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Characterizing temporal and spatial patterns of urban PM_{10} using six years of Finnish monitoring data

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Data from the Finnish Meteorological Institute's Air Quality Monitoring Data Management System (ILSE) for 1998–2003 were used to examine the temporal and spatial patterns of urban PM_{10} in Finland. Long term means of PM_{10} at 24 Finnish urban stations vary between 11 and 24 μg m^{-3}. The seasonal variation of PM_{10} at all stations was dominated by the spring maximum. A strong influence of traffic on the urban PM_{10} concentrations is shown. However the highly synchronized day-to-day variation at a variety of sites across the country highlights the role of large scale weather patterns also in the formation of spring episodes. Every year, most often in August, September and October, there were also 1–5 irregular regional PM_{10} episodes, lasting from one day to six days and most likely caused by long-range transported particles. During these regional events, the PM_{10} concentrations may well reach the typical spring peak concentration levels.

Introduction

In its recent review the World Health Organization confirmed the causal relationship between particulate matter (PM) exposure and health effects (WHO 2003). The present information shows that fine particles are strongly associated with mortality and cardio-pulmonary diseases. There is evidence that also coarse particles promote respiratory illnesses, but their effect on mortality is less clear. Epidemiological studies on large populations have been unable to identify a threshold concentration below which ambient PM has no effect on health (WHO 2003). It is estimated that in the European Union 55% of the urban population in 2002 was exposed to concentrations of particulate matter exceeding those of the EU limit values set for the protection of human health (European Environment Agency 2005).

Atmospheric particulate matter is a mixture of solid and aqueous species originating from anthropogenic and natural sources. Due to the large number of sources, PM may present diverse physical and chemical patterns in different areas. In addition to the emissions, both climatology (temperature, humidity, radiation, precipitation scavenging, re-circulation *vs.* dispersion of air masses) and geography (topography, soil cover, proximity of arid zones or the coast) have effect on the ambient PM characteristics in a given region. Therefore, wide variations in PM levels and characteristics may be expected when considering regions with different climatological and geographical patterns.

Here we present a broad, national-level summary of the spatial and temporal patterns of PM_{10} in Finland. Although PM_{10} is not a specific indicator for human health impacts, it represents the

overall thoracic fraction of the ambient particles and as such forms an inevitable part of human exposure.

At present in about 50 cities in Finland there are over a hundred fixed monitoring stations, equipped with on-line ambient air quality monitors. These sites are managed and operated by local bodies that also carry out regular assessments of air quality based on the needs of each territory. Whereas the routine monitoring data from e.g. the Helsinki metropolitan area have nowadays become highly exploited in combination with extended measurement and modeling research projects, the vast majority of these long term data have remained scientifically undiscovered. Especially integrated assessments aiming at broad national level have been very few and have mainly been focused on the establishment of the exceedances of the air quality guideline and limit values (Kukkonen *et al*. 1999, Pietarila *et al*. 2000, Anttila *et al*. 2003).

The Finnish long term PM_{10} monitoring data with the high time resolution and capture and encompassing different sized cities in different parts of the country make up a worthy data set for the establishment of this national PM_{10} summary.

Material and methods

The mass concentrations of PM_{10} presented in this paper were obtained with automatic analyzers using either the tapered element oscillating microbalance (TEOM) or the beta-attenuation method (Table 1). In the TEOM instrument particles are collected on an oscillating filter whose frequency changes as the mass loading of the filter increases. In these instruments, the aerosol stream towards the filter is typically heated to 50 °C to prevent moisture from affecting the mass measurements. The beta-attenuation analyzer is based on the measurement of the reduction in the intensity of beta particles passing through a dust-laden filter. The filter material or the air flow is not usually heated. The manufacturer provides calibration foils against which the performance of the instrument can be checked and adjusted. In both methods the particle size selection is determined by the inlet geometry and the flow rate used so flow calibration is an essential part of the routine maintenance of the instruments.

The data obtained by these automatic analyzers are not necessarily fully equivalent with those from direct gravimetric methods (or with each other), the unknown loss of semi-volatile species being an important cause of differences. To quantify these errors in the Finnish climatic conditions a field intercomparison was arranged in Helsinki (station Helsinki2 of this study) in autumn 2000 and winter/spring 2001 (http://www.fmi.fi/kuvat/FINal_PM_Report.pdf). Two beta-attenuation instruments and a TEOM instrument were compared with the reference samplers according to the European Standard EN12341. Based on the results of this comparison it was concluded that the equivalency of both these methods with the reference method was good enough and no correction factors were needed. These conclusions were proposed for all public monitoring networks in Finland.

However, the PM_{10} measurements of all local networks in Finland have not yet been systematically intercompared. The first field intercomparison campaign of the public monitoring networks (directed at gaseous compounds) was carried out in 2002 and 2003 (Walden *et al*. 2004). In this context also a field audit of the operation and quality control of the measurement stations was conducted. Almost all the networks have documented quality assurance procedures and orderly maintenance practices together with obviously functional data acquisition, transfer and processing methods. However, the methods and procedures often differ significantly between the networks. In the case of PM_{10} this lack of national harmonization creates a potential for discrepancies, which cannot be fully ruled out here.

PM_{10} data were extracted from the Finnish Meteorological Institute's Air Quality Monitoring Data Management System (ILSE), a database which aggregates all national monitoring results from local networks. Those PM_{10} measurement sites with a minimum annual data capture of 75% during at least four years between 1998 and 2003 were used. Altogether 25 sites in 20 cities in different parts of the country met these criteria (Table 1 and Fig. 1). In the past the PM_{10} data in the ILSE database were reported relative to

Table 1. Overview of the PM_{10} monitoring sites.

Place	Station name	Network	Population	Type of the measurement site	Nearest street		NO_x[1] median (μg m^{-3})	PM_{10} measurements 1998–2003		
					Distance (m)	Traffic flow (vehicles day^{-1})		Capture (%)	Height (m)	Instrument
Turku	Kauppatori	Turku	175000	urban/traffic	4	6000	53	98	3.5	Eberline FH62IN
Raisio	Keskusta	Turku	23000	urban/traffic	18	4500	46	98	3	Eberline FH62IN
Naantali	Keskusta	Turku	14000	urban/traffic	6	n/a[2]	27	82	3	TEOM 1400A
Pori	Itätulli	Pori	76000	urban/traffic	3	n/a[2]	45	82	4	Eberline FH62IR
Lohja	Nahkurintori	Lohja	36000	urban/background	4	2500	23	99	3	TEOM 1400A[4]
Hämeenlinna	Raatihuoneenkatu	Hämeenlinna	47000	urban/traffic	30	8500	48	63	3	TEOM 1400
Jyväskylä	Lyseo	Jyväskylä	82000	urban/traffic	25	12000	25	65	3.5	TEOM 1400A
Helsinki1	Töölö	YTV	560000	urban/traffic	2	25000	95	99	4.5	TEOM 1400AB
Helsinki2	Vallila	YTV	560000	urban/traffic	12	13000	45	98	4.5	Eberline FH 62IR
Helsinki3	Kallio	YTV	560000	urban/background	80	7700	31	82	4	Eberline FH62IR
Espoo1	Leppävaara	YTV	224000	suburban/traffic	30	14500	48	99	4.5	TEOM 1400AB
Espoo2	Luukki	YTV	224000	rural/background	800	4900	6	80	7[3]	Eberline FH 62 IR
Vantaa	Tikkurila	YTV	184000	suburban/traffic	8	14500	69	97	4	TEOM 1400AB
Lappeenranta	Keskusta	Imatra	59000	urban/traffic	25	24000	34	60	3	Eberline FH62IR
Imatra1	Rautionkylä	Imatra	30000	suburban/industry	350	10000	14	95	8	Eberline FH62IN[4]
Imatra2	Mansikkala	Imatra	30000	suburban/background	100	8000	19	65	3	Eberline FH62IR[4]
Kotka	Kirjastotalo	Kotka	55000	urban/background	40	6400	15	65	13	Eberline FH62IR
Kouvola	Keskusta	Kouvola	31000	urban/traffic	30	10000	41	87	4	TEOM 1400
Varkaus	Pääterveysasema	Varkaus	23000	suburban/industry	20	5500	11	96	6	TEOM 1400A
Oulu1	Keskusta	Oulu	126000	urban/traffic	5	7000	63	95	4	TEOM 1400
Oulu2	Pyykösjärvi	Oulu	126000	suburban/background	n/a[2]	n/a[2]	14	97	4	TEOM 1400
Kajaani	Keskusta	Kajaani	36000	urban/traffic	6	13800	23	99	2	TEOM 1400A
Kokkola	Keskusta	Kokkola	36000	urban/traffic	15	3000	24	95	4	TEOM 1400A[5]
Pietarsaari	Bottenviksvägen	Pietarsaari	19000	urban/traffic	10	3600	21	81	3	TEOM 1400
Kuopio	Keskusta	Kuopio	88000	urban/background	50	11800	19	77	4	TEOM 1400A

[1]whole period median of the parallel NO_x measurement as NO_2.
[2]not available.
[3]height changed to 4.5 meters 24 January 2000.
[4]instrument was changed to ThermoESMAndersen FH 62IN/R 1 January 2003.
[5]instrument was changed to Eberline FH 62IR 11 July 2002

Fig. 1. Locations of the stations. Division of the stations into four geographical regions is denoted with lines.

20 °C; now all historical data have been revised to local temperatures which is also the format used here.

The classification of the sites is based on the European Communities exchange of information decision (EC 2001). All except one (Espoo2) of the stations are located in urban or suburban surroundings. Espoo2 is located in the countryside about 20 km northwest from Helsinki and represents rural surroundings. The majority of the stations (16) are classified as traffic stations, with seven and two classified as background and industry stations, respectively. These nine background or industry stations are here referred to as non-traffic stations (Table 1).

The data were grouped into four broad geographical regions; southwestern Finland, the Helsinki metropolitan area, southeastern Finland and northern Finland (Fig. 1). The four groups were adjusted to nearly equal size to enhance the clarity of the graphical displays of data. This grouping is used in the text and figures unless stated otherwise.

All measuring sites included in this study had collocated NO_x measurements with the conventional chemiluminescent method (*see* the field comparison results in Walden *et al*. 2004). These NO_x concentrations in 1998–2003 were also retrieved from the ILSE database (the whole-period median is given in Table 1). Temperature, precipitation and depth of snow cover data from Turku, Helsinki, Lappeenranta and Oulu airport synoptic stations, i.e. one station from each of the four geographical groups, were retrieved from the database of the Finnish Meteorological Institute. These weather data were utilized to compare the seasonal variation of PM_{10} concentrations in different parts of the country.

Results and discussion

Annual variations

At each station the annual median PM_{10} concentrations spanned a relatively narrow range (Fig. 2). The station-specific difference in the highest and lowest annual median concentration was typically only 2–4 μg m^{-3}, and with no common trend. From station to station the concentrations differed more. The total-period median of PM_{10} varied by a factor of two between the lowest three, Varkaus, Kajaani and Espoo2 (9 μg m^{-3}), and the highest, Helsinki1 (20 μg m^{-3}). The non-traffic stations tended to have lower concentrations than the traffic stations, however these categories overlap. The rural background station Espoo2 has distinguishably lower concentrations than the rest of the stations in the Helsinki metropolitan area.

Traffic-induced dust has been reported to be a substantial source of elevated PM_{10} concentrations at Finnish urban sites especially in spring (e.g. Hosiokangas *et al*. 1999, 2004, Vallius *et al*. 2000, Pakkanen *et al*. 2001, Pohjola *et al*.

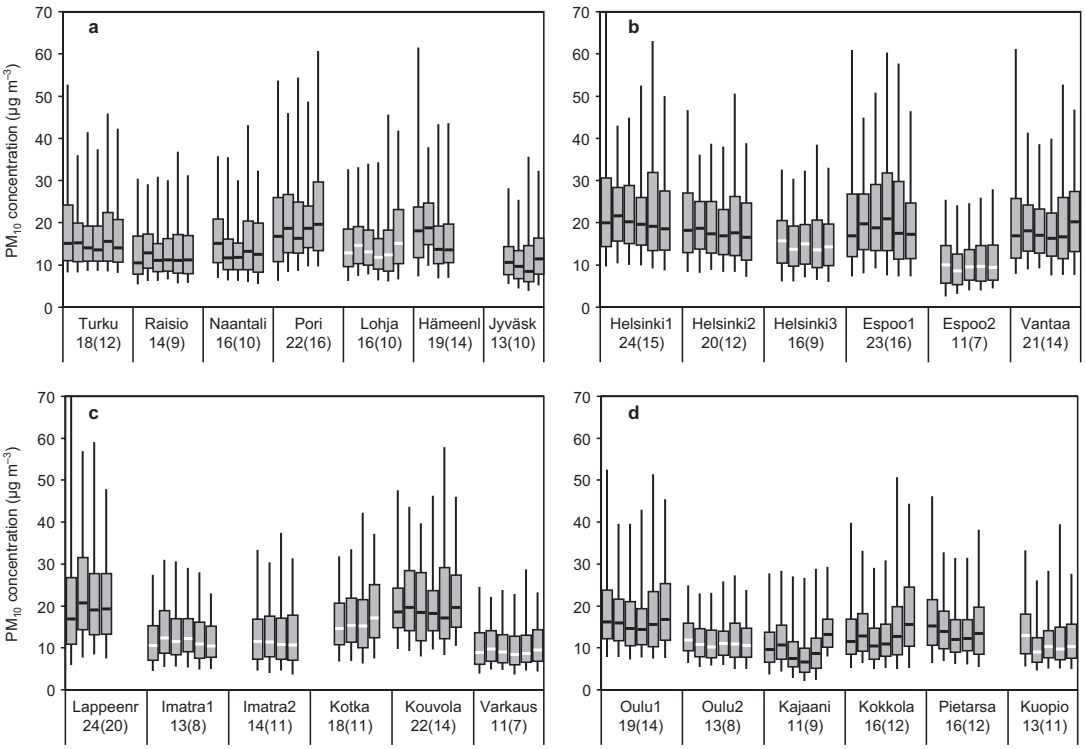

Fig. 2. Annual distribution of 24-hr average PM_{10} concentrations from 1998 to 2003 at the 25 stations in Finland. (**a**) Southwestern Finland, (**b**) the Helsinki metropolitan area, (**c**) southeastern Finland, and (**d**) northern Finland. Values for the 5th, 25th, 50th (median), 75th and 95th percentiles are shown. At the non-traffic stations the medians are denoted with white horizontal lines. The total means (1998–2003) and standard deviations (in μg m^{-3}) are shown at the bottom of each set of plots.

2002, Laakso *et al*. 2003, Kukkonen *et al*. 2005). However, the long-term mean concentrations of PM_{10} seemed not to be extremely sensitive to the very unequal traffic volumes contributing at the stations (Table 1). While the PM_{10} total-period median varied by a factor of two between the lowest and highest in this material, NO_x concentration varied by a factor of 15 between the lowest (rural Espoo2) and highest (Helsinki1) (*see* Table 1). This indicates that the long term PM_{10} concentration is less sensitive to the local traffic sources than is the NO_x concentration.

Van Dingenen *et al*. (2004) used the 5th percentile values of PM_{10} at the rural and near city background sites to derive a European continental background PM_{10} concentration 7.0 ± 4.1 μg m^{-3}. The use of the 5th percentile values was justified by the fact that in their extensive compilation of data the 5th percentile values of rural and near city background sites were similar to the annual average concentrations observed at natural background sites (i.e. Norwegian and Swedish EMEP sites, with distances from large pollution sources over 50 km). The mean 5th percentile values of the nine non-traffic stations here yielded an average of 5.2 ± 1.2 μg m^{-3} suggesting a lower background concentration for Finland than the European continental value given by Van Dingenen *et al*. (2004). Note that these background values are not purely natural by definition, but are affected by long-range transported particles.

As a whole the Finnish concentration levels were consistent with those obtained in European city or near-city data compilations (Lazaridis 2001, Van Dingenen *et al*. 2004, Querol *et al*. 2004). Heal *et al*. (2005) found the urban background annual mean concentration in central Edinburgh to be 15.5 μg m^{-3} (in 1999–2000). Harrison *et al*. (2001) found 26 μg m^{-3} as an

annual mean (1997–1998) for the urban background, 36.5 μg m^{-3} for a busy street canyon (Marylebone road) in London, and 16.8 μg m^{-3} and 19.5 μg m^{-3} at two rural sites in the UK. Three-year urban background data from Birmingham gave an annual mean of 22.5 μg m^{-3} (Harrison *et al.* 2001). Gehrig and Buchman (2003) reported 22.5–35.9 μg m^{-3} as long-term means (1998–2001) for urban/suburban stations in five cities in Switzerland, and 20.7 μg m^{-3} for a lowland rural site. Gomišček *et al.* (2004) reported PM_{10} annual means of 26 to 31 μg m^{-3} (1999–2000) for three urban sites in Vienna, Linz and Graz and 21 μg m^{-3} for a rural site in Austria. Rodríguez *et al.* (2003) reported PM_{10} annual means of 46 and 49 μg m^{-3} (year 2000) for two urban street canyons (L'Hospitalet and Sagrera), 59 μg m^{-3} for an industrial urban site (S. Andreau) in northeastern Spain, and 17 μg m^{-3} for a rural site (1998–2000). Chaloulakou *et al.* (2003) reported an annual mean PM_{10} concentration of 75.5 μg m^{-3} (1999–2000) for downtown Athens. These examples from the cities in different parts of Europe show that long-term means exceeding 30 μg m^{-3} are quite typical in urban environments and 15–20 μg m^{-3} are frequently given as rural long-term means. The Finnish concentrations tended to settle in the lower end of the European concentration range.

Seasonal variation

A highly synchronized temporal pattern could be seen not only within each geographical subgroup but also countrywide (Fig. 3). The overarching feature of the PM_{10} concentrations was the sharp maximum in March–April. This spring maximum was highest at the traffic stations but also present at the urban non-traffic stations. The concentrations started to increase already when temperature was well below zero, but reached their peak when temperature approached zero.

Numerous Finnish studies have shown that traffic-induced resuspension is the predominant source of coarse particles and forms a pronounced contribution also to the PM_{10} at urban traffic-influenced sites (e.g. Hosiokangas *et al.* 1999, 2004, Vallius *et al.* 2000, Pakkanen *et al.* 2001, Pohjola *et al.* 2002, Laakso *et al.* 2003, Kukkonen *et al.* 2005). Other identified contributors to PM_{10} in these studies are motor vehicle exhaust, wood combustion, long-range transport and sea salt. While the major role of the traffic related dust in the PM_{10} concentrations is widely accepted, the proportions of geological dust, traction sand, wear of asphalt enhanced by studded tires or even wear of break linings in road dust are not yet quantified (e.g. Kuhns *et al.* 2003, Kupiainen and Tervahattu 2004, Sternbeck *et al.* 2004). However, a commonly accepted hypothesis is that during winter the particles from different sources are accumulated onto road surfaces and shoulders and then resuspended by traffic-induced turbulence during favorable meteorological conditions.

During the study period (as in Finland generally), March–May was the period with least rain (Fig. 4). The elevated concentrations of PM_{10} coincided with this extended period of relatively low rainfall amounts. The PM_{10} concentrations settled down before the more rainy summer season started, so the dryness itself hardly was the ultimate cause of the concentration rise. On the other hand, the elevation of PM_{10} concentrations began quite concurrently with the onset of snowmelt period in the first half of March at the southern coast and a week or two later inland. So the concentration increase at these traffic stations occured well before the soils in general had become uncovered from snow. This may well be due to the fact that roads, and even entire city centers, are kept clear from snow and weather conditions conducive to melting the winter snow pack readily enhanced the particle resuspension from the roads.

Omstedt *et al.* (2005) managed to reproduce the spring peak in a street canyon and in a highway in Stockholm by modeling the road surface moisture and the dust layer growth on the road surface during the wet autumn/winter period and its rapid decrease due to resuspension in early spring when evaporation rates increase due to higher solar radiation and higher temperatures. The high number of cars with studded tires and the winter sanding of the streets were additional factors contributing to the development of the spring peak.

In Finland the use of studded tires is allowed from 1 November up to the week after the Easter

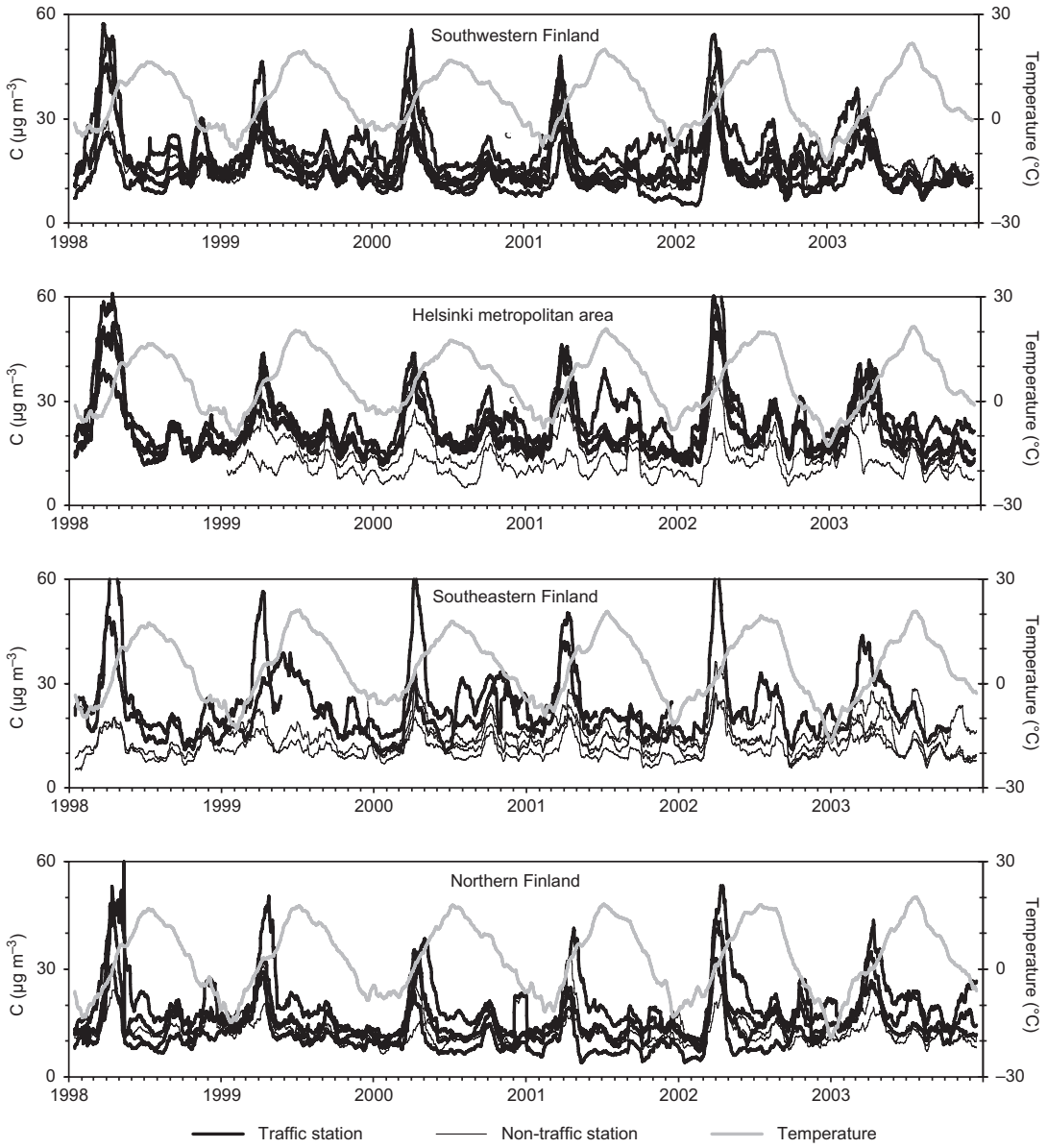

Fig. 3. Monthly running means of the daily PM_{10} concentrations (black lines) and daily mean temperatures in 1998–2003. Meteorological data are from Turku, Helsinki, Lappeenranta and Oulu airport synoptic stations.

Monday, the date of which varied between 8 April and 1 May during this study period. However the autumnal start of the usage of studded tires and/or the start of winter sanding did not instantly cause a large scale suspension of particles from the roads (Fig. 4). This can be due to the higher relative humidity and more frequent rain/snowfall events during the winter months which limit the suspension and rather enhance the accumulation of particles on road surfaces. Only the springtime dry period with higher temperatures, radiation and evaporation enables the effective suspension.

Figure 5 gives an impression of the magnitude of the spring peak at each station compared to the mean concentration of the rest of the year. Here we aggregated the baseline year from the June to January PM_{10} concentration; February

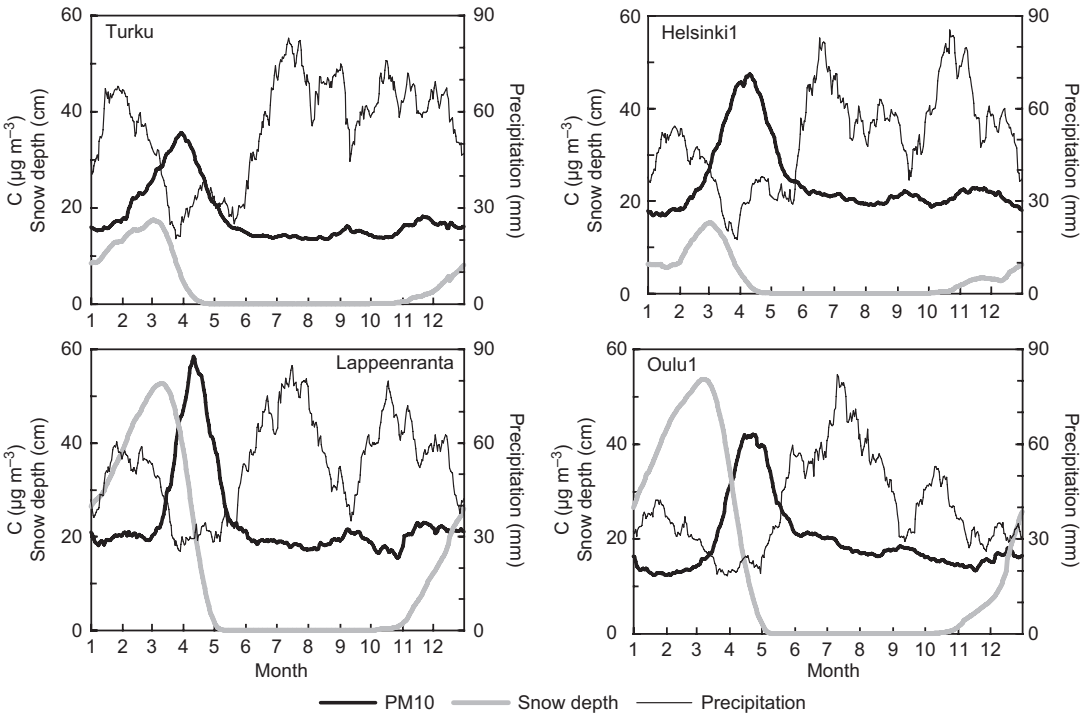

Fig. 4. Seasonal variations of the PM_{10} concentration and snow depth and precipitation averaged over years 1998–2003. PM_{10} concentrations and snow depths were first filtered with monthly running mean and precipitation with monthly running sum.

and May were excluded as transition months, and the spring peak was compiled of the March–April mean concentrations. These were clearly correlated; a high baseline concentration implied a high spring peak, with the long-term mean of the spring peak about twice as high as the rest of the year ($n = 25$, $r = 0.91$, $p < 0.0001$). This suggests that the spring peak is strongly connected to the stations' year-round source/sources which tend to strengthen/discharge during the spring months. At the non-traffic stations the PM_{10} concentrations tended to rise relatively less than at the traffic stations. For example Kotka in the southeastern coast was characterized by a high baseline concentration and a low spring peak. Of the non-traffic stations Kuopio in central Finland, for example, had a relatively high spring peak as compared with its baseline. However, also at the rural Espoo2 the March–April concentrations were about 30% higher than during the rest of the year.

The annual cycle of PM_{10} in Finnish cities differs much from the one observed e.g. in central Europe, where elevated concentrations occur during the winter season and are due to meteorological effects; frequent inversions in winter and good vertical mixing during summer (Gehrig and Buchmann 2003, Gomišček *et al*. 2004). In the Mediterranean region the restrained mesoscale circulations with slow scavenging potential favor the aging of air masses during summer causing subsequent summer elevation of PM_{10} (Rodríguez *et al*. 2003).

The highly synchronized temporal pattern of PM_{10} at a variety of sites across the country (Fig. 3) highlights the role of large scale weather patterns in the formation of PM_{10} episodes in Finland too.

The widespread moderate secondary rises of PM_{10} concentrations in the latter half of the year (*see* Fig. 3) further point toward synoptic scale meteorology. These latter episodes varied in magnitude and timing from year to year more than the spring maximum. For example in 1998 there were simultaneous concentration elevations at almost all stations in September and again at

the turn of November–December, as well as at the turn of September–October in 2000, in September in 2001, in August in 2002, and in July in 2003. We will focus more in these non-spring episodes later in this article.

Day-of-week pattern

The influence of the human activities and traffic flows were clearly seen (Table 2) in the weekly pattern of the PM_{10} concentrations. Traffic intensities are lower during weekends, especially on Sundays when opening hours of shops are regulated, a fact that may be meaningful in city centers.

When calculated for the total period 1998–2003 all stations showed significantly lower concentrations during Sundays than during weekdays, the difference varying between 10% and 30%. Also all non-traffic stations showed this difference. However, seasonally this pattern showed more variation. During the spring period all stations except rural Espoo2 exhibited significantly lower concentrations on Sundays than on weekdays while in the autumn period this was true at only about half of the stations. Thus in spring the lower traffic on Sundays also systematically reduced the PM_{10} concentrations, while in the autumn period equivalent reduction in traffic flows produced no significant reduction in PM_{10} at several sites. This suggests that in the development of the generally lower autumn concentrations the role of traffic is no more so overwhelming. However during the summer period with equally low level of the PM_{10} concentrations the Sunday reduction of the concentrations was yet again more prevalent (even without studded tires and traction sanding). This implies that in the autumn period some additional source disturbs the strong connection between the traffic and PM_{10}. This pattern was most systematic in the southeastern group and almost absent in the northern group.

The influence of the local traffic flows was clearly seen also on the diurnal cycle of PM_{10} concentrations (Fig. 6). The urban city centers Helsinki1, Lappeenranta and Oulu1 experienced the strong morning congestion peak at about 08:00 during weekdays. During the off-peak

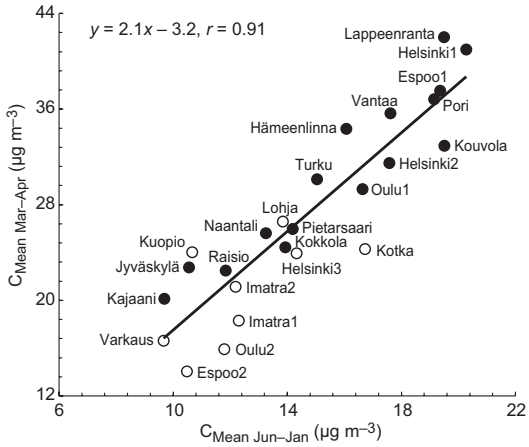

Fig. 5. Relationship (scatter plot and linear regression) between the mean June–January PM_{10} concentrations and the mean March–April concentration at each station in 1998–2003. Open circles are non-traffic stations.

midday hours the concentrations developed more smoothly, and the afternoon rush hour was not reflected as strongly as the morning one maybe due to the typically better atmospheric mixing conditions in the afternoons. The smaller towns Jyväskylä, Pietarsaari and Naantali had concomitant but weaker morning peaks and finally at the small town suburban stations the morning peak was hardly visible. At the rural station Espoo2, the direct weekday morning peak was missing and Sunday differed only slightly from the weekday daily cycle. At urban stations on Sundays, a morning minimum took place and was followed by slow increase during the day. Interestingly the Sunday night concentrations at urban traffic stations were systematically higher than those on weekday nights. Evidently the more intensive nightlife increased concentrations, the effect of which was further strengthened by the decreased mixing of the stable nighttime atmosphere.

The Sunday early morning minimum concentrations are evidently least affected by traffic and other human activities following weekly and diurnal cycles. As such they may give information on the contributions of other PM_{10} sources, such as windblown dust, sea spray, wildfires and anthropogenic secondary particles. Early Sunday morning mean concentrations varied between 7–16 μg m^{-3} (Table 3). Interestingly the Sunday minimums tended to be slightly lower at the

northern stations; all stations north of Jyväskylä and Varkaus had Sunday minimums below 10 μg m^{-3} while in the south only the rural and suburban background stations Espoo2 and Imatra2 had concentrations this low. This may reflect the longer distance and subsequently smaller influence of the southern long range transported pollution to the northern stations. The weekly and diurnally non-cyclic proportions of PM_{10} concentrations represented by the Sunday minimum encompassed on average two thirds of the long-term mean of the PM_{10} (Table 3). The lowest non-cyclic relative proportion was detected at the northern urban traffic station Oulu1 (48%) and highest at the eastern suburban industry station Imatra1 (83%).

Spatial variation

High spatial variability is often associated with the PM_{10} concentrations due to the potential effects of the very localized, sporadic particle suspension in the vicinity of the monitors and short atmospheric lifetime. The spatial distribution of the PM_{10} concentrations was addressed by analyzing the correlation (Pearson r) of the daily values between the different sites. The correlation between the stations declined linearly with the distance between them (Fig. 7a). The list of highest correlations (Table 4) was dominated by the dense network of the Helsinki metropolitan area, but was completed with other closely-spaced (~20 km) station pairs and also

Table 2. Statistically significant differences between the average Sunday and weekday (average Tuesday–Thursday) PM_{10} concentrations. Non-traffic stations are set in italics.

	Difference between Sunday and weekday (%)				
Station	All	Winter	Spring	Summer	Autumn
Southwestern					
Turku	–22***	–29**	–21***	–25***	
Raisio	–19***	–21**	–18***	–23***	–17*
Naantali	–15***		–13*	–24***	
Pori	–24***		–30***	–25***	–19*
Lohja	*–17****	*–18**	*–21****	*–19***	
Hämeenlinna	–20***	–16*	–25**	–16*	
Jyväskylä	–25***	–21**	–29**	–20*	–25**
Helsinki metropolitan area					
Helsinki1	–27***	–32***	–26***	–29***	–23***
Helsinki2	–24***	–23***	–24***	–29***	–19*
Helsinki3	*–26****	*–24****	*–28****	*–31****	
Espoo1	–29***	–25***	–29***	–35***	–26***
Espoo2	*–13***			*–21**	
Vantaa	–25***	–26***	–26***	–25***	–21**
Southeastern					
Lappeenranta	–28***	–21*	–40**	–23**	
Imatra1	*–15****	*–15**	*–15**	*–22****	
Imatra2	*–21****	*–23**	*–30**		
Kotka	*–22****		*–22**	*–25***	
Kouvola	–10**		–19**		
Varkaus	*–12****	*–8**	*–20***	*–15***	
Northern					
Oulu1	–30***	–21***	–32***	–34***	–30***
Oulu2	*–21****	*–22***	*–17***	*–22****	*–25****
Kajaani	–20***	–17**	–20**	–26***	–17*
Kokkola	–28***	–21**	–32***	–28***	–30***
Pietarsaari	–23***	–12*	–19*	–29***	–30***
Kuopio	*–20****		*–21***	*–24****	*–16**

Nonparametric Wilcoxon signed rank test: *$p < 0.01$, **$p < 0.001$, ***$p < 0.0001$.

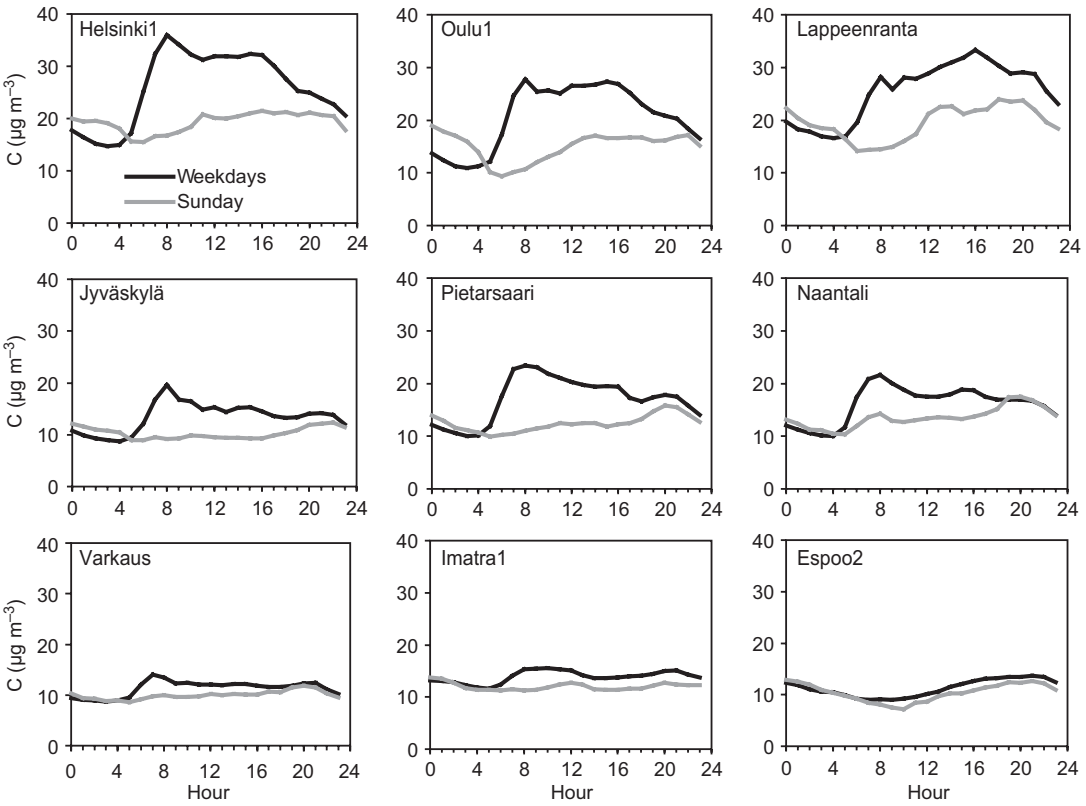

Fig. 6. Average diurnal variation of PM_{10} at selected stations during weekdays (Monday–Friday) (black lines) and Sundays (grey lines) compiled from years 1998–2003.

with a couple of reasonably distant station pairs as Kuopio and Jyväskylä (distance 122 km) or Imatra2 and Varkaus (135 km). The two stations in Oulu with a distance of 4 km and $r = 0.76$ settled only narrowly outside this list. Instead Espoo2 — the only station classified as rural — had outstandingly different behavior than the other stations (*see* Fig. 7a, square symbol) with its poor correlation with especially traffic-stations (squares with dots). Actually it correlated best ($r = 0.77$) with the non-traffic Kotka station at a distance of 123 km eastwards and also with the non-traffic stations of Imatra and Varkaus. So not only the distance but also the type of station affected the correlation between the stations. This suggests that the correlation between the urban traffic stations is based on similar source profiles whose strength varies in time following the common weather conditions.

Another possible explanation for the detected fairly high correlations between urban stations located even as far as 100 km from each other would be the long range transported particles. However the correlations were highest in spring (Fig. 7b) during the period with highly elevated concentrations caused by road dust suspension, which endorses the role of common weather patterns as the primary cause of the correlation. The poorest correlations i.e. the most locally varying PM_{10} concentrations were detected during the winter season (Fig. 7b).

Comparison of our results with the other European studies is complicated due to the more variable European terrains which distracts the simple distance dependence of correlations detected here. However, the extremely high correlations of PM_{10} between distant urban stations reported from e.g. the Swiss basin (Dübendorf and Payerne; distance 160 km; $r = 0.87$; data from 1998–2001 (Gehrig and Buchman 2003) were not present in the Finnish data. This may indicate a higher influence of local sources as compared with the long range transported particles in Finland.

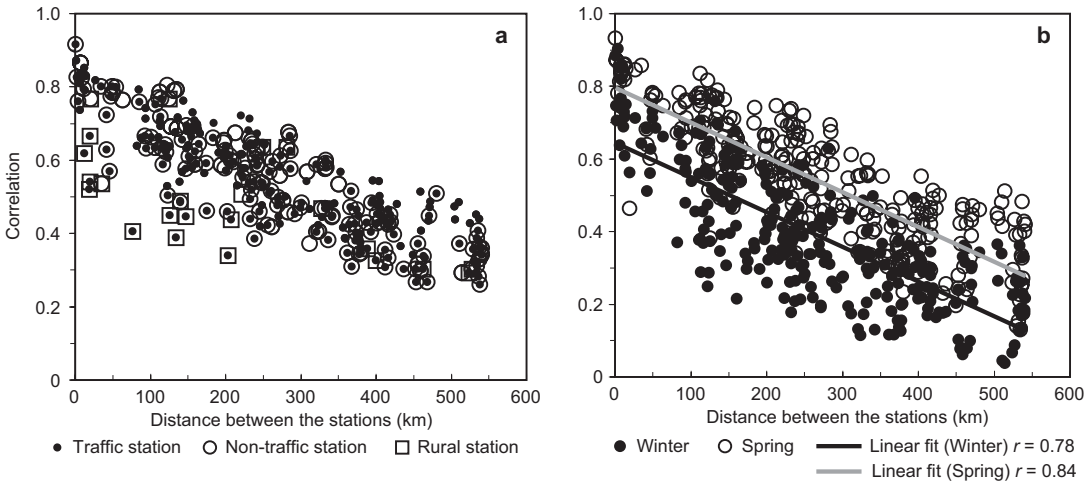

Fig. 7. Correlation (Pearson *r*) of the daily values of PM_{10} between the sites against their distance in 1998–2003. (**a**) All station pairs segregated by station type (note that each mark in the plot is generated as a combination of type marks of the paired stations), and (**b**) all urban station pairs (i.e. Espoo2 removed) segregated by season.

Table 3. Minimum concentrations of the diurnal distributions calculated from Sundays in years 1998–2003 at each station.

Station	Minimum of the Sunday distribution (μg m^{-3})	Proportion of the Sunday minimum of the total period mean (%)
Southwestern		
Turku	11.8	64
Raisio	10.0	72
Naantali	10.4	67
Pori	11.0	49
Lohja	11.6	72
Hämeenlinna	12.4	64
Jyväskylä	8.9	70
Helsinki metropolitan area		
Helsinki1	15.5	64
Helsinki2	13.4	66
Helsinki3	10.5	64
Espoo1	14.0	61
Espoo2	7.2	64
Vantaa	14.1	67
Southeastern		
Lappeenranta	14.1	59
Imatra1	11.2	83
Imatra2	8.9	63
Kotka	11.9	66
Kouvola	15.7	72
Varkaus	8.6	78
Northern		
Oulu1	9.4	48
Oulu2	8.6	69
Kajaani	7.0	61
Kokkola	9.4	60
Pietarsaari	9.9	62
Kuopio	9.3	70

Regional episodes

The repeated coincident elevated PM_{10} concentrations throughout the country were visible in the smoothed time series (Fig. 3). An episodic day for PM_{10} at a site was defined as a day on which the observed 24-hour average concentration exceeded the 90th percentile of all data of the site during 1998–2003. Further a regional

Table 4. Highest correlations of the daily values of PM_{10} between the sites; all measurements 1998–2003.

Station *vs.* station	Pearson *r*	Distance (km)
Helsinki2/Helsinki3	0.91	1
Helsinki1/Helsinki2	0.87	2
Imatra1/Imatra2	0.86	8
Helsinki2/Vantaa	0.85	12
Raisio/Naantali	0.84	8
Helsinki1/Helsinki3	0.83	3
Helsinki2/Espoo1	0.83	7
Espoo1/Vantaa	0.83	15
Helsinki1/Vantaa	0.83	13
Helsinki3/Espoo1	0.82	9
Kokkola/Pietarsaari	0.82	27
Helsinki1/Espoo1	0.81	7
Lohja/Helsinki2	0.8	50
Lappeenranta/Imatra2	0.8	35
Jyväskylä/Kuopio	0.8	122
Turku/Naantali	0.79	13
Hämeenlinna/Vantaa	0.79	85
Helsinki3/Vantaa	0.79	11
Imatra2/Varkaus	0.79	135

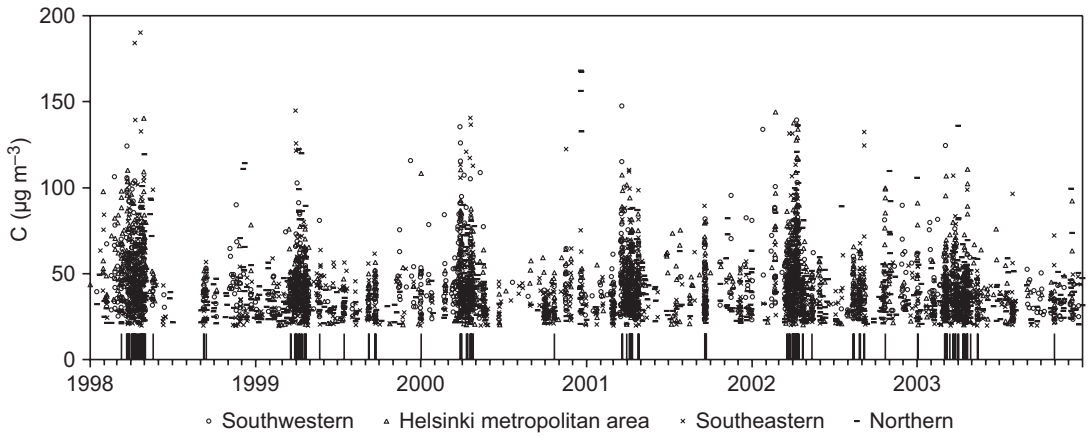

Fig. 8. Time series plot of the episodic day PM_{10} concentrations at each station in 1998–2003. Regional episodic days are indicated with black vertical markers on the abscissa.

episodic day was chosen to be a day when at least half of the measuring stations were observing an episodic day.

The PM_{10} episodic days tended to be grouped in dense clusters as a function of time; most regularly appearing were the March–April episodes (Fig. 8). Of the 2191 days of the total study period, altogether 148 (6.7%) days were regional PM_{10} episode days. Since about half of the episodes of each station occured in March–April (varying from 33% in Espoo2 to 72% in Hämeenlinna) the spring time co-occurrence was hardly a surprise.

More interestingly, every year there were also 1–5 non-spring irregular regional episodes lasting from one day to six days which cannot directly be connected to local dust but might rather be connected to the long range transport. These non-spring episodes typically occured in August, September and October. Only winterly regional episodes were related to New Year's Eves in 2000 and 2003. The winter months were characterized by dispersed occurrence of high concentrations while midsummer (June) was characterized by an absence of high concentrations. Perhaps the poor wintertime mixing conditions give rise to very local concentration development with increased influence of specific local sources such as small scale wood combustion. These low level inversions are most typical in the north. In this respect it is worth noticing that the highest concentrations during winter months were most frequent at northern sites Oulu2 and Kokkola (not shown).

The sources of a pair of the widespread autumn episodes of PM have been identified. Tervahattu *et al*. (2004) and Hongisto and Sofiev (2004) studied the sources and development of the PM_{10} episode on 17–23 September 2001. This episode was shown to be composed of soil dust particles raised by wind in the Kazakhstan Ryn Peski desert and mixed with the anthropogenic particle emission from Estonian and Russian oil-shale burning industries. The data here show that this episode was recorded throughout the country; on 19 September all 25 stations recorded daily PM_{10} concentrations which belonged to the highest tenth of the data at each station i.e. fully comparable to the spring dust episodes in magnitude. An almost equally widespread autumn episode was recorded on 14 September 1998 when 16 of the 17 stations working that day recorded the episode (*see* Fig. 8).

Hongisto and Sofiev (2004) further showed in their long term model simulation that the most important source of natural LRT dust in Scandinavia is the desert area near the Caspian Sea, whose influence is about an order of magnitude higher than that of the Sahara desert. LRT pollution episodes originating from the Caspian Sea region were shown to be most frequent during March; however severe episodes were estimated to be extremely rare.

Niemi *et al*. (2005) reported three fine particle LRT episodes in late summer 2002 (12–15 and 26–28 August and 5–6 September). These fine particle episodes were identified to be enhanced

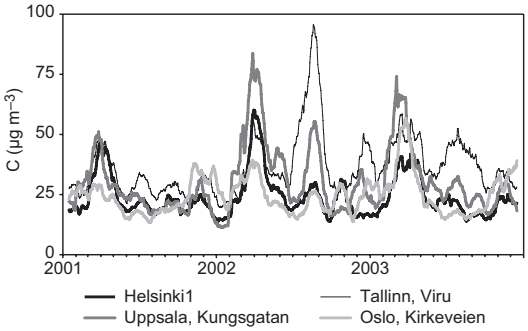

Fig. 9. Monthly running means of PM_{10} at Tallinn, Helsinki, Oslo and Uppsala 2001–2003.

by the forest fires and agricultural field burning in eastern Europe. Also here these fine particle episode days came forward as regional PM_{10} episode days.

These repeated widespread autumn episodes which occured during the period with no specific additional nationwide source or specifically difficult mixing conditions can plausibly be connected to the long range transport. On the other hand equally possible are the LRT-events of PM_{10} during the spring months with frequent co-occurrence of episodes, however the overwhelming influence of traffic-induced road dust may mask this LRT fraction during the spring months.

Niemi *et al*. (2004) identified sources of the fine particle LRT episode on 17–22 March 2002 over large areas of Finland. During this episode most of the PM_{10} particle mass was in the $PM_{2.5}$ size range with high sulphate, ammonium and nitrate concentrations (Niemi *et al*. 2004). Agricultural field burning in the Baltic countries, Belarus, Ukraine and Russia was shown to be a major source of the elevated fine particle concentrations. This fine particle episode coincided with the annual coarse fraction spring episode so its possible contribution to the PM_{10} concentrations at the sites included in this study could not be separated. However, perhaps coincidently, 19–20 March were also regional episode days for PM_{10}; 78% and 95% of stations recorded an episodic day.

Comparison with Oslo, Uppsala and Tallinn

Road dust reinforced by studded tires and small-scale residential wood combustion amplified by poor winterly mixing conditions have been identified as typical Scandinavian PM_{10} sources (e.g. Areskoug *et al*. 2004, Forsberg *et al*. 2005, Yttri *et al*. 2005). Comparison of the PM_{10} time series in Helsinki, Olso, Uppsala and Tallinn is given in Fig. 9. Estonian, Norwegian and Swedish raw data were downloaded from EEA's AirBase (http://air-climate.eionet.eu.int/databases/airbase/airview/index_html) and processed identically to the Finnish data. These cities have populations between 200 000–500 000. Traffic volumes are 25 000 vehicles per day in Oslo/Kirkeveien (Lützenkirchen and Lutnæs 2004) and 18 000 vehicles per day in Uppsala/Kungsgatan (Stockholm-Uppsala Air Quality Management Association http://www.slb.mf.stockholm.se). The traffic volume for Tallinn/Viru is not available.

The presence of the spring maximum was obvious in all four cities; however its dominance in the seasonal course of the PM_{10} concentrations varied a lot among these cities. In Helsinki and Uppsala the highest concentrations occured invariably in March–April. In Oslo the early-winter concentrations approached and even exceeded (in 2001) the spring concentrations, whereas Tallinn was characterized with extremely high summer and early autumn concentrations.

In Norway the spring dust is fought against by regulating the use of studded tires. In Oslo during the winter 2000/2001 the use of studded tires was substantially reduced by setting fees which resulted in significant reduction of the PM_{10} concentrations. This taxation charge was dropped for the next two years and was restored in 2004 due to its favorable effect on reducing the PM_{10} concentrations (Lützenkirchen and Lutnæs 2004).

The high winter concentrations — which were practically absent in the Finnish data — in Oslo are commonly connected to small scale fuel wood use in old wood-burning stoves. It has been estimated that as much as 25% of the PM_{10} mass in Oslo comes from wood burning (Yttri *et al*. 2005). The particulate emissions from small scale wood burning were estimated to be 384 tonnes in winter 2001/2002 (Finstad *et al*. 2004). For comparison, the particulate emissions from

small scale wood burning in the Helsinki metropolitan area have been estimated to be about 300 tonnes per year (Haaparanta *et al*. 2003). When calculated both per capita and per land area this yields roughly twice as high fuel-wood related particulate emission density in Oslo than in the Helsinki area. However, meteorology, topography and positioning of the stations relative to the residential areas evidently have a high impact on the concentrations, which makes this comparison only indicative.

The co-occurring summer and autumn episodes in all stations here were usually strongest in Tallinn which suggests its closer vicinity to the source. The total anthropogenic direct particulate emissions in Estonia were rapidly decreasing during the last 15 years and were in 2002 35 000 tonnes (as PM_{10}) as compared e.g. with the Finnish 55 000 tonnes (Vestreng *et al*. 2005). Combustion in power plants, residential combustion and traffic are the major sources of direct PM_{10} emissions in both countries. So the Estonian national direct emissions alone hardly explain these Scandinavian-wide summer episodes, though the fact is that Estonian emissions specifically originate from a couple of enormous point sources in northeastern Estonia (e.g. Treier *et al*. 2004) which indicates potential for episodic plume transportation and successive concentration enhancement. Sofiev *et al*. (2003) calculated that outside the radius of about 200 km from these Estonian sources, their influence on background deposition becomes negligible as compared with that of other sources. Both Tallinn and Helsinki are on the outskirts of this effective range.

A more probable explanation for these simultaneous but north and westwards attenuating episodes is the long-range transport from continental Europe and Asia. For example, the already mentioned fine particle episodes in August 2002 originating from forest fires in East Europe (Niemi *et al*. 2005) emerged here as a PM_{10} peak expanding over most of Scandinavia. At that time a long lasting hot and dry high pressure prevailed over Scandinavia, and the northwestern corner of Russia (from the Moscow Region to Sankt Petersburg) suffered from an extremely severe fire season; over ten thousand wildfires in forests and bog lands were registered due to long lasting dryness (Davidenko and Eritsov 2003).

The minimums and maximums in Helsinki and Uppsala followed each other almost perfectly, the only clear deviation was October–December 2002 when first Helsinki (and Oslo) experienced a maximum and Uppsala (and Tallinn) about one month later. The midwinter concentration rise was almost absent in Uppsala and Helsinki but very systematic in Oslo and also in Tallinn.

This non-exhaustive comparison implies that these four northern European cities share the spring peak phenomenon of PM_{10} as well as the long-range transported autumn episodes, however during the winter season the PM_{10} concentrations are more diverse.

Summary and conclusions

Long-term means of PM_{10} at twenty four Finnish urban stations varied between 11–24 $\mu g\ m^{-3}$, the urban centers with high traffic tending to have higher concentrations than the suburban stations or small towns. Year to year variation at each station was very low, typically only 2–4 $\mu g\ m^{-3}$. Several non-traffic stations in small towns were quite comparable in magnitude (Varkaus) and temporal variation (Kotka, Imatra, Varkaus) to the rural background station Espoo2 in the vicinity of Helsinki. The national background concentration was estimated to be about 5 $\mu g\ m^{-3}$, and there was some indication that when traffic influence was eliminated a decreasing trend of PM_{10} from south/southeast to north emerged.

The seasonal variation of PM_{10} at all stations was dominated by the maximum during spring; in March–April the PM_{10} concentrations were about twice as high as during the rest of the year. This spring peak of PM_{10} covered practically the whole snowmelt period, which also was the driest period of the year. In spring also the Sunday concentrations at all urban stations were significantly lower (from the 13% in Naantali to the 40% in Lappeenranta) than the weekday concentrations, which implies a strong effect of traffic on the PM_{10} concentrations. It is plausible that the spring dry period with increasing temperatures, radiation and evaporation enables the effective suspension of the dust accumu-

lated from multiple sources to road surfaces and shoulders and initiates the elevation of PM_{10} concentrations. However the highly synchronized day to day variation at a variety of sites across the country highlights the role of large scale weather patterns also in the formation of spring episodes. Thus they should not be regarded as just sporadic dust eruptions in the vicinity of the monitors.

Every year, most often in August, September and October, there were also 1–5 irregular regional PM_{10} episodes, lasting from one day to six days which most likely originated from long-range transported particles. During these regional events the PM_{10} concentrations may well reach the typical spring peak concentration levels. Similar regional LTR-events are probable also in spring but they get masked behind the overwhelming road influence. Summer was characterized by the lack of episodes and winter by spatially more scattered episodes.

Comparison of the PM_{10} concentrations from Oslo, Uppsala, Tallinn and Helsinki points to largely coincident but unequal spring peaks. Also the partly synchronized summer and autumn episodes may be considered characteristic of this region. During winter the PM_{10} concentrations were most diverse at these cities.

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