Characterisation of Fine and Coarse Atmospheric Aerosols at the Measuring Sites in Riga City Centre, Latvia

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Abstract: - Good air quality in urban environment is an important issue for city sustainable development and prosperity of the population. Despite different improvements in vehicle, fuel and heating technologies that have made possible to reduce particulate matter emissions, high ambient aerosol concentrations are still an important challenge in several European cities, among them in Riga city (56°56´N, 24°6´E). PM_{10}, $PM_{2.5}$ and PM_1 in Riga city is evaluated from direct monitoring results during time period from October 2009 until December 2010. Monthly means of PM_{10} concentrations ranged from 21 to 73 μg m^{-3}, $PM_{2.5}$ from 13 to 63 μg m^{-3} and PM_1 from 10 to 60 μg m^{-3}. The data were used to characterize correlations between particulate matter concentrations, some other pollutants and meteorological factors. It is concluded that PM_{10} concentrations exceeded both thresholds: average daily concentration above 50 μg m^{-3} more than 35 times a year, and annual average concentration - 40 μg m^{-3} only on the densely congested traffic streets – Brivibas and Kr. Valdemara Street.

Key-Words: - **Coarse and fine particulate matter, PM_{10}, $PM_{2.5}$, PM_1, meteorological factors**

1 Introduction

The ambient atmosphere contains different size and content particulate matter that is complex chemical mixture of naturally occurring materials and materials resulting from human activities. Since the impact of aerosols on human health [1-3] and climate [4-6] is an environmental key concern, particulate matter have been studied extensively [7,8]. 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]. As particulate matter contain 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 [11,12]. However the main air pollution problem - high mass concentration levels of 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 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 objective of this paper is to evaluate concentration variations of ambient particulate matter at three sampling stations and ascertain reasons of exceedances of PM.

2 Experimental

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 310.00 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 in a year are cloudy, average precipitation 700 mm a year.

2.1 Sampling sites

During this research three sampling sites in Riga city centre were chosen to monitor mass concentrations of coarse and fine fraction aerosol particles at traffic and industrial 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 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 [13]. The distance between both sampling places is about 0.8 km. The third monitoring station that belongs to Latvian Environment, Geological and Meteorological Centre (LEGMC) [14] is situated on Brivibas Str. 73 which is parallel to Kr. Valdemara Str. Two last mentioned stations are measuring particulate matter on street level.

2.2 Sampling and analysis

Overall for the current research sampling and measuring was carried out from October 2009 until December 2010. PM_{10}, $PM_{2.5}$ and PM_1 measuring data from Environmental Dust Monitor 165 (Grimm Aerosol Technique) placed 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 colour 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 fibre filter tape and is designed to measure the mass concentration continuously. The determination of the mass concentration is independent from the particle form, colour 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 by SM200, 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.

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3 Results and Discussion

3.1 Evaluation of ambient particulate mater concentrations

PM_{10} monitoring provided by LEGMC in typical background stations in Riga (Viestura Boulevard, Tvaika Str.) showed that mean annual PM_{10} mass concentrations 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 mean and 50 daily values) was observed over all monitoring period from 2004. Annual mean PM_{10} mass concentrations in background stations are almost twice lower than concentrations detected in urban sampling sites which are located directly over rather heavy traffic. Annual mean PM_{10} concentrations in 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 on ambient PM_{10} levels from previous years (2005-2008) in these monitoring stations recorded exceeds of threshold of annual mean PM_{10} mass concentration which is 40 μg m^{-3}. The numbers of days on which 24 h PM_{10} objective of 50 μg m^{-3} was exceeded more than 35 allowed days in a year. On the other hand, positive tendency has occurred during last years when 24 h PM_{10} objective exceedances are decreasing about 9 % in every year. Monthly means of PM_{10} mass concentrations over the sampling period from October 2009 until December 2010 from three sampling sites are showed in Table 1, where minimal and maximal values are daily mean values. In Riga, the highest monthly means of PM_{10} levels were recorded in January and April 2010. 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 approached to 330, 307 and 299 μg m^{-3} respectively. The same situation was detected in our neighbouring country Lithuania [15]. 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.

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

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

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 winter) 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 both sampling sites. Obtained data recorded that mass concentration values of fine aerosol particles are marginally high during colder days.

The highest mean monthly 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. 1). 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.

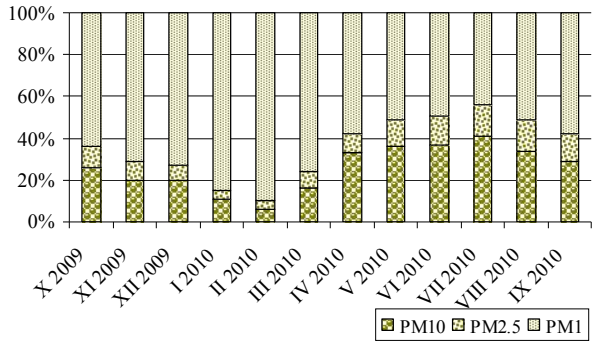

Fig.1 PM_{10}, $PM_{2.5}$ and PM_1 proportions during one year (data from sampling site on Kr.Valdemara Str.48)

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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.

3.2 Relationship between PM and other pollutants and meteorological factors

Pearson correlation coefficients were obtained between daily coarse and fine particulate matter concentrations and gaseous pollutants as well as meteorological parameters using SPSS (Statistical Package for the Social Sciences). Obtained correlation coefficients are shown in Table 3. The correlation between PM and CO is generally stronger among other correlation. Pearson correlation coefficient of PM_{10} is 0.639 and it is slightly higher than coefficient of $PM_{2.5}$ and PM_1 which in both cases is 0.620.

Table 3
Pearson correlation coefficients between daily PM_{10}, $PM_{2.5}$ and PM_1 and other parameters

Parameter	PM_{10}	$PM_{2,5}$	PM_1
Global Radiation, kw/m^2	-0.247**	-0.330**	-0.336**
Temperature, ^{o}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. 2. It is well seen that during high PM mass concentrations episodes there are observed also exceeded 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. 3 is clearly indicating that exceeded PM mass concentrations are observed while O_3 mass concentrations remain less than 25 µg m^{-3}.

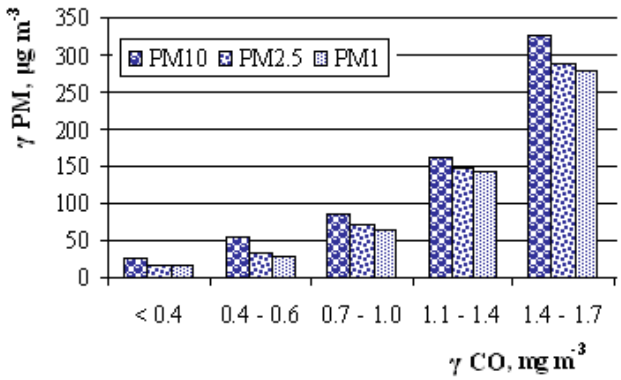

Fig. 2 PM and CO correlation in different concentration ranges

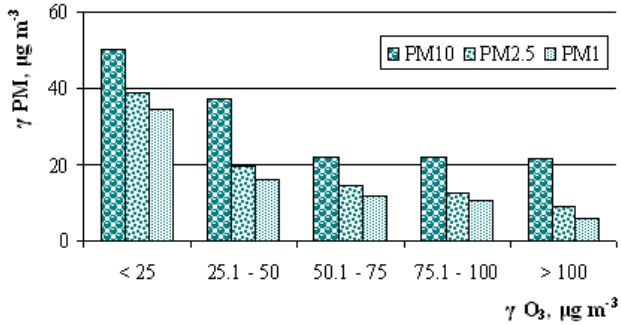

Fig. 3 PM and O_3 correlation in different concentration ranges

Obtained correlation coefficients indicated the activity of photochemical reactions during forming of O_3 pollution level. The influence of local origin pollution sources is observed during pollution formation process. 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 [16,17]. The correlation analysis between PM and temperature (Fig. 4) 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.

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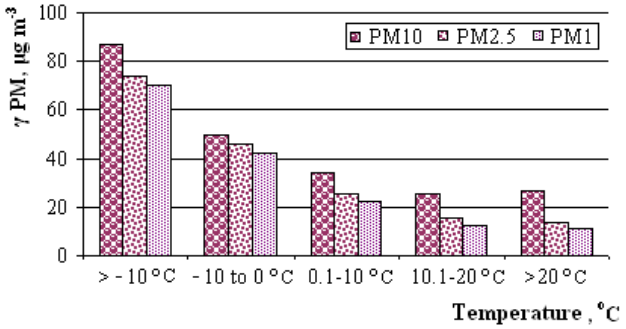

Fig.4 PM mass concentration and ambient air temperature correlation in different temperature ranges

4 Conclusion

Measurements of PM_{10}, $PM_{2.5}$ and PM_1 mass concentrations were carried out at three urban sampling sites in Riga (Latvia) during October 2009 until December 2010. Results of ambient particulate matter from all measuring sites are in good agreement. The monthly mean particulate matter concentrations recorded high concentration values during January, February and April 2010. The lowest concentrations were during summer and early autumn months. Contribution of PM_1 to total amount of particulate matter is the highest over one year period and it exceeded more than 80 % during cold season, while it is about 50 % in summer time. It can be stated that PM_1 is most important parameter for environmental pollution characterization of urban areas. The strongest Pearson correlation coefficients were between PM and CO mass concentrations. It testifies that large part of pollution originates from combustion processes in internal combustion engines of vehicles and in heat production plants. Strong correlation of PM and air temperature affirm that meteorological conditions prevailing in Riga are disadvantaged for dispersion of atmospheric pollution. Such adverse weather conditions dominate during autumn, winter and spring periods, when pollution from heat production plants additionally enter into urban atmosphere to existing background and anthropogenic pollution.

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