Particulate Matter and Polycyclic Aromatic Hydrocarbon Air Pollution in Areas of Bucharest with Heavy Road Traffic

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This study presents the results from two tests performed in one of Bucharest's main crossroads during the summer and winter of 2010. These tests monitored the air pollution based on PM 2.5 and PM 10 particulate matter as well as the total suspended particles (TSP) and polycyclic aromatic hydrocarbons (PAH). The PM2.5 concentration varied between 21.9 and 56.6 μg/m³, the higher concentrations being found during the winter. The PM2.5 concentration contributed 92% of the PM10 concentration in the winter and 79% in the summer. The relatively high molecular weight PAHs (4–6 rings), which are typical of traffic emissions, were found in all three particulate matter fractions. The most abundant PAH, benzo[k]fluoranthene, was found in PM2.5 between 1.96 - 22.5 ng/m³. Based on the results, we showed that the air pollution level with particulate matter PM 2.5 and PAH detected in the ruttier traffic conditions is frequently above the legal limit having a potential harmful effect on the population.

Keywords: ambient air, PAHs, PM2.5, PM10, Pearson correlation

Air pollution in urban areas is an important issue because of the effects it can have on the health of the local population. Particular attention has been paid to particulate matter pollution, especially to particles with diameters less than 10 μm, PM10 [1-4], and to particles with diameters less than 2.5 μm, PM2.5 [5-10] because their small sizes allow them to accumulate in the respiratory system, causing adverse health effects, such as asthma [11], lung cancer [12-14], and cardiovascular diseases [8].

Particulate matter air pollution can be caused by natural phenomena, human activities, and interactions between atmospheric pollutants [15-17]. The human activities that cause the most particulate matter pollution in the atmosphere include fossil fuel combustion (predominantly in industrial and energy-production plants) and road traffic [9,15-17]. Road traffic is considered to be one of the main sources of particulate matter and polycyclic aromatic hydrocarbon pollution in urban areas [18-20].

Airborne particulate matter emitted from road traffic is generated close to the ground, and toxic chemicals can be adsorbed to the particulate matter surface, enhancing the adverse effects that can be caused in the local population [2,4,21]. Exposure to PAHs, which are produced in combustion processes, is of particular concern because these compounds have strong carcinogenic and mutagenic effects [13].

It is necessary for action to be taken to decrease the particulate matter concentrations in urban air. The first steps to achieve this in any area are to measure the pollution concentrations and then to impose legal limit values, with the aim of limiting environmental and human health effects. The number of people with respiratory problems is increasing, especially in urban areas, and there is a tendency to decrease the legal limit values to reduce the risk of healthy people becoming ill. For example, the US Environmental Protection Agency National Ambient Air Quality Standard [22] daily PM2.5 limit was reduced from 65 μg/m³ to 35 μg/m³, and the European Union, through Directive 2008/50/EC [23] has set an annual PM2.5 limit value of 25 μg/m³ to be implemented by 2015, and a limit of 20 μg/m³ to be implemented by 2020.

The airborne particulate and PAH pollution in Bucharest, Romania is less understood, and measurements need to be made to provide the information needed to assess potential pollution control measures. In this study we aimed to assess the pollution level with total suspended particles (TSP), PM10, PM2.5 and PAH of ambient air near Razoare crossroads, the busiest intersections in Bucharest, by direct measurements made in two sampling campaigns in the winter (cold season) and summer (hot season) of 2010. We also studied the mass ratios of different particulate size fractions and the influence of traffic and meteorological conditions, especially precipitations, may have on the concentration of particulate matter in the air. To our knowledge, this was the first study of this kind conducted in Bucharest.

Experiemntal part
Materials and methods
Field Sampling

Bucharest (44°26'7"N; 26°6'10"E) is the capital, and the largest and most crowded city in Romania. Industrial activity within Bucharest has decreased significantly in recent years because most industrial enterprises have been moved to the outskirts of the city or closed. This has caused a decrease in emissions of particulate matter and other toxic air pollutants within the city. Despite this, Bucharest is still one of the highest polluted cities from Europe with particulate matter [24] and gaseous pollutant associated traffic emissions, NO_2, SO_2 and CO [25].

The concentrations of particulate matter and PAHs adsorbed to particulate matter were measured close to the Razoare crossroads, which is one of the busiest crossroads in Bucharest (fig. 1) that gathers and directs vehicles from three main neighborhoods to downtown.

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Fig. 1. The sampling area, Razoare crossroad, Bucharest

In Razoare's residential area there are not industrial activities which may generate particles emission or PAH; the soil characteristics from Bucharest, loosened clay without strong adhesive properties [26], is a possible source for particle emissions due to erosion process especially during the drought when the wind generates important amounts of dust in the air.

Two sampling campaigns were performed in 2010, in the winter (February 8th to February 17th) and the summer (July 26th to August 3rd). The air samples were analyzed following the Romanian SR EN 12341:2002 [27], SR EN 14907:2006 [28] and STAS 10813/76 [29] standards to ensure that representative samples were collected and that the particulate matter and PAH concentrations were determined properly.

For sampling was used 6 instrument (Sven Leckel Ingenieurbüro GmbH, Berlin, Germany) fitted with impactors for separating the size fractions of PM2.5, PM10, and TSP; samples were collected during 24 hwith a flow rate of 2.3m³/h. The collection point was 1.5m high from the ground and at 50m from a cross road without vegetation or building surroundings.

The suspended particles were collected on 47 mm quartz fibre filters from the same set/campaign; two filters from each set were tested by HPLC/FLD to check the possible interferences in PAH determination; there were no detectable data to suggest a thermic treatment of the filters before using. The samples were analyzed together with two field and laboratory blanks in order to rule out any contaminations during filter handling in the laboratory or in the field. Used and unused filters were transported in labeled Petri dishes. Half of the filters, for PAHs determination, were wrapped separately in aluminum foil and stored in a freezer at -20ºC until analysis.

Laboratory Analysis

For particulate matter determinations the quartz filters were weighed before and after exposure, using an analytical balance AG 135 (Mettler-Toledo GmbH,

Greifensee, Switzerland) with a 10 μg resolution. Before weighing the filters were equilibrated at 20°C and 50% humidity for minimum 48 h. The mass gained by the filter was taken to be the particulate mass in the air volume that was sampled.

For PAHs determination, half of the exposed filters, stored in a freezer until analysis, were extracted with 10mL toluene in an ultrasonic bath, at room temperature, for 30 min, twice, and the combined extracts were concentrated to dry in a nitrogen atmosphere, then solubilized in 1mL acetonitrile. The extracts were then refrigerated until they were analyzed. Immediately before analysis, the samples were filtered through 0.45-μm Acrodisc nylon syringe filters (Pall Corp., Port Washington, NY, USA). Field and laboratory blank filters were regularly analyzed together with a LGC Standards (Teddington, UK) US-106N PAH Mixture Certified Reference Material (16 PAHs in methylene chloride/ benzene 1:1, lot number CD-3568) as a quality assurance measure.

Chromatographic-quality water, toluen and acetonitrile were used throughout the extraction and analysis procedures. The PAHs in the particulate matter were analyzed following the ISO 16362:2005 method [30], using an Agilent 1200 high-performance liquid chromatograph with a fluorescence detector (Agilent Technologies Inc., Santa Clara, CA, USA). The LGC Standards PM-613A PAH mixture, which contains 16 PAHs between 5 and 10 μg/ mL in acetonitrile, was used to calibrate the instrument.

Separation was achieved using a Phenomenex Envirosep-PP-PAH column (125 mm long, 3.2 mm i.d.; Phenomenex Inc., Torrance, CA, USA), with a flow rate of 1mL/min. The injection volume was 10μL and the total analysis time was 40min. In table 1 there are presented the limits of detection (LD) and the limits of quantification (LQ) of the methods calculated as three times the standard deviation of 10 blank filters for LD and ten times the standard deviation for LQ, for an air volume corresponding to daily sampling at a flow rate of 2.3m³/h.

Results and discussions
Particulate Matter

The particulate matter concentrations were interpreted in relation to legal daily limit values. For example, the PM10 results were interpreted in relation to the European Union daily limit of 50 μg/m³ (Directive 2008/50/EC), but, because that directive only stipulates an annual limit for PM2.5, our PM2.5 results were interpreted in relation to the US Environmental Protection Agency daily limit of 35 μg/m³. Limits for TSP concentrations have not been established in the European Union or the USA, so the Romanian standard STAS 12574/87 [31] daily limit value of 150 μg/ m³ was used.

The particulate matter concentrations found in the air at the Razoare crossroads in the winter and summer 2010 are shown in table 2 and table 3, respectively. The mean, median, standard deviations and some information on meteorological parameters are given, along with the ratios

PAHs	LD, ng/m³	LQ, ng/m³	PAHs	LD, ng/m³	LQ, ng/m³
naphthalene	0.007	0.02	benz[a]anthracene	0.005	0.016
acenaphthen	0.004	0.015	chrysene	0.001	0.003
fluorene	0.0003	0.001	benzo[b]fluoranthene	0.005	0.015
phenanthrene	0.0003	0.001	benzo[k]fluoranthene	0.015	0.05
anthracene	0.00005	0.00015	benzo[a]pyrene	0.0035	0.01
fluoranthene	0.015	0.05	dibenz[a,h]anthracene	0.025	0.008
pyrene	0.0025	0.008	benzo[ghi]perylene	0.007	0.02

Table 1
DETECTION LIMITS (LD) AND QUANTIFICATION LIMITS (LQ) FOR PAH$_s$ METHOD

Sampling period	Temp, °C	RH,%	precipitations	PM concentrations, µg/m³			PM ratios		
				PM 2,5	PM 10	TSP	2,5/10	10/TSP	2,5/TSP
February 8–9,	-2	95	snow fall	31.5	32.3	39.6	97.5	81.6	79.5
February 10–11	1	94	snow/rain fall	21.9	23.7	30.5	92.4	77.7	71.8
February 11–12	-3	97	snow fall	34.1	43.1	48.5	79.1	88.9	70.3
February 14–15	-1	64	cloudy	41.3	40.7	42.6	101.5	95.5	96.9
February 16–17	2	59	cloudless	55.6	62.3	64.5	89.2	96.6	86.2
Mean				36.9	40.4	45.1	92	88.1	81
Median				34.1	40.7	42.6			
Stand. dev.				12.56	14.42	12.63			
Daily limit value				35	50	150			

Table 2
PARTICULATE MATTER CONCENTRATIONS IN THE AMBIENT AIR OF RAZOARE DURING THE WINTER SAMPLING CAMPAIGN

Sampling period	Temp, °C	RH, %	precipitations	PM concentrations, µg/m³			PM ratios		
				PM 2,5	PM 10	TSP	2,5/10	10/TSP	2,5/TSP
July 26–27	27	86	cloudy, rain fall	25.3	27.6	43.5	91.7	63.4	58.2
July 27–28	26	64	cloudy	27.5	37.1	55.8	74.1	66.5	49.3
July 28–29	30	48	sunny	36.9	46.7	80.9	79	57.7	45.6
July 29–30	28	62	cloudy	31.4	42.8	57.8	73.4	61.9	45.4
August 1–2	34	46	sunny	34.2	44.6	69.1	76.7	77.2	59.2
August 2–3	36	51	sunny	41.9	52.6	76.4	79.7	68.8	54.8
Mean				32.9	41.9	63.9	79.1	65.9	52.1
Median				32.8	43.7	63.5			
Stand.dev.				6.13	8.64	14.08			
Daily limit value				35	50	150			

Table 3
PARTICULATE MATTER CONCENTRATIONS IN THE AMBIENT AIR OF RAZOARE DURING THE SUMMER SAMPLING CAMPAIGN

between the PM2.5, PM10, and TSP fractions, and the daily limit values.

The mean particulate concentrations in the ambient air during February 8th to February 17th of 2010 were 45.1 ± 12.63 µg/m³ for TSP, 36.9 ± 12.56 µg/m³ for PM 2.5, and 40.4 ± 14.42 µg/m³ for PM10; only PM2.5 having exceeded the daily limit value.

In meteorological terms, during the test periods the weather was characterized by strong precipitation as snow in February 8th to February 12th, which resulted in a reduction of traffic in the area and its complete discontinuation on February 10th. From February 13th the snowfall stopped and traffic gradually returned to normal values. The temperature and relative humidity (RH) values are in the normal range characteristic for winter season.

During the summer, from July 26th to August 3rd of 2010, there is a raise of total suspended particles by 40%, but PM2.5 and PM10 particles maintained the same level as in the winter; so the average concentration of total suspended particles was 63.9 ±14.08µg/m³, where PM 10 was 41.9 ±8.64µg/m³ and PM 2.5 was 32.9 ±6.1µg/m³.

Studies conducted around the world (table 4) showed a comparable level of pollution with the samples PM2.5 and PM10 analyzed in this study.

The results shown in tables 2 and 3 as well as in the figure 2 highlighted the variation of PM2.5 and PM10 amounts detected in the composition of total suspended particles.

During the winter, PM2.5 represents 92% from PM10 and 81% from the total suspended particles detected. On the other hand, during the summer the amount of small particles (PM2.5) in TSP decreased to 52% and in those of larger particles (PM10) decreased to 79%. This changed balance between large (larger than 10 µm) and small particles may be influenced by the resuspension process [32, 34] due to soil erosion during the drought when the wind lifts in the air larger particles located on the ground.

The statistical analysis, realized by Pearson's method (Table 5), indicates a very good correlation (Pearson correlation coefficient, R, from 0.91 to 0.99) between the three sizes of suspended particles collected summer and winter 2010 to Razoare crossroad. This good correlation suggests a common source of pollution in the area: combustion products from the ruttier traffic and burning of fossil fuels for heating the households during the winter.

It had been observed an influence of the meteorological parameters such as temperature and humidity on the concentration of suspended particles in the air. Therefore, during the hot season, it is demonstrated (i) a very good inverse correlation between particle concentrations in the air and humidity (R values from -0.81 to -0.92) and (ii) a good direct correlation to temperature (R values from 0.71 to 0.87). During the cold season, there was maintained a

City/Country	Reference	PM concentrations, µg/m³			
		PM2.5		PM10	
		Winter	Summer	Winter	Summer
Bucharest, Romania	Current study	36.9	32.9	40.4	41.9
Milan, Italy	[7]	60	20	-	-
Yokohama, Japan	[21]	21.15	20.8	29.83	27.65
Palermo, Italy	[1]	33.7	-	44.2	-
Zonguldak, Turkey	[32]	28.1	-	44.1	-
Budapest, Hungary	[2]	-	-	18–50	-
Borgerhout, Belgium	[4]	-	-	33.7	-
Thessaloniki, Greece	[3]	-	-	51	42
Birmingham, UK	[33]	15.8	-	23.9	-

Table 4
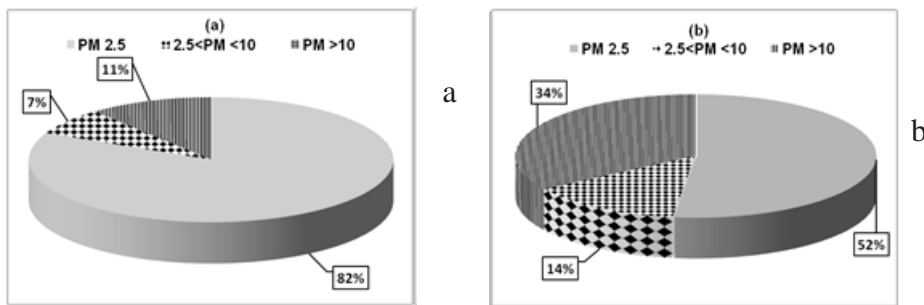CONCENTRATION OF PM2.5 AND PM10 IN AMBIENT AIR AROUND THE WORLD

Fig. 2. The contributions of the particle size fractions to the total suspended particle concentrations in the winter (a) and in the summer (b).

Table 5
PEARSON'S CORRELATION COEFFICIENT VALUES (R)

	Winter, 8–17 February						Summer, 26 July-3 August				
	PM 2,5	PM 10	TSP	Temp	RH		PM 2,5	PM 10	TSP	Temp	RH
PM 2,5	1	0.96	0.93	0.36	-0.86	PM 2,5	1	0.95	0.92	0.87	-0.81
PM 10	0.96	1	0.99	0.29	-0.72	PM 10	0.95	1	0.91	0.78	-0.9
TSP	0.93	0.99	1	0.26	-0.63	TSP	0.92	0.91	1	0.71	-0.92
Temp	0.36	0.29	0.26	1	-0.57	Temp	0.87	0.78	0.71	1	-0.69
RH	-0.86	-0.72	-0.63	-0.57	1	RH	-0.81	-0.9	-0.92	-0.69	1

Table 6
MEAN PAH CONCENTRATIONS (ng/m³) IN THE PARTICULATE MATTER, RAZOARE, 2010

PAH	PM 2.5 (ng/m³)		PM10 (ng/m³)		TSP (ng/m³)	
	Feb. 2010	July 2010	Feb. 2010	July 2010	Feb. 2010	July 2010
Pyrene	1.21	0.35	1.63	0.44	1.61	0.41
Benzo[a]anthracene	1.25	BDL*	1.61	BDL*	1.78	BDL*
Chrysene	1.68	0.21	2.33	0.23	2.31	0.16
Benzo[b]fluoranthene	1.93	0.26	2.62	0.32	2.74	0.29
Benzo[k]fluoranthene	15.87	1.96	22.17	1.97	22.51	1.83
Benzo[a]pyrene	1.27	0.15	1.83	0.15	1.91	0.13
Dibenzo[a,h]anthracene	1.29	BDL*	1.83	BDL*	1.88	BDL*
Benzo[g,h,i]perylene	1.7	0.21	2.25	0.2	2.44	0.19
∑PAHs	26.2	3.14	36.27	3.31	37.18	3.01

BDL* – below detection limit.

good inverse correlation between particle concentrations in the air and humidity (R values from -0.63 to -0.86), but it was a weak correlation to temperature (R values from 0.26 to 0.36).

Polycyclic aromatic hydrocarbons

The mean PAH concentrations in the airborne particulate matter fractions sampled at the Razoare crossroads during the winter and summer are shown in table 6.

Only eight of the 16 PAHs were identified in the particulate matter in the winter: benzo[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[g,h,i]perylene, benzo[k] fluoranthene, chrysene, dibenzo[a,h]anthracene, and pyrene; but, in the summer, benzo[a]anthracene and dibenzo[a,h]anthracene were not identified too.

The lower molecular weight PAHs (with 2–3 rings), such as acenaphthene, anthracene, fluoranthene, fluorene, naphthalene, and phenanthrene were not detected in either of the sampling periods. The dominance of PAHs with 4, 5, and 6 rings in the particulate matter indicates that road traffic was the major source of the PAH air pollution [10, 20, 35].

The PAH concentrations in the particulate matter were 10–15 times higher in the winter than in the summer because of the influence of meteorological conditions and the increased emission of PAHs from the combustion of fossil fuels in power plants and for household heating in the winter which is, in agreement with previous publications [18,19].

The PM2.5 contributed 72% of the PAH concentration in the PM10 and 70% of the PAHs in the TSP. This also shows that road traffic was the main source of PAH pollution, because road traffic emissions are characterized by small particles and relatively high PAH concentrations. The most abundant PAH, which was present in all three particulate fractions, was benzo[k]fluoranthene, which is well known for its carcinogenicity. The mean benzo[k]fluoranthene concentration was 1.83–22.51 ng/m³. Benzo[a]pyrene was found in the PM10 at concentrations of between 0.15 ng/m³ in the summer and 1.83 ng/m³ in the winter. Benzo[a]pyrene is considered to be a useful indicator of PAH air pollution according to the World Health Organization and the European Union [36]. The European Union has established an annual limit value of 1 ng/m³ for benzo[a]pyrene in PM10, and it can be seen from Table 5 that its concentration at the Razoare crossroads exceeded that limit value in the winter.

Conclusions

Concentrations of particulate matter and PAH's in air in the zone of Razoare crossroad in Bucharest during both warm and cold seasons 2010 present important variations due to meteorological factors and specific emission sources. We observed high concentrations of all three dimensional fractions of particulate matter with frequent shifts of the limits imposed for the environmental air quality in the case of PM2.5 and PM10.

There are no major industrial sources in the sampling area, which is surrounded by office buildings, education institutions, and residential buildings, so we concluded that the main source of particulate matter pollution in the area is road traffic, with supplementary emissions from the combustion of fossil fuels for heat during the winter. The high contributions of PM2.5 to the PM10 and TSP and the presence of higher molecular mass PAHs (with 4–6 rings), especially in the PM2.5, also indicate that road traffic is the most important source of pollution in the sampling area.

The paper also aims to be a starting point and a warning about the level of air pollution in Bucharest with breathable particulate matter and PAHs given the adverse effect that it can induce the population from urban area with heavy road traffic.

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