Variations of fine and coarse urban atmospheric aerosol concentrations in Riga City, Latvia

A. Osite, A. Viksna, J. Kleperis, and I. Steinberga

***Abstract*—**Atmospheric fine and coarse particulate matter were measured in Riga City during two sampling campaigns. The aim of the research was to determine the concentration levels of urban aerosols in Riga centre on typical canyon streets and at the monitoring station which is classified as urban-industrial, and it is located closely to Riga Freeport territory. The first sampling campaign was arranged at the urban-industrial monitoring station. The concentrations of airborne particulate matter (PM_{10} and $PM_{2.5}$) were measured from the end of April until the end of December 2007. During the second sampling campaign the concentrations of particulate matter of different fractions (PM_{10}, $PM_{2.5}$ and PM_1) were measured during the time period from October 2009 until December 2010 in the city centre on two canyon streets. Concentrations and seasonal variations of urban aerosols were evaluated analyzing correlations with meteorological parameters and other pollutant gases. At the Riga Freeport territory the maximum daily concentration of $PM_{2.5}$ and PM_{10} reached 52.5 μg m^{-3} in June and 83.4 μg m^{-3} in December, respectively. On the densely congested traffic streets – Brivibas and Kr. Valdemara Street PM_{10} concentrations exceeded both thresholds: an average daily concentration higher 50 μg m^{-3} more than 35 times per year, and the annual average concentration - 40 μg m^{-3}.

***Keywords*—**Coarse and fine particulate matter, meteorological factors, PM_{10}, $PM_{2.5}$, PM_1

I. Introduction

The ambient atmosphere contains different size and content particulate matter that is a complex chemical mixture of naturally occurring materials and materials resulting from human activities. During last decades, since the impact of aerosols on human health [1] – [3] and climate [4] – [6] has become of global importance, there has been increased concern about the monitoring and investigation of airborne aerosols and they have been studied extensively [7], [8]. These particles come in many different size ranges such as coarse, fine and ultra fine.

Particles with diameter between 2.5 and 10 μm can enter the lungs, however, those with diameter less than or equal to 2.5 μm can reach the alveolus and from there enter the blood-stream. It is also widely described in literature that particularly aerosol particles of fine dimensions exert an influence on the Earth's atmospheric energy budget directly through scattering and absorption of Solar radiation and indirectly by functioning as cloud condensation nuclei [9], [10]. But there is still a fundamental lack of understanding the underlying mechanisms of their toxicity; one of the widely accepted hypotheses is that toxicity of particulates depends not only on their size but also on their composition both of which depend on location, time of year and meteorological conditions [11]. As the particulate matter contains inorganic ions among them harmful metallic compounds, crustal compounds, black carbon and hundreds of organic compounds formed from incomplete combustion of fossil fuels and pyrolysis of organic materials, chemical characterization of aerosols is required in order to achieve a more complex picture.

Recently in many European countries the concentrations of particulate matter has been decreased due to modern technologies [12], [13]. However the main air pollution problem - high mass concentration levels of the particulate matter in Riga city centre is still associated with traffic intensity. Evaluation of mass concentrations of PM_{10} and $PM_{2.5}$ obtained from measuring places distributed around the Riga city indicated that excesses of PM_{10} and $PM_{2.5}$ air quality standards were found in measuring places with heavy traffic capacity.

On 28 January 2010 Latvia has received the European Commission's formal notification of the infringement procedure, Case No. 2008/2195, which indicates that the submitted air quality assessments during time period of 2007 and 2008 shows that the agglomeration of Riga has exceeded the threshold for human health for particulate matter PM_{10} - the annual (40 μg m^{-3}) and the daily threshold (50 μg m^{-3} of the calendar year may not exceed more than 35 times) defined

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by the Directives of European Parliament and Council (1999/30/EK and 2008/50/EK). This suggests that the first Action Program is not effective enough to ensure that air quality thresholds are met. The interest of this research is to evaluate concentration variations of ambient particulate matter in different areas of Riga, to find any relationships between particulate matter concentrations, meteorological parameters and some other pollutants, and ascertain reasons of exceedances of particulate matter.

II. Experimental

Study area

Riga (56°56´N, 24°6´E, 7 m above sea level) is the largest, most populated (about 704.000 inhabitants) capital city of Latvia. It is situated in the central part of Latvia, along the Baltic Sea, at the southern seashore of the Gulf of Riga and on the right bank of the River Daugava. The natural terrain of this area is a flat and sandy plain and the territory covers about 310 km^2 from which the industrial territories occupied approximately 17 %.

Humid continental climate is typical for Baltic States. The climate of Riga is maritime and temperate, influenced by its close proximity to the Baltic Sea. Summers tend to be short and comparatively warm with cloud cover, and temperatures average around +16 to +20 ºC, while the temperature on the hottest days can exceed +30 °C. The coldest months are January and February, when the average temperature is −5 °C, but temperatures as low as −20 °C to −25 °C can be observed almost every year on the coldest days. Snowfall is heavy and cover usually lasts from mid-December to mid-March. About 40% of the days per year are cloudy, average precipitation 700 mm a year.

During the last 15 years, a significant decrease in total emission of gaseous pollutants has been detected, e.g. the total SO_2 emissions decreased by 96 %, NO_x emissions by 43 % and NH_3 emissions by 72 %. The reduction is mainly due to use of fuels with lover content of sulphur as well as switching from solid and liquid types of fuel to natural gas and biomass; due to fuel consumption and increasing use of catalyst cars. The main sources of the particulate matter emission are the commercial and residential sectors. Combustion of wood and wood products accounts for about 72 % of total PM_{10} emission, against industry 10 %, traffic 9 % and agriculture 6 %. The long-range pollution transport significantly contributes to PM_{10} pollution level, caused by the geographical position and close proximity of the sea. Secondary particles are mainly formed by oxidation of SO_2 and NO_x and reaction with ammonia. Long-range pollution transport contributes about 80 % for oxidized sulphur, 75 % for oxidized and reduced nitrogen from total deposition in Latvia. In addition, there is a large contribution from natural sources, e.g. soil dust and sea spray.

Sampling sites

The first sampling campaign was organized at the monitoring station which is classified as urban-industrial, and it is located closely to Riga Freeport territory. The Freeport of Riga is a significant global and regional cargo supplier. In 2010 the volume of the transshipped cargoes has reached 30.5 million tons, 4040 vessels were mounted. Main types of cargo handled are containers, various metals, timber, coal, mineral fertilizers, chemical cargoes, oils and food products.

During the second campaign three sampling sites in Riga city centre were chosen to monitor mass concentrations of coarse and fine fraction aerosol particles at traffic sites. One of the monitoring places is situated on the Kr.Valdemara Str. 48. The sampling device was placed at the level of 2nd floor in the Faculty of Chemistry (University of Latvia). Kr. Valdemara Street is a typical canyon street. The second monitoring station is located on the same canyon street – Kr. Valdemara Str. 18. This monitoring station belongs to Riga City Council [14]. The distance between both sampling places is about 0.8 km. The third monitoring station that belongs to the Latvian Environment, Geological and Meteorological Centre (LEGMC) [15] is situated on Brivibas Str. 73 which is parallel to Kr. Valdemara Str. Two last mentioned stations are measuring particulate matter on street level.

Sampling and analysis

The campaign of particulate matter sampling and measuring at the monitoring station close to the Freeport was held from 28 April to 31 December 2007. Particulate matter PM_{10} and $PM_{2.5}$ measurements were done by beta gauge method, SM200 equipment. The field campaign was performed by Latvian Environment, Geological and Meteorological Centre (LEGMC).

The sampling and measuring of coarse and fine urban aerosols in the central part of Riga was carried out from October 2009 until December 2010 at the three urban monitoring sites. PM_{10}, $PM_{2.5}$ and PM_1 measuring data from Environmental Dust Monitor 165 (Grimm Aerosol Technique) placed at the first monitoring site on Kr. Valdemara Str. 48 are available for one year period from October 2009 to September 2010. The Environmental Dust Monitoring System provides precise particle size and count measurements, nearly independent of the particle color and moisture. A defined sample air volume containing suspended particles of various sizes is continuously drawn through a focused laser beam. Each scattered signal generated from a single particle is detected at 90 degrees by a high speed photo diode. These signals are counted and classified into 15 different size channels by an integrated pulse height analyzer. Finally, the counts are converted into a mass distribution and then formatted into the appropriate EPA categories PM_{10}, $PM_{2.5}$ and PM_1.

PM_{10} data obtained from the second monitoring station on Kr. Valdemara Str. 18 are represented from October 2009 to December 2010, except June – August 2010. PM_{10} was measured by ESM FH62. The apparatus utilizes the radiometric principle of beta-attenuation by an accumulated dust layer on glass fiber filter tape and is designed to measure the mass concentration continuously. The determination of the mass concentration is independent from the particle form, color and size. Ambient air is sucked through the sample system and the dust particles contained in the air are deposited on the filter continuously. The layer of dust is building up and this increasing dust mass weakens the intensity of the beta

radiation beam. The beta mass absorption shows only a very slight dependence on the chemical composition.

In the third monitoring station on Brivibas Str. 73 PM_{10} and $PM_{2.5}$ mass concentrations were measured from October 2009 to December 2010. Measuring was done also by beta gauge method, SM200 equipment, Opsis AB. The SM200 is an automatic semi-continuous particle sampler that is equipped with PM_{10} and $PM_{2.5}$ head. The SM200 can be operated unattended because of the large number of filters in its filter magazine. The aparatus loads one 47-mm filter from the clean filter magazine into the sampling chamber and, after sampling, unloads filter in the storage filter magazine. A Geiger-Muller detector detects the radioactivity before the filter is unloaded. A differential technique is used to measure particle mass and accounts for air density alternations and the effects of the natural radioactivity associated with a sample. The SM200 beta source is ^{14}C, and two interconnected microcontrollers allow sampling and measuring to be done simultaneously. The measurement chamber is thermo regulated to minimize air density alterations due to temperature variations.

III. Results and discussion

Evaluation of ambient particulate mater concentrations

Air quality in Riga is affected by pollution from stationary, mobile sources, as well as transboundary pollution. Pollutant concentrations in urban areas are affected also by climatic factors such as strength and direction of wind, as well as a rainfall. During the combustion of fuels the different pollutants are emitted in the atmosphere, while the quantity and chemical composition depends from the fuel quantity and type used. The largest source of particulate emissions in Latvia is heat and energy production, including the household sector. It is estimated that wood (biomass) burning is contributing about up to half of the total particulate emissions. Natural gas (imported from Russia), however, is the most widely used source in Latvia to produce heat and electricity; oil products and coal are used in relatively small quantities. Comparing with other co-generation forms that are used to produce heat and electricity, burning of natural gas is giving small particle emissions, while exhausts from combustion of oil products and coal consist of both fine and coarse particles and their precursors. Nevertheless the energy policy in Latvia in general and the support mechanisms for renewable energy sources in particular have suffered from inconsistencies and, despite public interest in these issues and the EU's legal requirements, the policy has been subject to political interests.

Exhausts form fuel combustion in vehicles contain particles (fine and ultra-fine fraction), as well as carbon oxides, nitrogen oxides, hydrocarbons. These emissions are dependent from the age of the vehicle and the fuel used. Black carbon (BC) or soot constituent in PM_{10} concentrations measured on Kr.Valdemara Str. in previous research showed that the main source of BC pollution is road transport and BC concentrations are directly proportional to the intensity of traffic flow on the street. It can be estimated that the exhausts from internal combustion engines gives an average of 15% from observed PM_{10} concentrations. To reduce this part of pollution, it is recommended to use an alternative vehicles – with cleaner fuel (biogas), electric (hydrogen) vehicles or to reduce number of cars on the streets.

The PM_{10} monitoring provided by LEGMC at the typical background stations in Riga (Viestura Boulevard and Tvaika Str.) showed that the average annual mass concentrations of PM_{10} varied from 32.0 μg m^{-3} (2005) to 20.4 μg m^{-3} (2009). No violation to the PM_{10} European air quality standards (40 μg m^{-3} annual average and 50 μg m^{-3} average daily values) was observed over all monitoring period from 2004. The annual average mass concentrations of PM_{10} in background stations are almost twice lower than concentrations detected in urban sampling sites which are located directly over rather heavy traffic. The annual average concentrations of PM_{10} at monitoring stations on Brivibas and Kr. Valdemara Str. in 2009 were 38.6 and 39.9 μg m^{-3}, but in 2010 39.9 and 41.9 μg m^{-3} respectively. Data of ambient PM_{10} levels from previous years (2005-2008) in these monitoring stations recorded exceeds of threshold of annual average PM_{10} mass concentration which is 40 μg m^{-3}. The numbers of days when the 24 h PM_{10} objective of 50 μg m^{-3} was reached were more than 35 allowed days in a year. On the other hand, the positive tendency has occurred during last years when 24 h PM_{10} objective exceedances are decreasing about 9 % in every year.

The concentrations of $PM_{2.5}$ and PM_{10} fractions of ambient airborne particulates were measured daily at the monitoring station in the industrial site of the Freeport. The highest daily concentration of $PM_{2.5}$ reached 52.5 μg m^{-3} (June 12, 2007), but the lowest 1.3 μg m^{-3} (October 13, 2007). Highest monthly variations of $PM_{2.5}$ were detected in June and November, unfortunately any substantial seasonal variations were not found. The highest daily concentrations of PM_{10} reached 83.4 μg m^{-3} (December 27, 2007), but the lowest 3.5 μg m^{-3} (April 20, 2007). Changes of monthly PM_{10} and $PM_{2.5}$ mass concentrations with corresponding standartdeviations from April until December, 2007 at the industrial monitoring site are showed in Fig. 1.

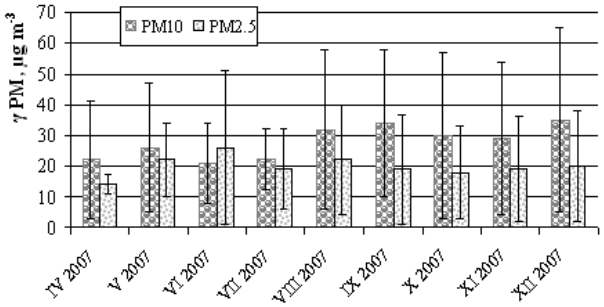

Fig. 1 Monthly variations of PM_{10} and $PM_{2.5}$ from April until December, 2007 at the industrial monitoring site

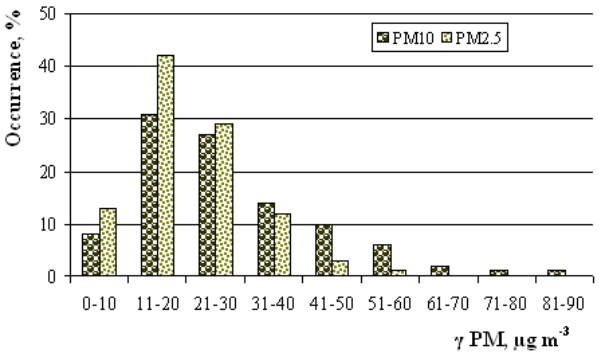

Fig. 2 Distribution of PM_{10} and $PM_{2.5}$ concentrations during April – December 2007

$PM_{2.5}$ concentration range from 11 until 20 µg m^{-3} was dominating for all period of measurements (42 % of all cases) (see in Fig. 2). Highest concentration variations are observed in the winter period, but lowest in the late spring-early summer period. PM_{10} concentration range from 11 until 30 µg m^{-3} was dominating during all period of measurements (58 % of all cases) (see in Fig. 2). Obtained results of PM_{10} concentrations showed that during the summer time concentrations are very stable, they are changing very slowly. It means that the influence of local sources is not so important and PM_{10} pollution mainly originates from long-range transboundary and background pollution. Relationships between $PM_{2.5}$ and PM_{10} mass concentrations were analyzed in terms of linear regression and the results revealed that coefficient of determination were 0.396. The relatively middle coefficient shows that $PM_{2.5}$ and PM_{10} could have some similar emission sources and different emission sources as well, and they could be influenced by the same local conditions. Such a relatively low coefficient of determination indicate significant contributions from primary sources such as resuspended soil/road dust and other mechanical activities [16], [17], [18].

Monthly means of PM_{10} mass concentrations over the sampling period from October 2009 until December 2010 from three urban sampling sites are showed in Table 1, where minimal and maximal values are daily average values. In Riga, the highest monthly average concentrations of PM_{10} were recorded in January and April 2010.

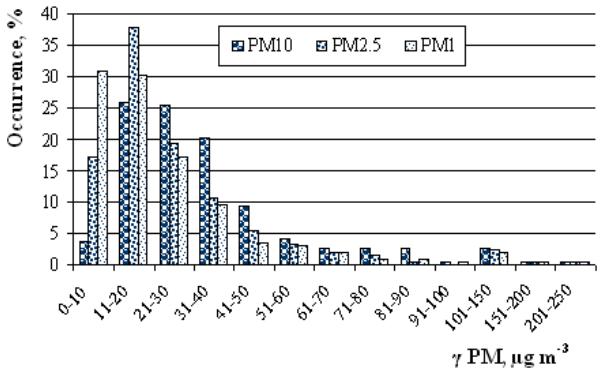

Fig. 3 Distribution of PM_{10}, $PM_{2.5}$ and PM_1 concentrations during 2009/2010

PM_{10} concentration range from 11 until 30 µg m^{-3} was dominating for all period of measurements (about 50 % of all cases). For $PM_{2.5}$ the dominating concentration range was from 11 until 20 µg m^{-3} (see in Fig.3).

But if we analyze the numbers of days with 24 h PM_{10} objective exceedances, it is seen that concentrations appeared to be marginally higher during entire cold season. It could be explained probably due to the high atmospheric stability and reduced air mixing on cold winter days. The lowest air temperature almost in all territory of Latvia in January 2010 dropped to minus 25 till minus 30 ºC. Due to such meteorological situation the highest hourly mean PM_{10}, $PM_{2.5}$ and PM_1 mass concentrations reached 330, 307 and 299 µg m^{-3} respectively. The same situation was detected in our neighboring country Lithuania [19]. During these cold days smog which is not typical for Riga was originated when concentrations of NO_2 were reaching even 200 µg m^{-3} and concentration of O_3 50 µg m^{-3} accordingly.

Analyzing the average daily PM_{10} concentration measured at the three urban stations, the dependence from the season is observed. So, for example, in winter (January 2010) the average daily PM_{10} concentrations measured in all three urban stations were changing very similarly (Fig. 4), but in spring (April 2010) the measured PM10 concentrations were sometimes even showing an opposite direction (Fig.5).

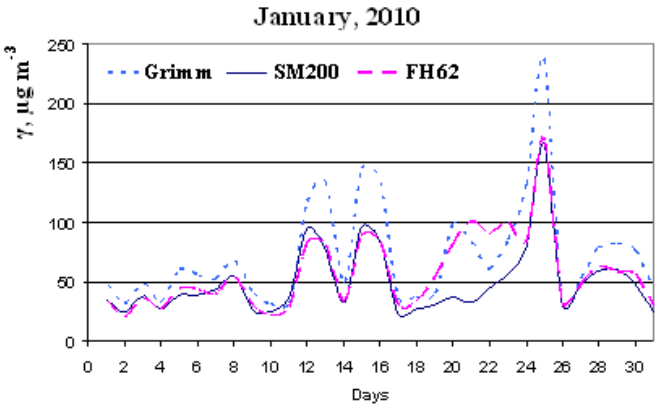

Fig. 4 Average daily PM_{10} concentrations in January 2010 (Grimm – Kr.Valdemara Str. 48; SM200 – Brivibas Str. 73; FH62 – Kr.Valdemara Str. 18; data from Riga City Council, LEGMC, University of Latvia)

This suggests that in the winter season when air temperature is low and humidity high (also high is probability to form aerosols) the air pollution in Riga centre is similar in different places and determined mainly by urban background pollution, which consists of all, both stationary and mobile emission sources. But in spring and summer seasons, air pollution in each local street develops differently, determined mainly by local emission sources in each site.

Table 1. Monthly mean PM_{10} mass concentrations (μg m^{-3}), corresponding standard deviations, minimal and maximal values at three urban measuring sites in Riga

	γ PM10, μg m^{-3}											
	Valdemara Str. 48				Valdemara Str. 18				Brivibas Str. 73			
	Mean	Std	Max	Min	Mean	Std	Max	Min	Mean	Std	Max	Min
2009/10	**39**	9	74 (2)	6	**34**	26	201 (4)	4	**36**	16	67 (7)	18
2009/11	**34**	11	70 (2)	18	**36**	20	91 (6)	4	**36**	11	56 (2)	14
2009/12	**41**	26	130 (4)	9	**43**	40	288 (10)	3	**33**	17	94 (4)	13
2010/01	**73**	46	237 (18)	31	**57**	33	171 (15)	21	**50**	31	166 (10)	24
2010/02	**51**	17	76 (5)	24	**43**	13	78 (7)	26	**42**	14	86 (6)	25
2010/03	**37**	19	77 (5)	9	**35**	18	80 (6)	10	**37**	17	89 (5)	11
2010/04	**39**	21	101 (8)	17	**52**	16	88 (17)	28	**55**	21	99 (13)	18
2010/05	**23**	10	43 (0)	8	**42**	17	77 (6)	19	**36**	12	63 (2)	19
2010/06	**21**	9	51 (1)	10	*				**33**	12	62 (2)	12
2010/07	**26**	7	40 (0)	13	*				**36**	14	66 (2)	7
2010/08	**22**	8	46 (0)	8	*				**36**	15	67 (3)	8
2010/09	**23**	11	47 (0)	11	**33**	12	59 (1)	17	**28**	10	52 (1)	7
2010/10	*				**37**	16	94 (4)	17	**42**	20	102 (7)	19
2010/11	*				**36**	14	76 (5)	14	**35**	15	77 (3)	17
2010/12	*				**35**	14	74 (4)	14	**38**	13	77 (4)	14

* Measurements were not done

In brackets number of days when PM_{10} > 50 μg m^{-3}

Table 2. Monthly mean $PM_{2.5}$ and PM_1 mass concentrations (μg m^{-3}), corresponding standard deviations, minimal and maximal values at two urban measuring sites in Riga

	γ PM2.5, μg m^{-3}				γ PM2.5, μg m^{-3}				γ PM1, μg m^{-3}			
	Brivibas Str. 73				Valdemara Str. 48							
	Mean	Std	Max	Min	Mean	Std	Max	Min	Mean	Std	Max	Min
2009/10	**23**	8	39	10	**29**	9	40	9	**25**	8	33	7
2009/11	**27**	6	45	17	**27**	10	58	14	**24**	10	52	13
2009/12	**25**	15	55	12	**32**	24	126	8	**29**	23	121	6
2010/01	**33**	15	57	18	**63**	41	206	29	**60**	40	198	28
2010/02	**31**	9	52	20	**48**	16	71	23	**46**	15	69	22
2010/03	**26**	10	47	12	**31**	14	66	8	**28**	13	60	7
2010/04	**42**	14	60	11	**26**	15	63	9	**24**	17	89	6
2010/05	**29**	9	45	14	**14**	5	22	5	**11**	4	18	4
2010/06	**26**	9	50	11	**13**	7	35	5	**10**	6	29	4
2010/07	**27**	7	43	11	**15**	4	26	9	**11**	4	18	5
2010/08	**24**	11	48	9	**14**	5	28	6	**11**	4	22	3
2010/09	**18**	6	27	7	**16**	8	32	8	**14**	8	28	6
2010/10	**25**	11	62	12	*				*			
2010/11	**21**	8	41	8	*				*			
2010/12	**33**	11	57	17	*				*			

* Measurements were not done

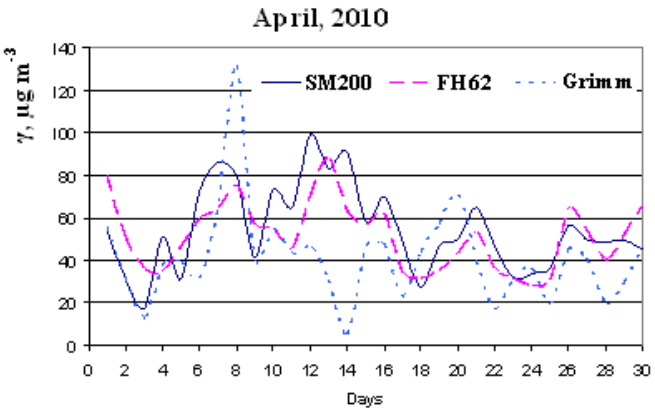

Fig. 5 Average daily PM10 concentrations in April 2010 (Grimm – Kr.Valdemara Str. 48; SM200 – Brivibas Str. 73; FH62 – Kr.Valdemara Str. 18; data from Riga City Council, LEGMC, University of Latvia)

Higher PM_{10} mass concentrations in April are related with snow melting and remaining of sand/salt mixture (what is used for safe driving during winters) on streets what causes particle resuspension. Also burning of branches and leaves in private gardens in spring and autumn seasons, as well as the burning of dead grass around Riga in the spring gives the urban pollution with particulate matter. Probably all before mentioned factors are reason for long PM pollution episode (even 16 days in 2010) what systematically occur during April. On the other hand, when the territory of Europe was exposed to the influence of volcanic origin emission (from Iceland volcano April 16 – 22, 2010), only one exceedance of PM_{10} (53 μg m^{-3}) was observed at Kr. Valdemara Str. 18. At the same time at other sampling stations no exceed of PM_{10} threshold were observed. Seasonally, the highest concentrations are measured during October – April, the lowest ones during May – September. Similar seasonal variations of $PM_{2.5}$ and PM_1 mass concentrations are observed. $PM_{2.5}$ and PM_1 mass concentrations from two measuring sites are shown in Table 2, where minimal and maximal values are daily mean values. From 2010 the target value for human health protection is set for fine particles $PM_{2.5}$ – annual average concentration 25 μg m^{-3}. However, monitoring data shows that mean measured concentration in 2009 - 2010 was 27 μg m^{-3} at the both sampling sites. Obtained data recorded that mass concentration values of fine aerosol particles are marginally high during colder days.

The highest monthly average values of $PM_{2.5}$ and PM_1 during entire sampling period were observed in January, 2010 - 63 and 60 μg m^{-3} respectively. Lowest ones of $PM_{2.5}$ and PM_1 - 13 and 10 μg m^{-3} in June 2010 at measuring site on Kr. Valdemara Str. 48. The difference between mass concentrations of coarse and fine particulate matter is strongly reduced during high pollution episode during January and February 2010. It should be noted that during these months contribution of PM_1 to total amount of particulate matter is the highest over one year period and it exceeded more than 80 % (Fig. 6). Fine particles are associated primarily with combustion in different stationary and mobile sources. Contribution of coarse particles is higher in spring and summer months than in winter time.

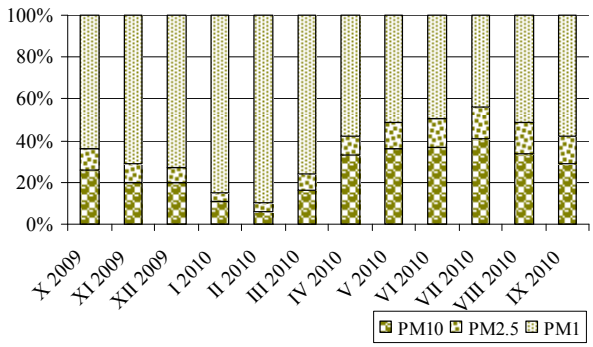

Fig. 6 PM_{10}, $PM_{2.5}$ and PM_1 proportions during one year (data from sampling site on Kr.Valdemara Str.48)

Results of ambient particulate matter from all measuring sites are in good agreement. Slight differences which were observed are allowable due to distance between sampling stations, different measuring equipment and slightly various traffic intensities.

Relationship between PM and other pollutants and meteorological factors

The relationships between PM, some gaseous pollutants and meteorological parameters (temperature, wind speed, relative humidity and precipitation) were investigated by Pearson's correlation analysis. Pearson correlation coefficients were obtained using SPSS (Statistical Package for the Social Sciences). The meteorological data, which were found to be significant in describing the variation in the 24-h averaged PM concentrations during sampling campaign in 2007, are: air temperature (ºC), relative humidity (%), wind speed (m s^{-1}) and precipitation (mm). The correlation coefficients of daily average PM, particle related substances and meteorological parameters are shown in Table 3. The correlation coefficients were marked in the table to indicate the significance levels ($P<0.01$ or <0.05). The statistically strongest correlations were detected for $PM_{2.5}$-temperature, PM_{10}-precipitation, PM_{10}-Ozone.

Table 3

Pearson correlation coefficients of PM_{10} and $PM_{2.5}$, particle related substances and meteorological parameters (during sampling campaign in 2007)

Parameter	PM_{10}	$PM_{2.5}$
O_3	-0.24*	0.09**
SO_2	0.166**	0.05**
NO_2	0.065**	0.07**
Precipitation, mm	-0.27*	-0.16**
Temperature, ºC	0.008**	0.28*
Wind speed, m s^{-1}	-0.07**	-0.03**
Relative humidity, %	0.122**	-0.13**

** Correlation is significant at the 0.01 level.
* Correlation is significant at the 0.05 level.

Wind speed long has been recognized as an important controlling factor on concentrations of air pollutants [20], [21], [22], [23] but in this study there is no any statistical relationship between wind speed and particulate matter pollution.

The effects of rainfall on particulate concentrations are detected as function of time gap, were found some evidences that after longer periods without rainfall particulate matter concentrations raising, what means that PM have tendency to accumulate in atmosphere. Moreover a precipitation is much more effective mechanism for particulate removal from atmosphere comparing to wind speed.

Pearson correlation coefficients of data obtained during the sampling campaign in 2009 – 2010 are shown in Table 4. The correlation of PM and CO is generally stronger among other correlations. Pearson correlation coefficient detected for PM_{10}-CO is 0.639 and it is slightly higher than coefficient detected for $PM_{2.5}$-CO and PM_1-CO which in both cases is 0.620.

Table 4
Pearson correlation coefficients of PM_{10}, $PM_{2.5}$ and PM_1, particle related substances and meteorological parameters (during sampling campaign in 2009/2010)

Parameter	PM_{10}	$PM_{2,5}$	PM_1
Global Radiation, kw/m²	-0.247**	-0.330**	-0.336**
Temperature, °C	**-0.445****	**-0.546****	**-0.568****
O_3, µg m^{-3}	**-0.377****	**-0.381****	**-0.374****
CO, mg m^{-3}	**0.639****	**0.620****	**0.620****
Precipit. duration, s	-0.074**		
Precipit. between measurements, mm	-0.135*		-0.128*
Pressure above sea level, hPa	0.329**	0.291**	0.288**
Pressure at station level, hPa	0.323**	0.282**	0.279**
Wind direction, deg	-0.212**	-0.200**	-0.199**
Wind speed, m/s	-0.218**	-0.234**	-0.228**

** Correlation is significant at the 0.01 level.
* Correlation is significant at the 0.05 level.

PM and CO correlation in different concentration diapasons is shown in Fig.7. It is well seen that during high PM mass concentrations episodes there are observed also high mass concentrations of CO.

The daily mean mass concentrations of all PM fractions negatively correlated reasonably well with O_3. Correlation coefficients are quite similar for both coarse and fine particulate matter fractions. Fig. 8 is clearly indicating that exceeded PM mass concentrations are observed while O_3 mass concentrations remain less than 25 µg m^{-3}.

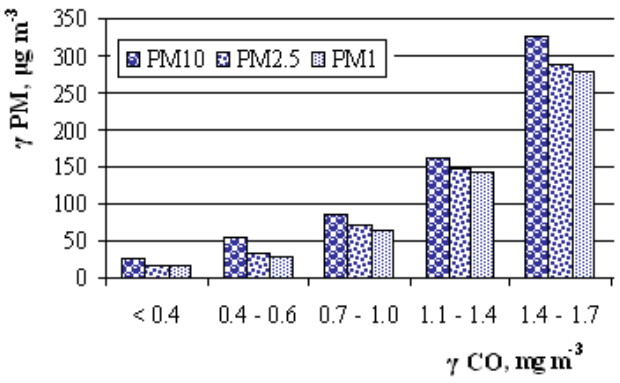

Fig. 7 PM and CO correlation in different concentration ranges

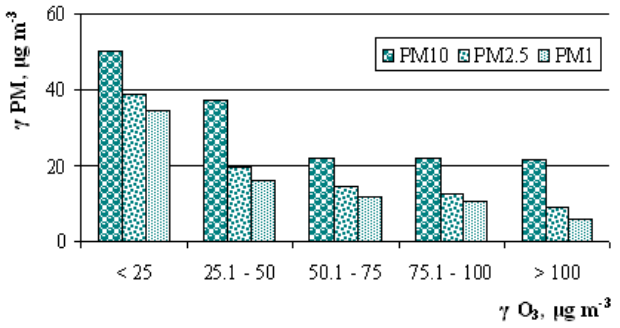

Fig. 8 PM and O_3 correlation in different concentration ranges

In this case rather close correlation of polluters probably pointed to comparatively insignificant pollution level of background and long range transport than the influence of local origin pollution sources.

It is well known that not only intensity or character of pollution sources influence level of particulate matter, but also meteorological factors [24], [25]. The correlation analysis of PM and temperature (Fig. 9) was the strongest among other meteorological factors and it was negative. For fine particulate matter correlation coefficients were even higher than for coarse particles. This could confirm hypothesis that during stable atmospheric conditions pollutant dispersion is reduced. This is also according to slight positive correlation between PM and atmospheric pressure observed in Riga. Also PM and global radiation correlation was negative. Finally, coarse and fine fractions of PM and wind speed, direction and precipitation anti-correlated.

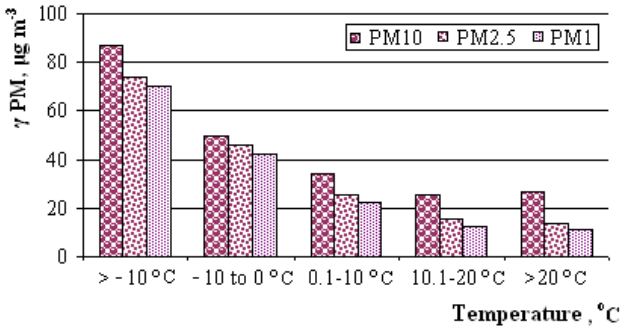

Fig. 9 PM mass concentration and ambient air temperature correlation in different temperature ranges

IV. CONCLUSION

Measurements of PM_{10} and $PM_{2.5}$ were performed at the industrial monitoring site close to the Freeport from April until December 2007 and measurements of PM_{10}, $PM_{2.5}$ and PM_1 mass concentrations were carried out at three urban sampling sites from October 2009 until December 2010 in Riga (Latvia). Results of urban ambient particulate matter from all sampling sites were in good agreement. In the measuring campaign during 2007 no seasonal variations were detected for $PM_{2.5}$, but for PM_{10} seasonal unstable and higher concentrations detected for winter period. Quite stable and inert $PM_{2.5}$ pollution levels prove that no strong impact of local sources and $PM_{2.5}$ pollution levels could be explained by long-range transboundary pollution and background pollution. But the monthly average concentrations of particulate matter recorded high concentration values during January, February and April 2010 and the lowest concentrations were during summer and early autumn months. The relatively middle coefficient of linear regression shows that $PM_{2.5}$ and PM_{10} could have some similar emission sources and different emission sources as well, and they could be influenced by the same local conditions. Such a relatively low coefficient of determination indicates significant contributions from primary sources such as re-suspended soil/road dust and other mechanical activities. Contribution of PM_1 to the total amount of particulate matter was the highest over one year period (2009-2010) and it reached more than 80 % during cold season, while it was about 50 % in summer time. It can be stated that PM_1 is the most important parameter for environmental pollution characterization of urban areas.

The statistically strongest correlations during 2007 were detected for $PM_{2.5}$-temperature, PM_{10}-precipitation, PM_{10}-Ozone. The effects of rainfall on particulate concentrations are detected as function of time gap, were found some evidences that after longer periods without rainfall particulate matter concentrations raising, what means that PM have tendency to accumulates in atmosphere. During the measuring campaign in 2009/2010 the strongest Pearson correlation coefficients were detected for PM-CO mass concentrations. It proves that major part of pollution originates from combustion processes in the internal combustion engines of vehicles and in the thermal power stations. Strong correlation of PM and air temperature affirms that meteorological conditions prevailing in Riga are not optimal for dispersion of atmospheric pollution. Such adverse weather conditions dominate during autumn, winter and spring periods, when pollution from the thermal power stations additionally enter into urban atmosphere to existing background and anthropogenic pollution.

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