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VARIATION OF PM_{10} MASS AND AEROSOL NUMBER CONCENTRATIONS IN ŠIAULIAI

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Abstract. The urban environment is distinguished by higher aerosol and gaseous pollutant concentrations than those in rural areas. A study of aerosol pollutant behavior was performed in an industrial Lithuanian city of Šiauliai. The PM_{10} mass concentration and meteorological parameter monitoring data were analysed. The aerosol number concentration was measured during a 10-day experiment in Šiauliai. Analysis of PM_{10} showed that the workdays-weekends phenomenon in the PM_{10} mass concentration distribution was prevailing. The PM_{10} mass concentration on workdays was higher in comparison with the concentration at weekends, 24,6 $\mu g/m^3$ and 21 $\mu g/m^3$, respectively. Clear PM_{10} mass concentration dependence on the wind parameters (speed and direction) was found. Linear relationship between aerosol number and PM_{10} mass concentrations was found at a high particle number concentration (more than 18000 cm^{-3}). PM_{10} level in Šiauliai was defined as a sum of three sources: regional background, urban background and local sources. Contribution of these sources to the total PM_{10} mass concentration was estimated to be 36 %, 30 % and 34 %, respectively, during June–October of 2005.

Keywords: PM_{10}, aerosol number concentration, weekend effect, daily variation, weekly variation, PM_{10} sources.

1. Introduction

Unceasing interest in atmospheric aerosols is high mainly because of their effect on human health [1, 2] and role in climatic change [3]. Particulate matter (PM) pollution consists of very small liquid and solid particles floating in the air. They originate from a variety of stationary and mobile sources and may be directly emitted (primary emissions) or formed in the atmosphere (secondary emissions) by transformation of gaseous emissions. Examples of atmospheric particulate matter cited in [4] include: combustion-generated particles, such as diesel, soot and fly ash; photochemically produced particles, such as those found in urban haze; salt particles produced by sea spray; and particles from re-suspended dust. There are also other natural sources like pollen or crushing and grinding rocks and soil. Of the greatest concern to human health are particles small enough to be inhaled into the deepest parts of the lungs. Particulates with an aerodynamic diameter below 10 μm are known as PM_{10}. These particles are about one-seventh the thickness of a human hair [5].

The concentration of particulate matter is highly influenced by human factors. In most European countries industrialization and high volumes of traffic mean that anthropogenic sources predominate, especially in urban areas, and sources of anthropogenic particles are similar throughout Europe. So, in urban areas traffic, factory fumes and use of domestic fuel can significantly influence PM level.

In Europe, ambient concentrations of PM_{10} have been monitored in some urban networks since 1990. According to European Environmental Agency information [6] emissions of fine particles have been reduced by 36 % from 1990 to 2001. The total emission reduction between 2000 and 2001 was 1.1 %. The emission reductions between 1990 and 2001 were mainly due to abatement measures in the energy industries (55 %), road transport (29 %) and energy use from industry (40 %).

The Air Quality Directive 1999/30/EC [7] set out limit values for PM_{10} concentrations. Annual and daily limit value of 50 $\mu g/m^3$ exceeds should not be more than 35 times per year in 2005 and 7 times in 2010. Wide variations in PM level were found in European cities [8]. Particulate emissions from road transport arise as direct emissions from vehicle exhausts, tire and brake wear and resuspension of road dust. In general, diesel engine vehicles emit a greater mass of fine particulate matter per vehicle than petrol engines.

The investigations of PM_{10} concentrations in cities [8] showed that in PM_{10} fraction at the kerbside site re-suspended mineral dust particles and re-suspension of mechanically generated particles prevailed.

Most of the air quality studies performed so far are based on measurements of PM_{10} in the urban air [2].

However, recent studies indicate that the aerosol number (N) concentration could be a much better predictor and indicator of the health effects of the particle matter than the mass concentration [9, 10]. On the other hand, Osunsanya et al [11] found no evidence to support the hypothesis that the component of particulate pollution responsible for effects on respiratory symptoms or function resides in the fraction below 100 nm diameters. Associations between symptoms and PM_{10} suggest that a contribution of the coarser fraction should not be dismissed.

The aerosol (d > 0,4 μm) number concentration measured at a rural site in Lithuania [12] varied in the range of $(5 \div 100)\ 10^3\ cm^{-3}$ in 1990. Similar concentrations were found in 1994 [13]. The Environmental Protection Agency performs PM_{10} concentration monitoring in the main Lithuanian towns [14]. The data obtained in 2005 showed that the highest PM_{10} concentration was observed in Klaipėda (253 $\mu g/m^3$) and a little lower one (196 $\mu g/m^3$) – in Vilnius monitoring stations [15]. However, the investigations of particulate matter concentration in Žvėrynas district of Vilnius [16] pointed up places where very high concentrations of more than 500 $\mu g/m^3$ were observed in 2005.

A wide investigation and assessment of gaseous pollutants was performed in Šiauliai [17]. The authors showed that the highest pollution in the central part of the city was caused by motor-transport.

The aim of this work was to establish the consistent pattern of PM_{10} variations, relationship between PM_{10} and aerosol number (N) concentration, to evaluate the input of possible PM_{10} sources in Šiauliai.

2. Experimental details

Šiauliai is one of the largest industrial centers in northern Lithuania with the population of about 140 000. The city is situated in a convenient geographical location. Šiauliai is famous for its well-developed transport infrastructure and the city is crossed by international routes.

The air monitoring data were obtained from a monitoring station (55º 56‘N, 20º 18‘E, 107 m above the sea-level) near the crossroad of Aušra Avenue and Žemaitė Street. This station is a part of the National Air Quality Monitoring network. The station is near the main town market, representing tendencies of the city traffic pollution and the highest concentration of visitors. Opposite the station high buildings are situated on Žemaitė Street. The area near the station can be characterized as a residential, commercial and industrial locality. Over 10 000 vehicles pass by the station per day. The monitoring data obtained during June–October of 2005 were analysed. The following pollutants are measured at the station: PM_{10}, CO, O_3, SO_2, NO, NO_2, NO_X and meteorological parameters: wind speed, wind direction, temperature, pressure, and humidity. Measurements are taken every 30 minutes [14].

The aerosol number concentration (d > 0,4 μm) was analysed during 10 days from 14 until 24 October 2005 with an optical aerosol particle counter AZ-5. The air sample rate was 1,2 l/min; measurement error according to specifications was ± 20 % [18].

3. Results and discussions

3.1. Weekly and daily variations of concentrations

The substantial weekend effect in the pollutant weekly course is mostly observed in highly polluted regions [19]. In many localities this effect is in direct response to human activity. The weekend effect is easily observed and generally associated with changes in ozone concentration during the week [19], but a similar situation was also established for PM. However, contrary to ozone the PM concentration is typically lower at weekends (Saturday–Sunday) compared to workdays (Monday–Friday).

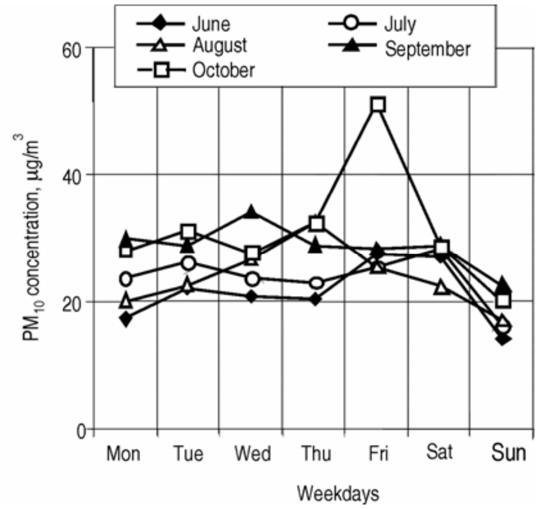

Fig 1. Average daily variation of PM_{10} concentrations in different months

In order to determine of existence of the weekend effect in Šiauliai, an average PM_{10} concentration during different days of the week was evaluated. Variations of an average daily PM_{10} concentration during the week of each month are presented in Fig 1. The highest concentrations were observed on Fridays during June and October, and the lowest ones – on Sundays during all the months. The highest average concentrations were observed during different workdays in July, August and September. As one of the main PM sources is traffic, increase in concentration on Fridays can be explained by the intensification of traffic density. Friday is the last workday, and many people are leaving the city for the weekend, then this street is quite crowded. The lowest concentrations were found on Sundays because that day most people do not use cars or are outside the city. Furthermore, low traffic of heavy-duty commercial and public vehicles is observed. The mean concentration of PM_{10} during workdays was 27 $\mu g/m^3$, and during weekends it was 22 $\mu g/m^3$, over the investigated period. It should be noted that the difference between mean concentrations during workdays and weekends was observed each month: 21,4 and 20,5, 24,2 and 22, 25,3 and 19,5, 29,9 and 25,7, 33,9 and 24,2 $\mu g/m^3$ in June, July, August, September and October, respectively.

The daily patterns of the PM_{10} mass concentration were evaluated separately for workdays and weekends. The results of daily variations during workdays and weekends over June–October are presented in Fig 2.

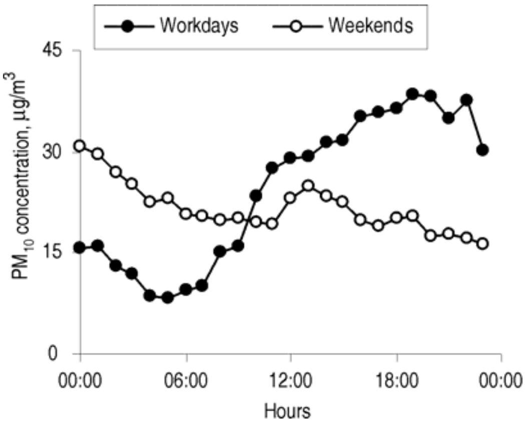

Fig 2. Daily variations of PM_{10} mass concentrations during workdays and weekends

Mean PM_{10} mass concentrations on workdays were higher in comparison with those at weekends, 24,6 µg/m^3 and 21 µg/m^3, respectively. The lowest level of PM_{10} was observed during early morning hours, but since 9:00 the concentration began to rise till 21:00. After 21:00 decrease in the concentration was observed till morning hours. Such variations of PM_{10} can be partially explained by traffic intensity changes.

Some different variations of the PM_{10} mass concentration were observed during weekends. Any morning peak could be seen in the daily pattern, and only after 12:00 the concentration started increasing slightly. The PM_{10} level dropped at 14:00 with some small growth after 18:00. A high PM_{10} mass concentration during nighttime indicates that transport is not the only source of particulate matter.

3.2. Meteorology

Analysis of relationship between PM_{10} concentration and the wind speed and direction was performed. The study showed that the strongest wind of up to 5,5 m/s was registered from the 200º–270º direction (Fig 3) because there are no high buildings in this direction. The registered wind speed from 45º–180º direction was slower by about 3,2 m/s because there are barriers in the southeastern direction, i e buildings. No wind was registered in the direction of 330º–30º during June–October. It seems that the main reason of this phenomenon is building No 66 on Aušra Avenue. It presents a high barrier for the wind.

Fig 4 shows relationship between PM_{10} concentration and the wind speed from different wind direction intervals during the investigation period. The highest PM_{10} concentration was found at an interval of 30º–60º which is distinguished by the lowest < 0,5 m/s wind speed.

The lowest PM_{10} concentration was observed under a stronger wind in the direction of 240º–300º because of particulate matter scattering. No PM_{10} concentration same as wind was registered in the direction of 360º–30º. The analyses clearly show that a low wind speed determines increase in PM_{10} concentration.

3.3. Mass concentration vs number concentration

The particle (d > 0,4 µm) number concentration was measured from 14 to 24 October of 2005. The 24-hour average of PM_{10} concentration and the aerosol number concentration are presented in Fig 5. The high particle number and PM_{10} concentrations were registered at the beginning of this period.

An elevated PM_{10} concentration was also observed on 21–22 October, whereas aerosol number concentration was even diminished. Fig 6 presents relationship between PM_{10} mass concentration and the particle number concentration.

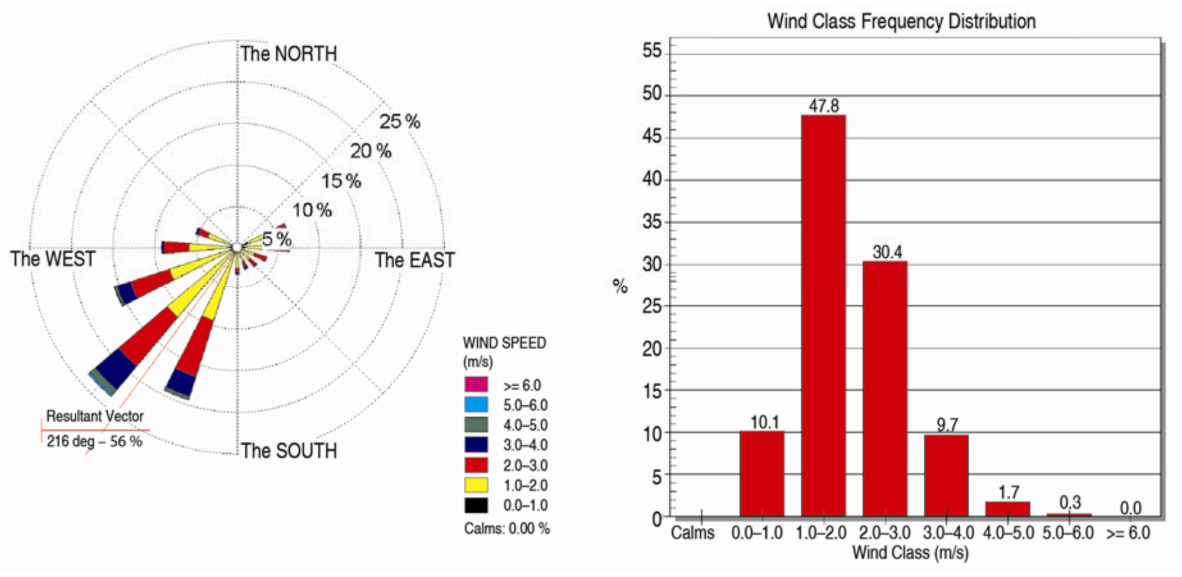

Fig 3. Relationship between wind speed and direction and frequency distribution of wind speed during June–October of 2005

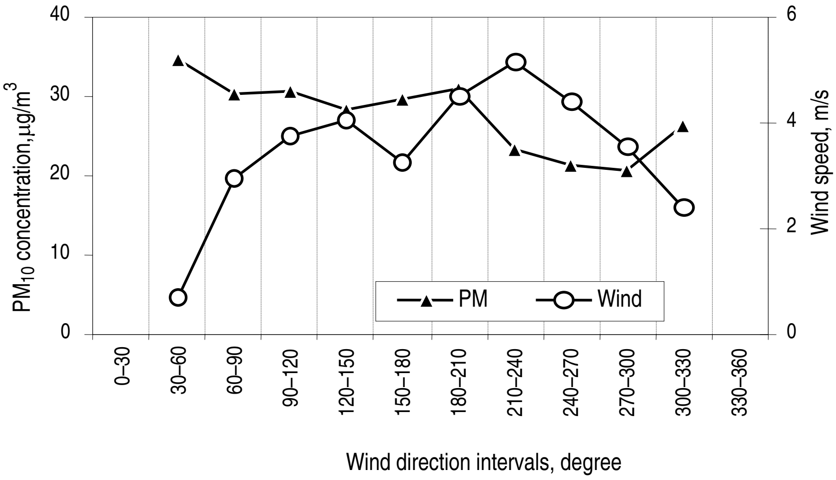

Fig 4. Relationship between PM_{10} concentration and wind speed at different wind direction intervals

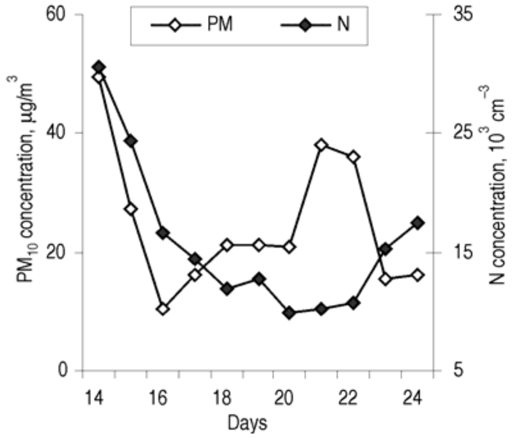

Fig 5. Variation of 24-hour average of PM_{10} concentration and aerosol number (N) concentration on 14–24 October 2005

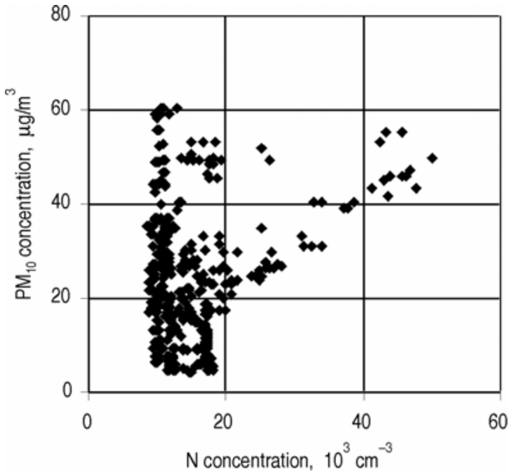

Fig 6. PM_{10} concentration *vs* aerosol number (N) concentration

An interesting relationship between these two variables was found. Two tendencies of the relationship were observed. At a low aerosol number concentration (below 18 000 cm^{-3}) PM_{10} concentrations can vary at a wide interval, and this relationship shows that it can be associated with local sources of PM_{10}. While at a higher particle number concentration (more than 18 000 cm^{-3}) relationship between aerosol number and PM_{10} concentrations was linear (Fig 7). A similar linear relationship was found by other investigators [19–22].

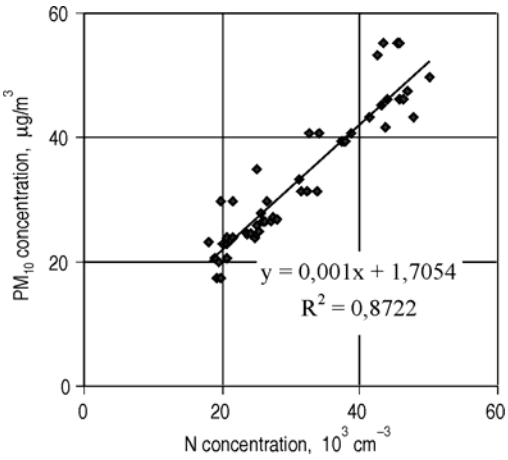

Fig 7. Relationship between aerosol number (N) and PM_{10} concentrations

The obtained results of PM_{10} increase of 1,7 $\mu g/m^3$ for every additional 1000 particles cm^{-3} agree with the results observed at the urban background locations in Birmingham, UK [21], where an increase of PM_{10} of 0,4 $\mu g/m^3$ for every additional 1000 particles cm^{-3} was found. In Delhi [23], the PM_{10} concentration increased at a rate of about 3,5 $\mu g/m^3$ for every additional 1000 particles cm^{-3} when the PM_{10} concentration was below 250 $\mu g/m^3$.

Fig 8 shows an average daily variation of aerosol number and PM_{10} concentrations during 14–24 October. The diurnal patterns of N concentration showed a direct relationship with the traffic density with indications of elevated concentrations during the traffic rush hours which started early in the morning (between 05:00 and 06:00). During the next hours the N level varied marginally till 12:00. Two clear peaks were observed during

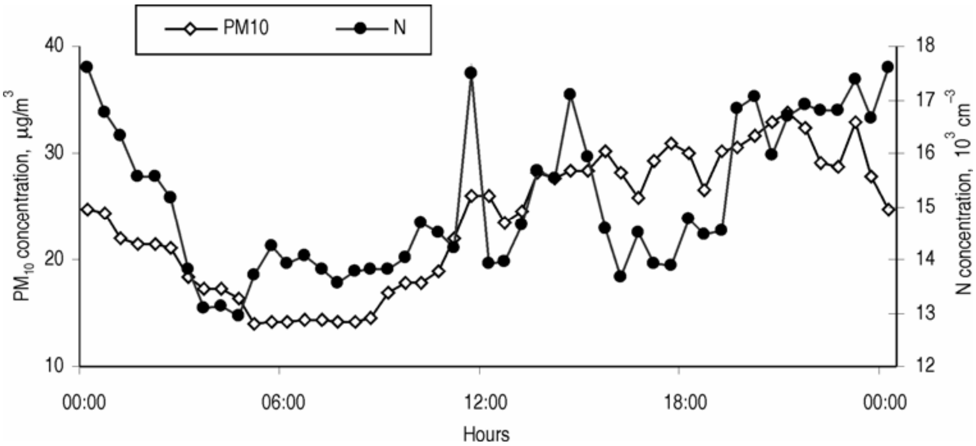

Fig 8. Average daily variation of aerosol number (N) and PM$_{10}$ mass concentrations on 14–24 October 2005

lunchtime 12:00–14:00. Then the N level dropped and began to rise in the evening at about 19:00. Some fluctuations were observed before midnight, and after 24:00 the concentration decreased up to the lowest level at 4:00. A similar average diurnal change of PM$_{10}$ concentration was observed during 14–24 October.

3.4. Sources of PM$_{10}$

The PM$_{10}$ concentration level at an urban site in principle is formed from the regional background level, urban background level and local sources of PM$_{10}$. The influence of these three sources on PM$_{10}$ level can be different in separate cities. This influence could be seen in the frequency distribution of PM$_{10}$ concentrations.

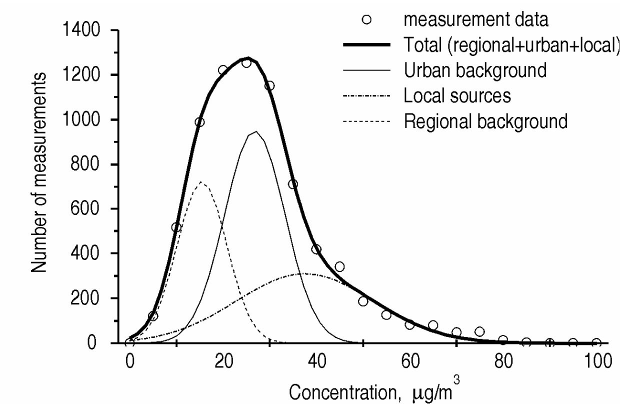

Fig 9. Frequency distribution of half-hour PM$_{10}$ mass concentration in Šiauliai on June–October 2005. The statistical parameters for the Gaussian fits are given in Table

The frequency distribution of the half-hour mean of PM$_{10}$ mass concentration obtained at Šiauliai city monitoring station during June–October of 2005 was investigated (Fig 9). The analysis showed that it could not be described by a simple normal Gaussian distribution, which was set if PM$_{10}$ concentration were determined only by natural sources, i e if no local anthropogenic source were present at the site. Further analysis showed that experimental data could be described by a sum of three bell-shaped curves (Eq 1) like the normal Gaussian distribution with a different center of the peak and standard deviation.

$$y = y_0 + \frac{A}{w\cdot\sqrt{\frac{\pi}{2}}}\, e^{-\frac{2(x-x_0)^2}{w^2}} ,$$

where y_0 – baseline offset, A – total area under curve baseline, x_0 – center of the peak, w^2 – "sigma", approximately 0.849 the width of the peak at half height.

This model describes a bell-shaped curve like the normal (Gaussian) probability distribution function. The center x_0 represents the mean, while w^2 is the standard deviation.

The obtained results revealed three prevailing groups of PM$_{10}$ mass concentrations, i e those with the mean of 16 µg/m^3, 27 µg/m^3 and 38 µg/m^3. As it is found by investigations [8], average PM$_{10}$ levels at regional background sites in Europe range from 14 to 24 µg/m^3, with the exception of Sweden where the PM$_{10}$ levels were lower (8–16 µg/m^3). This fact allows to propose that the PM$_{10}$ group with the mean of 16 µg/m^3 can be associated with broad-based European PM$_{10}$ background concentration.

The second group could be associated with Šiauliai urban background concentration with the mean of 28 µg/m^3. This value is close to other urban background value, which according to [8] varied around 25 µg/m^3. The third group could be related with local PM$_{10}$ sources, for example, traffic, re-suspended dust, domestic fuel burning and others. The mean concentration of this group is 38 µg/m^3. This value is close to the value in Central and North European countries where it varies at an interval of 26–53 µg/m^3.

The total area under curve, which outlines the separate PM$_{10}$ group distribution in Fig 9, enables us to estimate the contribution of an individual group to the total PM$_{10}$ level.

Statistical parameters for the Gaussian fits to regional background, urban background and local sources of PM10 concentrations

PARAMETER	Regional background	Urban background	Local sources
y^0 (baseline offset)	0 ± 0	0 ± 0	0 ± 0
x (centre of peak)	15,48 ± 1,61	26,78 ± 1,76	37,53 ± 6,39
w (width of the peak at half height)	10,64 ± 1,74	12,93 ± 2,25	29,47 ± 5,87
A (area under peak)	9628,58 ± 4781,98	15376,75 ± 5271,72	11432,81 ± 4670,30

Analysis shows that for the Šiauliai station, the regional, urban background and local contributions to the total PM_{10} are estimated to be 36 %, 30 % and 34 %, respectively, during June–October of 2005. It should be noted, that this proportion is somewhat different during summer months: 46 %, 30 % and 24 %, respectively, and this proportion can be different for each country and even city [24].

This method allows to define which part of the measured PM_{10} concentration can be controled. It is evident that the control of the regional background PM_{10} concentration is practically impossible. However, the influence of the other two sources can be reduced by local means.

4. Conclusions

1. The analysis of the PM_{10} mass concentration showed that the weekdays-weekends phenomenon was observed in Šiauliai. The mean concentration of PM_{10} mass concentrations on weekdays was higher in comparison with concentration at weekends, 24,6 $\mu g/m^3$ and 21 $\mu g/m^3$, respectively.

2. Clear relationship between PM_{10} mass concentration and the wind parameters (speed and direction) was found. The wind as well as PM_{10} concentration was not registered in the direction of 360°–30°.

3. Linear relationship between aerosol number and PM_{10} mass concentrations was found at a high particle number concentration (more than 18 000 cm^{-3}).

4. Analysis shows that PM_{10} level in Šiauliai is formed from three sources: the regional background, city background, and local sources. These contributions to the total PM_{10} mass concentration were estimated to be 36 %, 30 % and 34 %, respectively, during June–October of 2005.

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PM_{10} MASINĖS IR AEROZOLIO SKAITINĖS KONCENTRACIJŲ KITIMAI ŠIAULIUOSE

R. Girgždienė, R. Rameikytė

Santrauka

Remiantis Valstybinio oro monitoringo duomenimis, gautais Šiaulių stotyje, atlikta PM_{10} masinės koncentracijos pokyčių analizė. Papildomo eksperimento metu lygiagrečiai matuota aerozolio ($d > 0{,}4$ μm) skaitinė koncentracija Šiauliuose. PM_{10} koncentracijos analizė parodė, kad per savaitę ji kito, t. y. buvo nustatytas savaitgalio efektas. Vidutinė PM_{10} masinė koncentracija darbo dienomis buvo didesnė, palyginti su koncentracija savaitgaliais, atitinkamai 24,6 $\mu g/m^3$ ir 21 $\mu g/m^3$. Nustatytas ryšys tarp PM_{10} masinės koncentracijos ir vėjo parametrų (greičio ir krypties). Parodyta, kad prietaisai, esantys šioje stotyje, dėl jos specifinės padėties neregistruoja vėjo ir kartu PM_{10} koncentracijos 360^o–30^o krypties. Nustatytas tiesinis ryšys tarp aerozolio skaitinės ir PM_{10} masinės koncentracijos, kai aerozolio skaitinė koncentracija viršija 18 000 dalelių/cm^3. Gauta, kad, padidėjus aerozolio skaitinei koncentracijai 1 000 dalelių/cm^3, PM_{10} masinė koncentracija išauga apytikriai 1,7 $\mu g/m^3$. PM_{10} lygis Šiauliuose gali būti charakterizuotas kaip trijų šaltinių suma: regiono foninio, miesto foninio ir vietinio. 2005 m. birželio–spalio mėn. šių šaltinių indėlis į PM_{10} koncentracijos lygį buvo įvertintas apytikriai atitinkamai 36 %, 30 % ir 34 %.

Reikšminiai žodžiai: PM_{10}, aerozolio skaitinė koncentracija, savaitgalio efektas, dieniniai svyravimai, savaitiniai svyravimai, PM_{10} šaltiniai.

ИЗМЕНЕНИЕ МАССОВОЙ И СЧЕТНОЙ КОНЦЕНТРАЦИИ АЭРОЗОЛЯ PM_{10} В ШЯУЛЯЙ

Р. Гиргждене, Р. Рамейките

Резюме

Проведён анализ массовой концентрации PM_{10}. Счётная концентрация аэрозоля была измерена в течение десятидневного эксперимента. Анализ показал изменения в распределении массовой концентрации PM_{10} в течение недели. Средняя массовая концентрация PM_{10} в течение рабочих дней была выше (24,6 мкг/м^3) по сравнению с концентрацией в выходные дни (21 мкг/м^3). Была найдена отчетливая связь между массовой концентрацией PM_{10} и параметрами ветра (скоростью и направлением). Выявлено линейное соотношение между счётной концентрацией аэрозоля и массовой концентрацией PM_{10} при высокой концентрации частицы (больше 18 000 частиц/см ^{3}). Уровень концентрации PM_{10} в Шяуляй был определен как сумма трех источников: регионального, городского и местного. Вклад источников в общую массовую концентрацию PM_{10} составил 36 %, 30 % и 34 % в течение июня–октября 2005 года.

Ключевые слова: PM_{10}, счётная концентрация аэрозоля, эффект конца недели, суточное изменение, недельное изменение, источники PM_{10}.

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