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Chemical characterization and source apportionment of PM_{10} in Belgrade, Serbia: influence of local and regional anthropogenic and natural sources

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Abstract

Substantial efforts and improvements in air quality across Europe lowered levels of air pollutants including particulate matter (PM) in the last decades. However, significant proportion of the European population still lives in areas exceeding WHO recommendations, especially in Northern Italy, Balkans and Eastern Europe, including Serbia. Targeted PM mitigation strategies require extensive air quality monitoring and modelling including source apportionment (SA) studies. In the past, numerous SA studies were conducted for Belgrade city, capital of Serbia. Nevertheless, comparisons across the results are difficult, as they encompass different datasets of pollutants contained in PM fractions such as elements and/or ions and/or PAHs. Here, the aim is to offer a broader insight on PM_{10} sources at an urban background site in Belgrade by including 34 species as input variables for SA (carbonous aerosols, elements, ions and specific organic tracers). For SA, the USEPA PMF 5.0 software was applied. The factor that dominated PM_{10} mass was biomass burning (21%), primarily during heating season, followed by ammonium sulphate (18%) and mineral dust (17%). A mixed traffic and industrial activity accounted for 15% of PM_{10} mass while contribution of factors from biological origin, primary biological aerosol particles and biogenic secondary organic aerosols from isoprene, was 10% each. Mixed factor of long-range transport and road salt/local combustion contributed to the PM_{10} mass 9%. This analysis provides more detailed perspective on the composition and sources of PM_{10} in Belgrade, both from anthropogenic and natural, including biological origin. These findings are valuable for defining targeted PM_{10} mitigation strategies.

Key words: air pollution, particulate matter, PM_{10}, source apportionment, chemical composition, organic tracers

1. Introduction

The World Health Organization (WHO) estimated that 4.2 million premature deaths at the global level are caused by exposure to outdoor air pollution (WHO, 2024). Among the air pollutants, particulate matter (PM) represents the biggest health concern. PM may originate from both anthropogenic and natural sources. These sources emit particles that vary in size, morphology, and chemical composition, and these differences influence how particles may be harmful for human health (Liu et al. 2025). Coarse particles (<10 µm and >2.5 µm) primarily deposit in the extra thoracic airways, whereas fine (<2.5 µm) and ultrafine particles (10-100 nm) (Ruuskanen et al. 2001) can penetrate deeper into the pulmonary region of respiratory tract (Khan et al. 2022, Zhai et al. 2024). Exposure to particles lower than 10 µm (PM_{10})

is related to increased mortality and morbidity from cardiovascular and respiratory diseases (Polichetti et al. 2009; Newell et al. 2017). In 2016, the International Agency for Research on Cancer (IARC) classified outdoor air pollution, as well as PM, as carcinogenic to humans (group 1) based on sufficient evidence in both humans and experimental animals. According to IARC, exposure to PM contributes to the development of lung cancer (IARC, 2016).

Although substantial efforts and improvements in air quality across EU significantly lowered levels of air pollutants (especially SO_2, NO_x) in the period from 2000 to 2019 (Aas et al. 2024), there is still important proportion of the European population that lives in areas exceeding WHO recommendations (EEA, 2025a). The most severe situation is in low- and middle- income countries where is estimated that citizens are exposed to up to four times higher levels of ambient $PM_{2.5}$ (Health Effects Institute, 2024). Cohen et al. (2017) demonstrated that even modest increases in PM concentrations can result in disproportionately large increases in health risks, underscoring the need for further mitigation. Moreover, substantially stronger evidence now indicates that air pollution may impact human health at lower exposure levels than previously recognized (Stafoggia et al. 2022). Therefore, the WHO established new guidance levels in 2021 where new set of annual and daily average levels of PM_{10} is lowered and may be used by policy makers who are responsible for air pollution control. WHO recommended average annual PM_{10} level is 15 µg/m^3 while average 24-hour target level is 45 µg/m^3.

Targeted PM_{10} mitigation strategies require extensive air quality monitoring and modelling including source apportionment studies. Therefore, new EU Air Quality Directive underlines the importance of monitoring at supersites novel pollutants such ultrafine particles, black carbon and elemental carbon, ammonia and oxidative potential (Directive (EU) 2024/2881). Directive emphasize that it is crucial to systematically monitor air quality in areas where pollutants concentrations are high, and measurements are scarce. Annual PM_{10} levels in Serbia in 2023 and 2024 exceeded EU limit value (40 µg/m^3) on several monitoring sites in Belgrade, Valjevo, Užice, Zaječar, Novi Pazar and Šabac. In 2023 and 2024, PM_{10} was the pollutant with the highest number of exceedance days in the Republic of Serbia (SEPA, 2024, 2026). European Environment Agency reports show that Serbian PM_{10} levels in 2023 were higher than European averages (EEA, 2025b) and Health Effects Institute report that air quality monitoring data is still scarce in this area (Health Effects Institute, 2022). Liu et al. 2025 emphasised that sources like secondary aerosols and road traffic, show consistent chemical profile, temporal variability, and contribution to PM_{10} concentration in source apportionment studies conducted in twenty-four cities across Europe. However, emissions from industry, heavy oil combustion, and crustal sources vary considerably by location, making local source apportionment essential for effective pollution control.

Source apportionment studies using different approaches were also conducted in Belgrade Metropolitan area, collectively covering the sampling period from 2003 to 2015. The sources most commonly identified were coal and wood combustion, secondary aerosols, traffic (vehicle exhaust) and mineral matter (Mijić et al. 2012; Cvetković et al. 2015; Stojić et al. 2016; Todorović et al. 2020). The influence of marine aerosol, construction activities, fertilizer plant, oil combustion and industry were observed too (Rajšić et al. 2008; Đorđević et al. 2012) (Table S1). In these studies, source with the biggest contribution to PM concentration was coal and wood combustion which authors mainly attributed to the thermal power plants “Kolubara A” and “Nikola Tesla A and B”, located 50 km and 25 km from Belgrade, respectively, as well as the usage of coal and wood for district and household heating. (Đorđević et al. 2012) observed that the dominant ionic specie in 2008 in Belgrade was formation of ammonium sulphate. Sulphate in the atmosphere mainly originates from the oxidation of SO_2 in urban areas and ammonium ion originates from biogenic NH_3 but also from fertilizer plant located in Pančevo, 15 km away from Belgrade (Đorđević et al. 2012). However, the fertilizer plant in Pančevo is closed today. In the period when the majority of the abovementioned studies were conducted, the consumption of natural gas for district heating was only 30% of the total gas consumption in Serbia (period 2001 - 2010) (Ivezić et al. 2016). Today, natural gas is the main fuel used for district heating in “Belgrade Power Plants” (https://beoelektrane.co.rs/zastita-zivotne-sredine/). Further, by 2020, around 1300 heating boilers within the "Belgrade Power Plants" system had been shut down and reoriented to operate on natural gas (https://beoelektrane.co.rs/o-nama/osnovni-podaci/). In 2024, the flue gas desulphurisation plant at "Nikola Tesla A" started operating which resulted in reduction in SO_2 emissions (https://www.eps.rs/eng/Pages/Istorija.aspx). The international E-70 highway passes through Belgrade but today, it functions primarily for local transport since the new bypass road around Belgrade now serves as the main transit road (Ćirović et al. 2026).

Abovementioned studies used different receptor modelling (Unmix, principal component analysis, positive matrix factorisation (PMF)), making it difficult to compare obtained results. Furthermore, some studies used only elements or PAHs for source apportionment. In light of this, the present study aimed to perform PM_{10} source apportionment in Belgrade using PMF, a multivariate receptor model capable of identifying factors without prior knowledge of locally relevant source profiles (Hopke et al. 2020) and allows for the quantification of both the chemical composition of these factors and their contribution to PM concentrations (Weber et al. 2019). Incorporating OC, EC, ions, elements, and specific organic tracers as input variables, approach enabled the quantification of both natural, including biological, and anthropogenic sources, thereby offering a broader perspective on PM_{10} sources at an urban background site in Belgrade.

2. Material and methods

2.1. Sampling site

Our sampling campaign was performed at an urban background monitoring site Ada Marina (44.790166° N, 20.4169° E; 71 m above sea level) which is a part of the local air quality network in Belgrade Metropolitan, operated by Public Health Institute of Belgrade (Fig. 1). Site is located at Ada Ciganlija, the largest recreational area in the central area of Belgrade city, along the Sava River. Possible pollution sources include numerous restaurants in the vicinity of the site places around the lake, biomass burning from the nearby residential area and a high traffic arterial road nearby. In addition, E-70 highway is located approximately 250 meters away from the sampling site. Other potential sources of aerosol pollution include a natural gas heating plants for district heating, located about 1.5 km to the Northwest and Southeast. Airport "Nikola Tesla" is 10 km in the Northwest. A municipal landfill at Vinča settlement is 10 km to the East. Another possible pollution source is road traffic associated with 1.7 million citizens and the fact that passenger car fleet in Serbia primarily consist of imported, fossil-fuelled second-hand cars (Mijailović et al. 2019). Furthermore, the coal-fired thermal power plant "Nikola Tesla" is located around 25 km to the Southwest while power plant "Kolubara" is distanced 50 km to the South of the Ada Marina site. Outside the city, there are the areas of agricultural activity as well as Smederevo ferreous smelter and "Kostolac" thermal power plant, 30 km and 60km, respectively, to the East.

2.2 Aerosol filter samples

Three simultaneous ambient aerosol filter samples were collected from 6.6.2023 to 28.5.2024, each from three co-located and identical low-volume samples (LVS3, Leckel) equipped with PM_{10} inlets. The samplers were loaded with quartz fibre filters (PALLFLEX Tissuequartz 2500QAT-UP, 47 mm in diameter), which were conditioned at 20 ± 1 °C and 50 ± 1 % RH for 48 h prior to sampling to enable gravimetric determination of PM_{10} mass. Filters from one sampler were designated for the analysis of OC, EC, organic tracers, and oxidative potential. Filters from a second sampler were conditioned following exposure for gravimetric analysis, then stored in polystyrene Petri slides at –20 °C until elemental and ionic analyses were performed. PM_{10} filter samples from a third sampler were retained as backups. All filters were stored at 4 °C prior to exposure and at –20 °C afterward. Field blank filters were collected to assess potential contamination during handling and transport. OC, EC, ions, elements and organic tracers were analysed from 84 samples.

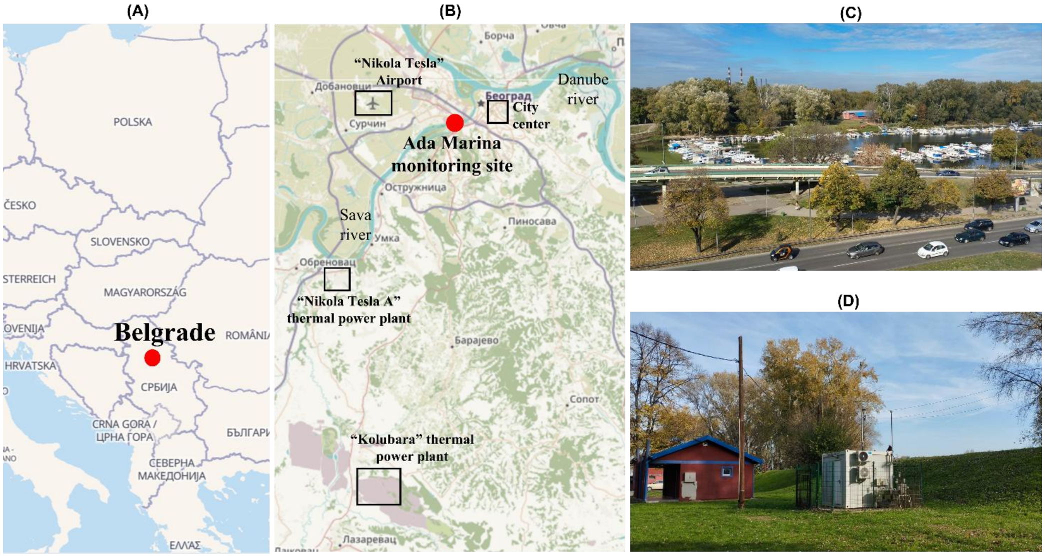

Figure 1. Location of the sampling site at Ada Marina, Belgrade, Serbia (Source: © OpenStreetMap contributors), (A) Map of Serbia with neighbouring countries, (B) wider region of the Belgrade area with points of interest, (C) and (D) the nearest surrounding of the monitoring site.

2.3 Measurement of OC and EC

Organic carbon (OC) and elemental carbon (EC) were measured by thermal-optical analysis using a Sunset Laboratory OC/EC aerosol analyser. The instrument operated according to the EUSAAR-2 temperature protocol (Cavalli et al. 2009) using optical transmission correction for charring.

2.4. Measurement of the elements and ions

The elements (Na, Mg, Al, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Cd, Ba, Pb, P, Ti, Zr, Mo, Sb, Sn) were quantified utilizing inductively coupled plasma – mass spectrometry (Agilent model 7700), following the procedure described in (Querol et al. 2001). Ions (Cl^-, NO_3^-, SO_4^{2-}, NH_4^+) were analysed from the solid phase of suspended particles in the air using ion chromatography (IC). The analysis was performed on Thermo Scientific Dionex™ ICS-1600 ion chromatograph with suppressed conductometric detection.

2.5. Measurements of organic tracers

Organic tracers (Table 1) were analysed using ultra-high-performance liquid chromatography (UHPLC; Vanquish UHPLC, Thermo Fisher Scientific) coupled with a Q Exactive™ Plus Orbitrap mass spectrometer (Thermo Fisher Scientific), following the method described by (Yttri et al. 2024).

Table 1. Organic tracers analysed in this study and their source categories.

Organic tracers	Sources
Levoglucosan, mannosan galactosan	Biomass Burning
Arabitol, mannitol, fructose, glucose, trehalose	Primary Biological Aerosol Particles
2-methylerythritol, 2-methylthreitol	Biogenic Secondary Organic Aerosol from oxidation of isoprene

2.6. Positive matrix factorization model

In the present study, the EPA PMF v.5.0 has been used. Our data matrix dimensions were 84 × 44 (sample number, number of species, respectively). As the input data we used concentration of species obtained from filter analysis and uncertainties calculated following procedure in (Norris et al. 2014). Uncertainties were calculated using Eq. (4) if the concentration of the species was higher than limit of detection (LOD) value.:

$$u = \sqrt{(error\ fraction\ \times concentration)^2} + \sqrt{(0.05\ \times LOD)^2} \qquad (4)$$

Otherwise, if the concentration data was below the LOD, it was replaced with half of the LOD and the uncertainties for the values below LOD were set to 5/6 of the LOD (Glojek et al. 2024). Error fraction for all species was 20%.

In this study, signal-to-noise ratio (S/N) and percentage of the samples below LOD (50%) were used to classify species as “strong”, “weak” and “bad”. Species selected for PMF are reported in Table S4. The species that had $S/N < 1$ and samples below $LOD > 50\%$ were considered as “bad” (in our study Se, Ga, Co, Zr), while species that had $S/N < 1$ and samples below $LOD < 50\%$ and vice versa ($S/N > 1$, below $LOD > 50\%$) were considered as “weak” (Cl^-, Cu, As, Cd, Ti, Sn). Species with S/N ratio > 1 and samples below $LOD < 50\%$ were considered “strong” (OC, EC, NO_3^-, SO_4^{2-}, NH_4^+, Na, Mg, Al, K, Ca, V, Mn, Fe, Rb, Sr, Pb, Sb, galactosan, mannosan, levoglucosan, 2-methylerythritol, 2-methylthreitol, glucose, fructose, mannitol, arabitol, trehalose). Few species, although having a satisfactory S/N and percentage of above LOD, were excluded from the model due to the contamination of the blanks (Cr, Ni, Mo, Ba) or higher analytical uncertainty of the results (Inositol, Erythritol, Zn, P).

3. Results and discussion

3.1 PM_{10} mass concentration and chemical composition

The average annual level of PM_{10} during WeBaSOOP campaign (23.9 µg m^{-3}) (Table S3), was below threshold value of 40 µg m^{-3} (Government of the Republic of Serbia, 2013). Regulation on conditions for monitoring and requirements for air quality is based on Serbian Law on Air protection (Government of the Republic of Serbia, 2025) which is in accordance with Directive from 2008 (Directive (EU) 2008/50/EC). However, average annual level measured in Belgrade exceed the latest limit value in new EU Directive 2024/2881, that will be in force from 2030, (20 µg m^{-3}) as well as WHOs guidelines from 2021 (15 µg m^{-3}). PM_{10} concentrations showed seasonal pattern with higher levels during heating (28 µg m^{-3}) (15th October to 15th April) than during non-heating season (19 µg m^{-3}). Daily limit value (50 µg m^{-3}) was exceeded four times and WHO recommended value (45 µg m^{-3}) seven times during campaign, in heating season, with the highest daily concentration recorded on December 19th (92 µg m^{-3}).

Carbonaceous fraction (OM + EC) was the major contributor to PM_{10} mass concentration with the highest contribution in winter (62%, 20 µg m^{-3}), then in autumn (58%, 13.5 µg m^{-3}), summer (49%, 9.9 µg m^{-3}) and spring (41%, 8.3 µg m^{-3}) (Fig. 2, Table S3). Organic matter was calculated as OC × 1.4 for all seasons (Turpin and Lim 2001). The second major contributor to PM_{10} mass concentration was secondary inorganic aerosol (SIA) with 19% on average. Sulphate and ammonium ions didn’t show seasonal variability, while nitrate was higher in winter (2.7 µg m^{-3}) and autumn

(1.6 µg m^{-3}) season, compared to summer (0.4 µg m^{-3}) and spring (0.6 µg m^{-3}). Semivolatile character of nitrate is responsible for its lower levels during warmer seasons due to increased temperatures and photochemical activity while sulphate is more stable under the same conditions (Waked et al. 2014). Mineral dust (MD) content was estimated following methodology described in (In't Veld et al. 2021). MD contributed to PM_{10} mass with 13% on annual average, without pronounced seasonality. Negligible contribution of trace elements (0.5%, 0.120 µg m^{-3}) and Cl^- (0.6%, 0.135 µg m^{-3}) was observed (Fig. 2).

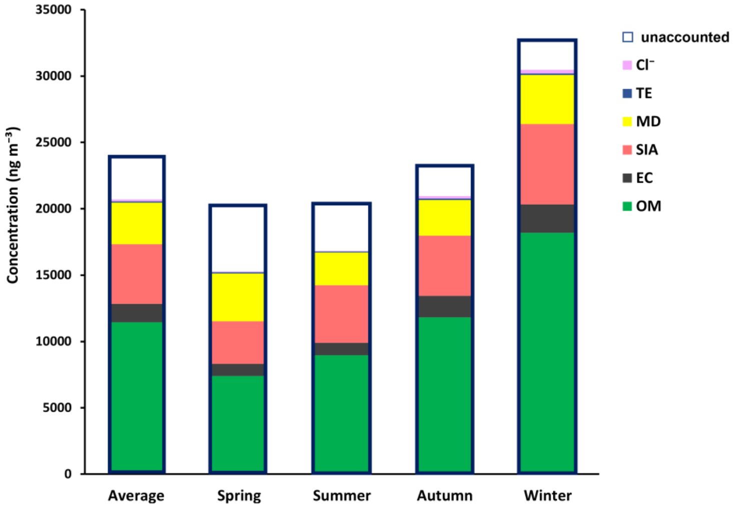

Figure 2. Major composition of PM_{10} in Belgrade on annual level and during different seasons. Every bar represents total PM_{10} on annual and seasonal average concentration. Major PM_{10} reconstructed components were: OM – organic matter, EC – elemental carbon, SIA – secondary organic aerosol, MD – mineral dust, TE – trace elements.

3.2 Identification of the factors

Using the PMF model, seven factors were identified based on their chemical profiles and the tracer species characteristic of specific emission sources (Fig. 3): Biomass burning (BB); Long range transport and local combustion (NaCl/COMB); Mineral dust (MD); Primary biological aerosol particles (PBAP); Biogenic secondary organic aerosol from isoprene ($BSOA_{Isopprene}$); Ammonium sulphate; Traffic/ Industry (TRA/IND). Factor fingerprints and relative contribution of the identified factors to PM_{10} are presented in Fig. S1 and S2 and in Table S4. Factor chemical profiles for each factor are presented in Fig. 3, whereas time evolution of daily contributions and wind polar plots are depicted in Fig. 4.

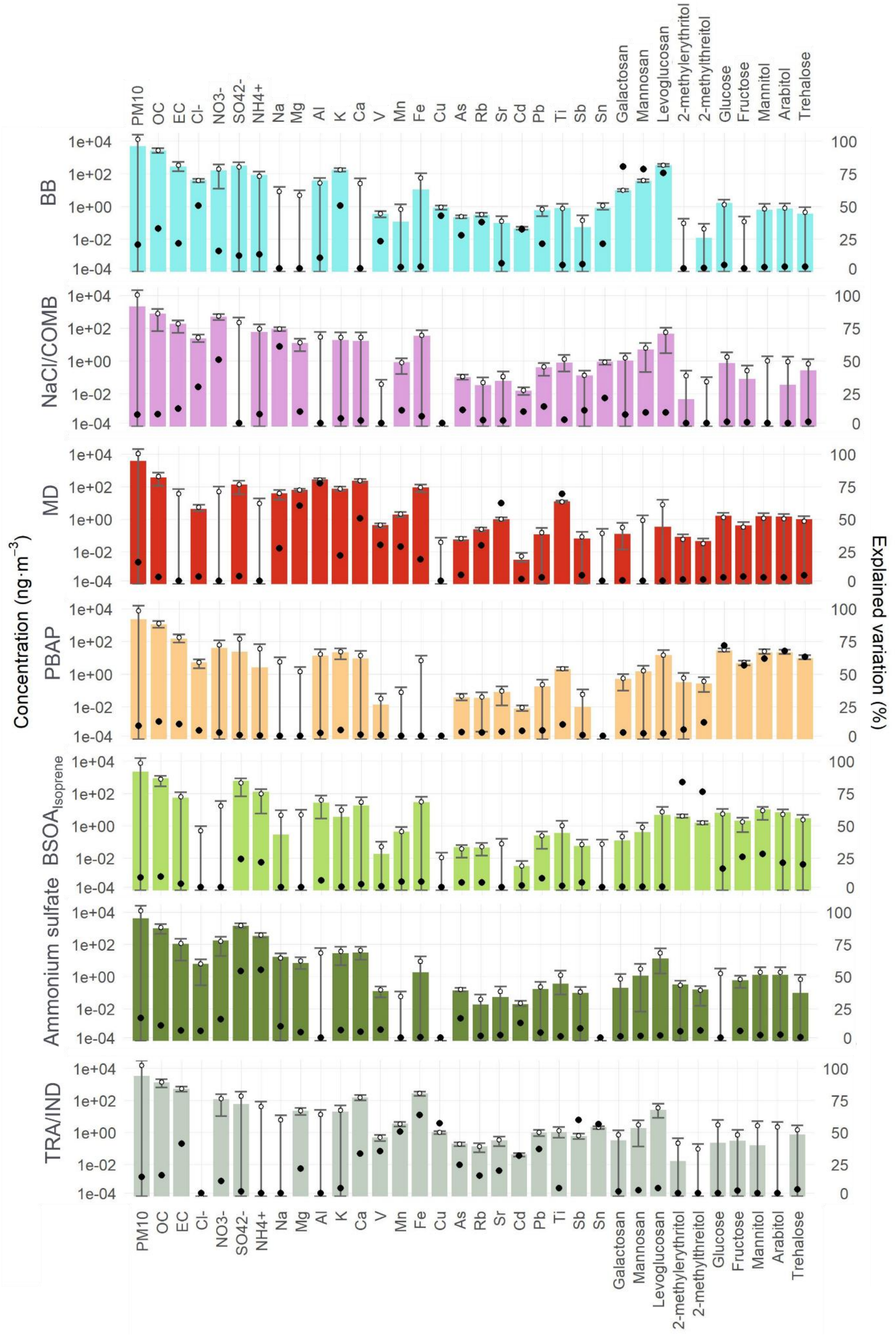

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Figure 3. Chemical profiles of the PMF factors in Belgrade Ada Marina. On the x-axis are shown species, on y-axis chemical concentration of the species in ng m^{-3} with uncertainties and, on the secondary y-axis are shown relative contributions of the species apportioned by the factor.

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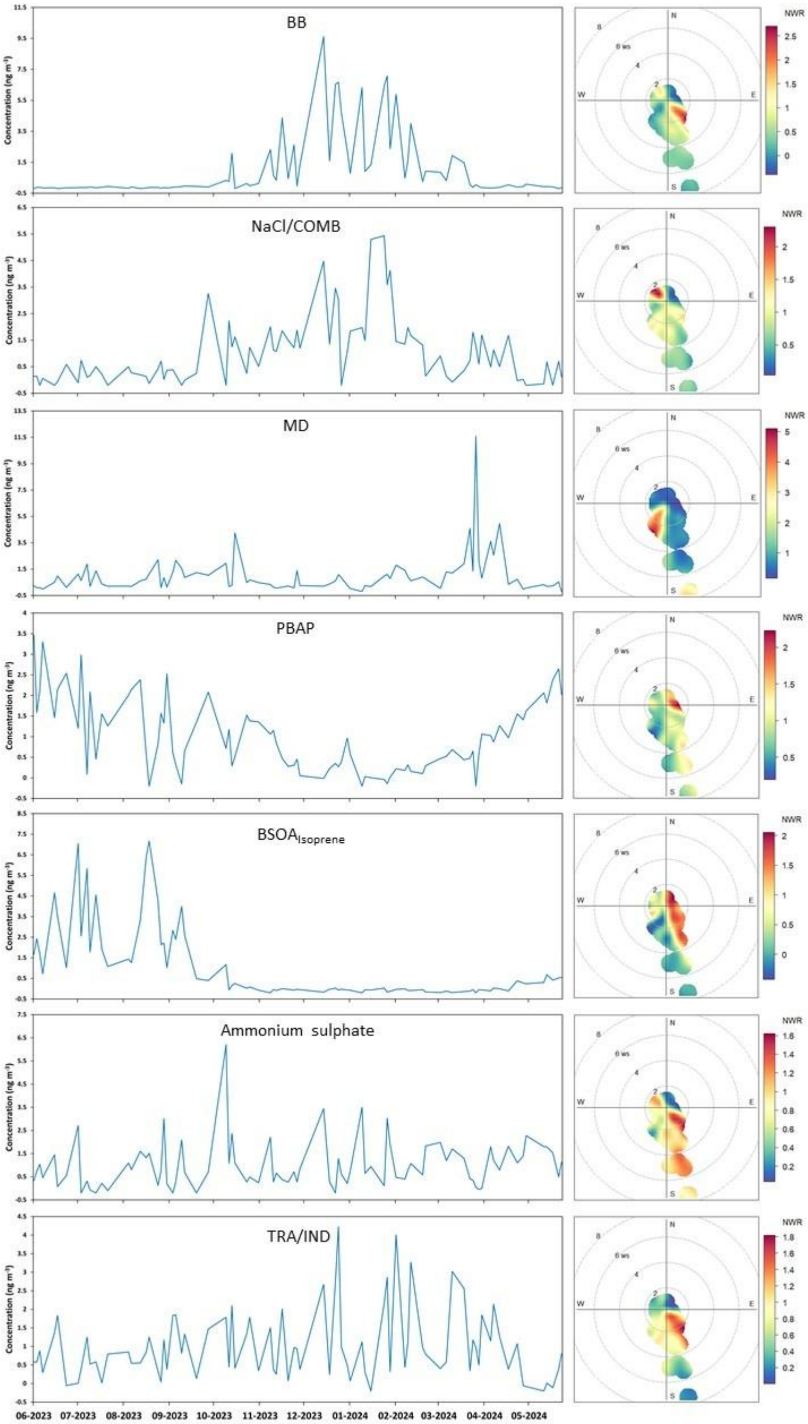

Figure 4. Daily factor contribution to PM$_{10}$ mass concentration during one-year campaign (left) and wind polar plots with normalized concentration of the apportioned PMF factors (right).

3.2.1 Biomass burning

Biomass burning (BB) factor contributed the most to the total PM_{10} concentration (21%, Fig. S2). It was characterized by the presence of galactosan (80% of total galactosan), 79% mannosan, 76% levoglucosan, 51% of K and Cl^-, 38% of Rb and 33% of OC (Fig. 3), typical indicators of BB. This factor also contains relatively high contribution of the As (28%), Cd (32%), Pb (21%). Unlike the species that are produced during incomplete combustion (like polycyclic aromatic hydrocarbons or black carbon), heavy metals emissions from BB are related to the biomass characteristic itself (e.g., natural metal accumulation from soil) (Yao et al. 2023). Metals may also originate from surface contamination from soil and dust or activities like wood treating (e.g., with arsenic-contained preservatives) (Stojić et al. 2016). Further examination of this factor included deriving OC/EC (9.4), OC/levoglucosan (8.3) and EC/levoglucosan ratios (0.88) which were in accordance with the values found in the literature (Table 2). Higher contribution of BB factor was observed during heating season in many European cities due to increased heating demand and stable atmospheric conditions in colder seasons (Liu et al. 2025). During cold seasons, as a result of near-surface cooling effect, temperature inversions occur, and stable boundary layer is formed which results in accumulation of the pollutants and prevention of their vertical mixing (Glojek et al. 2022). In our study, contribution of BB factor to total PM_{10} concentration during heating season was 34% and temporal variability with peak during heating season (from October till April) further confirmed this factor (Fig. 4). In urban areas, BB factor is associated with domestic heating (Belis et al. 2013). In Belgrade, it is estimated that around 300 000 households are not connected to district heating, and they mostly rely on wood as heating fuel (Belgrade's air quality action plan, 2021). This indicates that residential wood burning substantially influence local air quality.

Biomass burning was a commonly identified source in Belgrade, and Balkan region as well. In the study of Stojić et al. 2016, the contribution of solid fuel burning was almost 30% of PM_{10} in 2010-2011 in Belgrade. However, comparison with this study is difficult since sampling was conducted 13 years before our campaign and not all tracers needed for the complete PMF were included. The source apportionment of PM_{10} in Sofia, Bulgaria resolved biomass burning factor with contribution of 23% (Hristova et al. 2020) while source apportionment study in urban area of Skopje, Macedonia, resulted in much higher contribution of BB (around 70%) (Mirakovski et al. 2020). In case of $PM_{2.5}$, contribution of BB factor was 24% of total $PM_{2.5}$ in Budapest, Hungary and 28% in Zagreb, Croatia (Perrone et al. 2018).

Table 2. OC/levoglucosan, EC/levoglucosan and OC/EC ratios from PMF BB factor, dataset and other European studies.

	BB factor (PMF)	Gilardoni et al. (2011)¹⁾	Zotter et al. (2014)²⁾
OC/EC	9.4	6.54 ± 0.25	8.6 ± 2.9
OC/ Levoglucosan	8.3	5.62 ± 0.3	7.8 ± 2.7
EC/ Levoglucosan	0.88	0.89 ± 0.06	0.87 ± 0.27

1) Po Valley; 2) South of the Alps;

According to (Rowell et al. 2012), wood biomass is grouped into two groups: softwood and hardwood. Levoglucosan to mannosan (L/M) ratio may be used to apportion biomass burning emissions to burning from hardwood (14-15) or softwood (<4) (Schmidl et al. 2008). The L/M ratio derived from our PMF factor was 8.9, indicating probably a mixed contribution from both soft and hardwood burning, which is further confirmed with spruce (softwood) percentage of 53%. As shown in Fig. S3, the L/M ratios were different between summer (18.9) and winter (8.0) season suggesting possible origin of levoglucosan from other sources rather than burning of biomass for heating only. Ratios in spring and fall were consistent, 12.7 and 12.3, respectively. On the 2nd of October, increases in levoglucosan, mannosan and galactosan concentrations were observed. The heating season in Belgrade officially begins on October 15th and if unusually low temperatures occur earlier, the season may start sooner. However, on 2nd of October, the ambient temperature was 15°C which indicates another possible source of anhydrosugars beside burning of biomass for heating. (Gajović and Todorović, 2013) observed open fire phenomena in Serbia every year in October in the period from 2000 to 2013. The authors attributed this phenomenon to burning of agricultural waste in the autumn. During WeBaSOOP campaign, on the October 2nd, satellite data showed fire occurrence in Vojvodina (Fig. S4), a common region where agricultural fires were distributed (Gajović and Todorović, 2013). The same sudden increase in biomass burning tracers' concentrations was observed in Belgrade in 2008 in study of (Zangrando et al. 2016), which was also attributed to burning of agricultural waste. In their study, L/M ratio in October was in range from 2.5 – 16.7, which agrees with our L/M average ratio in October (14.9). Beside agricultural waste burning, the high L/M ratio in summer may also be explained by wood combustion (e.g. charcoal) in restaurants located in the Ada Ciganlija recreational area in the vicinity of Ada Marina monitoring site. The wind polar plots indicate strongest influence of air masses coming from the West in non-heating season, which coincides with direction of recreational area. Northeast, East, and Southeast wind direction may be observed during heating season, indicating influence from residential area (Fig. S5), thus supporting this interpretation.

3.2.2 Long-range transport and road-salt/local combustion

The next factor was a mix of NaCl from long-range transport and local emission from road salting with contribution of local combustion. It accounts for 9% of PM_{10}. The factor is dominated by Na (61% of total Na), NO_3^- (51%), and Cl^- (30%). As NaCl travels toward the Balkans, it is likely reacting with HNO_3, forming $NaNO_3$ and releasing HCl that is a typical aging process for sea salt aerosol (Pandolfi et al. 2020). Although Na-Cl high Pearson correlation coefficient (0.75) (Table S5) and Mg/Na (0.14) ratio (Seinfield and Pandis, 2016) confirm marine origin, K/Na (0.22) and Ca/Na (0.18) ratios (Ooki et al. 2002) from PMF output do not comply with ratios typical for marine aerosol and thus indicate mixing with other salt sources. Extensive road salting on 20th of January contributed to the pronounced peak on that day (https://nsuzivo.rs/srbija/na-auto-putevima-celu-noc-99-kamiona-prosuto-100-tona-soli-vesic-na-terenu-cak-200-radnika-veceras-cisti-saobracajnice/).

A slight seasonality and peaks during the heating season (Fig. 4) point to local sources of salt from road mixed with fossil fuel and biomass combustion. This factor accounts for 13% of EC, 11% of levoglucosan, and 9% of OC. Wind rose data (Fig. 4) show the dominant influence of air masses from the Northwest, aligning with the location of the "Novi Beograd" heating plant. Belgrade's district heating system covers 360,000 households and 4.4 million m² of commercial space, with 90% of its thermal energy derived from natural gas (https://beoelektrane.co.rs/zastita-zivotne-sredine/). Although natural gas emits less particulate matter than other fuels, it remains a potential source of PM. The influence of nearby plants is supported by wind patterns corresponding to the positions of the Novi Beograd (Northwest) and, to a lesser extent, "Banovo Brdo" (Southeast) heating plants. Additional fuels used by Belgrade Power Plants include CNG, light heating oil, low- and medium-sulphur heating oil, coal, and pellets which may also contribute to elevated PM_{10} levels during the heating season.

Flexpart footprint analysis (Fig. S6) for the two peaks in the time series of this factor shows air masses from the Atlantic and Northern Sea (Fig. S6-1) but also local influence of combustion on the 19th December which is the day with the highest concentration in the whole WeBaSOOP campaign (Fig. S6-2). The NaCl/COMB factor contributed the least to PM_{10} mass concentration, 9%. Similar findings were found in an urban background site in Northwest Germany, located 250 km from the coast. The aged sea salt factor in this study was explained mainly by Na, NO_3^-, Mg and OM and contributed to the PM_{10} mass same as our factor, 9% (Beuck et al. 2011). Although located around 300 km away from closest sea, other source apportionment studies in Belgrade observed the influence of marine aerosol also. In the study of

(Đorđević et al. 2012), marine aerosol was confirmed by the presence of Na and Cl in the coarse mode of measured particles.

3.2.3 Mineral dust

The mineral dust (MD) factor accounts for 17% of PM_{10} and is characterized by high contributions from crustal elements: Al (78% of total Al), Ti (69%), Sr (62%), Mg (60%), and Ca (51%), which are typical markers of soil and mineral matter. Moderate enrichments of V (30%), Mn (29%), and Fe (20%) are also observed, although less pronounced than in the TRA/IND factor. Diagnostic ratio of K/Fe (0.8) complies with typical ratio in dust (Liu et al. 2022). Wind polar plots confirm the strongest influence of air masses coming from the Southwest (Fig. 4). Moreover, on 31st March 2024, high PM_{10} concentration (48.5 $\mu g/m^3$), partially attributed to a Saharan dust outbreak, as confirmed by characteristic Na/Al (0.14) and K/Al (0.27) ratios, closely matching values reported for Saharan dust (Na/Al = 0.12; K/Al = 0.31) by (Moreno et al. 2006). (Nicolas et al. 2008) observed that Saharan outbreaks are marked by a more pronounced increase in Ti relative to Si and Ca. Therefore, Ti/Ca ratio is good marker for Saharan dust episodes. In our study, Ti/Ca (0.05) and Ti/Fe (0.14) ratios were both consistent with the values from the literature (Ti/Ca = 0.02; Ti/Fe = 0.10) (Nicolas et al. 2008) and aligned with ratios obtained in Debrecen, Hungary (Ti/Ca = 0.07; Ti/Fe = 0.09) (Borbely-Kiss et al. 2004). Although these observations were made during the March to April 2024 transition, no clear seasonal pattern was evident (Fig. 4).

Beside the long-range transported dust, the influence of local dust was also observed. The WeBaSOOP campaign coincided with preparatory work for the starting station of Belgrade metro. Sand and crushed stone were applied around the future depot site in Makiš, roughly 7 km Southwest of the Ada Marina site. This proximity is reflected in the mineral dust polar plot, which shows a joint influence of construction activity and natural mineral dust (Fig. S8).

The MD factor accounts for 17% of PM_{10}, but only 5% to OC was attributed to MD, indicating no mixing of dust and organic matter by aging and thus making it the lowest contributor to OC among all identified factors. In the study of (Mijić et al. 2012), mineral matter contributed to PM_{10} with 19%, with the most dominant southwest wind, which agrees with our results and indicates mineral dust constant influence in Belgrade. Our results complied with other European cities where contribution of MD factor accounted for 10% in Milan, Italy to 25% in Athens, Greece (Amato et al. 2016).

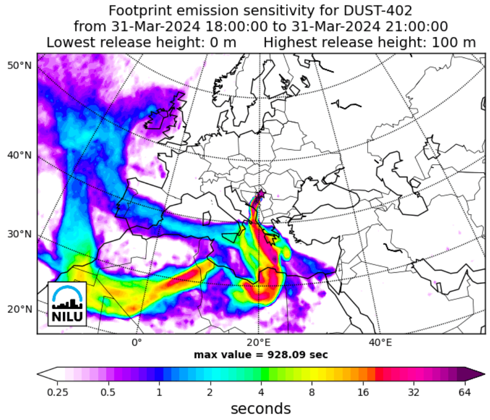

Figure 5. FLEXPART Footprint analysis for 31st of March confirming influence of Saharan dust on that day.

3.2.4 Primary biological aerosol particles

Primary biological aerosol particles (PBAP) factor contributes to 10% of PM_{10} and it was traced by sugars (Glucose, Fructose, Trehalose) and sugar alcohols (Mannitol and Arabitol). It accounted for 72% of total glucose, 68% arabitol, 63% trehalose, 62% mannitol, and 57% fructose. The remaining sugars and sugar alcohols were apportioned by PMF in the $BSOA_{Isoprene}$ factor. This factor shows a clear seasonal variation with higher levels during warmer seasons (Fig. 4) due to increased solar radiation and higher surface temperatures (Waked et al. 2014). PBAP factor is indicator of a highly heterogenic source of particles coming from bacterial and fungal cells or spores, their fragments (e.g. endotoxins, mycotoxins), viruses, pollen grains, and fragments of plants and insects (Barbaro et al. 2024; Samake et al. 2019). Mannitol and arabitol are established tracers of fungal spores while glucose is being used as tracer for plant material like pollen and fragments of plant material (Tonon et al. 2017, Samake et al. 2019). In our study, fungal spores were estimated using mannitol to arabitol ratio (M/A) (Bauer et al. 2008). The M/A ratio (1.0) derived from the PBAP factor and dataset, aligns closely with values reported for fungal spores (Bauer et al., 2008) and for aerosol filter samples collected across Europe (Yttri et al., 2011, 2024; Samake et al., 2019) (Table 3). Additionally, by applying specific OC to mannitol ratio of 5.2 and 10.8 to our dataset (Bauer et al. 2008), we estimated that 21-44% of OC_{PBAP} comes from fungal spores' source (Platt et al. In preparation). Further resolving this heterogeneous group and estimating its contribution to the total PBAP factor remains challenging, as a broader range of primary biological organic tracers has yet to be examined (Samake et al. 2019). The polar wind diagram shows a Northeasterly influence, likely related to the vegetation zones of the Ada Ciganlija recreational area near Ada Marina. A slight Southeasterly influence is also observed, where the Košutnjak forest is located, 3 km away and covering an area of 300 ha (Fig. S9). Borlaza et al. 2021 indicated that the influence of this source is more local than large-scale impact, complying with our conclusion that biggest influence of this factor is from vegetation at Ada Ciganlija.

The PBAP factor accounts for 10% of PM_{10}, 14% to OC and 11% to EC which was less expected result. However, in the study of Waked in Lens, France, contribution of EC to PBAP factor was 16% on average annual level. Such results were obtained due to possible atmospheric mixing process, limitations in PMF modelling or OC/EC analysis artifacts. Possible explanation for this may be that some types of PBAP, char and evolve as a modern carbon EC during OC/EC analysis (Yttri et al. 2020). Similarly to our study, PBAP contributions at two urban sites in France were 9% Lens (Waked et al. 2014) and 11% in Grenoble, (Srivastava et al. 2018). Slightly higher contributions (16%) were measured in a peri-urban area 25km away from Rome, Italy (Marcovecchio and Perrino, 2021).

Table 3. Mannitol to arabitol ratios for PBAP factor from both PMF and dataset compared with other studies in Europe.

	PBAP factor (PMF)	Ratio from Belgrade dataset	Bauer et al. (2008)$^{1)}$	Yttri et al. (2011)$^{2)}$	Yttri et al. (2024)$^{3)}$	Samake et al. (2019)$^{4)}$
Mannitol/Arabitol	1.0	1.0 ± 0.3	1.5 ± 0.5	1.4 ± 0.3; 1.1 ± 0.3	1.1 ± 0.5; 1.0 ± 0.0	1.15 ± 0.59

1) Vienna, Austria; 2) Oslo, Norway, summer and winter season, respectively; 3) Zeppelin Observatory, Birkenes Observatory, respectively; 4) France

3.2.5 Biogenic secondary organic aerosol from isoprene

This factor represents biogenic secondary organic aerosol formed from isoprene oxidation ($BSOA_{Isoprene}$), a major volatile organic compound emitted from land vegetation (Van Drooge and Grimalt, 2015). It accounts for 10% of PM_{10} (Fig. 3) and is characterized by the presence of 2-methylerythritol (84% of total 2-methylerythritol) and 2-methylthreitol

(76%), both known isoprene oxidation products (Claeys et al., 2004). A modest contribution from PBAP tracers (17–28% of their total content) likely reflects co-varying biological activity, as these two factors exhibit similar temporal patterns (Fig. 4), although these species do not define this factor. Liu et al. (2025) emphasised that including secondary biogenic organic aerosol tracers, such as 2-methyltetrols or pinic acid and 3-MBTCA (markers for α-pinene oxidation), in PMF analysis led to the separation of a substantial OC fraction in the form of biogenic secondary organic aerosol from a sulphate-rich factor. A similar split was observed in our study. This is supported by the co-occurrence of SO_4^{2-} (24%) and NH_4^+ (22%), present as NH_4HSO_4 and/or $(NH_4)_2SO_4$, suggesting that 2-methyltetrols are formed via acid-catalysed aqueous-phase reactions, confirming the secondary origin of this factor. Wind polar plot shows influence from East, and both North and Southeast, indicating influence from the nearby vegetational sources as well as influence from Košutnjak forest, even higher than for PBAP factor (Fig. 4). The $BSOA_{Isoprene}$ factor appears exclusively in the growing season, peaking in summer. It serves as a clear proxy for isoprene-derived SOA formed under conditions of high photochemical activity and enhanced biogenic emissions. It contributes 10% to the PM_{10} mass concentration and 11% to OC. Similarly to our result, average annual contribution to PM_{10} of biogenic secondary organic aerosol was in range from 8 % - 11 % in the study of (Borlaza et al. 2021) in Grenoble, France. The similar contribution to OC of 10 % was observed in Norway (Yttri et al. 2020). To date, to our knowledge, no source apportionment study including tracers for secondary biogenic aerosol particles has been conducted in Belgrade, except to other WeBaSOOP campaign papers.

3.2.6 Ammonium sulphate

This ammonium sulphate is a secondary factor accounting for 18% of PM_{10} and represents the long-range transport of aged air masses and secondary aerosol formation. It is dominated by NH_4^+ (55% of total NH_4^+) and SO_4^{2-} (54%), with the coexistence of NH_4HSO_4 and $(NH_4)_2SO_4$ indicating only partial neutralization of H_2SO_4. An elevated NO_3^- fraction (17%) further supports its secondary origin. Trace contributions from As (18%), Cd (14%), and levoglucosan (5%) point to minor influences from industrial and combustion sources, likely associated with long-range transport. A high OC/EC ratio (9.1) corroborates the secondary and aged nature of this factor.

The occurrence of NH_4HSO_4 and $(NH_4)_2SO_4$ in both, this and in the $BSOA_{Isoprene}$ factor likely reflects a common regional background that persists year-round but co-varies with distinct seasonal processes. In summer, partially neutralized sulfuric acid acts as an acid seed facilitating IEPOX (Isoprene Epoxydiol) uptake and subsequent 2-methyltetrol formation. During other seasons, it primarily represents regional SIA. The $(NH_4)_2SO_4$ factor accounts for 18% of PM_{10} and 12% of OC, making it the second largest contributor to PM_{10} after the BB factor. The contribution of this factor is similar to the ones previously reported for various urban sites in Europe (Beuck et al. 2011; Borlaza et al. 2021) as well as in Belgrade. (Đorđevic et al. 2012) found that the formation of $(NH_4)_2SO_4$ has the dominant influence on the contents of aerosol ionic species in Belgrade in 2008. (Belis et al. 2019) indicated that secondary sulphate in the Danube and Western Balkan region is mostly associated with coal combustion since its precursor is gas SO_2. Wind polar plots showed highest influence from the Southeast and South. The "Kolubara" thermal power plant is located around 50 km South of Belgrade, and "Kostolac" is approximately 60 km East. Peak on 14th October showed HYSPLIT backward trajectories from the Southwest and influence on that day may be attributed to operation of “Nikola Tesla A” thermal power plant, located around 25 km way from Belgrade (Fig. S10). Our results complied with results obtained previously in Belgrade. In the study of (Stojić et al. 2016), secondary aerosol contributed to PM_{10} 19.5%, while contribution of 16% was obtained in Sofia, Bulgaria (Hristova et al. 2020). Our results are withing the European range too, 15% to 26% (Liu et al. 2025).

3.2.7 Traffic/industry

This Traffic/Industry factor (TRA/IND) is likely a mixture of tail pipe and non-tail pipe emissions with influence of industrial activity based on presence of As, Cd, Pb and V. It accounts for 15% of PM_{10} and it is characterized by EC (41% of total EC) and a broad suite range of trace metals, notably Fe (64%), Sb (60%), Cu (57%), Sn (57%), Mn (51%), Pb (37%), V (35%), Cd (32%), and As (25%). Fe, Sb, Sn and Cu are typical non-exhaust tracers (e.g., from brake wear), V relates to heavy oil combustion and As, Cd and Pb are associated with coal combustion and metallurgic activities (Yttri et al. 2020; Pandolfi et al. 2016). The enrichment of Ca (34%) and Mg (22%) suggests road dust resuspension and influence of construction activity. Therefore, this factor is considered as a mix of traffic exhaust and non-exhaust emissions and industrial sources. Low contributions from levoglucosan (6 %), 2-methyltetrols (< 0.5 %), and sugars and sugar alcohols (< 5%) confirm a minor to negligible influence by biomass burning, biogenic SOA, and PBAP. SIA species are moderate for NO_3^- (12%), minor for SO_4^{2-} (2%) and absent for NH_4^+, consistent with limited regional influence. This factor does not change throughout a year. The TRA/IND factor contributed 15 % of PM_{10} and 17% of OC, making it the second largest contributor to OC. The OC/EC ratio of 2.5 is higher than the typical OC/EC traffic ratio (1.4) found in literature (Pio et al. 2011), which indicates the contribution of other combustion sources to this factor (probably SOA mixed with primary emissions). Since the PMF model results are based on internal correlations among species that have similar time series, it is difficult to differentiate sources that do not vary independently. The limitation of 24h sampling methodology is that it couldn't capture the diurnal variations of traffic source, therefore, some primary and secondary sources are not separated (Waked et al. 2014).

Traffic with metal industry influence was also observed in Rijeka, Croatia and accounted for 11 % of total PM_{10} mass concentration (Mifka et al. 2021). In other countries of the Balkan region, traffic contributed 9% in Sofia, Bulgaria, and 18.9% in Istanbul, Turkey, with slight seasonality observed in winter (Hristova et al. 2020; Kocak et al. 2011). Our results also complied with range observed in other European urban background sites (5-25%) (Srivastava et al. 2018).

4. Conclusion

This study presents the chemical composition and source apportionment of PM_{10} mass concentrations during the WeBaSOOP campaign (June 2023 to May 2024). Although average PM_{10} concentration complied with current EU and national legislation, 23.9 $\mu g\ m^{-3}$, it exceeded the limit value proposed by the new EU 2024/2881 AQ Directive on ambient air quality and cleaner air for Europe, to be in force from 2030, and the WHO AQ Guidelines updated in 2021. By the application of mass reconstruction model it was estimated that carbonaceous fraction (OM + EC) constituted the dominant PM component (54% on annual average), followed by secondary inorganic aerosol (19%) and mineral dust (13%). Source apportionment using USEPA PMF 5.0 resolved seven factors contributing to PM_{10} as follows: biomass burning (21%), ammonium sulphate (18%), mineral dust (17%), traffic/industry (15%), primary biological aerosol particles (10%), biogenic secondary organic aerosol from isoprene (10%), long-range transport and road salt/local combustion (9%). Evaluation of ratios of PM elements and species, back trajectory analysis and wind polar plots verified the local and regional sources contributing to PM_{10} in Belgrade. Compatibility of the two approaches, mass reconstruction and SA, indicated that more than 40% of PM_{10} mass was related to different combustion sources, between 30%-35% may be devoted to mineral dusts and inorganic aerosols of anthropogenic and natural sources, while specific organic tracers proved that about 20% of PM_{10} mass originated from biological sources. Seasonal patterns varied among factors. Biomass burning and the long-range transport and road salt/local combustion factors exhibited elevated levels during heating season. The biomass burning driven primarily by residential wood burning and the long-range transport and road salt/local combustion reflecting contributions from local sources during winter period of road salt events and local combustion. In contrast, primary biological aerosol particles and biogenic secondary organic aerosol from isoprene peaked during the non-heating season, coinciding with increased biological activity. Mineral dust, ammonium sulphate, and traffic/industry showed no clear seasonal variability. Ammonium sulphate was resolved as a distinct factor, but its influence was also evident in the $BSOA_{Isoprene}$ factor, supporting the secondary origin of this aerosol and its likely role in facilitating isoprene oxidation. Traffic factor has more enhanced diurnal variation than seasonal which cannot be observed with resolution of 24 hours sampling. Therefore, time series of traffic and industry tracers were not different enough to separate them which is a limitation of this 24h sampling resolution.

Incorporation of specific organic tracers in PMF enabled the identification of biogenic activity-related factors and provided deeper insight into the composition and sources of PM_{10} in the Belgrade Metropolitan area. Our results showed influence of both, local (biomass burning, traffic) and regional sources (mineral dust, salt from long-range transport) on PM_{10} concentration in this area. Continuous measurements and source apportionment are needed in data-scarce areas of Europe to better understand source impacts on air quality. Therefore, such findings may offer a scientific basis for air quality management and support policy makers in identifying priority sectors and targeted measures to mitigate PM_{10} pollution. However, further work for better understanding of PM_{10} sources in Belgrade is needed, including combination of offline and online PMF analysis and long-term measurements.

Data availability

All data on OC, EC, ions, elements and organic tracers are available at EBAS (https://ebas.nilu.no/).

Author contributions

Conceptualization: MJS, AA, KEY. Data curation: BP, BR, RK, MD. Formal analysis: BP, AA, KEY, AB, MJS, DS, MP. Funding acquisition: MJS. Investigation: BP, MJ, BR, RK, DS, MD, MJS. Project administration: MJS. Supervision: MJS, AB, AA. Validation: AA, KEY, MJS, MP, SMP. Visualization: BP. Writing (original draft preparation): BP. Writing (review and editing): BP, MJS, AA, MP, DS, SMP.

Competing interests

The authors declare that they have no conflict of interest.

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