SOURCE APPORTIONMENT OF PM10, PM2.5 AND PM1 IN THE LARGER CITY IN THE NORTH OF ALBANIA

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Abstract:
In this paper we are focused on the determination of particulate matter concentrations in different locations in Shkodra city, Albania, and rural areas around it. Monitoring results show that principal aerosol sources in the city of Shkodra are traffic and residential activities. Overall measurement results show a particularly problematic situation, where PM concentrations in the city clearly exceed international recommendations. To improve this situation, it is recommended to order traffic, re-construct existing roads, substitute old vehicles with new ones, and improve also the fuel quality.

Keywords: PM concentrations, monitoring campaign, air quality

1. Introduction

Aerosols are classified as solid or liquid particles suspended in the air. Their size ranges from few nanometers up to hundreds micrometers. Particles smaller than 1 μm are found in free troposphere in concentrations 103-104 cm-3, while particle greater than 1 μm are found in concentrations less than 1 cm3. Aerosol particles play an important role in atmospheric processes. The principal aerosol impacts on atmosphere are air pollution (Schneider et al., 2004), related particulate matter especially in heavy populated industrial centers and global warming which is related with radiation budged of the atmosphere (Viana et al., 2008; Stier et al., 2004).

Aerosol particles are characterized by many parameters as physical, chemical or geometrical ones (Harrison et al., 1999; Harrison et al., 2006; Dall'Osto et al., 2006). These parameters are related with size, form, internal structure, chemical composition (Oliviera et al., 2007; Hu et al., 2008; Beekmann et al., 2007), and with many other characteristics (Van Dingenen et al., 2004). Over all geometrical parameters, the size of aerosol particles is the most important (Makela et al, 2000). Almost all aerosol particles are size dependent. Based on their size, aerosol particles are classified in fine mode (up to 2.5μm) and coarse mode (up to 10 μm). The fine mode can be divided into other finer modes; nucleation mode (up 20 nm), Atiken mode (20-90 nm), and accumulation mode (90-1000 nm). Nucleation and Atiken particles are both called ultrafine particles.

The major number concentration of aerosol particles are in ultrafine mode, while the major surface and mass concentrations are respectively in the accumulation and coarse modes.

Aerosol particles are divided into two main groups according to their origin; primary and secondary aerosols. Principal sources of primary aerosol particles are produced by natural and anthropogenic origin like volcanic activity, re-suspended dust, sea salt, industrial processes, etc (Harrison et al., 2008; Charron et al., 2007; Wang et al., 2006). On the other hand sources of secondary aerosol particles are physical and chemical processes that cause the formation of new particles in the atmosphere. Nowadays atmospheric studies aerosol research is one of the most important topics. Aerosol research is related with many atmospheric and environmental problems, such as new particle formation, particle growth rate, radiation balance, atmospheric electricity, air quality, etc (Liu et al., 2004). Various scenarios of aerosol concentrations in the atmosphere in nowadays are retrieved using sophisticated measurement instruments and information and communication technologies (Bushati, et al., 2010).

In urban areas or industrial centers air quality becomes one of the hottest topics in environmental and health problems (Maesano et al., 2007; Jones et al., 2004). The monitoring of air quality consists on the measurement of several meteorological parameters, chemical agents, particulate matter, etc (Yin et al., 2005; Zhao et al., 2009). The last parameter, particulate matter mass concentration, called PM, is one of the physical parameters in air quality monitoring. The most investigated environmental aerosol parameters are PM modes, like PM2.5 and PM10 (particles respectively with aerodynamic diameter up to 2.5 μm and 10 μm) (Kousa, 2002). Particles in PM2.5 mode can reach till alveolus, while PM10 particles can't penetrate, but they reach only on lung pathways. For an accurate PM analysis, the parameters of the distribution of aerosol mass concentration must be determinate (Zhang et al., 2004; Makkonen et al., 2010). The effects of PM on health occur at levels of exposure currently being experienced by most urban and rural populations in both developed and developing countries. Chronic exposure to particles contributes to the risk of developing cardiovascular and respiratory diseases, as well as of lung cancer (Jacqes et al., 2000). In few words, estimating aerosol concentrations gives a clear picture of atmospheric properties related with our living environmental.

Traffic is one of the most important factors influencing on air quality (Tiitta, 2009). Diesel vehicles contribute significantly to the air pollution burden (Oahn et al., 2010; Jones et al., 2005). The emissions depend on number factors such as vehicle age, engine design and operating conditions, lubrificant oil and fuel (Lim et al, 2010). Diesel engines in developing countries are generally old, and exhaust emission controls are rare. Most of particles emitted from diesel exhaust are in submicron size. The presence of particulate matter below ten microns size (PM10) is significant due to the associated respiratory health implications (Byrd et al., 2010). Especially diesel exhaust particle is a complex that has been linked to acute cardiopulmonary and vascular responses, chronic health effects, and lung cancer in a number of epidemiologic studies (Lee et al., 2010; Yi et al., 2010).

2. Methods and experimental setup

Several measurement campaigns of PM1, PM2.5 and PM10 have been realized in several points of Shkodra city. Shkodra is the biggest city in the north of Albania (42°4'14.20"N; 19°30'49.79"E). The city has about 120,000 inhabitants. The altitude of the city is around 6-12 m and it is bounded by mountains in the north and east.

Measurement campaign was spread in seven months, May-November 2009. The measured sites are selected based on a number of criteria, including some common characteristics between the sites, as well as some key differences. These locations have been selected for PM measurements:

- Two points in the center of the city (site 1)
- Main exit road of the city (site 2)
- Inner part of the city, distant from the main roads (site 3)
- Rural area of Zogaj, near the lake of Shkodra (site 4)

Two points in the center of the city and one point of main exit road are distant from the main roads by about 10-20 m. Traffic rate in two center points is about 15,000 vehicles /day, whereas in the rural site Zogaj the traffic rate is very limited, about 100 vehicles /day. In the rural area of Zogaj (42° 4'16.76"N; 19°23'55.89"E), near the lake of Shkodra, where there are also conducted measurements, traffic impact can be neglected. Numbers of light/heavy duty vehicles in the Shkodra city are given by the table 1.

Since Shkodra city cannot be considered as industrial center, main contributions on PM concentrations are traffic and residential wood combustion, the latter especially during the colder season. In the inner part of the city, the measurements are carried out in the gardens about 55 m far from the main roads, and protected from direct impact of traffic effect by several 10-20 m high buildings. Altitude measurements were about 1.5 m above the ground. Reading the PM concentrations in this measurement location, one can determine the city background PM concentration.

Monitoring process is realized in various meteorological conditions; fair weather, cloudy, rainy and foggy days. This diversity of meteorological conditions lets us notice also their effects on PM concentration. Number of days with and without precipitation is presented in the table 2.

For measurements and data collection an environmental dust monitor had been used, model GRIMM EDM 107, which enables simultaneous measurements of PM1, PM2.5 and PM10 (Martuzevicius et al., 2004). Measurement principle is light-scattering and the measurement range of this instrument is

0.25-32 μm. This size range is divided in three PM modes (PM1, PM2.5 and PM10) for environmental purposes. Concentration range is $1\text{-}2\cdot10^6$ particles/liter. Time response of the instrument is 6s, but it can be obtained by hourly or daily averaged data.
Also the dust whose mass distribution is measured can be collected in filters for further chemical analysis. After each measurement in every site, the filter of the instrument is taken off. Then these filters are subjected to subsequent chemical analysis. Three filters have been analyzed; for measurements in the center and in the main exit road of the city, as well as in the inner part of the city. Filters are conserved in plastic sachets. Then the samples are dried at $100\text{-}110^OC$ temperature and they are transported in chemical glasses. The digestion of dust samples is done by using aqua regna (ratio 3:1 HCl and HNO_3) with analytical grade reagents. Samples were heated to boil for about 30 min and were filtered by using Sinta Glass filter and were marked to 25 ml volumetric flask. Calibration standards were done by using the matrix of digestion samples. In the last step, samples were analyzed for metals content by spectrophotometer NovaA400, Analytik Jena.

3. Monitoring results

About 70,000 data of PM concentrations have been revealed from our measurement campaign. PM1, PM2.5 and PM10 concentrations in fair weather conditions in the urban centre of Shkodra and in rural area of Zogaj are summarized in the tables 3-6. In these tables are given principal statistical parameters of PM distribution. PM concentration values in tables 2-5 are expressed in $\mu g/m^3$.
After concluding with PM concentrations analysis, we are focused on the estimation of metals in the samples taken from the measurement process in the urban cent (Lithgow et al., 2004; Bekteshi et al., 2010). Three samples for analysis have been taken; site 1, site 2 and site 3. The results obtained from the estimation of metal presence in analyzed samples are given in table 7. An overall estimation of metals in PM of Shkodra city shows that their average concentrations are in low levels. Greater concentrations are encountered for calcium, magnesium and lead.
From the table 7 it can be seen that most "problematic" metals on PM are calcium, magnesium and lead.

4. Discussion

The principal PM sources in the urban centre of Shkodra city are traffic and residential wood combustion. The second PM source becomes more evident during November measurements, when ambience temperatures fall, and heating processes begins through populated areas. Residential wood combustions are spread almost uniformly across all the city area. But these activities play an important role especially in the inner part of the city, far from the main roads. Since Shkodra city has only a modest industrial activity, it cannot be considered as an industrial centre. Shkodra city, being simply an urban centre, the main contributors on PM concentrations would be traffic and residential wood combustion. Traffic is uniformly distributed over all monitoring campaign, while residential wood combustion was evidenced mostly on colder days of November measurements. This is the main reason that why we have conducted measurements in road-sides and in the inner part of the city (Krecl et al., 2008).
Traffic activity has two major effects; mechanical particle re suspension (mainly in coarse mode) and particle emission form vehicle exhausts (mainly in fine mode). Form the comparisons between values of table 3 and 4 we can see that average values of PM1 and PM2.5 are almost equal in the center and exit road of the city, with respectively only 3.0% and 3.3% higher values in the center. PM10 values differ more evidently, getting 8.8% higher values on the exit road. Higher values of PM10 in the exit road compared with the center of the city can be justified by the worse conditions of main exit road which enables the re-suspension of coarse mode particles.
"Surprising" results are taken from the measurements in the inner part of the city. In these measurements PM1 and PM2.5 modes were respectively 51.4% and 21.1% higher than in the center of the city. While PM10 mode has about 42.3% lower values than in the center of the city. This happens because of greater efficiency on gravitational sedimentation and low efficiency transport of PM10 mode particles (Chang et al., 2010). Meanwhile PM2.5 and especially PM1 mode have greater transport efficiency, because of their longer life times, and so can be moved from main roads where traffic is more effective to the inner part of the city. Another factor except traffic, which is also the

most important, is the contribution of residential wood combustion especially on fine mode of particle concentrations. These aerosol sources were quite near the measurement point in site 3.

Effectiveness of aerosol sources is also influenced by meteorological conditions at the measuring site. Precipitation is the most important meteorological factor which influences PM concentrations. In table 2 we have presented the number of days with and without precipitation during monitoring campaign. During precipitations on rainy days there was evidenced a clear reduction of PM concentrations at all measuring sites. Average PM Concentrations during fair-weather and rainy days in the site 3 are presented in table 8.

Values in brackets in the table 8 are percentage reductions of PM concentrations during days with precipitations. Based on the reduction values of table 8 it can be affirmed that both PM modes are reduced during precipitations. The percentage reductions of each PM mode were quite equal, with only small differences between them.

Wind is another important meteorological factor which ha a great influence on the PM concentrations. In the figure 3 there is presented wind rose during monitoring campaign and wind class distribution.

Let us analyze the contributions of different aerosol sources in the measuring sites taking into account wind influence. Wind rose suggests that moderate wind come usually from the east. All the three measuring sites in the urban centre are located in the west side of the main axis of the Shkodra city. In this road passes the major part of the traffic in the city. This fact makes this road a great aerosol source in this area. The distances of measuring sites in urban centre from this road are respectively: d_{site1} = 20 m, d_{site2} = 10 m and d_{site3} = 55 m. In general wind influence on PM dispersion, decreasing so PM concentrations. But because the short distances from the main road and relatively high win frequency coming from the east, we can affirm that in the days with wind turbulences, the decreasing of PM concentrations is compensated somewhat by aerosols coming from the traffic sources.

PM concentrations in rural areas near the urban centre were lower than in two sites of urban centre, for all PM modes. PM1 concentration was 13.7%, PM2.5 26.5% and PM10 61.7% lower than the average PM values in the urban centre of Shkodra. This situation is in a good accordance with theoretical expectations. This rural area is characterized by very low traffic rate and low PM emissions for both modes. But transport mechanism from urban centre of Shkodra, contributes mainly on fine particles; PM2.5 and especially PM1 mode. This is why PM2.5 and especially PM1 mode in rural area have relatively smaller differences from these modes in urban centre, compared with the concentrations of PM10 mode. Transport effect from urban centre of Shkodra is facilitated by the fact that Zogaj area and Shkodra city are separated only by Lake of Shkodra, and there are not any natural obstacles between them. Their distance (about 9.5 km) is relatively short for transport mechanisms of fine mode particles. Not only the absence of natural obstacles, but also the air masses coming pronominally from the east (Zogaj lay in the west side of Shkodra city) influence in the high level of PM concentrations in this rural area.

Based on environmental viewpoint we have compared averaged PM concentrations in the city of Shkodra and rural area of Zogaj with international recommendations for PM2.5 and PM10 concentrations. These comparisons are presented by the table 9.

Values in the first column of the table 9 are averaged concentrations of the road-side measurements; in the center and in the main exit road of the city of Shkodra. Values of second column of this table are averaged values of PM concentration in the rural area of Zogaj. From this table can clearly be seen that PM concentrations in Shkodra urban centre exceed many international recommendation, while PM concentrations in the rural area of Zogaj are almost in the upper limits of these recommendations. This result has a special environmental significance because Zogaj is one of the most frequented tourist locations in the Shkodra Lake.

It can be possible also to separate different contributors on PM concentrations, in order to have a clear picture of their sources in the area of monitoring (Querol et al., 2004). The values of regional background can be estimated by minimal PM values obtained over all monitoring area. The values of city background (local background) can be estimated by minimal PM values in the three sites of city measurements (sites 1, 2 3). On the other hand, average values of local sources, in our case traffic and residential wood combustion, can be estimated by the differences between averaged and minimal values respectively in the road-side measurements and the measurements in the inner part of the city. In the table 10, are presented PM concentrations of local sources, local background and regional background.

Values in the table 10 are obtained from averaging diurnal variations of PM concentrations in all measurement locations in fair weather conditions. So, local PM sources are obtained from differences between maximal and minimal values of these diurnal variations. In the figure 4-6 are presented the percentile contributions of these sources on PM concentrations given in table 8.

From the fig.6 it can be noticed that traffic contributes more in PM10 mode (30%). This is due to re-suspension effect of the traffic, which influences more on the coarse mode. Residential wood combustion (domestic activities) has almost equal contribution for all modes (35-42%), but for fine modes this contribution is about 33% higher than in PM10 mode. Residential wood combustion like heating or cooking contributes mostly on fine particle mode. These measurement results make residential wood combustion the most effective source on PM emission in the city of Shkodra during the cold season, while traffic effects have almost the same contribution all over the year.

Remote sensing by satellite measurements of PM10 suggest that in our region PM10 concentrations lay is about 1 0$\mu g/m^3$ (Weijers et al.,1998). This regional PM concentration is dominated by Saharan dust fluxes (Langmann et al., 2008). In the southern European region the atmospheric aerosol has an important contribution from natural dust due local emissions and to the influence of African dust intrusions (Contini et al., 2010).

From the chemical viewpoint, based on the values of the table 7, it is evident that high content of calcium and magnesium in environmental dust, especially in the site 1, derives from mechanical processes in construction activities carried out in that location. Furthermore, the high concentration of lead in the samples shows high presence of lead in gasoline. This concentration is much higher in the site 1, where traffic rate is higher, and the used gasoline contains high lead concentrations. But low lead concentrations were encountered in the sites 2 and 3, despite the relatively high traffic rates, low lead presences in site 2 were related with the fact that the traffic is composed mainly by heavy-duty vehicles which usually use diesel. Even lower lead concentrations are encountered in site 3, which is a residential site far from the main roads of the city.

The air quality state can be worsened by continuous increase of the number of vehicles moving in this city. But some interventions in due time can reduce this phenomenon, or even improve the air quality in the city. Most immediate interventions can be considered the construction of new roads, re-construct many of existing roads, initiate the substitution process of old vehicles, and setting a quality standard for fuel used by vehicles in our country.

5. Conclusions

During the period of May-November continuous monitoring campaigns have been realized on PM concentrations in urban centre of Shkodra and around areas. Measurements are located in three principal locations; road-side, inner part of the city and around rural areas.

In this monitoring campaign we have developed a methodology of measurements in selected sites inside and outside urban area. This methodology permits the estimation of activation rates of different aerosol sources, and to predict the trends of PM concentrations.

Measurement results show that higher PM10 concentrations are found in the road-side measurements, because the traffic effect in these locations has a major contribution on re suspended particles. But high PM2.5 and PM1 concentrations are also encountered in road-side measurements. This is because of emission particles from the traffic. Meanwhile higher PM1 and PM2.5 values are encountered in the inner part of the city, near residential wood combustion, especially in the colder part of the year. But also PM10 concentrations were relatively high in these areas of the city. This is due to the contribution of residential wood combustion in this measurement location. So we can conclude that major contributors on PM concentrations in the Shkodra city are traffic and residential wood combustion. Traffic contributes over all the year, while the residential combustion contributes only on the cold season of the year. It is important to emphasize the fact that from the environmental viewpoint PM2.5 and PM10 exceed international recommendations on air quality in the city of Shkodra. But the situation is somewhat better in the tourist location of Zogaj, having also high levels of PM concentrations.

Even though Shkodra isn't an industrial centre, chemical analyses of samples show that the air in the city is relatively polluted by heavy metals, relatively to other cities in Albania. Metal concentrations vary in different measurement sites. Lead has a more evidenced presence in the sites characterized by heavy traffic rate, indicating the high lead concentrations in benzene. The relatively high presence of

calcium and magnesium is related with high construction activities carried out in the center of the city. The situation is somewhat better in the inner part of the city, far from the main roads.
To improve this situation it is recommended to order traffic by constructing new roads which release the traffic, re-construct existing roads, substitute old vehicles with new ones, and improve also the fuel quality. Such interventions, might mach the air qualities in the city of Shkodra according to international recommendations.

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Table 1. Number of vehicles in Shkodra city

	Diesel	Gasoline
Light duty vechicles	15,500	3,200
Heavy duty vehicles	1,800	10

Table 2. Precipitations during monitoring campaign

	Fair-weather (days)	Precipitations (days)	Precipitations (%)
Site 1	37	24	39.3
Site 2	47	15	24.2
Site 3	57	34	37.4

Table 3. PM results in the center of Shkodra (site 1)

	Minimal	*Average*	*Maximal*	*Medium*	*Mode*	*St.Deviation*
PM1	1.9	29.2	393.4	22.2	15.9	21.1
PM2.5	2.2	39.9	1796.3	33.8	22.6	30.6
PM10	2.7	124.7	8139.2	96.4	46.6	150.8

Table 4. PM results in the main exit road of Shkodra (site 2)

	Minimal	*Average*	*Maximal*	*Medium*	*Mode*	*St.Deviation*
PM1	6.9	28.3	147.5	25.4	24.1	15.7
PM2.5	8.7	38.6	266.5	33.3	29.2	23.5
PM10	13.2	135.4	3854.3	95.8	73.3	154.3

Table 5. PM results in the inner part of Shkodra (site 3)

	Minimal	*Average*	*Maximal*	*Medium*	*Mode*	*St.Deviation*
PM1	6.8	44.2	160.7	32.3	10.2	33.6
PM2.5	8.5	48.3	175.5	35.6	29.0	33.6
PM10	11.3	71.9	293.8	51.8	24.2	54.7

Table 6. PM results in the rural area of Zogaj (site 4)

	Minimal	*Average*	*Maximal*	*Medium*	*Mode*	*St.Deviation*
PM1	1.5	25.3	42.1	24.6	24.8	2.9
PM2.5	1.7	29.4	82.2	28.1	27.3	4.9
PM10	2.1	47.7	416.3	39.7	39.3	34.1

Table 7. Concentration of metals on PM in Shkodra city

	Ca (μg/m^3)	*Mg* (μg/m^3)	*Zn* (μg/m^3)	*Cu* (μg/m^3)	*Pb* (μg/m^3)
Site 1	1.90	1.67	0.36	0.19	14.96
Site 2	1.03	0.84	0.51	0.11	1.12
Site 3	0.46	0.37	0.36	0.10	0.15
Average	1.13	0.96	0.41	0.13	5.41

Table 8. Average PM concentration 'with' and 'without' precipitations

	PM 1	PM 25	PM 10
Fair-weather	42.40	46.36	69.41
Precipitations	25.98 (38.7)	29.05 (37.3)	45.40 (34.6)

Table 9. Comparisons of measured PM concentrations with international recommendations

	Shkodra	*Zogaj*	*EU*		*WHO*		*EPA*	
			24 hour mean	*Annual mean*	*24 hour mean*	*Annual mean*	*24 hour mean*	*Annual mean*
PM2.5	42.3	29.3	-	-	25	10	35	15
PM10	110.7	47.7	50	40	50	20	150	50

Table 10. Main PM contributors in the Shkodra region

	Local sources		*Local background*	*Regional background*
	Traffic	*Domestic*		
PM1	3.76	20.69	24.8	1.5
PM2.5	5.35	22.54	27.1	1.7
PM10	31.77	37.02	37.0	2.1

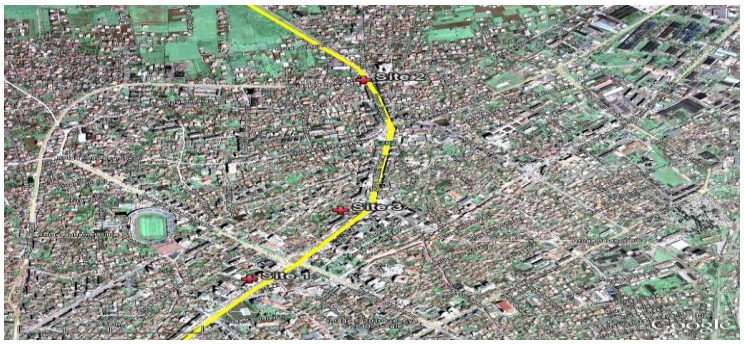

Fig.1. Measurement sites in Shkodra urban centre

Fig. 2. Positions of sites in Zogaj and in Shkodra city

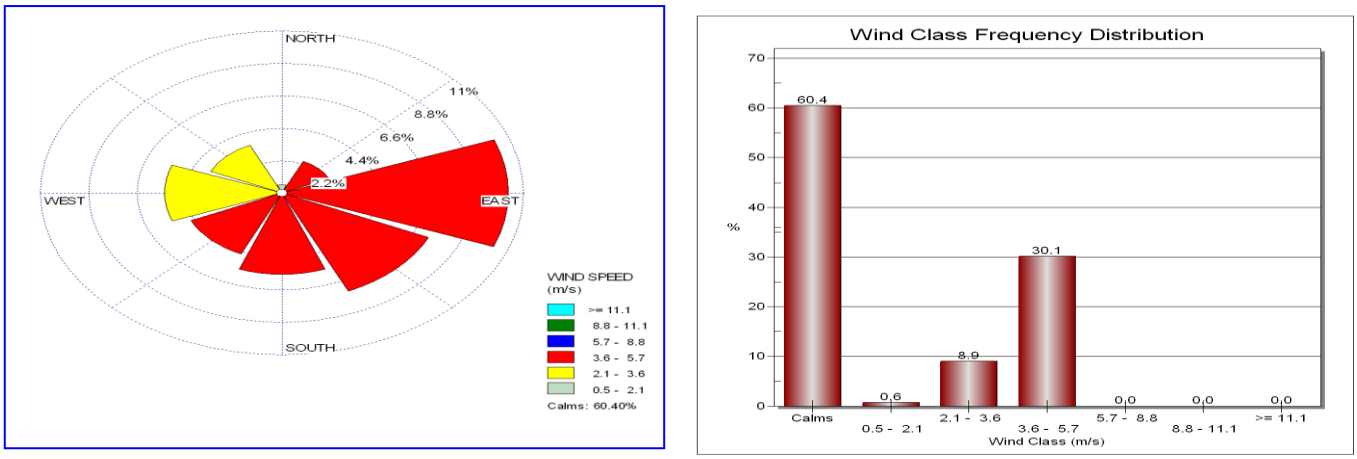

Fig. 3. Wind rose and wind class distribution

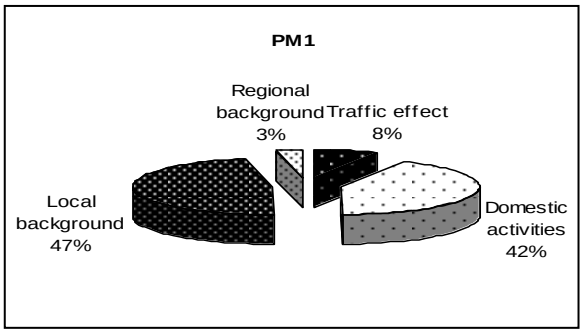

Fig. 4. Source contributions on PM1 concentration in the Shkodra region

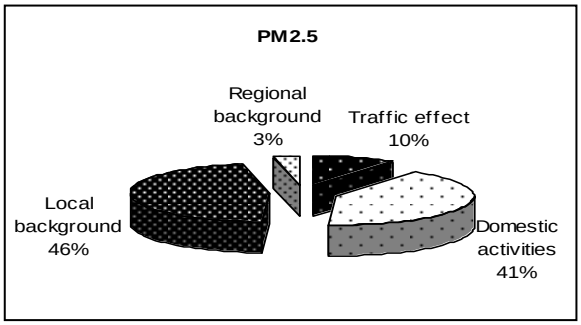

Fig. 5. Source contributions on PM2.5 concentration in the Shkodra region

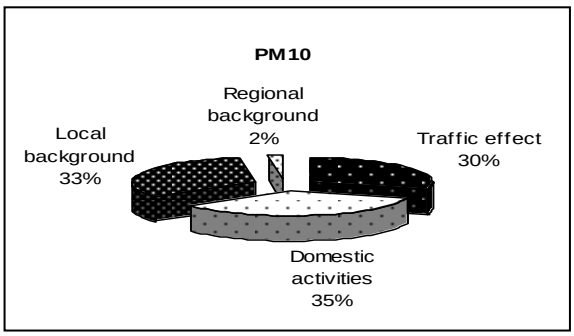

Fig. 6. Source contributions on PM10 concentration in the Shkodra region