

Original Research

SOURCE-ASSOCIATED HEALTH RISKS RELATED TO METALLIC AND CARBONACEOUS PM₁₀ FRACTION AT TWO URBAN SITES IN SERBIA

DOI: 10.5937/EcoTER26602P

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Abstract

Source apportionment studies and continuous monitoring of air pollutants are crucial for developing effective PM₁₀ mitigation strategies. In addition to source identification and quantification, assessment of health risks posed by PM sources may contribute to more complete understanding of human exposure to this pollutant. Following the USEPA methodology, source-specific health risk assessment was performed in two urban background sites in Belgrade and Bor. For non-carcinogenic risk assessment included species were As, Cd, Pb, Ni, Sb, Se and EC, while for carcinogenic As, Cd, Pb, Ni, and EC were used. In Belgrade, non-carcinogenic risk was characterized as acceptable while smelter factor in Bor presented a hazard quotient (HQ) of 1.3, resulting in HI of 2.07, indicating unacceptable risk. The biggest contributor to total non-carcinogenic risk in Belgrade was traffic/industry and smelter in Bor. Carcinogenic risks (CR) for each source in both cities fell within the range of potential concern but total carcinogenic risk (TCR) was above 1×10^{-4} so the carcinogenic risk in both cities was classified as unacceptable. Results of one-year long WeBaSOOP campaign indicate that contributions of sources to PM₁₀ mass concentration, as determined by positive matrix factorization, may differ from their contributions to health risks. Therefore, health risk assessment findings can provide valuable insights for policymakers when deriving PM₁₀ mitigation measures.

Keywords: particulate matter, health risk, elemental carbon, heavy metals, source apportionment.

INTRODUCTION

Air pollution is a global challenge that affects both climate and human health. Although mitigations measures resulted in decrease of air pollutants' levels in Europe (especially NOx

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and SO₂) in the last decades [1], World Health Organization estimates that still 99% of the population lives in areas with unacceptable levels of pollutants, especially in areas like Northern Italy, Balkans and Eastern Europe [2,3]. In Serbia, the city of Bor, is known for its extensive copper deposits, which are among the largest in Europe. Therefore, the Mining-Metallurgical Complex Bor is considered as the primary source of environmental pollution in the region, due to emissions from the copper smelter, open pits, tailing ponds and associated industrial activities [4]. On the other hand, Belgrade, the capital of Serbia, with a population of approximately 1.7 million, and around 3 000 km² surface area, including inner city and surrounding settlements, due to its high-traffic arterial load, natural gas heating plants, proximity to an international airport, municipal landfill, coal thermal power plants, represents a complex urban environment with multiple sources of air pollution [5].

Among air pollutants, particulate matter (PM) represents the biggest health concern as it is classified as group 1 carcinogen, according to International Agency for Research on Cancer (IARC) [6]. IARC classified both outdoor polluted air and PM as carcinogenic based on the enough evidence that they may lead to development of lung cancer. PM represents heterogeneous mixture of chemicals which composition depends on the sources from which they originate [7]. Therefore, PM may consist of crustal elements (Al, Ti, Ca, Fe) if particles originate from mineral dust, trace elements (As, Cd, Pb, Ni, Cr, Cu, Sb, Sn, Se) if PM originates from industrial or traffic sources, as well as from wood or coal combustion [8]. Organic component includes presence of products of uncomplete combustion, like polycyclic aromatic hydrocarbons (PAHs) or products of fungal metabolism and growth like polyols [9]. These PM₁₀ compounds exhibit varying toxic potentials, making both the size of the particles and their chemical composition critical predictors of PM toxicity [10].

According to the studies conducted so far, heavy metals (HMs) (e.g., As, Cd, Pb, Ni, Cr, Hg) are considered as important PM compounds from toxicological point of view [11,12]. WHO classified As, Cd, Pb and Hg among the chemicals of public concern while As, Cd, Ni and Cr are classified as carcinogenic to humans (group 1) by IARC. So, when evaluating health risk from PM₁₀ exposure, it is important to take into consideration HMs even though they do not contribute substantially to the PM mass concentration. On the other hand, carbonaceous aerosols (organic carbon (OC) and elemental carbon (EC)) are often the largest component of PM. EC is emitted from the incomplete combustion of carbon-containing materials and is correlated with negative effects on both climate and health. However, health risk assessment studies that included EC are still limited [13]. Therefore, the aim of our study was to assess health risks from PM₁₀ sources in relation to the presence of metallic and carbonaceous component, observed in Belgrade and Bor, Serbia.

MATERIALS AND METHODS

PM₁₀ sampling, chemical characterization and PM₁₀ source apportionment

PM₁₀ sampling, chemical analysis of PM₁₀-bound HMs and EC and source apportionment of PM₁₀ mass concentration during WeBaSOOP campaign in Belgrade (June 2023 – May 2024) and Bor (March 2023 – February 2024) were described in Petrovic *et al.* [14]. The source apportionment using positive matrix factorization (PMF) was used to identify independent factors that serve a proxy for PM₁₀ emission sources. The PMF included 33

species (OC, EC, 4 ions, 27 elements, 10 organic tracers) in the model. For the source-specific health risk assessment, concentrations from the output that model assigned to each resolved factor were used in Eq. 1.

Source-related health risk assessment

To assess non-carcinogenic and carcinogenic health risk of specific factors apportioned by PMF in Bor and Belgrade during WeBaSOOP campaign, the Inhalation Dosimetry Methodology by USEPA was followed [15]. The risk analysis included only those species used in the PMF model that had available reference values for Inhalation Toxicity Reference Concentration (RfC) and Inhalation Unit Risk (IUR) in the Risk Integrated Information System (IRIS). HMs used for non-carcinogenic risk assessment in Belgrade were As, Cd, Ni, Pb and Sb while in Bor As, Cd, Ni, Pb, Sb and Se were considered. As more than 50% of Se values were below limit of detection, it was not included in non-carcinogenic risk assessment in Belgrade. In case of carcinogenic risk, As, Cd, Ni and Pb were included in both cities. To account for the influence of the carbonaceous fraction alongside the metallic fraction in the health risk, EC concentration values were incorporated. In case of EC, RfC and IUR values from IRIS database for diesel engine exhaust were used [13]. Inhalation exposure concentrations (IEC) for resident adult population in Belgrade and Bor were calculated following Eq. 1:

$$IEC = (CA \times ED \times EF \times ET) / AT \quad (1)$$

Values of exposure parameters were taken from USEPA Exposure Factor Handbook [16]. CA represents HMs and EC concentration in the air ($\mu\text{g}/\text{m}^3$) obtained from PMF output where concentrations of HMs and EC were assigned to every factor. Exposure duration (ED) was 26 years, Exposure frequency (EF) 350 days per year (for residents), for Exposure time (ET) 24 hours per day were considered and Averaging time (AT) for non-carcinogenic risk was calculated as $ED \times 350 \text{ days/year} \times 24 \text{ hours/day}$ while for carcinogenic risk, AT was $70 \text{ years} \times 350 \text{ days/year} \times 24 \text{ hours/day}$ [15].

For non-carcinogenic risk, Hazard Quotients (HQ) for every HM and EC for each apportioned factor were calculated first (Eq. 2). The sum of HQs for each identified PM_{10} factor was Hazard Index (HI) (Eq. 3) which was used to classify the non-carcinogenic risk. It was considered acceptable when the HI value was below 1, and unacceptable when the HI value exceeded 1.

$$HQ = IEC / (RfC \times 1000 \mu\text{g}/\text{mg}) \quad (2)$$

$$HI = \sum HQs \quad (3)$$

Carcinogenic risk (CR) was calculated by multiplying IUR value for each HM and EC with IEC (Eq. 4) and total carcinogenic risk (TCR) (Eq. 5) was calculated as a sum of CRs for each factor in Belgrade and Bor. The acceptable TCR value is below 1×10^{-6} , TCR between

1×10^{-6} and 1×10^{-4} represent possible risk and values higher than 1×10^{-4} indicate unacceptable risk [17,18].

$$CR = IEC \times IUR \quad (4)$$

$$TCR = \sum CRs \quad (5)$$

RESULTS AND DISCUSSION

PM₁₀ source apportionment

After the PMF analysis, seven factors were resolved in Belgrade and eight in Bor. In Belgrade, resolved factors were mineral dust (MD), biomass burning (BB), ammonium sulphate (SUL), primary biological aerosol particles (PBAP), biogenic secondary organic aerosol from isoprene (BSOA_{Isoprene}) and long-range transport and local NaCl mixed with combustion (NaCl/comb) [14]. In Bor, PMF resulted in 8 factors: BSOA_{Isoprene}, Mixed combustion (MIXcomb), MD, secondary inorganic aerosol (SIA), Industrial dust (ID), PBAP, Smelter (SM) and BB [In preparation].

Source-related health risk assessment

Non-carcinogenic health risk assessment

Traditional health risk assessment methods do not differentiate risks from natural, anthropogenic, and secondary PM sources. Therefore, allocating health risks to the distinct sources identified by PMF is essential for better understanding source-specific health impacts. [13]. In Belgrade, obtained HQ values for every factor were below 1 and their sum resulted in HI value of 0.46 so non-carcinogenic risk from exposure to HMs and EC in the capital of Serbia was classified as acceptable (Table 1). The biggest HQ value was obtained for traffic/industry factor, then biomass burning and NaCl/comb factors, however, all values were below 1. In Bor, obtained HQ values were below 1 except for smelter factor (HQ = 1.3). The HI value for Bor was 2.07 so non-carcinogenic risk was characterized as unacceptable. After the smelter factor, biomass burning, and mixed combustion contributed the most to non-carcinogenic risk in Bor, but its values were negligible in comparison with HQ_{smelter}. The city of Bor is considered as a pollution “hot spot” at both the local and regional levels due to the nonferrous metallurgical industrial complex located in close proximity to the urban area, within sight and walking distance of residential zones. Until 2016, the outdated pyrometallurgy technology for copper production was applied [19] and since 2016, new technology for the melting copper concentrates in the smelter is used. However, according to our results, smelter activities still pose health risk for Bor inhabitants, mainly due to high As content in this factor. A study from 10 years ago, conducted by Jovanovic *et al.* [20] emphasised that Bor is more polluted by HMs, while Belgrade is more polluted by carbonaceous aerosols. EC, emitted from the combustion of fossil fuels such as diesel, coal, and biomass, was observed at higher levels in Belgrade during the heating season due to biomass burning, while elevated levels during spring and summer are likely associated with more pronounced traffic emissions in the city, compared to Bor [14].

The contributions of individual HMs and EC to the HQs varied among the PM₁₀ factors. In Belgrade, the highest HQ values were obtained for EC in case of BB, TRA/IND and BSOA_{Isoprene} factors. EC contributed around 50% to the HQs of SUL, NaCl/comb and PBAP factors also. Beside EC, Ni was the second dominant compound in risk contribution, with biggest contribution in NaCl/comb, MD, SUL and PBAP factors. The presence of EC in PBAP and BSOA_{Isoprene} factors was less expected result. EC in these factors was observed in other PMF studies in Europe [9]. EC presence in factors from biological origin may be explained by the fact that, during OC/EC analysis, some types of PBAP char and evolve as EC [21]. In Bor, unlike in Belgrade, HQ values of HMs contributed the most to the HI (Table 2). HQ_{As} had the highest contribution to most of the apportioned factors – ID, SM, SIA, MD, BB and BSOA_{Isoprene}. Beside As, Ni contributed significantly to the health risk from MIXcomb, PBAP and ID, and, in case of Cd, SM and BB. The presence of Ni in the PBAP and BSOA isoprene factors in both Belgrade and Bor may result from atmospheric mixing processes, such as road dust resuspension, leading to the adsorption of elements onto particles of biological origin [22].

Table 1 HQ values in Belgrade

	MD	BB	SUL	PBAP	BSOA _{Isoprene}	TRA/IND	NaCl/comb	HI
As	3.6E-03	1.4E-02	9.1E-03	2.3E-03	2.9E-03	1.3E-02	6.5E-03	
Cd	3.2E-04	4.3E-03	1.9E-03	7.5E-04	3.0E-04	4.1E-03	1.4E-03	
Pb	2.2E-04	1.1E-03	3.2E-04	3.2E-04	4.8E-04	1.9E-03	7.8E-04	
Ni	3.2E-02	3.9E-03	1.7E-02	3.2E-02	2.1E-03	2.2E-02	3.1E-02	
Sb	2.1E-04	1.7E-04	3.2E-04	3.0E-05	1.8E-04	1.9E-03	4.0E-04	
EC	0	5.5E-02	2.0E-02	2.9E-02	1.0E-02	1.0E-01	3.3E-02	
HQ	3.6E-02	7.9E-02	4.9E-02	6.4E-02	1.6E-02	1.5E-01	7.3E-02	4.6E-01

MD – mineral dust, BB – biomass burning, SUL – ammonium sulphate, PBAP – primary biological aerosol particles, BSOA_{Isoprene} – biogenic secondary organic aerosol from isoprene, TRA/IND – traffic/industry, NaCl/comb – long-range transport and road salt/local combustion.

Table 2 HQ values in Bor

	BSOA _{Isoprene}	MIXcomb	MD	SIA	ID	PBAP	SM	BB	HI
As	2.6E-02	5.4E-03	2.0E-02	3.2E-02	8.9E-02	2.1E-02	6.7E-01	4.9E-02	
Cd	9.3E-03	1.1E-03	0	1.1E-02	0	1.1E-02	4.5E-01	5.5E-02	
Pb	2.0E-03	1.3E-03	2.5E-03	4.8E-03	1.4E-02	2.8E-03	5.9E-02	2.4E-03	
Ni	1.5E-02	1.2E-01	0	9.8E-03	4.2E-02	4.1E-02	6.0E-02	2.9E-02	
Sb	7.1E-04	1.7E-04	1.6E-03	4.9E-04	5.2E-04	4.1E-04	1.4E-03	1.8E-04	
Se	4.3E-06	2.9E-06	8.8E-08	1.4E-05	4.3E-05	6.4E-06	1.7E-04	6.2E-06	
EC	1.3E-02	4.0E-02	1.3E-02	1.4E-03	3.0E-05	1.1E-02	5.8E-02	7.2E-02	
HQ	6.6E-02	1.7E-01	3.7E-02	5.9E-02	1.4E-01	8.7E-02	1.3E+00	2.1E-01	2.07

BSOA_{Isoprene} – biogenic secondary organic aerosol from isoprene, MIXcomb – mixed combustion, MD – mineral dust, SIA – secondary inorganic aerosol, ID – industrial dust, PBAP – primary biological aerosol particles, SM – smelter, BB – wood burning. HI value higher than 1 is in bold.

Carcinogenic health risk assessment

In case of carcinogenic risk, independent CR values for factors in Belgrade and Bor were within the range of possible health risk (between 1×10^{-6} and 1×10^{-4}), except mineral dust in Belgrade, where CR value was in range of acceptable risk (Table 3). However, the total

carcinogenic risk in both Belgrade and Bor were above 1×10^{-4} so the carcinogenic risk in Belgrade was classified as unacceptable and so does in Bor. Although having similar TCR values, factor that contributed the most to TCR in Belgrade was TRA/IND while SM contributed the most in Bor. EC was the dominant species contributing the most to CR in both Belgrade and Bor. Exceptions were the ID, SM, and SIA factors in Bor, where As contributed significantly to the CR values (Tables 3 and 4). The study of Li *et al.* [23] observed the highest total health risks, both carcinogenic and non-carcinogenic, from mining and smelting activities in region of Central China. According to WeBaSOOP campaign results smelting activities still represent the biggest health concern, in case of both non-carcinogenic and carcinogenic risk even though new technologies in smelting facility were applied in 2016 [19]. In the case of Belgrade, higher EC concentration and, consequently, the high contribution of the traffic factor to carcinogenic risk, may be attributed to the fact that passenger car fleet in Serbia primarily consist of imported, fossil-fuelled second-hand cars [24]. In concordance of our study were results of source-oriented health risk assessment where factor associated with road dust and petroleum volatilization contributed the most to the total cancer risk in Belgrade city centre [25].

Table 3 CR values in Belgrade

	MD	BB	SUL	PBAP	BSOA _{Isoprene}	TRA/IND	NaCl/comb	TCR
As	8.60E-08	3.45E-07	2.19E-07	5.47E-08	7.11E-08	3.05E-07	1.55E-07	
Cd	2.14E-09	2.85E-08	1.24E-08	4.99E-09	2.00E-09	2.77E-08	9.59E-09	
Pb	3.23E-09	1.64E-08	4.78E-09	4.80E-09	7.16E-09	2.83E-08	1.16E-08	
Ni	3.98E-08	4.89E-09	2.15E-08	3.97E-08	2.68E-09	2.73E-08	3.82E-08	
EC	0	3.06E-05	1.14E-05	1.61E-05	5.83E-06	5.76E-05	1.85E-05	
CR	1.31E-07	3.10E-05	1.17E-05	1.62E-05	5.91E-06	5.80E-05	1.87E-05	1.42E-04

MD – mineral dust, BB – biomass burning, SUL – ammonium sulphate, PBAP – primary biological aerosol particles, BSOA_{Isoprene} – biogenic secondary organic aerosol from isoprene, TRA/IND – traffic/industry, NaCl/comb – long-range transport and road salt/local combustion.

Table 4 CR values in Bor

	BSOA _{Isoprene}	MIXcomb	MD	SIA	ID	PBAP	SM	BB	TCR
As	6.17E-07	1.30E-07	4.74E-07	7.58E-07	2.13E-06	5.03E-07	1.61E-05	1.18E-06	
Cd	6.19E-08	7.32E-09	0	7.50E-08	0	7.46E-08	3.02E-06	3.66E-07	
Pb	3.04E-08	1.94E-08	3.76E-08	7.18E-08	2.01E-07	4.09E-08	8.75E-07	3.58E-08	
Ni	1.88E-08	1.54E-07	0	1.22E-08	5.25E-08	5.16E-08	7.44E-08	3.61E-08	
EC	7.33E-06	2.22E-05	7.24E-06	7.68E-07	1.65E-08	5.92E-06	3.24E-05	4.01E-05	
CR	8.06E-06	2.25E-05	7.75E-06	1.68E-06	2.40E-06	6.59E-06	5.25E-05	4.18E-05	1.43E-04

BSOA_{Isoprene} – biogenic secondary organic aerosol from isoprene, MIXcomb – mixed combustion, MD – mineral dust, SIA – secondary inorganic aerosol, ID – industrial dust, PBAP – primary biological aerosol particles, SM – smelter, BB – wood burning.

Comparing the contributions of the apportioned factors to total PM₁₀ mass concentrations and to non-carcinogenic and carcinogenic risks, differences may be observed. In Belgrade, the main contributors to PM₁₀ mass were BB, SUL, and MD, whereas TRA/IND, BB, and NaCl/comb were the dominant contributors for the assessed risks. Although SM contributed with small percent to PM₁₀ mass in Bor, it was the dominant contributor to both HI and TCR.

This difference in contributions to PM₁₀ mass and health risks supports the hypothesis that PM mass alone is insufficient when dealing with health risks posed by atmospheric particles.

CONCLUSION

Conducting source apportionment studies in order to identify and quantify main sources of PM₁₀ is essential for targeted mitigation measures. The mass concentration of PM is not the most relevant metric when dealing with health impacts of airborne particles since major properties (chemistry, shape, size distribution, solubility, speciation) driving PM toxicity are not taken into account within this single mass metric. Therefore, to examine health risk from PM exposure, health risk assessment may be used as additional tool to identify sources that pose the biggest health concern regardless its contribution to the PM₁₀ mass.

Results from non-carcinogenic health risk assessment showed unacceptable risk in Bor based on HI value of 1.3 for smelter factor. On the other hand, results for Belgrade showed acceptable risk. In case of carcinogenic risk, all values in both Belgrade and Bor were within range of possible risk. These findings may give additional information for policymakers when deriving PM₁₀ mitigation strategies, to take into consideration source toxicity, beside solely its contribution to PM₁₀ mass.

ACKNOWLEDGEMENT

This research was funded by the European Union's Horizon Europe Research and Innovation Program under GA 101060170 (WeBaSOOP project) and the Ministry of Science, Technological Development, and Innovation of the Republic of Serbia under GA 451-03-33/2026-03/ 200017.

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