VARIATIONS OF CONCENTRATION OF AEROSOL PARTICLES (<10 μm) IN VILNIUS

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Long-term observations of atmospheric constituents such as particulate matter (PM) are increasingly important in evaluating their adverse effects on human health and climate. Time series (2005–2007) of the hourly mass concentrations of PM10 recorded at three air quality monitoring (AQM) sites characterized by a different exposure to traffic emissions in Vilnius city have been used for the investigation of spatial and temporal variations as well as the input of pollutants to PM10 levels by long-range transport of air masses. The distribution plots of PM10 24-h mean mass concentrations showed a unimodal lognormal distribution with the mode centered on 34.4, 21.6, and 19.9 μg/m^3, respectively, at the traffic-exposed (Žirmūnai), semi-urban (Senamiestis), and urban background (Lazdynai) sites. The difference between traffic-exposed and urban background data indicates average concentration increment of 12.5 μg/m^3 of PM10. Site-specific, clear diurnal and seasonal fluctuations were observed for PM10 concentrations at all AQM sites. Two major pollution episodes when PM10 24-h mean exceeded 50 μg/m^3 were differentiated. The first one (25 April – 15 May 2006) was characterized by the long-range transport of pollutants emitted by biomass burning in southeastern Europe. In the second episode (9–17 January 2009), high levels of PM10 were determined, tracing the polluted air mass transport from southwestern Europe where large combustion plants are responsible for two thirds of the European Union (EU) total sulphur emissions.

Keywords: PM10, mass concentration, statistical analysis, intraurban uniformity, long-range transport, air mass backward trajectories

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1. Introduction

Particulate matter (PM) is a general term used for a mixture of solid particles and liquid droplets, in a wide range of size and chemical composition, suspended in the atmosphere. They may be emitted directly by a source (primary particles) or can be formed in the atmosphere (secondary particles) by transformation of gaseous precursors emitted from a variety of sources. Primary particles, which may be coarse or fine, are commonly associated with combustion sources including traffic, industry, domestic heating, as well as sea salt, pollen, volcanic emissions, and earth crust materials. Secondary particles are finer and formed in the atmosphere through chemical and physical conversions of gaseous precursors such as nitrogen oxides (NO_x), sulphur dioxide (SO_2), volatile organic compounds (VOCs), etc. The impact of ambient particulate matter on public health, global climate, and local visibility has been a longstanding concern of the air quality management for community and regulatory authorities. A large number of epidemiological studies have

been conducted worldwide that demonstrated associations between concentrations of atmospheric particles of the aerodynamic diameter smaller than 10 μm and excesses in daily mortality and morbidity, especially in urban areas [1, 2]. Scientific studies show that there is a strong relation between PM and a series of significant adverse impacts on the environment via reduced visibility and changes in the nutrient balance through deposition processes. The atmospheric particulate matter plays an important role in the trace material cycle of the atmosphere and may influence the atmospheric chemistry. PM has climate-forcing impacts, either contributing to or offsetting the warming effects of greenhouse gases and can also act as condensation nuclei in the cloud formation [3, 4]. Furthermore, secondary inorganic particle formation and transport have been studied for decades as they contribute to acidification of soils. Hence, for all these issues a thorough knowledge of the concentrations of PM as well as their sources and sinks is needed. Particles may present diverse physical (size, surface area, density, number) and chemical patterns in

different areas due to a large number of natural and anthropogenic sources. In addition to the local and regional anthropogenic particulate matter emissions, both the concentration and composition of airborne PM depend on the climatology (temperature, humidity, solar radiation, rain scavenging potential, re-circulation of air masses) and on the geography (topography, soil cover, proximity to arid zones or to the coast) of a given region. Therefore, wide variations in PM concentrations and characteristics may be expected when considering European regions such as the Mediterranean and Scandinavian countries with different climatologic and geographical patterns. Data on particulate matter physical and chemical characteristics and source apportionment of a wide variety of sites in Europe may be found in Putaud et al. (2004) [5] and van Dingenen et al. (2004) [6]. However, the current European Union (EU) limit value (LV) for airborne PM10 comprises both an annual mean concentration (40 μg/m^3) and daily concentration of 50 μg/m^3 which should not be exceeded more than 35 days a year [7].

Atmospheric particulate matter in Vilnius city may originate from three predominant sources, i. e. primary combustion particles (originating largely from motor vehicles), secondary particles (arising from the oxidation of SO_2, NO_x, and VOC) and dust/soil resuspension processes. Previous measurement data from the air pollution studies in Vilnius city have shown that 24 h mean concentrations of various pollutants (i. e. benzo(a)pyrene, total particulate matter, carbon monoxide) have sometimes exceeded permissible levels [8–10]. However, in Vilnius, where emissions from industrial sources and stationary combustion are modest, the motor vehicles' contribution has been estimated to represent about 64% of the emissions of particles in the area in 2006 [11].

The main objectives of this study were: (i) to estimate an intraurban variation in PM10 mass concentrations; (ii) to determine diurnal and monthly variations in PM10 concentrations; (iii) to evaluate the importance of air masses' origin for the PM10 concentration in Vilnius city.

2. Measurement sites and methods

Spatial and temporal variations of PM10 within Vilnius city were investigated using the data sets collected from three air quality monitoring (AQM) sites of the Lithuanian Environment Protection Agency during a 3-year period (2005–2007) [12, 13]. These AQM sites were chosen in order to represent an urban background

(Lazdynai, residential) site, semi-urban (Senamiestis, mixed residential and commercial) site, and the site with an intensive road traffic flow (Žirmūnai) in Vilnius. The Žirmūnai air pollution monitoring station is located on Kareivių street close (about 150 m) to one of the significant and busiest junctions in Vilnius city with the traffic density of 30,000–40,000 vehicles per 24 h. The sampler was situated at about 2 m from the edge of the curbside and about 6 metres from the street with a 4-lane road and a frequent congestion. Braking is frequent near the measurement site due to the presence of traffic lights on the nearest junction.

The Senamiestis air pollution monitoring station is located in the old part of Vilnius, in a densely populated area with residential and commercial buildings. The sampler is located in a square (K. Sirvydo) surrounded on two sides by streets (Pilies and Švarco) with a moderate intensity of traffic.

The Lazdynai air pollution monitoring station is located within a residential district nearby the TV tower in a relatively open area. There are no residential buildings and heavily travelled streets within a few hundred metres from the station. There are also no significant stationary sources within 2–4 km.

The mass concentrations of PM10 were obtained with the automatic analyzers using either the Environment PM101M monitors based on the β-attenuation method (in Žirmūnai and Lazdynai) or the tapered-element oscillating microbalance (TEOM) series 1400a monitor (at Senamiestis). The data of the traffic density used in this study are taken from [14].

The monitoring data of PM10 concentrations used in this paper have been available originally on the basis of 1 h averages. The daily means of PM10 have been calculated from the 1 h averages and were used for statistical considerations. For the calculation of daily means a minimum number of 20 one-hour values was required, otherwise the value was considered as missing. Logarithms of all daily means of PM10 mass concentrations were calculated and after determining the maximum and minimum values the whole interval of numerical values of logarithms of daily means of PM10 concentrations was divided into 18 segments. The segment had a value of 10 μg/m^3. Later, the number of concentration logarithms corresponding to a particular segment was divided by the numerical value of this segment interval. In such a way, the density distribution was obtained. Finally, the distribution mode, the averages of mode concentrations, and their standard deviation were determined by the least-squares method. A more specific explanation of application of the log-

normal distribution law can be found in the study by Šakalys et al. (2004) [15].

The potential source area of PM10 during the long-range transport (LRT) episodes was detected using backward air mass trajectories. 72-h air mass backward trajectories at heights of 50, 1000, 1500 m above the ground level were obtained by the atmospheric model Hybrid Single-Particle Lagrangian Integrated Trajectories (HYSPLIT), version 4 [16].

3. Results and discussion

3.1. Mass concentration of PM10 and spatial distribution patterns

The concentrations of air pollutants are usually random variables and every area is a special case. The lognormal distribution is the most popular distribution used for fitting the air pollutant concentration. Distributions of pollutant concentrations measured in the environment often appear to be approximately unimodal lognormal ones [17, 18]. As seen in Fig. 1, the PM10 concentration distribution has a rather similar shape and can be approximated by unimodal lognormal distributions at all three sites. The distribution plots of PM10 24-h mean mass concentrations showed the unimodal lognormal distribution with a mode centered on 34.4, 21.6, and 19.9 μg/m^3, respectively, at the traffic-exposed (Žirmūnai), semi-urban (Senamiestis), and urban background (Lazdynai) sites. The occurrence of maximum frequency differs between the traffic site and two other urban sites. A shift of maximum frequency to the higher concentrations is observed at the traffic site. The daily means of PM10 concentrations falling in the range of 25.7–40.2 μg/m^3 constituted the dominant fraction (75%) of total values at the Žirmūnai site, while it was in the range of 16.4–26.0 and 16.0–24.0 μg/m^3 at Senamiestis and Lazdynai sites, respectively. The data indicate that the EU limit value of 50 μg/m^3 as a daily mean concentration of PM10 was annually exceeded 52, 15, and 11 days, respectively, at traffic, semi-urban, and urban background sites. The exceedences (Table 1) of the limit value most often occurred at the station with great influence of traffic during 2006 when 66 days had daily means of PM10 concentrations higher than 50 μg/m^3. The results of this comparative analysis indicate that PM10 levels at the Žirmūnai sites can be distinguished by such a factor as traffic intensity. As seen in Table 1, PM10 annual mean concentrations vary between 17.5 and 37.3 μg/m^3 and cover a relatively narrow range with the inter-annual variability not more
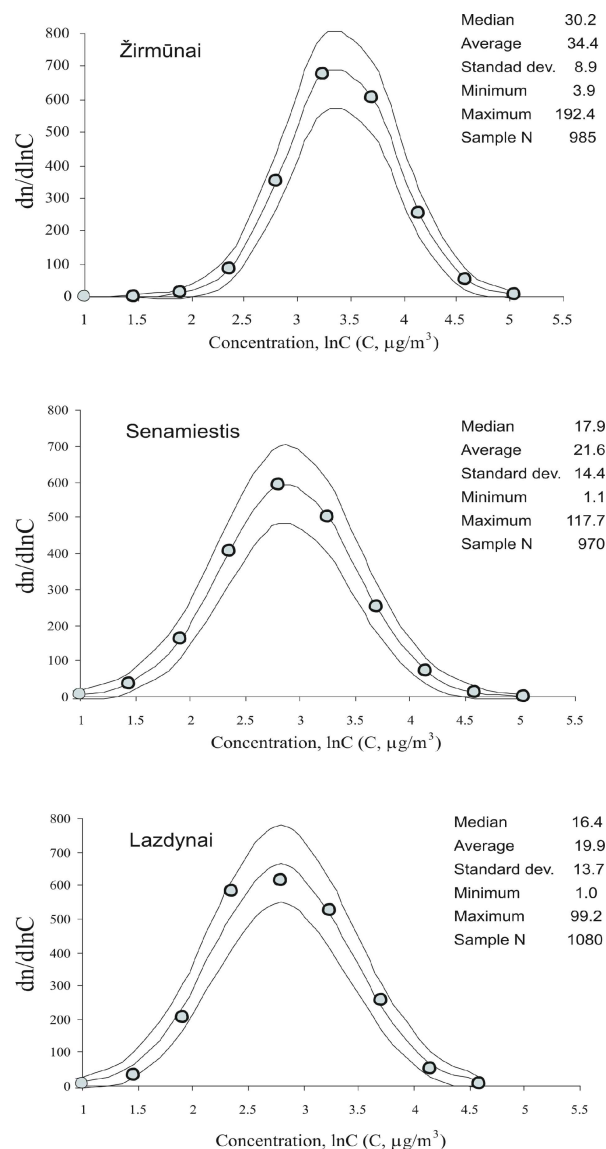

Fig. 1. Frequency distributions of 24-h average PM10 concentrations at 3 AQM sites (Vilnius) in the period of 1 January 2005 – 31 December 2007 (black solid line is distribution of concentration logarithms, thin line marks 2σ intervals).

than 30% at each station. Meteorological conditions affect PM concentrations in the atmosphere by determining formation, dispersion, and removal of atmospheric pollutants. Thus, year-to-year variability in PM10 concentration is partly due to the inter annual meteorological variability. PM10 annual mean concentrations differed more from site to site, the lowest concentrations were observed at the urban background site (Lazdynai) and increased values were measured at the semi-urban site (Senamiestis). Remarkably higher PM10 levels determined for the site with a heavy traffic (Žirmūnai) point up a relatively high contribution from the traffic.

The data show that the limit value of the EU directive (PM10 annual mean of 40 μg/m^3) was not exceeded at

Table 1. Annual mean, standard deviation (SD), range of daily average PM10 mass concentration (μg/m^3), and the exceedences (days) of the EU 24-h PM10 limit value at three sites in Vilnius city.

Sites	2005		2006		2007	
	Mean (SD)	Min/max exceedences	Mean (SD)	Min/max exceedences	Mean (SD)	Min/max exceedences
Žirmūnai	33.1 (16.5)	5.9/192.0 44	37.3 (19.6)	6.2/130.5 66	32.2 (17.2)	5.1/122.0 47
Senamiestis	23.7 (11.8)	4.1/110.3 18	18.4 (8.4)	4.3/78.0 4	21.7 (13.0)	5.0/117.7 22
Lazdynai	21.9 (10.9)	4.4/83.3 15	20.9 (10.8)	4.1/99.2 11	17.5 (10.2)	3.8/82.9 7

study sites in Vilnius city. Variation of the daily means expressed by the coefficients of variation, i. e. the standard deviation (SD) divided by the mean, is very similar to an average of 50–58% over that period for the three sites. The difference for the 1-year mean concentration of PM10 between the urban background and semi-urban sites is from 1.8 to 4.2 μg/m^3. These values demonstrate rather small differences and support the idea of regional pollution by PM10 and its strong connection to the long-range transport of particulate and gaseous pollutants on the regional scale [19].

On the whole, PM10 levels in Vilnius city were consistent with those obtained in European cities and can be ascribed to sites with lower annual average concentrations. Van Dingenen et al. [6] reported PM10 concentrations of 27.5, 25.1, and 23.0 μg/m^3 for three different sites in London. Long-term (1998–2003) PM10 annual means at the Finnish urban sites ranged from 11 to 24 μg/m^3 [20]. For PM10 concentrations at several urban and rural sites in Switzerland Monn et al. [21] reported 10–33 μg/m^3, Roosli et al. [22] reported 28 μg/m^3, and Gehrig et al. showed [23] 22.5–35.9 μg/m^3. A two to threefold concentration range (from 41 to 98 μg/m^3) was found for PM10 in Central and East European countries: Bulgaria, the Czech Republic, Hungary, Poland, Romania, and Slovak Republic [24]. The concentrations in the Czech Republic, Slovak Republic, and Hungary were considerably higher than the Austrian data [1]. At urban background sites, the highest PM10 levels were obtained in Spain and Germany where the annual PM10 mean varied from 28 to 42 μg/m^3. At the other urban background sites PM10 levels reached around 25 μg/m^3, with the lowest values recorded in Sweden (17–23 μg/m^3). A similar variation was found for the traffic-exposed sites. The highest annual means for PM10 of 37–55 μg/m^3 were obtained in Austria, Germany, and Spain. At the traffic sites in the Netherlands and the United Kingdom PM10

concentration varied from 30 to 35 μg/m^3, and from 26 to 51 μg/m^3 in Sweden [25]. Marcazzan et al. [26] reported PM10 winter concentrations of 103 μg/m^3 and summer concentrations of 68 μg/m^3 in Milan, Italy. The presented data from the cities in different parts of Europe show that annual means for PM10 exceeding 30 μg/m^3 are quite typical of urban environments.

The method [27], which is frequently used to derive information about the spatial uniformity of pollutant concentrations within the study area, is the comparison of PM10 mass concentrations measured at the sites. Three patterns of relationships between PM10 daily concentrations measured on the same days for Lazdynai and Žirmūnai, for Senamiestis and Žirmūnai, for Lazdynai and Senamiestis are illustrated in Fig. 2. The data of the relationship between PM10 concentrations from the traffic site (Žirmūnai) and the urban background site (Lazdynai) (Fig. 2(a)) show a slope larger than 1.0, a positive mass increment of 12.6 μg/m^3 of PM10 at the traffic site and the correlation coefficient $r = 0.71$. A similar pattern (Fig. 2(b)) was observed for the relationship between the traffic site (Žirmūnai) and the simultaneously measured PM10 concentrations at the urban site with moderate traffic (Senamiestis). The Lazdynai and Senamiestis sites appeared to show a somewhat intermediate pattern (Fig. 2(c)), having a slope close to 1.0, relatively slight positive PM10 mass increment of 3.1 μg/m^3 at the moderate traffic site, and the correlation coefficient $r = 0.82$. Rather similar averages for PM10 concentrations over the period at those two sites (21.6 and 19.9 μg/m^3) and a high correlation between PM10 values suggest regionally homogeneous sources. The increment of PM10 concentrations between the traffic site and the urban background site can be attributed to the local influence of traffic on the adjacent street. High and significant ($0.71 \leq r \leq 0.82$) correlation coefficients between the daily average concentrations of PM10 from three sites show relatively
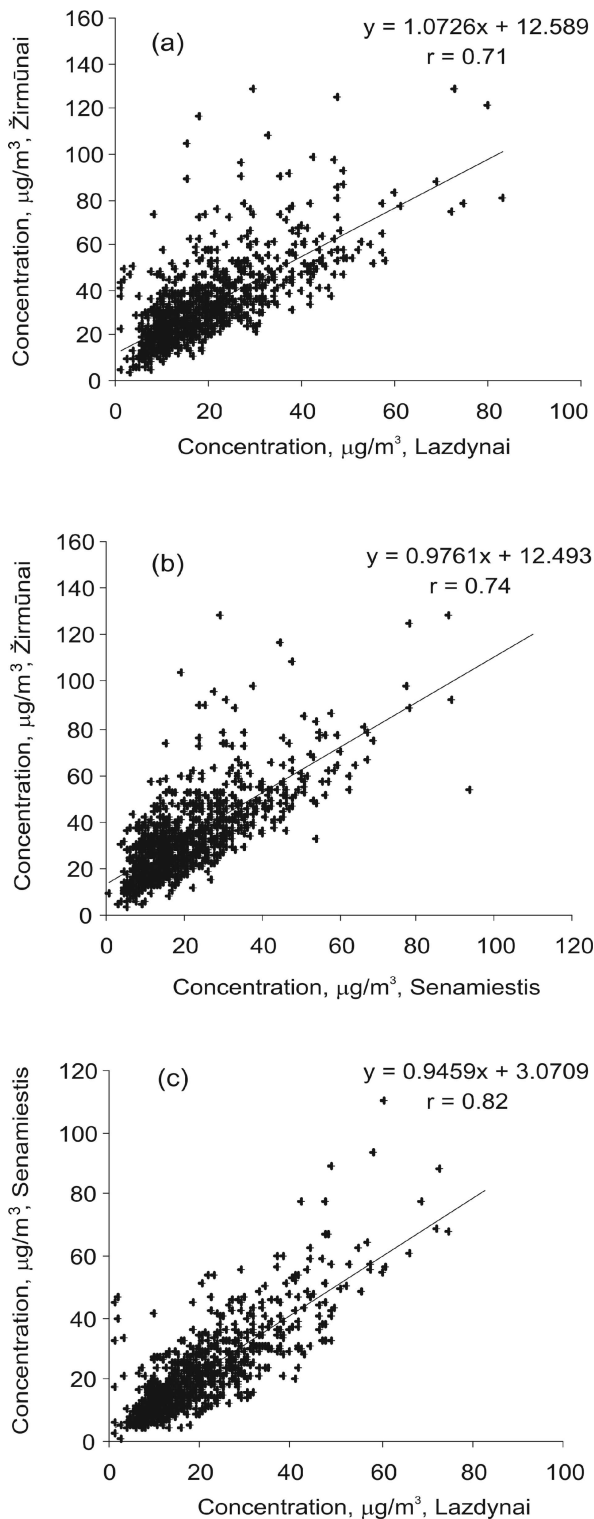
Fig. 2. Scatter plot and linear regression line between 24-h average PM10 concentrations measured on the same day at air quality monitoring sites (Vilnius).

uniform temporal distribution of PM10 mass concentrations in the city.

In addition to correlation coefficients, the calculation of the coefficient of divergence (COD) as a relative measure of PM concentration uniformity within the study area is recommended by Wilson et al. [28]. The COD is defined mathematically as follows:

$$\mathrm{COD}_{jk} = \sqrt{\frac{1}{p} \sum_{i=1}^{p} \left(\frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}} \right)^2}, \qquad (1)$$

here x_{ij} and x_{ik} represent the 24-h average PM concentration for sampling day i at sampling sites j and k, and p is the number of observations. A COD value of zero or close to zero indicates similarity between concentrations at the two investigated sites, while a value approaching one indicates maximum differences and absolute heterogeneity. The application of this procedure to the PM10 daily dataset yielded relatively low COD values ($<$0.25). Source-similar sites (Lazdynai and Senamiestis) had a COD of 0.19, while Lazdynai and Žirmūnai sites, measuring PM10 concentrations from different types of sources, resulted in a COD of 0.23. This reveals that throughout the whole study period the PM10 levels of all three sites were mostly influenced by similar sources, i. e. regional emissions or long-range transport, and only to a certain extent by site-specific local emissions, which were assumed to be responsible for the differences in PM10 concentration between the Žirmūnai and Lazdynai sites. The COD value of 0.19 for Lazdynai and Senamiestis indicates a better uniformity.

3.2. Diurnal and seasonal distribution patterns of PM10 mass concentrations

In addition to spatial variation, particulate matter has been shown to fluctuate over time [29]. Many urban areas experience a diurnal cycle in air pollutant concentrations, as anthropogenic sources make up a large portion of measured concentrations. Over short sampling durations (1 or 24 h), pollutant concentrations for different sites in a city will usually differ to a greater degree than measurement data averaged over longer periods. The diurnal variation of PM10 concentrations depends on the day of the week; we have selected data for analysis from the working days (Monday–Friday) and weekends (Saturday–Sunday). The diurnal patterns of PM10 mass concentration prepared for each of the three study sites are shown in Fig. 3. Initially, the number of vehicles passing the sampling point at the Žirmūnai
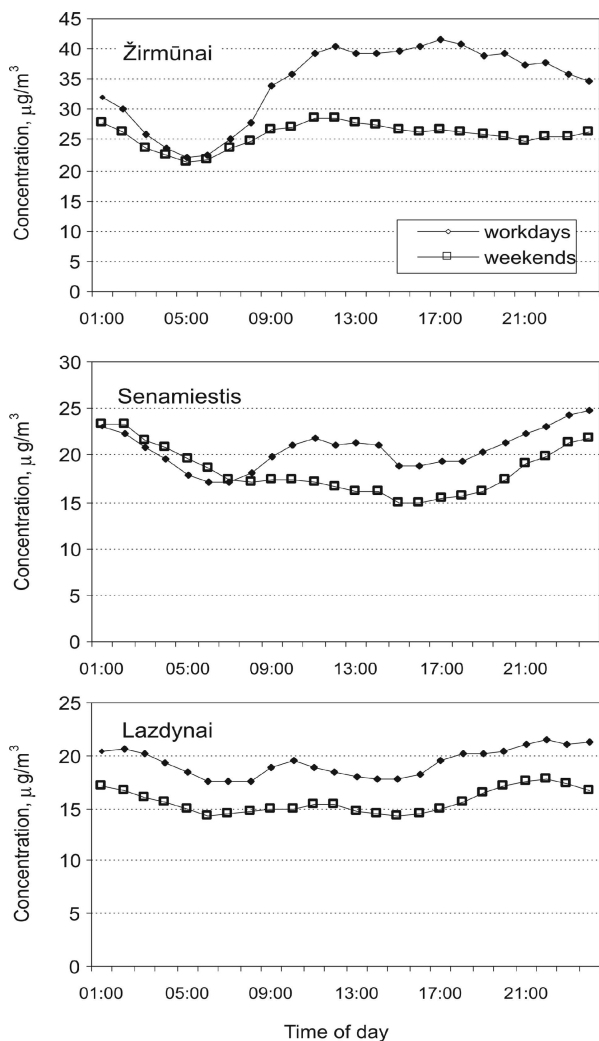
Fig. 3. Mean diurnal variation of PM10 concentration during workdays and weekends at 3 AQM sites in Vilnius during the period of 1 January 2005 – 31 December 2007.

site was low, with 1300 vehicles/h at 06:00 rising to 2500 vehicles/h from 08:00 to 10:00, corresponding to the morning rush-hours. The traffic intensity dynamics at the Žirmūnai site was found in [19]. The concentrations of PM10 at the Žirmūnai site show a distinct daily variation caused by emission changes stimulated by vehicles during the day and the dispersion conditions. The concentrations continuously increase during the morning rush hours, from approximately 6:00 to 12:00, during working days. Subsequently, the concentrations decrease slowly during the rest daytime hours, in some cases also showing peak values during the afternoon rush hours, from approximately 16:00 to 18:00. The maximum (41 μg/m^3), recorded at 10:00–12:00, is by approximately 98% higher than the daily minimum. The more moderate diurnal variation of the PM10 concentrations is seen on weekends; the maximum (29 μg/m^3) is by approximately 37% higher than

the daily minimum. The diurnal patterns of PM10 mass concentration at Lazdynai and Senamiestis sites are similar to those of Žirmūnai site, but variation of PM10 concentrations is not so significant. At all stations lower levels of PM10 are observed on the weekends than during workdays. The weekend PM10 levels were, on average, by 9 and 4 μg/m^3 lower than the workday levels, respectively, at a traffic site and an urban background site, and this gives indirect evidence that the PM10 concentrations are originated mainly from local vehicular transport by direct emissions and suspension of road dust particles from the road surfaces. The latter process is most important during early spring when the roads are dry and the sand is not removed from roads, and the studded tyres are not removed from the vehicles. The correlation between the traffic density, NO$_x$ and CO gases, and the PM concentration in cities has been investigated in the studies [30, 31] and by means of receptor modelling the PM from the local traffic could be successfully attributed to four sources: exhaust, road dust, brake wear, and winter salting. Roughly half of the road dust could be attributed to asphalt wear or other mineral dust sources. The other half must be due to tyre wear and probably other sources of organic material. Figure 4 presents the monthly mean, daily maximum and minimum concentrations of PM10 measured at traffic, urban background, and urban sites from 2005 to 2007. At all three sites the highest monthly PM10 levels were recorded in late winter and spring (from February to May). Conversely, the lowest PM10 levels were measured in summer. Data show much larger fluctuations of PM10 daily concentrations during winter and spring than during summer and, therefore, the exceedences of the daily limit value of 50 μg/m^3 for PM10 were not evenly distributed over the year at all stations. These exceedences were more frequent in March and April with hourly mean PM10 concentrations rising up to 200 and 130 μg/m^3 at Žirmūnai and Lazdynai sites, respectively. The winter–spring maximum of PM10 concentrations can be explained by seasonal change in the meteorological conditions, such as wind speed, wind direction, and restricted atmospheric mixing conditions. It was established [9, 10] that serious pollution episodes in Vilnius city were not generally caused by the sudden increase in the emission of pollutants, but resulted from the unfavourable meteorological conditions that influence pollutant dispersion: the wind speed and direction, the boundary layer depth, the vertical temperature gradient, and stability class of the atmosphere. A limited dispersion during periods with low temperatures causes higher PM10 concentrations.
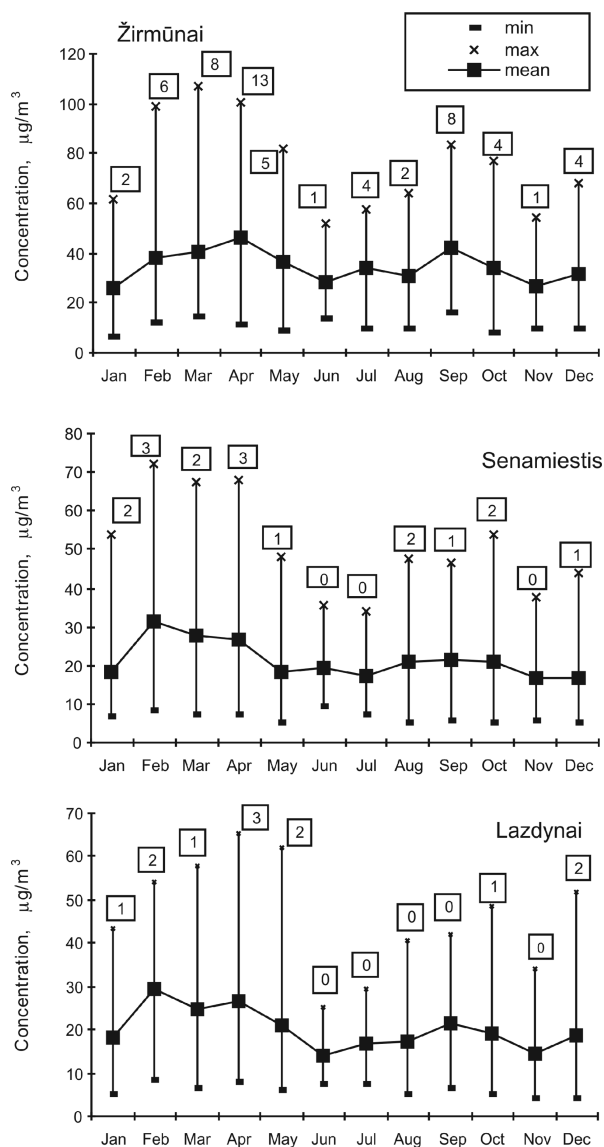
Fig. 4. Monthly mean, daily minimum and maximum concentrations of PM10 at all 3 sites in Vilnius during the period of 1 January 2005 – 31 December 2007. In rectangles is the number of exceedances of the EU 24-h limit value for PM10 of 50 $\mu g/m^3$.

Moreover, the springtime dry periods with increasing temperatures, heating, and evaporation cause the effective suspension of the dust accumulated from multiple sources to road surfaces and initiate the elevation of PM10 concentrations. However, the synchronized day-to-day variation at study sites across Vilnius city points up the role of weather patterns in the formation of pollution episodes. Overall, the mass concentrations of PM10 were clearly higher at the traffic site (Žirmūnai) where the local emission from the intense traffic considerably contributes to a surplus of the PM10 burden. At the urban background and semi-urban sites, which are less influenced by traffic emission, the monthly means of PM10 are almost the same and are less than half of those at the traffic site. The seasonality of PM10 has also been proved in a number of studies [23, 25, 32].

3.3. Examination of selected PM10 pollution episodes in Vilnius

As already mentioned above, PM10 concentrations in Vilnius city are influenced by the combination of emission sources and meteorological conditions, which cause the accumulation as well as transport of pollutants. As shown in the study [8] the concentration of pollutants including particulate matter close to the road is higher by factor of 8.1 than that determined at a 40 m distance from the road and the spatial distribution of PM10 concentrations originating from local traffic occurs substantially on the microscale (i. e. on a scale of tens or hundreds of metres). The atmospheric lifetime of PM10 is of a magnitude allowing it to be advected over distances of several hundreds of kilometres and to be accumulated in air mass over several days usually during the stable high pressure situations. Therefore, a simultaneous increase in PM10 levels at all three sites in Vilnius city may be associated with regional episodes or with "external" contributions by long-range transport of pollutants from the regions in Europe.

Two types of episodes with simultaneous increase in PM10 daily values exceeding 50 $\mu g/m^3$ at the urban traffic and urban background air quality monitoring sites in Vilnius during different seasons in 2006 and 2009 were analysed.

In spring of 2006, a large part of Europe experienced reduced air quality for a prolonged period of time caused by emissions from agricultural waste burning in eastern Europe. This was caused by specific meteorological conditions that resulted in the transport of fire plumes well beyond the northern and western continental Europe up to high Arctic latitudes. For investigation of wild fire areas, daily active fire hot spot detections were extracted from the MODerate-resolution Imaging Spectroradiometer (MODIS) Rapid Response System [33, 34].

Figure 5 shows PM10 daily concentrations measured at urban background and traffic sites in Vilnius from 10 April to 8 May 2006, the MODIS produced fire map, and air mass backward trajectories to Vilnius. Analysis of air mass backward trajectories and fire map confirmed that the long-range transported pollutants emitted by biomass burning in southeastern Europe had a strong impact on PM10 concentrations in Vilnius in April–May of 2006. As shown in Fig. 5, in the period of 29 April – 4 May air masses arrived from

the agricultural waste burning area (moving over Russia, the Ukraine, and Belarus), and an increase in the PM10 concentrations was recorded on 29 April (76 and 73 μg/m^3 at Žirmūnai and Lazdynai sites, respectively). The PM10 daily values reached a maximum (130 and 99 μg/m^3 at Žirmūnai and Lazdynai sites, re-

spectively) on 2 May 2006 and remained at elevated level until 7 May. On 7–8 May 2006 the Atlantic air masses were advected towards Vilnius city causing a drop in PM10 concentrations. This episode was extensively reported in papers [35, 36]. Saarikoski et al. [37] showed that PM10 concentrations during the pe-
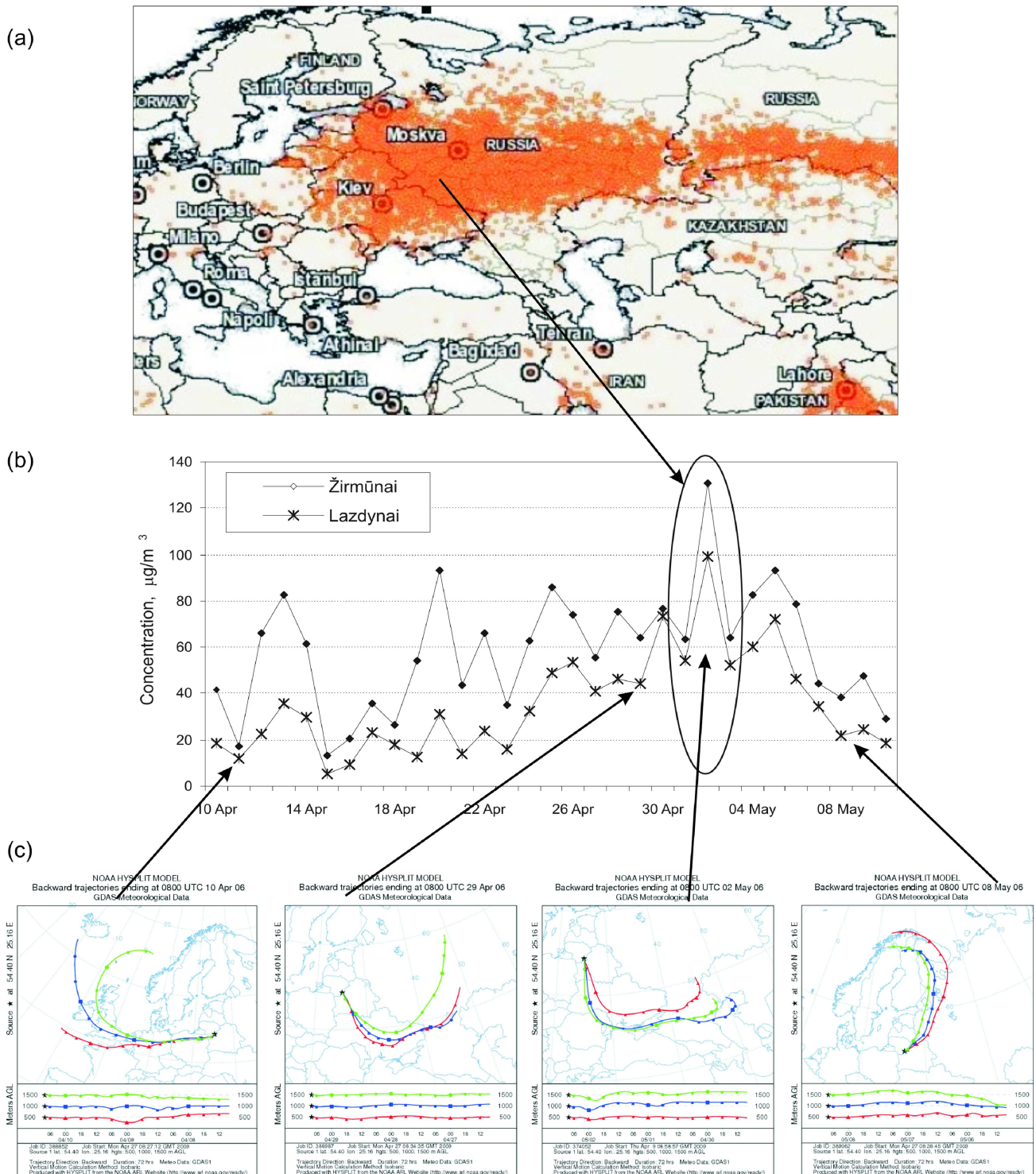

Fig. 5. (a) MODIS produced fire map during 25 April – 15 May 2006, (b) daily mean concentrations of PM10 measured at traffic and urban background sites, and (c) backward air mass trajectories during the period of 10 April – 9 May 2006.
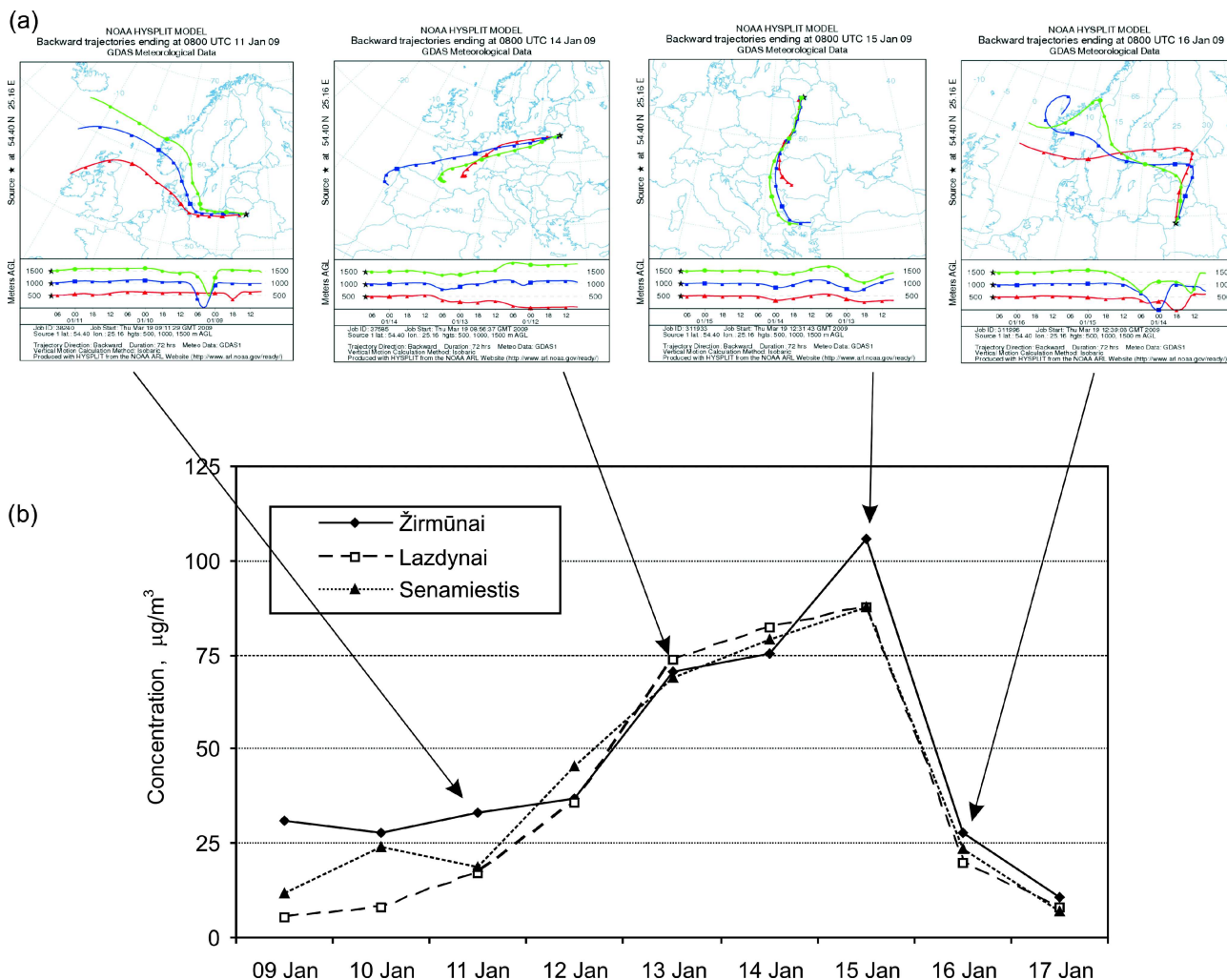
(a)

(b)

Fig. 6. (a) Backward air mass trajectories and (b) daily mean concentrations of PM10 measured at traffic and urban background sites during the period of 9–17 January 2009.

riod of 25 April to 5 May 2006 were in the range of 68–81 μg/m^3 at an urban background site in Helsinki. Seven urban sites in England and Wales [38] registered elevated levels of PM10 with maximum hourly concentrations of 163 μg/m^3 at the Glasgow roadside site in Scotland and concentrations >130 μg/m^3 at seven urban sites in England and Wales from 8 to 10 May 2006.

Emissions from agricultural waste burning in fields and wildfires deteriorate air quality over very large areas, even at the distance of over 1000 km from the fire areas. Therefore, the smoke from open biomass burning is an important factor in the deterioration of air quality in Europe during dry periods in spring and late summer. In future, climate change may increase extreme meteorological conditions (e. g. dry heat waves) and smoke episodes if more efficient fire prevention measures are not implemented.

The second highest PM10 level at the monitoring sites in Vilnius city was recorded during the period of

13–15 January 2009. Daily mean PM10 levels were in the range of 73–88 and 70–105 μg/m^3 at the urban background and traffic sites, respectively. Figure 6 depicts examples of typical situations with long-range transport from regions in southern Poland, northern Moravia (the Czech Republic), and Bulgaria where large combustion plants are responsible for two thirds of EU total sulphur emissions [39] during 14–15 January 2009 coinciding with high PM10 concentrations in Vilnius city. Before and after this event, significantly lower PM10 levels (<30 μg/m^3), when air masses' flow was from the northwestern Europe, were related with local emission sources. In previous studies it has been shown [40, 41] that the air mass flow from the southeasterly direction dominates in April and May, and south-southwesterly directions are most frequent during winter months and early spring. In general, the air flows over the large area sources of SO$_2$ and NO$_2$ emissions in the southern part of Europe can be consid-

ered as a peculiar feature of transport of air pollutants to the Lithuanian territory.

4. Conclusions

The distribution plots of PM10 24-h mean mass concentrations showed the unimodal lognormal distribution with a mode centered on 34.4, 21.6, and 19.9 μg/m^3, respectively, at the traffic-exposed (Žirmūnai), semi-urban (Senamiestis), and urban background (Lazdynai) sites in Vilnius city.

High and significant ($0.71 < r < 0.82$) correlation coefficients between the 24-h mean concentrations of PM10 from three sites show relatively uniform temporal distribution of PM10 mass concentrations in the city. This implies that throughout the whole study period the PM10 levels of all three sites were mostly influenced by similar sources (e. g. regional emissions or long-range transport) and only to a certain extent by site-specific local emissions. Inter-site data comparison shows that highest concentrations of PM10 have been observed at the traffic-exposed site and the lowest ones at an urban background site. The roadside increment in PM10 mass concentration of 12.6 μg/m^3 has been observed from paired traffic-exposed and urban background sites and may be attributed to vehicle exhaust emissions and to other sources such as resuspension, brakes and tyre wear.

Diurnal cycles displayed a morning maximum in PM10 concentrations correlating with the onset of vehicle activity on the road. Monthly averaged concentrations of PM10 were generally higher during late winter and spring (from February to May) at three sites. Conversely, the lowest PM10 levels are measured in summer. PM10 levels (annual mean concentration) in Vilnius city ranged from 17.5 to 37.3 μg/m^3. The EU limit value of 50 μg/m^3 for PM10 (24-h mean) was exceeded more than 35 days annually only at the traffic-exposed site (Žirmūnai) and this exceedence was less than 35 days at the urban background (Lazdynai) and semi-urban (Senamiestis) sites.

The simultaneous increase in PM10 daily values exceeding 50 μg/m^3 at three sites in Vilnius city was related to "external" contribution of pollutants by long-range transport from open biomass burning in eastern Europe and from ordinary anthropogenic sources (e. g. energy production, traffic, industry, and wood combustion) in western Europe.

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AEROZOLIO DALELIŲ (<10 μm) KONCENTRACIJOS VILNIUJE KAITA

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Santrauka

Per pastaruosius kelis dešimtmečius pastebimai didėja mokslinių publikacijų, kuriose įvairiais aspektais pateikiami duomenys apie atmosferos aerozolio daleles [5, 6]. Dėl savo fizinių ir cheminių savybių įvairovės jos tiesiogiai veikia daugelį atmosferos procesų, tuo pačiu globalų ir regiono klimatą [4]. Be to, nustatytas ryšys tarp aerozolio dalelių koncentracijos ir žmonių sergamumo bei mirtingumo, ypač urbanizuotuose rajonuose [2]. Aerozolio dalelių savybės ir koncentracija kinta laike ir erdvėje priklausomai nuo jų emisijos šaltinių tipo, stiprio ir geografinės vietos. Taigi, įvairiais aspektais vykdomi aerozolio dalelių tyrimai skirtingose pasaulio vietose yra labai aktualūs ir dažnai įjungiami į įvairių pasaulinių programų sudėtį [19, 24, 25]. Pagrindinis šio darbo tikslas buvo nustatyti aerozolio dalelių, kurių skersmuo yra mažesnis nei 10 μm, masės koncentracijos erdvinės sklaidos Vilniaus mieste ypatumus, paros ir sezoninę jos kaitą bei įvertinti toli esančių taršos šaltinių poveikį. Trijų oro kokybės tyrimų stočių Vilniuje aerozolio dalelių masės koncentracijos duomenys paimti iš Lietuvos aplinkos apsaugos agentūros interneto puslapio [13]. Pasirinktose stotyse nenutrūkstamai matuojamos ore aerozolio dalelių koncentracijos: esant intensyviam transporto eismui Kareivių gatvėje (Žirmūnuose), gausiai žmonių lankomame ir tankiai gyvenamame rajone (Senamiestyje) ir atokiau nuo intensyvaus transporto eismo gatvių bei stacionarių taršos šaltinių esančiame gyvenamajame rajone (Lazdynuose).

Atlikus statistinę 2005–2007 m. aerozolio dalelių 24 val. (t. y. paros) vidutinių masės koncentracijų analizę, nustatyta, kad visose trijose oro kokybės tyrimo vietose gautus duomenis gerai aprašo lognormalinis pasiskirstymas, turintis tik po vieną modą, kuri Žirmūnuose, Senamiestyje ir Lazdynuose atitinkamai yra 34,4, 21,6 ir 19,9 μg/m^3. Dideli koreliacijos koeficientai (0,71 $< r <$ 0,82) su patikimumu $p > 0,99$, gauti lyginant aerozolio dalelių koncentracijas dviejose jų matavimo stotyse (2 pav.), rodo gana vienodą jų paros vidutinių koncentracijų eigą ir tai, kad aerozolio dalelių koncentracijas lemia tie patys taršos šaltiniai (lokalinė teršalų emisija bei jų pernaša iš tolimesnių regionų). Tačiau Žirmūnuose 12,5 μg/m^3 vidutiniškai didesnės nei Lazdynuose ir Senamiestyje (2(a, b) pav.) aerozolio dalelių koncentracijos rodo vietinio taršos šaltinio (autotransporto) įtaką. Lazdynuose matuojamos aerozolio dalelių koncentracijos gali būti traktuojamos kaip Vilniaus miesto foninės koncentracijos. Visose matavimo stotyse aerozolio dalelių koncentracijos turėjo ir paros, ir sezoninį sandą. Didesnės koncentracijos buvo būdingos laikotarpiui nuo vasario iki gegužės mėn., o mažiausios – vasarą.

Nagrinėjant dviejose stotyse vienu metu stebėtus du (5 ir 6 pav.) didelių aerozolio dalelių koncentracijų ($>$50,0 μg/m^3) atvejus ir analizei panaudojus atgalines oro masių pernašos į Vilnių trajektorijas, nustatyta, kad aerozolio dalelių dideles koncentracijas lėmė teršalai, atnešti iš pietrytinės ir pietvakarinės Europos emisijos šaltinių.