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Analysis and evaluation of selected PM_{10} pollution episodes in the Helsinki Metropolitan Area in 2002

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Abstract

In this study, we developed two methods to distinguish the long-range transport (LRT) episodes from local pollution (LP) episodes. The first method is based on particle number concentrations ratio between accumulation mode (diameter >90 nm) and Aitken mode (diameter 25–90 nm). The second method is based on a proxy variable (interpolated ion sum) for long-range transported $PM_{2.5}$. The ion-sum is available from the measurements of sulphate, nitrate and ammonium at the nearest EMEP stations. We also utilised synoptic meteorological weather charts, locally measured meteorological data, and air mass back-trajectories to support the evaluation of these methods. We selected nine time periods (i.e. episodes) with daily average $PM_{10} > 50\,\mu g\,m^{-3}$ in the Helsinki Metropolitan Area during year 2002. We characterized the episodes in terms of PM_{10} and $PM_{2.5}$ concentrations and the fraction of fine particles in PM_{10} at an urban traffic and regional background air quality monitoring sites. Three of these episodes were clearly of local origin. They were characterized by a low average fraction of $PM_{2.5}$ (<0.2) in PM_{10} at the urban traffic monitoring site, low ratio between PM_{10} concentrations at the regional background site and at the urban traffic site (<0.2), low average ion sums (1.5–2.5 $\mu g\,m^{-3}$) and low accumulation to Aitken mode ratios (0.13–0.26). Four of the episodes had distinct LRT characteristics: a high fraction of fine particles in PM_{10} (0.5–0.6) at the urban traffic site, a high ratio between PM_{10} concentrations at the regional background site and at the urban traffic site (0.7–0.8), high interpolated values for the ion sum (6.6–11.9 $\mu g\,m^{-3}$), and high accumulation to Aitken mode ratios (0.75–0.85). During the remaining two episodes there was significant contribution from both local sources and LRT. A detailed analysis of meteorological variables and air mass back-trajectories gave support to these findings. These characteristics can be utilised in a simple procedure to distinguish between LRT and LP episodes. Further quantitative investigations to these characteristics provide an indication to the episode strength. The quantitative results presented in the current study are applicable to the Helsinki Metropolitan Area and similar cities. Nevertheless, developing these methods for other cities require analyses of the meteorological conditions, behavior of the PM concentrations, and air-mass back trajectories for that specific city.

Keywords: Particulate matter; Long-range transport; Local characterization; Ion sum; Number concentration

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1. Introduction

It has been shown that a number of health effects (e.g. mortality, morbidity, respiratory and cardiovascular problems, etc.) are related to elevated concentration levels of particulate matter (PM) (WHO, 2003; de Hartog et al., 2003; Pope et al., 2002; Laden et al., 2000). Even though some studies have shown that coarse particles (diameter $>2.5\,\mu$m) cause some of these health effects (Brunekreef and Forsberg, 2005; Tiittanen et al., 1999), there have been a strong evidence that fine particle (FP, diameter $<2.5\,\mu$m) and also ultrafine particles (UFP, diameter $<0.1\,\mu$m) are more harmful (Penttinen et al., 2004; Hoek et al., 2002; Schwartz et al., 1999; Peters et al., 1997). In addition, the adverse health effects of PM may not be due to aerosol particle mass concentration, but instead, due to aerosol particle number concentration (e.g. Penttinen et al., 2001) or even due to the chemical content and physical properties of aerosol particles.

According to the EU directives on air quality, the 24-h average of the PM_{10} (particulate mass concentrations of aerosol particles smaller than $10\,\mu$m in diameter) concentrations is not supposed to exceed a maximum value of $50\,\mu g\,m^{-3}$ for more than 35 times in a calendar year and the annual average is not supposed to exceed $40\,\mu g\,m^{-3}$. According to the Council Directive (96/62/EC) on ambient air quality assessment and management, member states have to report the occurrence of levels exceeding the limit values and also the reasons for these exceedances. In Finland, the first daughter directive was brought into force in the Air Quality Degree in September 2001. In 2002, the PM_{10} concentrations did not exceed the limit value in the Helsinki Metropolitan Area (Haaparanta et al., 2003). However, in both 2003 and 2005 the limit values were exceeded in street canyon environments (Myllynen et al., 2004).

Re-suspended dust episodes are a common problem in many sub-arctic cities during spring time (e.g. Kukkonen et al., 1999; Johansson et al., 1999; Pohjola et al., 2002). Although the annual average PM_{10} concentrations in these cities may be lower than those in major Central European cities (Ruuskanen et al., 2001), the PM concentrations characteristically exceed the 24 h limit value in various episodic conditions such as re-suspended dust and winter-time temperature inversion induced episodes. This former category of episodes commonly occurs in spring and to a lesser extent in autumn. The street sanding, the use of studded tires, and the meteorological conditions are the major reasons for the elevated PM_{10} concentrations during such episodes (e.g. Hosiokangas et al., 1999, 2004; Kupiainen and Tervahattu, 2004; Kupiainen et al., 2003; Räisänen et al., 2003, Kukkonen et al., 2001).

Long-range transport (LRT) of aerosol particles also mainly contributes to the elevated urban $PM_{2.5}$ and to a less extent to PM_{10} concentrations. It has been evaluated that more than half of the $PM_{2.5}$ concentrations is due to LRT for the Helsinki urban areas (e.g. Karppinen et al., 2004; Vallius et al., 2003). Some of the LRT episodes observed in the Helsinki Metropolitan Area were previously investigated by Niemi et al. (2004, 2005) and Tervahattu et al. (2002, 2004).

Valkama and Kukkonen (2004) have compiled an inventory of episodes within COST 715 action and the FUMAPEX project. A total of 21 episodes from seven cities or metropolitan areas in six countries were examined. Partly based on this inventory, Kukkonen et al. (2005) analysed and evaluated selected local-scale PM_{10} episodes in four European cities (Helsinki, Oslo, London and Milan) in relation to prevailing meteorological conditions, local emissions, and regionally and long-range transported background concentrations. They aimed at a structured and homogeneous analysis in all the four cities, and selected for the analysis recent episodes that were predominantly caused by various local emission sources; these episodes can also be considered characteristic for each region, in terms of their frequency of occurrence. The best meteorological predictors for the elevated concentrations of PM_{10} were found to be the temporal evolutions of temperature inversions and atmospheric stability and in some cases the wind speed.

In the current study we designed, applied, and evaluated two novel methods to distinguish between LRT and local-pollution (LP) episodes. The first method is based on evaluation of the particle number concentrations ratio between the accumulation (diameter >90 nm) and Aitken (diameter between 25 and 90 nm) modes. The second one is a linear regression model that defines a proxy variable for LRT based on the available measurements of sulfate, nitrate, and ammonium at the EMEP (Co-operative program for monitoring and evaluating of the long-range transmission of air pollutants in Europe) stations. The latter method was previously presented by Karppinen et al. (2004).

We analysed selected re-suspension and LRT episodes of PM_{10} in the Helsinki Metropolitan Area during 2002. The criterion for selecting these particular episodes was the 24-h average of PM_{10} exceeded the limit value of $50\,\mu g\,m^{-3}$ at several air quality monitoring sites. In the analyses we used these two novel methods together with the PM concentrations, meteorological conditions, and air mass back trajectories to systematically study the factors that contribute to these exceedances. The aim was also to develop easy-to-use methods for the air quality assessment and management to be used by the local authorities.

2. Materials and experimental set-up

2.1. Description of the Helsinki Metropolitan Area

Helsinki Metropolitan Area and its surrounding regions are situated on a fairly flat coastal area by the Baltic Sea at the latitude of 60°N (e.g. Hussein et al., 2004b). Because the Gulf Stream and the prevailing global atmospheric circulation have a warming effect, the climate is relatively mild compared with many other areas in the same latitudes. The Helsinki Metropolitan Area comprises four cities (Helsinki, Espoo, Vantaa, and Kauniainen) with a total area of $743\,km^2$ and population of about 950,000 by the end of year 2001.

In the Helsinki area the PM_{10} concentrations in street level air are dominated by the combustion, non-combustion and suspension emissions originating from vehicular traffic, and the LRT (e.g. Hussein et al., 2005b). All the largest stationary sources, such as power plants use natural gas or coal and heavy fuel oil additive. The contribution of minor residential heating plants and industrial plants on the total PM emissions is negligible. At the ground level, the traffic emissions have a larger relative influence than the stationary emissions, which are mostly released from higher altitudes.

2.2. Aerosol particle measurements

2.2.1. Particulate mass

The PM_{10} and $PM_{2.5}$ concentrations were monitored as a part of the municipal air quality-monitoring network of the Helsinki Metropolitan Area (Fig. 1 and Table 1). PM_{10} concentrations were measured continuously with β-attenuation monitors (Eberline FH 62 I-R, Eberline Instruments, GmbH, Germany) or with the Tapered Element Osccillating Microbalance (TEOM 1400ab, Rupprect and Patashnick Co.). At Vallila and Kallio monitoring sites also $PM_{2.5}$ concentrations were monitored with the β-attenuation method (Eberline FH 62 I-R).

Because none of the above mentioned continuous methods used to monitor PM_{10} was a reference method determined in the standard EN 12341, the equivalency of these methods with the reference was demonstrated in a measurement campaign conducted by the Finnish Meteorological Institute and the Helsinki Metropolitan Area Council (Sillanpää et al., 2002). The comparison showed that both continuous methods gave equivalent results with the reference method and thus no correction factors were required. The equivalency of the β-attenuation method was tested by the Finnish meteorological Institute at a rural EMEP background station in Finland with low PM_{10} concentrations (Salminen and Karlsson, 2003).

The flow rates of the PM_{10} monitors were calibrated with a mass flow metre every 6 months. The mass measurements of the TEOM monitors were calibrated once a year by determining the oscillation frequency with a known mass. For the β-attenuation method, the mass measurements were calibrated by measuring the β absorption of the calibration plate once a year.

2.2.2. Ultrafine and fine particle number size distributions

The ultrafine and fine particle number size distributions have been measured continuously in Helsinki since 1997 with a Differential Mobility Particle Sizer (DMPS) system (Hussein et al., 2004b). The measurement site during year 2002 was located at Kumpula, Helsinki, 3 km from the downtown. The surrounding area of Kumpula is typical urban background. There was a major highway nearby (less than 200 m) the measurement site. The area itself is populated by residential buildings except for the western side, which was mostly covered by small trees and greenswards.

The aerosol particle measurements took place on the fourth floor of the Department of Physical Sciences of the University of Helsinki. The measured particle diameter range covered by the DMPS was 8–400 nm (dry sizes) with 6-min resolution. The DMPS measurement is based on differential mobility classification of aerosol particles. The differential mobility analysis consists of three major parts:

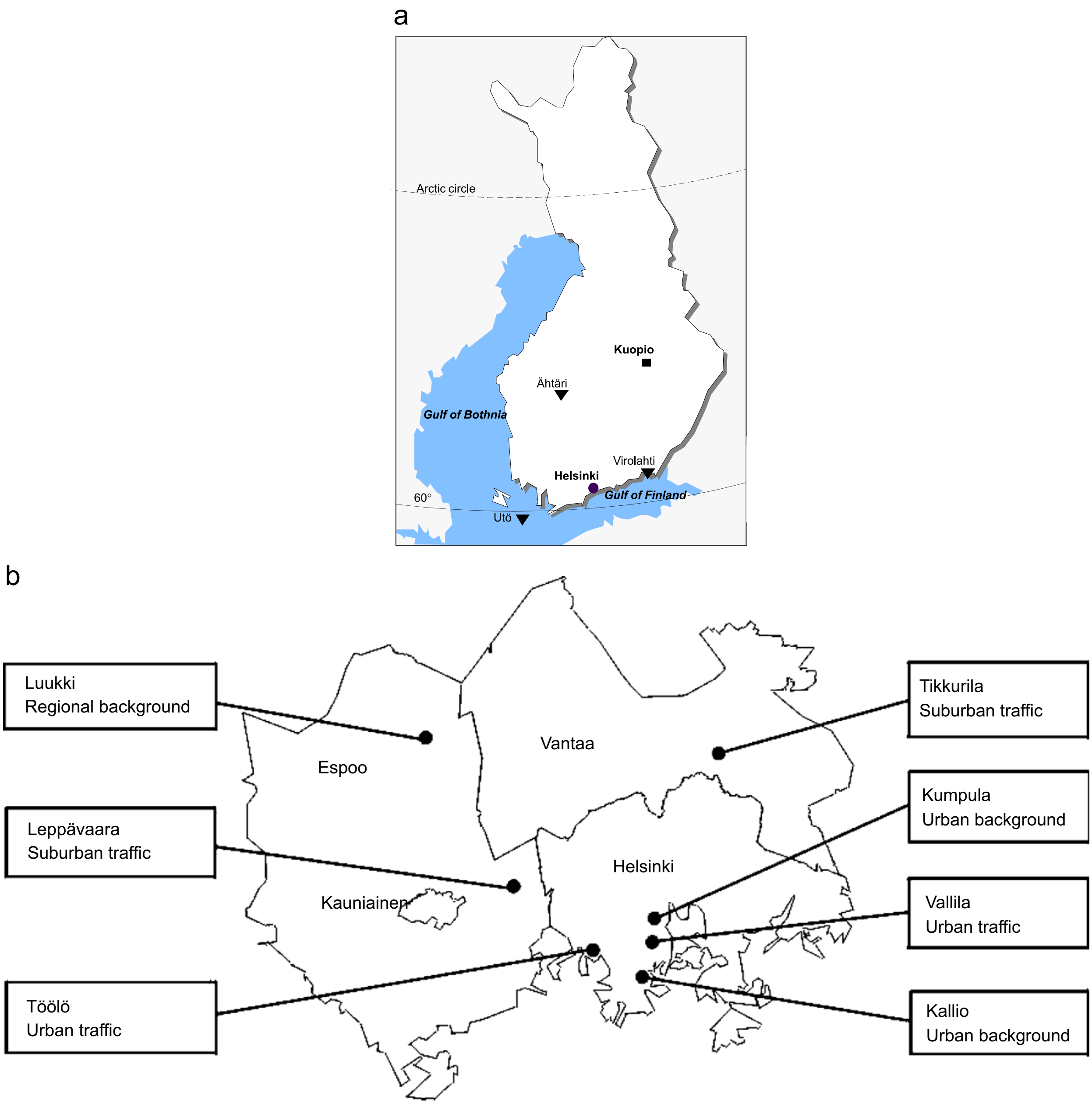

Fig. 1. (a) Location of the Helsinki Metropolitan Area and its three nearest EMEP monitoring stations. (b) The air quality monitoring network of the Helsinki Metropolitan Area Council. The location of the joint measurement station of the Helsinki University and the Finnish Meteorological Institute in Kumpula is also included in the map.

charging aerosol particles with a 74 MBq KR-85 neutralizer (Liu and Pui, 1974); aerosol particle classification by a differential mobility analyser (DMA) TSI 3071 (Knutson and Whitby, 1975); and counting aerosol particles with a Condensation Particle Counter (CPC) TSI 2025 (Stolzenburg and McMurry, 1991) and TSI 3010 (Quant et al., 1992). The aerosol flow was 1.5 litre per minute (lpm) and the sheath flow was 8.5 lpm. The sheath air was circulated to the DMA after drying and filtration (Jokinen and Mäkelä, 1997; Birmili et al., 1999). Sampling line was 2 m long stainless steel tube with 4 mm inner diameter; no special inlet was used except for a rain cover only.

In the weekly maintenance, both flows and CPC zero concentration were checked. Yearly maintenance

Table 1
PM and NO concentrations at the air quality monitoring sites in the Helsinki Metropolitan Area in 2002

Station	Station classification	PM_{10} median ($\mu g\, m^{-3}$)	PM_{10} 24 h max ($\mu g\, m^{-3}$)	PM_{10} >50 ($\mu g\, m^{-3}$) no of days	$PM_{2.5}$ median ($\mu g\, m^{-3}$)	$PM_{2.5}$ 24 h max ($\mu g\, m^{-3}$)	$PM_{2.5}/PM_{10}$ median ($\mu g\, m^{-3}$)	NO median ($\mu g\, m^{-3}$)
Töölö	Urban traffic	19	97	32				35
Vallila	Urban traffic	18	137	19	8	52	0.48	11
Kallio	Urban background	14	85	10	7	43	0.57	4
Leppävaara	Suburban traffic	17	144	27				10
Tikkurila	Suburban traffic	17	118	22				23
Luukki	Regional background	10	58	2				1

included system calibration and cleaning. The instruments were calibrated according to Aalto et al. (2001).

2.3. Meteorology

The meteorological conditions in this study were obtained from the measurement station in the city center (Kaisaniemi) and three other measurement stations at the Helsinki–Vantaa airport, Kivenlahti mast, and Isosaari. Helsinki–Vantaa is a synoptic station located at the airport in a suburban area northern part of the Helsinki Metropolitan Area. Kivenlahti is a World Meteorological Organisation (WMO) station and it is a 327 m high radio tower situated in a partly rural, partly suburban environment. Isosaari station is located on a small island.

The Monin–Obukhov length, ambient air temperature, and pressure in the Helsinki city center were evaluated using a meteorological pre-processing model (MPP-FMI) that has been adapted for an urban environment (Karppinen et al., 2000a). This model was originally based on the energy budget method of van Ulden and Holtslag (1985). The model utilises meteorological synoptic and sounding observations, and its output consists of estimates of the hourly time series of the relevant atmospheric turbulence parameters and the boundary layer height. The computation is based on a combination of the data from the stations at Helsinki–Vantaa airport and Isosaari.

2.4. Air mass trajectories

We investigated back trajectories (at 760 and 1450 m altitude and 96 h backward) of air masses arriving (at 00 h UTC) in Helsinki during the period of interest. We evaluated the back -trajectories with the HYSPLIT_4 model (Draxler and Hess 1998), which was developed by NOAA/ARL. HYSPLIT_4 is a single-particle Lagrangian trajectory dispersion model. The model utilises the Global FNL meteorological archive with a spatial resolution of $190 \times 190\,km^2$.

3. Determination of LRT and LP episodes: methods development

3.1. Ion sum

We utilised a linear regression model to evaluate the LRT contribution to $PM_{2.5}$ (Karppinen et al., 2004). We have utilised the data of the three EMEP-stations that are located nearest to Helsinki. Their locations are shown in Fig. 1: Utö (59°47′N, 21°23′E), Ähtäri (62°35′N, 24°12′E) and Virolahti (60°32′N, 27°41′E). The following ion concentrations are measured daily at the EMEP stations: (i) sulphate (SO_4^{2-}), (ii) the sum of nitrate (NO_3^-) and nitrogen acid (HNO_3), and (iii) the sum of ammonium (NH_4^+) and ammonia (NH_3). The sulphate, nitrate and ammonium ions are in particulate form, while nitrogen acid and ammonia are gaseous compounds in atmospheric conditions.

The above mentioned measurements (i), and (ii) together with (iii) are reported as equivalent masses of sulphur and nitrogen, respectively. We therefore define the so-called ion sum as follows

$$C_{ion} = 3.0[{SO_4}^{2-}]_S + 4.4[({NO_3}^- + HNO_3)]_N + 1.3[({NH_4}^+ + NH_3)]_{N,} \qquad (1)$$

where the subscripts S and N denoted that the mass had been given as the equivalent mass of sulphur or nitrogen. These values were converted to equivalent masses of the ions SO_4^{2-}, NO_3^- and NH_4^+, using the conversion factors 3.0, 4.4 and 1.3, respectively. This conversion was necessary in order to treat the particulate concentration variables in a comparable manner.

The C_{ion} variable (Eq. (1)) was assumed to be a suitable proxy variable for LRT. However, it should be noted that this variable contained in part measurements of gaseous substances (HNO_3, NH_3), and on the other hand, that LRT always contains also other compounds, such as elemental and organic carbon.

Each of the three ion terms in Eq. (1) could be assumed to be even alone a suitable proxy variable for LRT. However, it was convenient to define a combined variable that was based on all three measurements of ion concentrations. This variable represented a larger fraction of the $PM_{2.5}$ mass than any of its components, which made it a better candidate for prediction of temporal variations of LRT.

We utilise a combination of data from the three nearest EMEP stations to Helsinki, in order to smooth out any possible disturbances caused by local emission sources. This procedure also removed the difficulties that might have arised from missing data at any individual station. Whenever possible, we utilised an interpolated value of the ion sum, defined as

$$C_{\mathrm{ion}} = \sum_{i=1}^{n} \chi_i C_{\mathrm{ion},i}, \tag{2}$$

where the subscript i referenced to the EMEP stations considered, n was the total number of stations to be applied, χ_i was the weight coefficient and $C_{\mathrm{ion},i}$ was the ion sum at a specific site. The weight coefficient was defined on the basis of the number of EMEP stations with available data, as the normalised inverse value of the distance between the urban measurement location and the EMEP station. The normalization implies simply that the sum of all χ_i values is equal to unity, and using the inverse values of the distance results in larger weight coefficients for the EMEP stations that are closer to the location considered.

3.2. *Accumulation-to-Aitken modes concentrations ratio*

Based on the short and long-term data analysis of the particle number size distributions within the Helsinki Metropolitan Area, a LRT pollution episode is characterised with elevated number concentrations for aerosol particles larger than 100 nm (Hussein et al., 2004b, 2005a–c). Typically, local traffic emissions dominate the particle number concentrations of UFP within the Helsinki Metropolitan Area during the daytime and especially during traffic rush hours (e.g. Hussein et al., 2004b, 2005b).

In general, and in the absence of LRT pollution episodes and during traffic rush hours, the UFP contributes more than 90% of the total number concentration of FP in the urban air of the Helsinki Metropolitan Area and nearby a highway (e.g. Hussein et al., 2004a, b, 2005b). On the other hand, during night time and in the absence of a LRT episode, the UFP contribution to the total number concentrations can be as high as 80% in the urban air of the Helsinki Metropolitan Area.

Based on the meteorological conditions, PM concentrations, back trajectory analysis, ion sums, and the number concentration ratio between the accumulation mode particles (diameter >90 nm) and the Aitken mode particles (between 25 and 90 nm in diameter), we can define the existence and strength of LRT episodes. The stronger the episode is the higher the ion sum and the concentration ratio.

4. Results and discussion

The annual median values of PM_{10}, $PM_{2.5}$, $PM_{2.5}/PM_{10}$ ratio, and the NO concentrations at the different air quality monitoring stations within the Helsinki metropolitan area are listed in Table 1. The highest 24-h averages of PM_{10} and $PM_{2.5}$ and the number of exceedances for the daily average over the limit values of $50\,\mu g\,m^{-3}$ are also given. Most of these exceedances occurred in March and April. A small number of the exceedances were observed in February, August, and October.

The average values of the PM_{10}, $PM_{2.5}$, NO concentrations, and $PM_{2.5}/PM_{10}$ at the urban traffic monitoring site Vallila (referred as "utr" in the tables) during the selected episodes are listed in Tables 2a and 2b. The PM_{10} concentrations at the regional background monitoring site in Luukki (referred as "rb" in the tables) and the ratios of PM_{10} concentrations at the regional background and urban traffic monitoring site are also presented in Table 2a. The interpolated ion sums from the EMEP stations for Helsinki (Eq. (2)) are given in

Table 2a
PM_{10}, $PM_{2.5}$, and NO concentrations at the urban traffic (Vallila) monitoring site during the episodes in 2002

Episode	Urban traffic PM_{10} average ($\mu g\,m^{-3}$)	Urban traffic PM_{10} max ($\mu g\,m^{-3}$)	Urban traffic $PM_{2.5}$ average ($\mu g\,m^{-3}$)	Urban traffic $PM_{2.5}$ max ($\mu g\,m^{-3}$)	Urban traffic $PM_{2.5}$/PM_{10} ave	Regional backgr PM_{10} average ($\mu g\,m^{-3}$)	Regional backgr PM_{10} max ($\mu g\,m^{-3}$)	PM_{10}rb/PM_{10}utr	Urban traffic NO average ($\mu g\,m^{-3}$)	Classification
20–22 February	60	78	6	6	0.1	6	11	0.10	11	Local
14–15 March	48	55	10	12	0.2	7	10	0.14	45	Local
17–21 March	47	78	30	52	0.6	33	58	0.70	11	LRT
24 March–4 April	41	63	11	16	0.3	13	27	0.33	16	Local + LRT
7–11 April	87	137	27	35	0.3	29	35	0.35	65	Local + LRT
12–14 April	72	79	34	38	0.5	48	52	0.67	10	LRT
12–15 Aug	42	68	24	34	0.6	35	50	0.84	13	LRT
24–30 Aug	38	64	23	41	0.6	21	29	0.71	16	LRT
21–22 October	77	99	18	25	0.2	12	13	0.16	143	Local

The values are based on the 24 h averages.

Table 2b
The interpolated ion sums and the particle number concentrations at the urban background (Kumpula) monitoring site during the episodes in 2002

Episode	Ion sum ($\mu g\,m^{-3}$)			PN (cm^{-3})			Acc/Ait			Classification
	Average	Min	Max	Average	Min	Max	Average	Min	Max	
20–22 February	1.5	1.2	1.9	17270	12607	21800	0.13	0.12	0.14	Local
14–15 March	2.5	2.5	2.5	23137	15657	30617	0.21	0.21	0.21	Local
17–21 March	10.7	5.6	15.8	21703	9967	34135	0.85	0.45	1.23	LRT
24 March–April 4	3.8	1.7	6.1	16153	9847	31363	0.29	0.17	0.50	Local + LRT
7–11 April	8.2	5.4	10.9	20425	18249	22482	0.41	0.34	0.50	Local + LRT
12–14 April	11.9	12.3	13.5	15941	15831	19180	0.85	0.64	0.95	LRT
12–15 August[a]	9.3	7.7	10.6	15824	13860	17788	0.75	0.72	0.79	LRT
24–30 August	6.6	4.6	9.1							LRT
21–22 October	1.6	1.5	1.7	18904	15433	22377	0.26	0.22	0.30	Local

The values are based on the 24 h averages.
[a] Ntot ja Acc/Ait for the 12–13 August.

Table 2b. Also the number concentrations and the accumulation to Aitken mode ratios at the Kumpula monitoring site are included in this Tables 2a and 2b. All these values are based on 24-h averages. A summary of the main meteorological features of the episodes are given in Table 3.

There was a good correlation ($R^2 = 0.73$) between the 24-h averages of $PM_{2.5}$/PM_{10} at the urban traffic site (Vallila) and the accumulation to Aitken mode ratio at the urban background site (Kumpula). There was a moderate to good correlation also between $PM_{2.5}$/PM_{10} ratio and the interpolated ion sum, and between the ion sum and the accumulation to Aitken mode ratio ($R^2 = 0.44$ and 0.58, respectively). The scatter plots for these correlations are shown in Figs. 2a–c, which also show the grouping of the different types of episodes.

4.1. LP episodes

We characterised the episodes as being LP, LRT, or a mixture between LP and LRT episodes. The episodes in 20–22 February, 14–15 March, and in 21–22 October showed a very clear LP characteristics. The $PM_{2.5}$/PM_{10} ratios (<0.2) at the urban traffic site were significantly lower than the annual median value and the lowest among the episodes studied. The PM_{10rb}/PM_{10utr} ratios were also low (<0.2). In the Helsinki Metropolitan Area the annual average of the accumulation mode at the urban background monitoring site (Kumpula) is 0.35 times Aitken mode. The average interpolated ion sum value for the year 2002 was 4.2 $\mu g\,m^{-3}$. During these LP episodes the interpolated ion sums and the accumulation to Aitken mode ratios were

Table 3
Summary of the main meteorological variables and features of the episodes in the Helsinki metropolitan area in 2002

Episode duration	Synoptic scale meteorological conditions	Local scale meteorological conditions	Type on temperature inversion	Main pollutant source for PM_{10}	Brief classification of the episode
20–22 February	High-pressure ridge	Low wind speed, stable stratification on the 20–21, high wind speed on the 22	Nocturnal ground based on the 20th	Local suspended matter	Traffic and wind induced suspension episode
14–15 March	High-pressure area	Low wind speed	Nocturnal ground based	Local suspended matter	Spring dust episode, traffic induced suspension
17–21 March	High-pressure ridge	Moderate wind speed, high daily temperatures	No inversion	Long-range transport	Long-range-transport
24 March–4 April	Strong centre of high pressure	Very warm weather, low wind speeds	Short term nocturnal inversion	Local suspended matter and long-range transport	Spring dust episode, traffic induced suspension mixed with long-range-transport
7–11 April	High-pressure area	Low wind speed, stable stratification, temperature inversion	Ground-based, radiation	Local suspended matter and long-range transport	Spring dust episode, traffic induced suspension mixed with long-range-transport
12–14 April	High-pressure area	Moderate wind speed	No inversion	Long-range-transport	Long-range-transport
12–15 August, 24–30 August	Extensive high-pressure area	Light winds	No inversion	Long-range transport	Long-range-transport
21–22 October	High-pressure ridge	Temperature inversion, low wind speed, stable stratification	Strong ground based, radiation	Local suspended matter	Autumn dust episode, traffic induced suspension

low (1.5–2.5 $\mu g\,m^{-3}$ and 0.13–0.26, respectively), indicating a negligible contribution from LRT episodes.

Typical meteorological features for these LP episodes were low wind speeds, stable stratification and ground based inversions.

Based on the various studies by e.g. Hosiokangas et al. (1999, 2004), Kupiainen et al. (2003) and Kupiainen and Tervahattu, (2004), Räisänen et al. (2003) we concluded that the source of the particles was traffic induced suspension from the street surfaces. The origin of the suspended material was street sanding and the use of studded tires. Due to low wind speeds and ground based inversions the primary tailpipe emissions contributed also to the high concentrations, but the effect of suspension was dominating.

On 22 February, however, the wind speed increased from moderate to strong, which significantly contributed to re-suspension of aerosol particles. Also the back-trajectories of air masses (not shown here) confirm the local origin of the aerosol particles. During all these time periods the air masses arriving at Helsinki originated from the relatively non-polluted regions in the westerly to northerly directions.

4.2. LRT episodes

Three LRT episodes with 24-h average PM_{10} concentrations exceeding 50 $\mu g\,m^{-3}$ were clearly observed during 17–22 March, 12–15 August, and in 24–30 August 2002 (Tables 2a and 2b). These episodes were characterised with relatively high $PM_{2.5}$ concentrations and high $PM_{2.5}$–PM_{10} ratios (0.5–0.6) at the urban traffic site. The PM concentrations at the regional background site were rather close to those at the urban traffic sites (ratio $\geqslant 0.7$),

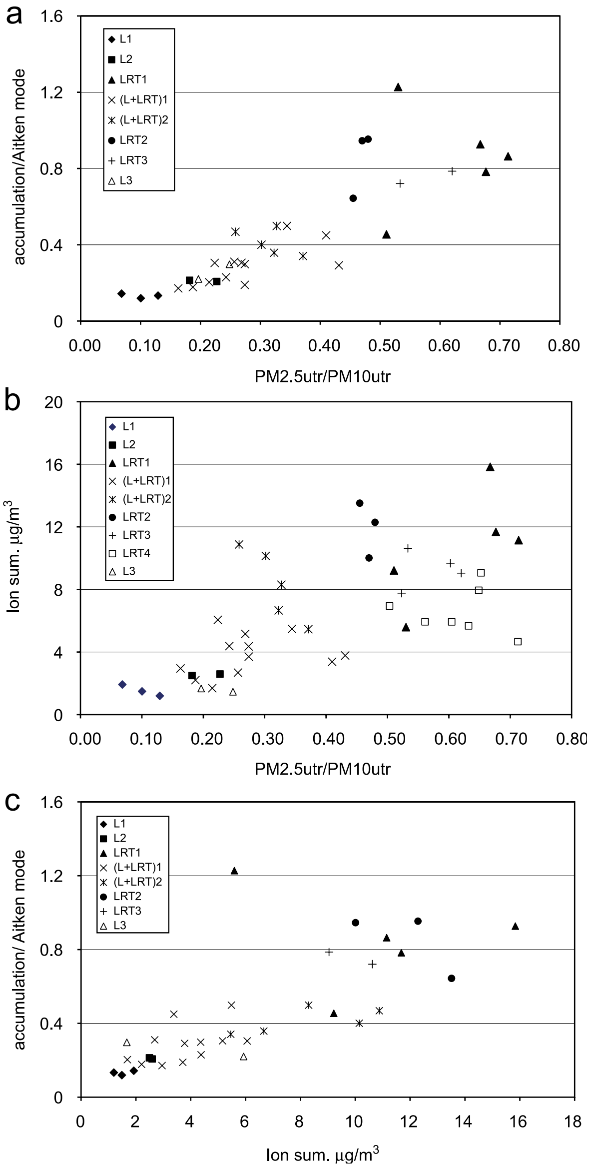

Fig. 2. (a) The correlation between the $PM_{2.5}/PM_{10}$ ratios at the urban traffic monitoring site and the accumulation to Aitken mode ratios measured at the urban background site. (b) The correlation between the $PM_{2.5}/PM_{10}$ ratios at the urban traffic monitoring site and the interpolated ion sums. (c) The correlation between the interpolated ion sums and the accumulation to Aitken mode ratios.

and the NO concentrations were low. Furthermore, the interpolated values for the ion sums and the accumulation to Aitken mode ratios were high (6.6–11.9 $\mu g\,m^{-3}$ and 0.75–0.85, respectively). Typical characteristics for the local scale meteorological factors during the LRT episodes were light to moderate wind speeds, high temperatures, and lack of inversions.

The analyses for the periods during 17–21 March and 12–15 August indicated that the air masses arrived at Helsinki from the south or south-east. The trajectories for the period of 24–30 August showed that the air masses arrived at Helsinki from various directions. During 24–26 August, the air masses arrived from north-east and east. After that they turn to south and south-west (27–30). This indicated that the air masses may contain substantial amounts pollutants originating outside the Finnish borders.

These three LRT episodes were previously characterised and sources identified by Niemi et al. (2004, 2005). Backward air mass trajectories, satellite detections of fire areas, dispersion modeling results, as well as chemical analyses indicated that large-scale agricultural field burning in the Baltic countries, Belarus, Ukraine, and Russia were the source of the episode during March and the emissions from wildfires in Russia and other Eastern European countries were the source for the LRT episodes during August.

Based on the PM concentrations, meteorological conditions and back trajectories alone the episode during 12–14 April was difficult to be distinguished from the preceding episode (7–11 April) with a strong local characteristics. However, the high ion sum and the high accumulation to Aitken mode ratio supported the strong LRT characteristic of this episode. Also, NO concentrations were lower than during the preceding episode whereas the PM_{10rb}/PM_{10utr} and $PM_{2.5utr}/PM_{10utr}$ ratios were higher. The meteorological conditions and the air mass trajectories of this episode are discussed in the following section.

4.3. Episodes with LRT and LP contribution

We classified the episodes in 24 March–4 April and 7–11 April as being caused mainly by local sources, i.e. LP, with some contribution from LRT. The $PM_{2.5}/PM_{10}$ ratios at the urban traffic site and the PM_{10rb}/PM_{10utr} ratios were higher (0.3 and 0.3–0.4, respectively) than during the above mentioned local episodes. The interpolated values for the ion sum (3.8–8.2 $\mu g\,m^{-3}$) and the accumulation to Aitken mode ratios (0.29–0.41) were slightly higher than for the local episodes.

During the period 24 March–4 April the dry (relative humidity 50–70%) and warm weather, combined with low wind speeds (mostly 3–5 $m\,s^{-1}$) was favourable for an episode of suspended dust in

Helsinki. The southerly winds, on the other hand, seem to indicate that there was some contribution from the LRT.

During this episode the trajectories first arrive from easterly (24–25), then from south-westerly (26–29) or a westerly (30 March–4 April) directions. The over-all transport pattern indicates that this is a "mixed" type of an episode, containing contributions both of local emissions and of LRT.

The local episode in 7–11 April and the LRT episode in 12–14 April differed meteorologically, especially regarding wind speed and air temperature. In the early stages of the episode, from 7 to 10 of April, the wind speed was low or moderate ($<3\,\mathrm{ms}^{-1}$), but increased towards the end of the period. The relative humidity was low, 40–70%. The wind direction varied considerably, from north and south-west to north and south-east. During the 12 to 14 of April, the winds were predominantly from south-east, and somewhat stronger, up to 4–6 m/s. The relative humidity was higher, c. 50–80%.

The atmosphere was stable (i.e., $L>0$) or neutral during the whole period considered, except for short periods on 7th and 8th April. There were several periods of very stable ($\mathrm{L}^{-1}>0.1\,\mathrm{m}^{-1}$) atmospheric conditions, especially at night-time. The measured temperature profiles at midnight showed moderate to strong ground-based inversions during 7–12 April (Fig. 4). On the 13th and 14th a cold front passed over Southern Finland and the sky became overcast, which caused the break-up of the inversions.

The back-trajectories for 7–11 April, 2002, show a complicated pattern. The trajectories arrived to Helsinki mainly from southerly directions. However, there was considerable "looping" and veering of trajectory lines shortly before they arrived, and the air-masses actually had their origins at the eastern directions. The over-all pattern is rather complicated, which indicates that frontal activities were affecting the transport. The same pattern appears at both elevations (760 and 1450 m), showing that a deep air-layer was being affected. The over-all pattern of trajectories combined with the prevailing weather (high pressure over Finland) seems to indicate that the episode was mainly of local origin, but may have had some contribution from long-range transport, as well.

The analysis for the period 12–14 April, 2002, is clearer. The back-trajectories were arriving from south or south-west. This indicates that the air-masses may have contained substantial amounts of pollutants originating outside the Finnish borders.

As an example, the evolution of the episodes observed in 7–14 April is described in Figs. 3–5. In Fig. 3 the concentrations of PM_{10}, $\mathrm{PM}_{2.5}$, and NO at the urban traffic site (Vallila), the number concentrations at the urban background site (Kumpula) and the interpolated ion sum values from the EMEP sites are depicted. The measured temperature profiles at the Kivenlahti mast and the air mass back trajectories during the episode are given in Figs. 4 and 5.

5. Summary and conclusions

We have studied the episodes of high PM concentrations in the Helsinki Metropolitan Area. An episode can be determined as a period of abnormally high concentrations of air pollutants. In this paper the criterion for selecting these particular episodes was that the 24 h average concentration of PM_{10} exceeded $50\,\mu\mathrm{g\,m}^{-3}$ at several air quality monitoring sites in the Helsinki metropolitan area. These situations contribute to the exceedances of the 24 h EU limit value for PM_{10}. Local authorities are obliged to report these episodes and the factors contributing to them to the Ministry of the Environment and further to the European Commission. They also have to inform the public on these events.

We studied the PM_{10} and $\mathrm{PM}_{2.5}$ concentrations and the fraction of fine particles in PM_{10} at an urban traffic and regional background air quality monitoring sites. We also applied two novel methods to distinguish the LRT episodes from episodes of a more LP origin. The first method was based on the evaluation of the ratio of particle number concentrations in the accumulation and Aitken modes. The second was a proxy variable (interpolated ion sum) based on the available measurements of sulfate, nitrate and ammonium at the nearest EMEP stations. Meteorological parameters and air mass back trajectories were also used, to evaluate the findings based on the two novel methods.

We distinguished nine episodes of high PM_{10} concentrations during the year 2002. Three of these episodes were clearly of local origin. They were characterised by low average fraction of $\mathrm{PM}_{2.5}$ (<0.2) in PM_{10} at the urban traffic monitoring site, low ratio (<0.2) between PM_{10} concentrations at the regional background site and at the urban traffic

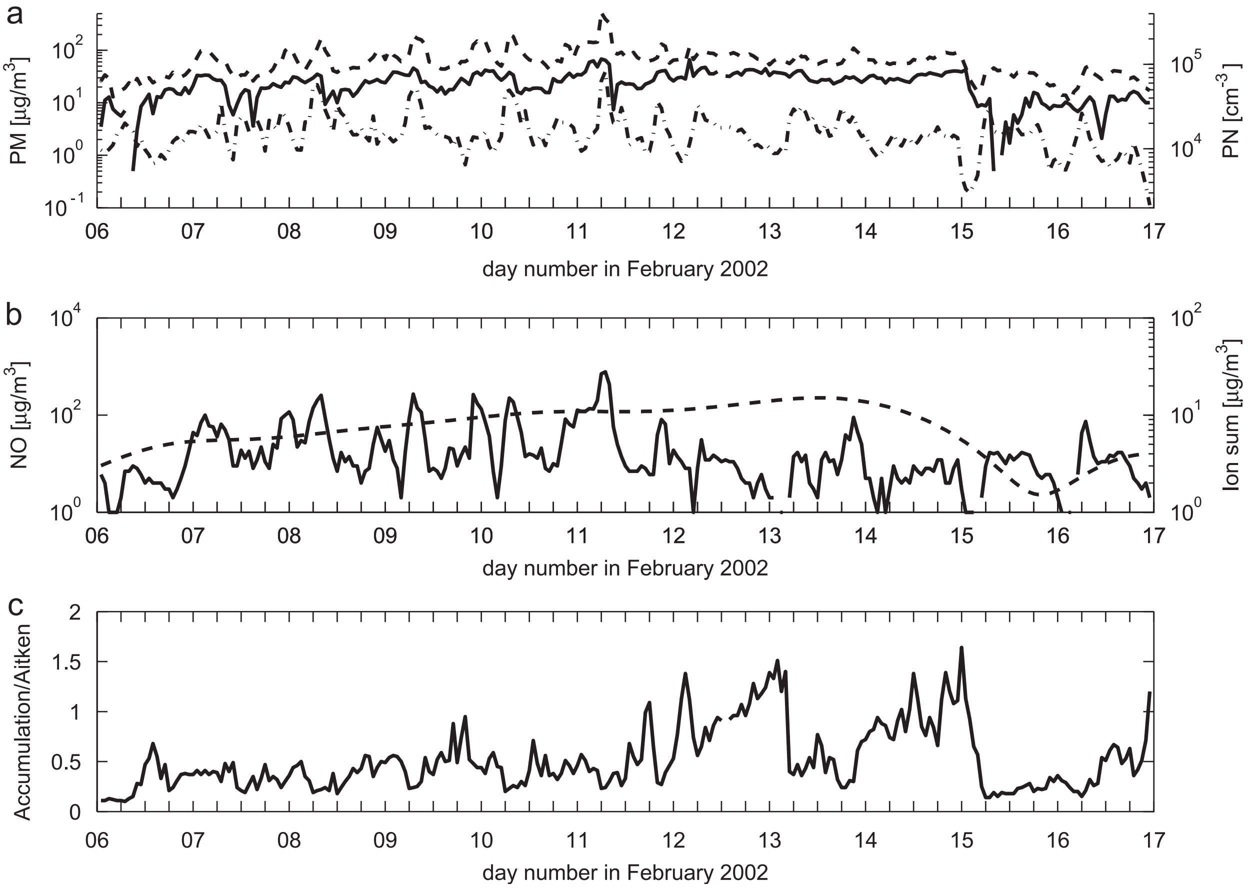

Fig. 3. Measurements from the 6 to 17 of April 2002. (a) Particulate matter at the urban traffic monitoring site Vallila (PM_{10} dashed line and $PM_{2.5}$ solid line, both in left axis), and the particle number concentration at the urban background monitoring site Kumpula (dash-dot line in right axis). (b) The NO concentrations at Vallila (dashed line, left axis) and the interpolated ion sum of the background stations (solid line, right axis). (c) The accumulation to Aitken mode ratio at the at the urban background monitoring site (Kumpula).

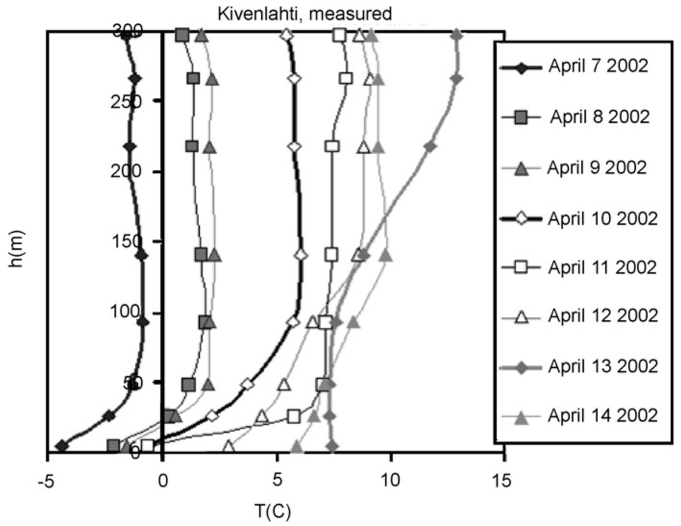

Fig. 4. The measured temperature profiles at the Kivenlahti mast during the episodes in 7–14 April.

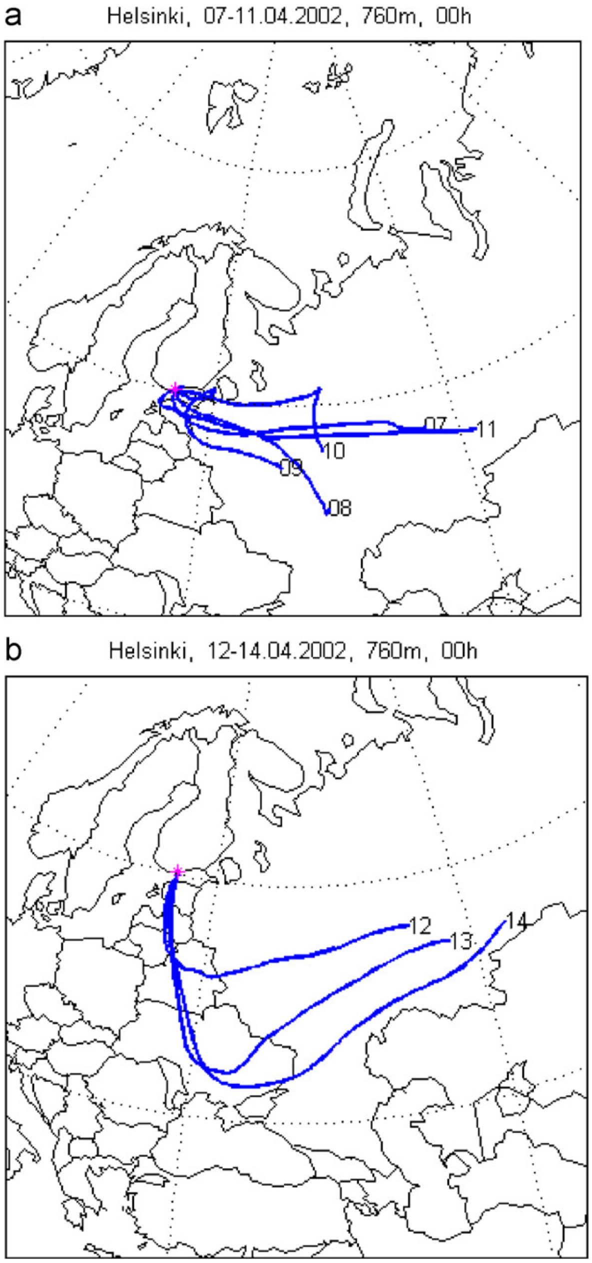

Fig. 5. Air mass back trajectories during the episodes in 7–14 April.

site, low average ion sums (1.5–2.5 μg m^{-3}) and low accumulation to Aitken mode ratios (0.13–0.26). Four of the episodes had distinct LRT characteristics: a high fraction of fine particles (0.5–0.6) in PM$_{10}$ at the urban traffic site, a high ratio between PM$_{10}$ concentrations at the regional background site and at the urban traffic site (⩾0.7), high interpolated values for the ion sum (6.6–11.9 μg m^{-3}) and high accumulation to Aitken mode ratios (0.75–0.85). During the remaining two episodes there was some contribution from both local sources and LRT.

A detailed analysis of meteorological variables and air mass back-trajectories gave support to these findings. Meteorological features such as extensive high-pressure areas, lack of local inversions and moderate to high wind speeds as well as air masses arriving from polluted areas indicate a higher probability of LRT. On the other hand certain features of local meteorological conditions such as low wind speeds and occurrence of strong ground based temperature inversions are typical of the LP episodes.

The results presented in the current study are applicable to the Helsinki Metropolitan Area and similar cities. Developing these methods for other cities require analyses of the meteorological conditions, behaviour of the PM concentrations, and air mass back trajectories for that specific city. However, the method is not applicable to all LRT episodes, such as those of mineral dust. Also, identifying the sources of LRT episodes requires further chemical analyses.

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