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PARTICULATE MATTER STATUS IN ROMANIAN URBAN AREAS: PM10 POLLUTION LEVELS IN BUCHAREST

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Abstract

In the present study, the results of PM10 pollution levels assessment in the most populated town from Romania, namely Bucharest, the capital of the country are presented for a 10 years period, 2004 – 2013, respectively, with the aim of emphasizing the progress recorded after the implementation of Directive 2004/107/EC and Directive 2008/50/EC provisions. Data were collected from 8 monitoring stations located in different areas of Bucharest. PM10, Cd, Pb and Ni from PM10 averages are presented by each sampling point, and by year, during a 10 years period (2003 - 2014). The raw data were available from the Romanian National Agency of Environmental Protection. IBM SPSS software, v. 6.0 was used for data processing. Even if some progress was reported for air quality in Bucharest (mainly in heavy traffic areas) from the point of view of PM10 pollution subsequently to directives implementation, important issues have to be solved, taken into account that industrial areas are still important PM10 sources. In this respect, further research will be opportune for identifying the potential contribution of PM10 background pollution in concerned areas.

Keywords: cadmium, dust, lead, monitoring, nickel

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1. Introduction

Air quality from urban environments is a critical component in ensuring a healthy environment. There are lots of studies conducted, in concerned areal, demonstrating the importance of NO_x, CO, SO₂, VOC, PM emissions (but also of O₃ concentration), resulting from traffic and industrial processes (Carslaw and Rhys-Tyler, 2013).

These, although have potential natural origin (volcanic emissions, soil erosion, forest fires, dust storms etc.), represent an important challenge in urban communities, and rural areas (Anderl, 2009; Querol et al., 2001), due to anthropic sources. In urban areas, there are a number of sources generating

consistent particulate matter (PM) quantities, such as urban traffic, industrial processes, fossil fuel combustion (Malina and Fischer, 2012; Stefan et al., 2013). The airborne particles are involved in complex atmospheric mechanisms that in most cases result in undesirable effects (Querol et al., 2004; Turšič et al., 2008), especially on human health, contributing to worsening cardiovascular and respiratory diseases (Pope et al., 2002; Zhou et al., 2011). Their formation is a phenomenon involving a series of intake factors with specific conditionality (Gehrig et al., 2004; Luca and Ioan, 2012).

The airborne particles structure is complex, including organic and inorganic elements. Numerous studies, conducted worldwide during the past two

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decades, reveals their structure in urban areas (Alberta Government, 2013; Lopez et al., 2005; Marković et al., 2008; Terzi et al., 2010; Thomaidis, 2003), and the degree of pollution produced by particulate matter (Clarke et al., 1999; Grivas et al., 2004; Marcazzan et al., 2001; Massagué and Puig, 2001; Puxbaum et al., 2004; Stefan et al., 2013; Wang et al., 2002).

The PM increasing levels in urban areas, especially after 2000, constituted an urgent requirement for the competent EU bodies, to establish limit values for both their concentration in the ambient air, and their components (Malina and Fischer, 2012). These concentration limits are set by EC Directive (2004) and EC Directive (2008).

Romania, after joining the European Union, has committed to implement the normative acts in force at Community level, and made important progresses (Koch, 2009; Proorocu et al., 2009). However, Romania failed in accomplishing the deadline for transposing the two Directives and was in a position to provide the European Commission reasoned opinion for delays (Bota et al., 2011).

Now, a legislative progress is recorded by promulgation of the Law 104 (2011) concerning the quality of environmental air quality, but at the implementation level, the problems are not fully resolved - not all trace elements are monitored, there are deficiencies in the functioning and coordination monitoring activities.

Even studies were performed in this respect, in Bucharest and at national level (Gavrilescu, 2007; Lăzăroiu, 2006; Moldoveanu, 2005), further research is need in order to establish the PM10 pollution levels, in the most crowded Romanian area, such as Bucharest City. The objective of this study was to emphasize the legislative improvement of air quality management concerning PM10 pollution (particulate matter with $\leq 10\mu\text{m}$ in diameter) during 2012 – 2013, in Bucharest, the most crowded city in Romania, compared to the previous period 2004 – 2011.

2. Materials and method

2.1. Study area and sampling area description

Bucharest is the largest urban agglomeration in Romania, with 1,926,334 inhabitants, according to the last census conducted in 2012, with a density of 8510 inhabitants/m². It is located in the Romanian Plain, with the following geographical coordinates: latitude: 25°49'50" - 26°27'15" East; longitude 44°44'30" - 44°14'05" North. It is located at a maximum altitude of 94.63 m and it is crossed by two rivers, Dâmbovița and Colentina. The two valleys formed around rivers divide the city into several areas.

The climate is temperate-continental, influenced by the characteristics of the contact area of the eastern continental masses with western and southern masses. The prevailing eastern air masses transmit excessive climate traits, with hot summers

and often harsh winters. During the analyzed period 2004 – 2013, the average annual temperatures, precipitation and relative humidity values were close to the average annual limits. The wind regimen reported for the studied period (Fig. 1), had an average value similar to the multiannual averages, close to the North-East and East predominance.

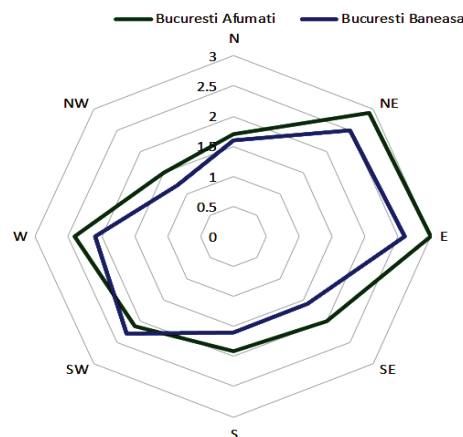


Fig. 1. The average wind regime during 2004–2013 in Bucharest (km/h)

In the entire area, the main sources of PM pollution are: traffic (responsible for about 70% of air pollution), which generates dust fine particles by impact with roads, thermal power plants by specific emissions, industry and construction, by fine dust particles resulted from friction of particles and sped in air, and or handling of materials, which produce dust (Amato et al., 2009).

In Bucharest, there are defined three monitoring areas, with 8 monitoring stations:

1. Central area - bounded by: Calea Văcărești, Sos. Mihai Bravu, Sos. Ștefan cel Mare, Bd. Titulescu, Bd. Vasile Milea, Str. Progresului, Sos. Olteniței (internal belt) - 2 traffic stations, Cercul Militar and Mihai Bravu;

2. Peripheral area - bounded by the inner and outer belt - 1 urban background station (Lacul Morii) and 3 industrial stations (Berceni, Drumul Taberei and Titan);

3. Exterior area - beyond the outer belt - 1 regional background station (Balotești) and 1 suburban station (Măgurele).

The monitoring network of the National Agency of Environmental Monitoring consists of 8 fixed monitoring points (Fig. 2, Table 1). PM10 were sampled by all 8 monitoring stations with Derenda LVS 31./PNS 2.1-15 equipment, and PM10 mass concentration ($\mu\text{g}/\text{m}^3$) was quantified gravimetrically, 0.1 mg precision, 4-5 m pelevation point height and 1.8 m length of the prelevation line, according to standardized method SR EN 12341 (2002). PM10 mass concentration ($\mu\text{g}/\text{m}^3$) averages by year during a 10 years period (2003 - 2014) are presented by each prelevation point. Cd, Pb and Ni from PM10 mass concentration ($\mu\text{g}/\text{m}^3$) were also quantified, using ICP-MS equipment. Cd and Pb raw data are available for all analyzed period, but Ni only for the interval

2012-2013. All data are available from the Romanian National Agency of Environmental Protection.

2.2. Statistics

Basic statistics was used for quantifying averages and dispersion parameters. The significance of differences was calculated using Analyze of Variance (ANOVA) test. It is applied in order to compare the PM₁₀ pollution levels in monitored areas during two distinct periods, 2004-2010, and

2011-2012, respectively, before and after implementation of EC Directive (2004) and EC Directive (2008), by Law 104 (2011) concerning environmental air quality.

The Principal Component Analysis (PCA) was applied in order to highlight areas with greatest contribution to total PM₁₀ pollution in Bucharest. According to PCA principles, the most important two factors, Factor 1 and Factor 2, are mentioned, because they emphasize the elements, which bring highest share from the total pollution.



Fig. 2. The network of the air quality monitoring in Bucharest (www.google.ro)

Table 1. The location and characteristics of 8 monitoring points fixed in Bucharest

No.	Station	Code	Characterization
1	Cercul militar	B001	Urban area characterized by residential and commercial geographic coordinate 44°25'44" N latitude, 26°7'15" E longitude, and 75 m altitude, being representative for 10 - 100 m. The main emission source is traffic.
2	Mihai Bravu	B002	Urban residential area characterized by geographic coordinate 44°26'26" N latitude, 26°9'4" E longitude and altitude 75 m, being representative for 10 - 100 m. The main emission source is traffic.
3	Lacul Morii	B007	Peripheral urban residential area characterized by geographic coordinate 44°26'33" N latitude, 26°3'36" E longitude, and 75 m altitude, being representative for 1 - 5 km. It is characterized by a moderate volume of traffic.
4	Berceni	B008	Urban industrial peripheral area characterized by geographic coordinate 44°24'40" N latitude, 26°11'5" E longitude and 75 m altitude, being representative for 100 m - 1 km. It is characterized by a moderate volume of traffic.
5	Drumul Taberei	B004	Urban industrial peripheral area characterized by geographic coordinate 44°24'42" N latitude, longitude 26°3'8" E and 75 m altitude, being representative for 100 m - 1 km. It is characterized by a high volume of traffic.
6	Titan	B003	Urban industrial peripheral area characterized by geographic coordinate 44°24'40" N latitude, 26°11'5" E longitude and 75 m altitude, being representative for 100 m - 1 km. It is characterized by a low volume of traffic.
7	Măgurele	B006	Suburban residential area characterized by geographic coordinate 44°20'56" N latitude, 26°2'1" E longitude and 75 m altitude, being representative for 25 km - 150 km. It is characterized by a moderate volume of traffic.
8	Balotești	B005	Regional rural area, whose geographical coordinates cannot be mentioned, because in the area is located a military unit. It is representative for 200 m - 500 km, and has low volume of traffic.

Correlations were established between of Cd, Pb, Ni concentrations and total content of PM₁₀, using Pearson correlation, due to linear variation between parameters. IBM SPSS software, v. 6.0. was used.

3. Results and discussion

Table 2 emphasizes the PM₁₀ averages by 10 years in Bucharest minimum, and maximum values of both intervals analyzed. The reason of this presentation of Directives 2004/107/EC and 2008/50/EC implementation, exceeding of the annual limit value for the protection of human health (40 $\mu\text{g}/\text{m}^3$) was recorded in almost all monitoring points. The single exception was Balotesti (32.15 $\mu\text{g}/\text{m}^3$), rural area with low traffic, characterized by the largest variability of PM₁₀ concentration (77.35%).

During the two years period subsequent to air directives implementation, very significant decrease ($p < 0.001$) of PM₁₀ concentration was reported in urban traffic stations Cercul Militar and Mihai Bravu. It proves that the problem of PM₁₀ air pollution produced by traffic in areas of maximum agglomeration in Bucharest was well managed.

For the industrial area Drumul Taberei, it is reported even higher PM₁₀ average concentration compared to previous years (50.36 $\mu\text{g}/\text{m}^3$ during 2012-2013 compared to 47.81 $\mu\text{g}/\text{m}^3$ in 2004 - 2011).

This is the result of several inputs: enhanced industrial PM₁₀ polluting activities, increased traffic,

wind direction (Fig. 1), which favors particles resuspension, insufficient and/or inadequate measures for air quality protection.

Exceeding of the limit values were recorded at Magurele (41.05 $\mu\text{g}/\text{m}^3$, compared to previously recorded 41.89 $\mu\text{g}/\text{m}^3$, respectively) and Lacul Morii (44.31 $\mu\text{g}/\text{m}^3$ compared to previously recorded 43.19 $\mu\text{g}/\text{m}^3$). The study of PM₁₀ annual averages in monitoring points (Table 1) highlights important exceeding of annual limit value for human health protection, in areas of heavy traffic during the first analyzed time interval (2004 – 2011) and in suburban industrial areas, in the second (2012-2013).

Pb, Cd and Ni share in PM₁₀ (Fig. 3 and 4) differs depending on monitoring points. Thus, during 2004 - 2011 the highest proportion of Cd is recorded in Berceni (18%) and Mihai Bravu (14%), while the highest proportion of Pb in Titan (23%) and Mihai Bravu (14%). These areas are characterized by heavy traffic and industrial activities. During 2012-2013 Cd has the highest concentration in industrial type area Drumul Taberei (22%), and Lacul Morii (15%) and their proportion increased versus the previous time interval (2004-2011). The highest Pb share was reported in Berceni (18%) and Drumul Taberei (16%), while Ni (18%) in Magurele suburban point and Berceni industrial monitoring point and (16%). This repartition may be explained by high share of metallic salts in particulate matters from urban areas (Balachandran et al., 2000; Celo and Dabek-Zlotorzynska, 2010; Chalbot et al., 2013).

Table 2. Basic statistics for PM₁₀ ($\mu\text{g}/\text{m}^3$) concentrations in 8 monitoring stations (Bucharest)

Issue	n	Mean $\mu\text{g}/\text{m}^3$	Min $\mu\text{g}/\text{m}^3$	Max $\mu\text{g}/\text{m}^3$	Std.Dev.	Std.Er.	Coef.Var.	p (ANOVA)
2004 - 2011								
Cercul Militar	1586	49.900	3.000	361.000	26.452	0.664	53.010	
Mihai Bravu	1586	55.316	2.000	220.000	27.559	0.692	49.822	
Titan	1586	45.195	1.000	226.000	25.478	0.640	56.374	
Drumul Taberei	1586	47.810	4.000	267.000	25.278	0.635	52.873	
Balotesti	1586	32.153	0.001	170.000	24.769	0.622	77.035	
Magurele	1586	41.896	0.000	296.000	23.307	0.585	55.630	
Lacul Morii	1586	43.199	1.000	199.000	23.754	0.596	54.989	
Berceni	1586	42.487	0.001	198.000	23.489	0.590	55.284	
2012 - 2013								
Cercul Militar	146	37.055	17.000	134.000	20.191	1.671	54.489	***
Mihai Bravu	146	35.404	6.000	107.000	17.233	1.426	48.675	***
Titan	146	39.596	1.000	198.000	26.277	2.175	66.362	**
Drumul Taberei	146	50.363	4.000	163.000	29.145	2.412	57.870	0.249
Balotesti	146	35.596	4.000	110.000	24.495	2.027	68.814	0.107
Magurele	146	41.055	6.000	117.000	24.109	1.995	58.723	0.677
Lacul Morii	146	44.315	8.000	143.000	27.146	2.247	61.257	0.591
Berceni	146	37.021	4.000	140.000	21.742	1.799	58.728	**

** - $p < 0.01$; *** - $p < 0.001$

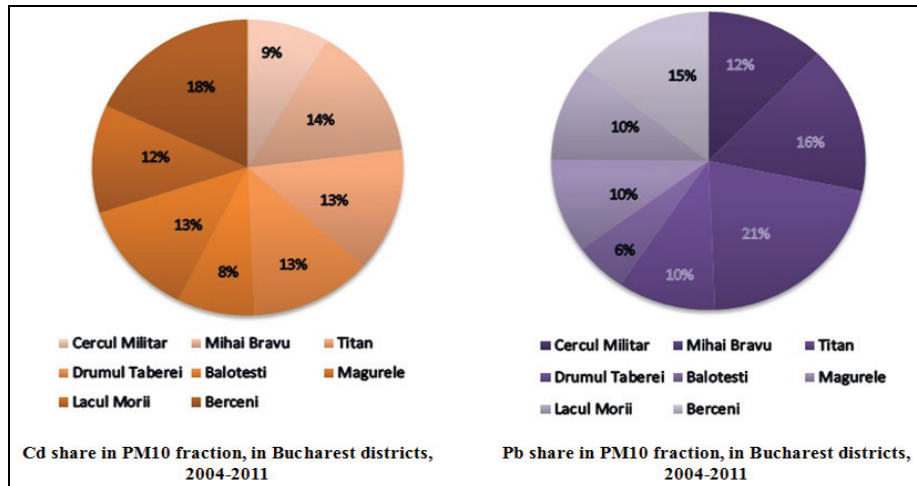


Fig. 3. The share of PM₁₀, Cd, and Pb distribution in Bucharest monitoring points, 2004-2011

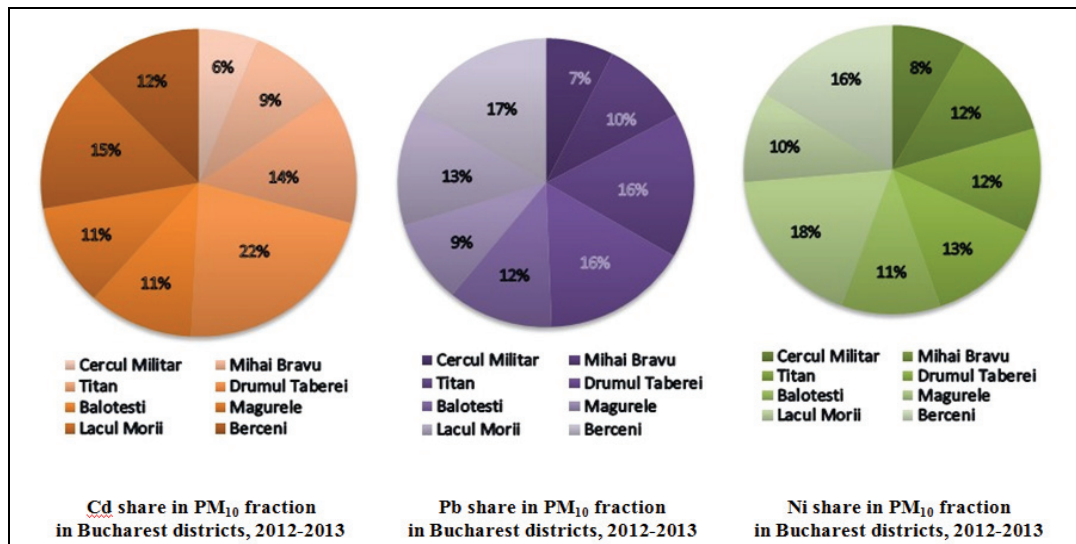


Fig. 4. The share of PM₁₀, Cd, Pb and Ni distribution in Bucharest monitoring points, 2012 – 2013

Nickel was quantified only during second studied time interval (2012 - 2013). It is of interest for industrial areas with heavy traffic, due to its pollutant potential when concentration exceeds target limit. It results mainly from combustion of fossil fuels (Cempel and Nikel, 2006).

Regarding the proportion of trace elements in areas characterized by heavy traffic, Cercul Militar and Mihai Bravu, respectively, they decrease as follows: Cd < Pb < Ni, contrary to the mentions of the European Environmental Agency (EEA, 2007), according to which traffic trace metals emissions decrease as Pb < Ni < Cd. On the other hand, studies conducted in the USA on PM composition in areas bordering highways (Xia and Gao, 2011), show the same results as our study. The highest nickel share was reported in both industrial and heavy traffic areas from Bucharest (Fig. 4).

If during 2004 - 2011 an exceeding of the PM₁₀ annual limit value was recorded (44.74 µg/m³), in the subsequent interval of directives implementation (years 2012 and 2013), the value

decreased, almost reaching the allowed concentration (40.05 µg/m³), but the difference is not statistically significant at a significance threshold of 0.05% (Fig. 5a). Statistically, very significant differences are reported for Cd and Pb between both time intervals (Fig. 5b and 5c). Concerning Cd, Pb and Ni levels, we noticed that no exceeding of the target values (Law 104, 2011). For Cd, a multiannual average of 4.338 ng/m³ was reported for 2004 – 2011 time interval and 0.810 ng/m³ for 2012 – 2013, both much lower compared to annual target value of 5 µg/m³ (Fig. 5b). For lead, a multiannual average of 0.0933 µg/m³ was reported for 2009 – 2011 time interval and 0.0148 µg/m³ for 2012 – 2013, lower than annual target value of 0.5 µg/m³ (Fig. 5c). Nickel, quantified only during second time interval framed in admitted limit for annual target value (20 ng/m³), 6.65 ng/m³ ± 0.61 ng/m³, respectively (Fig. 5d).

The correlation matrix between PM₁₀ components identified in this study, Pb, Cd, and Ni, respectively, and between these metals, was obtained using Person algorithm, as result of testing the

linearity of dependence between the correlated components. During 2004-2011, average correlations were reported between Cd and Pb concentrations (Table 3), and during 2012-2013, strong correlations emphasized by the values of the Pearson coefficients of correlation, r . Thus, during first studied period, between total PM₁₀ and Pb a positive average correlation was reported ($r = 0.567$), and between PM₁₀ and Cd, a positive weak correlation ($r = 0.295$). During the second studied period, 2012-2013, very strong positive correlation between PM₁₀ and Pb ($r = 0.877$) was reported, and positive moderate PM₁₀ with Cd ($r = 0.430$).

These results demonstrate the high contribution of lead in PM₁₀ formation (Table 3), also emphasizing the enhanced contributions of industrial sources to PM₁₀ air pollution. Principal Component Analysis (PCA) was implemented for emphasizing the areas representing source for the most important impact upon PM₁₀ pollution in Bucharest, during both considered periods, 2004 –

2011 previously to EC Directives (2004) and EC Directive (2008) implementation, and subsequent, 2012–2013, respectively (Fig. 6).

Figures 6a and 6b emphasizes the distributions of most important areas contributing to PM₁₀ pollution in Bucharest, by both periods analyzed, before and after EU Directives transposition by Romanian Law 104 (2011). For the period 2004–2011, 2 factors may be taken into consideration, and they explain 56.23% of variance (Fig. 6a).

The most important contributions are brought by Factor 1 is responsible of 44.47% of variance and emphasizes the most important contribution of industrial monitoring area Drumul Taberei to PM₁₀ pollution (coefficient > 0.7), Magurele and Miha Bravu monitoring areas. Factor 2, is responsible for 11.60% of variance and it is mainly represented by the industrial monitoring area Titan and traffic monitoring area Cercul militar (coefficient > 0.7).

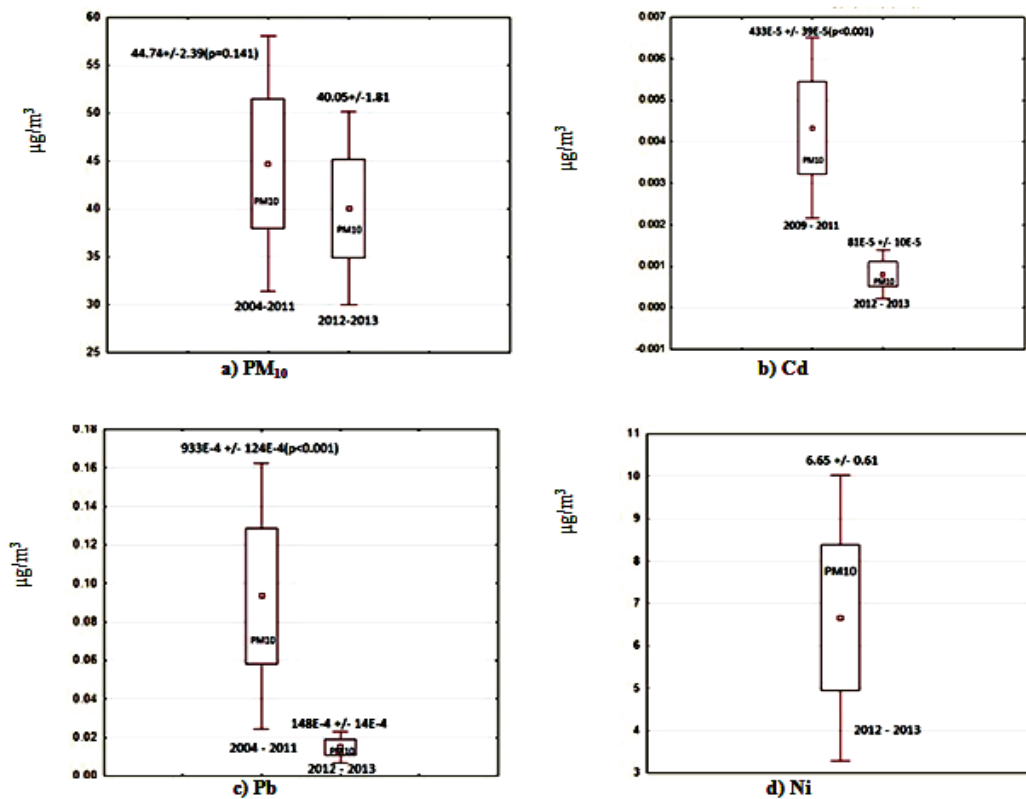
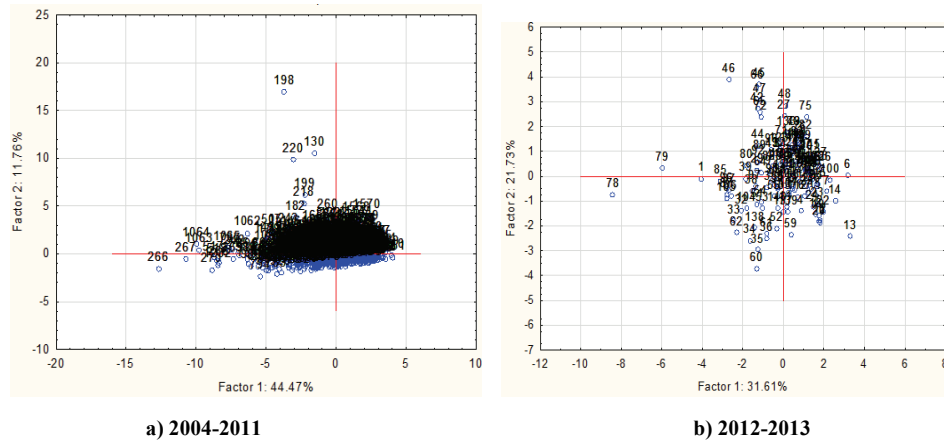


Fig 5. The Box-Plot diagrams for PM₁₀, Cd, Pb, and Ni by entire monitored area of Bucharest, during both studied time periods 2004 – 2011, and 2012 – 2013, respectively

Table 3. The matrix of correlations between PM₁₀ ($\mu\text{g}/\text{m}^3$), Pb ($\mu\text{g}/\text{m}^3$), Cd (ng/m^3), and Ni (ng/m^3) concentrations in Bucharest area, during 2004-2013

	<i>Period</i>	<i>PM₁₀</i>	<i>Pb</i>	<i>Cd</i>	<i>Ni</i>
PM ₁₀		-	0.567**	0.295 ^{ns}	-
Pb	2004-2011	-	-	0.563**	-
Cd		-	-	-	-
PM ₁₀		-	0.877**	0.430 ^{ns}	0.074 ^{ns}
Pb	2012-213	-	-	0.769***	0.146 ^{ns}
Cd		-	-	-	0.143 ^{ns}
Ni		-	-	-	-

ns – not significant; ** - $p < 0.01$; *** - $p < 0.001$.



a) 2004-2011

b) 2012-2013

Area	Factor 1	Factor 2
	% variance = 44.47%	% variance = 11.76%
Cercul Militar	-0.200	0.811
Mihai Bravu	0.415	-0.194
Titan	0.884	0.841
Drumul Taberei	0.963	-0.065
Balotesti	-0.058	0.578
Magurele	0.532	0.301
Lacul Morii	0.344	0.361
Berceni	0.219	0.119

Area	Factor 1	Factor 2
	% variance = 31.61%	% variance = 21.73%
Cercul Militar	-0.262	0.362
Mihai Bravu	0.213	-0.312
Titan	0.820	0.116
Drumul Taberei	0.915	-0.045
Balotesti	-0.049	0.215
Magurele	0.645	0.039
Lacul Morii	0.698	0.115
Berceni	0.354	0.749

Fig. 6. Principal Component Analysis for PM10 distribution in monitoring areas from Bucharest, for two studied temporal intervals

During second monitored period, 2012 – 2013, respectively, two factors are also noticed.

They are responsible for 53.34% of variance (Fig. 6b). 31.61% of variance is explained by Factor 1, with the components Titan and Drumul Taberei industrial monitoring areas (coefficient > 0.7) and at lowest extent Magurele and Lacul Morii. 21.73% of variance is explained by Factor 2 by Berceni industrial monitoring area (coefficient > 0.7). The PCA shows that in both experimental time intervals, the main contribution to PM10 air pollution in Bucharest is brought by industrial areas, represented by Drumul Taberei and Titan.

4. Conclusions

The level of PM10 pollution in Bucharest during 2004–2011, previous to Directives 2004/107/EC and 2008/50/EC implementation, except rural monitoring area Balotesti ($32.15 \mu\text{g}/\text{m}^3$), exceeded the annual limit for human health protection ($40 \mu\text{g}/\text{m}^3$), but trace elements (Cd, Pb and Ni) framed within target values.

Subsequently to directives implementation, the time interval 2012–2013, respectively, is characterized by noticeable better air quality in Bucharest, from point of view of PM10 pollution, mainly in heavy traffic areas. Even though, the present study demonstrates that air quality in Bucharest is not yet satisfactory, because the industrial areas (Drumul Taberei and Titan) still represent an important issue to be solved. Further research is needed in order to identify the possible

contribution of PM10 background pollution in concerned areas.

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