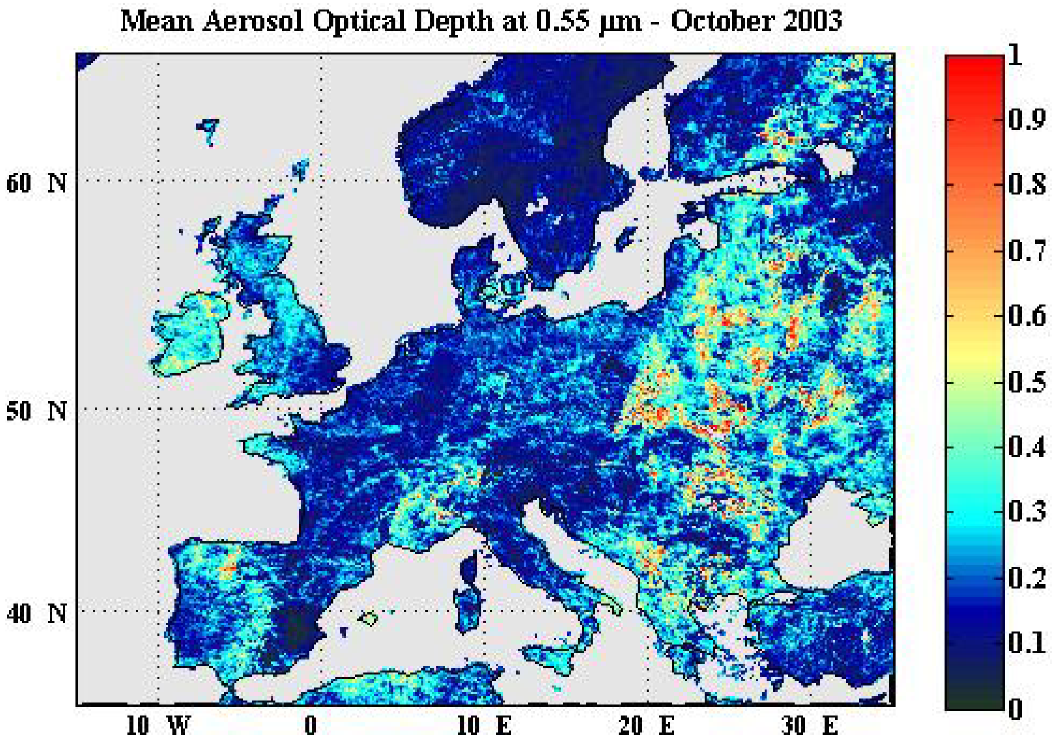


Figure 6.9: Monthly mean AOD at 550 nm over Europe in October 2003 from AATSR onboard ENVISAT.

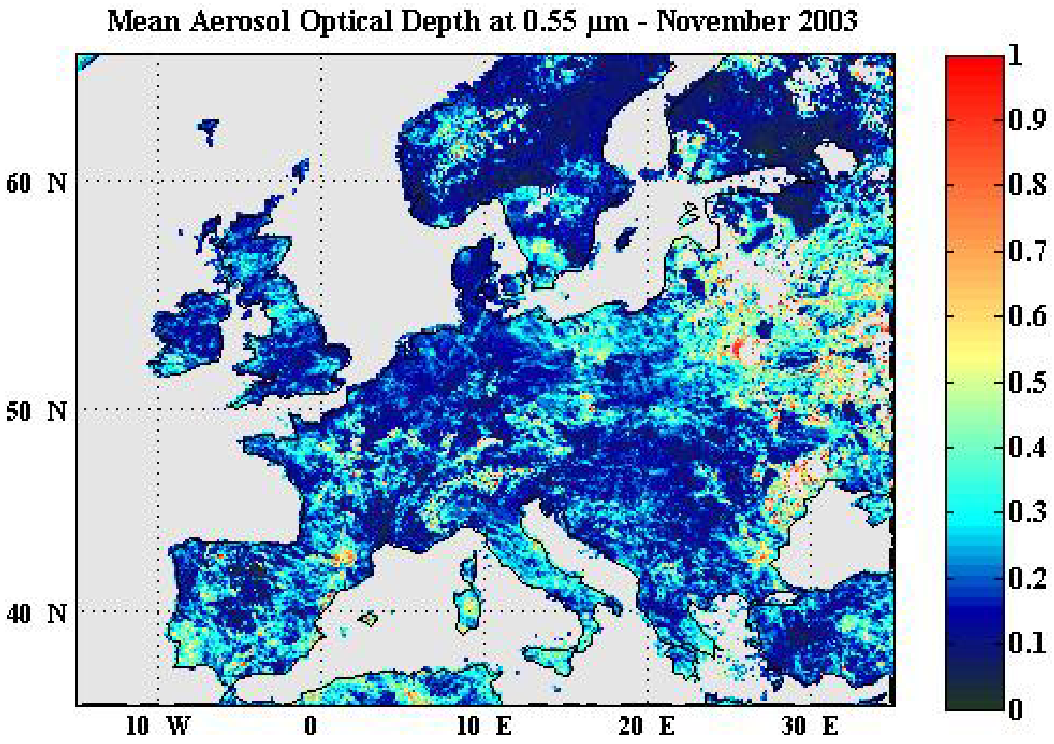


Figure 6.10: Monthly mean AOD at 550 nm over Europe in November 2003 from AATSR onboard ENVISAT.

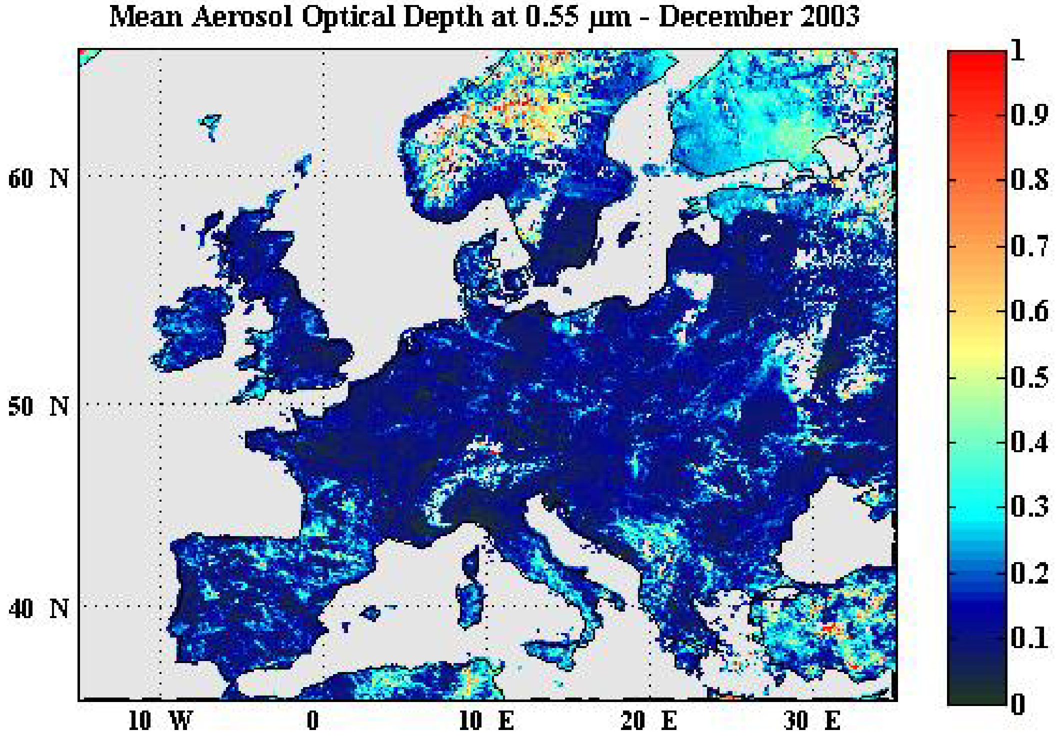


Figure 6.11: Monthly mean AOD at 550 nm over Europe in December 2003 from AATSR onboard ENVISAT.

Validation of the data done through the PROMOTE project shows a slight positive bias when the product is compared with Groundbased AOD measurements from AERONET. AOD measurements are known to have a large variability during the day and from one day to the next. The satellite product is a compilation of a series of snapshots taken every third day during a month while the groundbased measurements may give results every clear day. Even when taken at the same hour of the day as the satellite product, the ground-based measurements averaged over a month will therefore most likely show different results. Taking this into account, the satellite AOD product compare satisfactorily with the groundbased measurements of AOD.

AOD measurements are also performed by several instruments like the two American MODIS sensors onboard the Terra and Aqua satellites and the European MERIS onboard Envisat. Maps of monthly means are available from these sensors and show similar large-scale trends as those seen from AATSR. An example from MODIS (Terra) is shown for August 2003 in Figure 6.12. Some variations are observed between the AATSR and MODIS data, most likely due to differences in measurement principle and resolution of the two instruments. Extensive comparison between MODIS and AATSR for 2003 over Europe is underway as part of a Dutch national project.

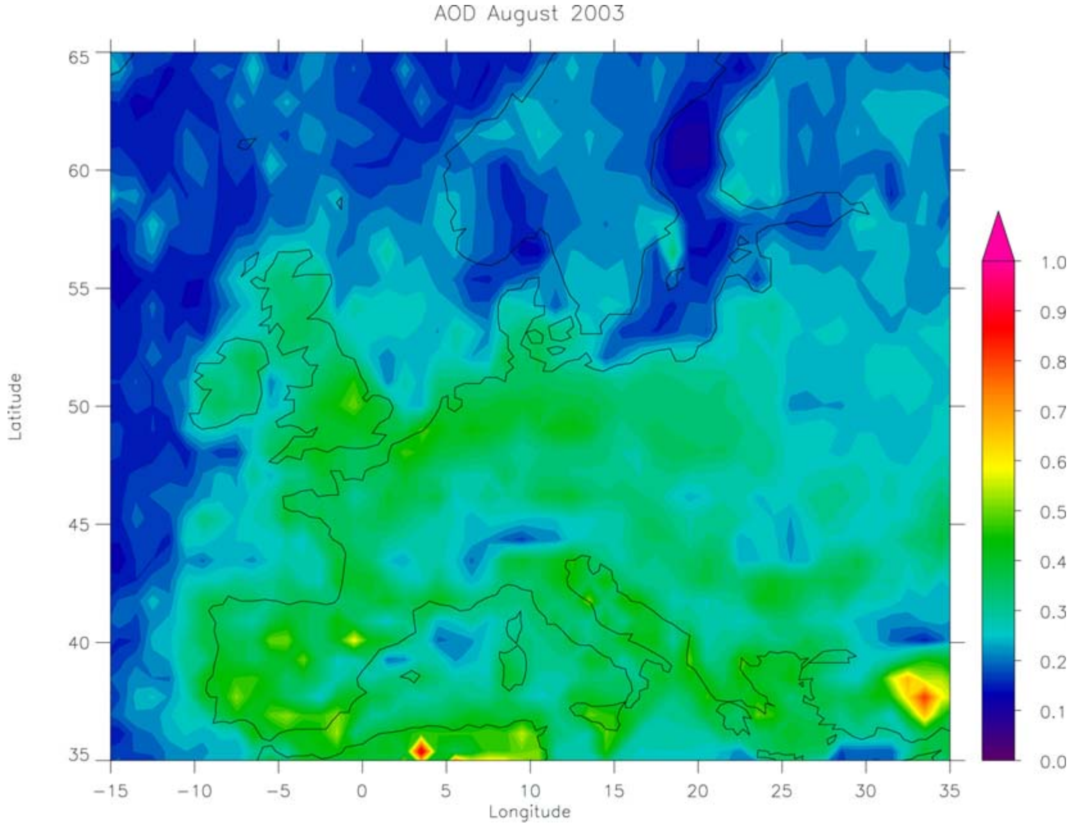


Figure 6.12: Monthly mean AOD at 550 nm over Europe in August 2003 from MODIS onboard Terra. Data are courtesy of Langley Research Centre, NASA.

As an additional possibility to monitor European air quality with respect to aerosols, the main advantage of the AATSR product is its large geographical coverage and ability to observe areas with little or no data coverage by the EMEP network. The high spatial resolution is also beneficial for some applications, but the practical use is limited due to low temporal coverage. Because of this, and the fact that only cloud-free scenes are observed, one may not interpret every detail in a monthly mean map as a pattern in regional scale air quality. The relation to PM is furthermore unsure since aerosol composition is unknown and the product does not differentiate between boundary level and free troposphere air pollution. However, the products may be a very useful supplement to existing groundbased networks, especially in remote areas. The possibility of creating an inter-comparable time-series of data from the ATSR-1, ATSR-2, AATSR and in the future AVHRR on MetOp, may provide an opportunity to establish long-term trends in air-pollution by aerosols.

6.2 Particulate matter in Europe as observed with SCIAMACHY and AATSR in the SYNAER retrievals

The SYNAER algorithm, which was described in detail in EMEP/CCC (2005), derives aerosol properties by exploiting complementary information from the radiometer AATSR and the spectrometer SCIAMACHY, both onboard ENVISAT. The combination of these two instruments allows retrieving aerosol optical depth at 550 nm and aerosol speciation from a choice of pre-defined aerosol types. For air quality purposes, the most important products are the PM$_{10}$, PM$_{2.5}$ and PM$_1$ parameters that, in addition to existing monitoring networks, may provide useful spatial information on the PM pollution levels. These products are therefore tested and validated against EMEP monitoring stations in the following. It should be noted that the SYNAER PM data used in this study were derived from the SYNAER AOD and speciation information, but assuming a fixed homogeneous boundary layer of 2 km height.

The SYNAER data are provided through the ESA-GSE project PROMOTE and are available as daily maps (images) of Global aerosol load and composition on www.gse-promote.org. Maps that cover only Europe are also available. In addition, the data may be downloaded as HDF files (Hierarchical Data Format, binary files) where one file contains all data from one complete satellite orbit. An example of a daily map of AOD over Europe is shown in Figure 6.13.

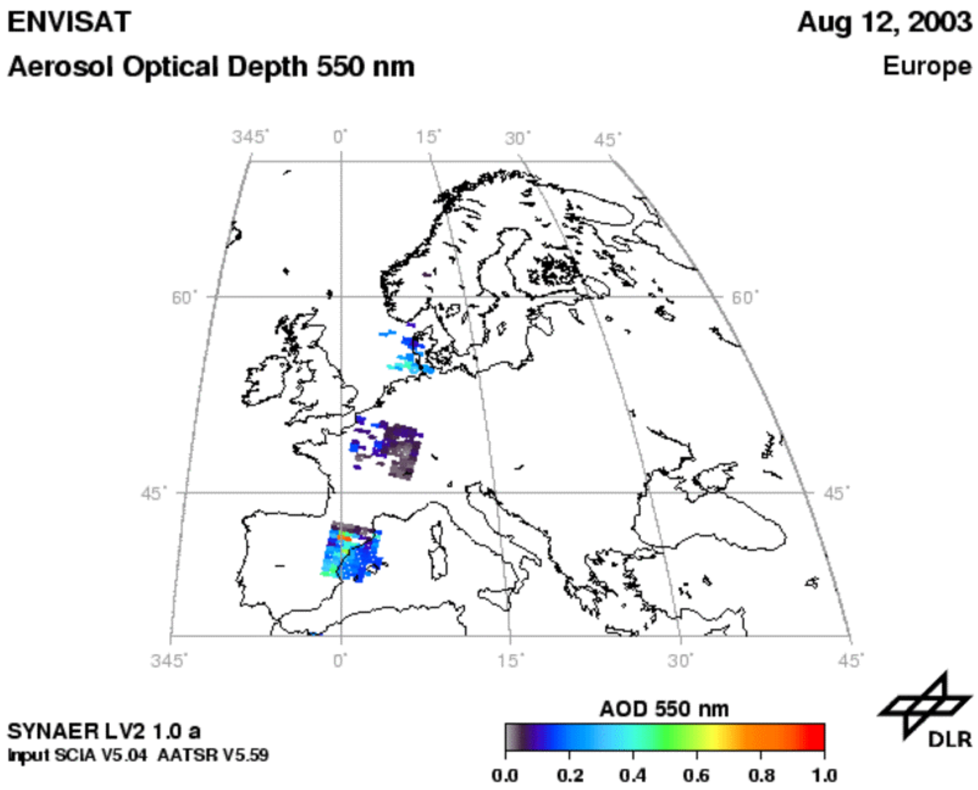
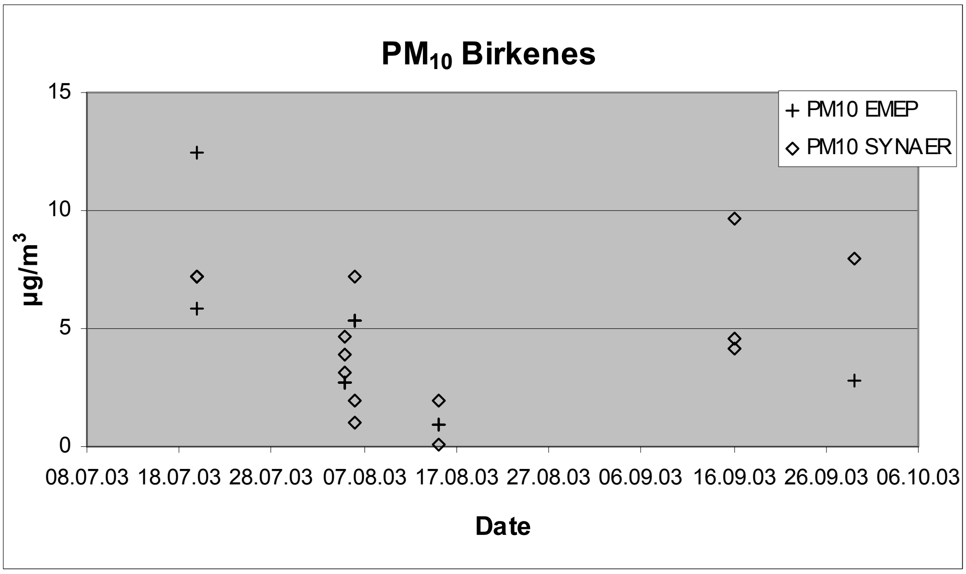


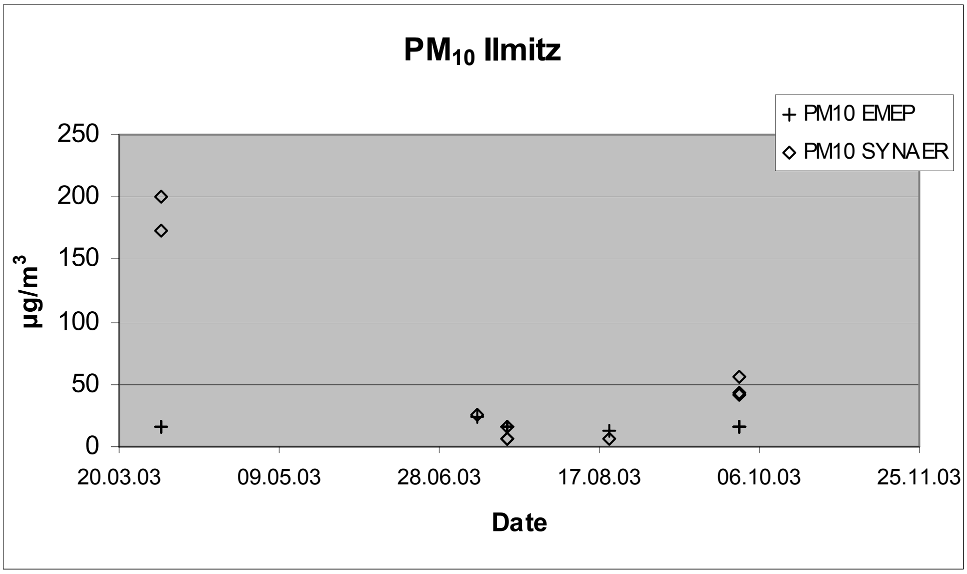
Figure 6.13: Daily Aerosol Optical Depth at 550nm over Europe from the SYNAER product.

Five EMEP stations were chosen for comparison against the satellite product: Birkenes (NO0001R), Illmitz (AT0002R), Ispra (IT0004R), Payerne (CH0002R) and Zarra (ES0012R). The selected stations needed to have daily measurements of at least PM_{10} and $PM_{2.5}$ and it was essential that they were not above the boundary layer. All satellite data for April, July, August and September (May and June were not available) were downloaded and pixels that were within ± 0.5° longitudes and latitudes were used for the comparison (the centre of the satellite pixel and the coordinates of the station were used as reference). This limited the number of possible matches significantly, but the number of measurements was still sufficient. Values of PM_{10}, $PM_{2.5}$ and for Illmitz also PM_1 are shown in Figure 6.14–6.16. Illmitz was the only station measuring PM_1 in 2003 for the selected stations.

The overall impression of the EMEP vs. SYNAER comparison is that the results compare rather well. Taking into account that a 24-hour average is compared with a snapshot, the two methods compare favourably. SYNAER seems to overestimate PM_{10} and $PM_{2.5}$ on certain occasions, especially at Zarra. This may be due to Saharan dust events that occur frequently over Spain, which may increase the tropospheric aerosol load without increasing ground level PM. Similar overestimation of PM_{10} at Illmitz in April and Ispra in August, may probably not be explained by this. At Payerne, the SYNAER product generally provides lower levels than the EMEP data. This may be due to high mountains with clean air surrounding the station, which are undoubtedly also sampled by the satellite. A spectrometer with higher spatial resolution than SCHIAMACHY is probably needed to solve these cases.

As for the AOD product from AATSR, the SYNAER retrievals may be a very valuable source of data in addition to the regular EMEP monitoring network. Compared to the AATSR product, its main advantage (for air quality purposes) is the ability to estimate aerosol composition and PM concentrations. The quality of the PM products looks very promising, but a larger validation exercise is probably needed to assess the product properly. The main disadvantage is the low spatial resolution and relatively low temporal data coverage of SYNAER/ENVISAT. The data presented in Figure 6.14–6.16 show all available measurements for four months above the EMEP monitoring sites, and emphasizes that the measurement frequency is too low for monitoring purposes. A small amount of data is, however, better than no data at all, and the product is valuable in remote areas and over oceans. The possibility of creating a long-term trend from ATSR-2/GOME on ERS-2, AATSR/SCIAMACHY on Envisat and in the future AVHRR/GOME-2 on MetOp, is also very interesting.





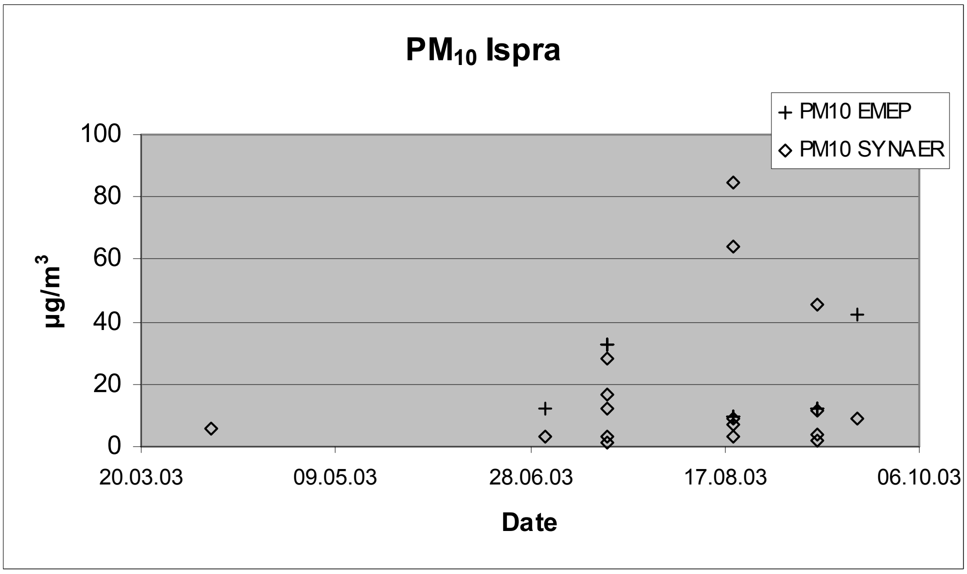
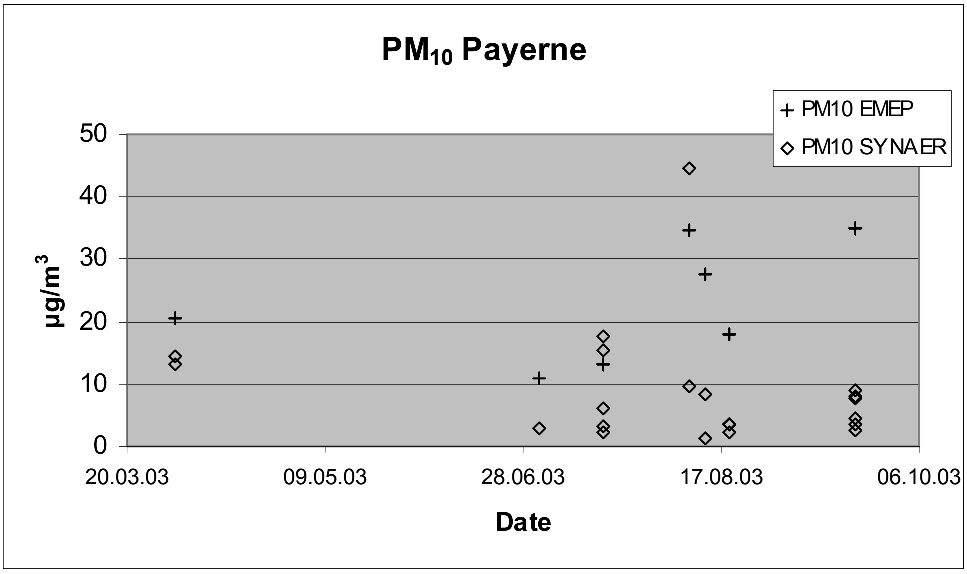


Figure 6.14: Daily averages of PM$_{10}$ at five EMEP stations compared with values from the SYNAER product.



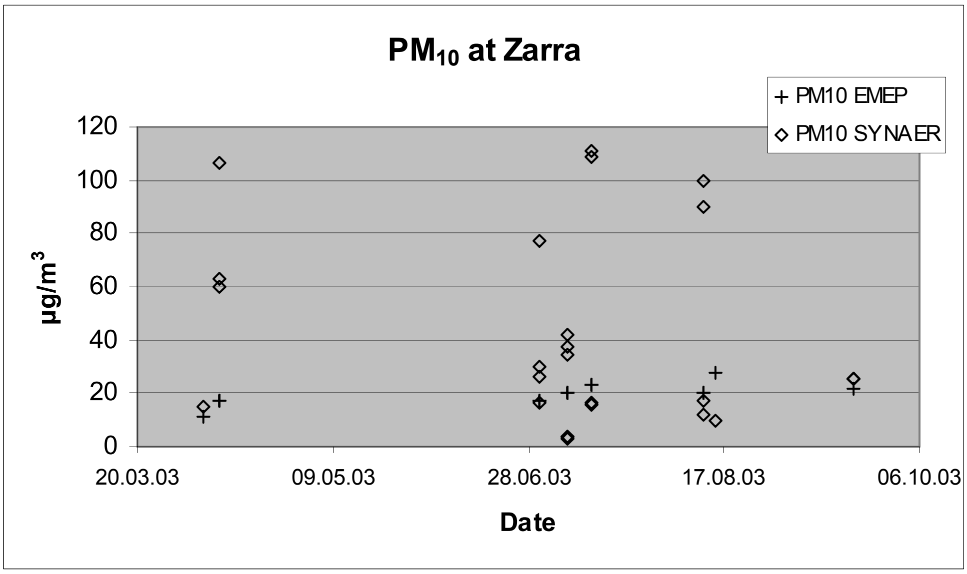


Figure 6.14, cont.

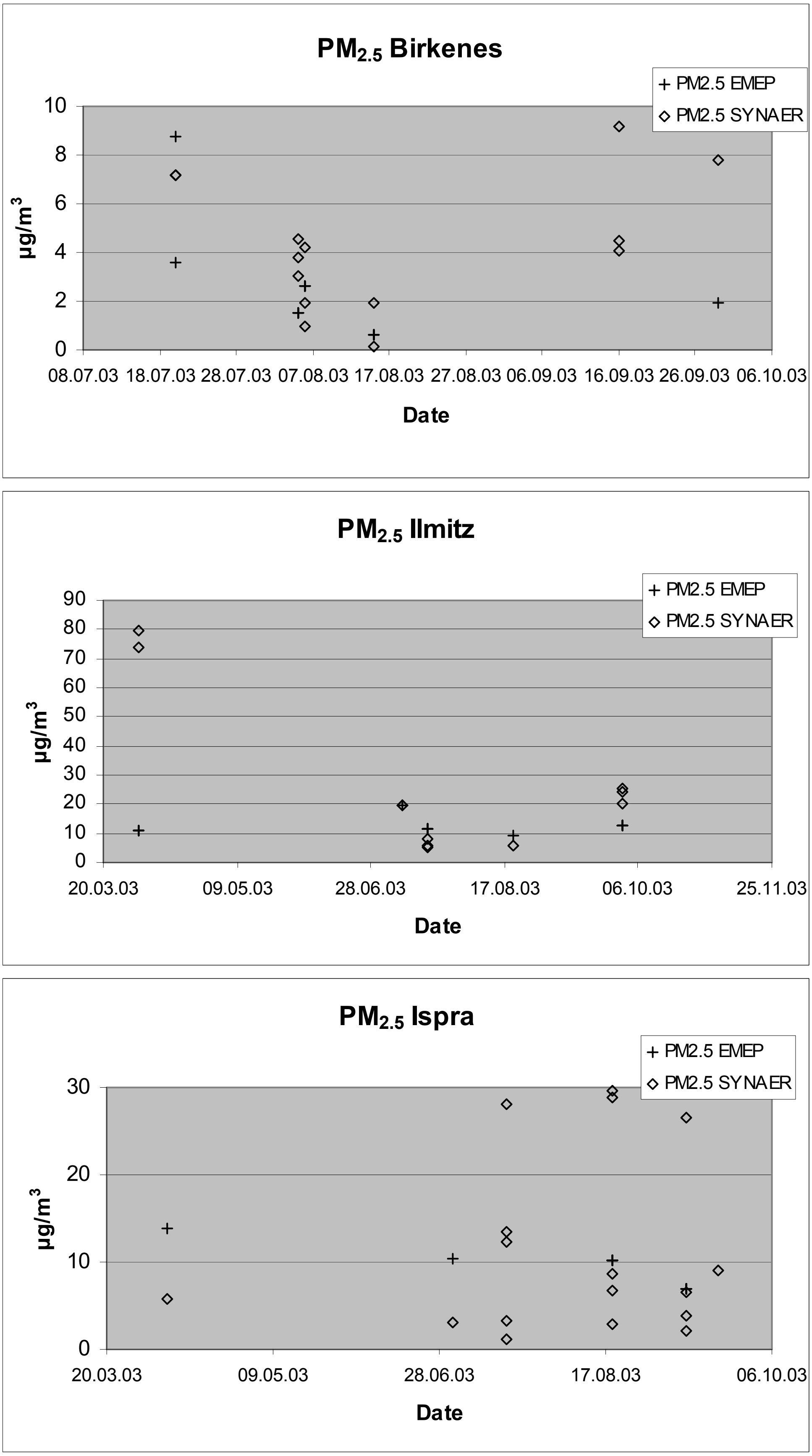
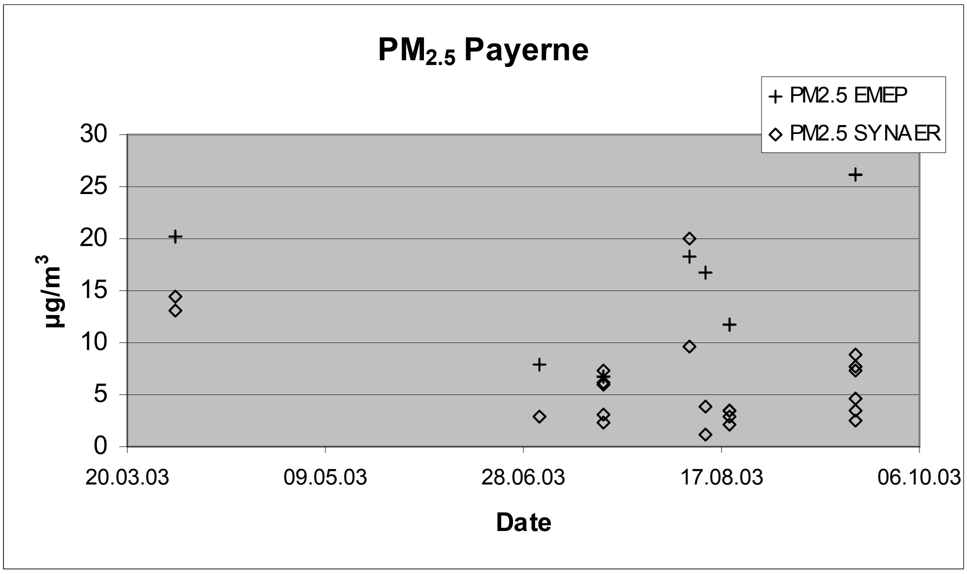


Figure 6.15: Daily averages of $PM_{2.5}$ at five EMEP stations compared with values from the SYNAER product.



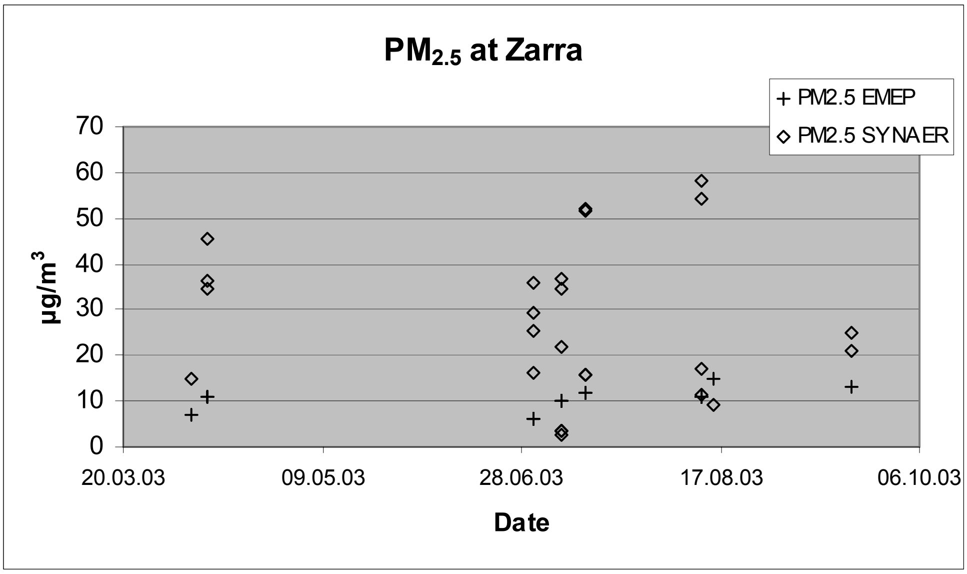


Figure 6.15, cont.

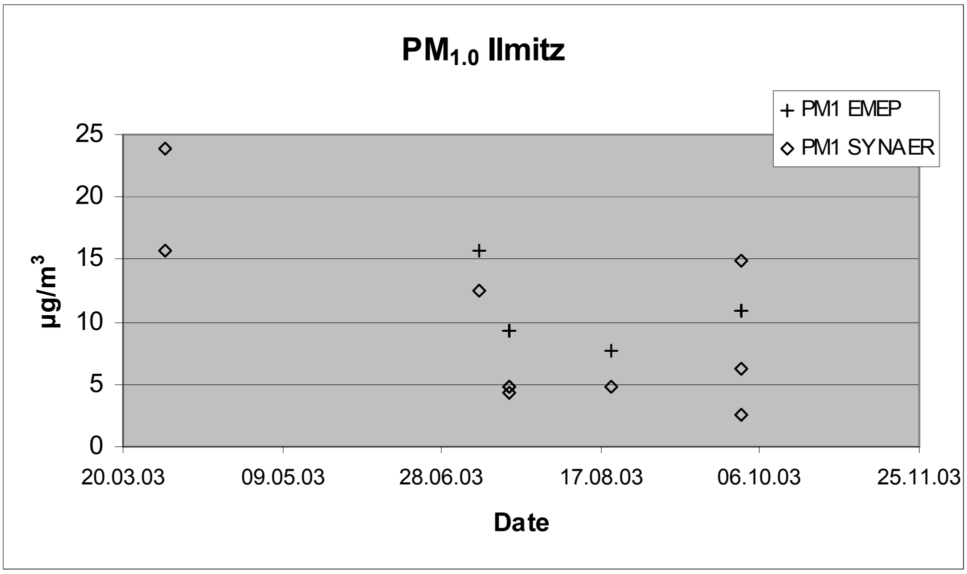


Figure 6.16: Daily averages of PM$_1$ at one EMEP station compared with values from the SYNAER product.

6.3 Vertical profiling of aerosols

The usefulness of active optical remote sensing for aerosol measurements has already been pointed out in EMEP/CCC (2003). Vertical profiling of aerosols is included in EMEP's monitoring strategy as a level 3 activity, contributing to the understanding of transport of air pollutions on the European and intercontinental level.

Already more than one decade ago, in 1994, the Lidar In-space Technology Experiment (LITE) onboard the Space Shuttle has demonstrated that lidar can bring valuable information on the vertical structure on aerosols and cloud layers on a regional and global scale. On April 28, 2006 NASA has launched CALIPSO into a sun-synchronous polar orbit. The first data from the nadir-viewing two-wavelength Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) was obtained on June 7 (see Figure 6.17).

Satellite systems require extensive validation by ground-based systems and networks.

The European Aerosol Research Lidar Network (EARLINET) contributes to the CALIPSO validation program. EARLINET has been established in February 2000. Nowadays 23 sites, well distributed over Europe, are contributing to the network. Vertical profiles of aerosol backscatter and extinction can be derived independently from more advanced (compared to CALIOP) Raman systems.

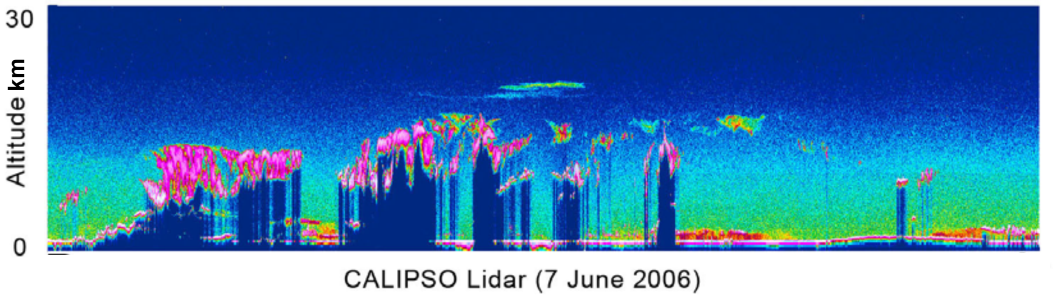


Figure 6.17: Total attenuated backscatter profile (532 nm) from the CALIPSO spacecraft (from NASA Langley Research Center).

6.4 Discussion and conclusions

Data from the current space-borne satellites are not alone able to monitor levels of air pollution by aerosols over Europe. The accuracy of the data are probably sufficent or are within reach to become so by ongoing algorithm developments, but temporal data coverage is not sufficient to properly monitor the regions of interest for EMEP. The AOD data from AATSR and the PM-products from the SYNAER process are probably representative for the time of satellite overpass for cloudless conditions, but it is doubtful if the data alone can describe the regional ambient aerosol concentrations for 2003, as seen from Figure 6.14–6.16. The satellite retrievals may, however, provide valuable information, as an additional data source to groundbased measurements. The SYNAER product and especially the AATSR retrieval may provide a spatial representation of air pollution, which is very useful for tracking special events. Of particular importance is the satellite’s ability to measure over oceans and remote areas. This monitoring is essential for the Task Force on Hemispheric Transport of Air Pollutants and satellite data like AOD from AATSR and PM data from SYNAER are needed to realise the goals of this activity. This includes access to observations to capture and monitor transport of polluted air masses, but also access to satellite data for model validation or assimilation. The convention for Long Range Transport of Air Pollutants (CLRTAP) under the United Nation Economic Commission for Europe (UN-ECE), currently has a strong interest in this task force and EMEP (which is under CLRTAP) is therefore more and more looking towards satellite retrievals. EMEP-CCC is, as a result of this, currently taking a more active part in the validation and use of future satellite products through its involvement in the ESA project PROMOTE Stage II.

The products described in this chapter are being continued and further developed through both national and international projects. TNO will be undertaking an effort to study synergy between AATSR and MSG/SEVIRI (geo-stationary weather-satellite) to couple AATSR accuracy to the MSG high temporal resolution (15 minutes repeat interval). The SYNAER product will be further improved in PROMOTE Stage II. This includes adaptation of the method to use GOME-2/AVHRR data to significantly improve the spatial coverage and enable daily observations of Europe (for cloud-free pixels) instead of 1 observation every 12 days (with ENVISAT AATSR/SCIAMACHY). The SYNAER AOD and PM products will become available on 10 km x 10 km grid to exploit the 1 km resolution of the radiometer, and the PM conversion will be improved by using daily EURAD model vertical profiles instead of the homogeneous fixed boundary layer.

In addition to currently flying satellite sensors, there are a number of instruments that in the future will provide aerosol records with even higher spatial and temporal resolution. The CALIPSO satellite will provide important aerosol profiling in the years to come, and experience from CALIPSO/EARLINET will be valuable for the next European EarthCARE mission. This is due for launch into a sun-synchronous orbit in 2012, carrying the European Atmospheric Lidar (ATLID) onboard. ATLID will be a nadir-looking multi-FOV single wavelength lidar with a high-spectral-resolution (HSR) receiver and polarization capability. The system is planned to detect radiatively significant clouds / aerosols (optical depth of 0.05), with vertical resolution of 100 (Mie-channel) and 300 m (Rayleigh-channel), and 10 km horizontal integration time. Similar to Raman lidar, aerosol backscatter and extinction coefficients can be derived independently. A similar active remote sensing instrument, the atmospheric Laser Doppler instrument ALADIN on ESA's atmospheric dynamic mission ADM-Aeolus will be launched already in 2008.

In addition to improving current data products and launching new satellite instruments, there is a clear need to further integrate the different data sources to better monitor e.g. levels of European ambient PM air pollution, and trends therein. This includes synergistic use of groundbased, airborne and satellite aerosol measurements, most likely through assimilation in aerosol models. Such activities are currently ongoing and further work on assimilation of aerosol data into air quality models will be done (e.g. in PROMOTE Stage 2 to provide historic records and daily 3-day forecasts based on in situ and satellite measurements). Other international strategies and programmes are currently being established to better integrate data sources, and IGACO, GMES and GEOSS will likely be important for the developments of future aerosol measurement capabilities. The EU-funded GEOMON project, a European atmospheric composition component to GEOSS, is starting up next year and will play an important role in this aspect.

7. References

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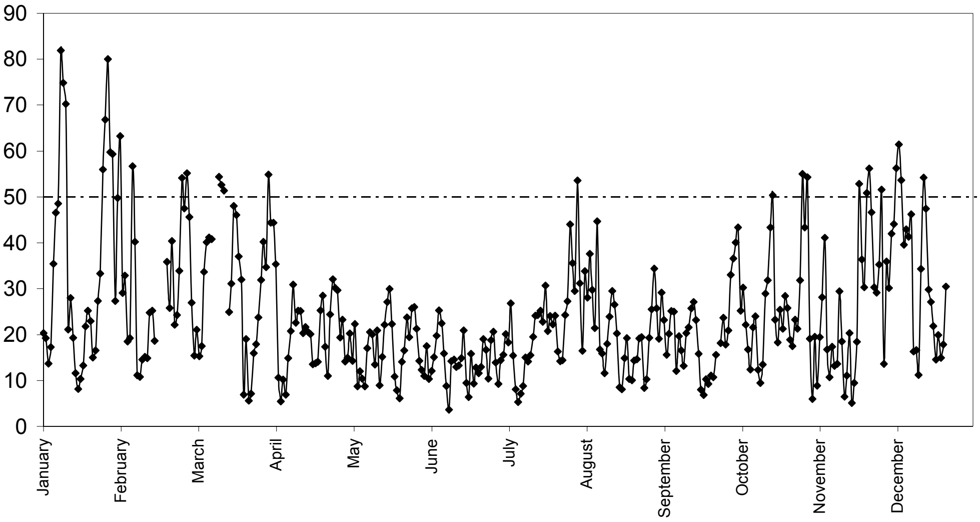
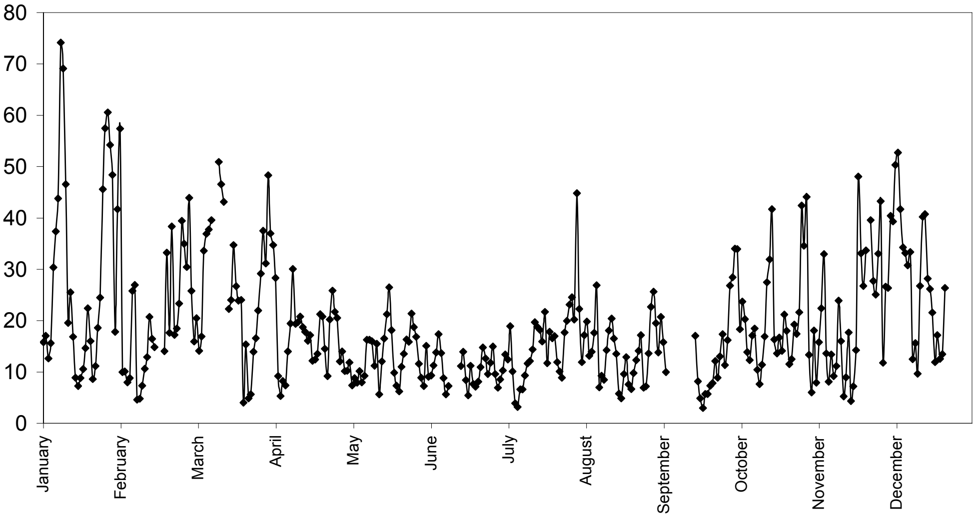
Annex 1

Time series of particulate matter mass concentrations at EMEP station in 2004

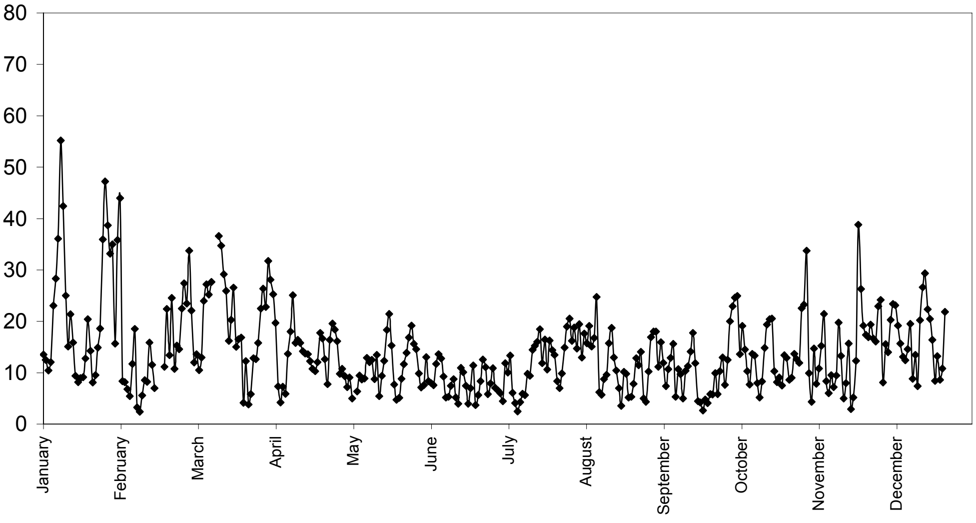
AT0002R 2004 PM_{10} (µg/m^3)



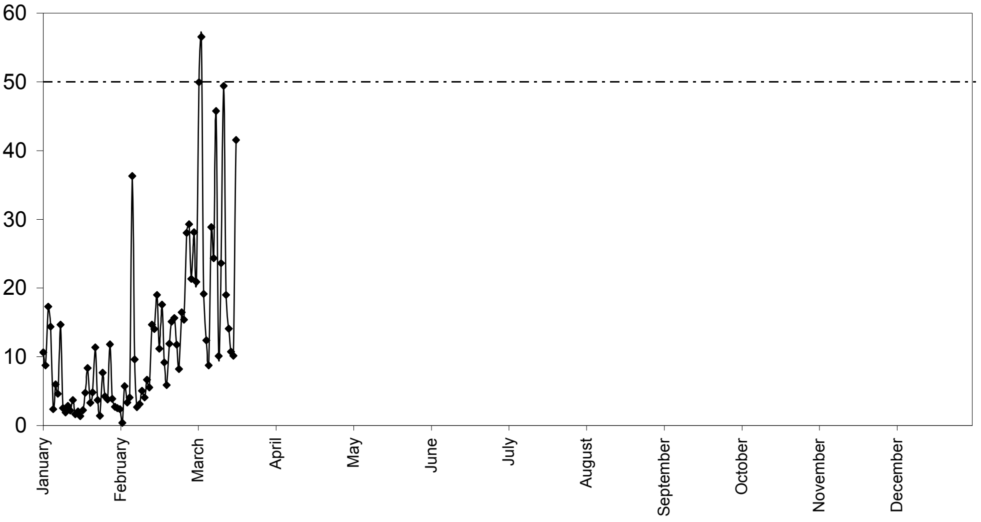
AT0002R 2004 $PM_{2.5}$ (µg/m^3)



AT0002R 2004 PM_1 (µg/m^3)



AT0004R 2004 PM$_{10}$ (µg/m^3)



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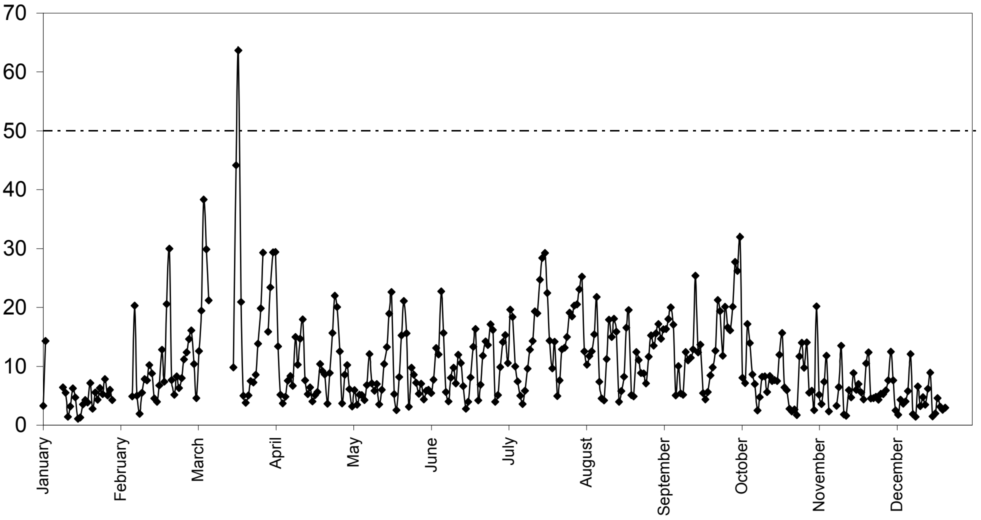
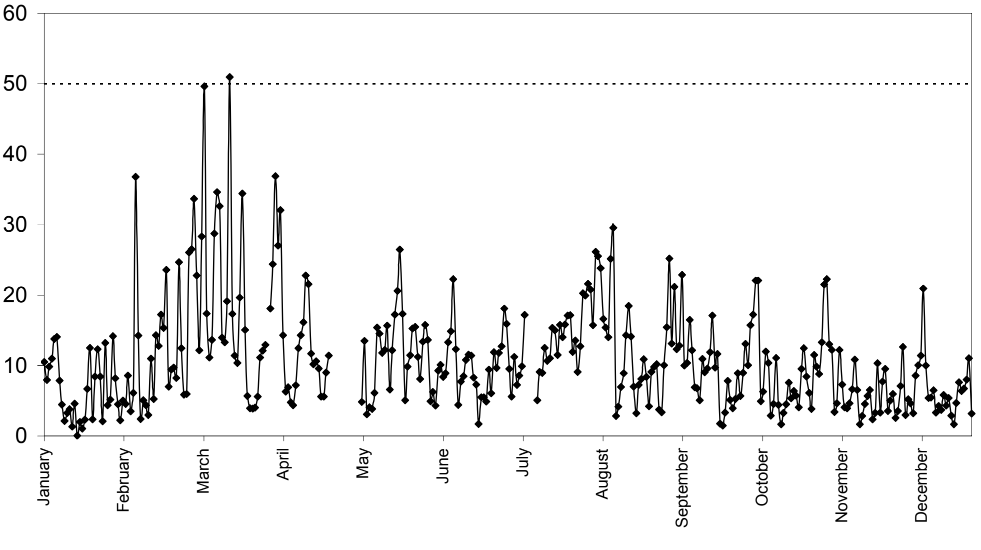


AT0048R 2004 PM$_{10}$ (µg/m^3)



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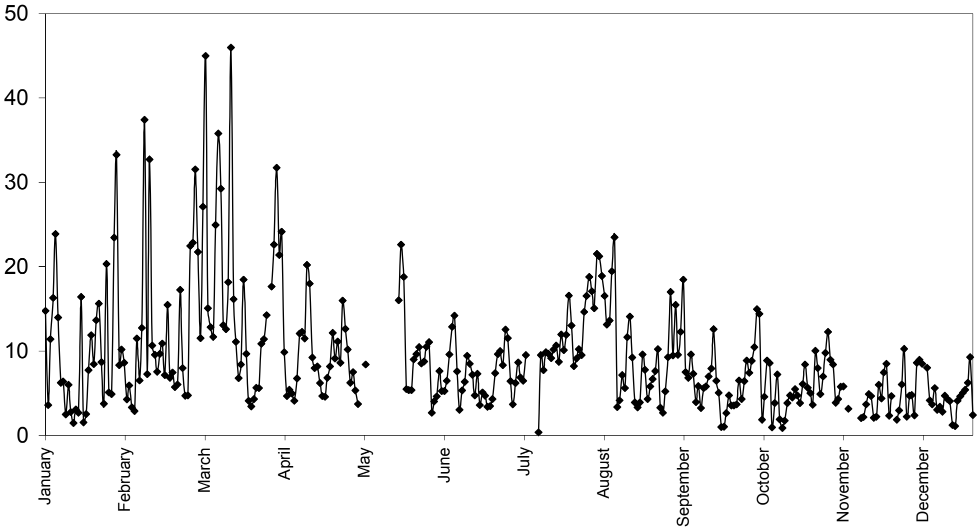
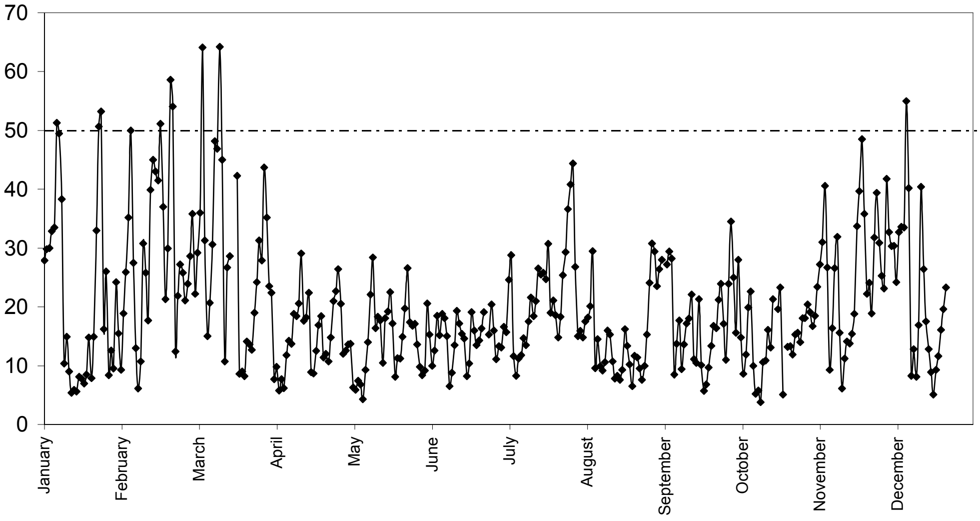
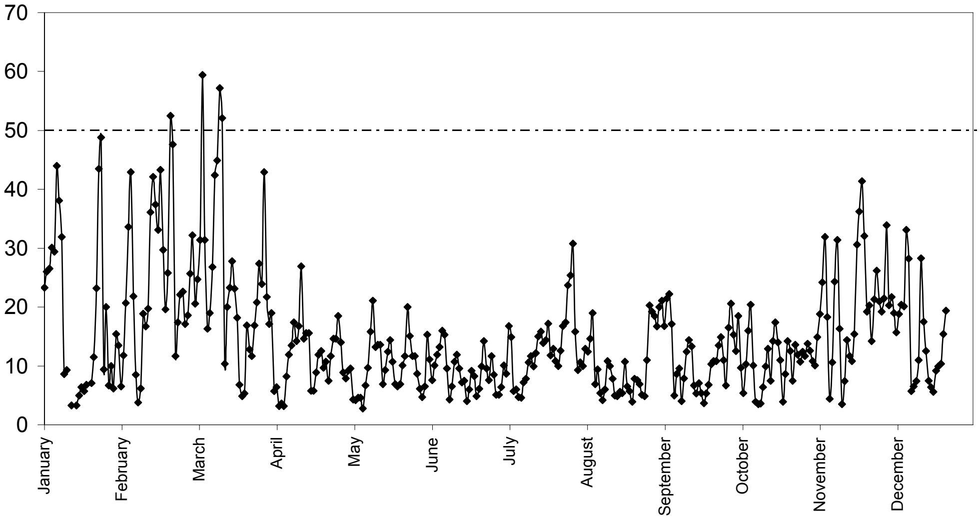


CH0002R 2004 PM$_{10}$ (µg/m^3)



CH0002R 2004 PM$_{2.5}$ (µg/m^3)



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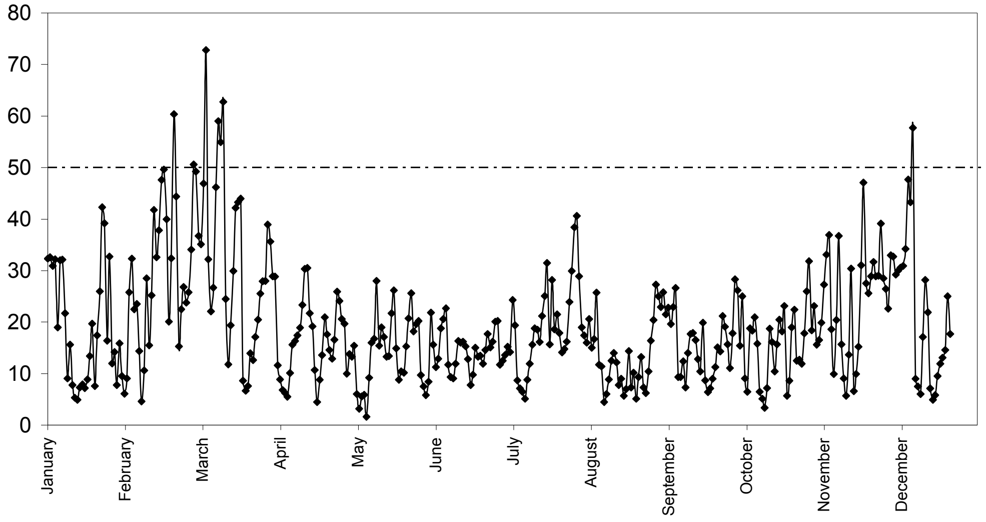

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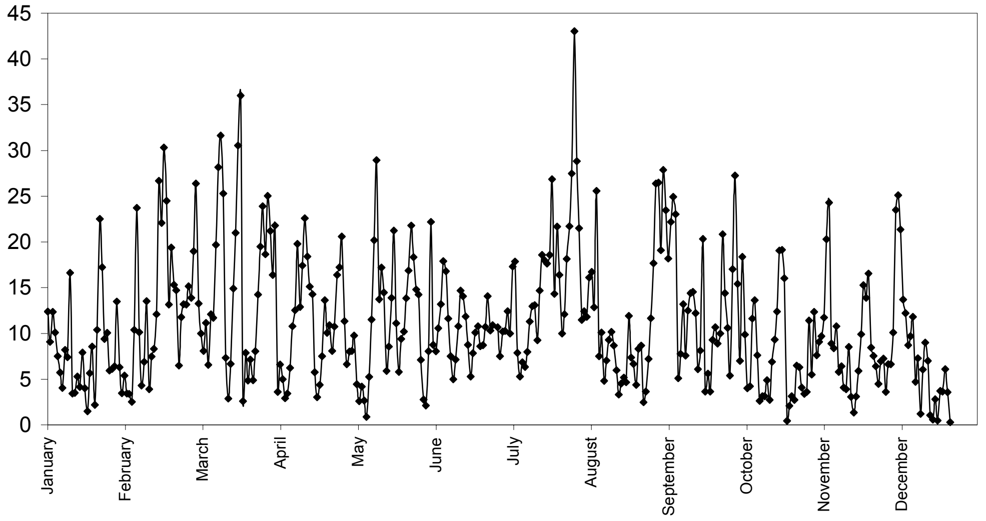
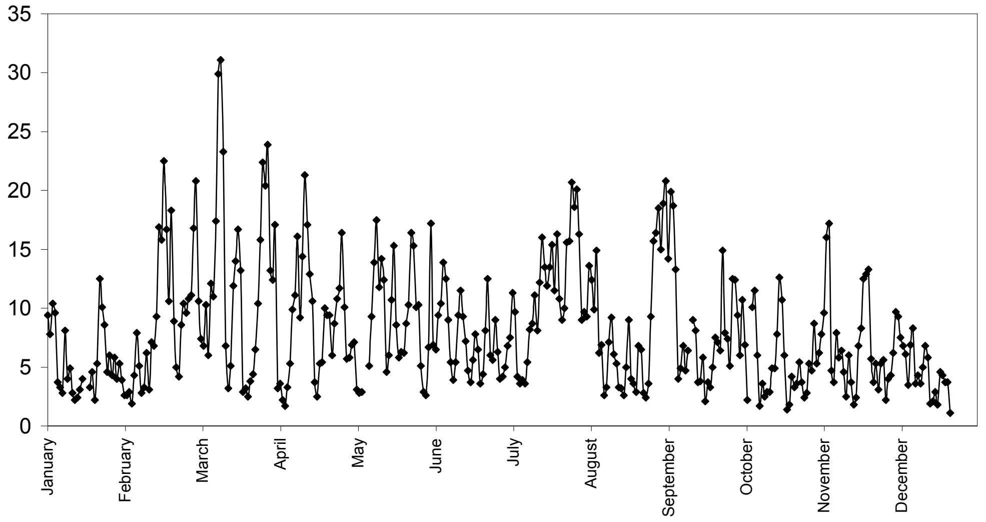
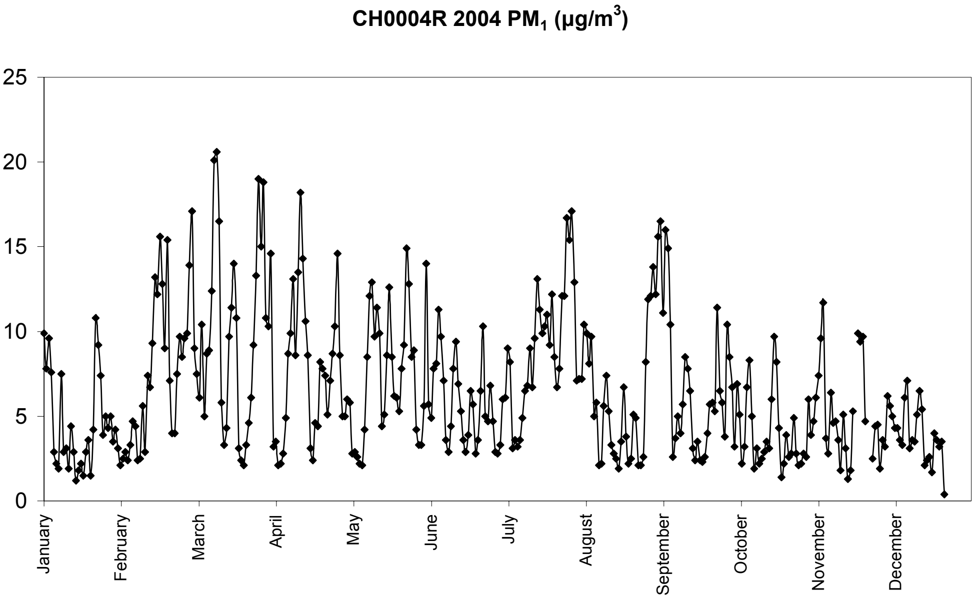
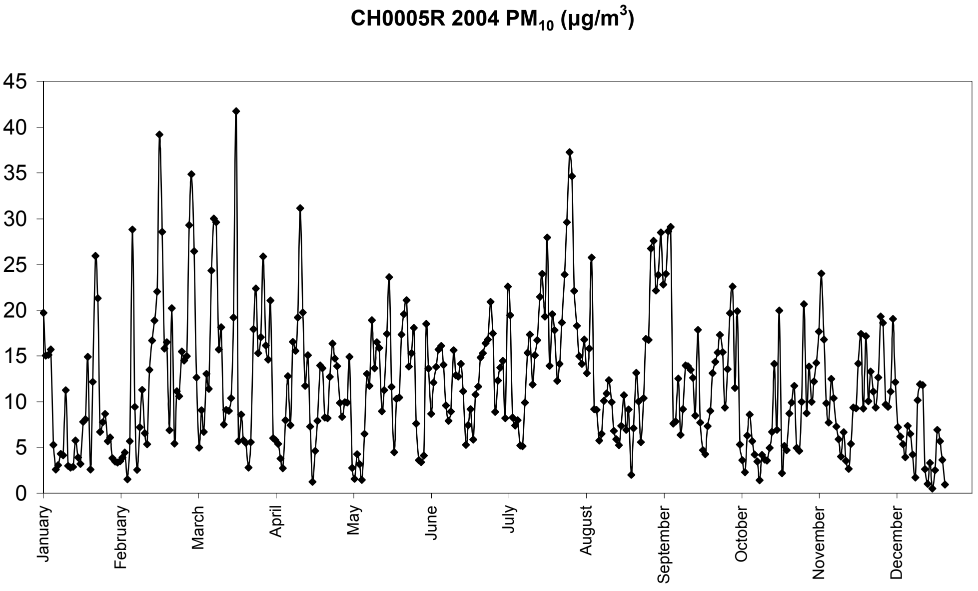
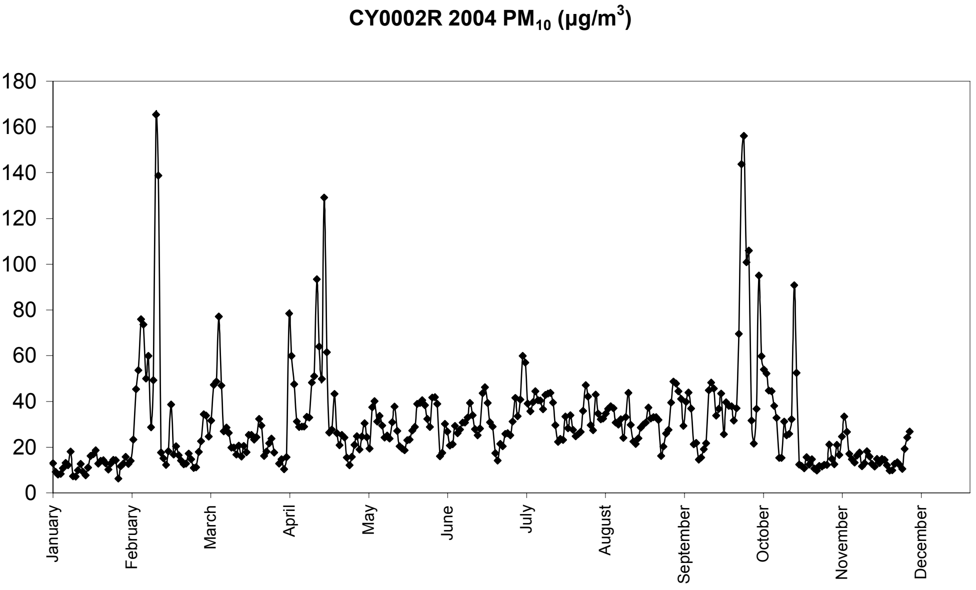


CH0004R 2004 PM$_{2.5}$ (µg/m^3)

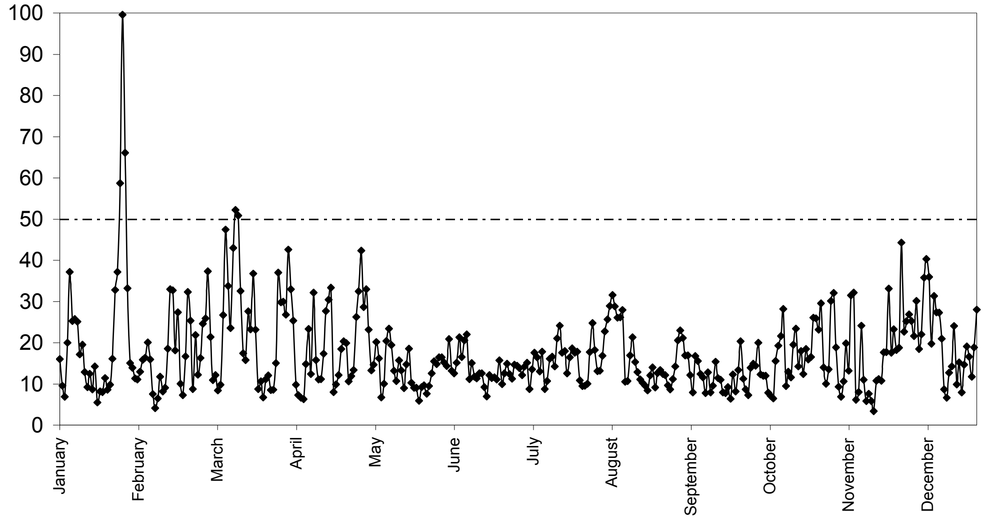



CH0004R 2004 PM$_1$ (µg/m^3)
25
20
15
10
5
0
January
February
March
April
May
June
July
August
September
October
November
December

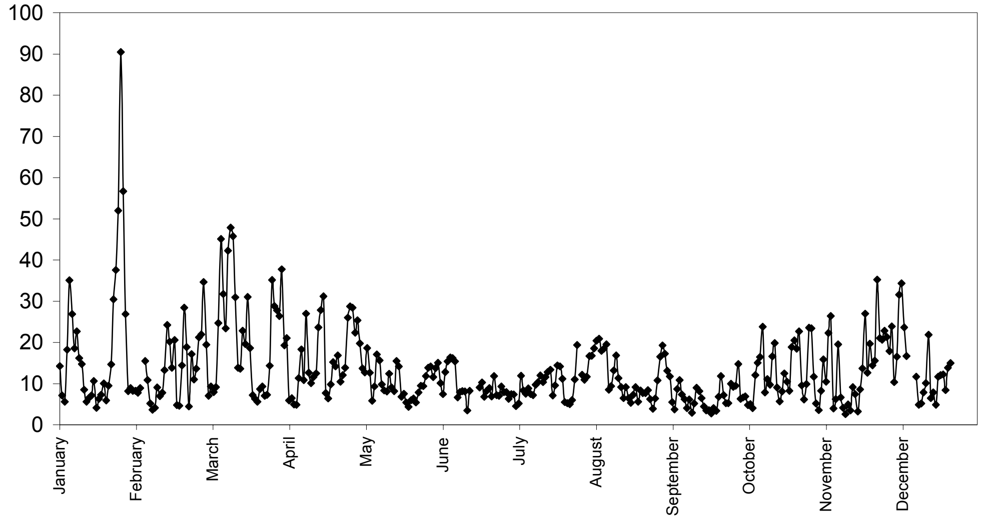

CH0005R 2004 PM$_{10}$ (µg/m^3)
45
40
35
30
25
20
15
10
5
0
January
February
March
April
May
June
July
August
September
October
November
December


CY0002R 2004 PM$_{10}$ (µg/m^3)
180
160
140
120
100
80
60
40
20
0
January
February
March
April
May
June
July
August
September
October
November
December

DE0002R 2004 PM_{10} (µg/m^3)



DE0002R 2004 $PM_{2.5}$ (µg/m^3)



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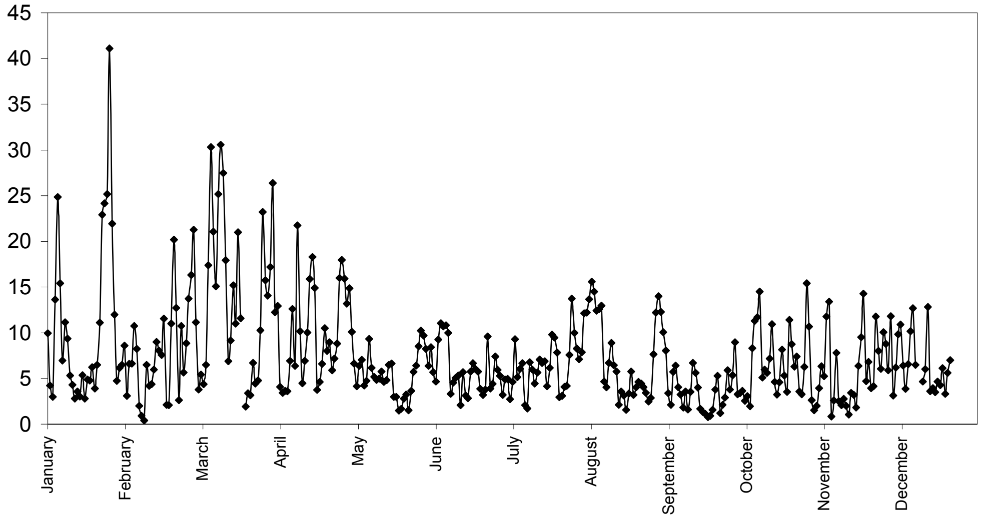
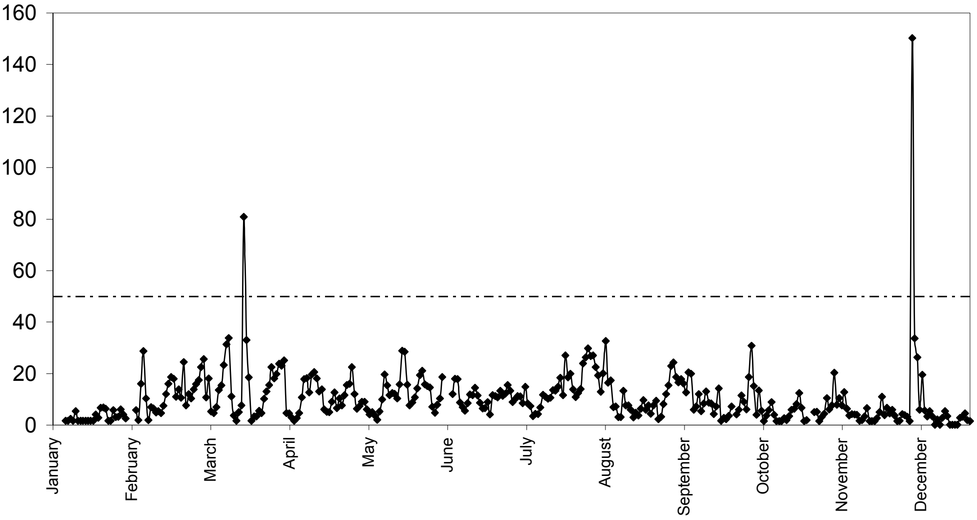
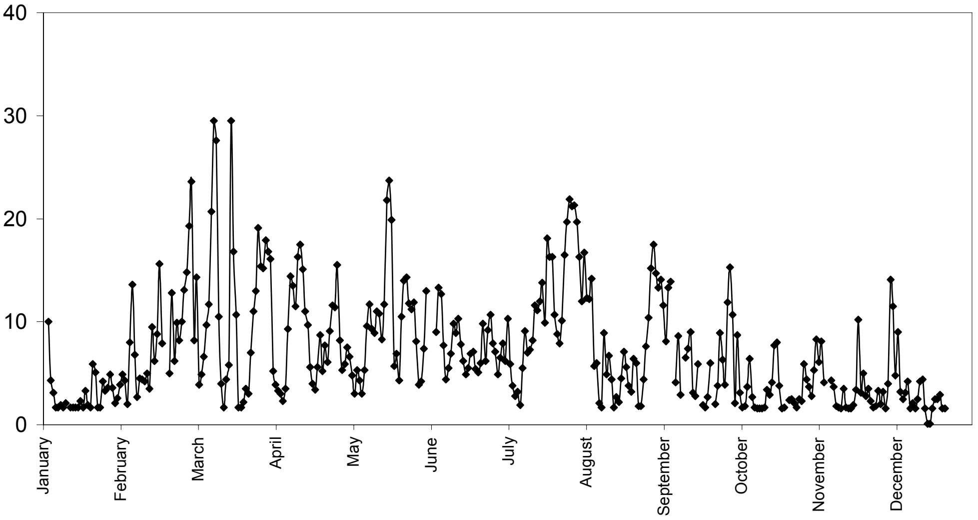


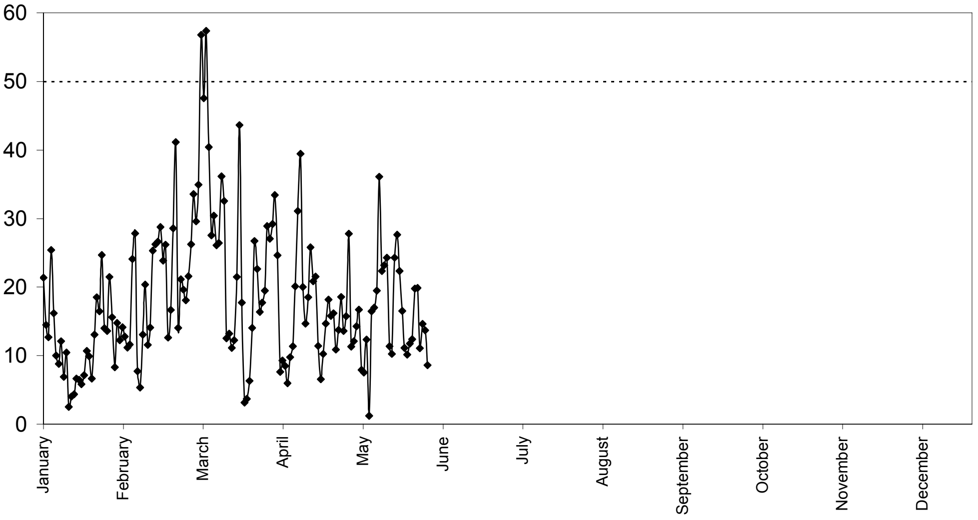
DE0003R 2004 PM_{10} ($\mu g/m^3$)

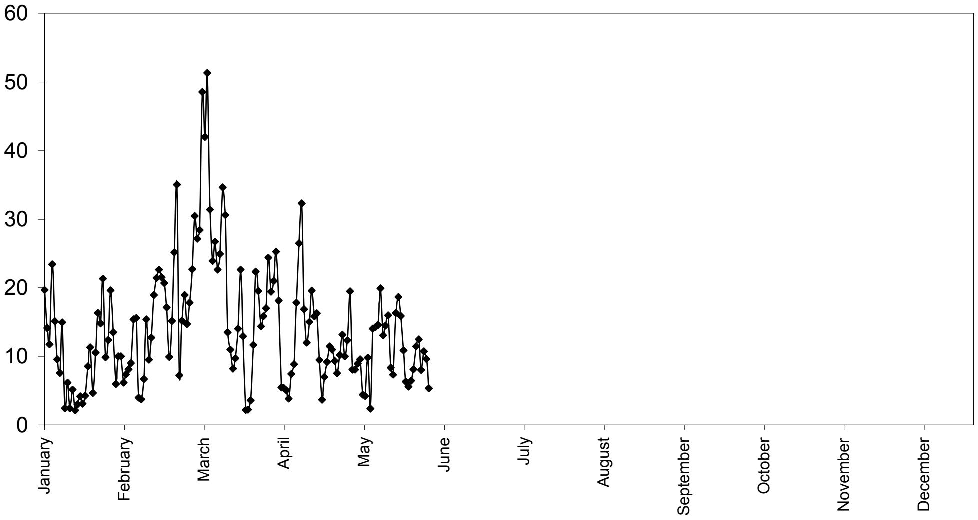


DE0003R 2004 $PM_{2.5}$ ($\mu g/m^3$)

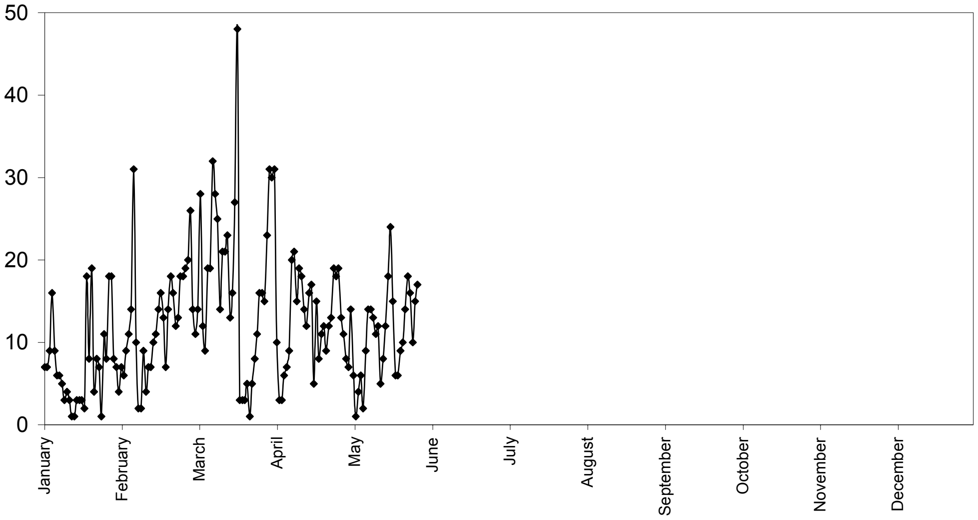


DE0004R 2004 PM_{10} ($\mu g/m^3$)

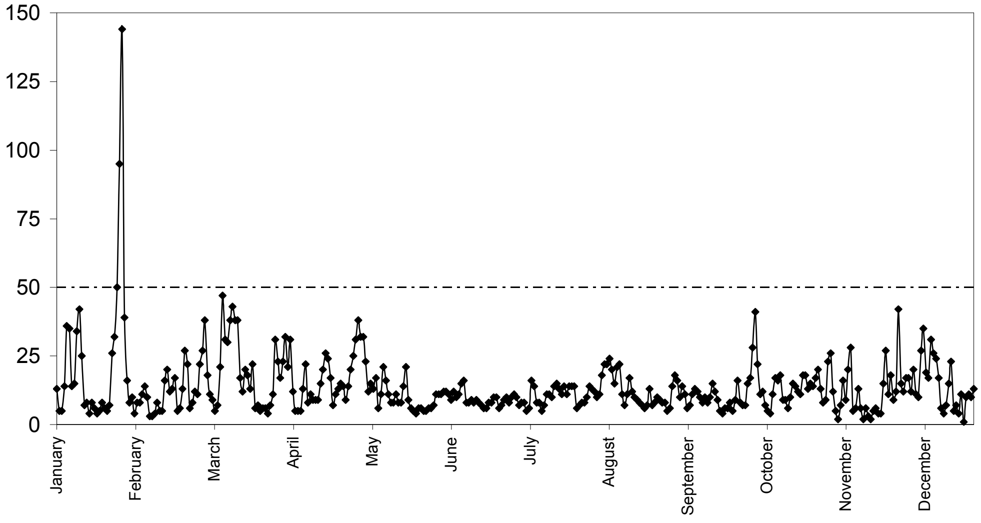




DE0004R 2004 $PM_{2.5}$ (µg/m^3)

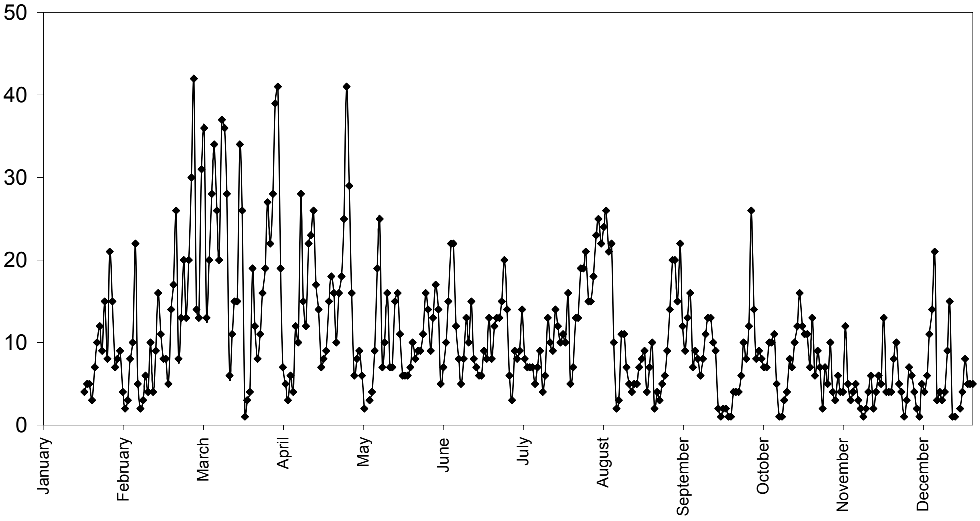


DE0005R 2004 PM_{10} (µg/m^3)

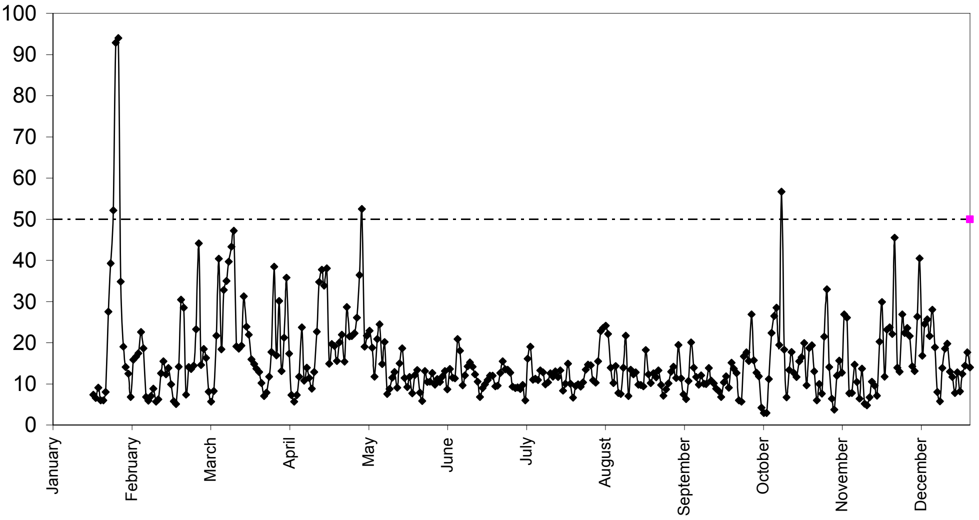


DE0007R 2004 PM_{10} (µg/m^3)

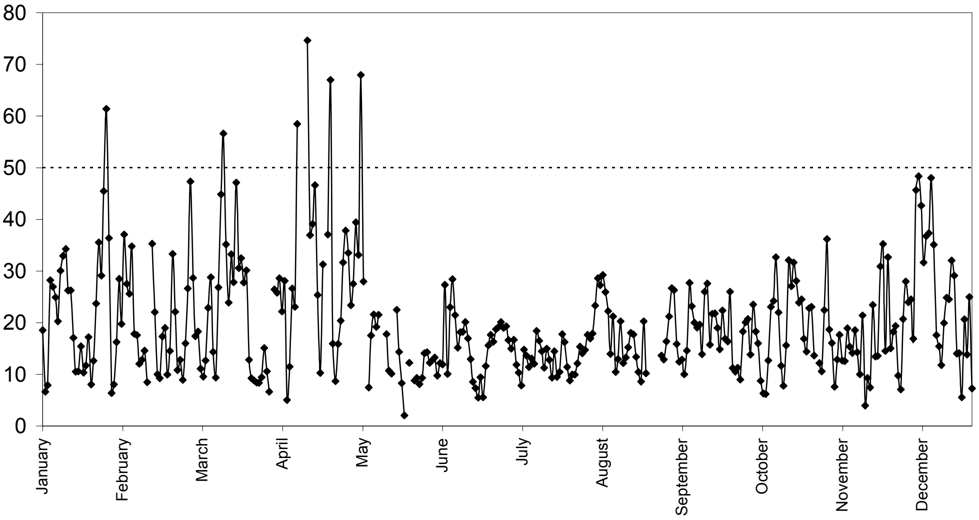
DE0008R 2004 PM_{10} (µg/m^3)



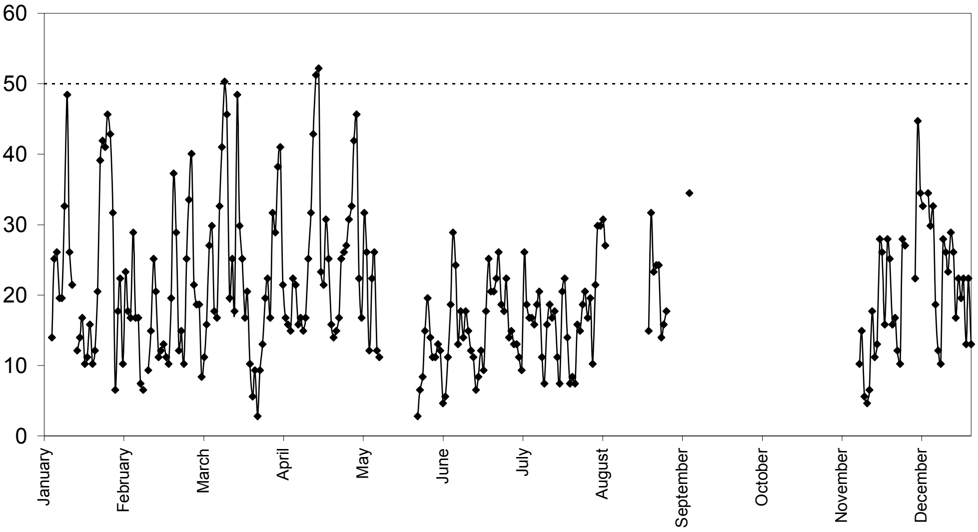
DE0009R 2004 PM_{10} (µg/m^3)



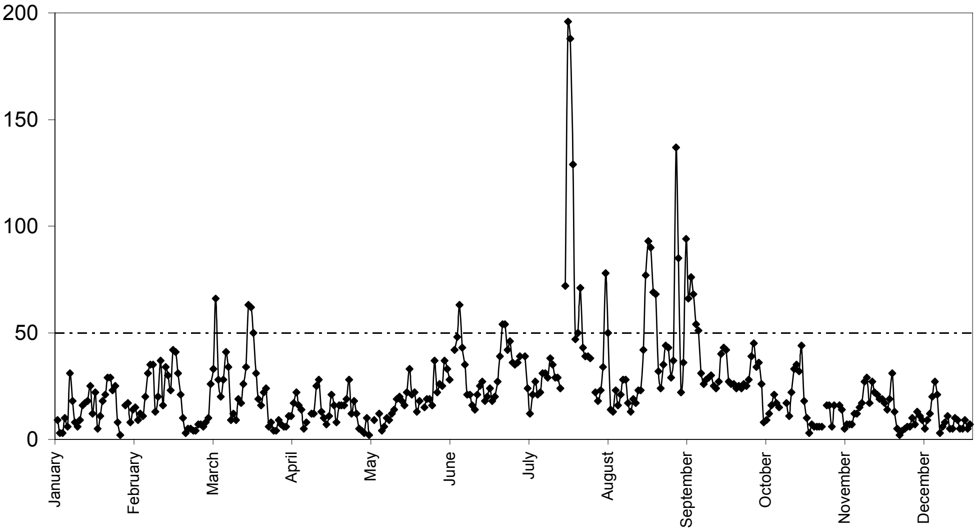
DE0041R 2004 PM_{10} (µg/m^3)



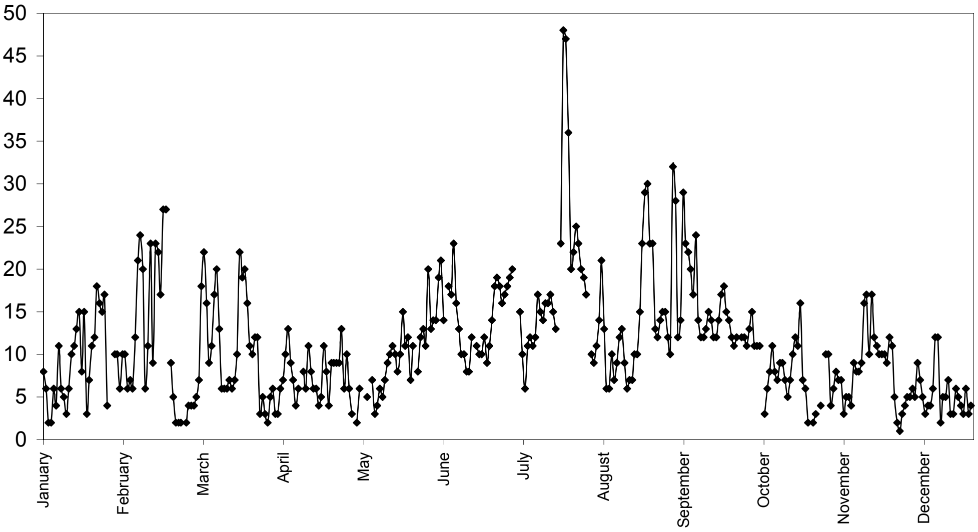
DK0005R 2004 PM$_{10}$ (µg/m^3)

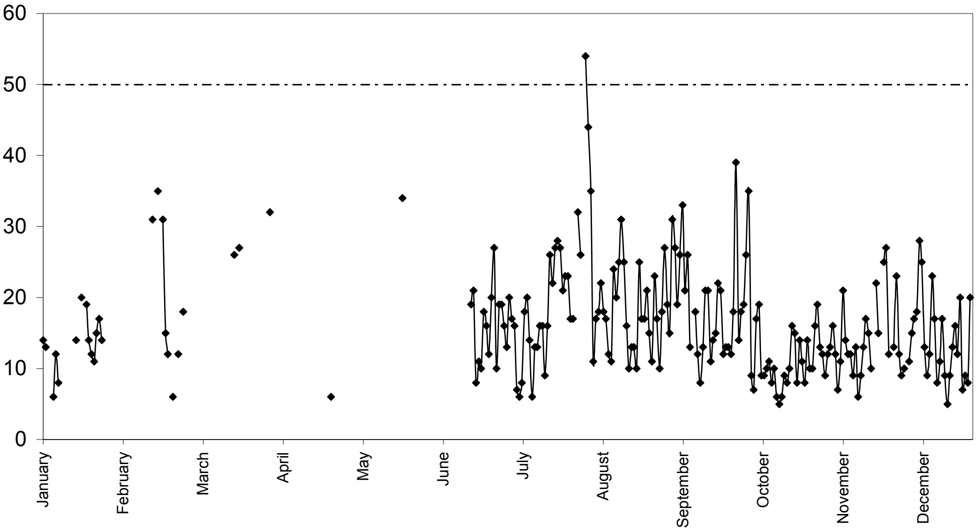


ES0007R 2004 PM$_{10}$ (µg/m^3)

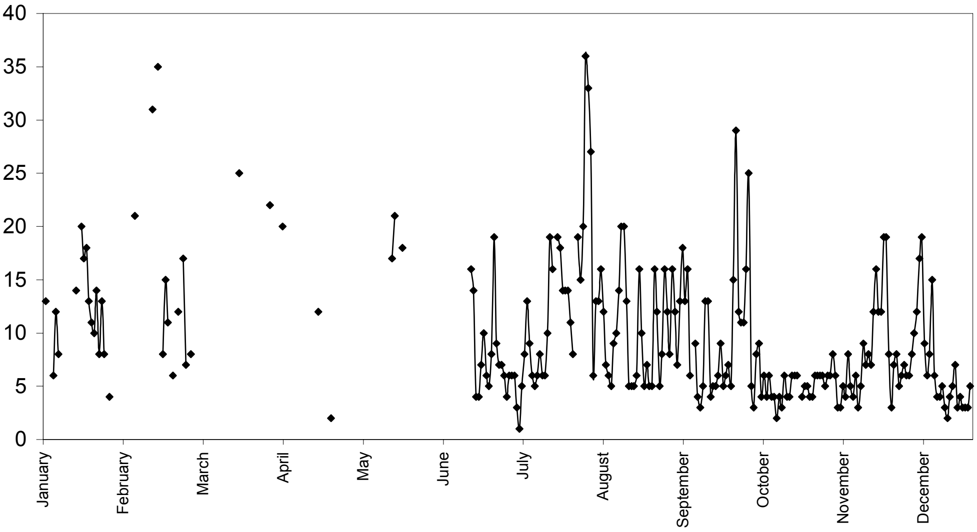


ES0007R 2004 PM$_{2.5}$ (µg/m^3)

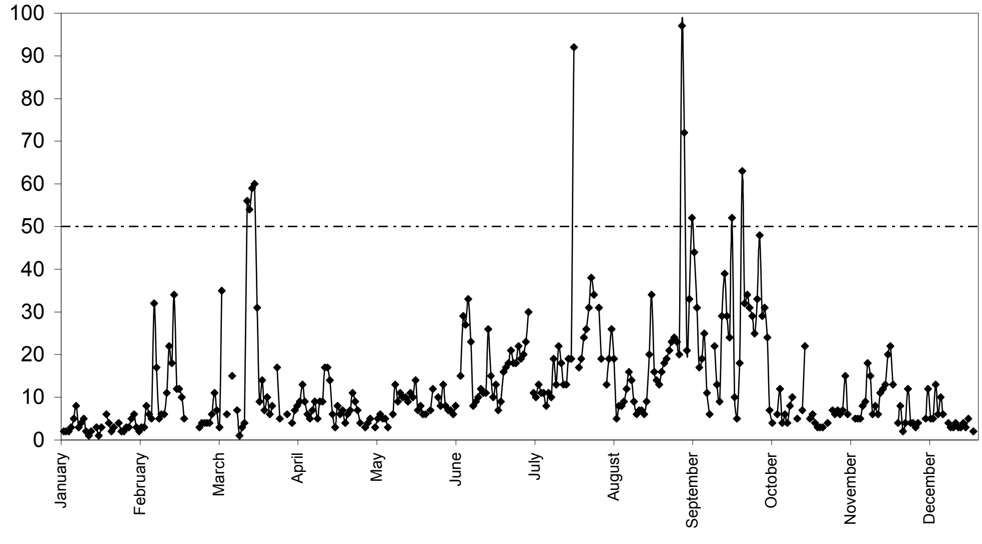




ES0008R 2004 PM_{10} (µg/m^3)

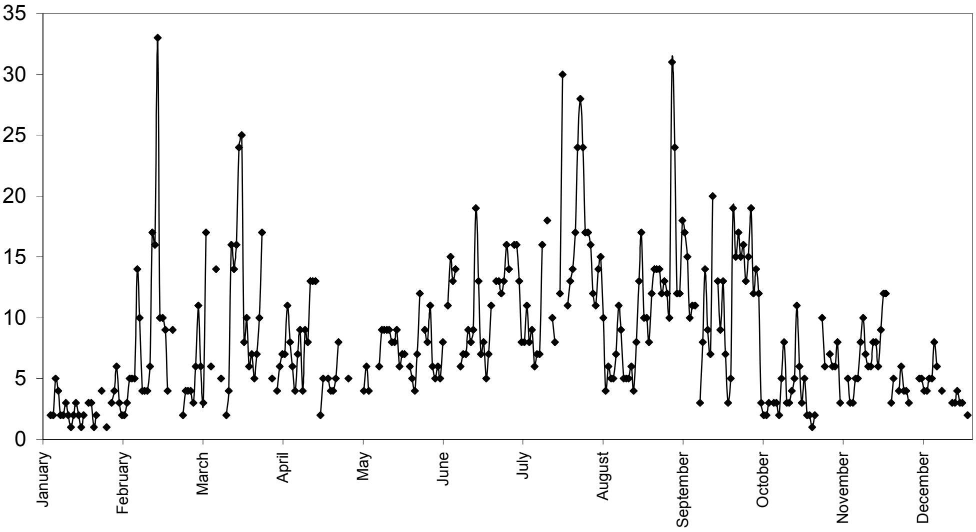


ES0008R 2004 $PM_{2.5}$ (µg/m^3)

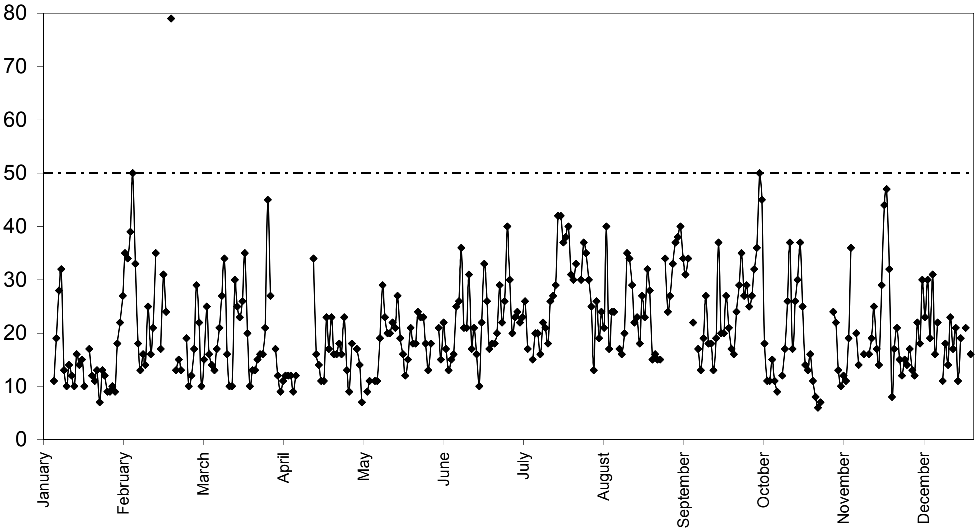


ES0009R 2004 PM_{10} (µg/m^3)

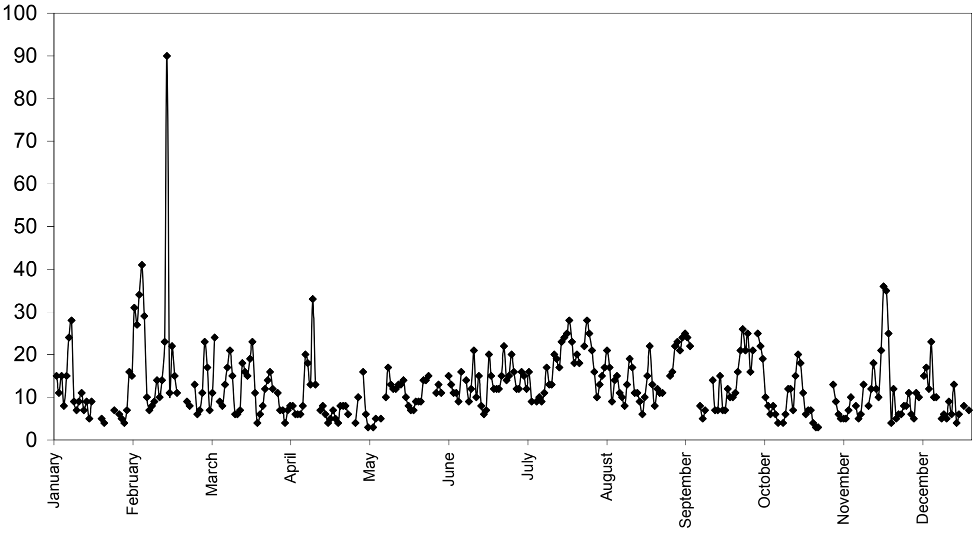
ES0009R 2004 $PM_{2.5}$ (µg/m^3)



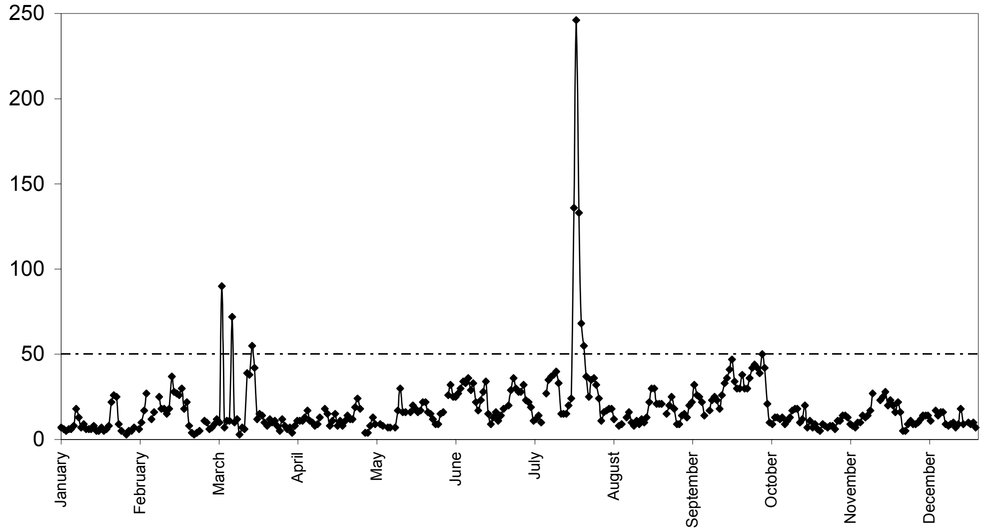
ES0010R 2004 PM_{10} (µg/m^3)



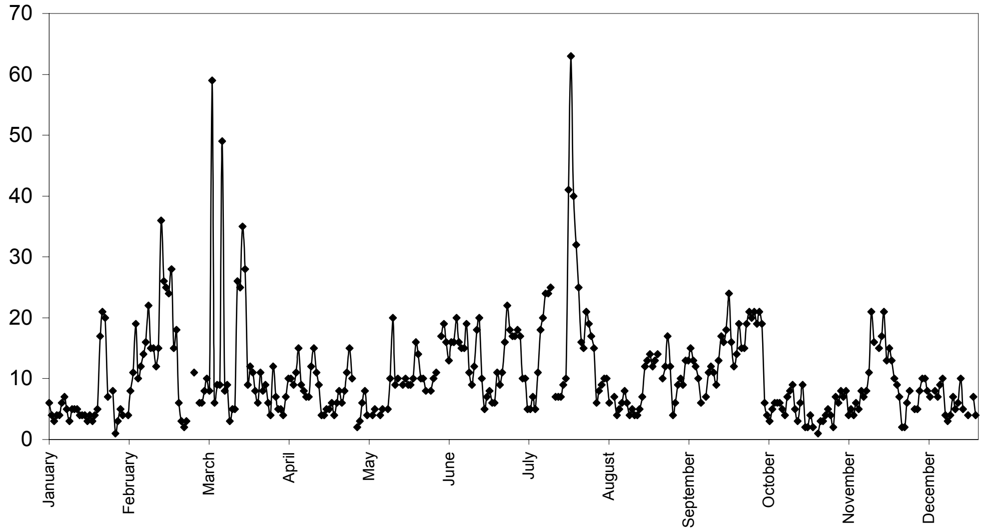
ES0010R 2004 $PM_{2.5}$ (µg/m^3)



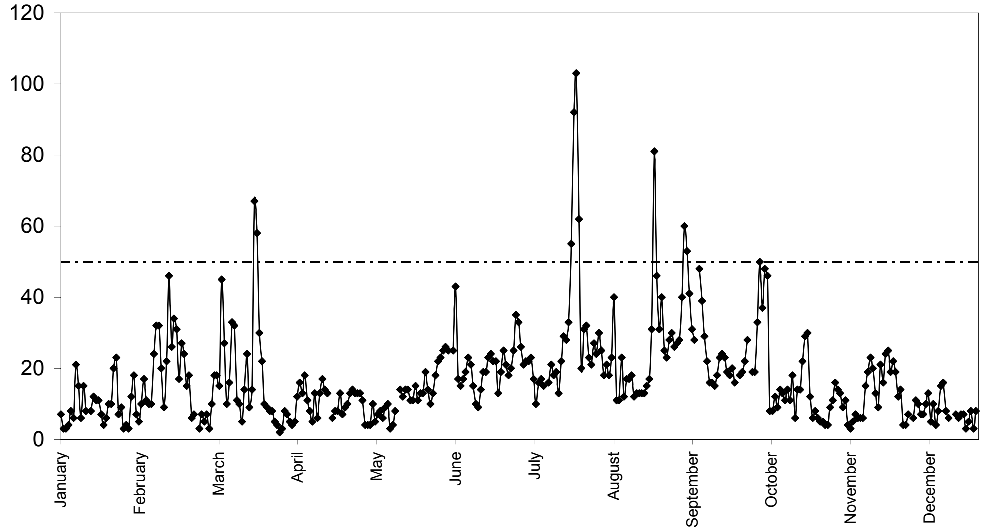
ES0011R 2004 PM_{10} ($\mu g/m^3$)



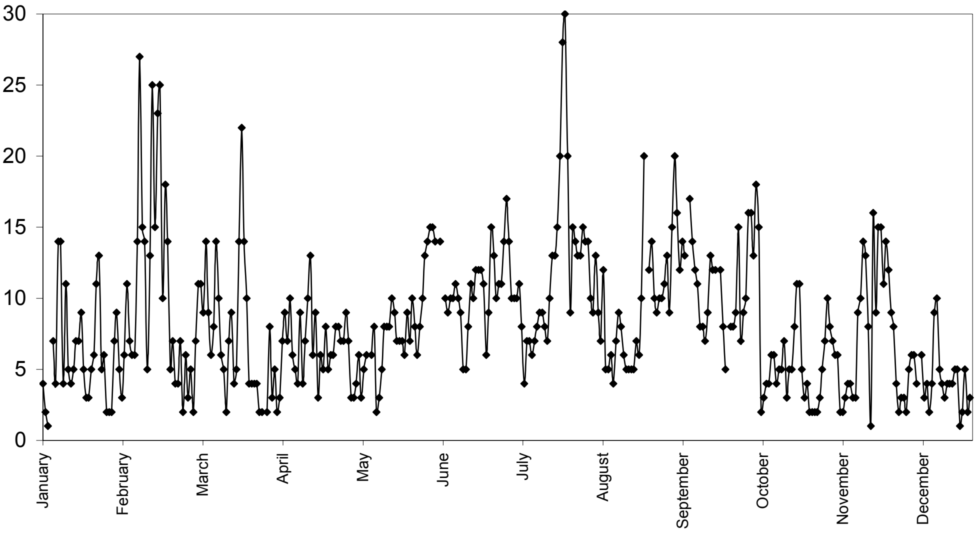
ES0011R 2004 $PM_{2.5}$ ($\mu g/m^3$)



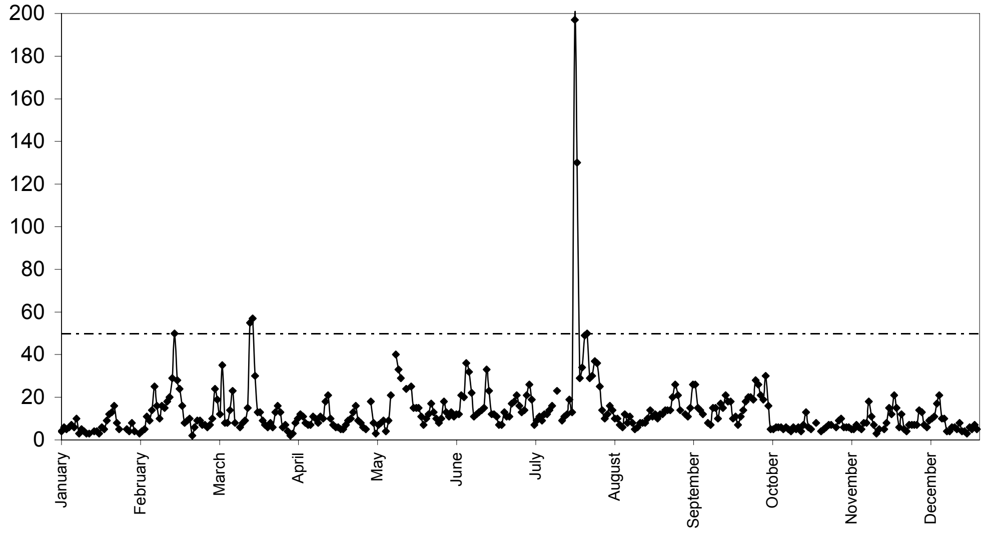
ES0012R 2004 PM_{10} ($\mu g/m^3$)



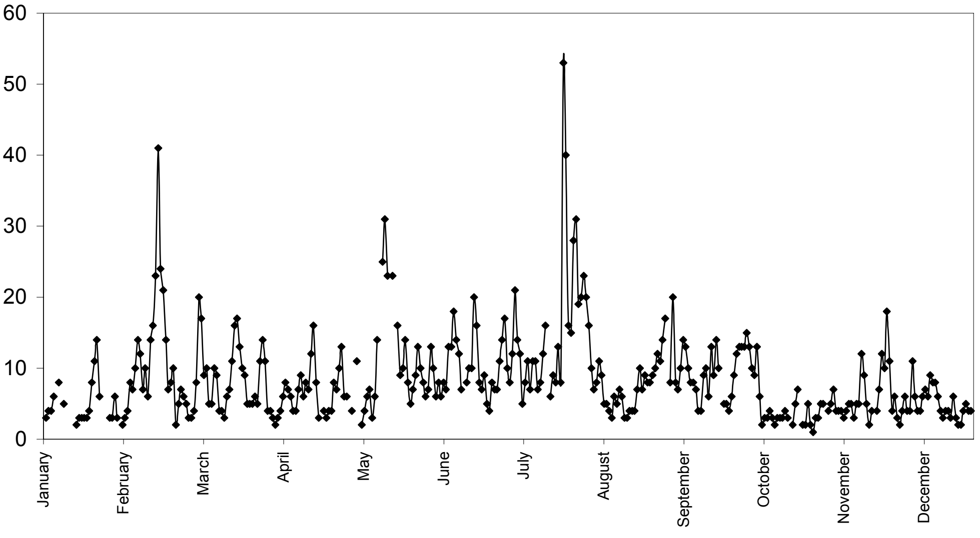
ES0012R 2004 $PM_{2.5}$ (µg/m^3)



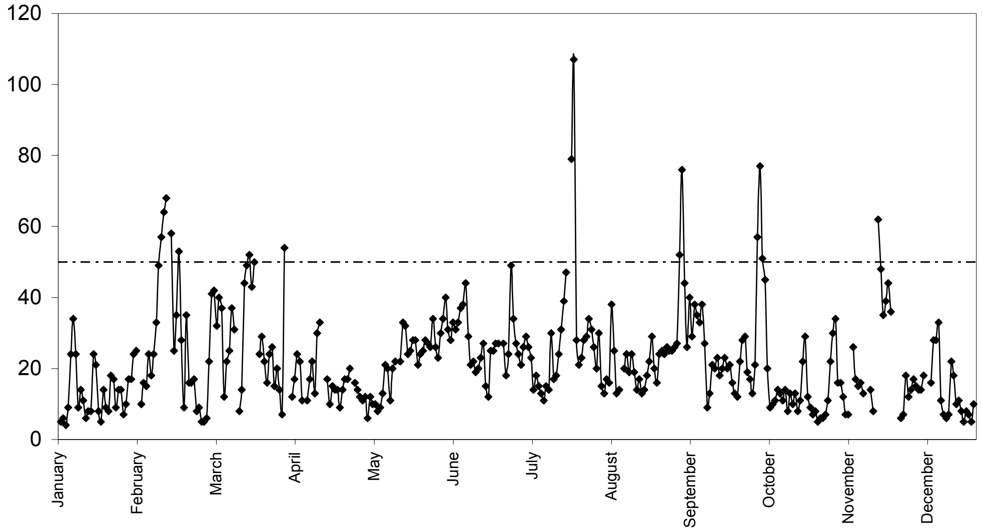
ES0013R 2004 PM_{10} (µg/m^3)



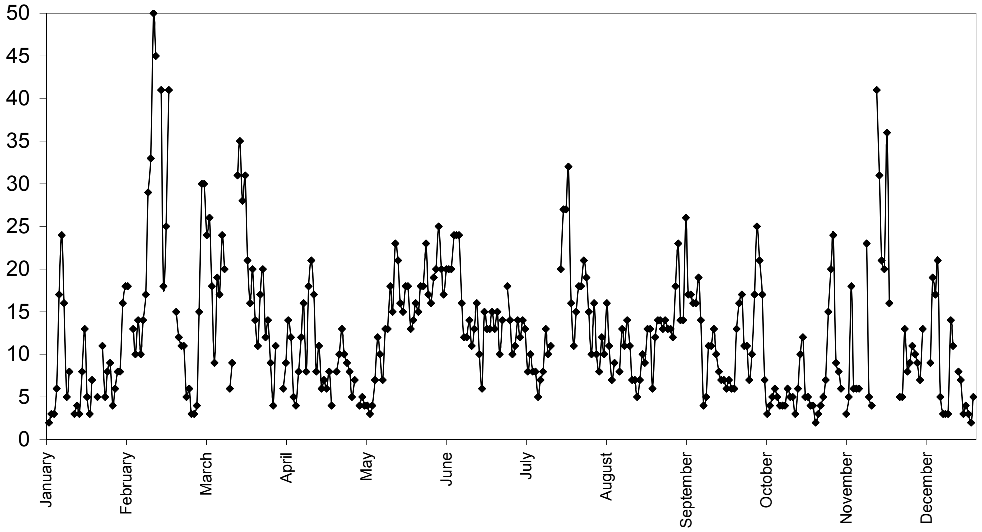
ES0013R 2004 $PM_{2.5}$ (µg/m^3)



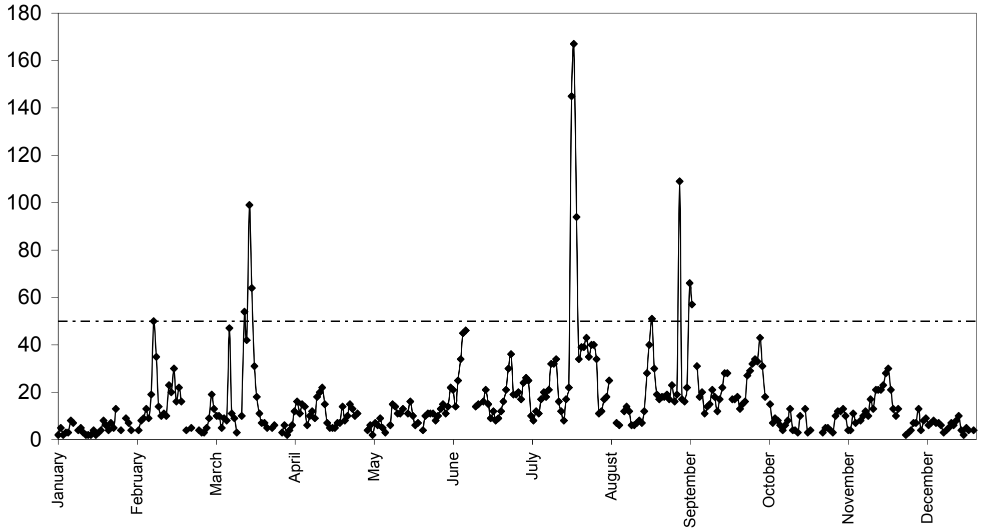
ES0014R 2004 PM$_{10}$ (µg/m^3)

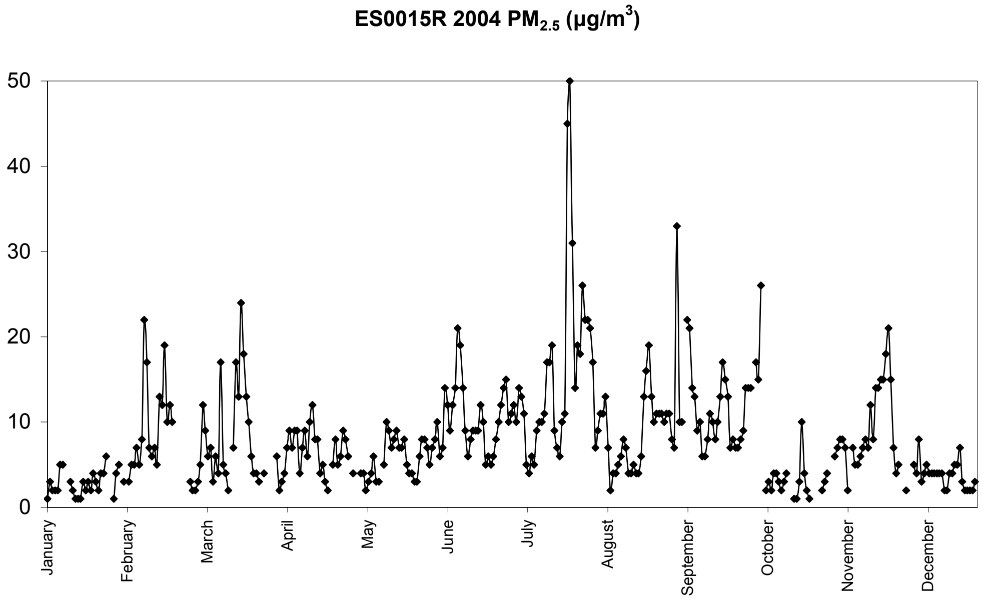


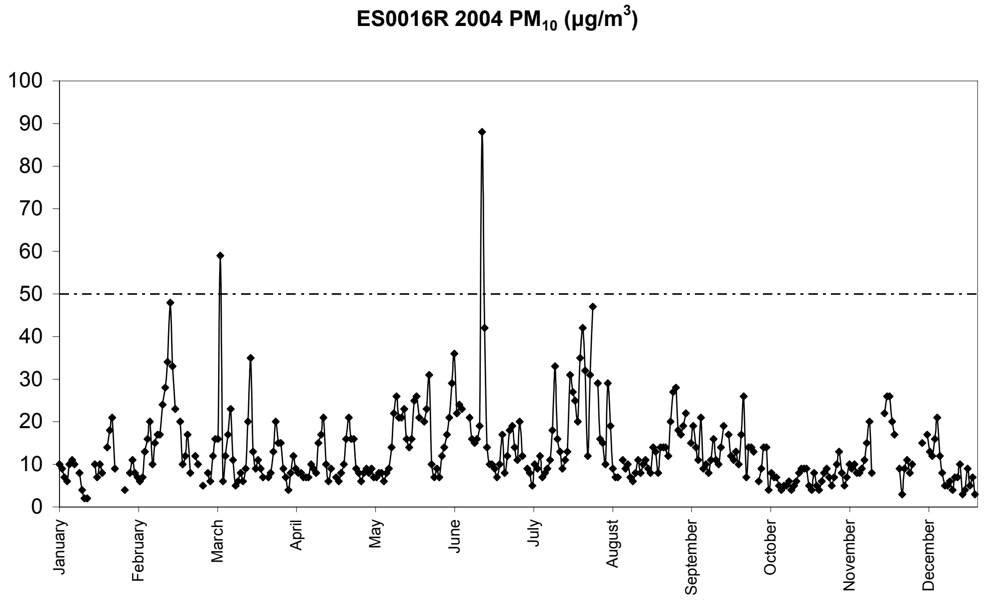
ES0014R 2004 PM$_{2.5}$ (µg/m^3)

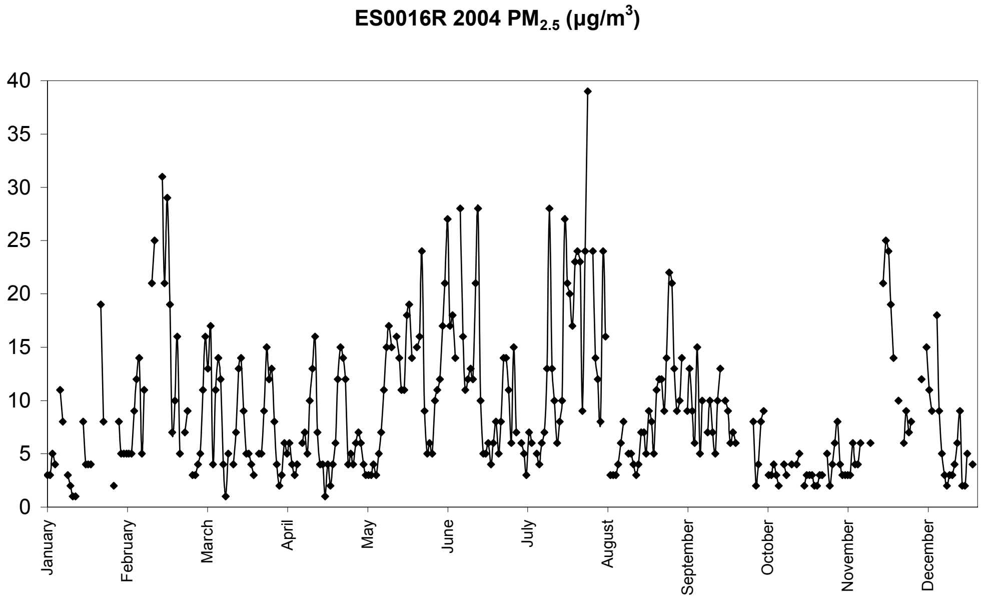


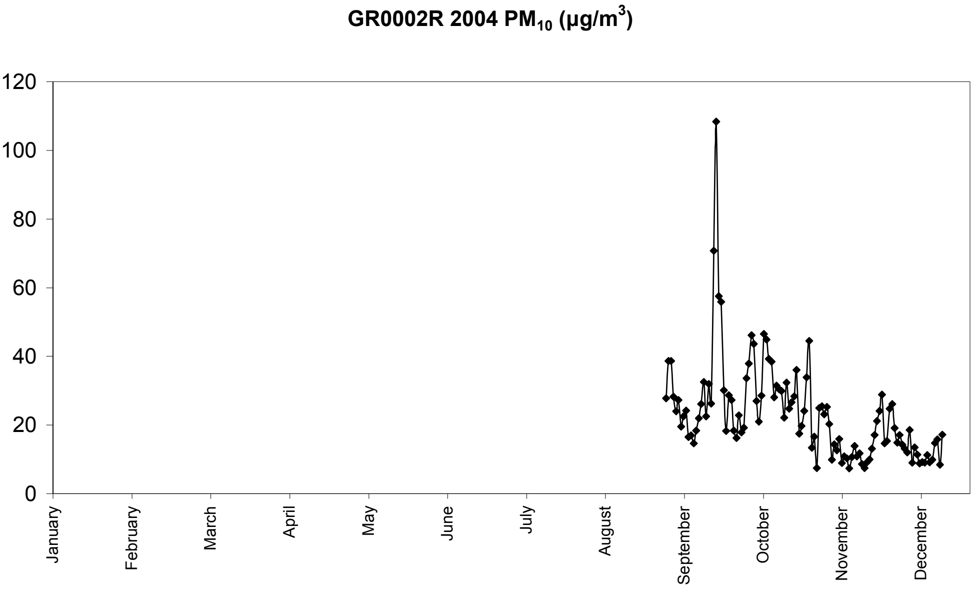
ES0015R 2004 PM$_{10}$ (µg/m^3)

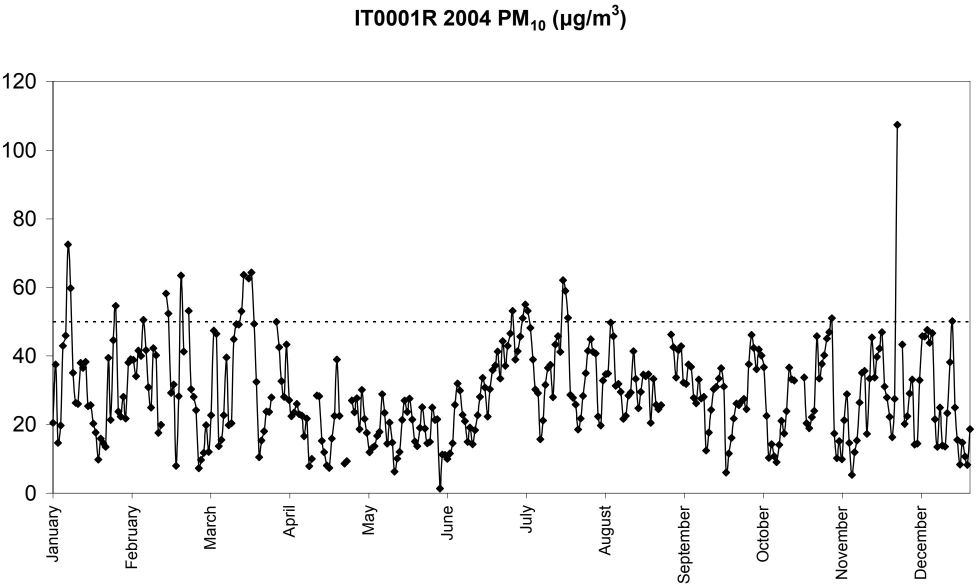


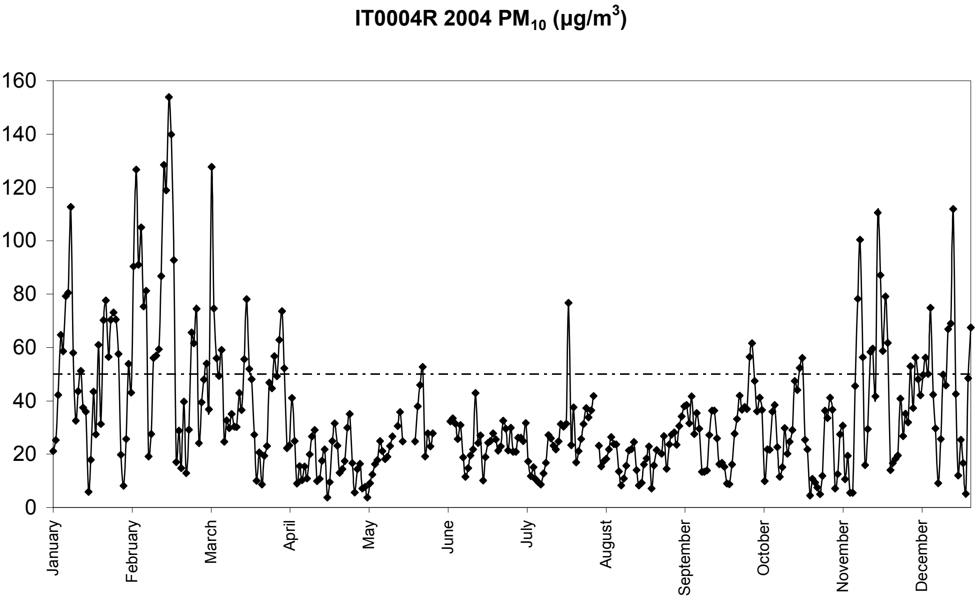

ES0015R 2004 PM$_{2.5}$ (µg/m^3)
0
10
20
30
40
50
January
February
March
April
May
June
July
August
September
October
November
December


ES0016R 2004 PM$_{10}$ (µg/m^3)
0
10
20
30
40
50
60
70
80
90
100
January
February
March
April
May
June
July
August
September
October
November
December

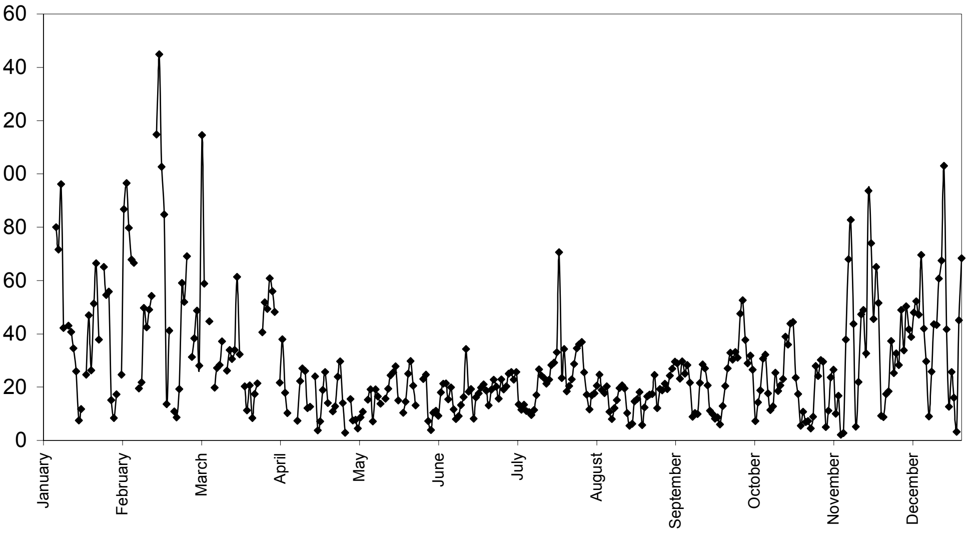

ES0016R 2004 PM$_{2.5}$ (µg/m^3)
0
5
10
15
20
25
30
35
40
January
February
March
April
May
June
July
August
September
October
November
December


GR0002R 2004 PM$_{10}$ (µg/m^3)
0
20
40
60
80
100
120
January
February
March
April
May
June
July
August
September
October
November
December

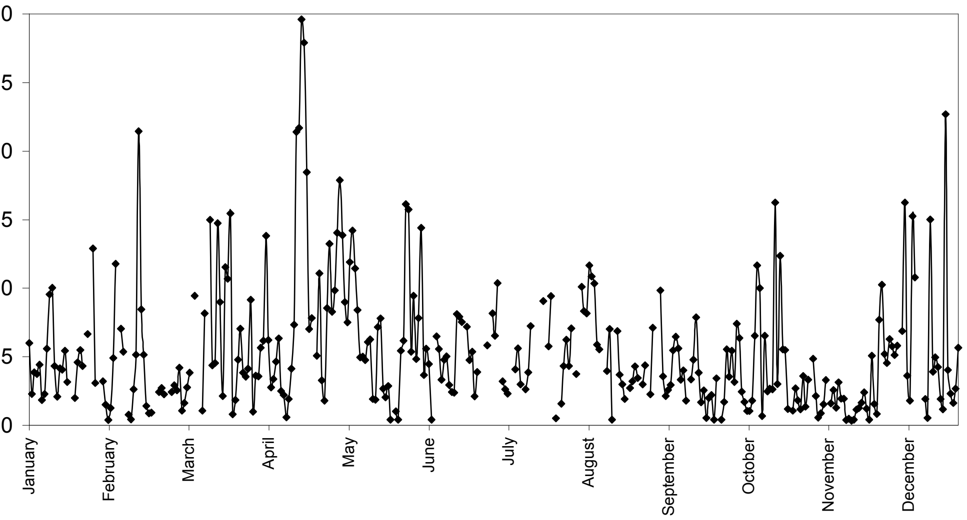

IT0001R 2004 PM$_{10}$ (µg/m^3)
0
20
40
60
80
100
120
January
February
March
April
May
June
July
August
September
October
November
December


IT0004R 2004 PM$_{10}$ (µg/m^3)
0
20
40
60
80
100
120
140
160
January
February
March
April
May
June
July
August
September
October
November
December

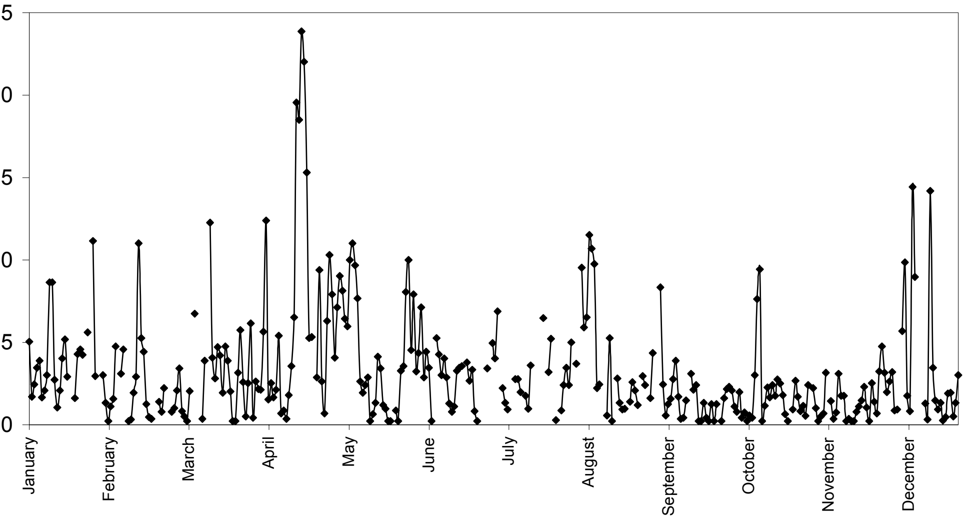
IT0004R 2004 $PM_{2.5}$ ($\mu g/m^3$)



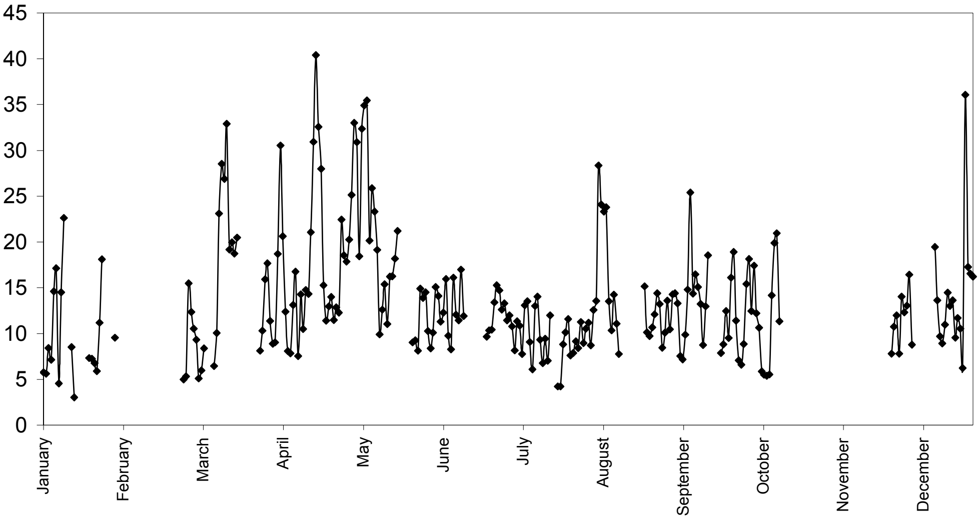
NO0001R 2004 PM_{10} ($\mu g/m^3$)



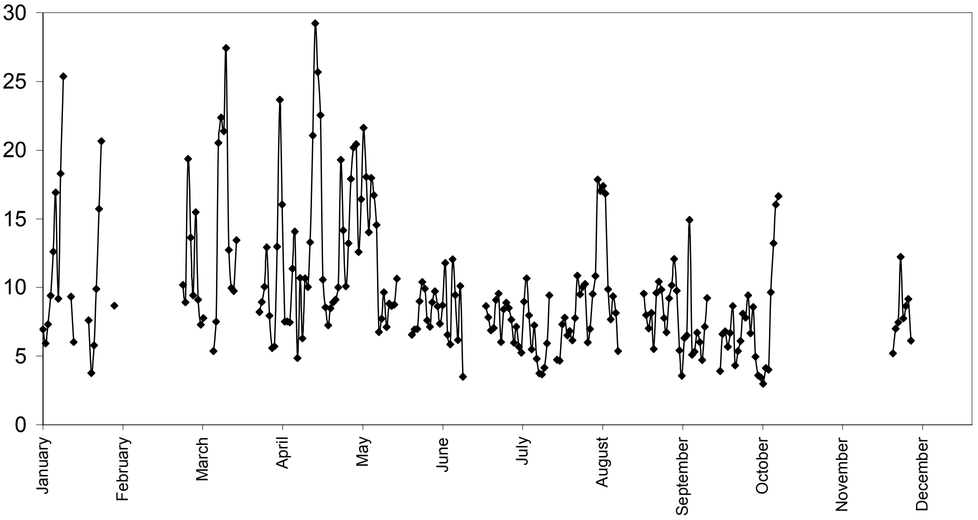
NO0001R 2004 $PM_{2.5}$ ($\mu g/m^3$)



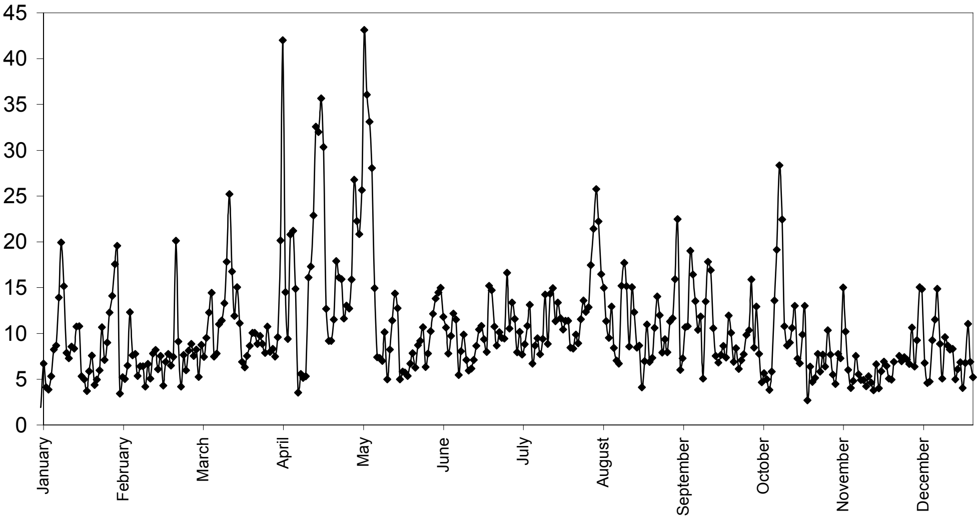
SE0011R 2004 PM_{10} (µg/m^3)



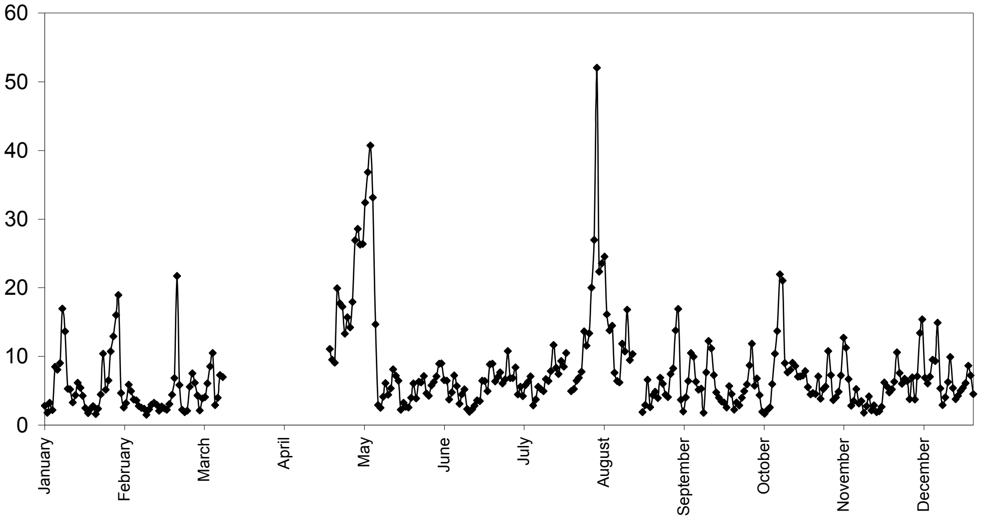
SE0011R 2004 $PM_{2.5}$ (µg/m^3)



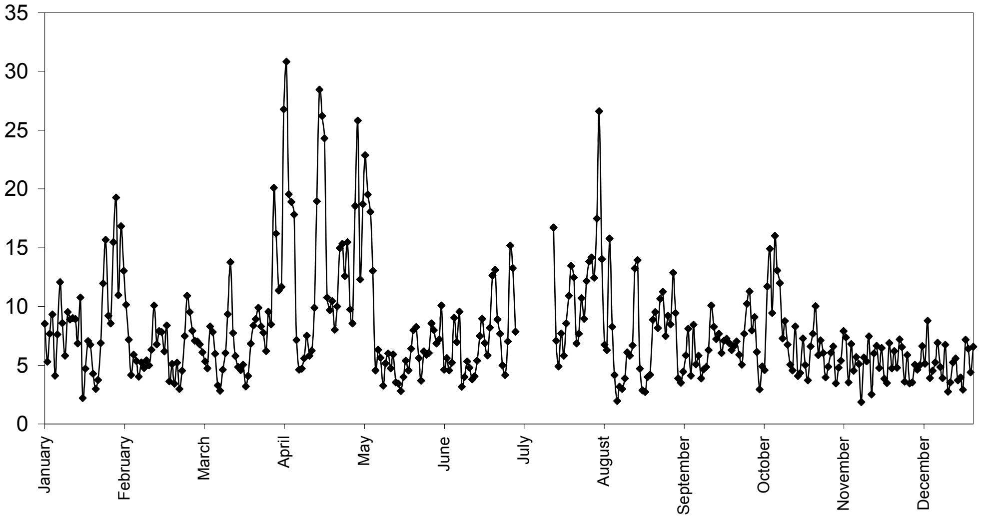
SE0012R 2004 PM_{10} (µg/m^3)



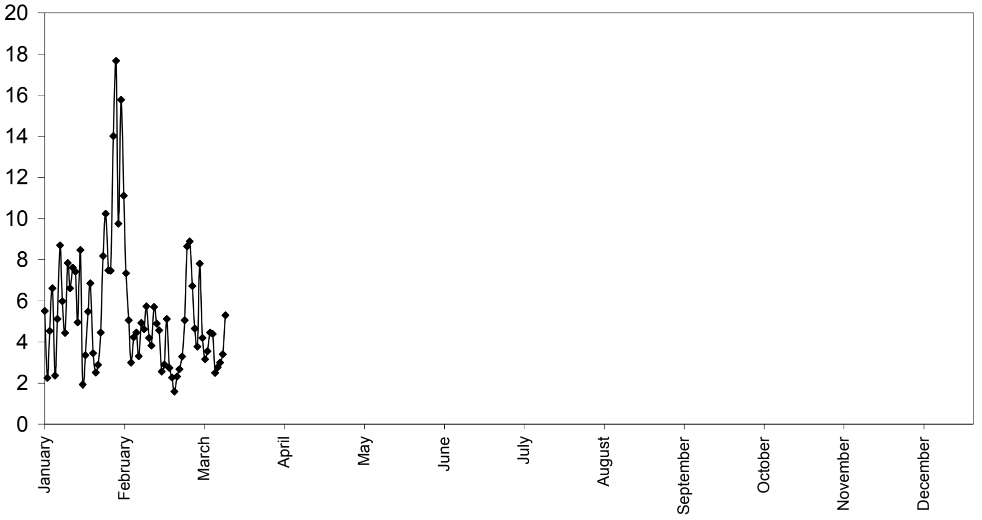
SE0012R 2004 PM$_{2.5}$ (µg/m^3)



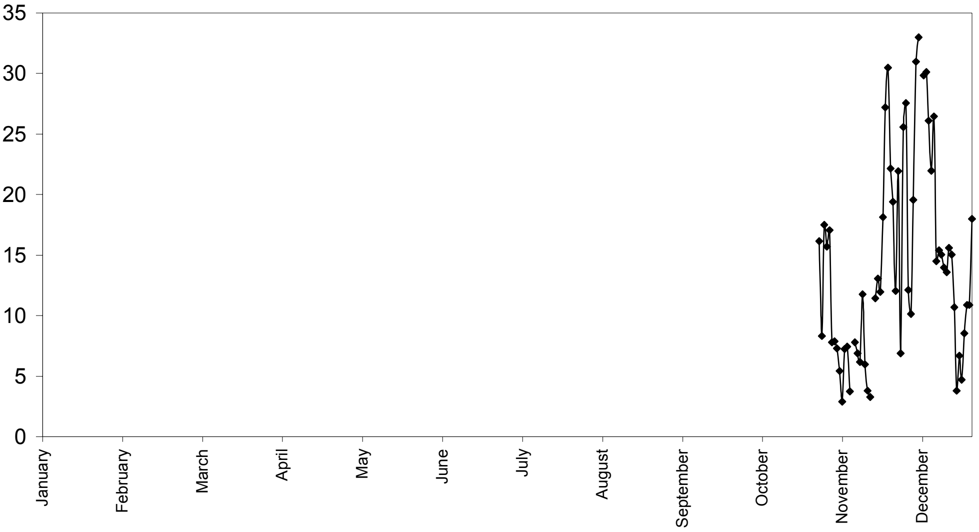
SE0035R 2004 PM$_{10}$ (µg/m^3)



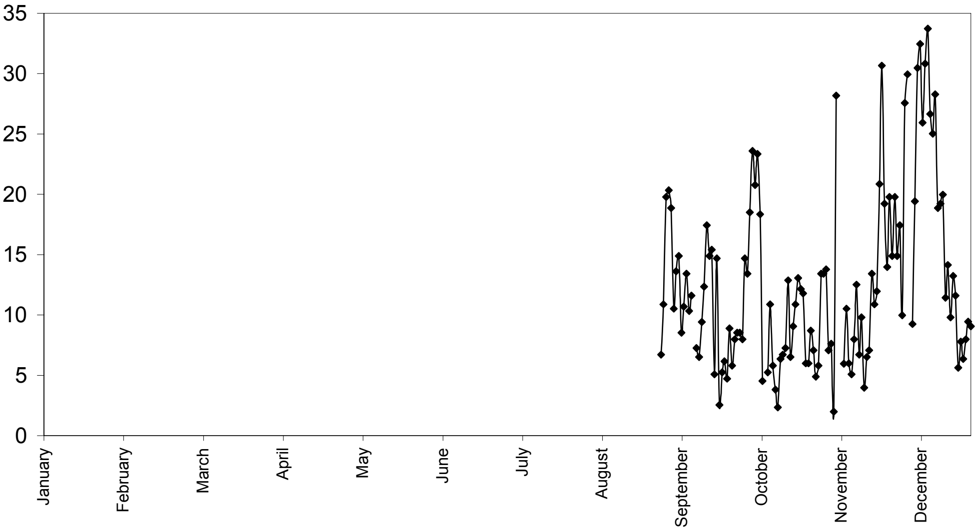
SE0035R 2004 PM$_{2.5}$ (µg/m^3)



SI0008R 2004 PM_{10} (µg/m^3)



SI0008R 2004 $PM_{2.5}$ (µg/m^3)

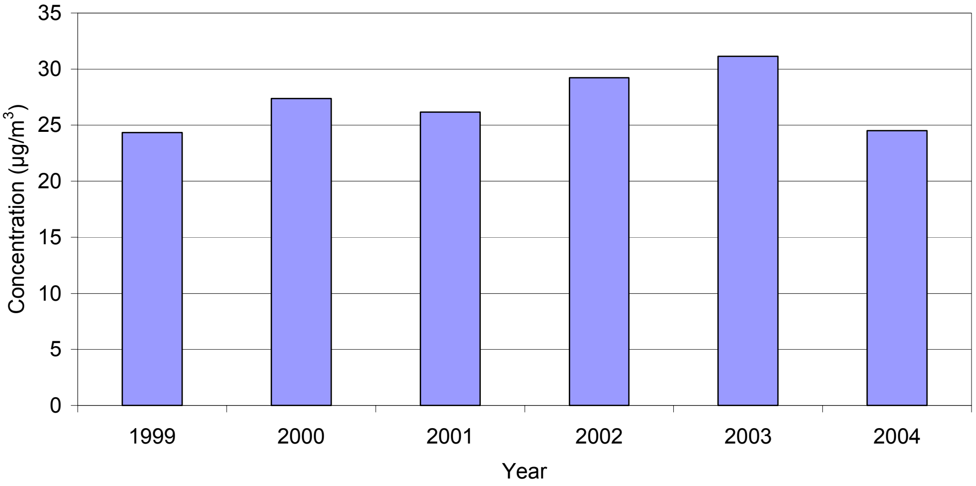


Annex 2

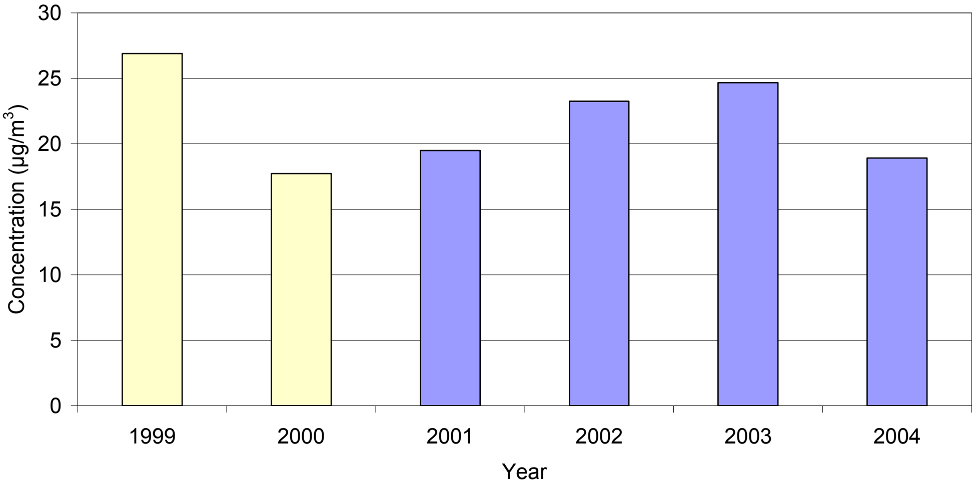
Series of annual mean particulate mass concentrations at selected EMEP stations

The annual average concentration of $PM_{2.5}$ reported in some stations was higher than that of PM_{10} values due to the fact that the data were collected at different time periods (i.e. the station AT0002 – Austria during 1999).

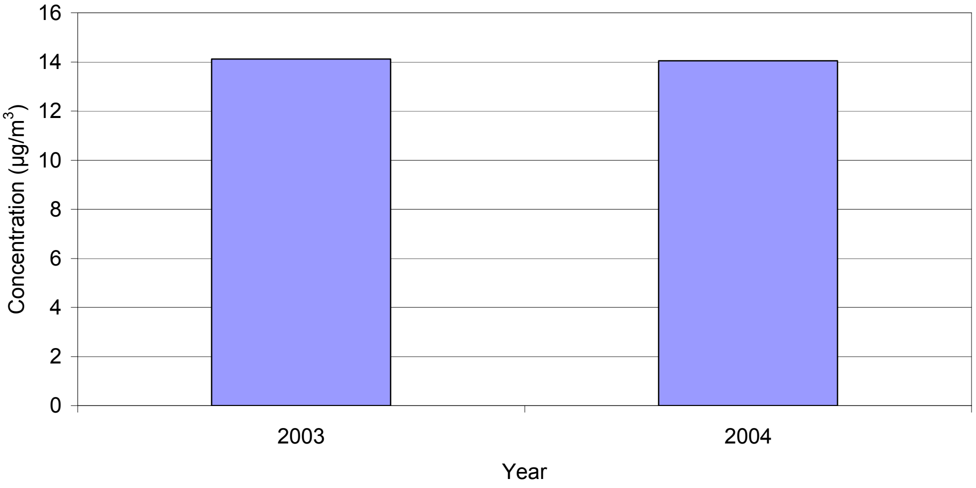
Data columns depicted in ivory colours correspond to stations with data capture < 50% in a specific year.



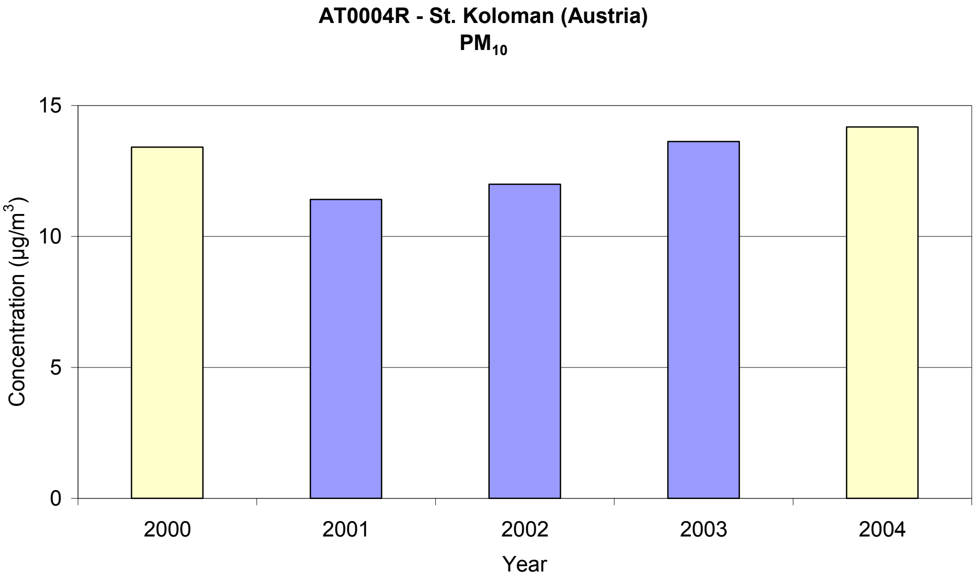
AT0002R - Illmitz (Austria)
PM_{10}

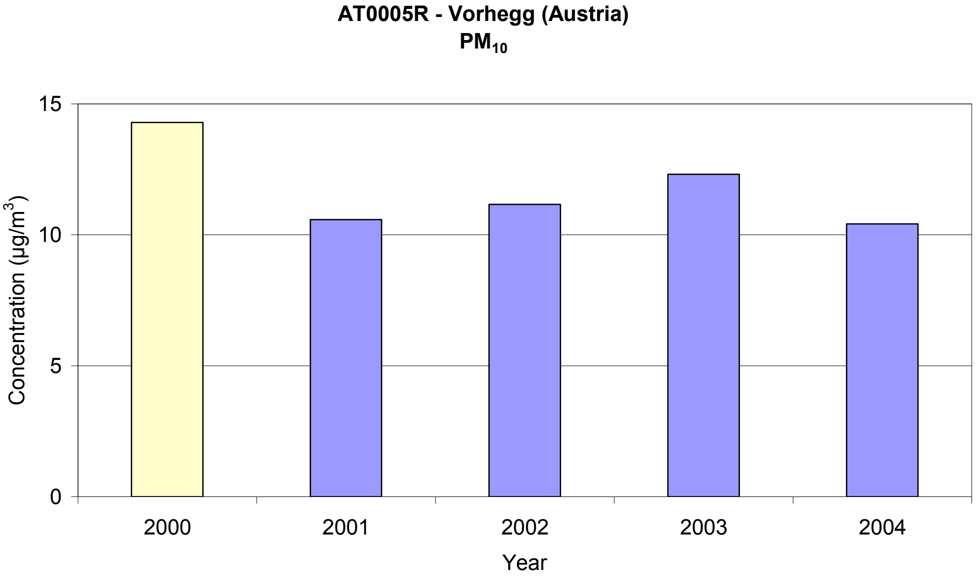


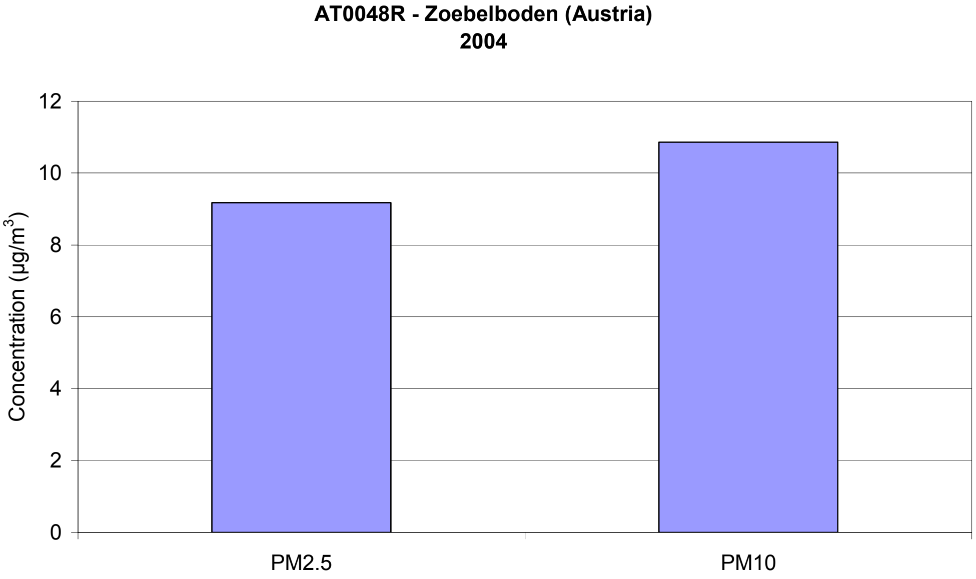
AT0002R - Illmitz (Austria)
$PM_{2.5}$



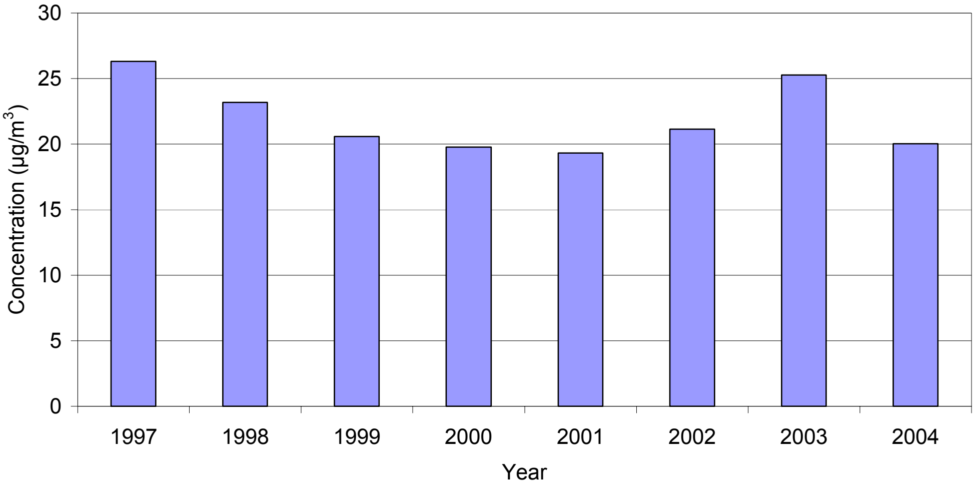
AT0002R - Illmitz (Austria)
PM_1


AT0004R - St. Koloman (Austria)
PM$_{10}$
Concentration (µg/m^3)
0
5
10
15
2000
2001
2002
2003
2004
Year

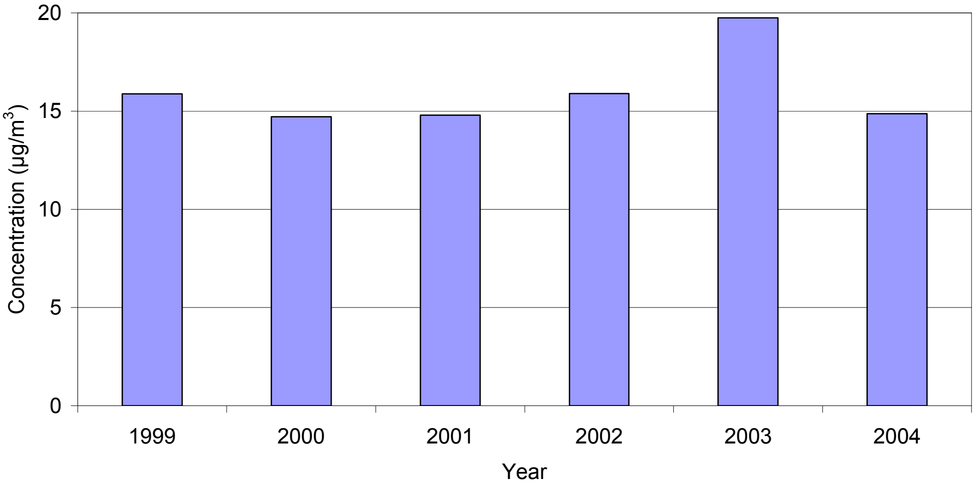

AT0005R - Vorhegg (Austria)
PM$_{10}$
Concentration (µg/m^3)
0
5
10
15
2000
2001
2002
2003
2004
Year


AT0048R - Zoebelboden (Austria)
2004
Concentration (µg/m^3)
0
2
4
6
8
10
12
PM2.5
PM10

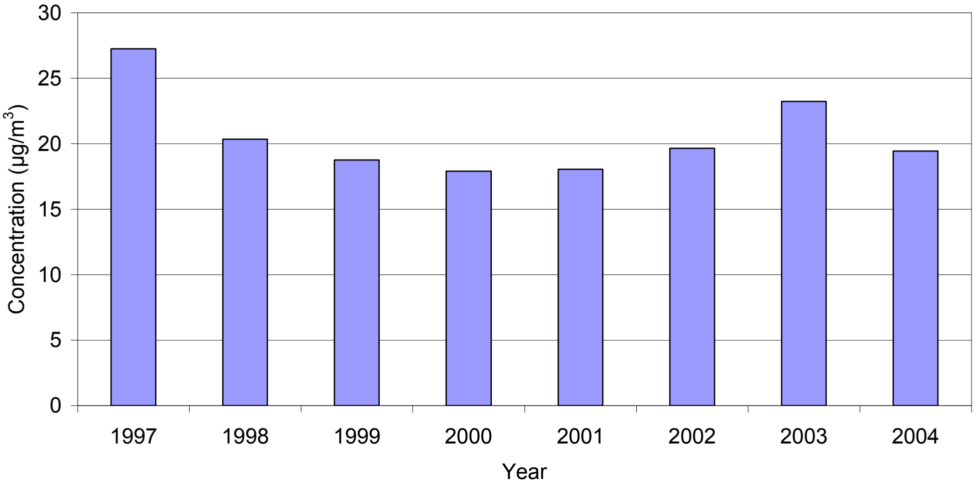
CH0002R - Payerne (Switzerland)
PM_{10}

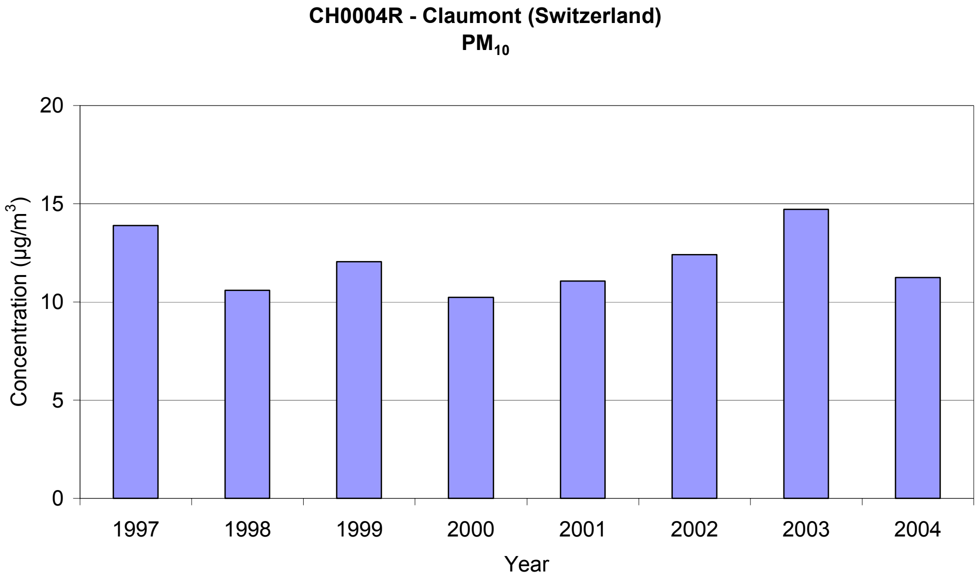


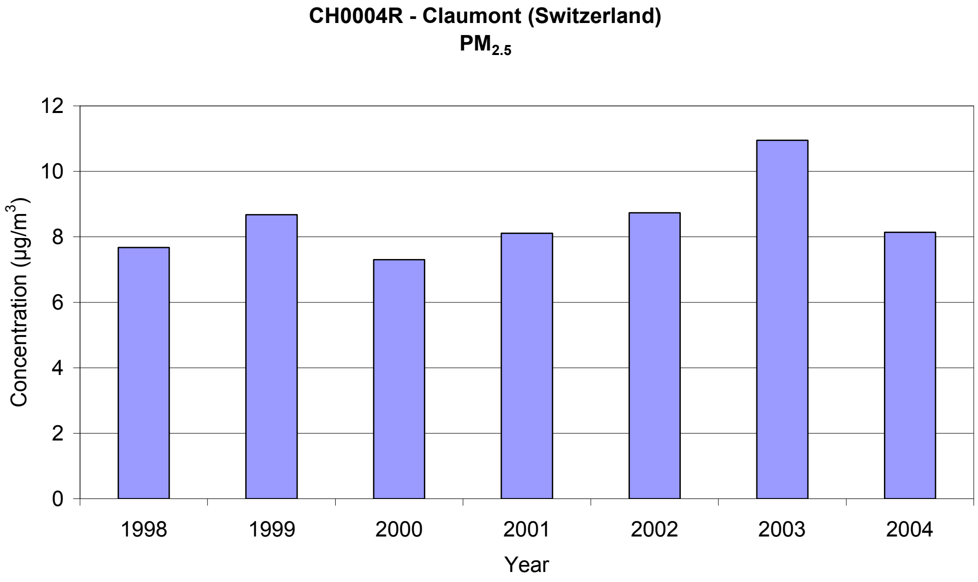
CH0002R - Payerne (Switzerland)
$PM_{2.5}$

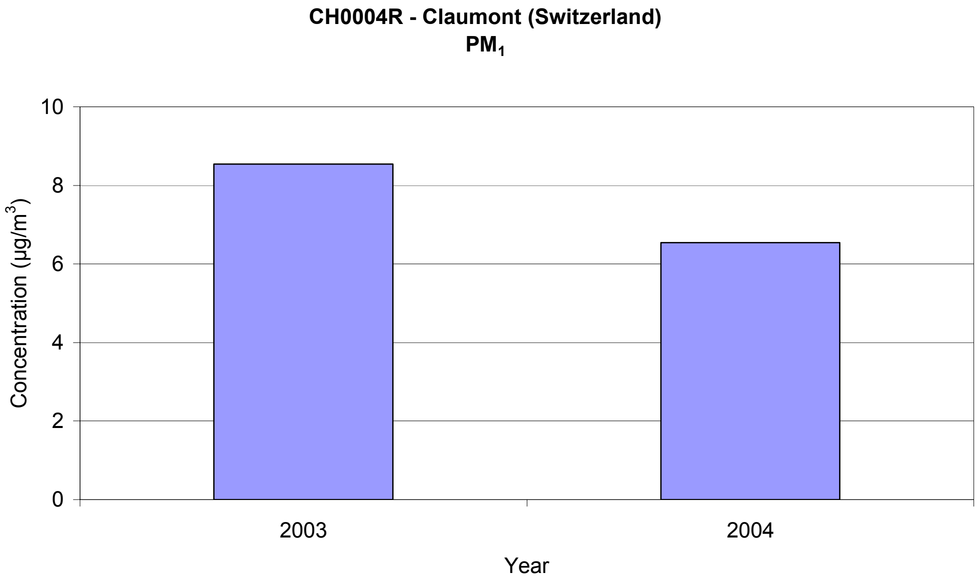


CH0003R - Tanikon (Switzerland)
PM_{10}

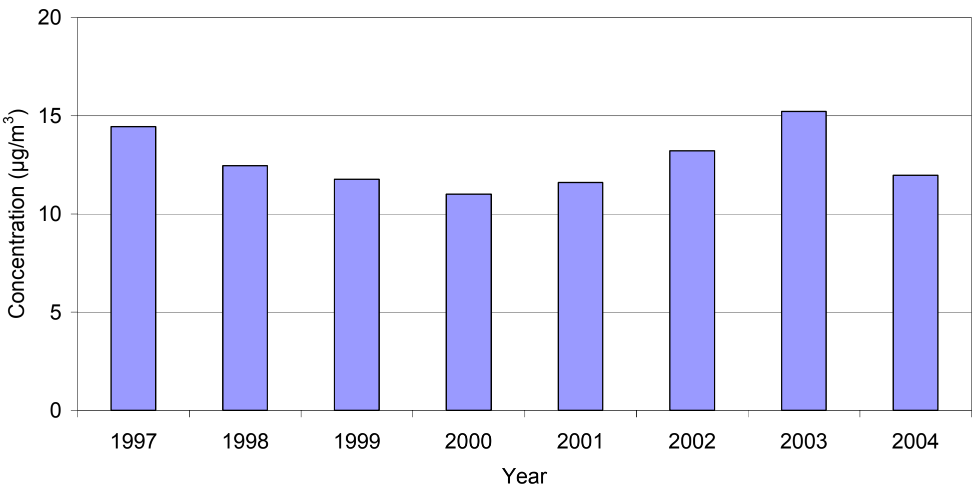



CH0004R - Claumont (Switzerland)
PM_{10}
Concentration (µg/m^3)
0
5
10
15
20
1997
1998
1999
2000
2001
2002
2003
2004
Year

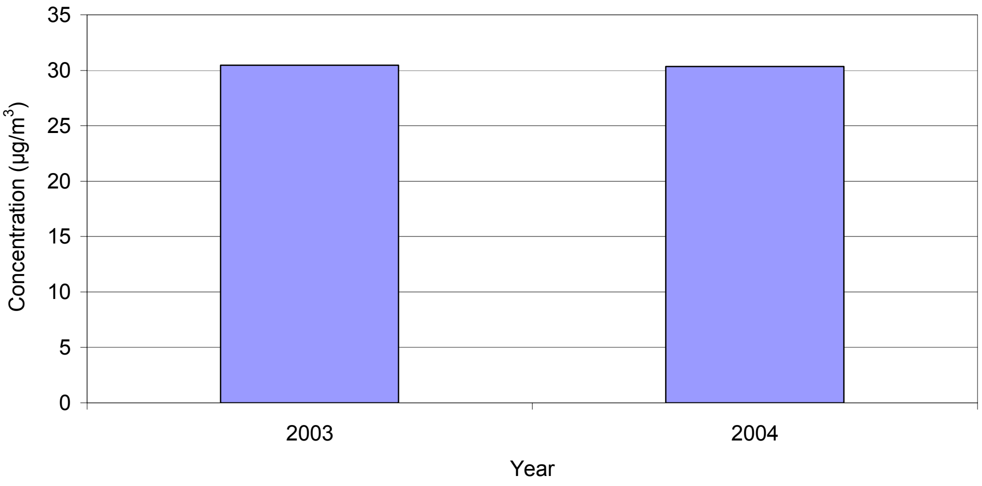

CH0004R - Claumont (Switzerland)
$PM_{2.5}$
Concentration (µg/m^3)
0
2
4
6
8
10
12
1998
1999
2000
2001
2002
2003
2004
Year


CH0004R - Claumont (Switzerland)
PM_1
Concentration (µg/m^3)
0
2
4
6
8
10
2003
2004
Year

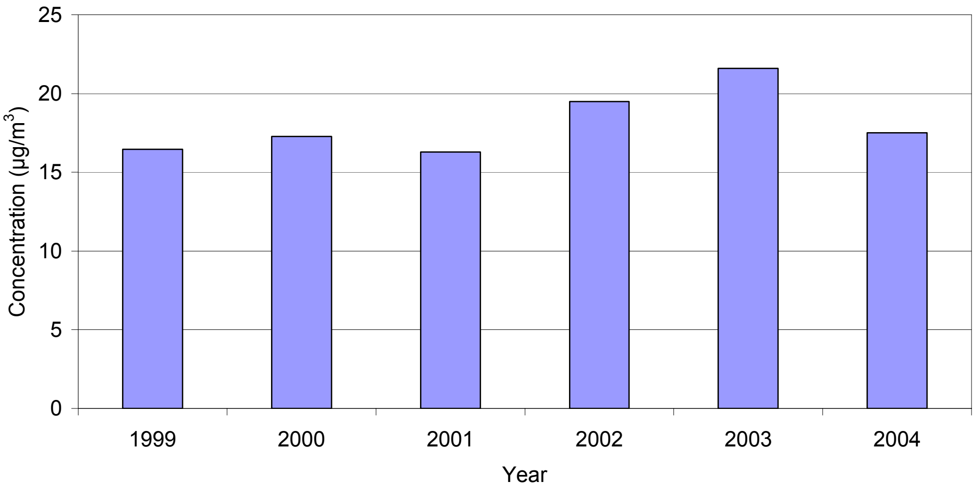
CH0005R - Rigi (Switzerland)
PM_{10}

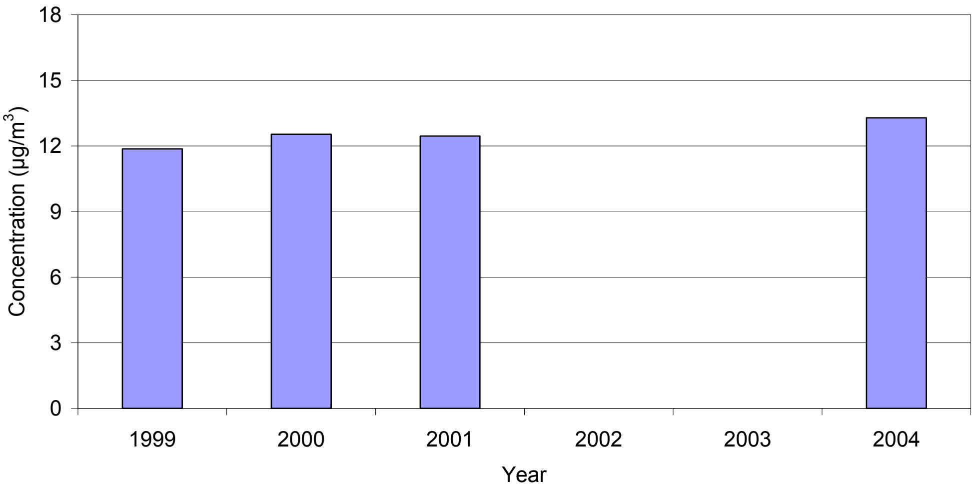


CY0002R - Ayia Marina (Cyprus)
PM_{10}

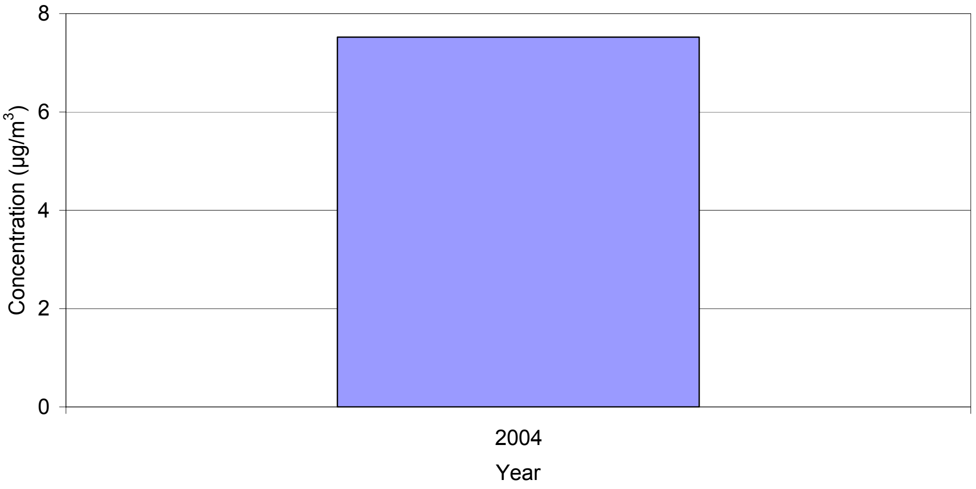


DE0002R – Langenbrügge (Germany)
PM_{10}

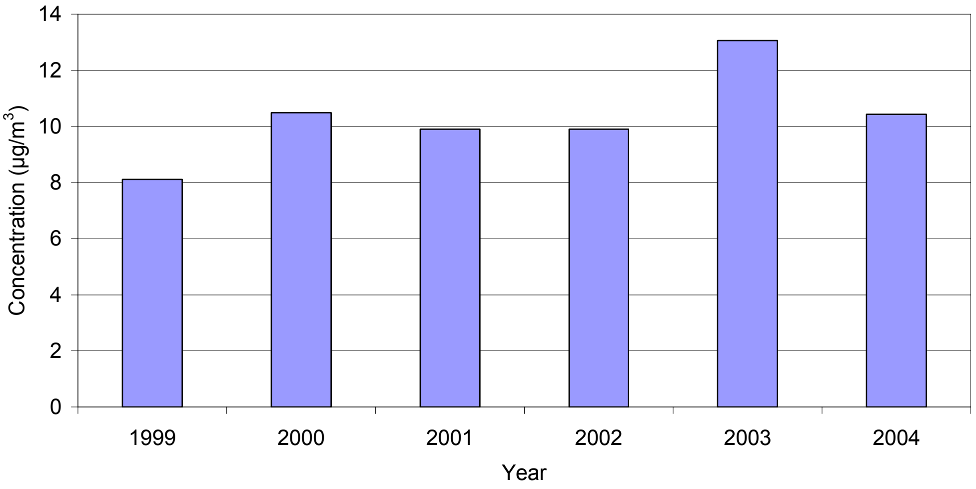




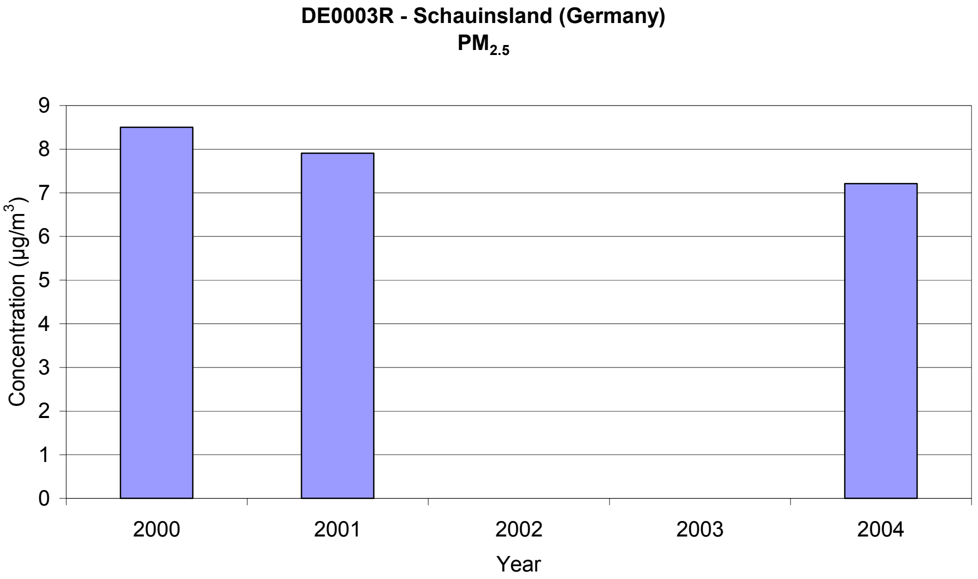
DE0002R – Langenbrügge (Germany)
PM$_{2.5}$

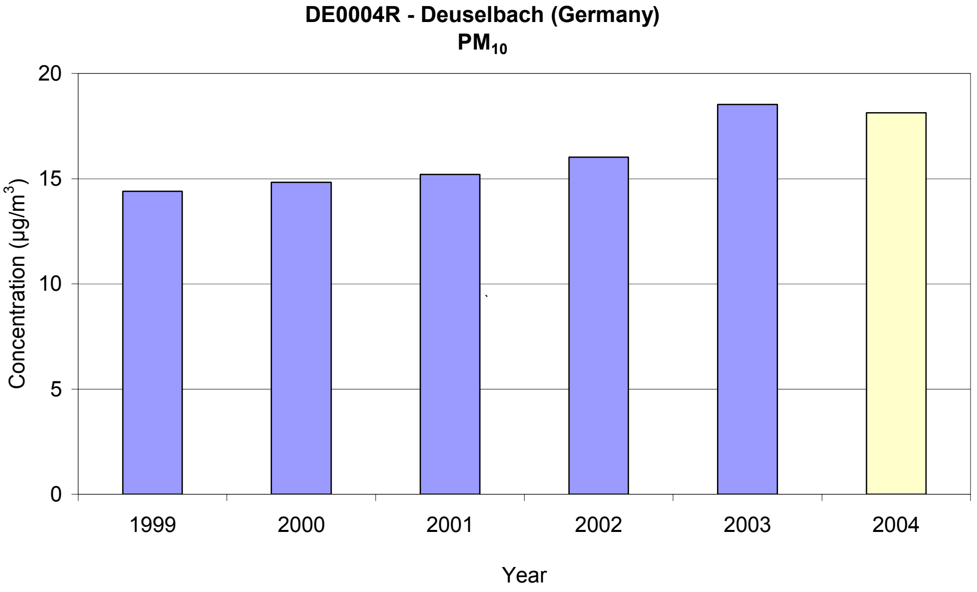


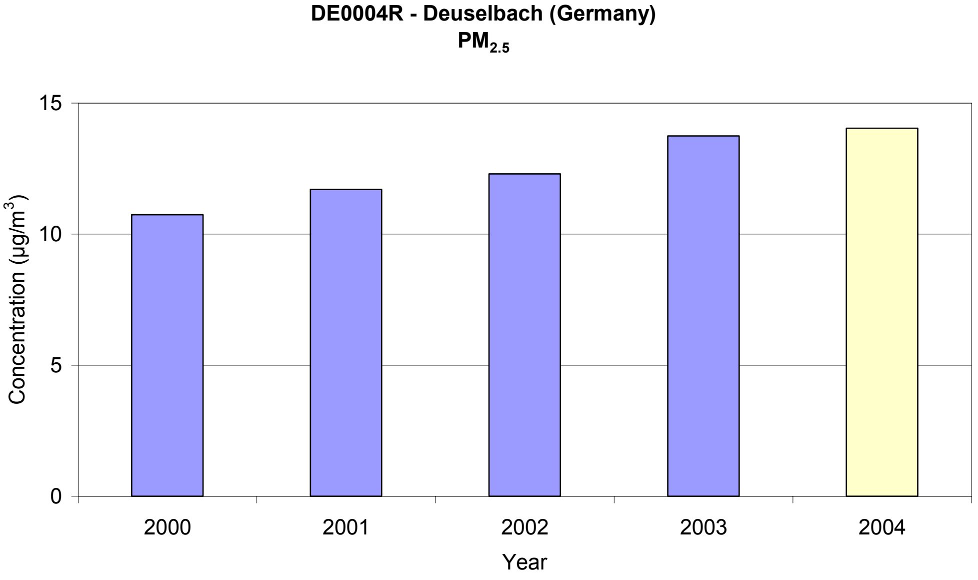
DE0002R – Langenbrügge (Germany)
PM$_1$



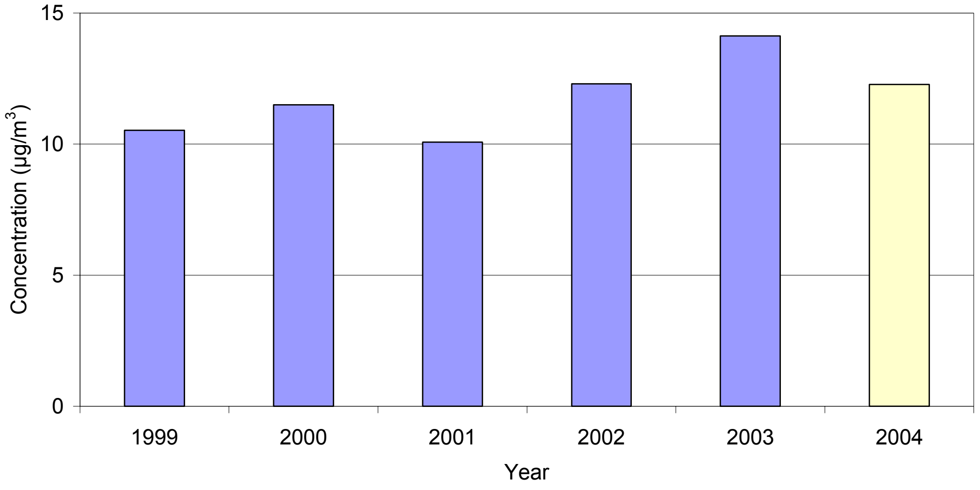
DE0003R - Schauinsland (Germany)
PM$_{10}$


DE0003R - Schauinsland (Germany)
$PM_{2.5}$
Concentration (µg/m^3)
0
1
2
3
4
5
6
7
8
9
2000
2001
2002
2003
2004
Year

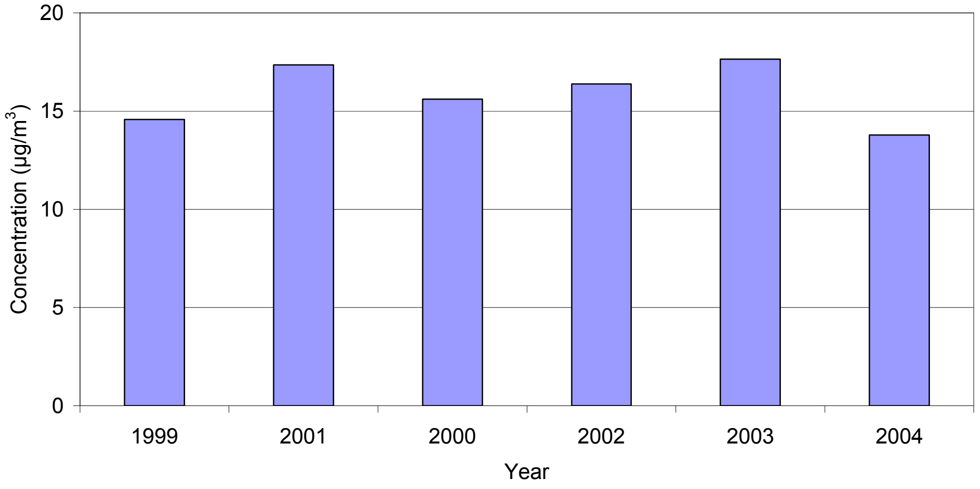

DE0004R - Deuselbach (Germany)
PM_{10}
Concentration (µg/m^3)
0
5
10
15
20
1999
2000
2001
2002
2003
2004
Year


DE0004R - Deuselbach (Germany)
$PM_{2.5}$
Concentration (µg/m^3)
0
5
10
15
2000
2001
2002
2003
2004
Year

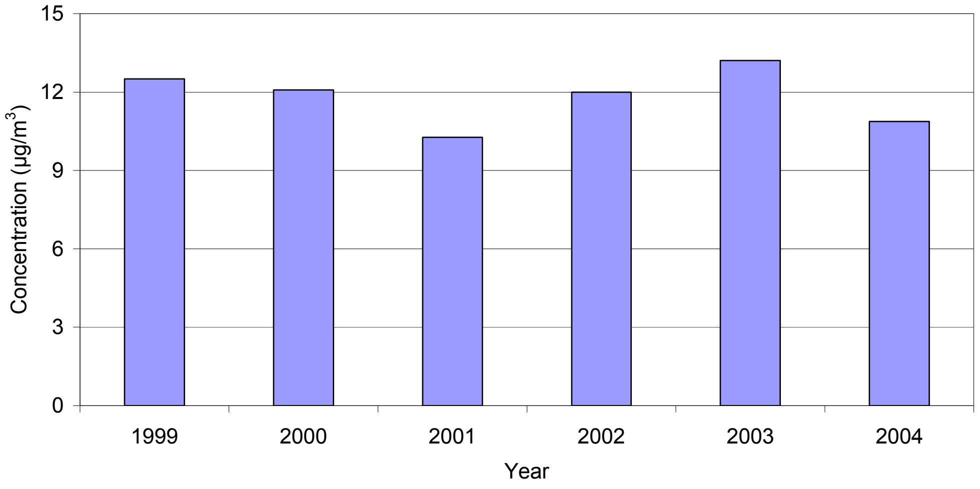
DE0005R - Brotjacklriegel (Germany)
PM_{10}



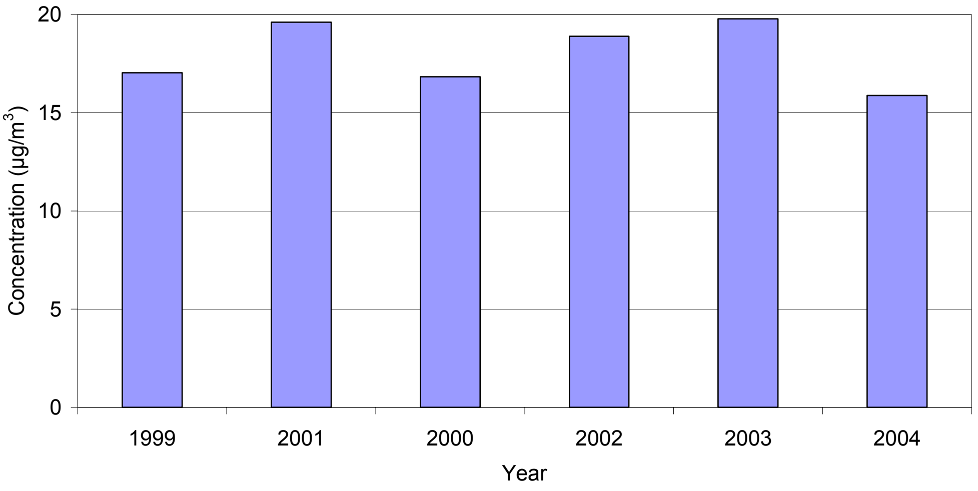
DE0007R - Neuglobsow (Germany)
PM_{10}



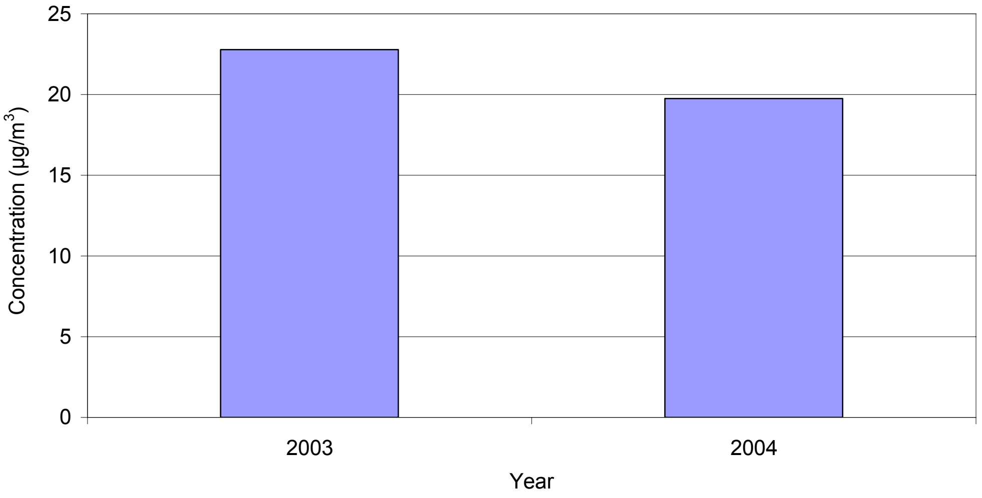
DE0008R - Schmuche (Germany)
PM_{10}



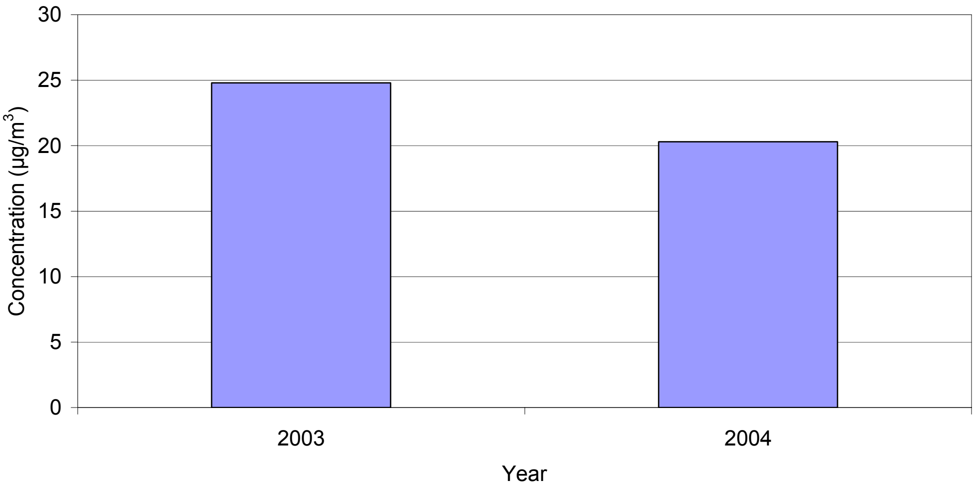
DE0009R - Zingst (Germany)
PM_{10}

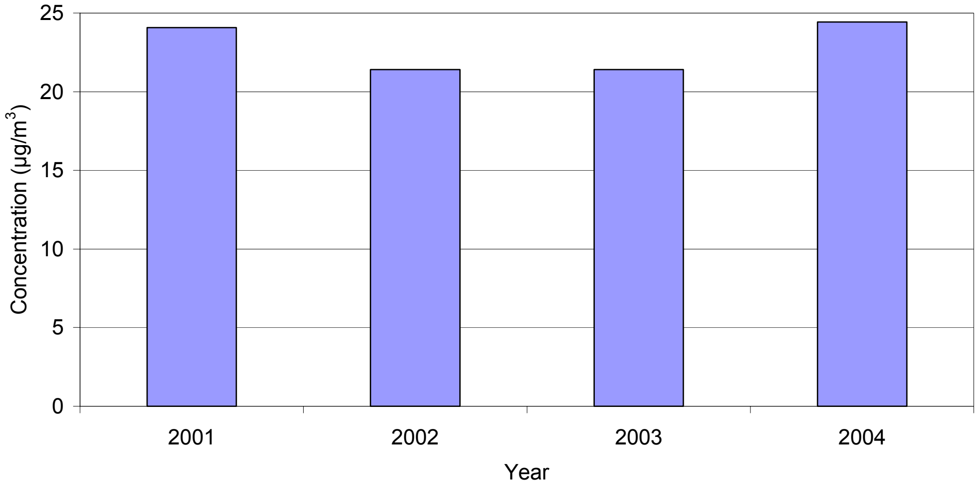


DE0041R - Tinnum (Germany)
PM_{10}

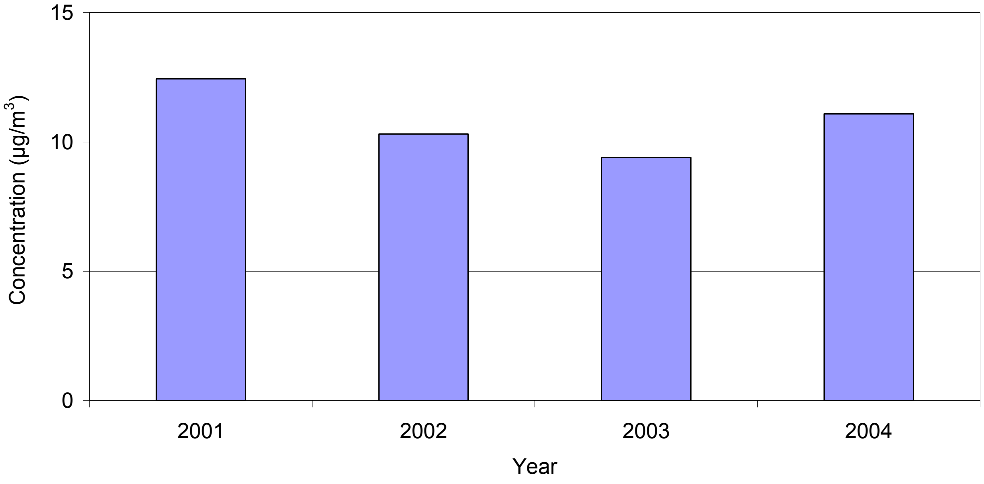


DK0005R - Keldsnor (Denmark)
PM_{10}

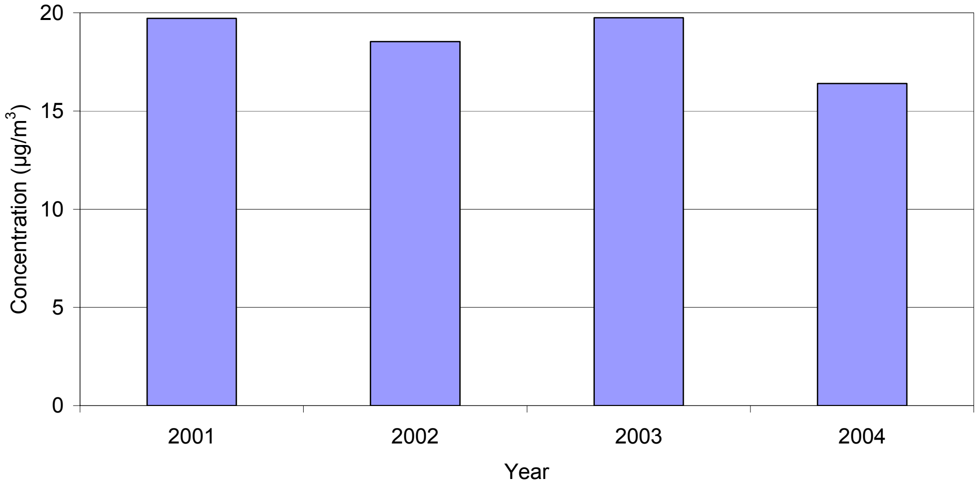




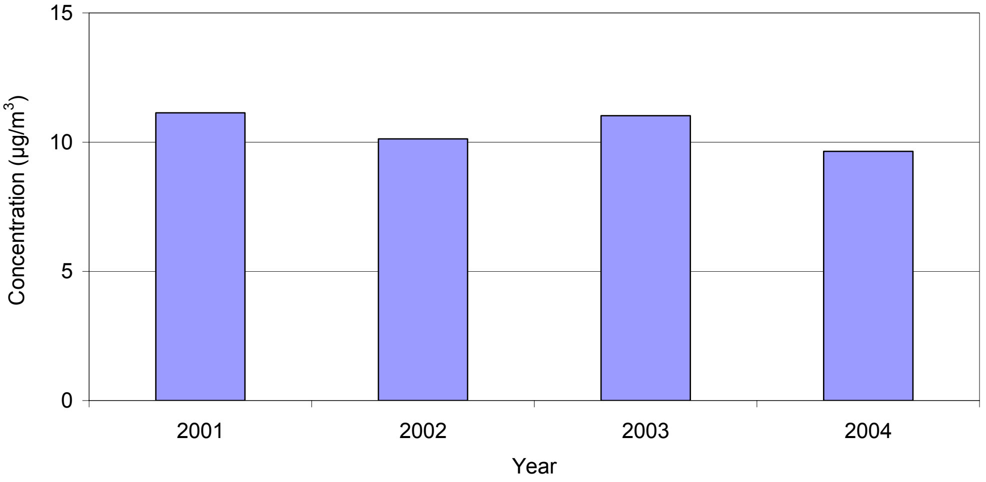
ES0007R - Vyznar (Spain)
PM_{10}



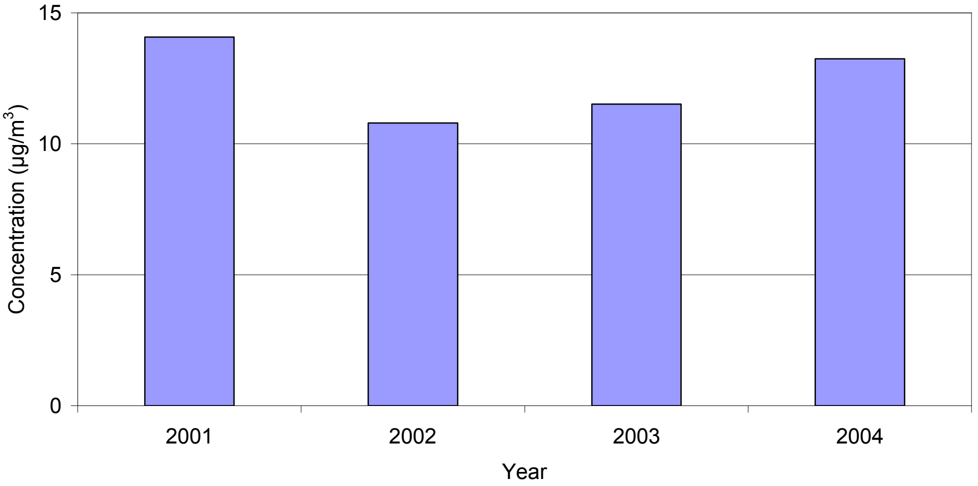
ES0007R - Vyznar (Spain)
$PM_{2.5}$



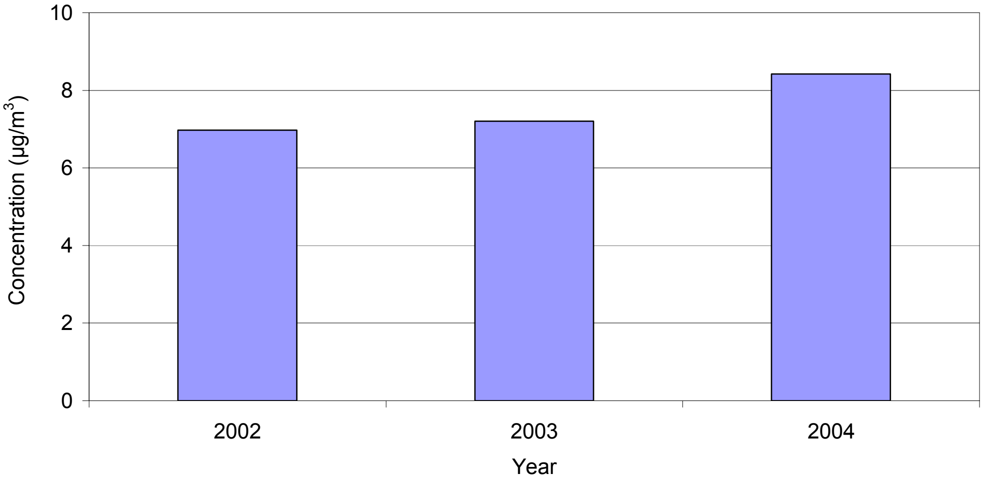
ES0008R - Niembro (Spain)
PM_{10}



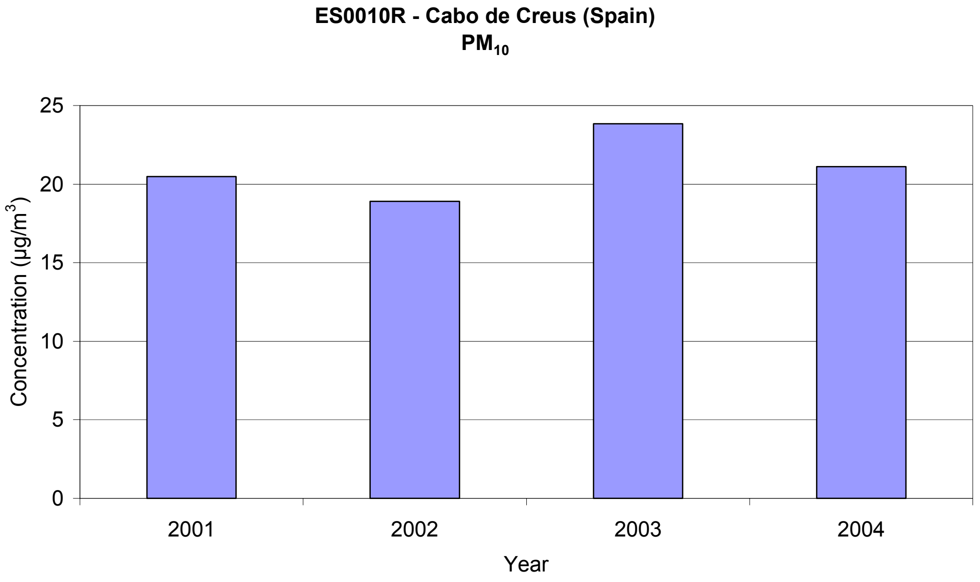
ES0008R - Niembro (Spain)
$PM_{2.5}$

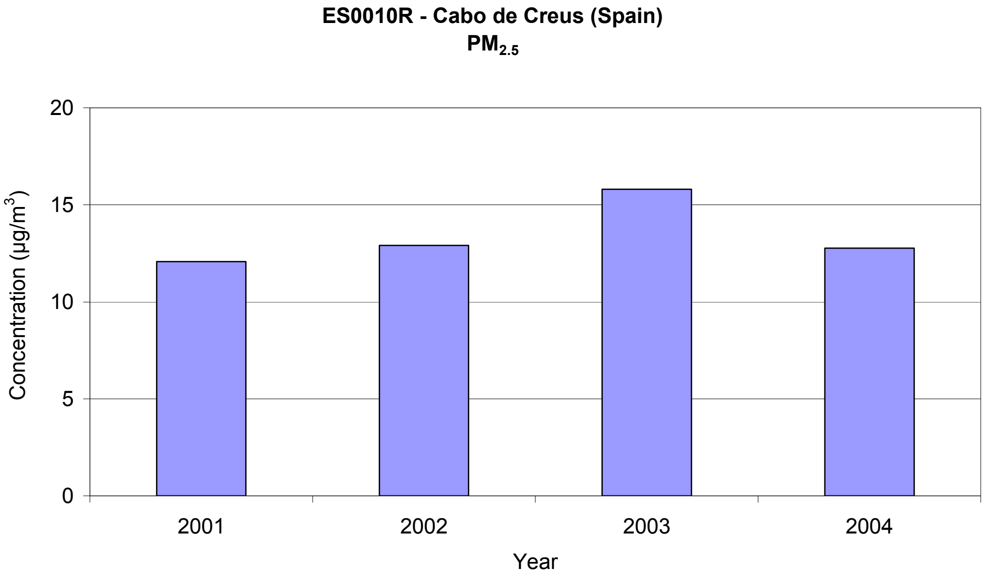


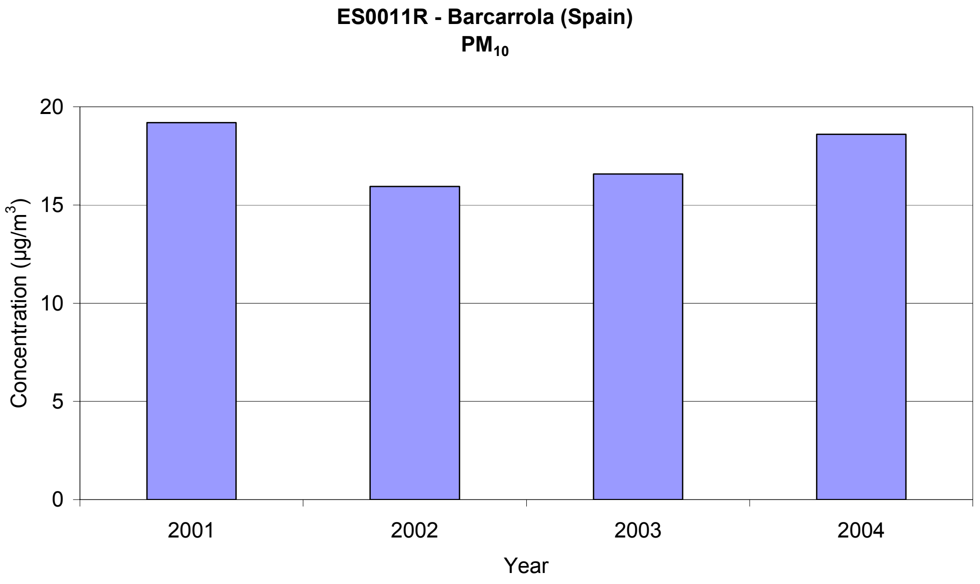
ES0009R - Campisabalos (Spain)
PM_{10}



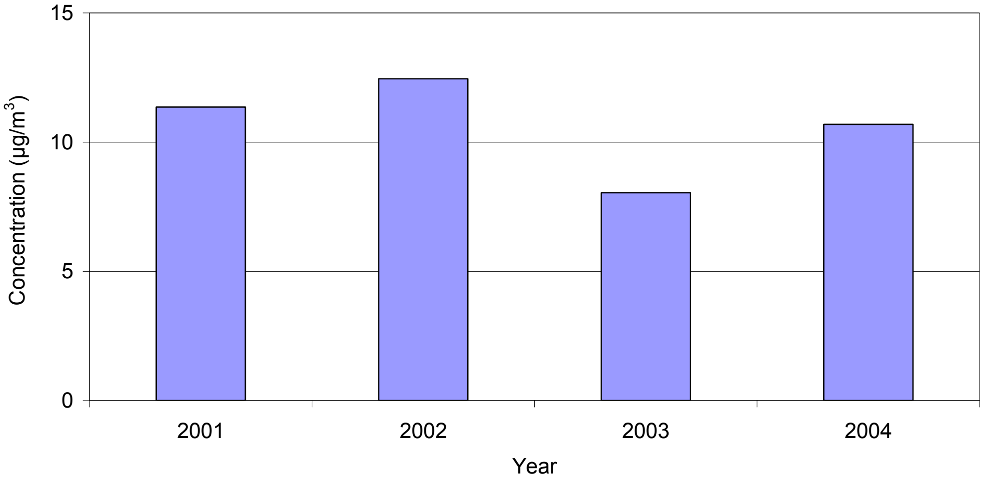
ES0009R - Campisabalos (Spain)
$PM_{2.5}$


ES0010R - Cabo de Creus (Spain)
PM_{10}
Concentration (µg/m^3)
0
5
10
15
20
25
2001
2002
2003
2004
Year

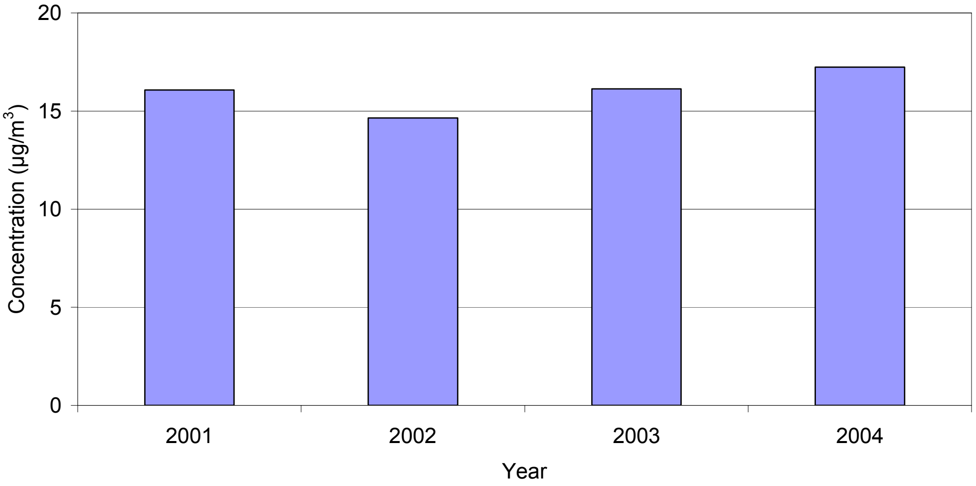

ES0010R - Cabo de Creus (Spain)
$PM_{2.5}$
Concentration (µg/m^3)
0
5
10
15
20
2001
2002
2003
2004
Year


ES0011R - Barcarrola (Spain)
PM_{10}
Concentration (µg/m^3)
0
5
10
15
20
2001
2002
2003
2004
Year

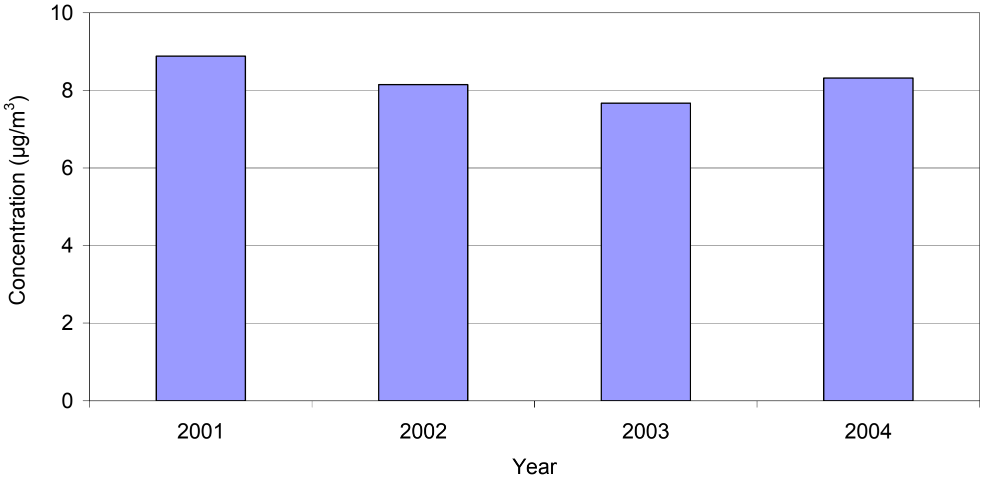
ES0011R - Barcarrola (Spain)
PM$_{2.5}$



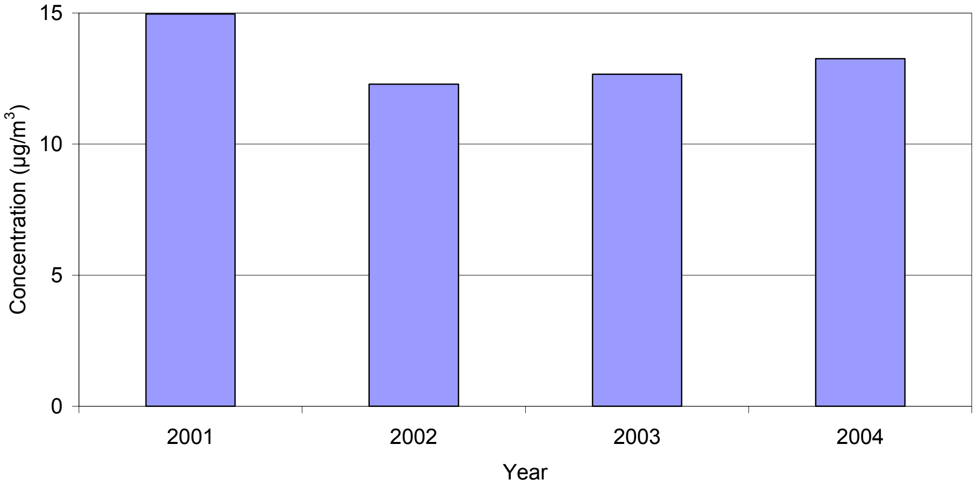
ES0012R - Zarra (Spain)
PM$_{10}$



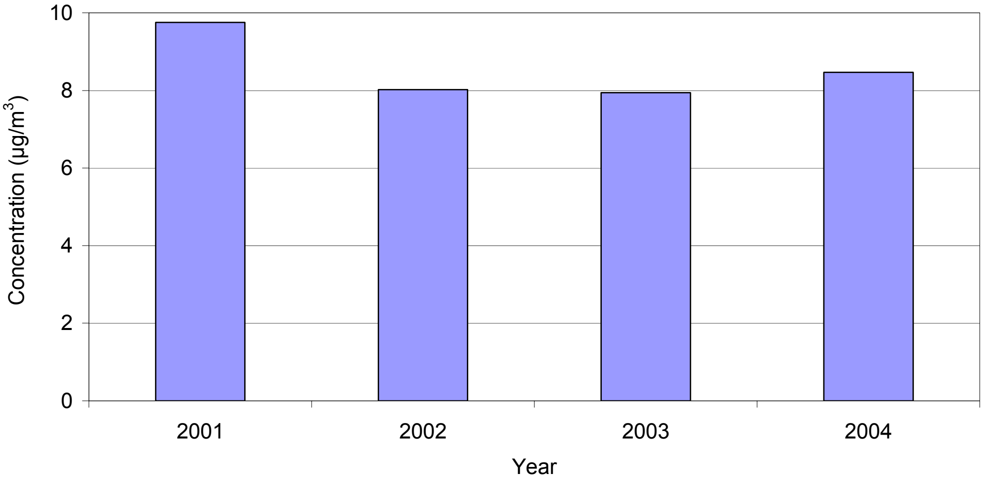
ES0012R - Zarra (Spain)
PM$_{2.5}$



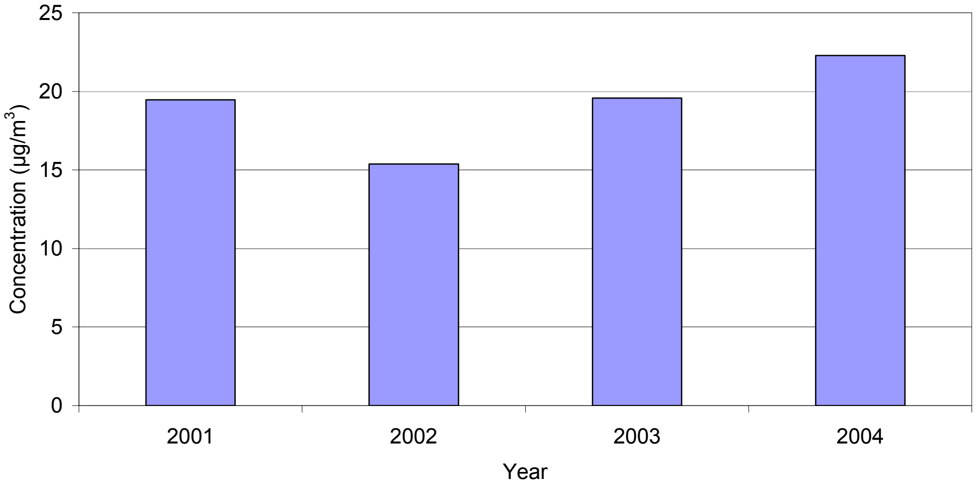
ES0013R - Penausende (Spain)
PM_{10}



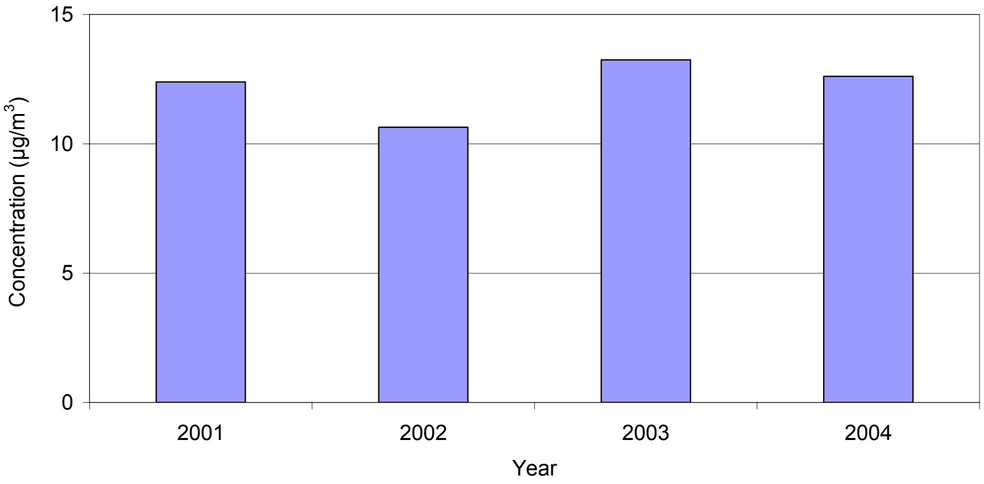
ES0013R - Penausende (Spain)
$PM_{2.5}$



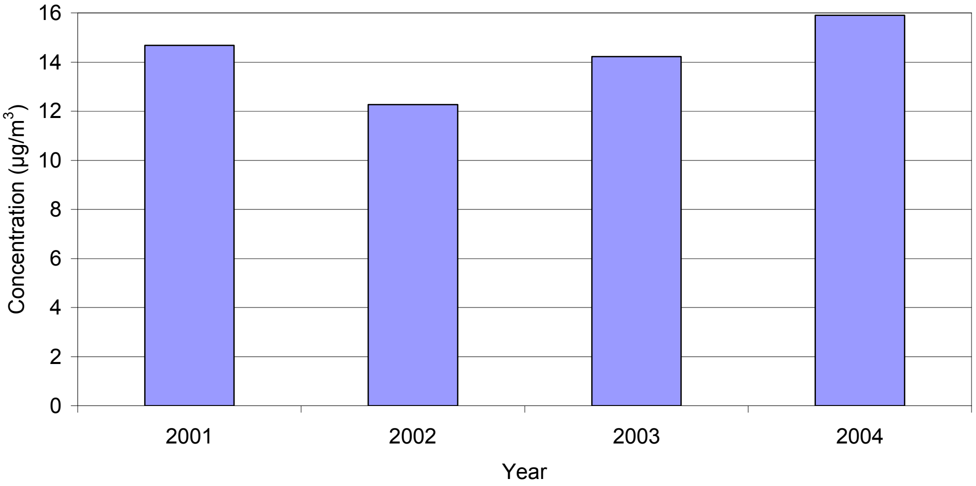
ES0014R - Els Torms (Spain)
PM_{10}



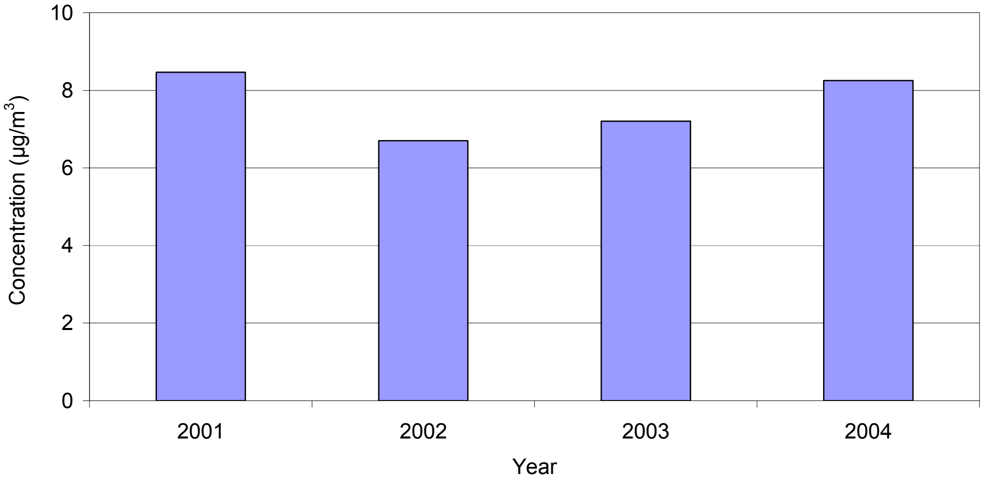
ES0014R - Els Torms (Spain)
$PM_{2.5}$



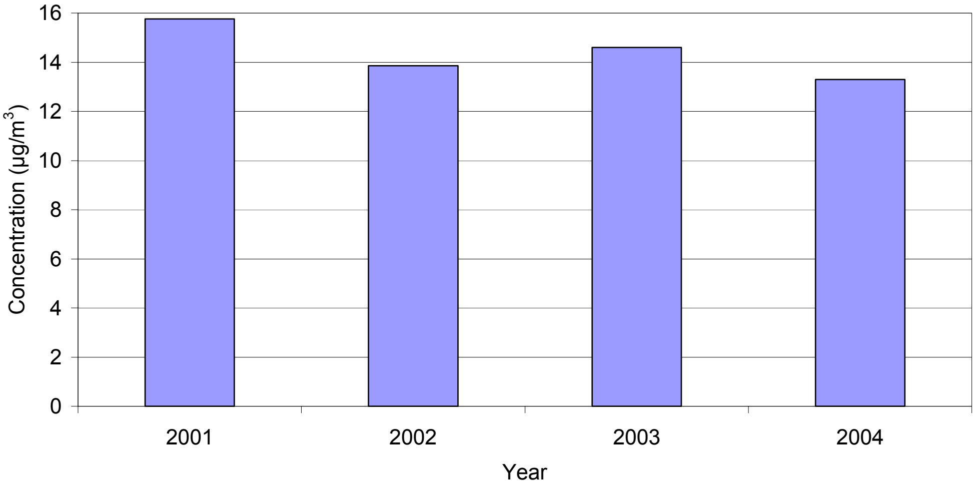
ES0015R - Risco Llamo (Spain)
PM_{10}



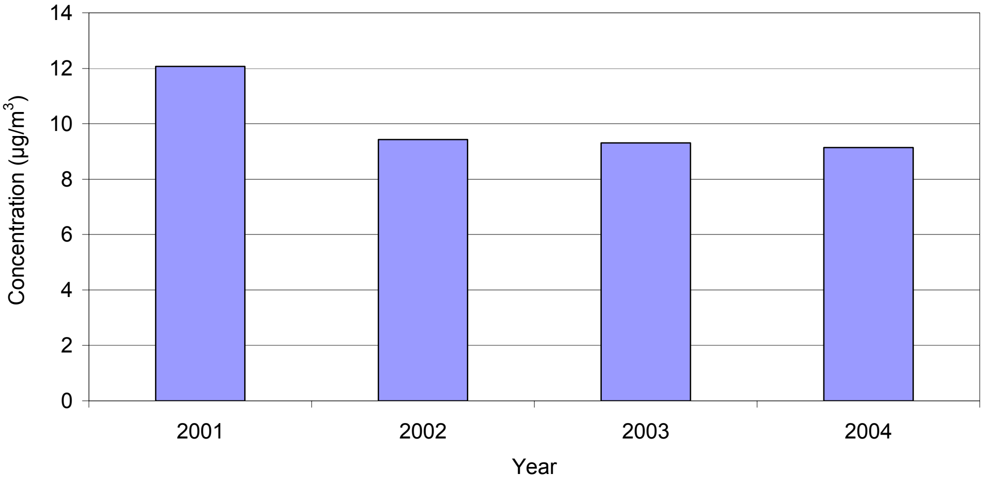
ES0015R - Risco Llamo (Spain)
$PM_{2.5}$



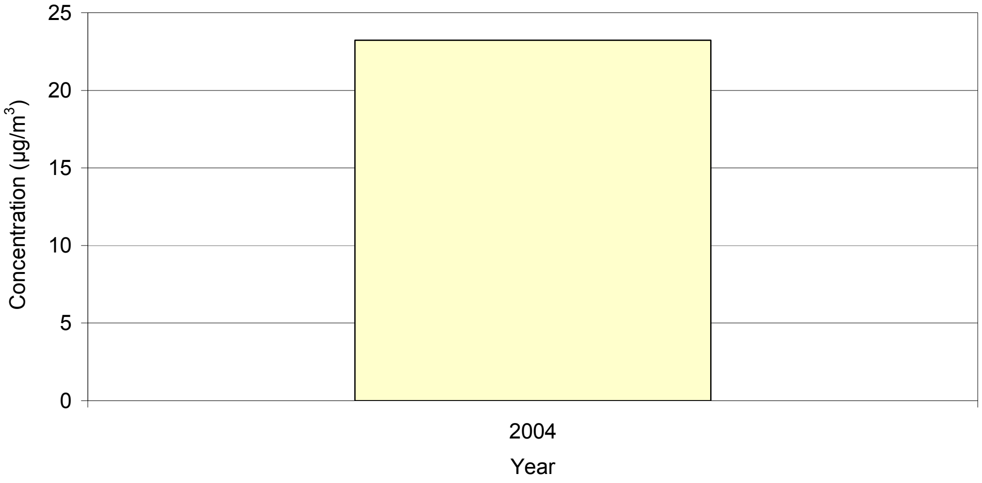
ES0016R - O Savipao (Spain)
PM_{10}

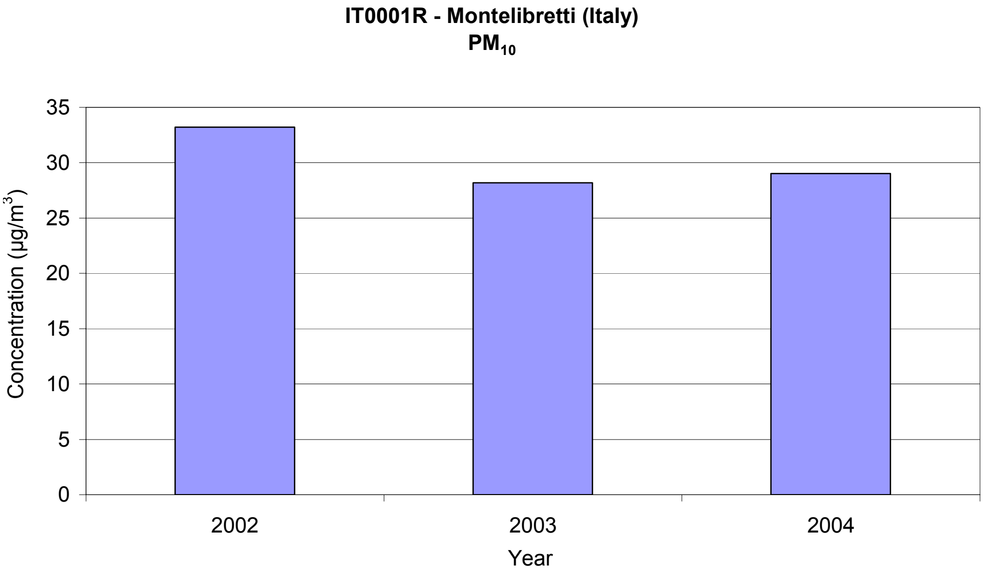


ES0016R - O Savipao (Spain)
$PM_{2.5}$

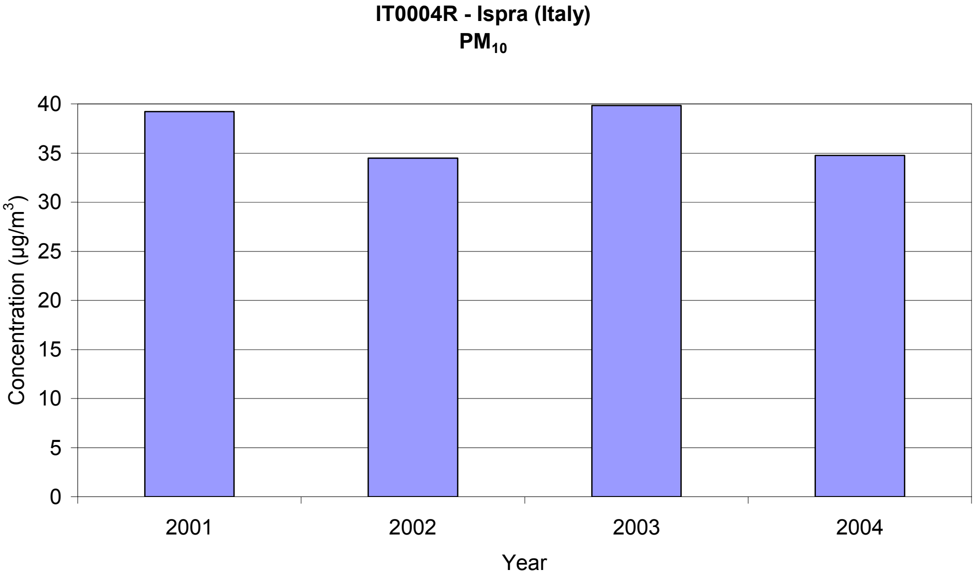


GR0002R - Finokalia (Greece)
PM_{10}

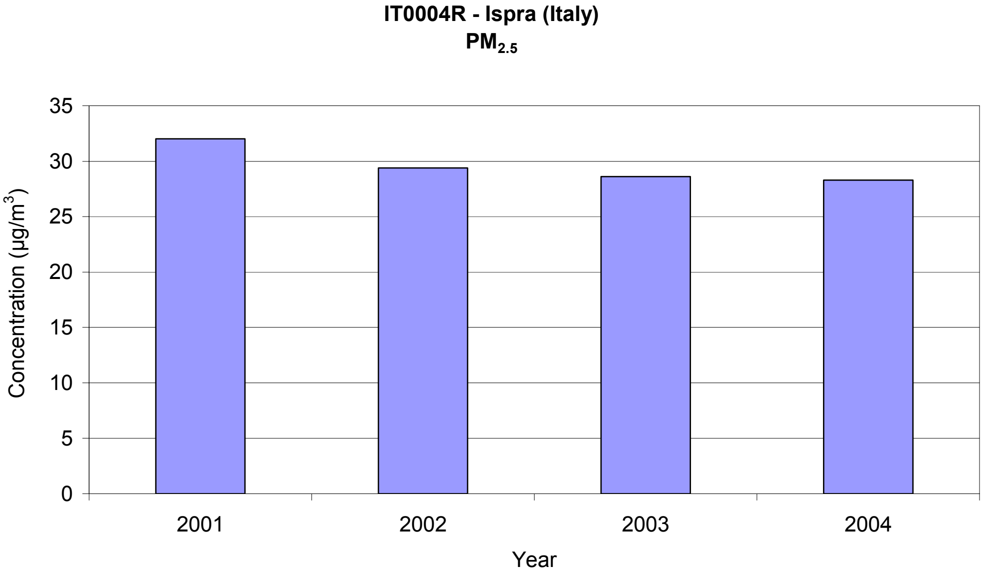




IT0001R - Montelibretti (Italy)
PM_{10}

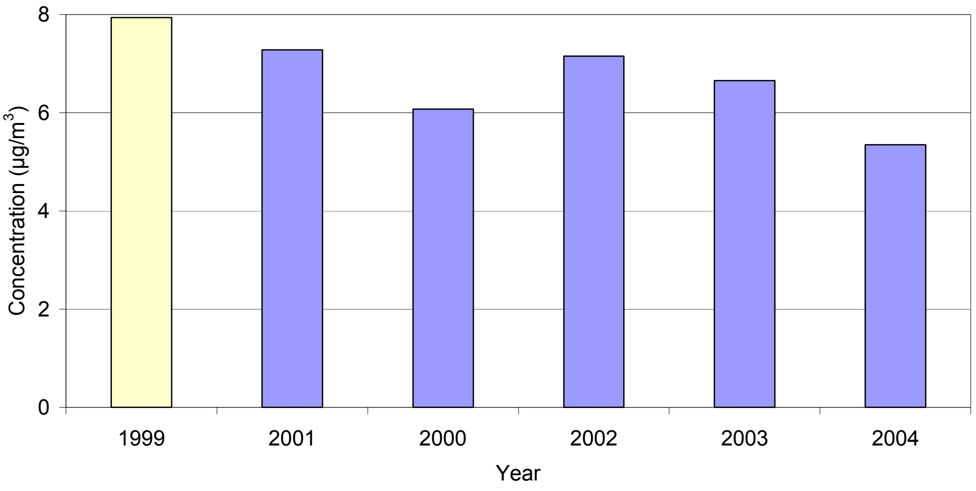


IT0004R - Ispra (Italy)
PM_{10}

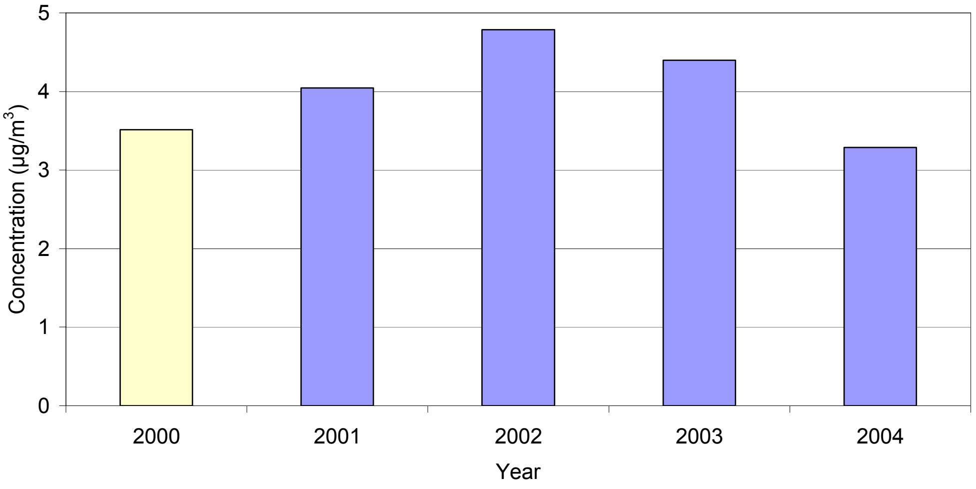


IT0004R - Ispra (Italy)
$PM_{2.5}$

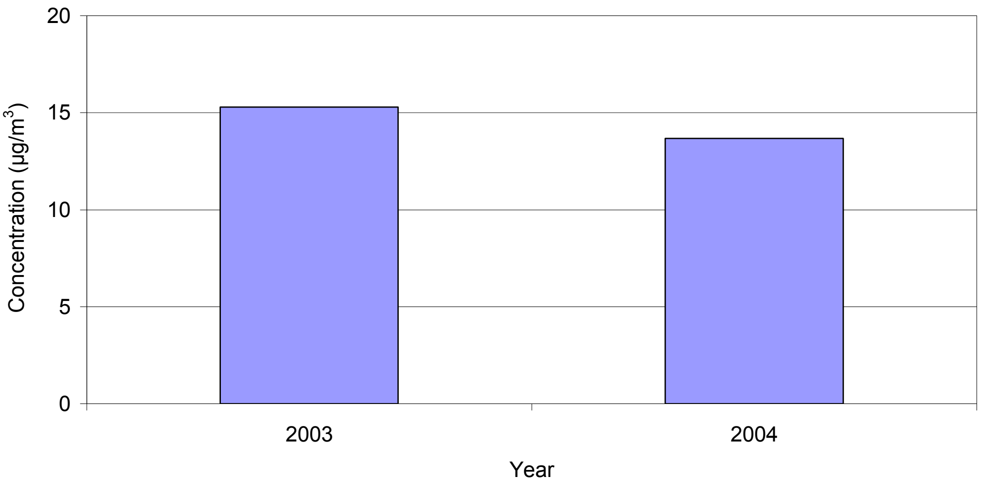
NO0001R - Birkenes (Norway)
PM$_{10}$



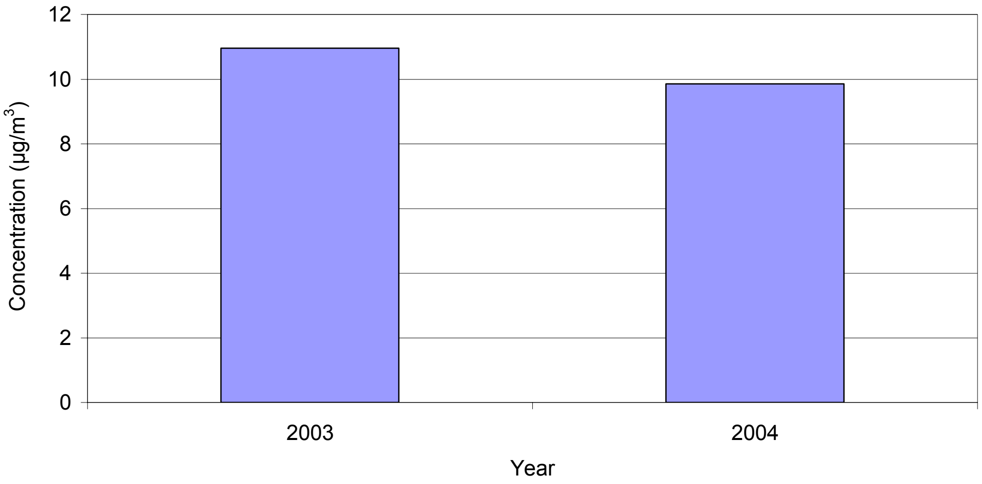
NO0001R - Birkenes (Norway)
PM$_{2.5}$



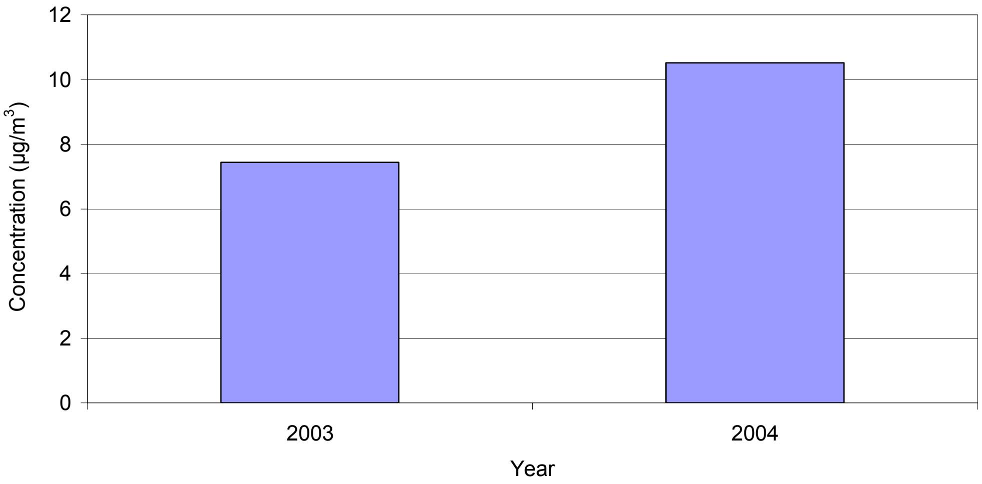
SE0011R - Vavihill (Sweden)
PM$_{10}$



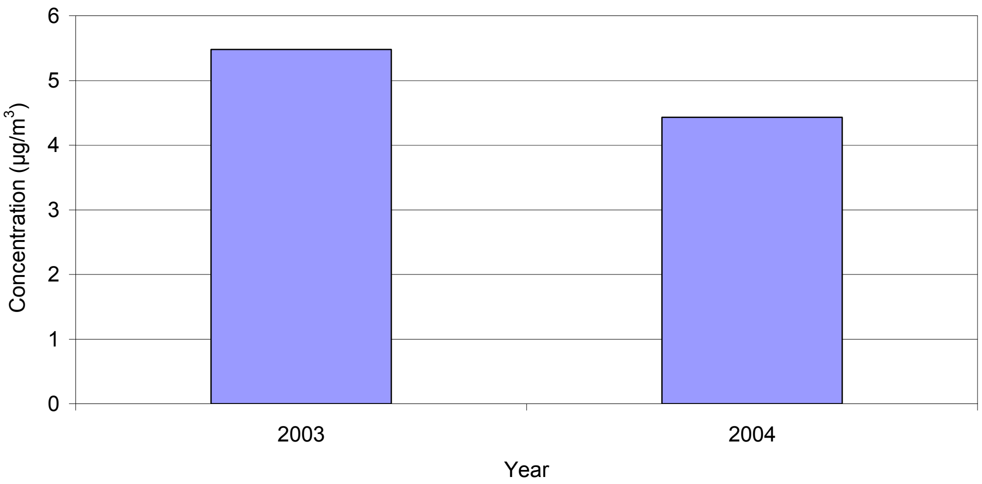
SE0011R - Vavihill (Sweden)
PM$_{2.5}$

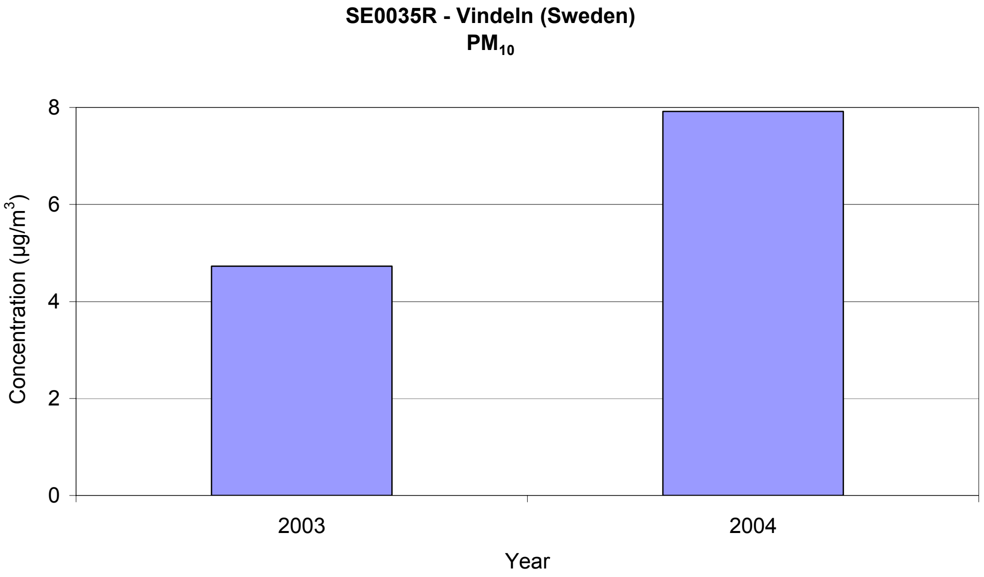


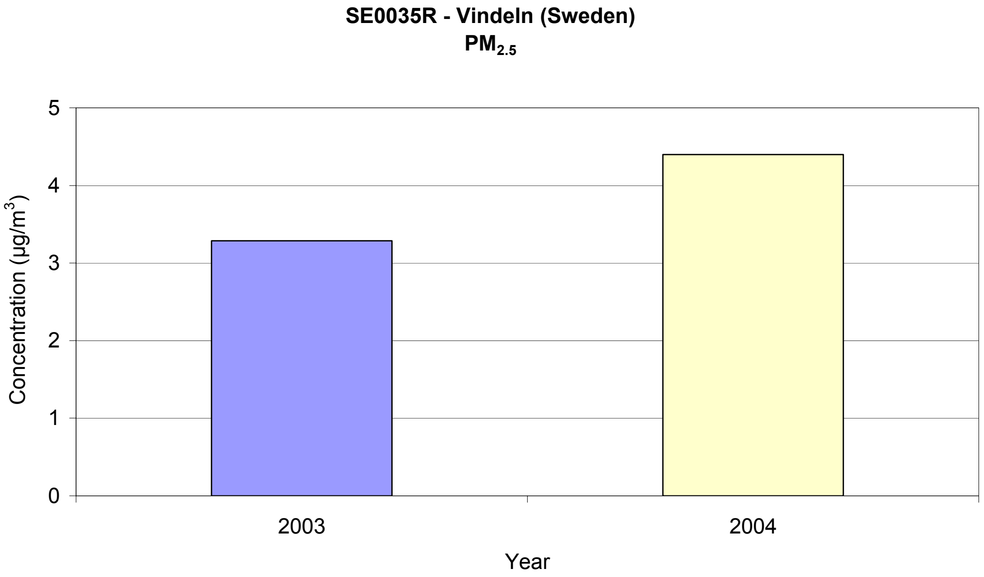
SE0012R - Aspvreten (Sweden)
PM$_{10}$

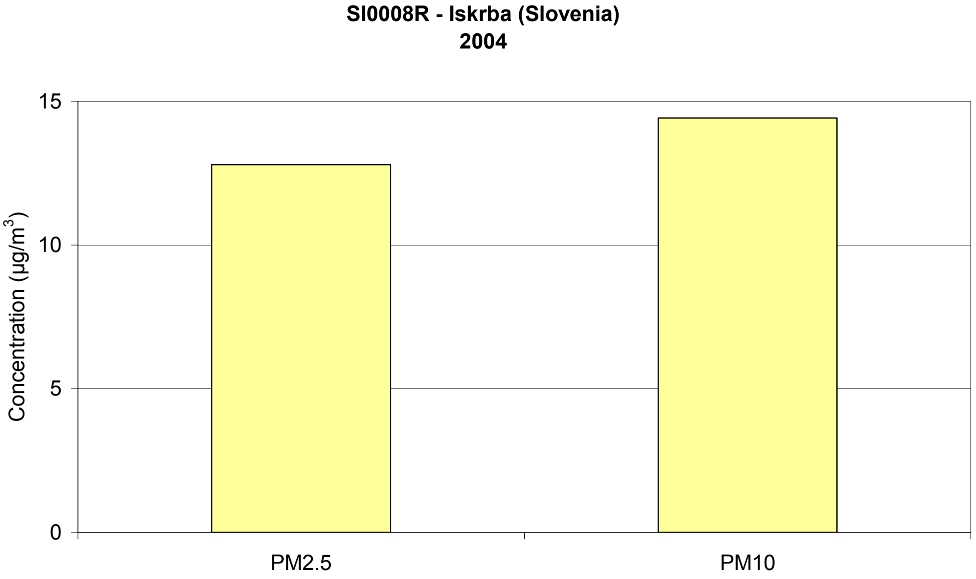


SE0012R - Aspvreten (Sweden)
PM$_{2.5}$

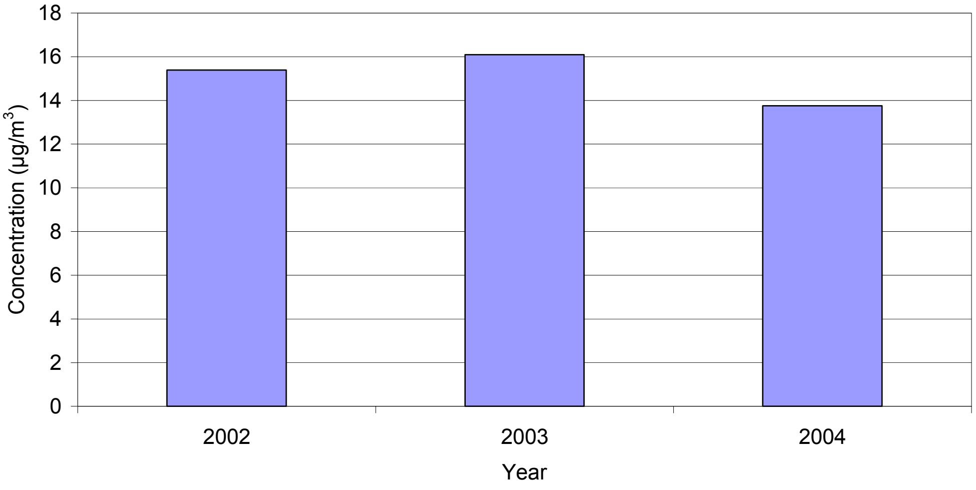



SE0035R - Vindeln (Sweden)
PM10
Concentration (µg/m3)
8
6
4
2
0
2003
2004
Year

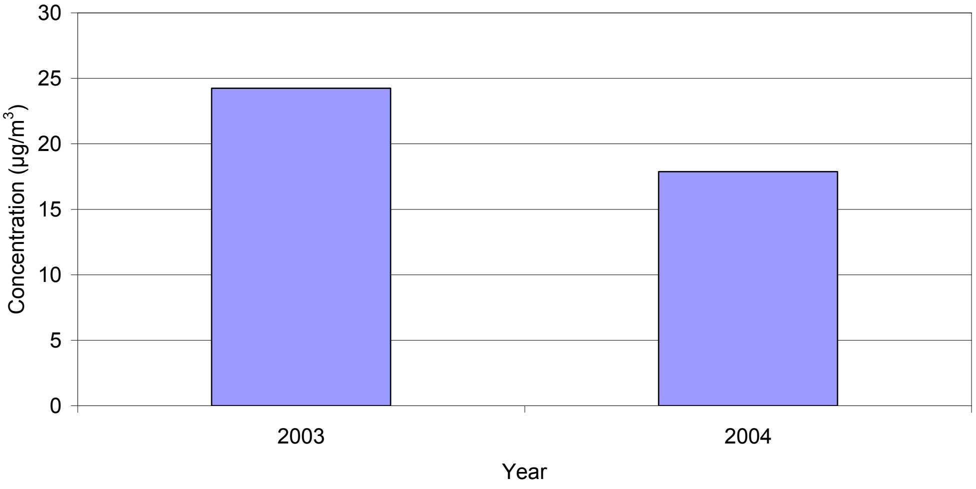

SE0035R - Vindeln (Sweden)
PM2.5
Concentration (µg/m3)
5
4
3
2
1
0
2003
2004
Year


SI0008R - Iskrba (Slovenia)
2004
Concentration (µg/m3)
15
10
5
0
PM2.5
PM10

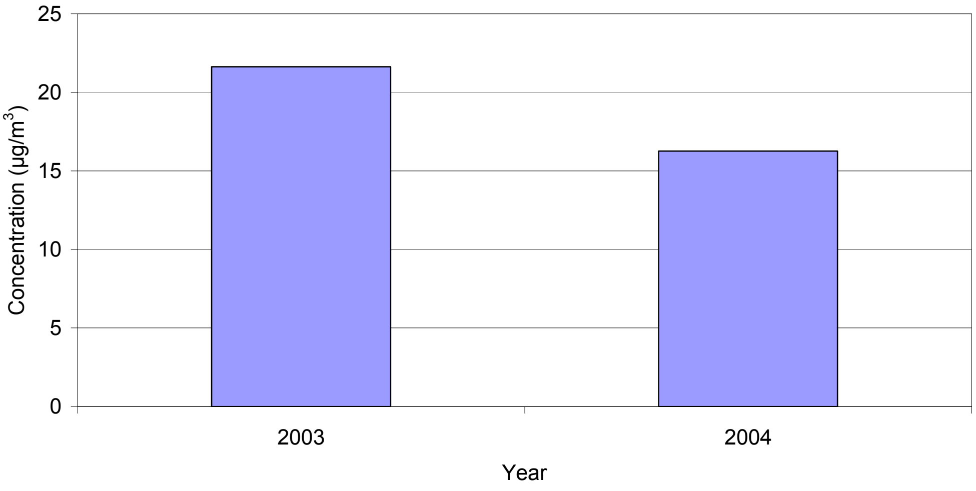
SK0004R - Stara Lesna (Slovakia)
PM_{10}



SK0005R - Liesek (Slovakia)
PM_{10}

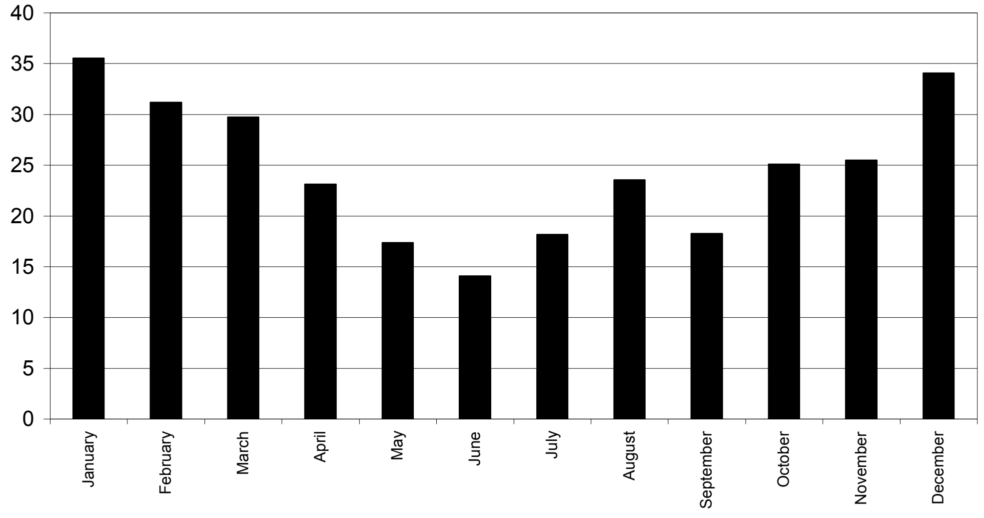


SK0006R - Starina (Slovakia)
PM_{10}

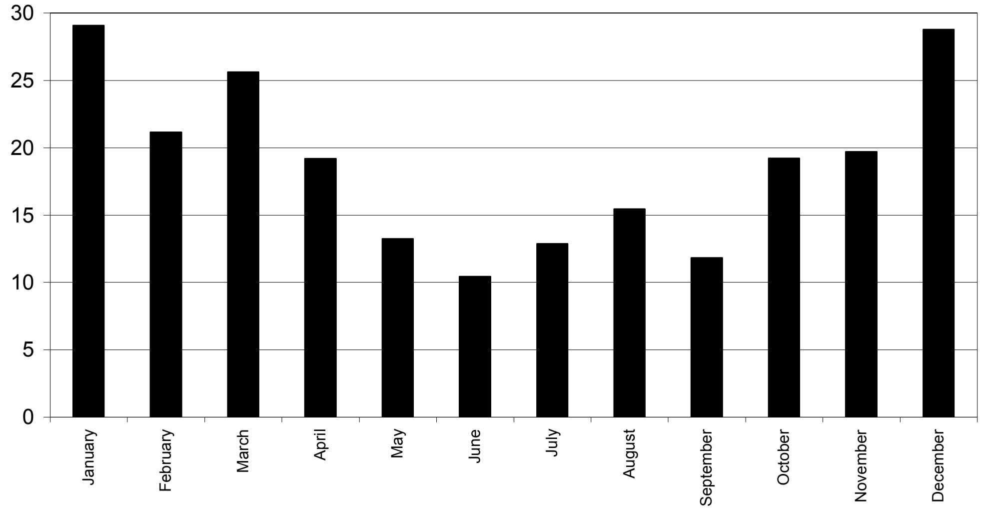


Annex 3

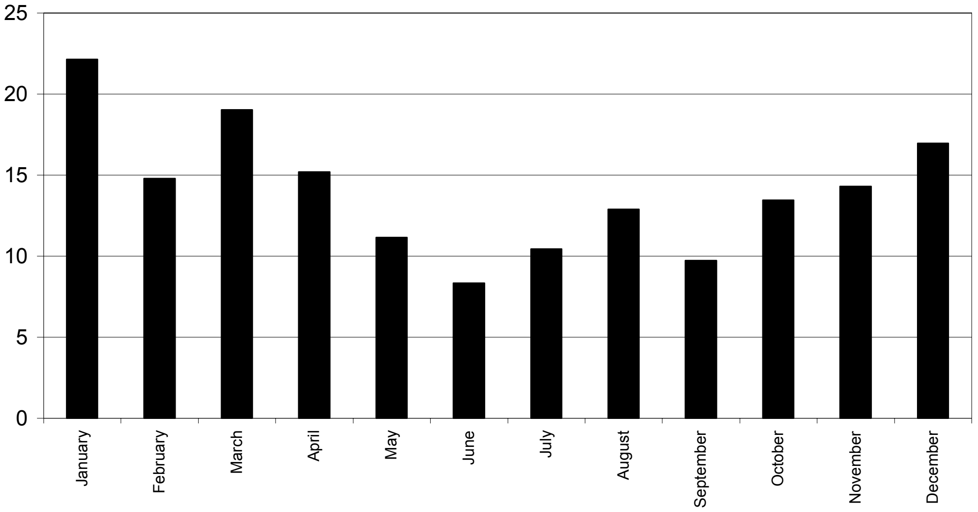
Trends in monthly mean particulate mass concentrations at EMEP stations in 2004



AT0002R 2004 PM$_{10}$ (µg/m^3)

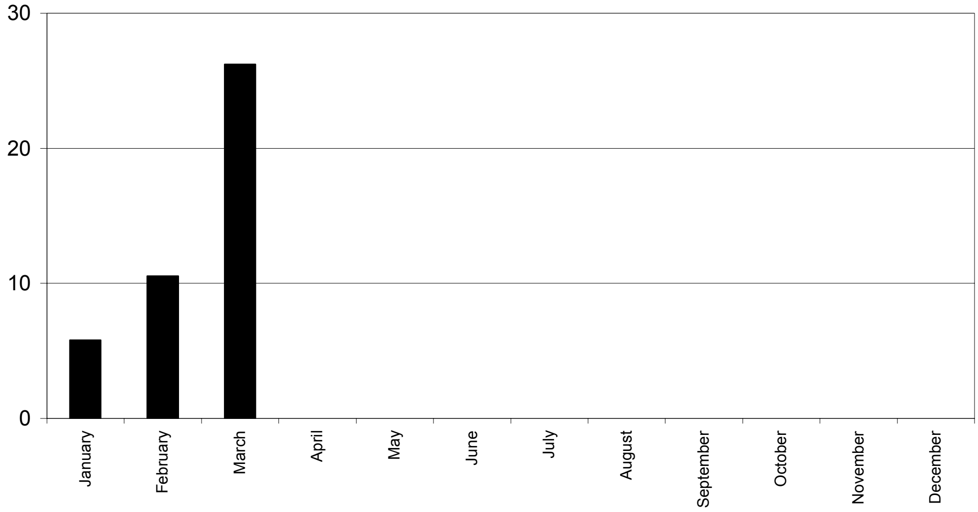


AT0002R 2004 PM$_{2.5}$ (µg/m^3)

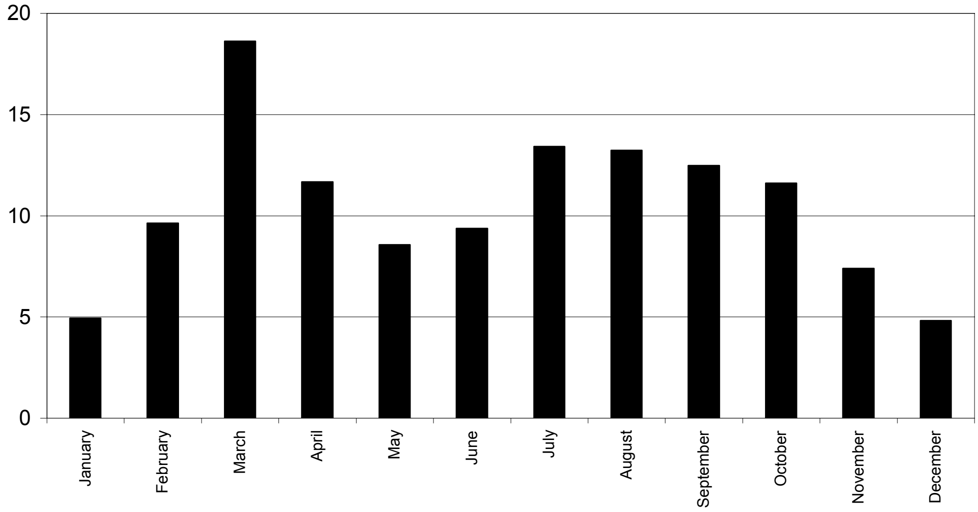


AT0002R 2004 PM$_1$ (µg/m^3)

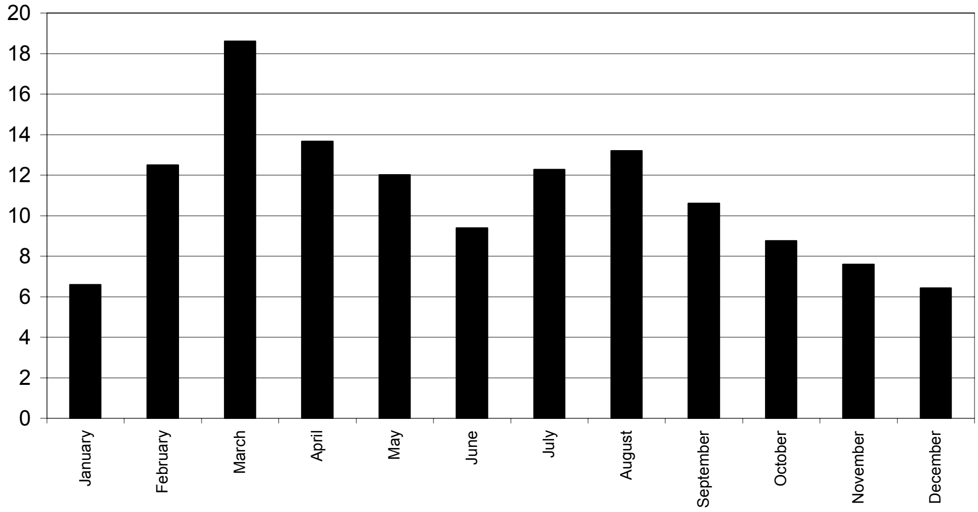
AT0004R 2004 PM_{10} ($\mu g/m^3$)

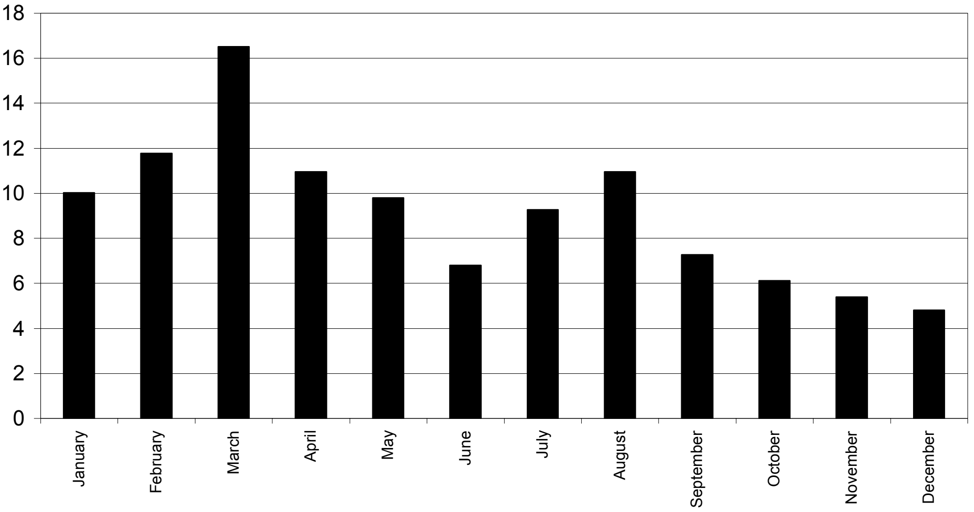


AT0005R 2004 PM_{10} ($\mu g/m^3$)

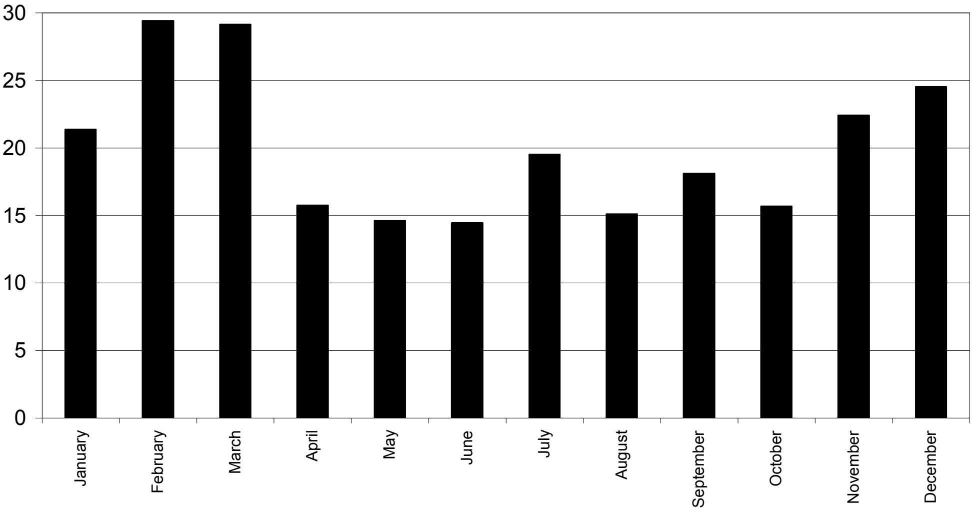


AT0048R 2004 PM_{10} ($\mu g/m^3$)

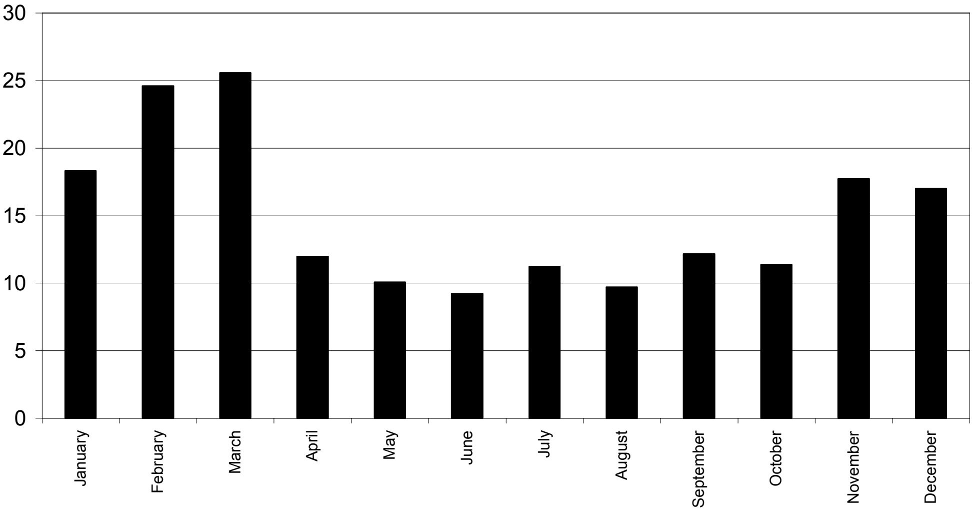




AT0048R 2004 $PM_{2.5}$ (µg/m^3)

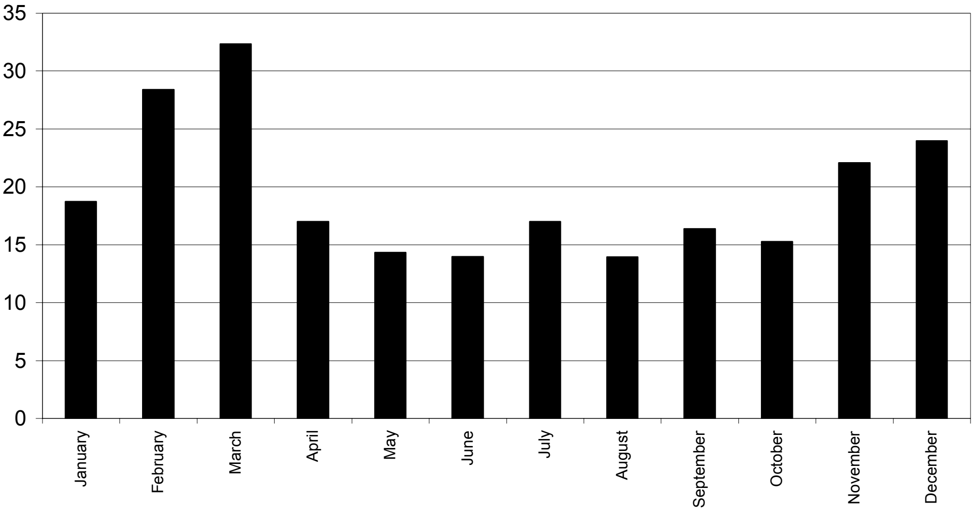


CH0002R 2004 PM_{10} (µg/m^3)

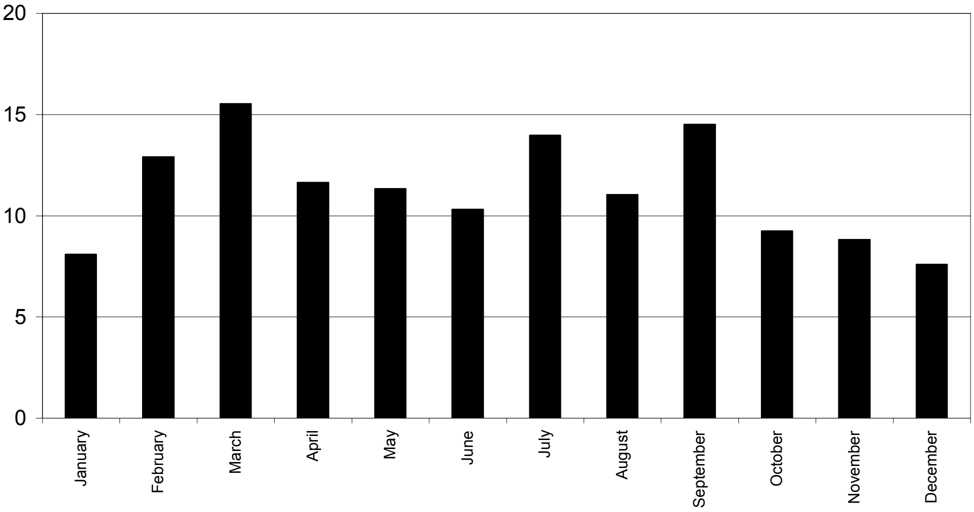


CH0002R 2004 $PM_{2.5}$ (µg/m^3)

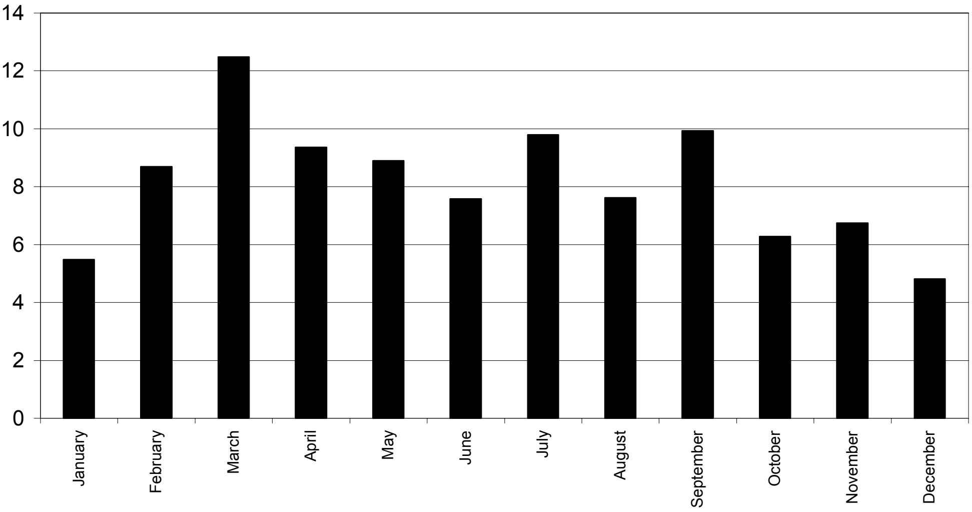
CH0003R 2004 PM_{10} (µg/m^3)



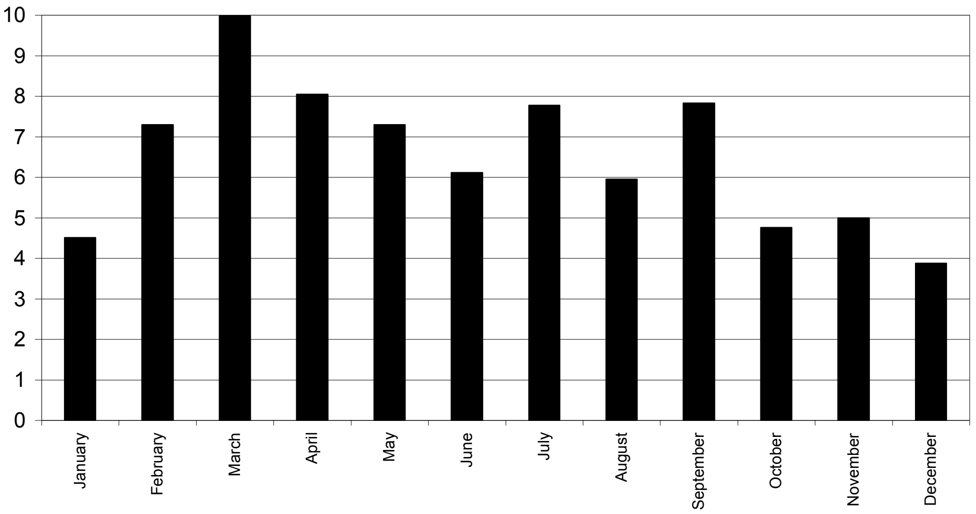
CH0004R 2004 PM_{10} (µg/m^3)



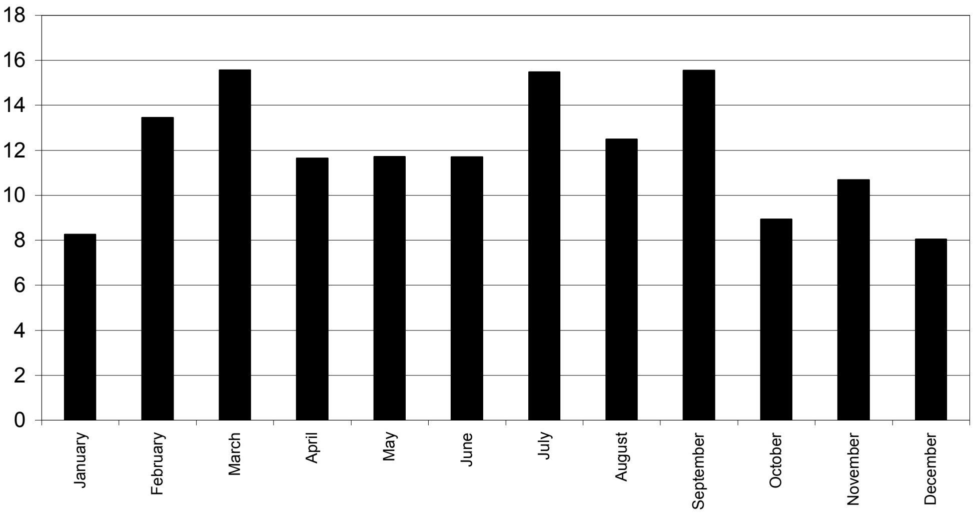
CH0004R 2004 $PM_{2.5}$ (µg/m^3)



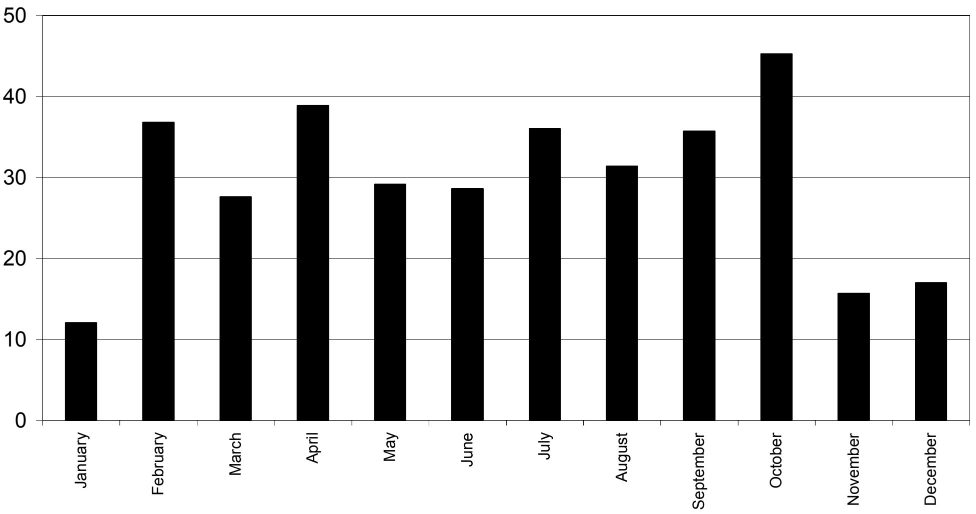
CH0004R 2004 PM_1 (µg/m^3)



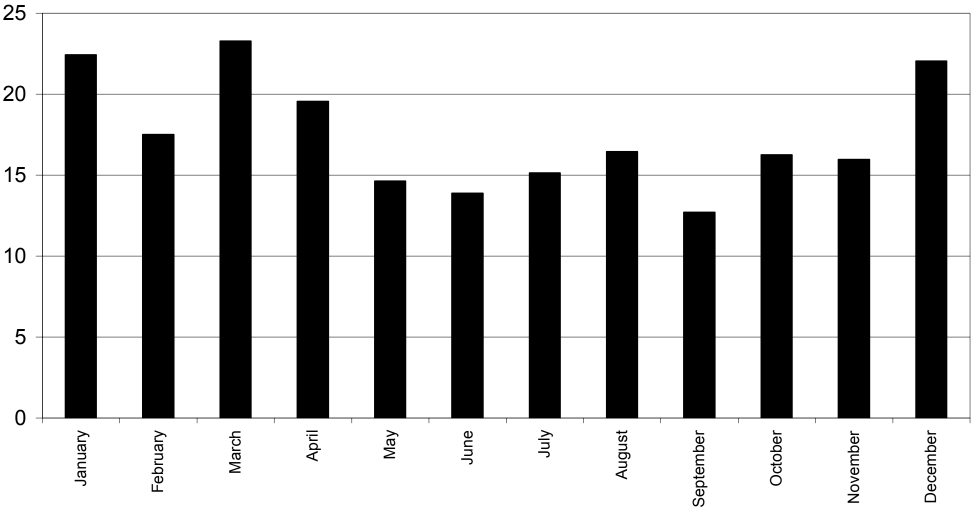
CH0005R 2004 PM_{10} (µg/m^3)



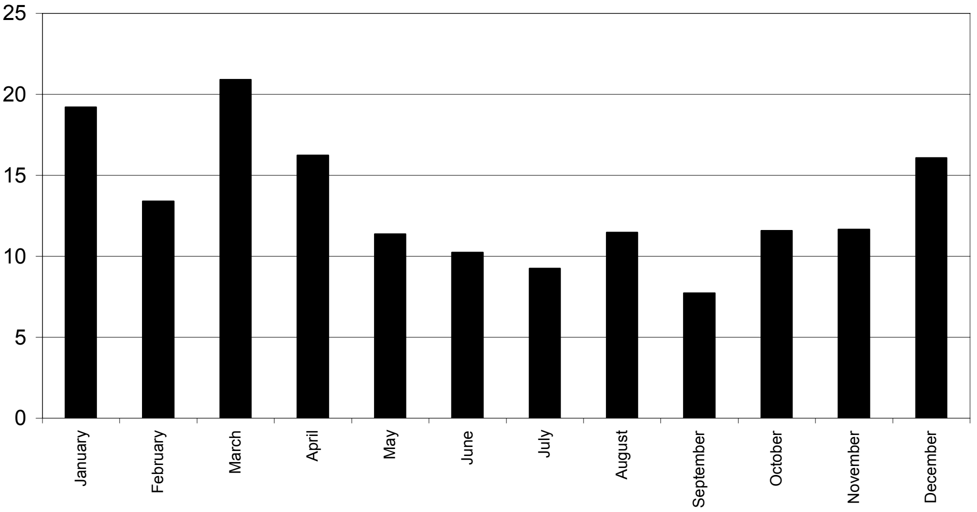
CY0002R 2004 PM_{10} (µg/m^3)



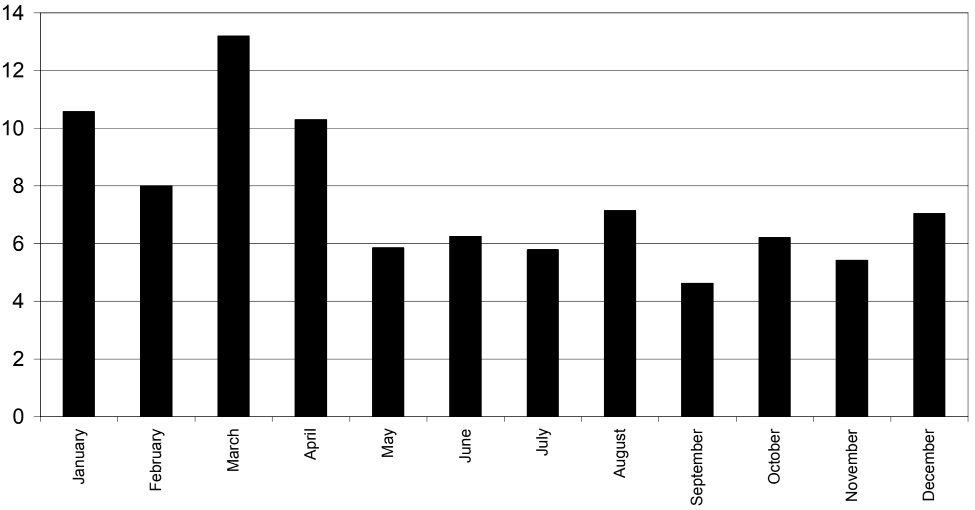
DE0002R 2004 PM$_{10}$ (µg/m^3)

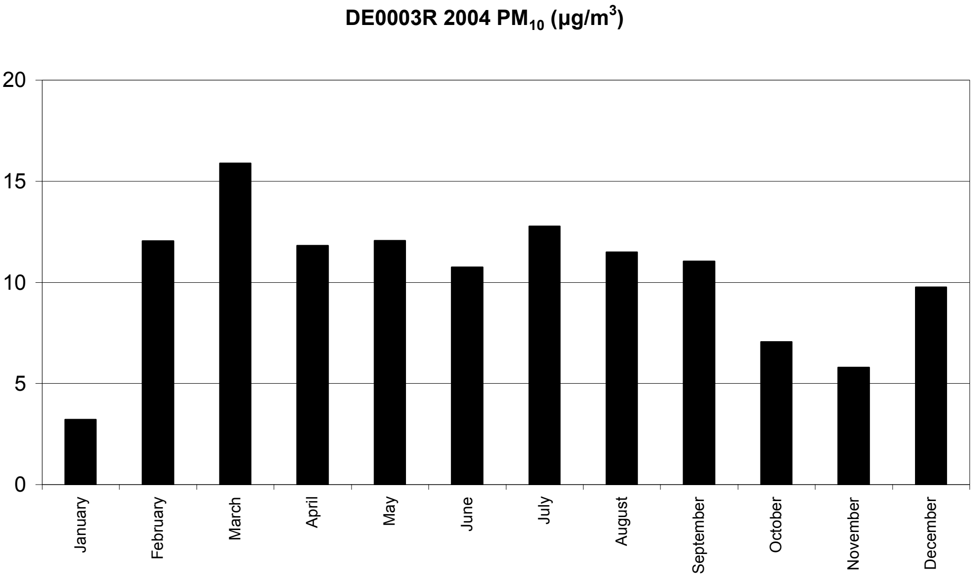


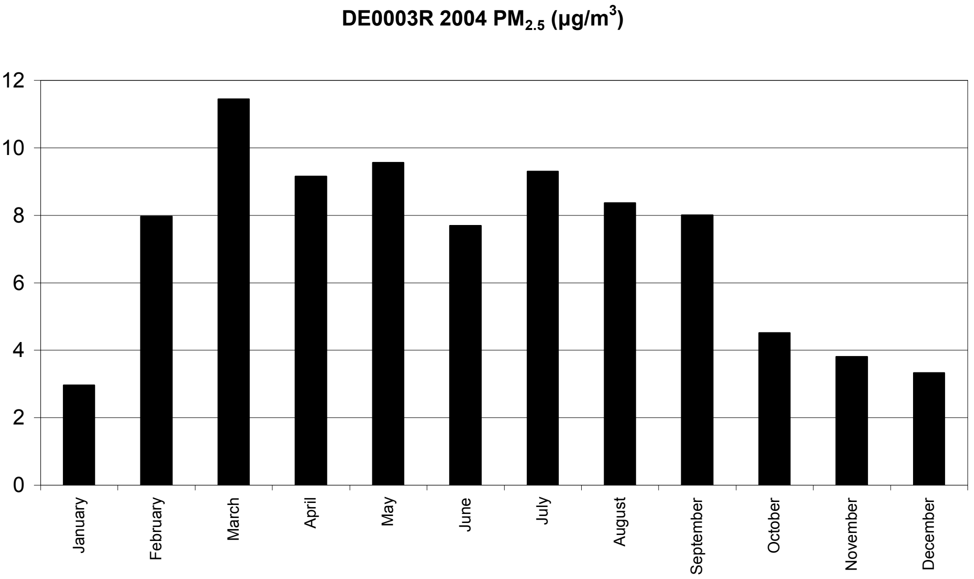
DE0002R 2004 PM$_{2.5}$ (µg/m^3)

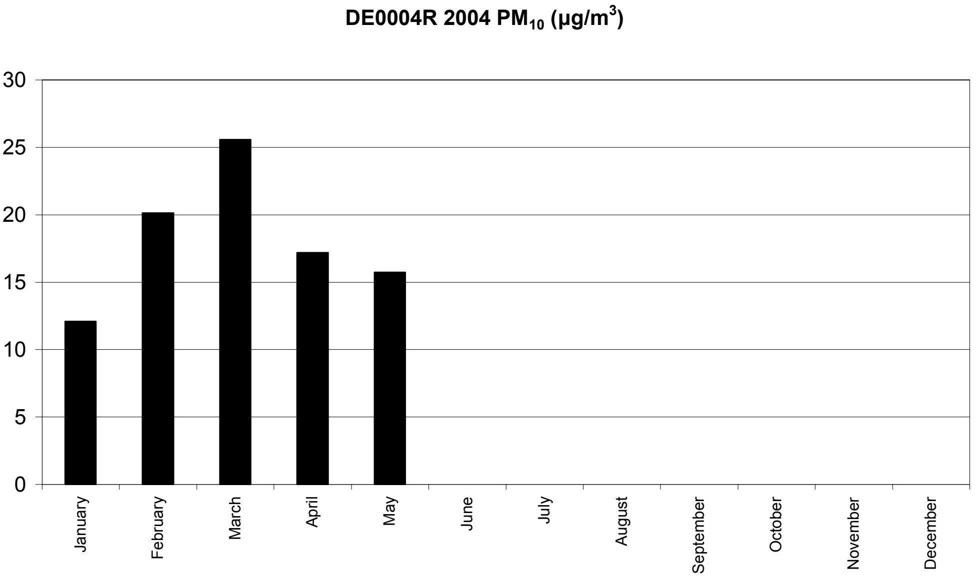


DE0002R 2004 PM$_1$ (µg/m^3)

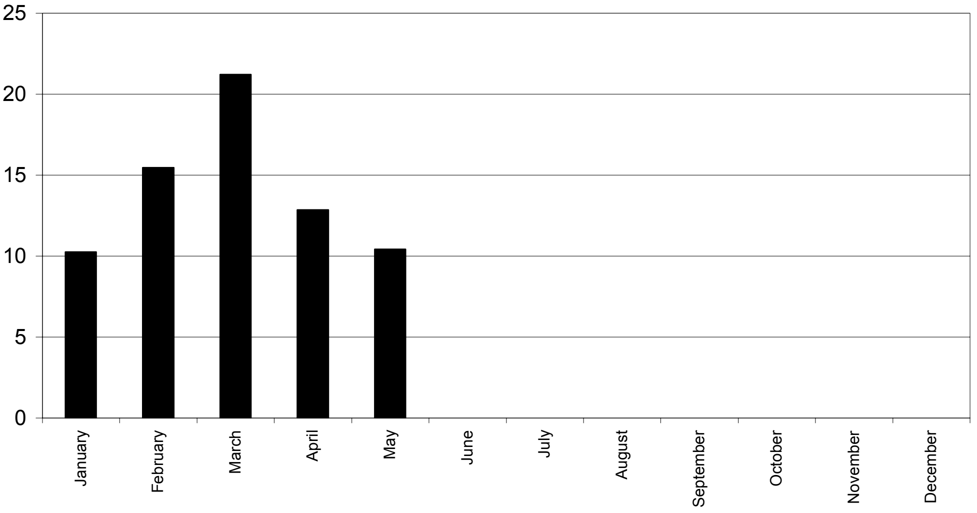



DE0003R 2004 PM_{10} (µg/m^3)
20
15
10
5
0
January
February
March
April
May
June
July
August
September
October
November
December

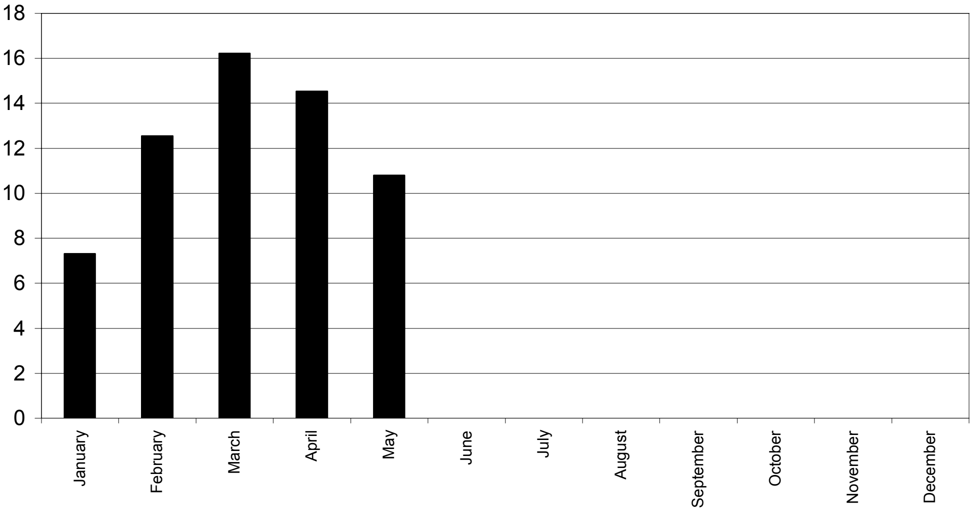

DE0003R 2004 $PM_{2.5}$ (µg/m^3)
12
10
8
6
4
2
0
January
February
March
April
May
June
July
August
September
October
November
December


DE0004R 2004 PM_{10} (µg/m^3)
30
25
20
15
10
5
0
January
February
March
April
May
June
July
August
September
October
November
December

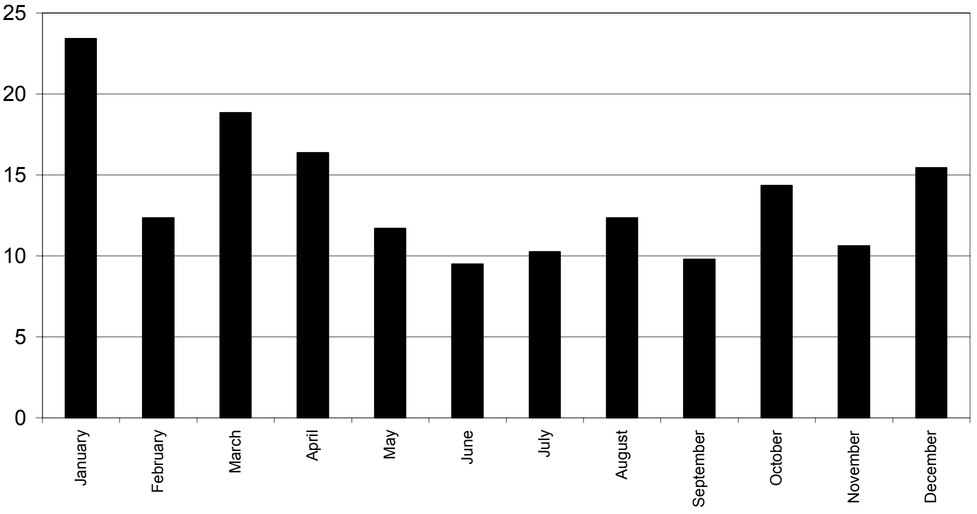
DE0004R 2004 PM$_{2.5}$ (µg/m^3)



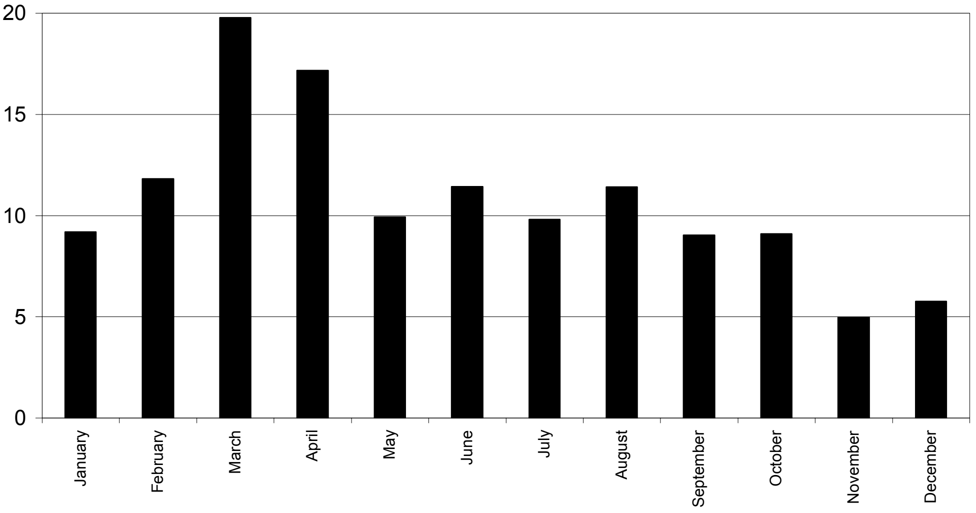
DE0005R 2004 PM$_{10}$ (µg/m^3)



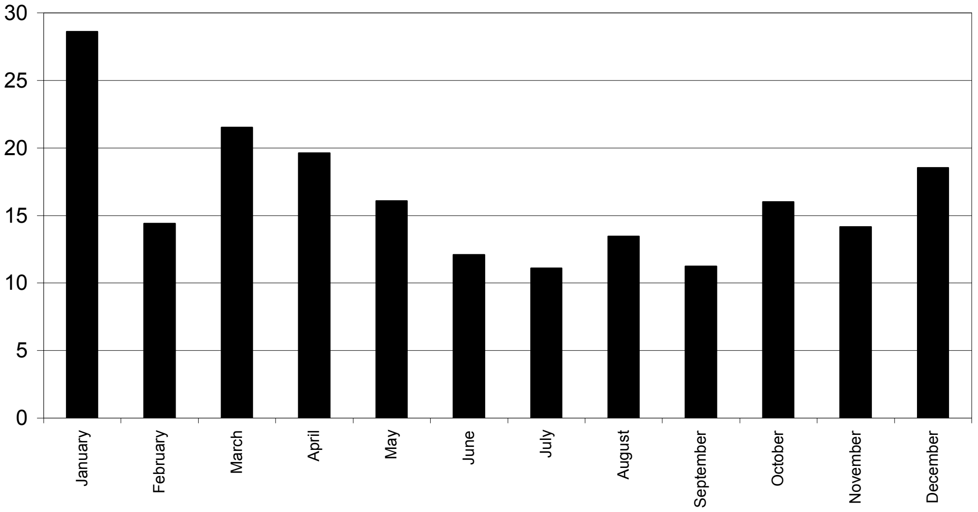
DE0007R 2004 PM$_{10}$ (µg/m^3)



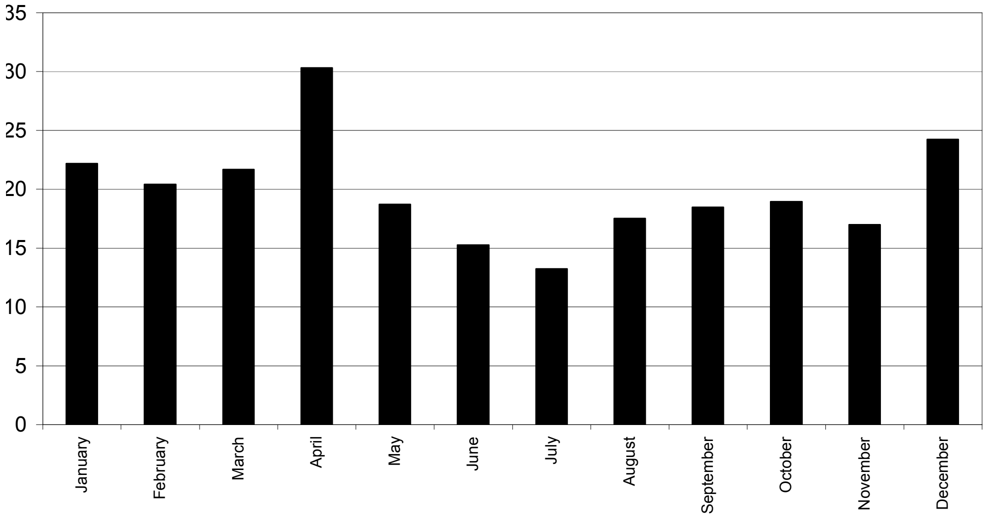
DE0008R 2004 PM_{10} (µg/m^3)



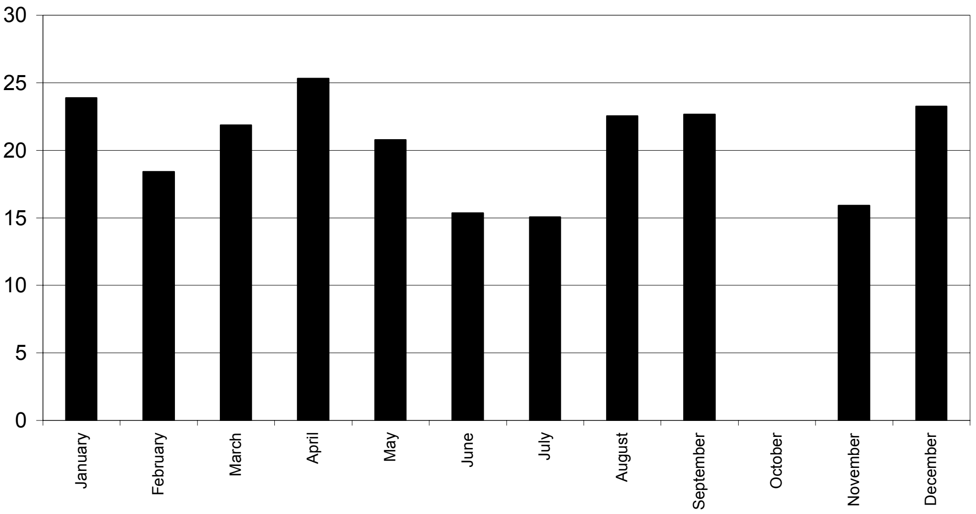
DE0009R 2004 PM_{10} (µg/m^3)



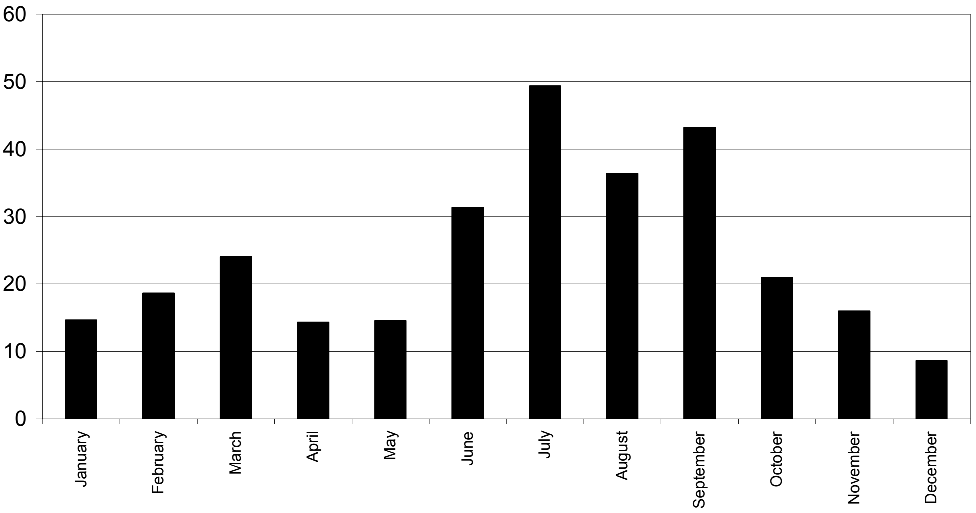
DE0041R 2004 PM_{10} (µg/m^3)



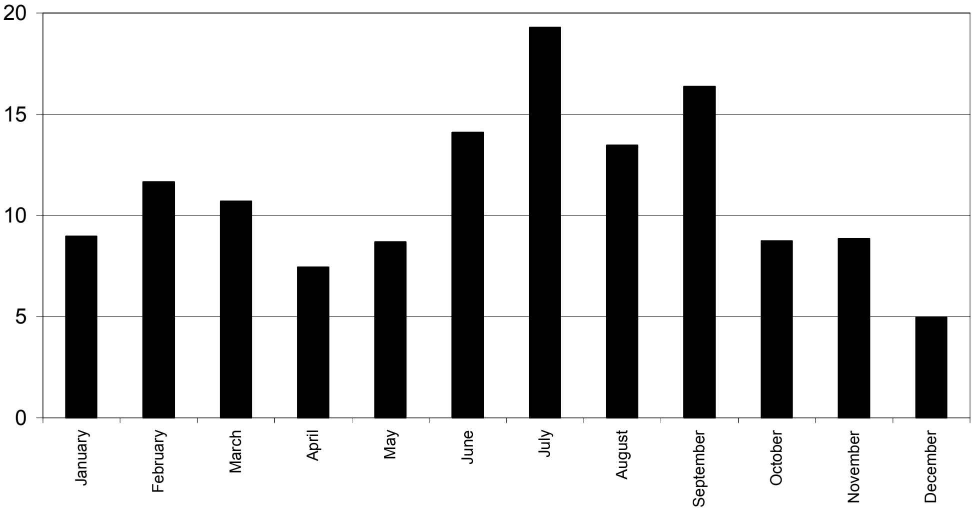
DK0005R 2004 PM_{10} (µg/m^3)

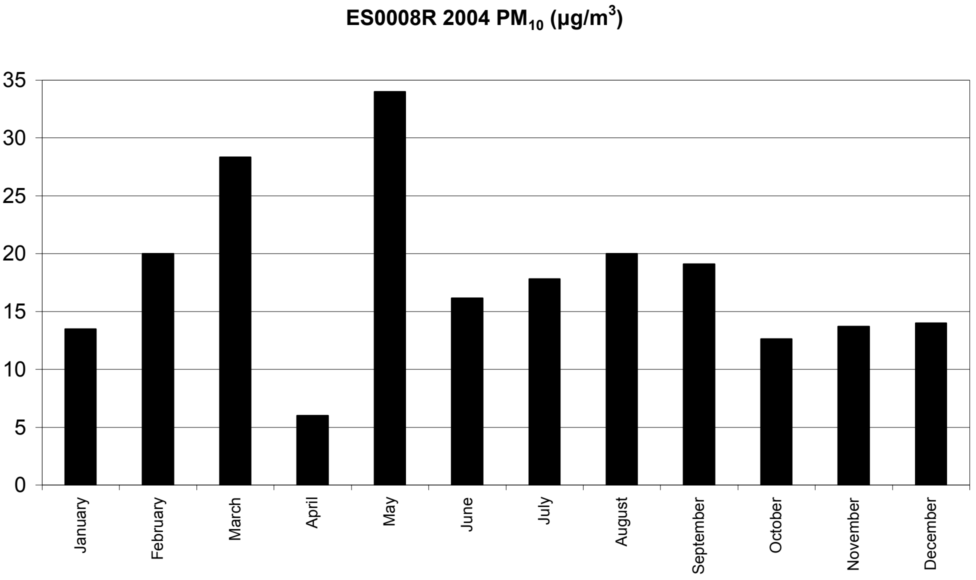


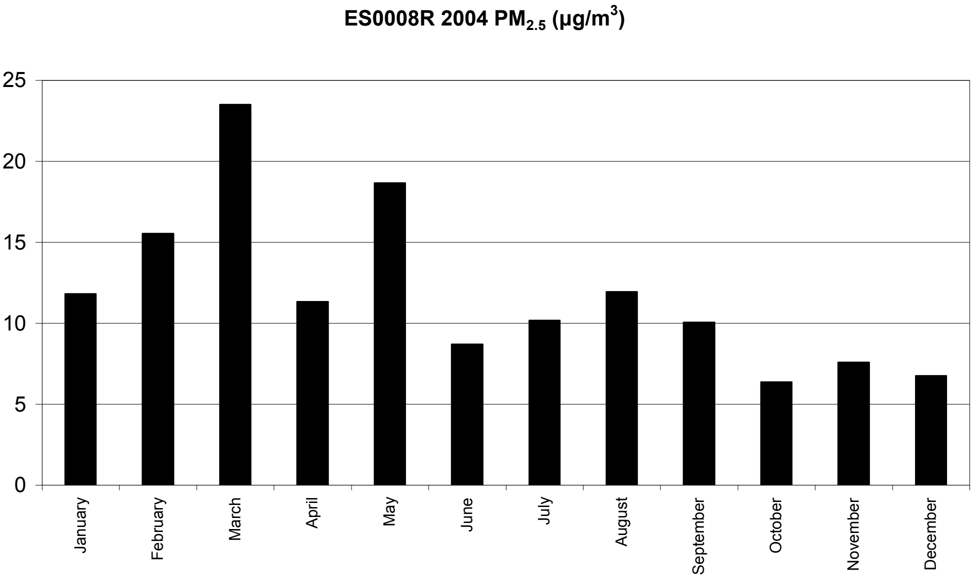
ES0007R 2004 PM_{10} (µg/m^3)

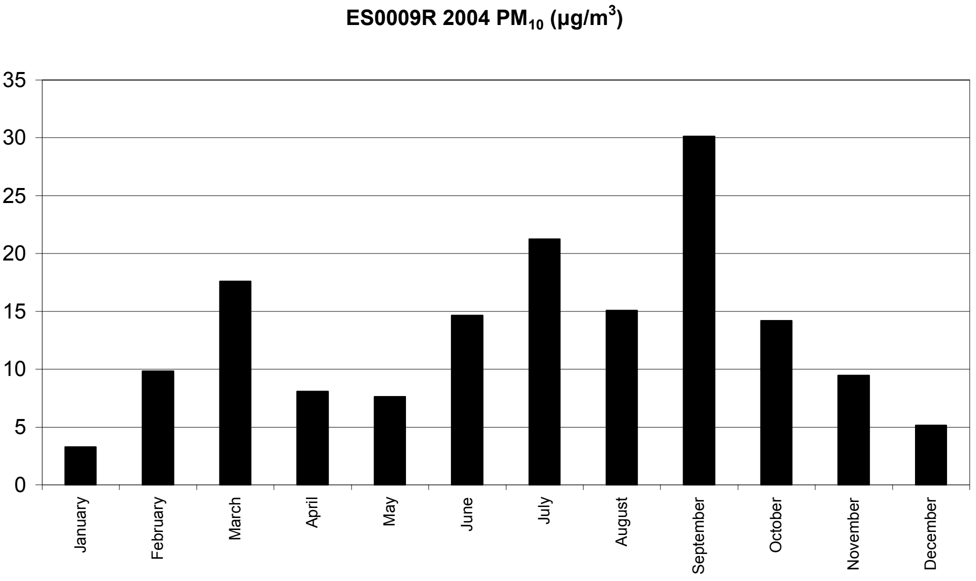


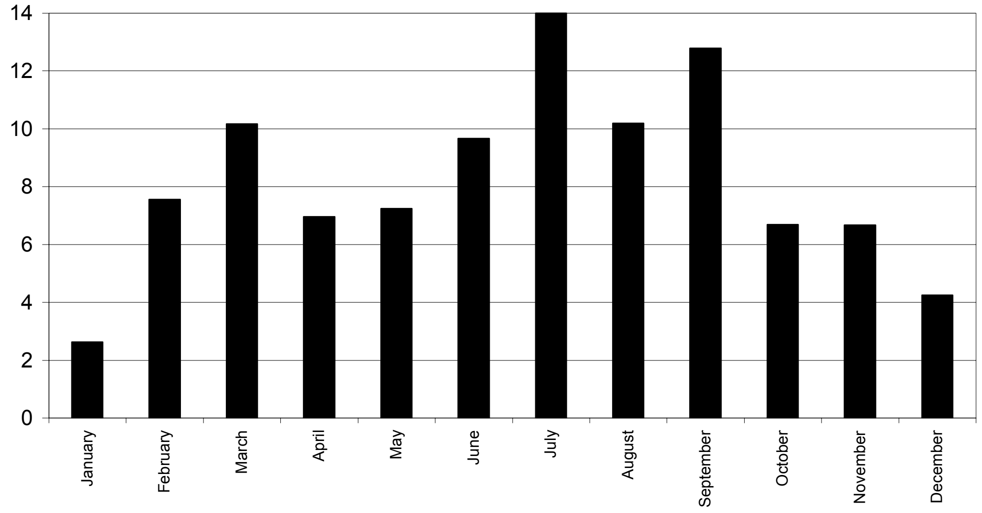
ES0007R 2004 $PM_{2.5}$ (µg/m^3)



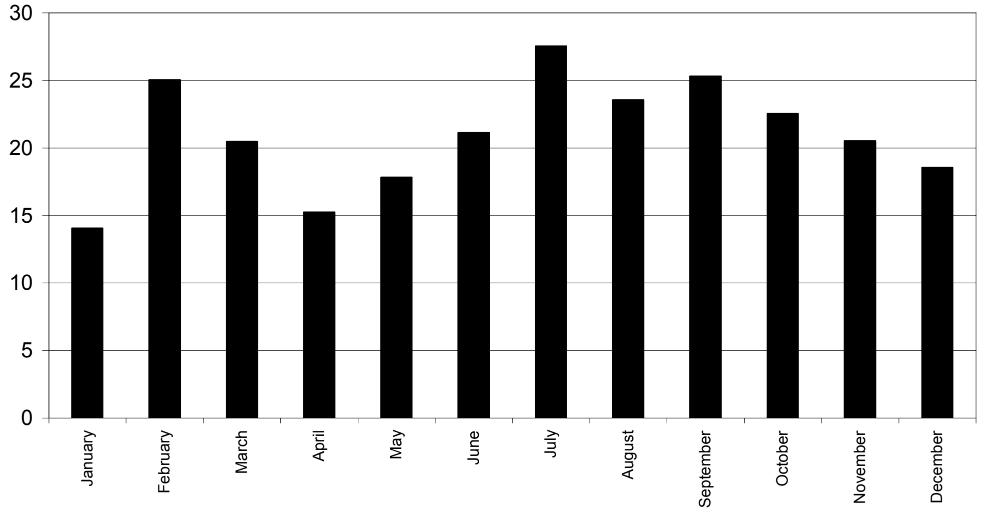

ES0008R 2004 PM$_{10}$ (µg/m^3)
35
30
25
20
15
10
5
0
January
February
March
April
May
June
July
August
September
October
November
December


ES0008R 2004 PM$_{2.5}$ (µg/m^3)
25
20
15
10
5
0
January
February
March
April
May
June
July
August
September
October
November
December

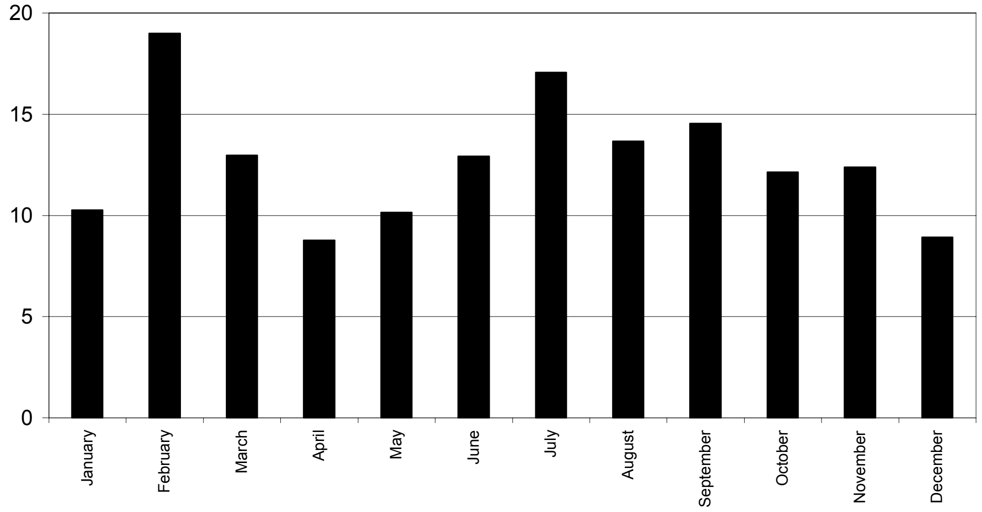

ES0009R 2004 PM$_{10}$ (µg/m^3)
35
30
25
20
15
10
5
0
January
February
March
April
May
June
July
August
September
October
November
December



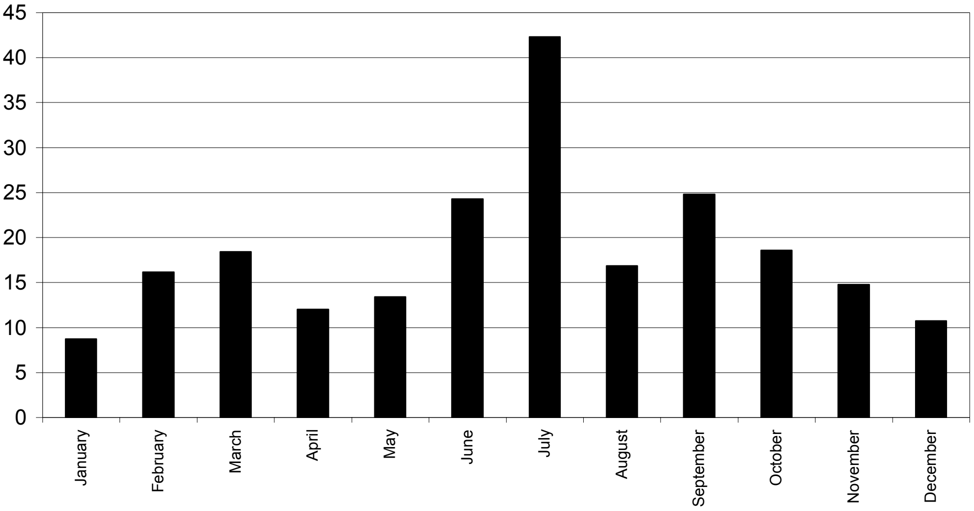
ES0009R 2004 $PM_{2.5}$ ($\mu g/m^3$)

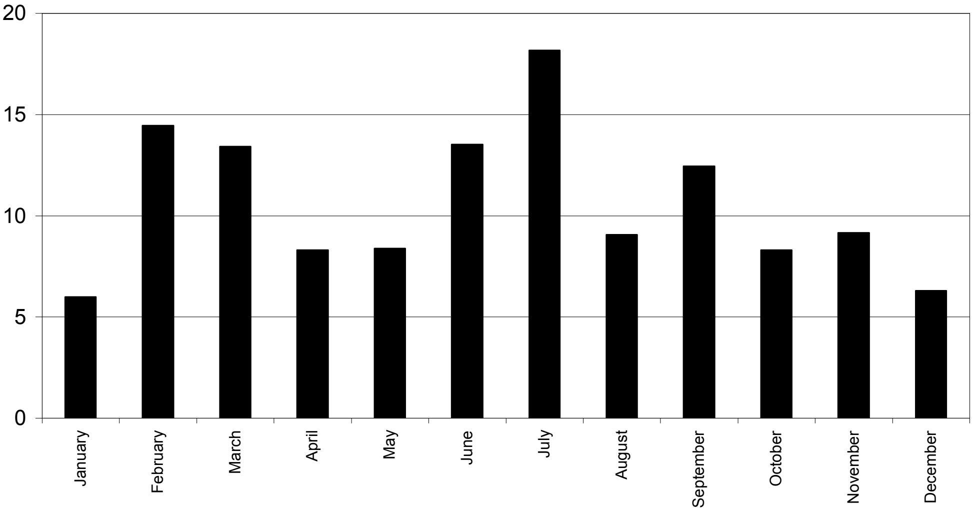


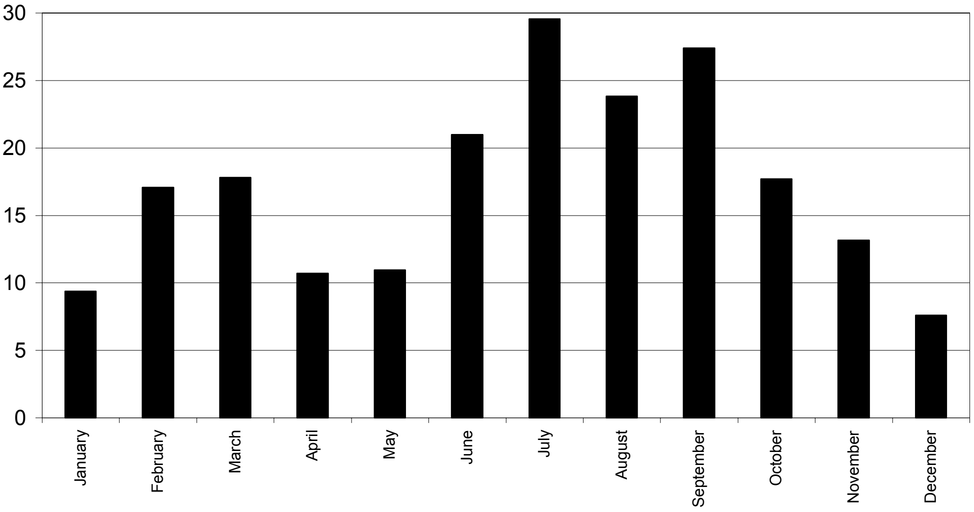
ES0010R 2004 PM_{10} ($\mu g/m^3$)



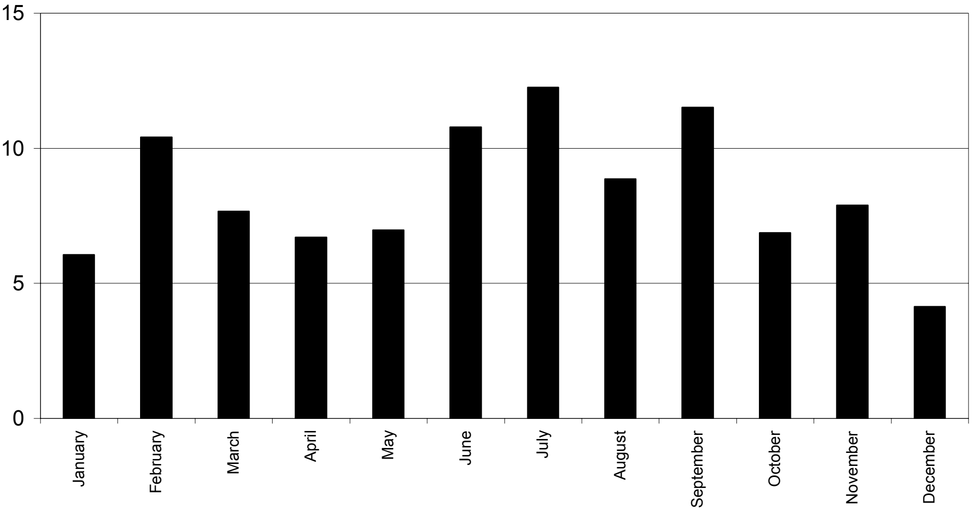
ES0010R 2004 $PM_{2.5}$ ($\mu g/m^3$)


ES0011R 2004 PM_{10} ($\mu g/m^3$)

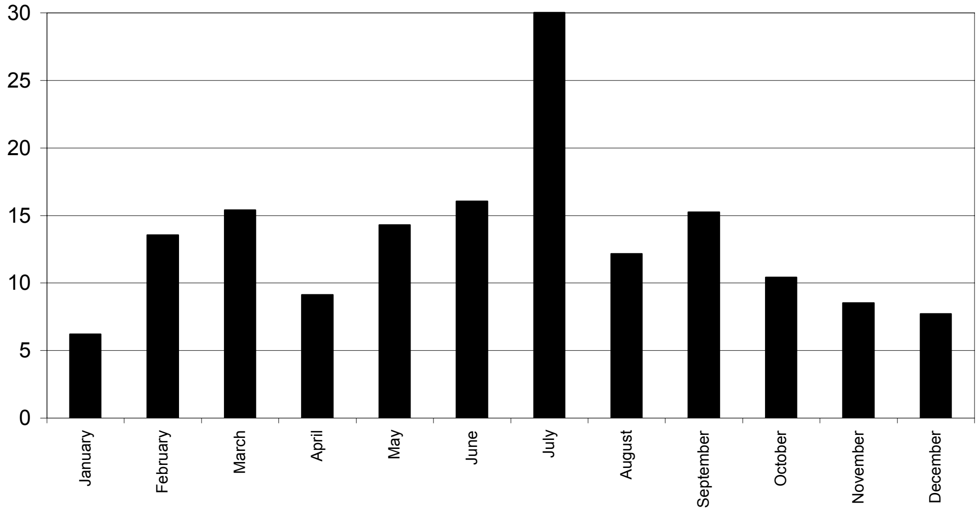

ES0011R 2004 $PM_{2.5}$ ($\mu g/m^3$)


ES0012R 2004 PM_{10} ($\mu g/m^3$)

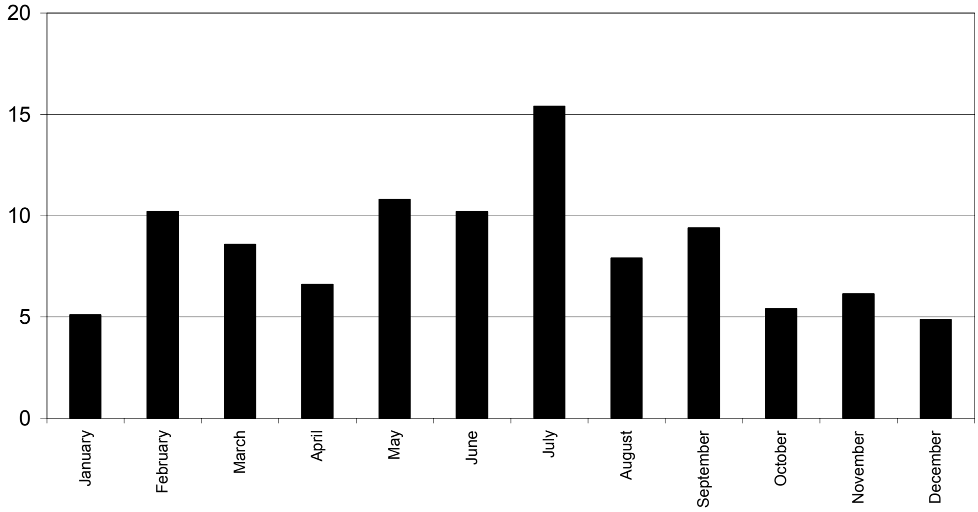
ES0012R 2004 PM$_{2.5}$ (µg/m^3)



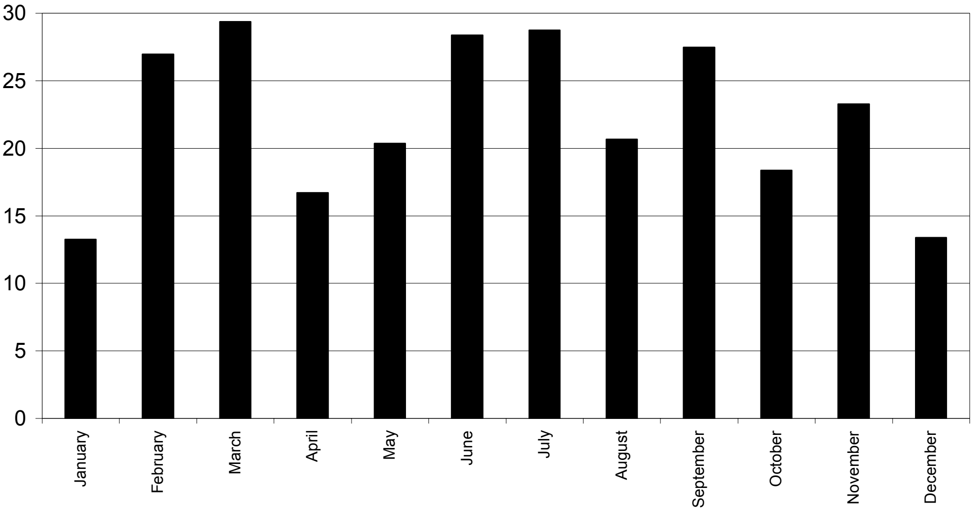
ES0013R 2004 PM$_{10}$ (µg/m^3)



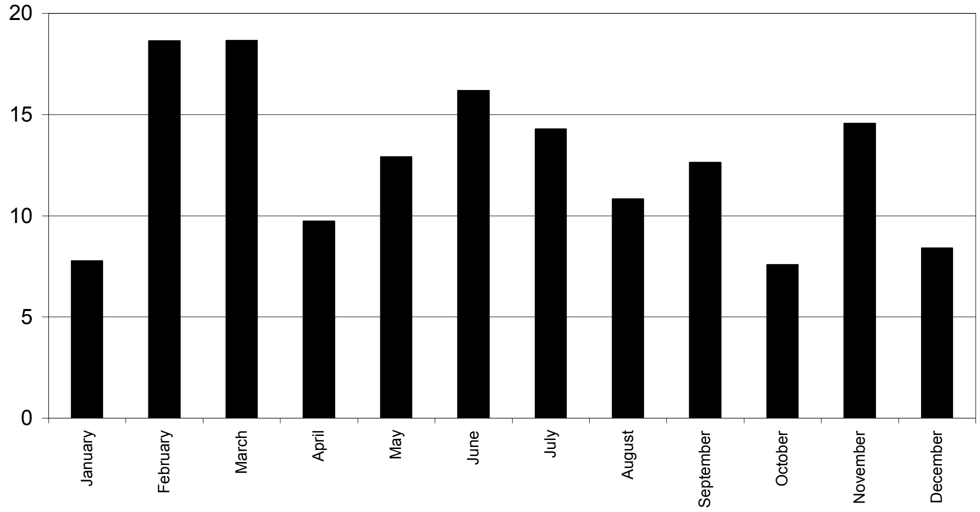
ES0013R 2004 PM$_{2.5}$ (µg/m^3)



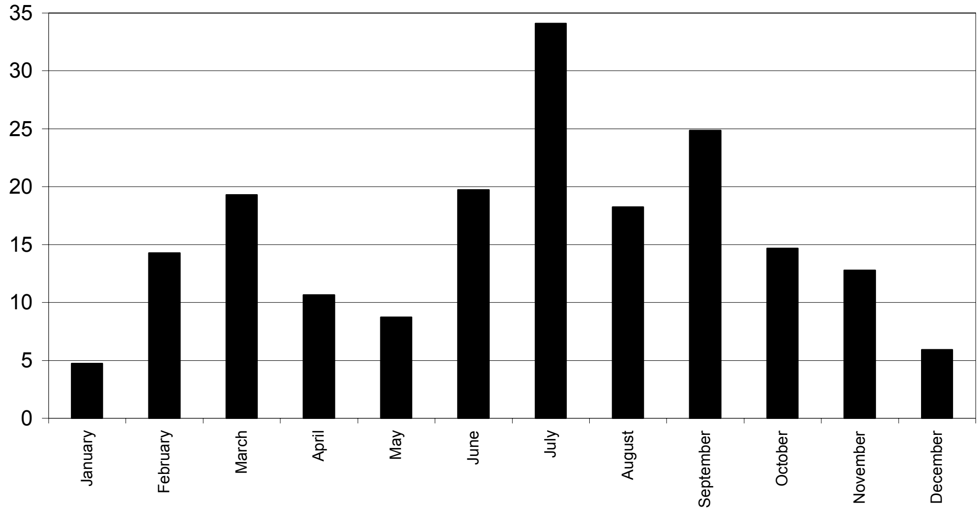
ES0014R 2004 PM_{10} (µg/m^3)



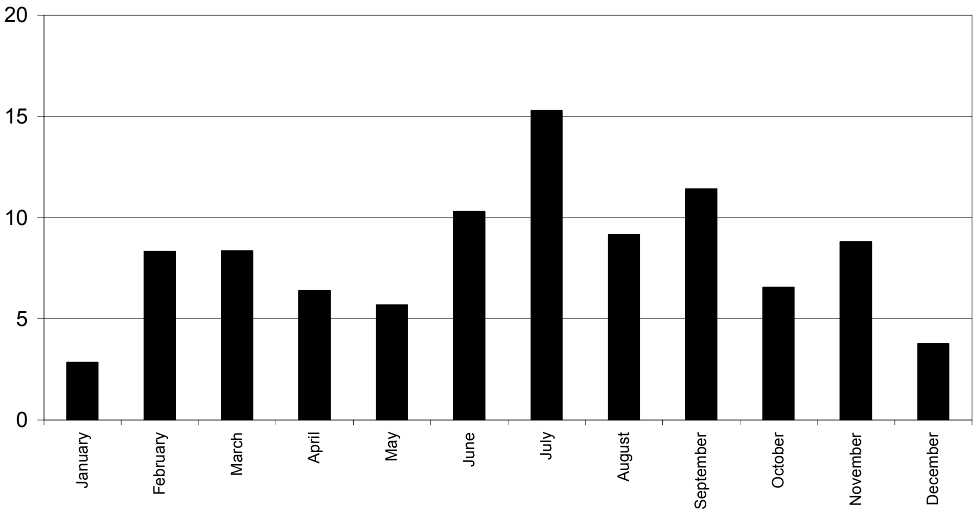
ES0014R 2004 $PM_{2.5}$ (µg/m^3)



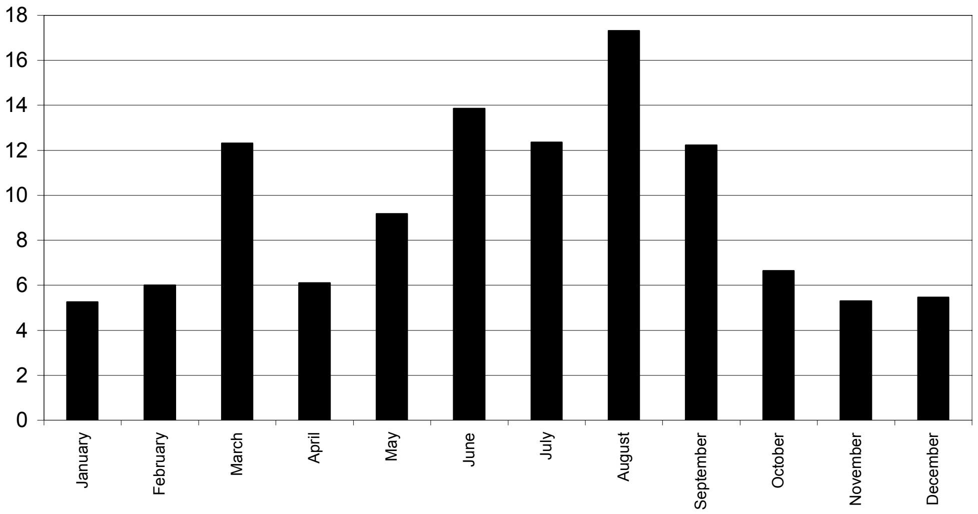
ES0015R 2004 PM_{10} (µg/m^3)



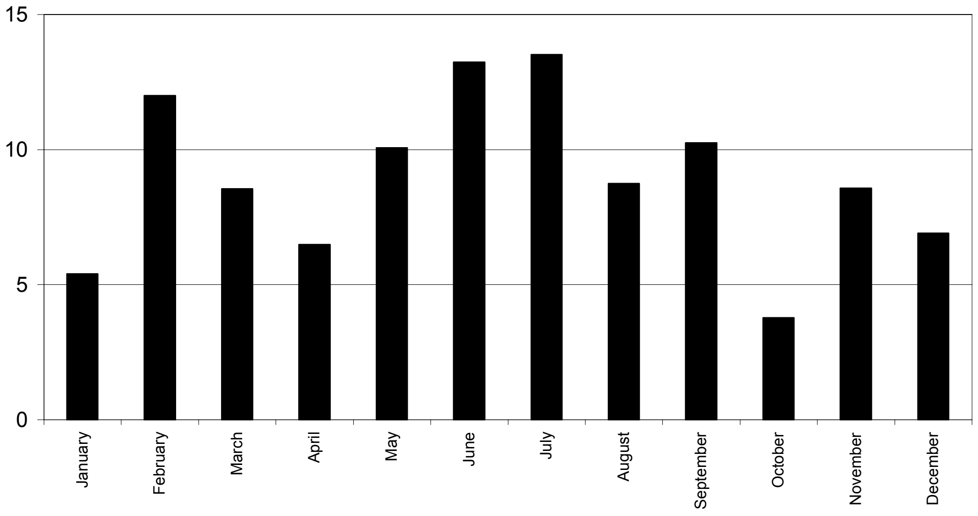
ES0015R 2004 PM$_{2.5}$ (µg/m^3)



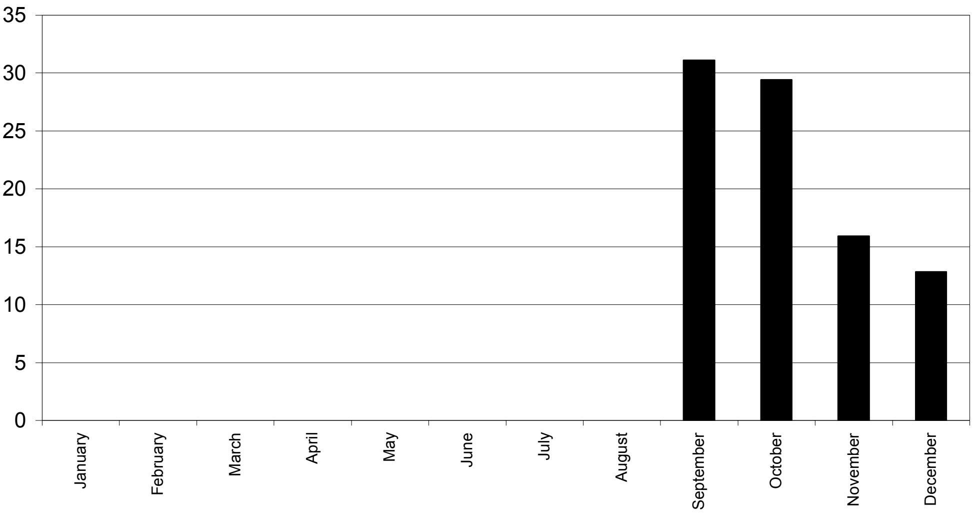
ES0016R 2004 PM$_{10}$ (µg/m^3)



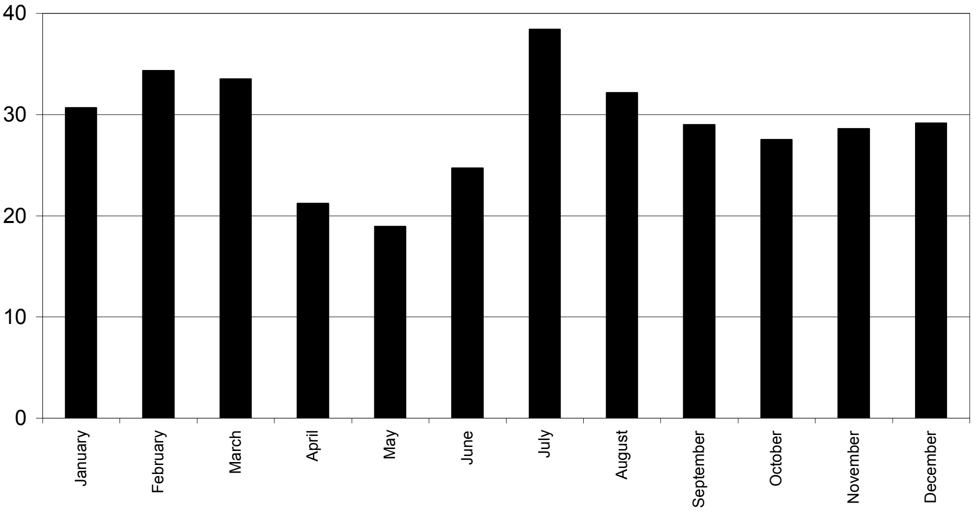
ES0016R 2004 PM$_{2.5}$ (µg/m^3)



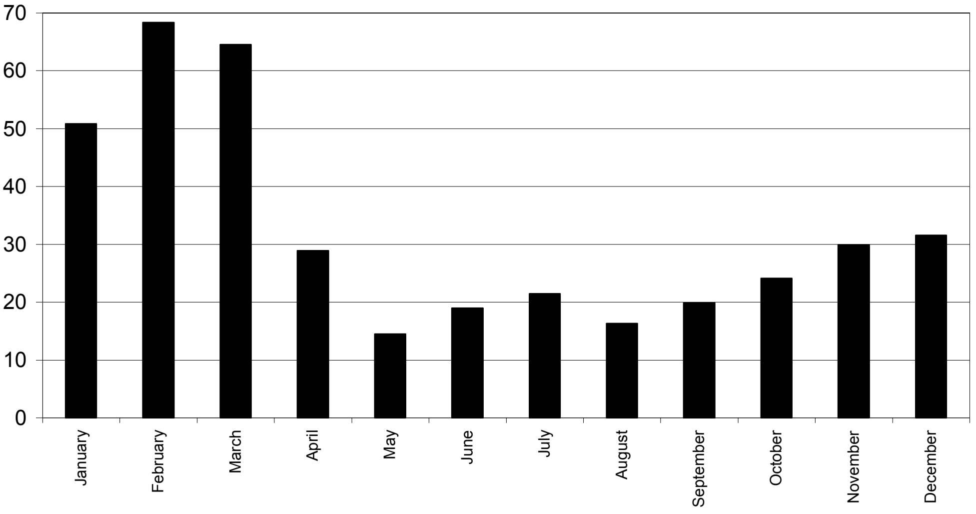
GR0002R 2004 PM$_{10}$ (µg/m^3)



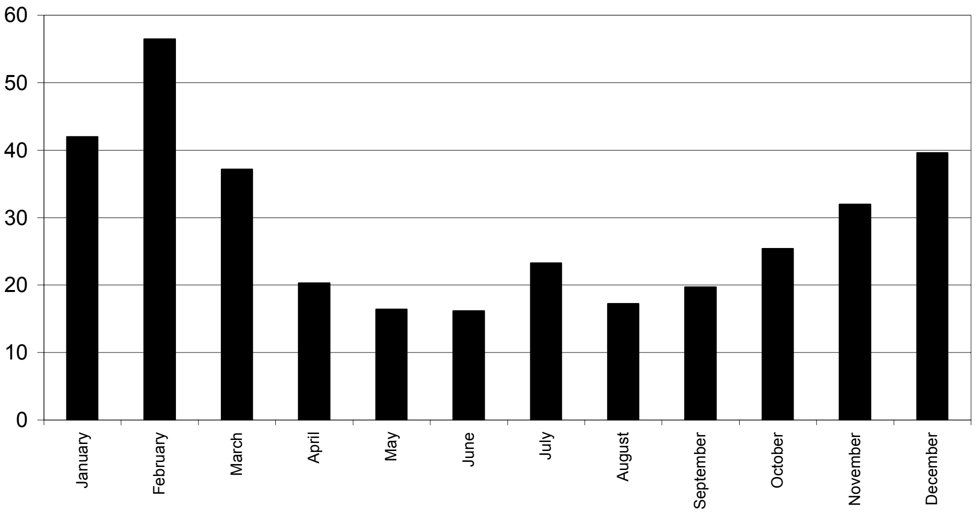
IT0001R 2004 PM$_{10}$ (µg/m^3)



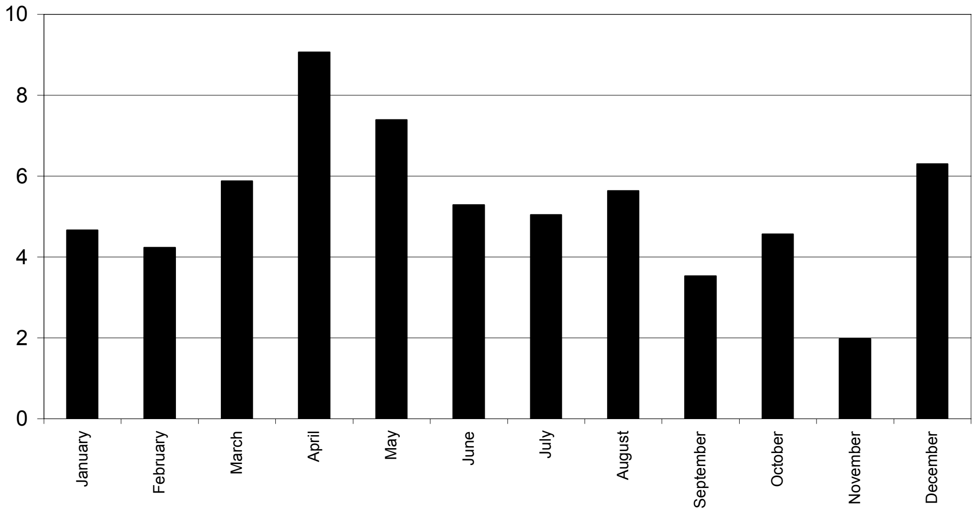
IT0004R 2004 PM$_{10}$ (µg/m^3)



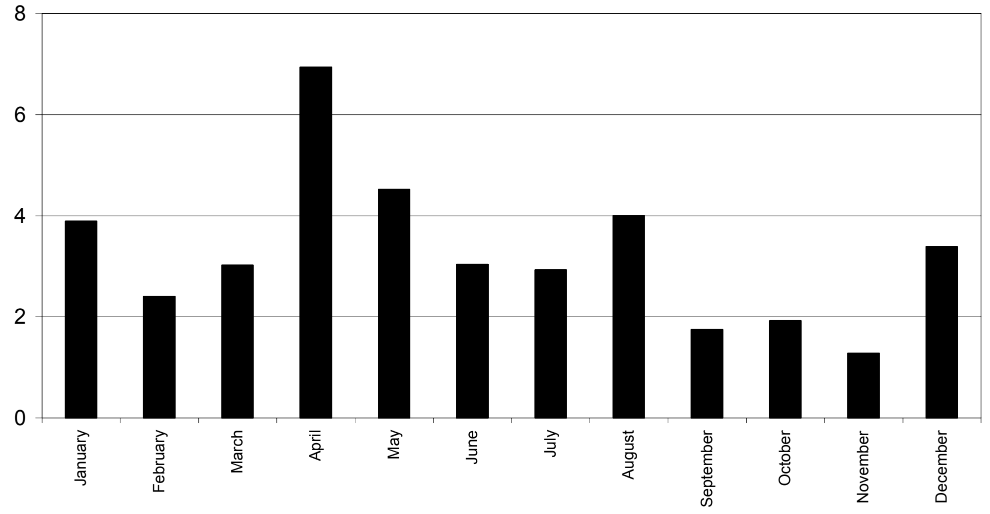
IT0004R 2004 $PM_{2.5}$ ($\mu g/m^3$)

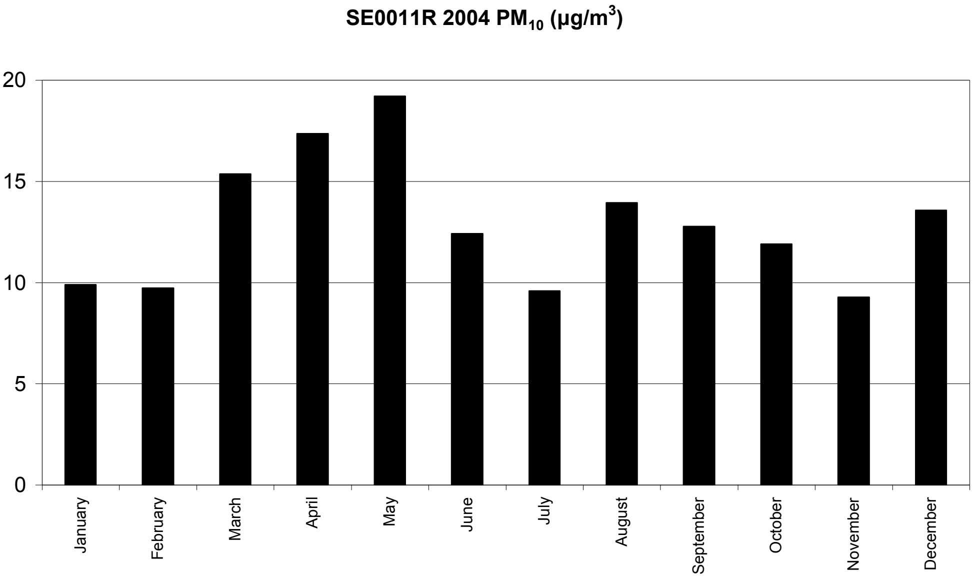


NO0001R 2004 PM_{10} ($\mu g/m^3$)

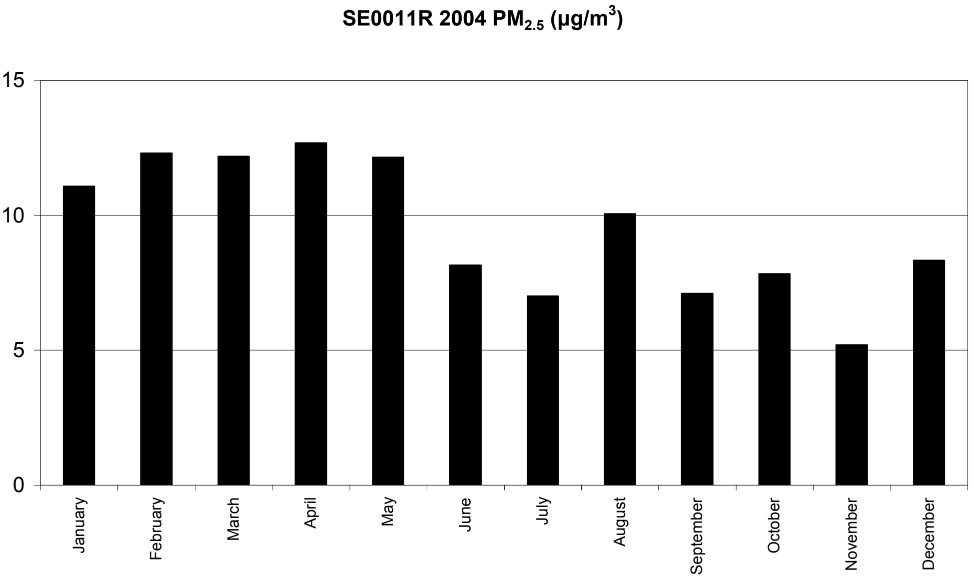


NO0001R 2004 $PM_{2.5}$ ($\mu g/m^3$)

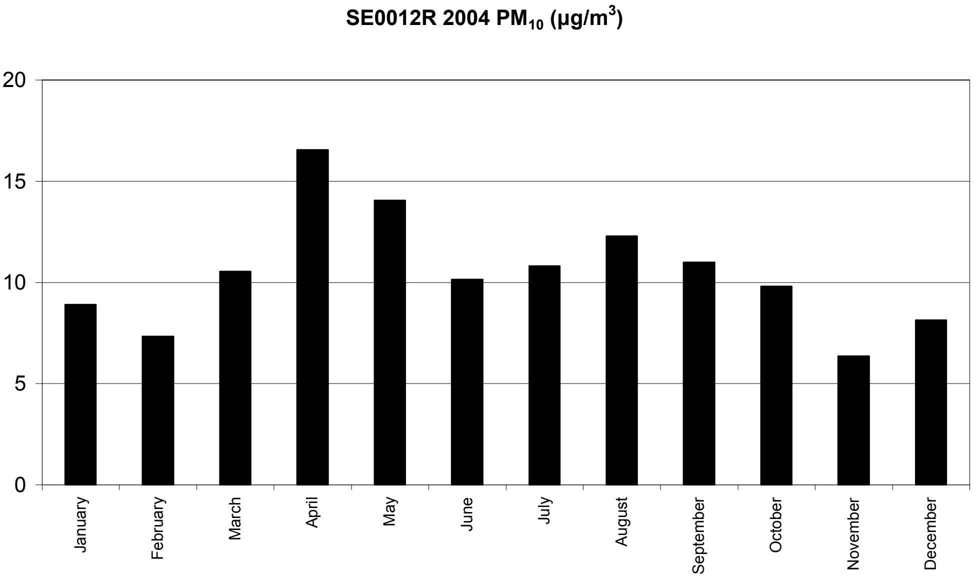




SE0011R 2004 PM_{10} ($\mu g/m^3$)

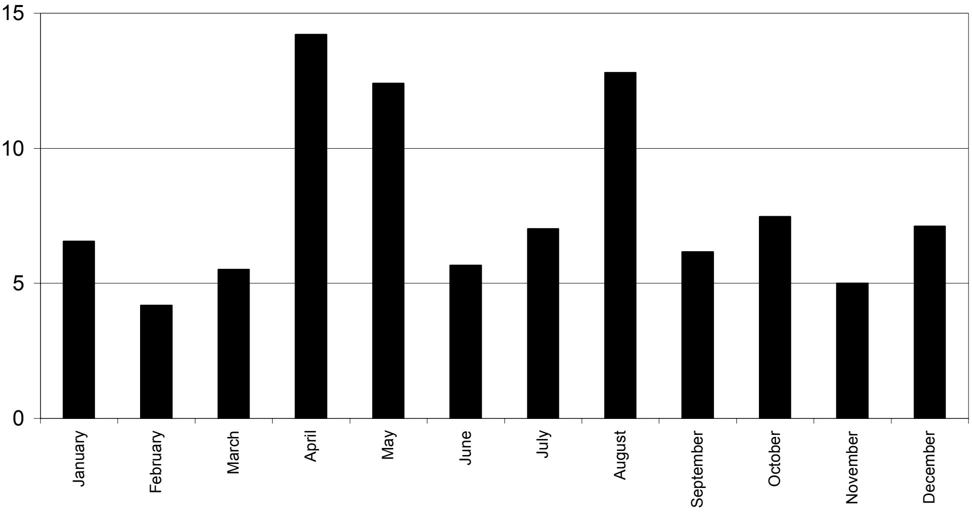


SE0011R 2004 $PM_{2.5}$ ($\mu g/m^3$)

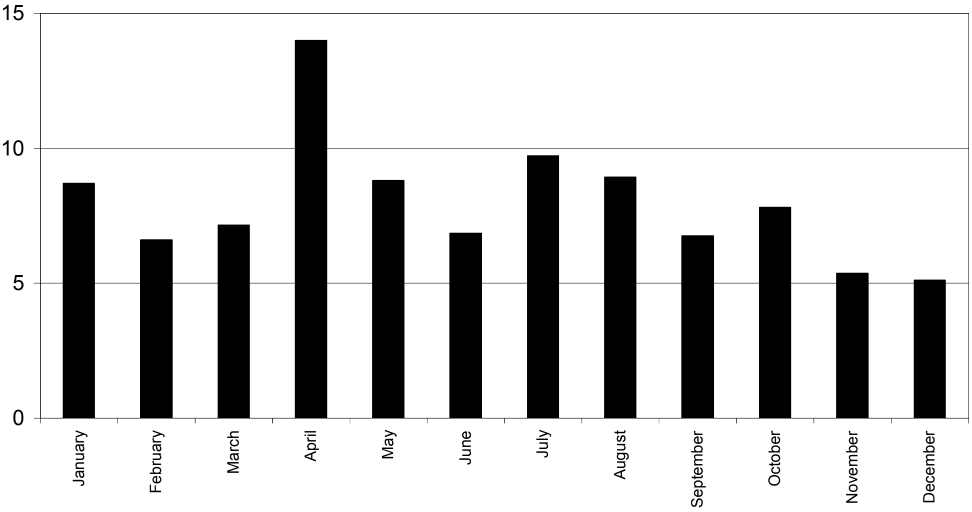


SE0012R 2004 PM_{10} ($\mu g/m^3$)

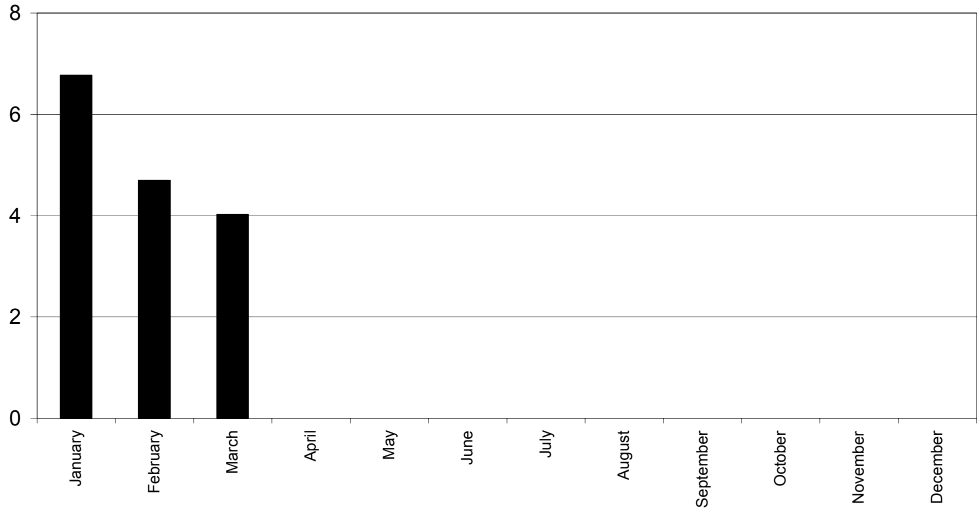
SE0012R 2004 $PM_{2.5}$ (µg/m^3)

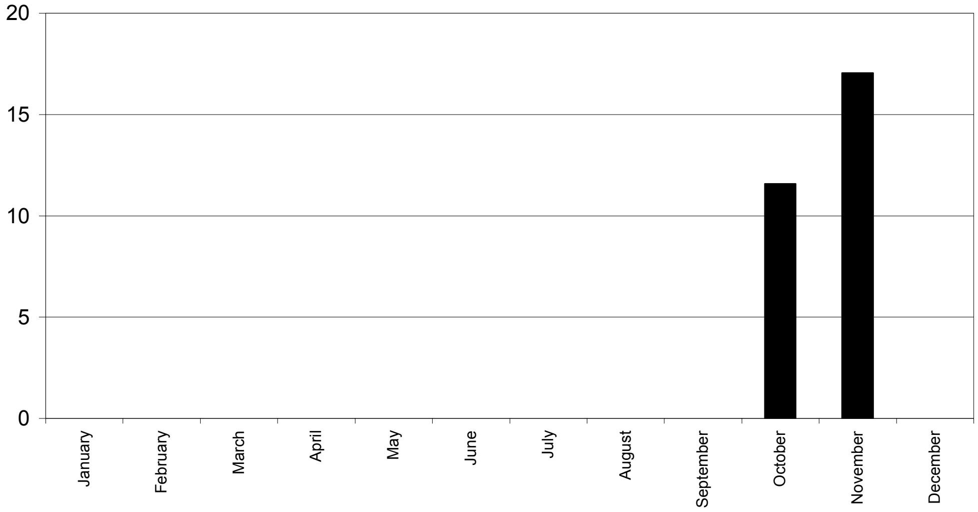


SE0035R 2004 PM_{10} (µg/m^3)

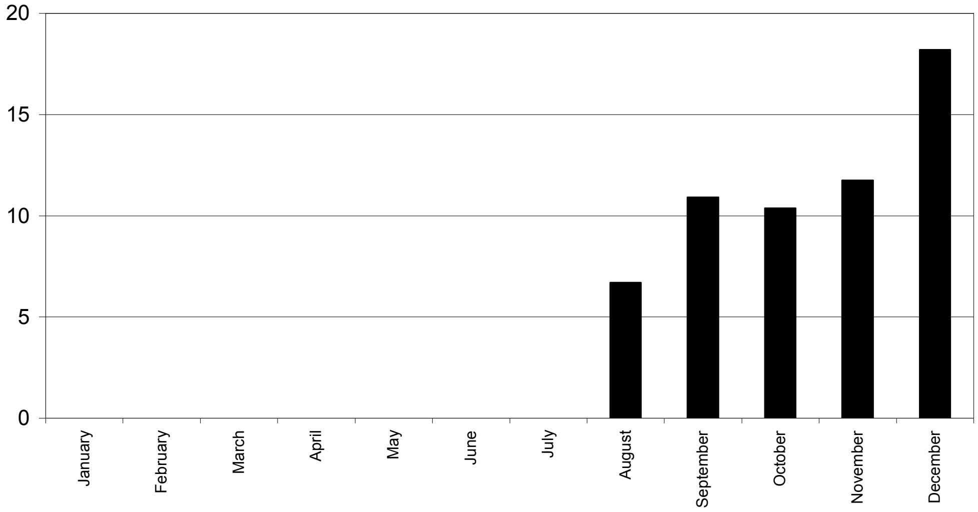


SE0035R 2004 $PM_{2.5}$ (µg/m^3)





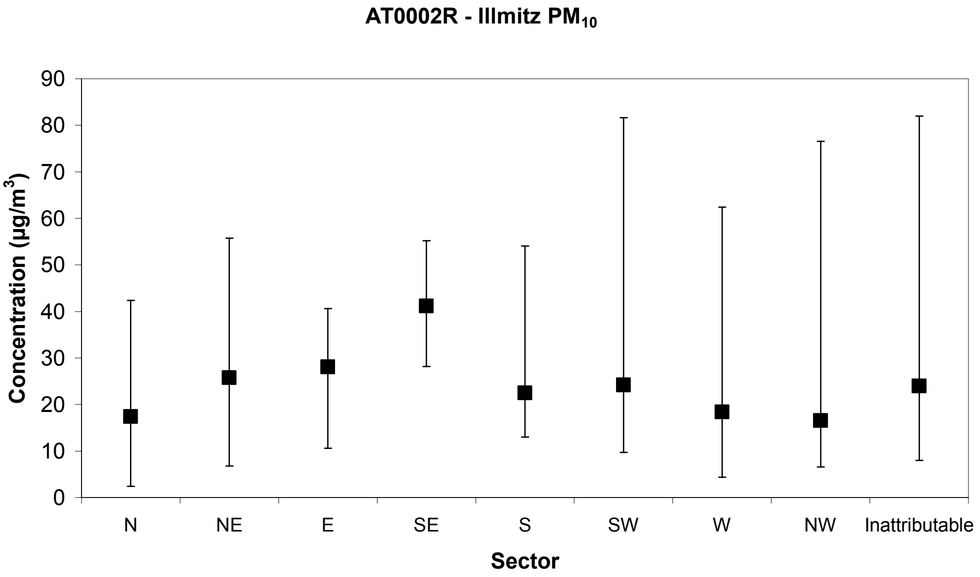
SI0008R 2004 PM$_{10}$ (µg/m^3)

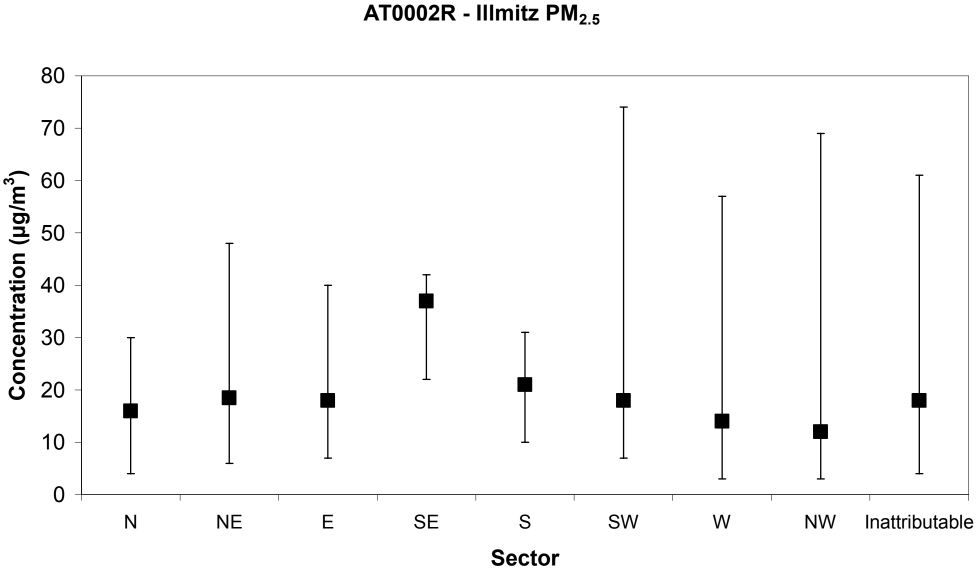


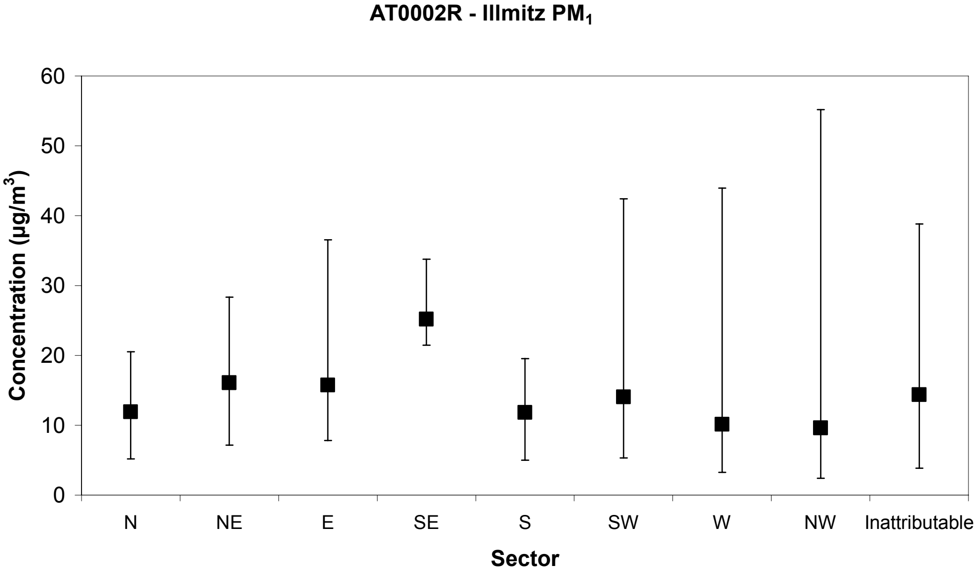
SI0008R 2004 PM$_{2.5}$ (µg/m^3)

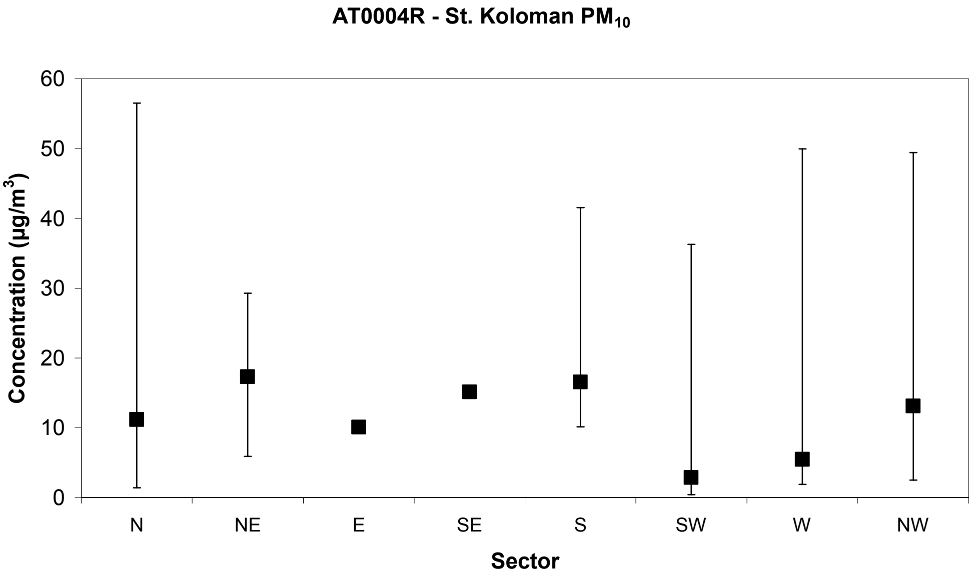
Annex 4

Particulate mass concentrations averaged by origin of air mass trajectories at EMEP stations in 2004

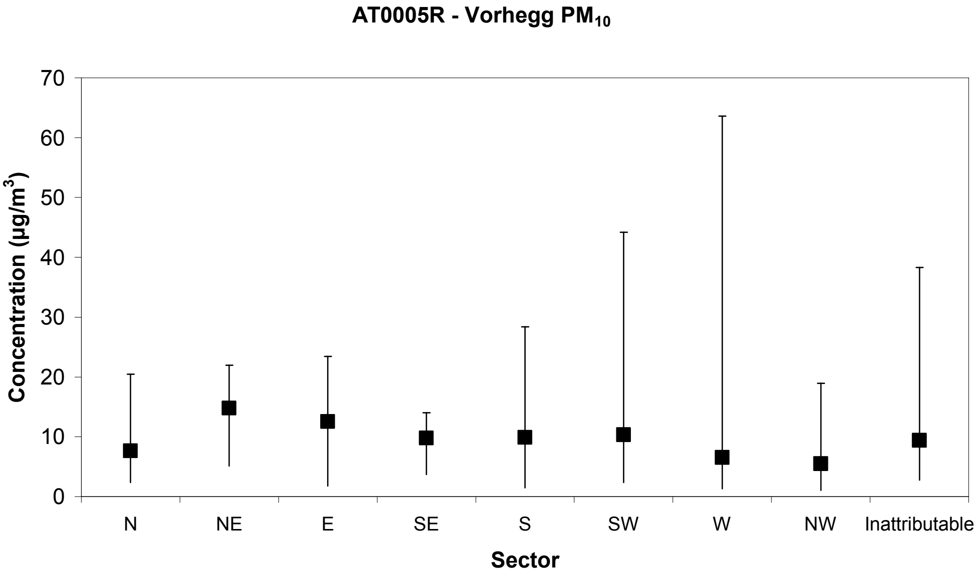

AT0002R - Illmitz PM$_{10}$
Concentration (µg/m^3)
0
10
20
30
40
50
60
70
80
90
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector


AT0002R - Illmitz PM$_{2.5}$
Concentration (µg/m^3)
0
10
20
30
40
50
60
70
80
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

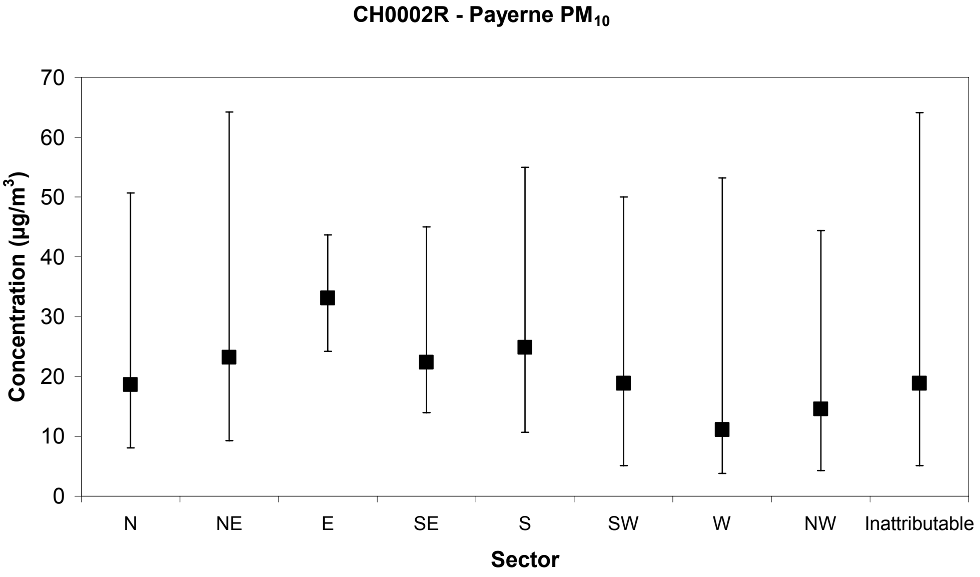

AT0002R - Illmitz PM$_1$
Concentration (µg/m^3)
0
10
20
30
40
50
60
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector



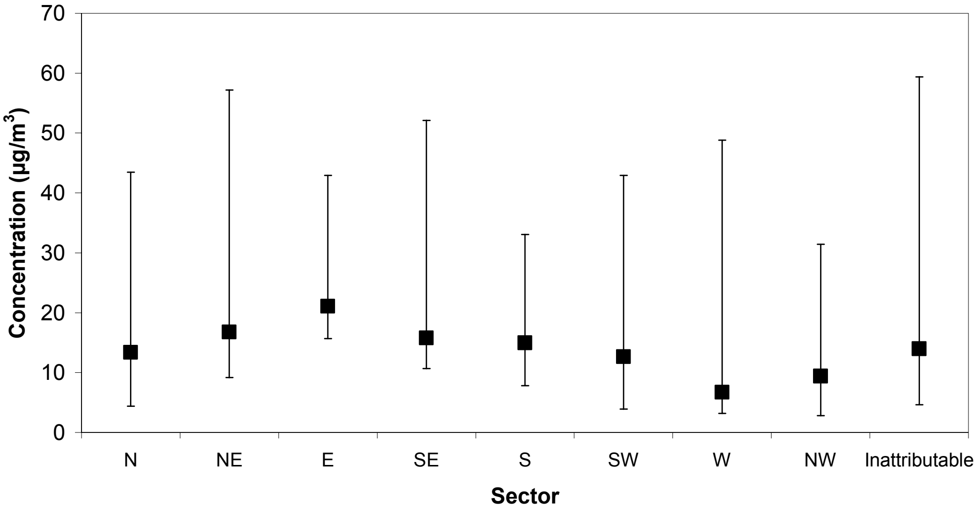
AT0004R - St. Koloman PM_{10}

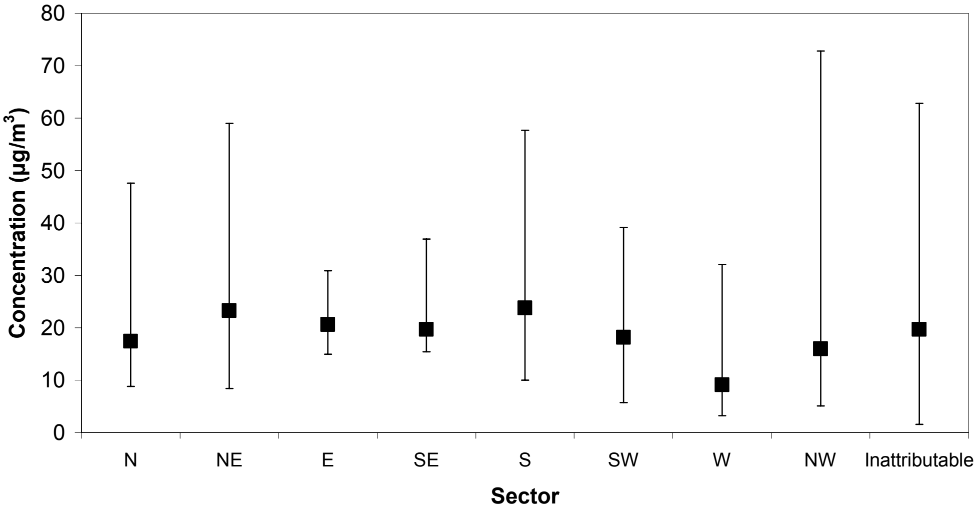


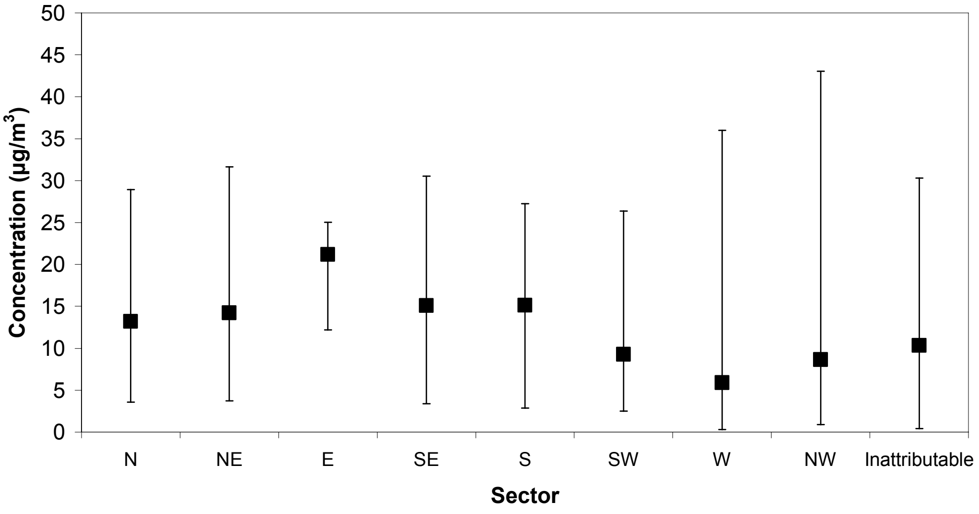
AT0005R - Vorhegg PM_{10}

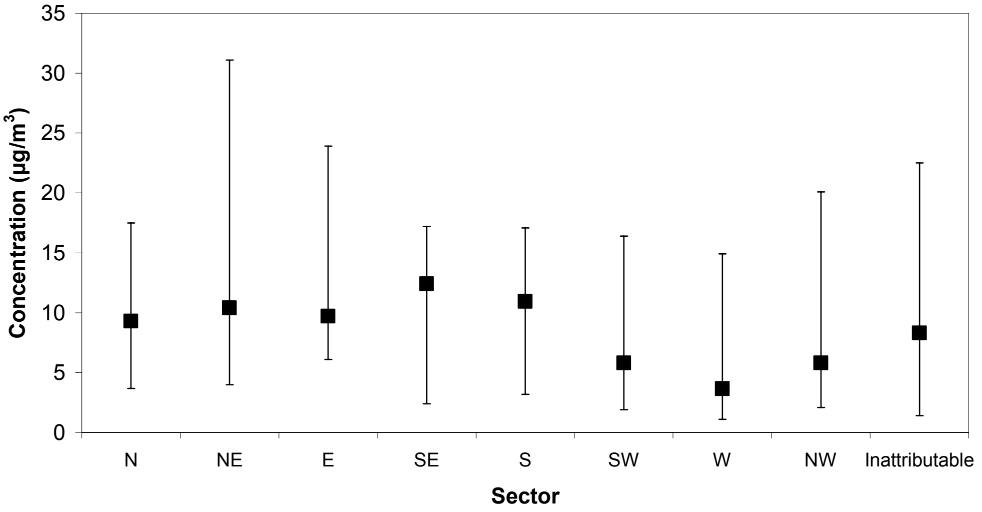


CH0002R - Payerne PM_{10}

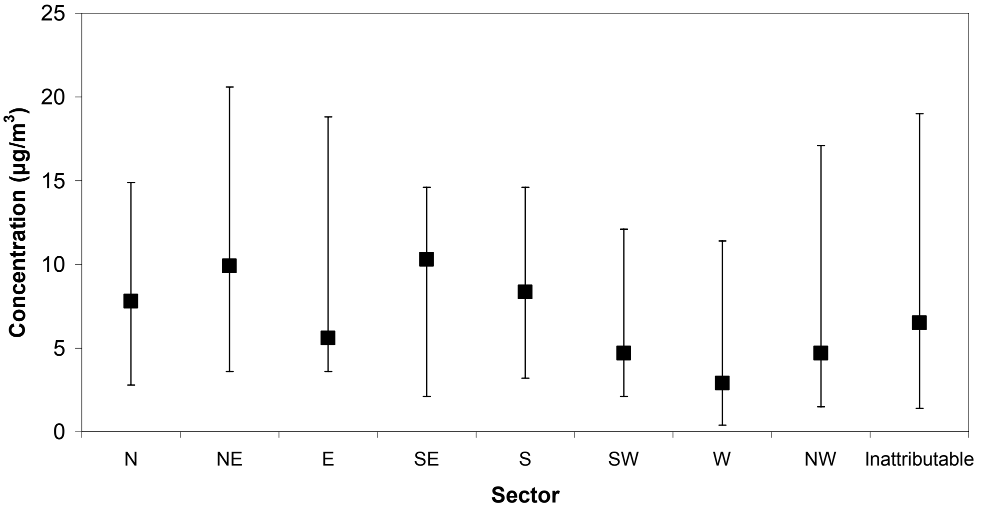

CH0002R - Payerne $PM_{2.5}$


CH0003R - Tanikon PM_{10}

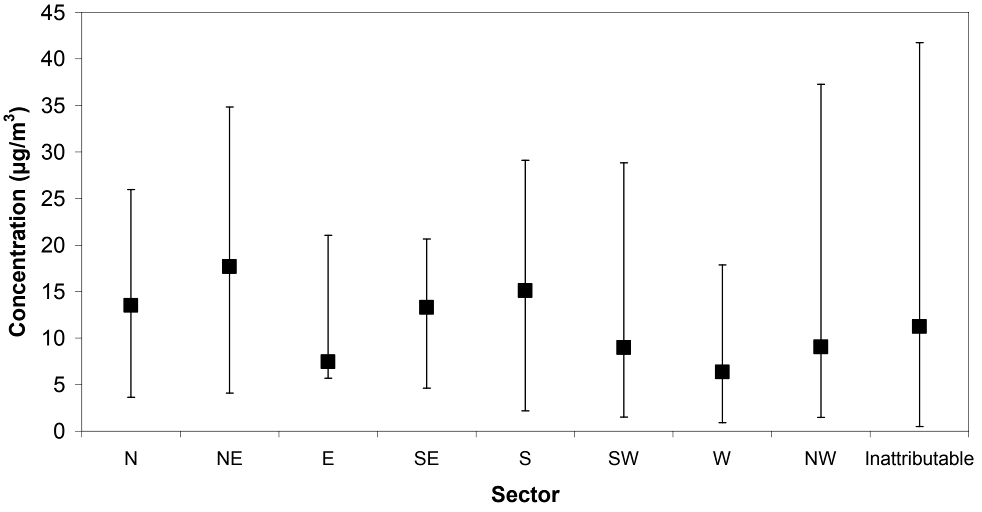

CH0004R - Chaumont PM_{10}



CH0004R - Chaumont $PM_{2.5}$

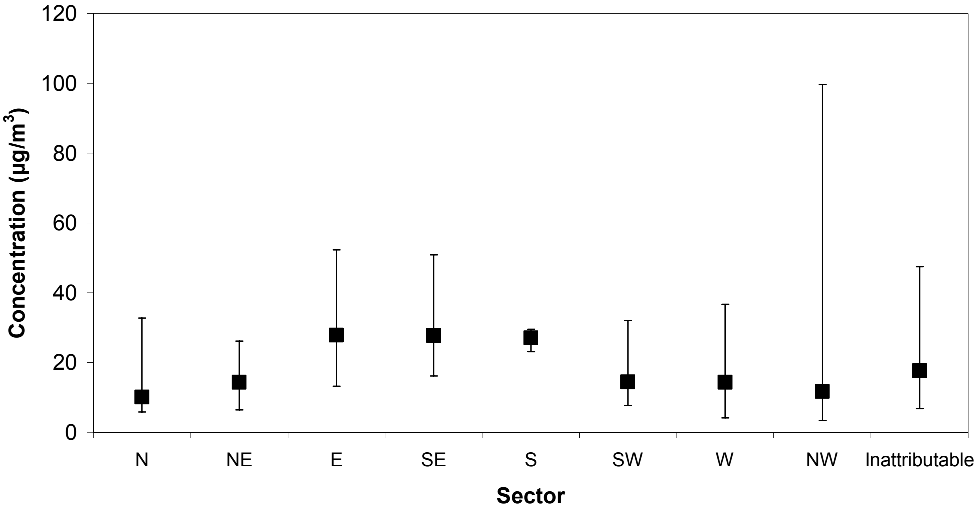


CH0004R - Chaumont PM_1

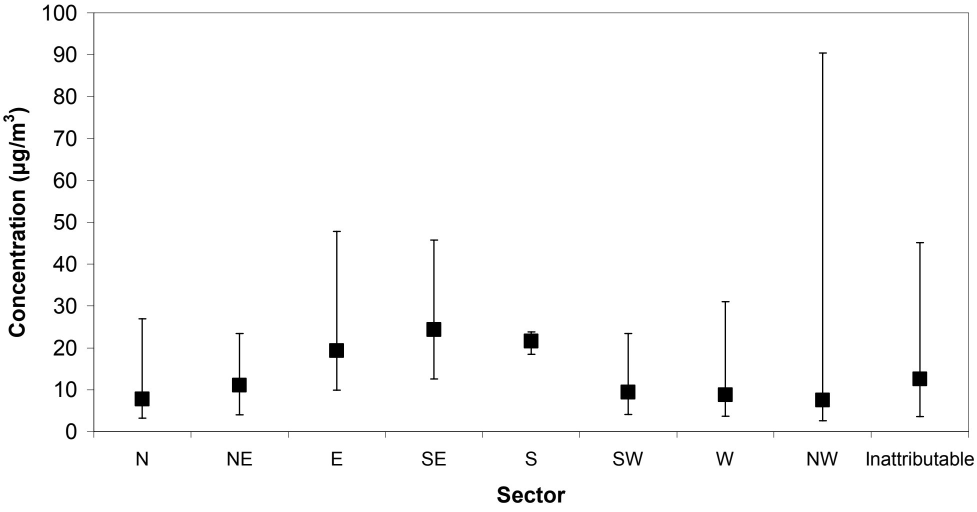


CH0005R - Rigi PM_{10}

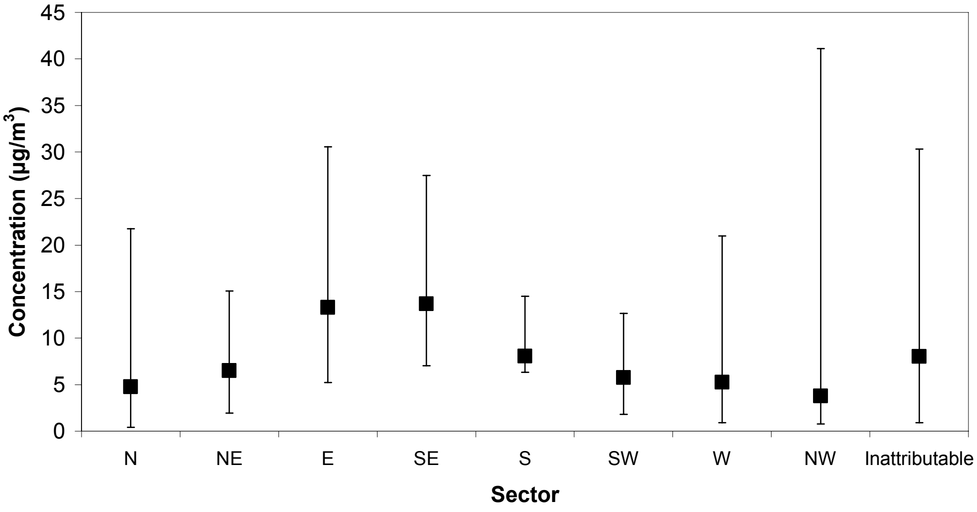
DE0002R - Langenbrugge/Waldhof PM_{10}



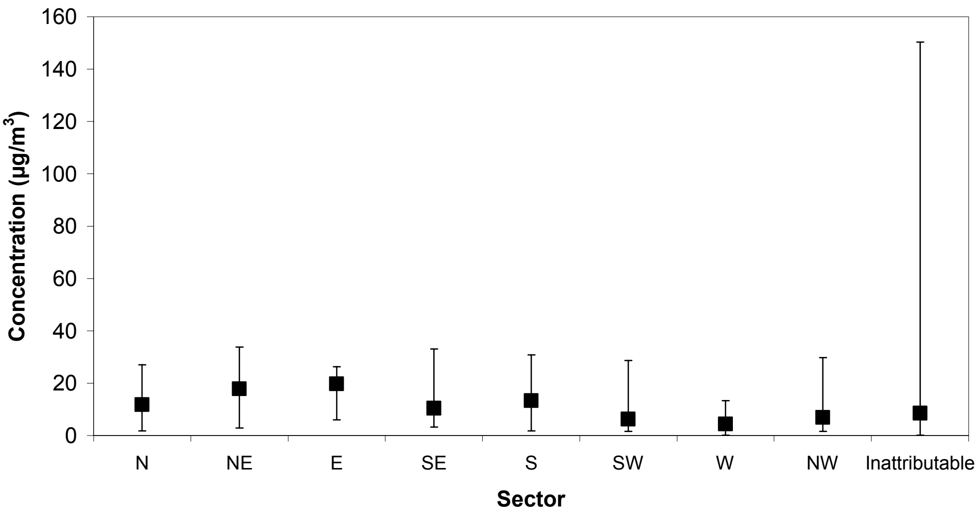
DE0002R - Langenbrugge/Waldhof $PM_{2.5}$



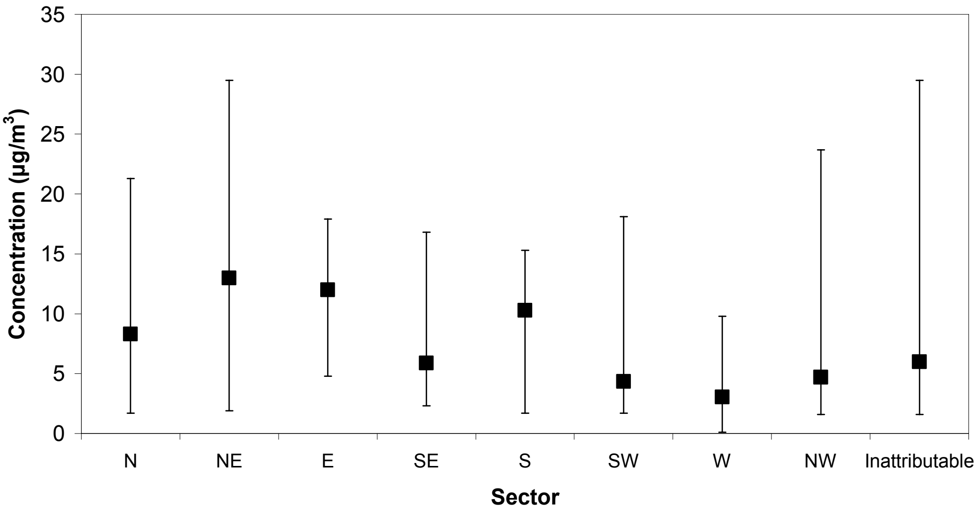
DE0002R - Langenbrugge/Waldhof PM_1



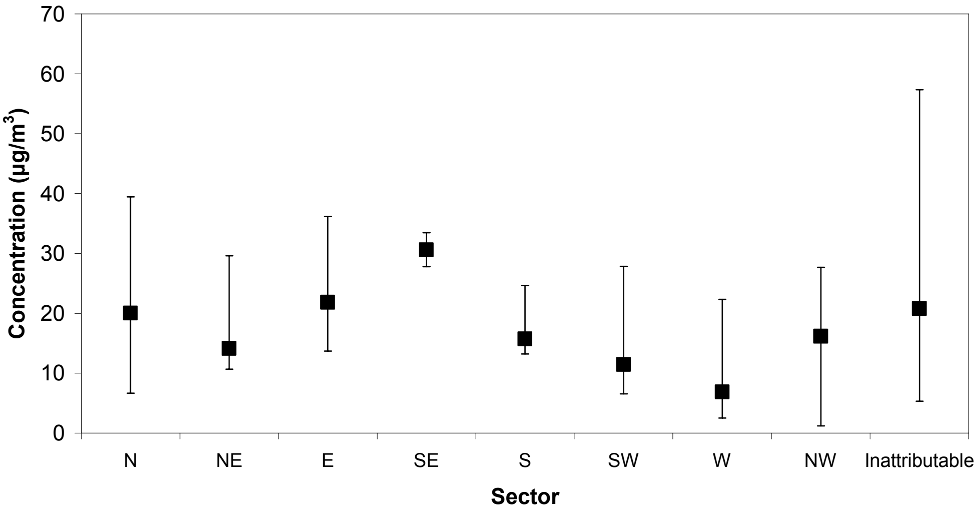
DE0003R - Schauinsland PM_{10}

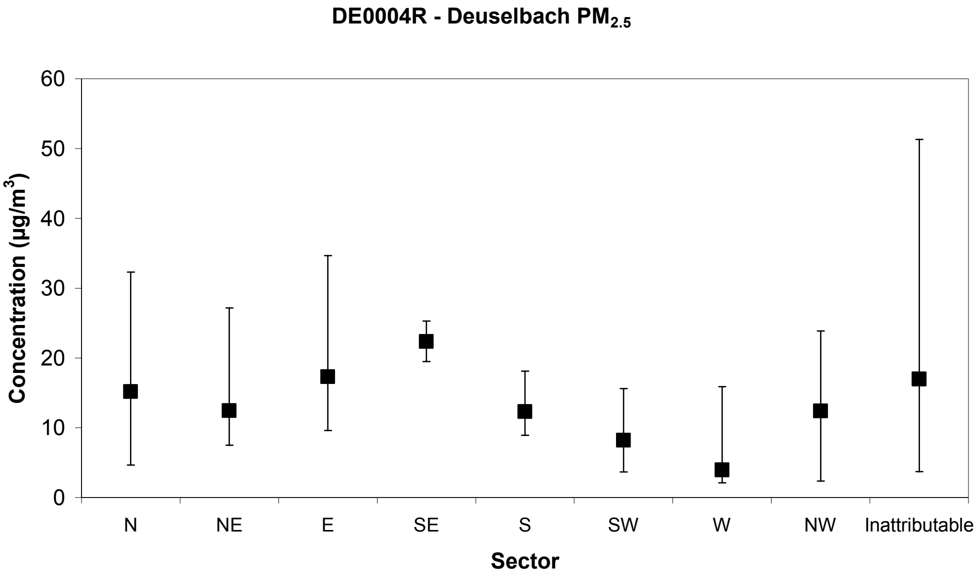


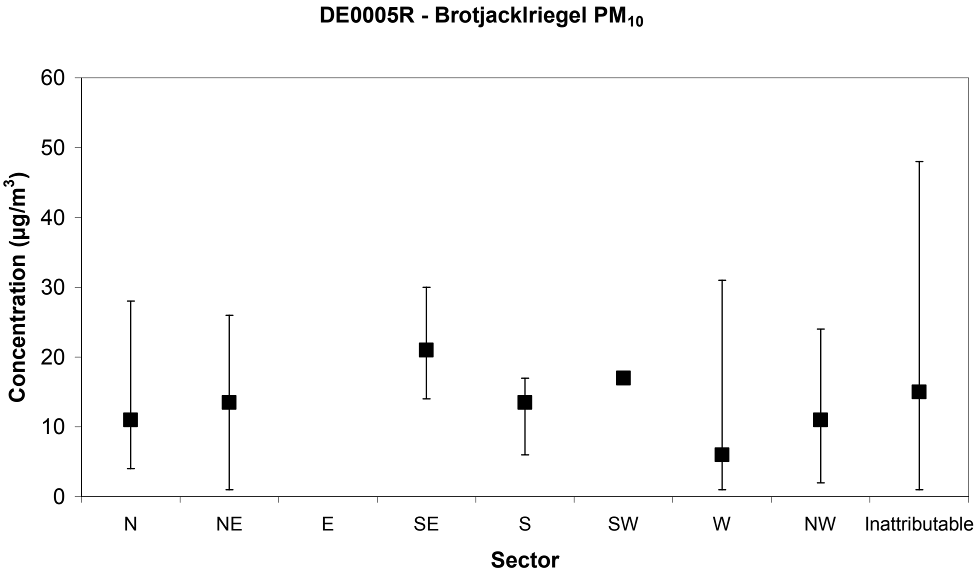
DE0003R - Schauinsland $PM_{2.5}$

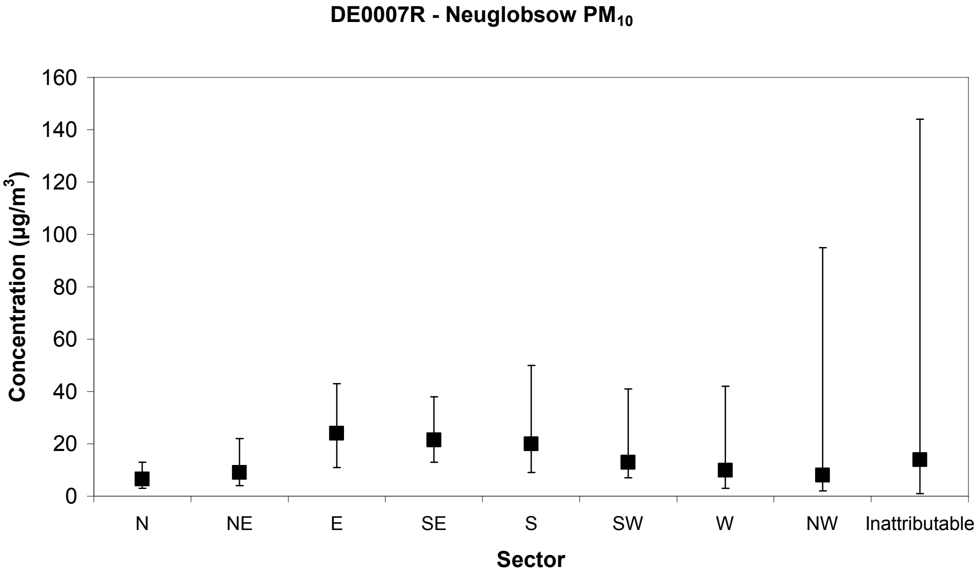


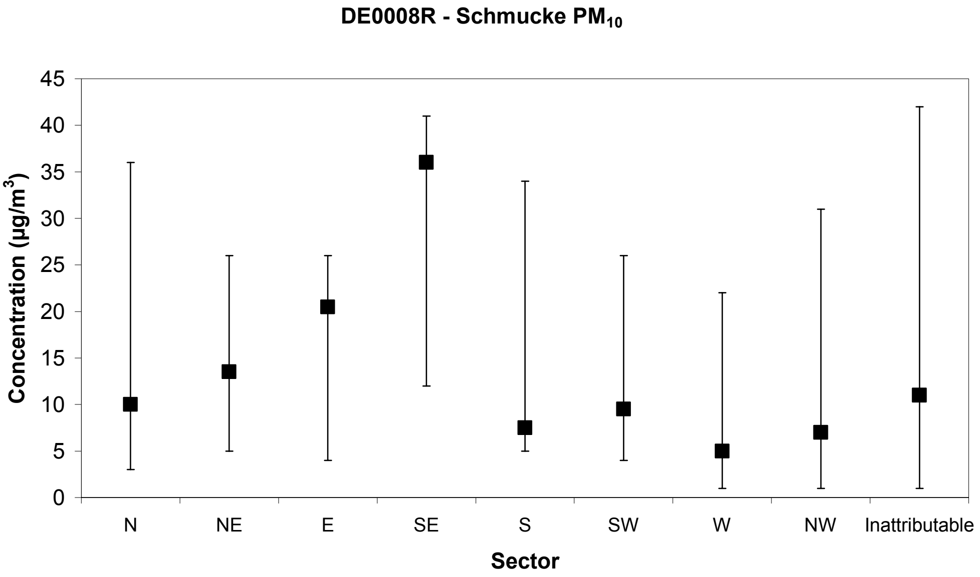
DE0004R - Deuselbach PM_{10}

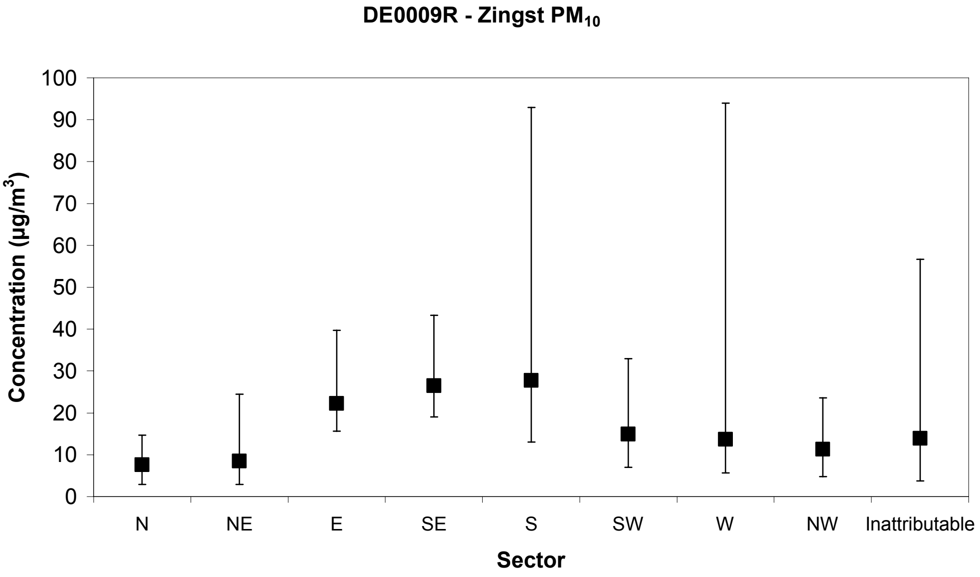


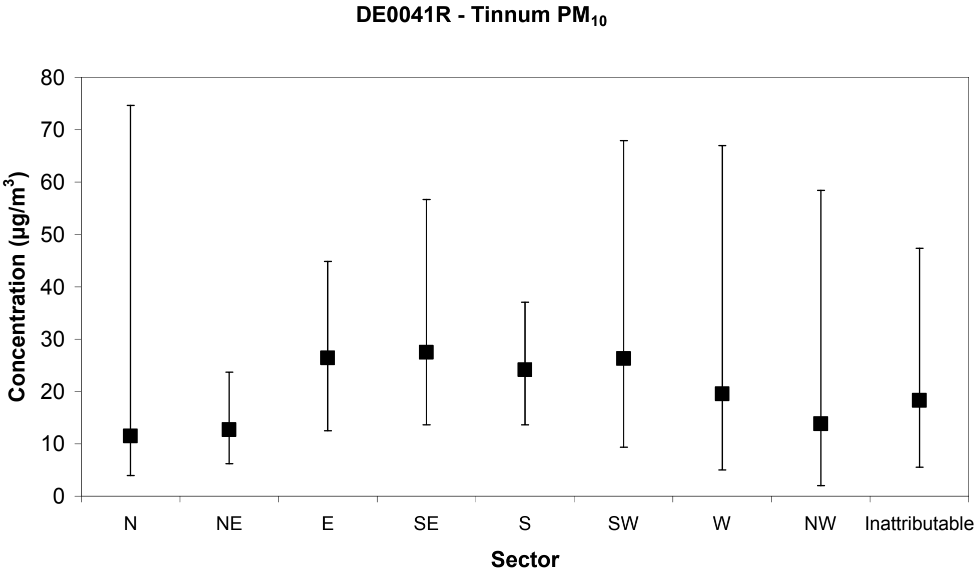

DE0004R - Deuselbach $PM_{2.5}$

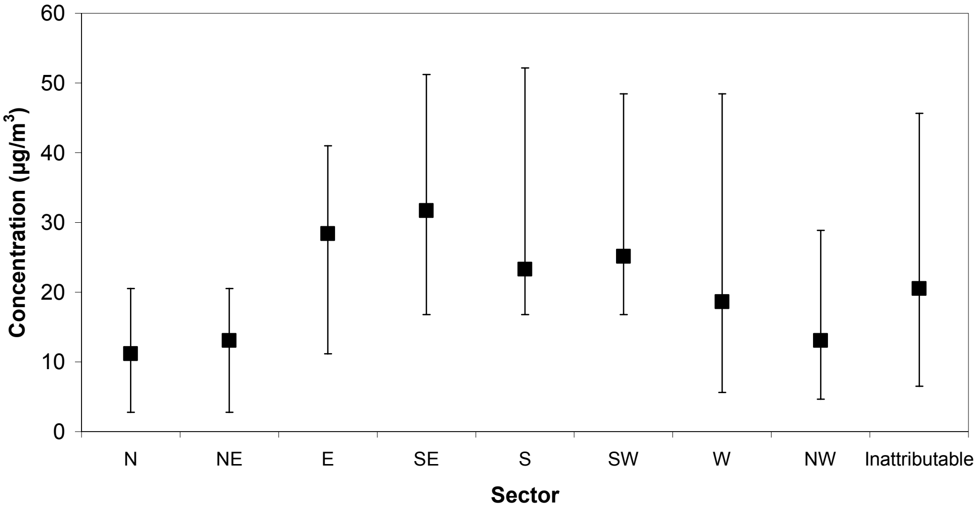

DE0005R - Brotjacklriegel PM_{10}


DE0007R - Neuglobsow PM_{10}

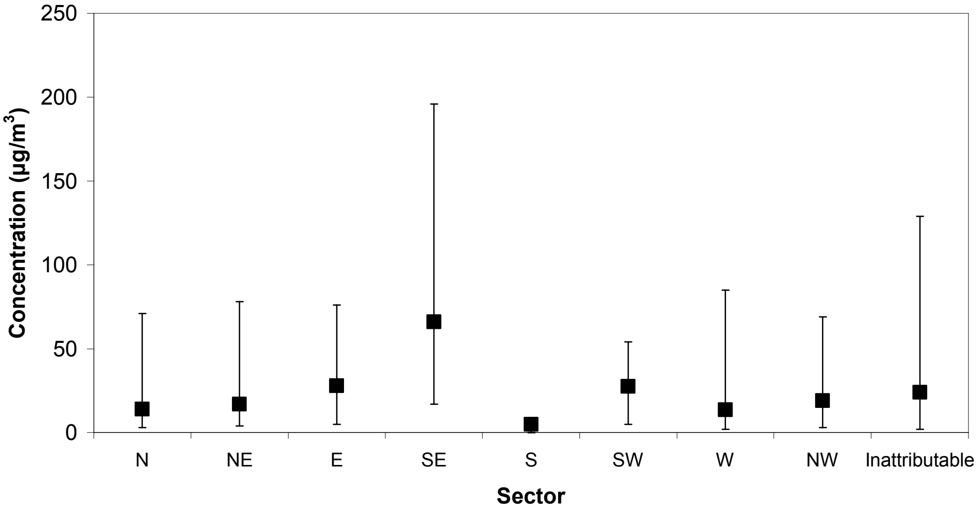

DE0008R - Schmucke PM$_{10}$
Concentration (µg/m^3)
45
40
35
30
25
20
15
10
5
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector


DE0009R - Zingst PM$_{10}$
Concentration (µg/m^3)
100
90
80
70
60
50
40
30
20
10
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

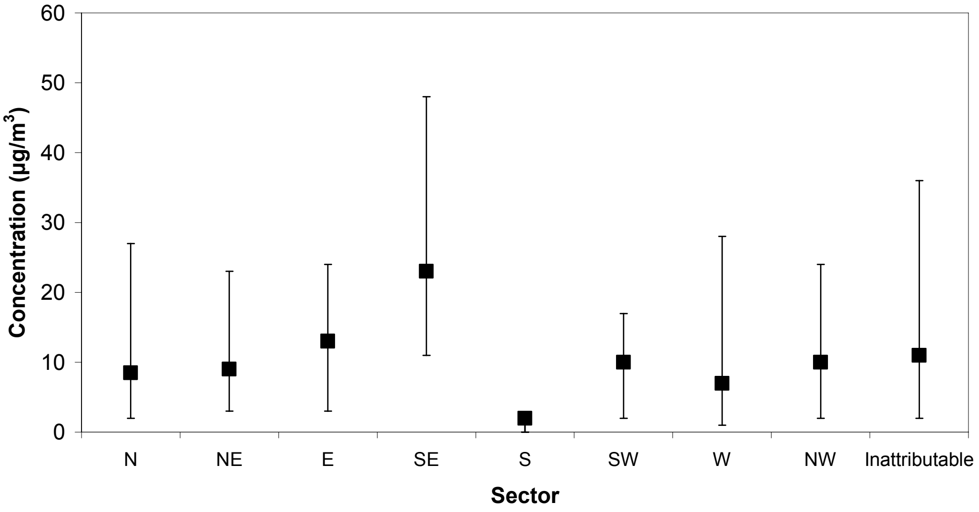

DE0041R - Tinnum PM$_{10}$
Concentration (µg/m^3)
80
70
60
50
40
30
20
10
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector



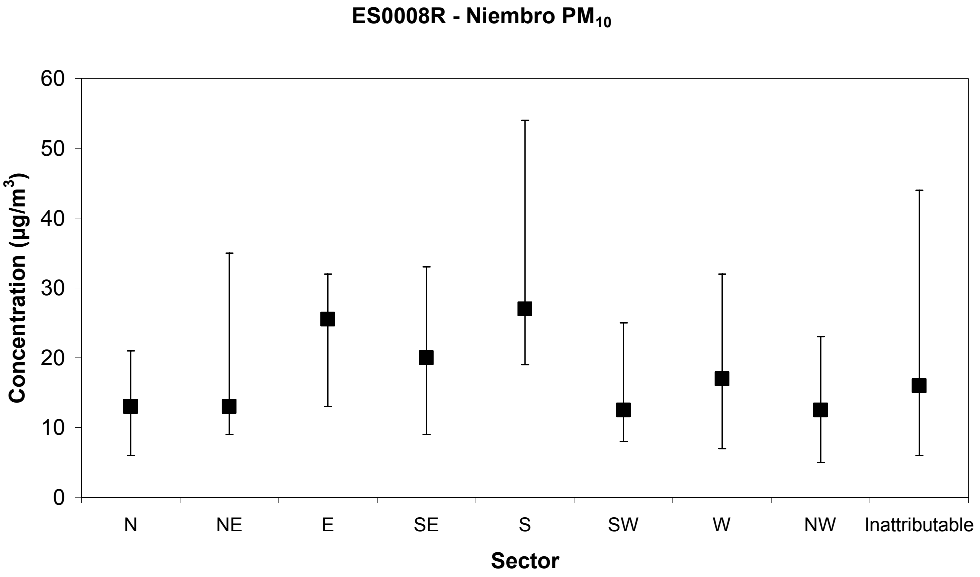
DK0005R - Keldsnor PM_{10}

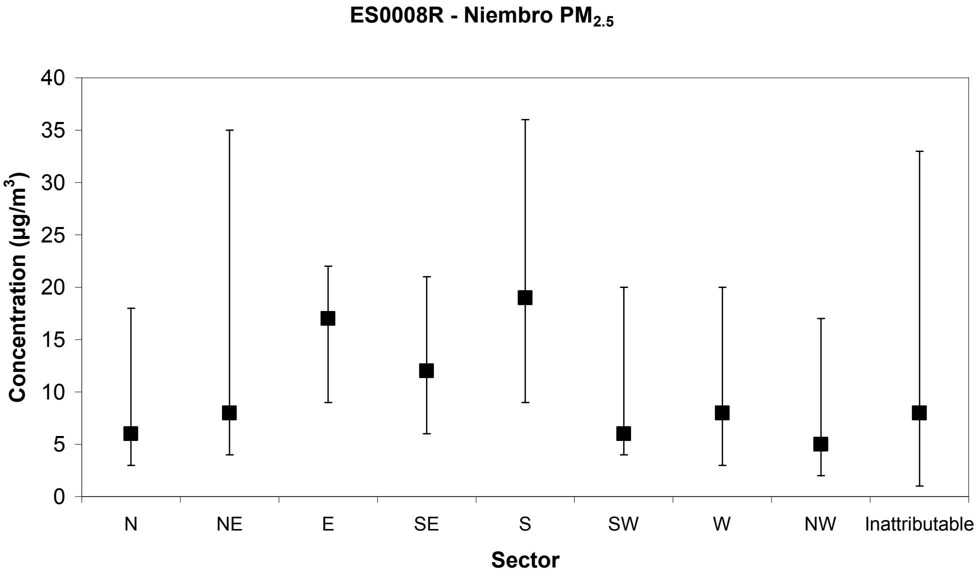


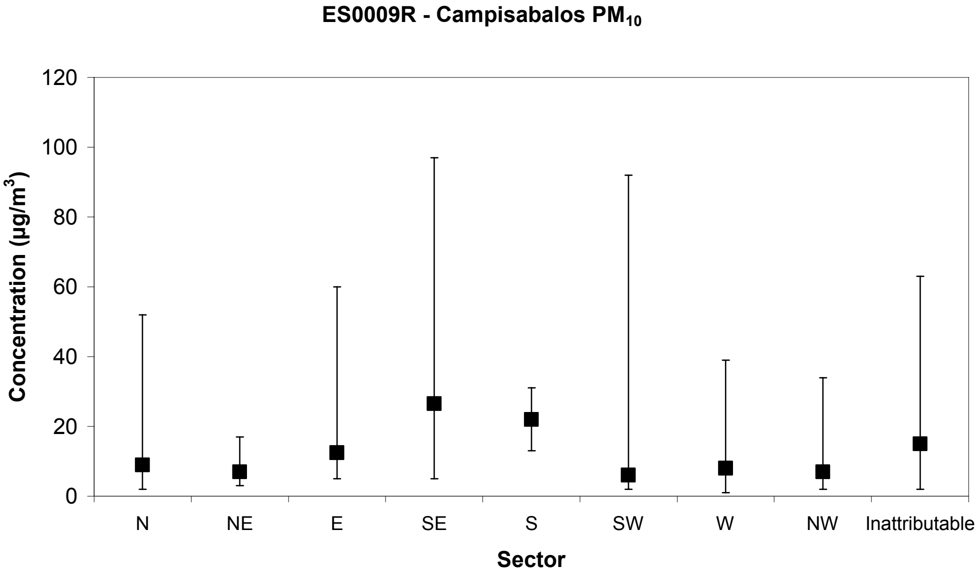
ES0007R - Vvznar PM_{10}

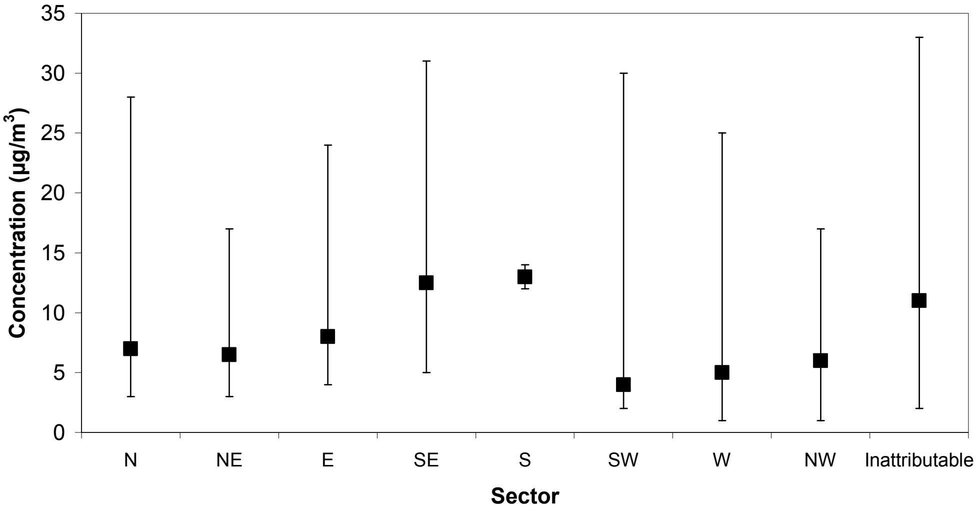


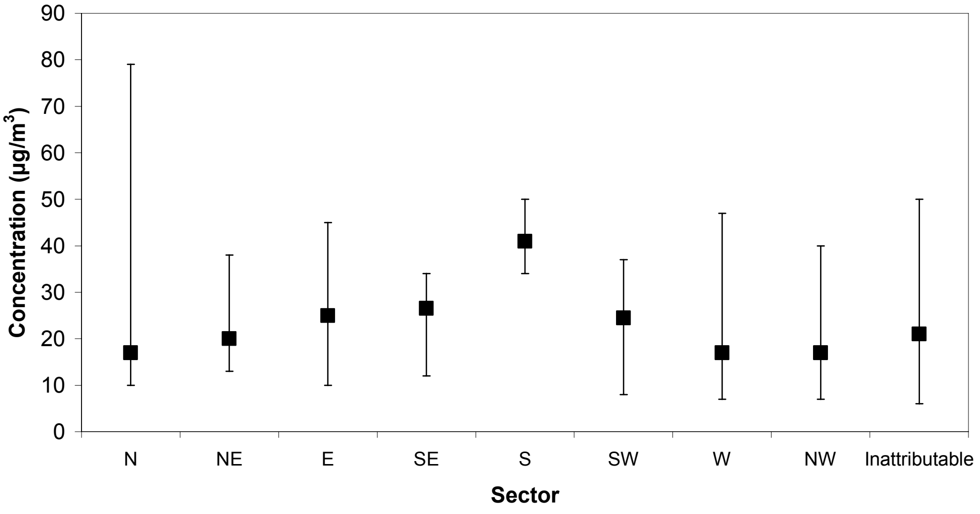
ES0007R - Vvznar PM_{25}

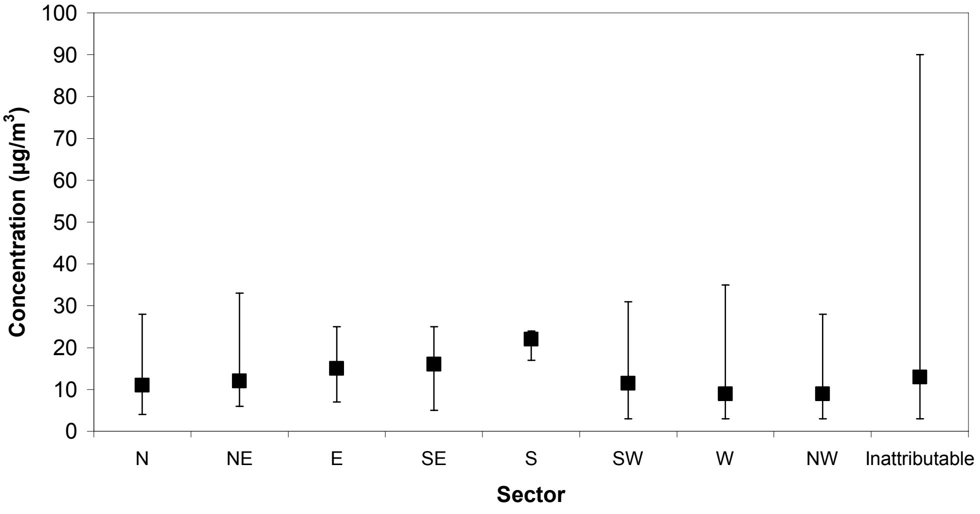

ES0008R - Niembro PM_{10}
Concentration (µg/m^3)
0
10
20
30
40
50
60
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

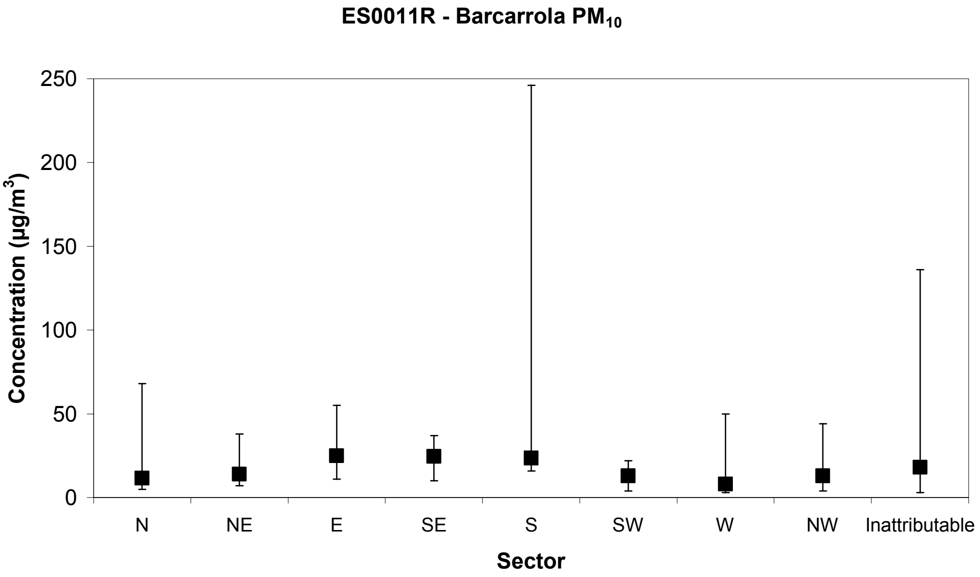

ES0008R - Niembro $PM_{2.5}$
Concentration (µg/m^3)
0
5
10
15
20
25
30
35
40
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

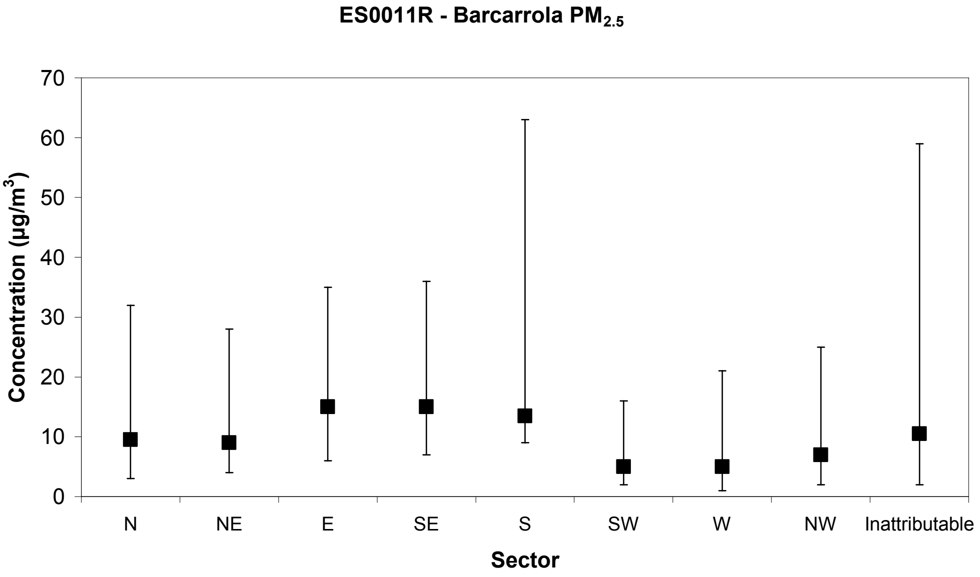

ES0009R - Campisabalos PM_{10}
Concentration (µg/m^3)
0
20
40
60
80
100
120
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

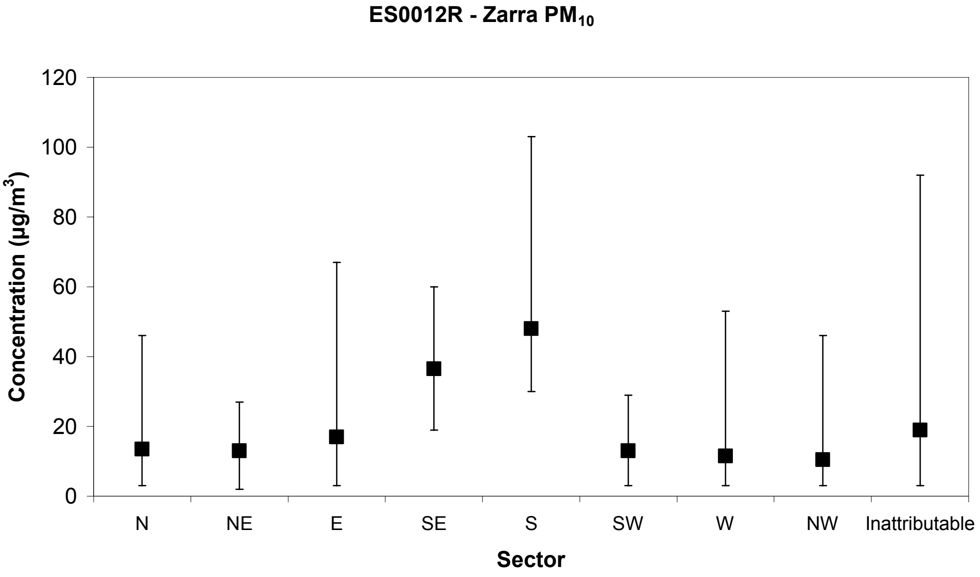

ES0009R - Campisabalos $PM_{2.5}$


ES0010R - Cabo de Creus PM_{10}

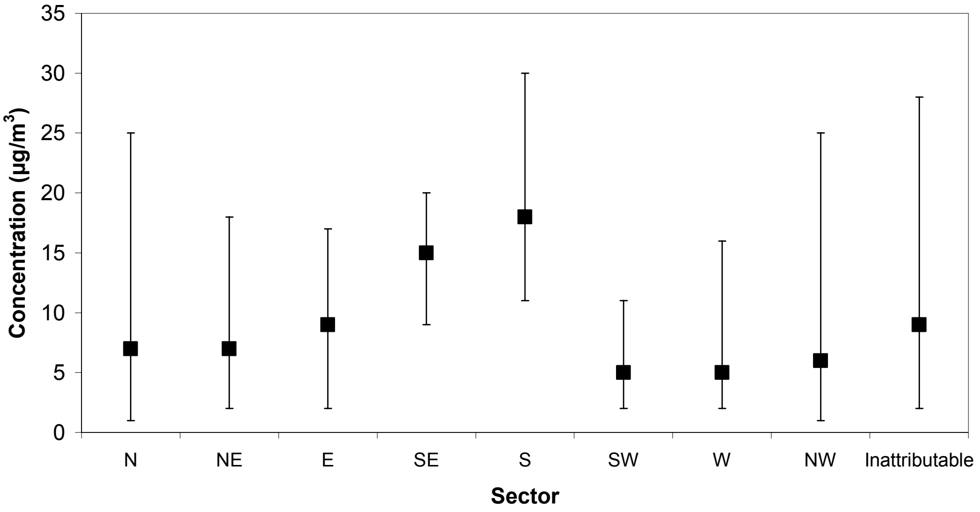

ES0010R - Cabo de Creus $PM_{2.5}$


ES0011R - Barcarrola PM$_{10}$
Concentration (µg/m^3)
250
200
150
100
50
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

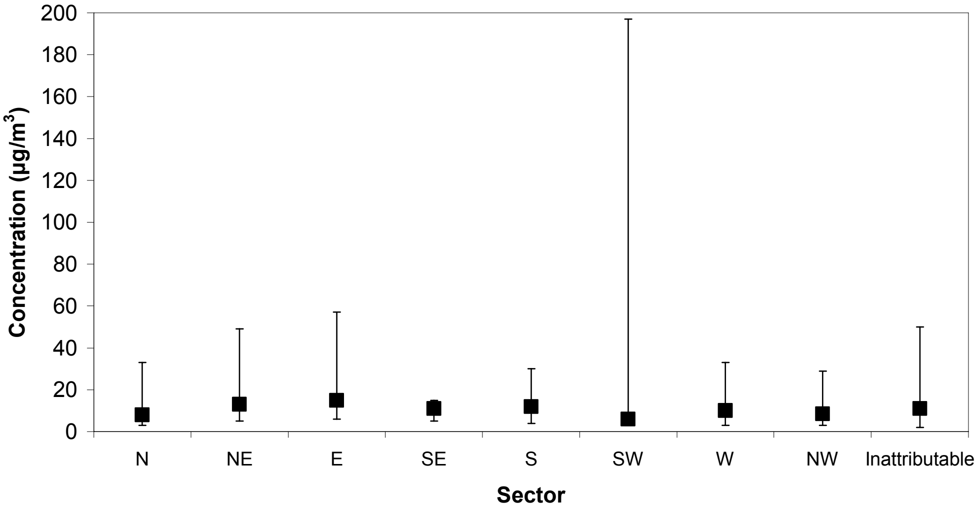

ES0011R - Barcarrola PM$_{2.5}$
Concentration (µg/m^3)
70
60
50
40
30
20
10
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector


ES0012R - Zarra PM$_{10}$
Concentration (µg/m^3)
120
100
80
60
40
20
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

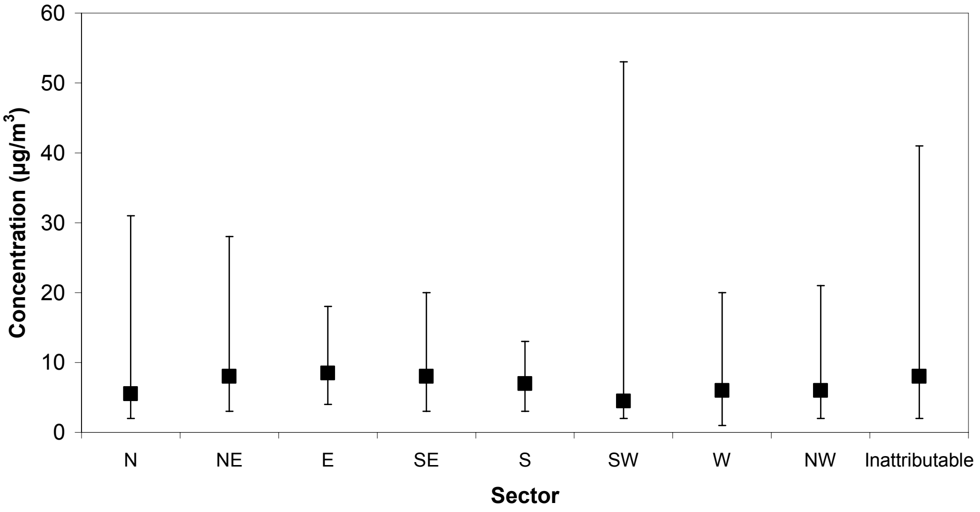
ES0012R - Zarra $PM_{2.5}$

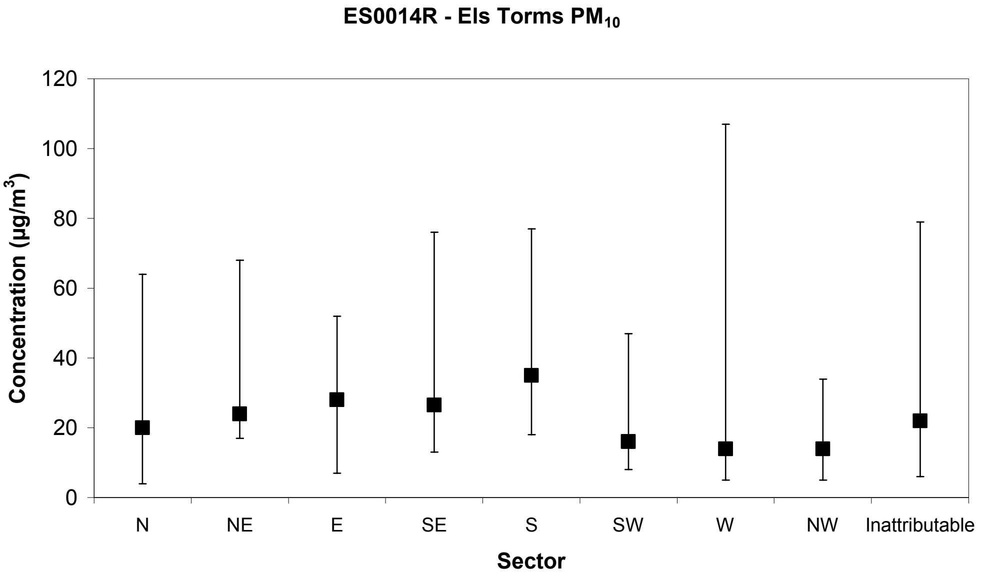


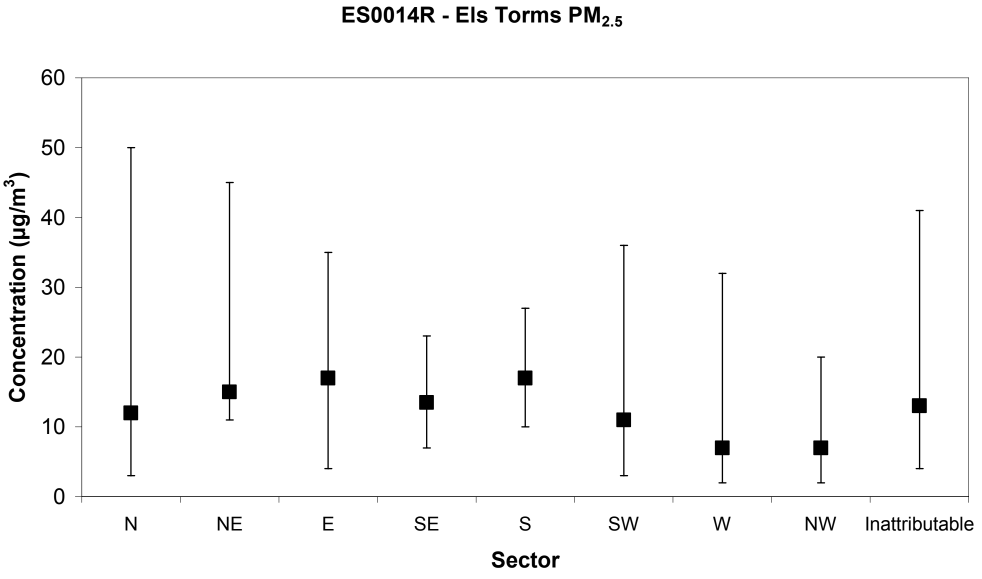
ES0013R - Penausende PM_{10}

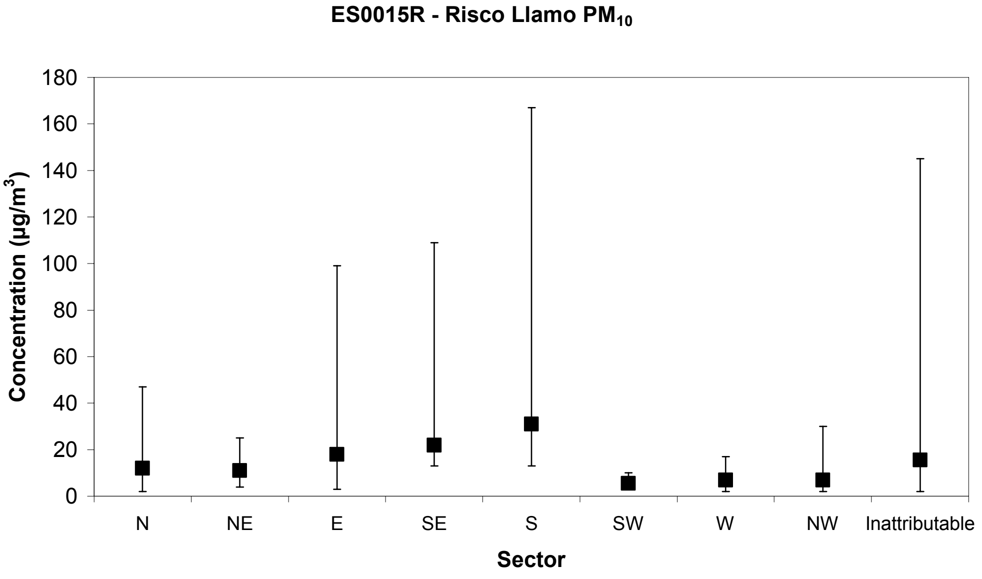


ES0013R - Penausende $PM_{2.5}$

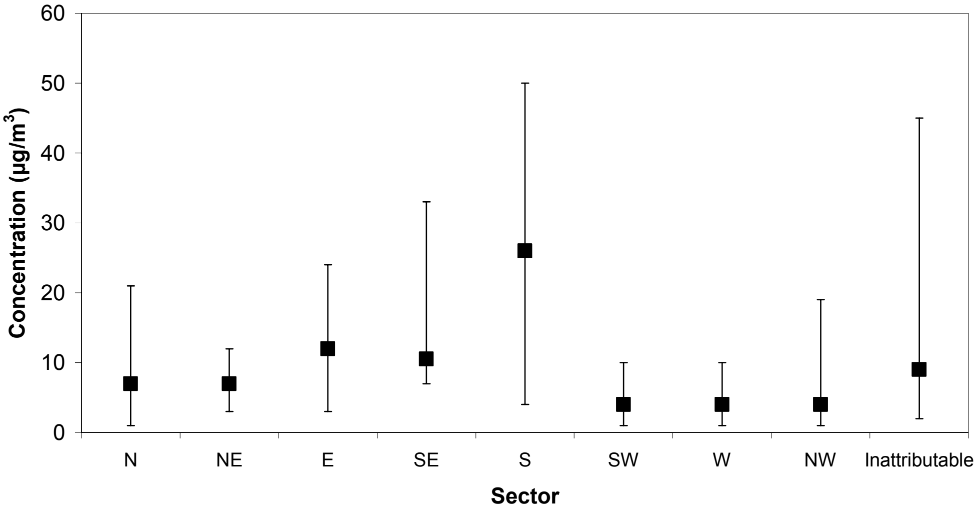



ES0014R - Els Torms PM_{10}
Concentration (µg/m^3)
120
100
80
60
40
20
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

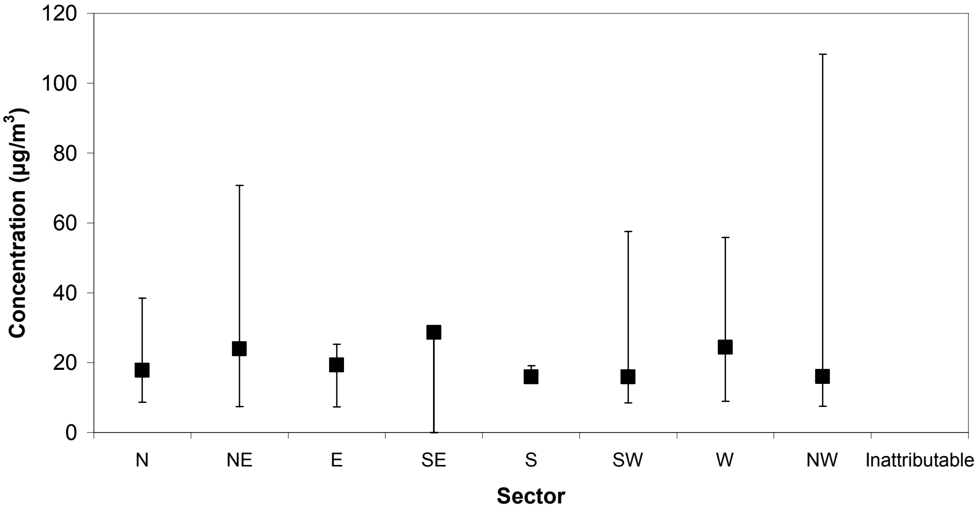

ES0014R - Els Torms $PM_{2.5}$
Concentration (µg/m^3)
60
50
40
30
20
10
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector


ES0015R - Risco Llamo PM_{10}
Concentration (µg/m^3)
180
160
140
120
100
80
60
40
20
0
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

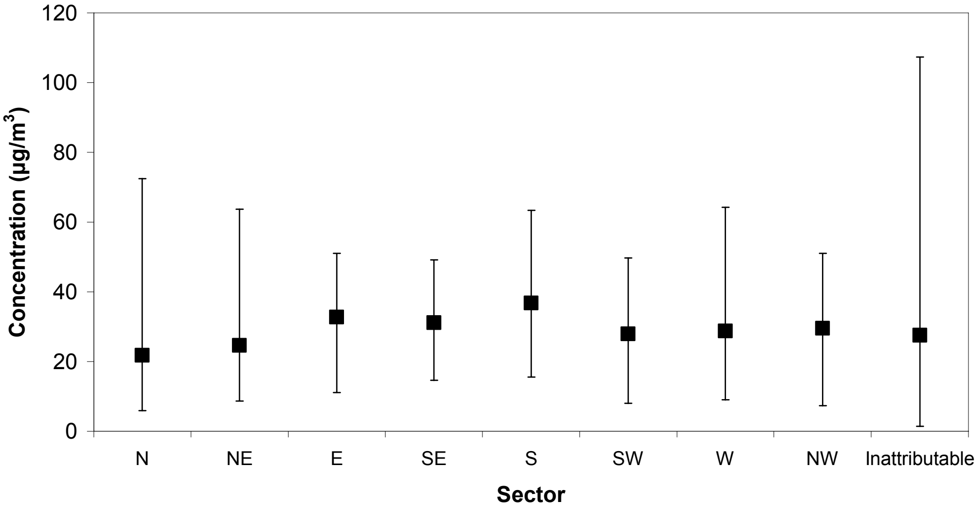
ES0015R - Risco Llamo $PM_{2.5}$

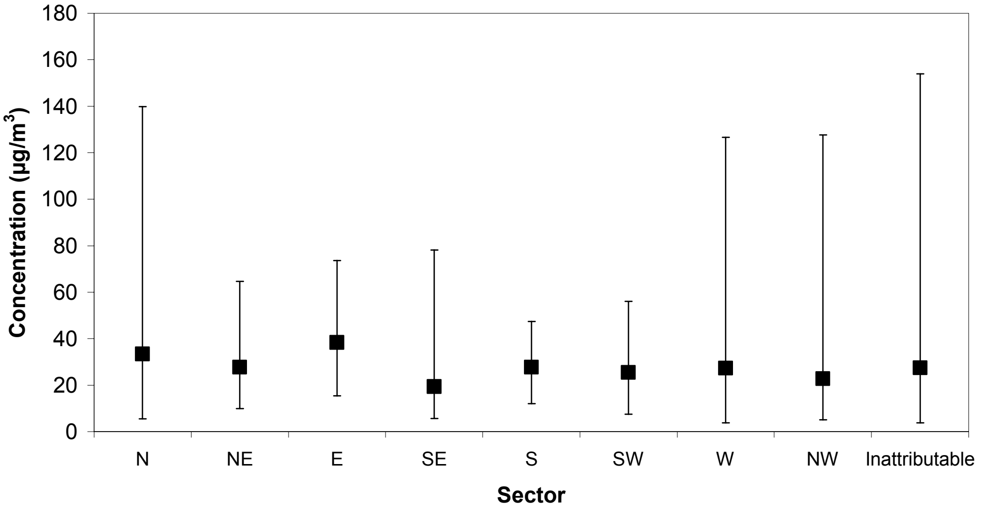


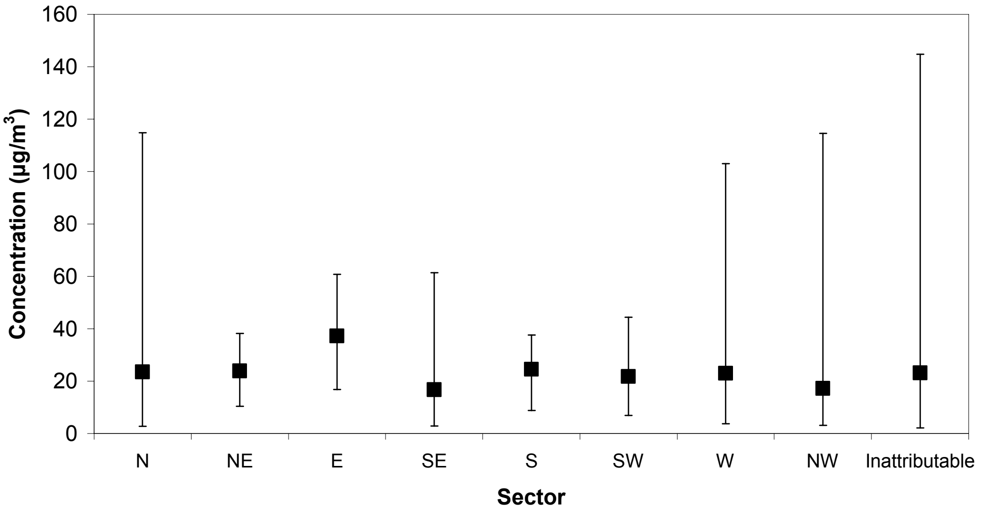
GR0002R - Finokalia PM_{10}

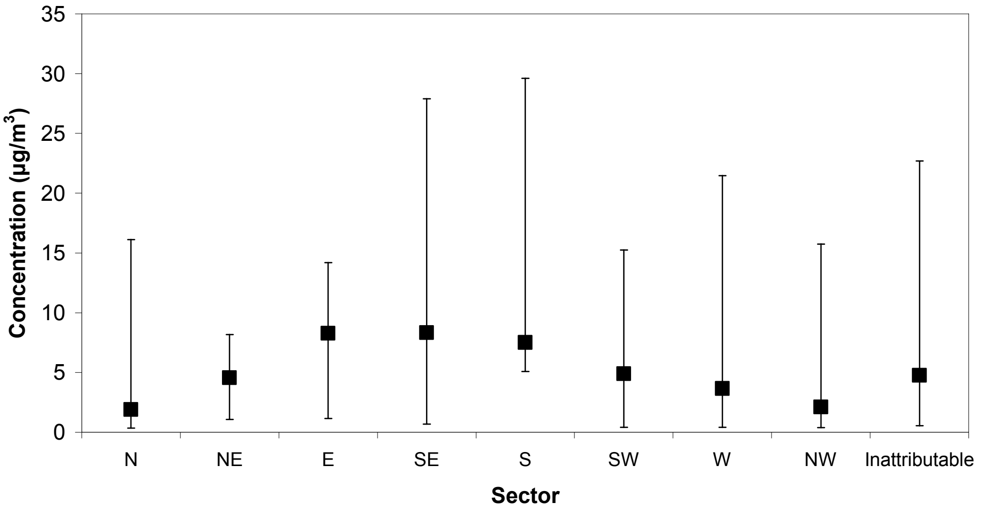


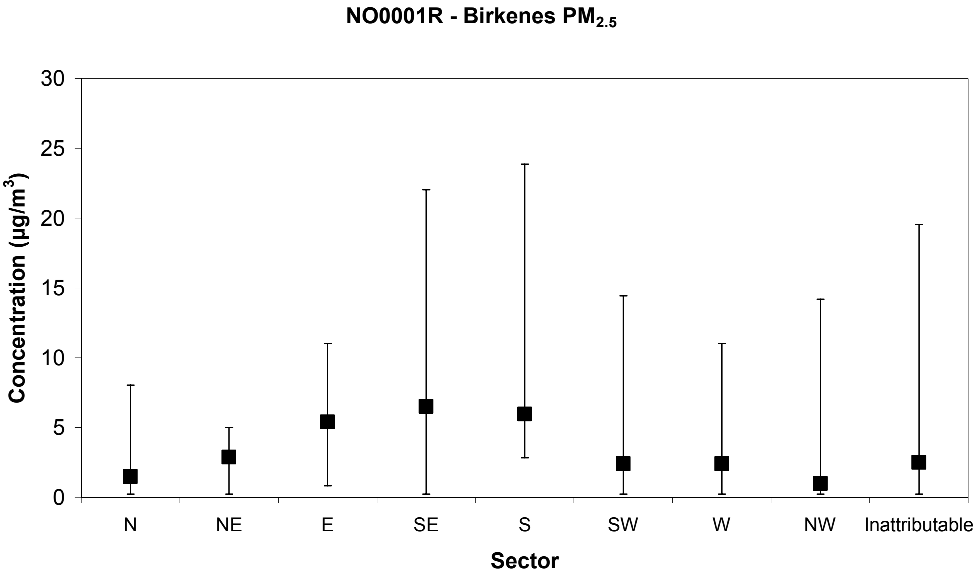
IT0001R - Montelibretti PM_{10}

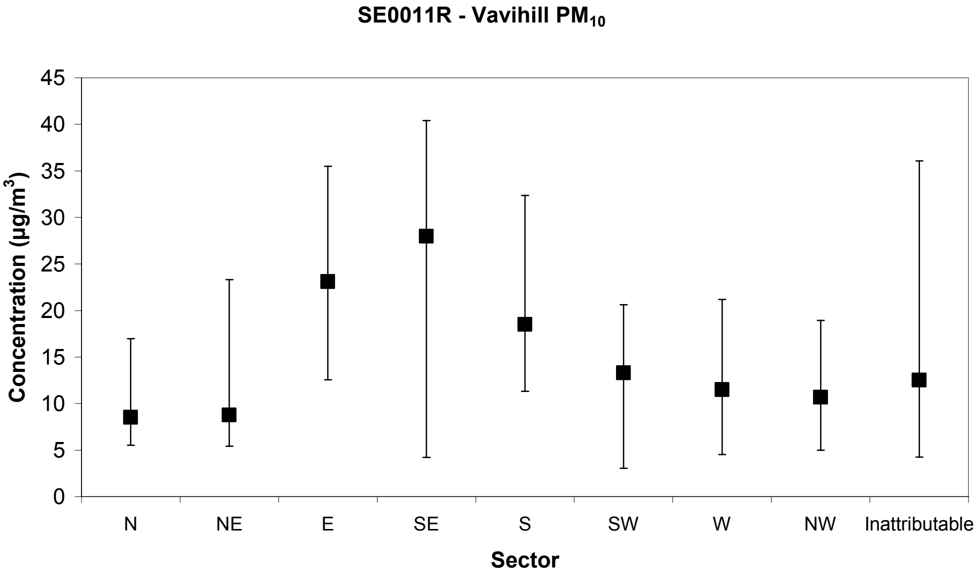


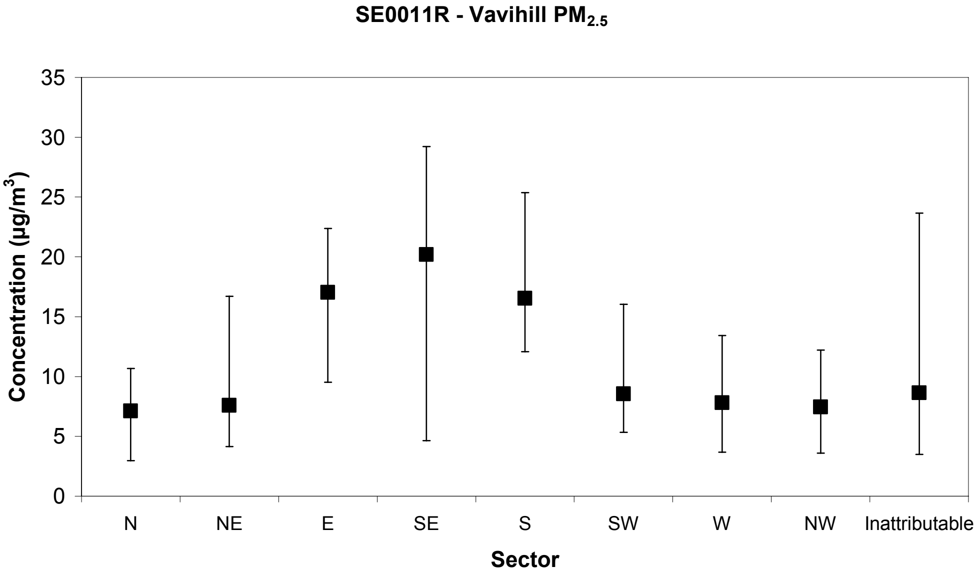

IT0004R - Ispra PM_{10}

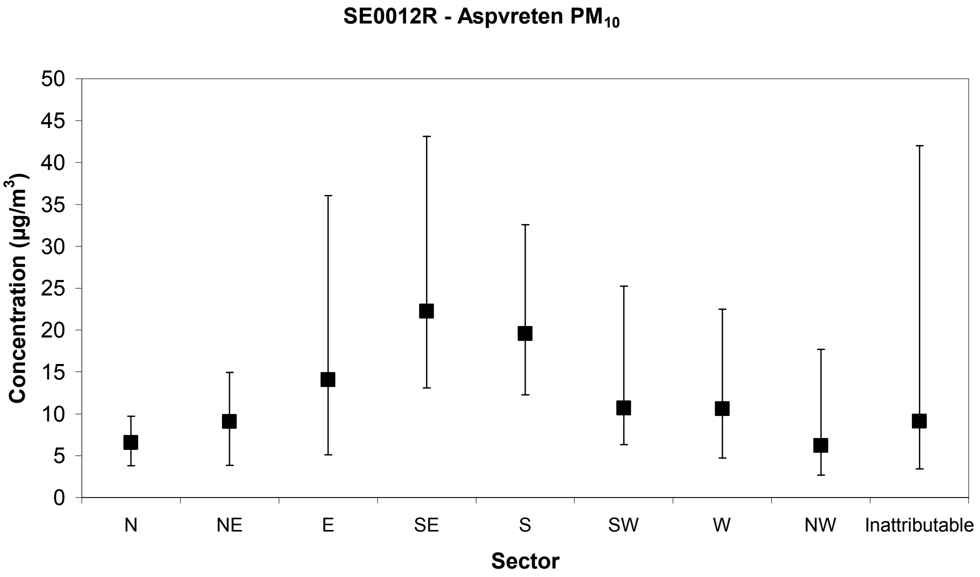

IT0004R - Ispra $PM_{2.5}$

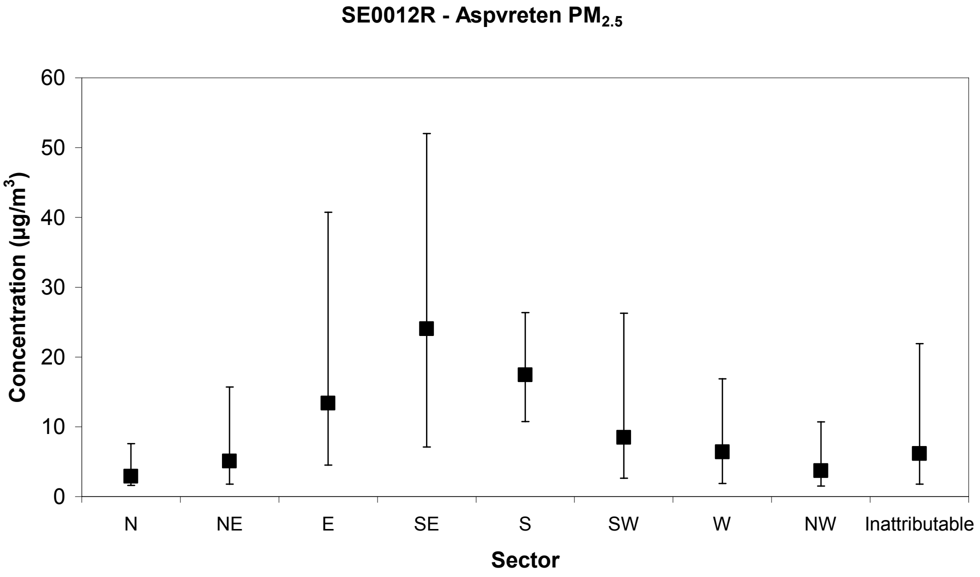

NO0001R - Birkenes PM_{10}

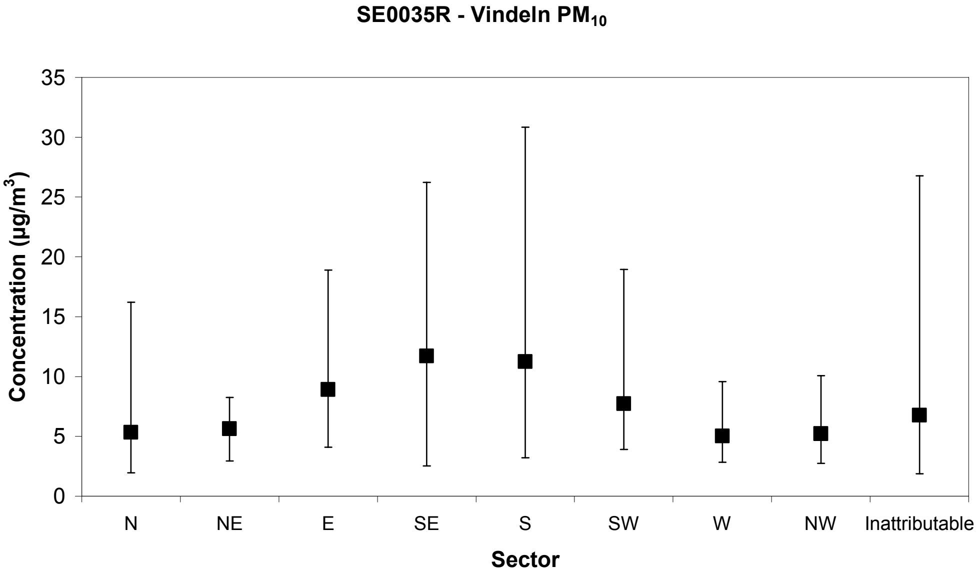

NO0001R - Birkenes $PM_{2.5}$

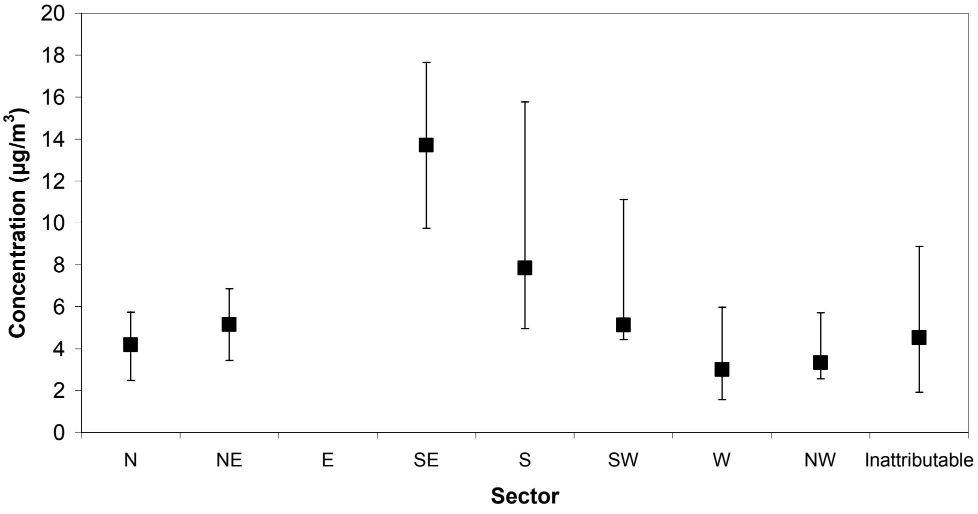

SE0011R - Vavihill PM_{10}


SE0011R - Vavihill $PM_{2.5}$

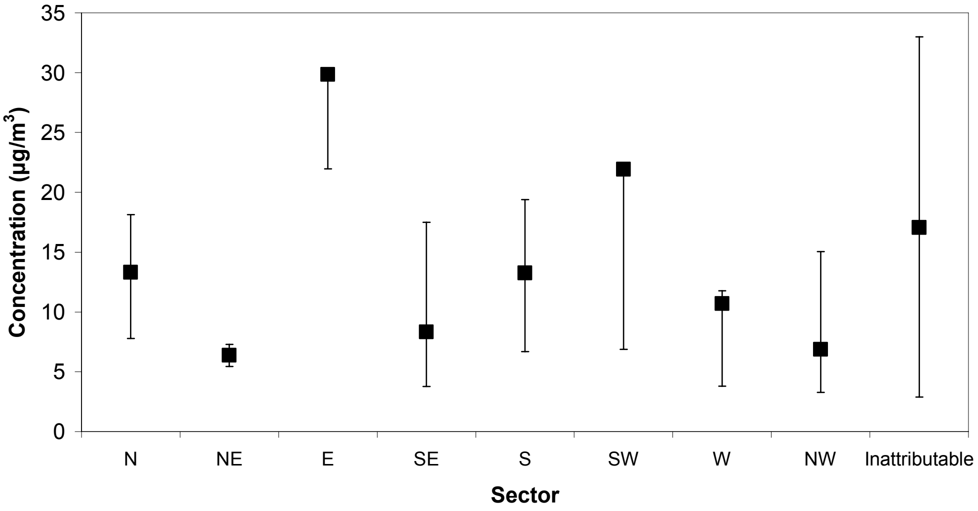

SE0012R - Aspvreten PM$_{10}$
Concentration (µg/m^3)
0
5
10
15
20
25
30
35
40
45
50
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector


SE0012R - Aspvreten PM$_{2.5}$
Concentration (µg/m^3)
0
10
20
30
40
50
60
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector

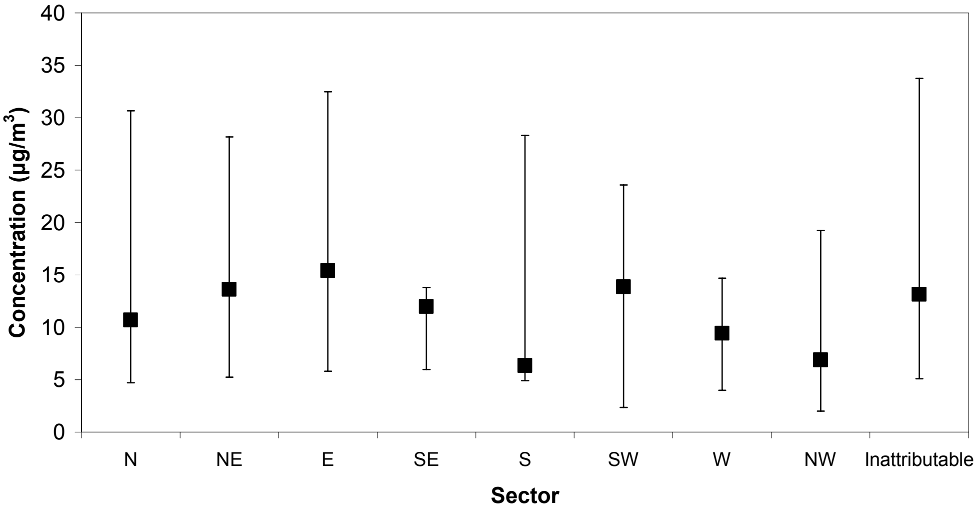

SE0035R - Vindeln PM$_{10}$
Concentration (µg/m^3)
0
5
10
15
20
25
30
35
N
NE
E
SE
S
SW
W
NW
Inattributable
Sector



SE0035R - Vindeln $PM_{2.5}$



SI0008R - Iskrba PM_{10}



SI0008R - Iskrba PM_{10}

Annex 5

Data on PM reported to EMEP and partly AIRBASE during 2004

		K^+	Na^+	Ca^{2+}	Mg^{2+}	Cl^-	NO_3^-	$HNO_{3\,(g)}$	$NO_3^- + HNO_{3\,(g)}$	SO_4^{2-} (t)	SO_4^{2-} (cor.)	NH_4^+	$NH_4^+ + NH_{3\,(g)}$	TSP	PM_{10}	$PM_{2.5}$	PM_1
Austria	AT0002	✓	✓	✓	✓		✓	✓		✓		✓			✓	✓	✓
	AT0004														✓		
	AT0005														✓		
	AT0030														✓#		
	AT0040														✓#		
	AT0042														✓#		
	AT0043														✓#		
	AT0047														✓#		
	AT0048														✓	✓	
Belgium	BE0033														✓#		
Switzerland	CH0001									✓				✓			
	CH0002								✓	✓			✓		✓	✓	
	CH0003														✓		
	CH0004														✓	✓	✓
	CH0005								✓	✓			✓		✓		
Cyprus	CY0002														✓		
Czech Republic	CZ0001								✓	✓			✓		✓#		
	CZ0003								✓	✓			✓		✓#		
Germany	DE0001								✓	✓			✓				
	DE0002														✓	✓	✓
	DE0003								✓	✓			✓		✓	✓	
	DE0004								✓	✓			✓		✓	✓	
	DE0005														✓		
	DE0007								✓	✓			✓		✓		
	DE0008														✓		
	DE0009								✓	✓			✓		✓		
	DE0012														✓#		
	DE0016														✓#		
	DE0017														✓#		
	DE0026														✓#		
	DE0035														✓#		
	DE0039														✓#		
	DE0041								✓	✓			✓		✓		

		K^+	Na^+	Ca^{2+}	Mg^{2+}	Cl^-	NO_3^-	$HNO_{3\,(g)}$	$NO_3^- + HNO_{3\,(g)}$	SO_4^{2-} (t)	SO_4^{2-} (cor.)	NH_4^+	$NH_4^+ + NH_{3\,(g)}$	TSP	PM_{10}	$PM_{2.5}$	PM_1
Denmark	DK0003		✓						✓	✓			✓				
	DK0005														✓		
	DK0008		✓						✓	✓			✓				
	DK0020														✓		
	DK0041														✓#		
Spain	ES0001														✓		
	ES0007						✓		✓	✓			✓		✓	✓	
	ES0008						✓		✓	✓			✓		✓	✓	
	ES0009						✓		✓	✓			✓		✓	✓	
	ES0010						✓		✓	✓			✓		✓	✓	
	ES0011						✓		✓	✓			✓		✓	✓	
	ES0012						✓		✓	✓			✓		✓	✓	
	ES0013						✓		✓	✓			✓		✓	✓	
	ES0014						✓		✓	✓			✓		✓	✓	
	ES0015						✓		✓	✓			✓		✓	✓	
	ES0016						✓		✓	✓			✓		✓	✓	
Finland	FI0007														✓#		
	FI0009								✓	✓			✓				
	FI0017								✓	✓			✓				
	FI0022								✓	✓			✓				
	FI0037								✓	✓			✓				
France	FR0008									✓							
	FR0009									✓							
	FR0010									✓							
	FR0012									✓							
	FR0013									✓							
	FR0014									✓							
	FR0015									✓							
	FR0016									✓							
	FR0017									✓							
Great Britain	GB0002									✓							
	GB0006									✓					✓#		
	GB0007									✓							
	GB0013									✓							
	GB0014									✓							
	GB0036														✓#		
	GB0043														✓#		

		K^+	Na^+	Ca^{2+}	Mg^{2+}	Cl^-	NO_3^-	$HNO_{3\,(g)}$	$NO_3^- + HNO_{3\,(g)}$	SO_4^{2-} (t)	SO_4^{2-} (cor.)	NH_4^+	$NH_4^+ + NH_{3\,(g)}$	TSP	PM_{10}	$PM_{2.5}$	PM_1
Greece	GR0002														✓		
Hungary	HU0002						✓	✓		✓		✓					
Ireland	IE0001	✓	✓	✓	✓				✓	✓			✓				
Iceland	IS0002									✓							
	IS0091					✓	✓			✓							
Italy	IT0001						✓	✓		✓		✓			✓		
	IT0004						✓			✓		✓			✓	✓	
Lithuania	LT0015								✓	✓			✓	✓			
Latvia	LV0010						✓		✓	✓		✓	✓				
	LV0016						✓		✓	✓		✓	✓				
F.Y.R.O.M.	MK0007														✓#		
Netherlands	NL0007														✓#		
	NL0009											✓			✓#		
	NL0010														✓#		
Norway	NO0001	✓	✓		✓	✓	✓	✓	✓	✓	✓	✓	✓		✓	✓	
	NO0008	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓				
	NO0015	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓				
	NO0039	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓				
	NO0042	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓				
	NO0055	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓				
Poland	PL0002						✓		✓	✓		✓	✓				
	PL0003						✓		✓	✓		✓	✓				
	PL0004						✓		✓	✓		✓	✓				
	PL0005								✓	✓			✓				

		K^+	Na^+	Ca^{2+}	Mg^{2+}	Cl^-	NO_3^-	$HNO_{3\,(g)}$	$NO_3^- + HNO_{3\,(g)}$	SO_4^{2-} (t)	SO_4^{2-} (cor.)	NH_4^+	$NH_4^+ + NH_{3\,(g)}$	TSP	PM_{10}	$PM_{2.5}$	PM_1
Russia	RU0001						✓			✓		✓					
	RU0016						✓			✓		✓					
	RU0017						✓			✓		✓					
Sweden	SE0005								✓	✓			✓	✓			
	SE0008									✓				✓			
	SE0011								✓	✓			✓	✓	✓	✓	
	SE0012														✓	✓	
	SE0014								✓	✓			✓	✓			
	SE0035														✓	✓	
Slovenia	SI0008	✓	✓	✓	✓	✓			✓	✓			✓		✓	✓	
Slovakia	SK0002									✓							
	SK0004														✓		
	SK0005						✓	✓		✓					✓		
	SK0006						✓	✓		✓					✓		
	SK0007						✓	✓		✓							
Turkey	TR0001						✓	✓	✓	✓		✓	✓				

#: Stations are listed as EMEP stations at (http://www.nilu.no/projects/ccc/onlinedata/pm/stations_pm.html). However data on PM_{10} concentrations have not been reported directly to EMEP for the year 2004. The concentrations presented in this report for these stations have rather been retrieved from the AIRBASE.