



Air quality challenges in Central Asian urban areas: a PM_{2.5} source apportionment analysis in Dushanbe, Tajikistan

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Abstract

This work presents the first comprehensive assessment of PM pollution sources in Dushanbe, Tajikistan. A total of 138 PM_{2.5} samples were collected during 2015–2016 and 2018–2019 and were analyzed through gravimetric, ED-XRF, and multi-wavelength absorption techniques. The results show that PM_{2.5} concentrations were substantially higher than the European annual limit value and WHO Air Quality Guidelines annual average value, with an average of $90.9 \pm 68.5 \mu\text{g m}^{-3}$. The PMF application identified eight sources of pollution that influenced PM_{2.5} concentration levels in the area. Coal burning (21.3%) and biomass burning (22.3%) were the dominant sources during the winter, while vehicular traffic (7.7%) contributed more during the warm season. Power plant emissions (17.5%) showed enhanced contributions during the warm months, likely due to high energy demand. Cement industry emissions (6.9%) exhibited significant contribution during the cold period of 2018–2019, while soil dust (11.3%) and secondary sulphates (11.5%) displayed increased contribution during the warm and cold months, respectively. Finally, waste burning (1.5%) displayed the lowest contribution, with no significant temporal variation. Our results highlight the significant impact of anthropogenic activities, and especially the use of coal burning for energy production (both in power plants and for residential heating), and the significant contribution of biomass burning during both warm and cold seasons.

Keywords Source apportionment · PMF · PM_{2.5} · Tajikistan · Central Asia

Introduction

Many cities and metropolitan areas around the world are experiencing rapid growth due to the expansion of the urban population, industrialization, and intensive human activities. This can lead to increased energy consumption and combustion emissions, which contribute to the deterioration of air quality and human health.

Suspended particulate matter (PM) is one of the most important atmospheric pollutants, originating from both natural and anthropogenic sources; many studies have demonstrated the detrimental effects on the environment, human health (Samoli et al. 2013; Katsouyanni et al. 2001; Beelen et al. 2015), and climate change (Chen et al. 2021; Tai et al. 2010; Yang et al. 2022). Atmospheric particles consist of many different chemical components, depending on their sources. Among the major anthropogenic sources contributing to PM concentrations levels are traffic, biomass burning, industry, and coal burning, while natural sources include local dust resuspension, long-range dust transport events and

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sea salt (Manousakas et al. 2018; Diapouli et al. 2017). In addition to primary sources, PM concentration levels may be impacted by secondary aerosol formation (e.g., secondary inorganic and organic aerosol).

The process of identifying air pollution sources and quantifying their contribution to pollutants' concentration levels is an important step in managing air quality, as it may assist towards developing targeted effective air pollution mitigation strategies. Positive matrix factorization (PMF) is a mathematical receptor modeling tool that has been used widely for source apportionment analysis (Belis et al. 2020; Gunchin et al. 2019; Hopke 2016). PMF aims to quantify the contribution of the major aerosol sources to the PM concentration levels recorded at a receptor site, based on the PM chemical composition and its temporal variability.

The escalating issue of the poor air quality across Central Asia is raising significant concerns among researchers and the general populace alike. A study in Almaty, Kazakhstan (Kerimray et al. 2020), reveals increasing trends in PM concentrations from 2013 to 2017. Winter peaks highlight coal combustion's significant contribution. Coal-fired power plants are a potential culprit, highlighting the need for stricter regulations and cleaner energy sources. Bishkek, Kyrgyzstan, faces similar struggles (Dzushupov et al. 2022). Over 40% of the urban population relies on poor quality coal for heating, resulting in deteriorating air quality throughout the city. Traffic is another significant contributor to PM pollution, with old vehicles running on low-quality fuel dominating the streets, intensified by insufficient public transport options. Air quality challenges are evident in Uzbekistan as well (Salomova et al. 2022). While industrial activity has decreased, a rise in car ownership has led to increased traffic emissions. Dust storms from surrounding deserts further complicate the issue along with landfill burning, particularly during the summer. Despite some progress in implementing cleaner technologies and waste management, air pollution remains a serious public concern.

Tajikistan also faces similar struggles since it has grown significantly in recent decades in terms of population, industrialization, and urbanization, which has led to declining air quality, due to increased anthropogenic emissions, such as biomass and coal burning (Chen et al. 2022). Tajikistan is also affected by frequent dust transport events, which further contribute to the increase of PM in the atmosphere and air quality deterioration (Abdullaev and Sokolik 2019; Hofer et al. 2017). In the study conducted by Tursumbayeva et al. (2023), it is highlighted that Central Asian countries, including Tajikistan's capital, Dushanbe, rank very high in IQAir's air quality index (IQAir 2022). The researchers also highlight that, when it comes to research publications, the number is significantly lower compared to other countries worldwide.

Although a few studies have investigated crustal and organic aerosols in the region (Chen et al. 2022; Abdullaev and Sokolik 2019), it is important to note the scarcity of scientific literature addressing the source apportionment of PM pollution in Central Asian countries. In fact, only one study provided insight into PM_{2.5} source apportionment across multiple countries, including Dushanbe (Tajikistan) and Kurchatov (Kazakhstan) (Almeida et al. 2020); nevertheless, this study did not specifically delve into the unique characteristics and PM sources in the different countries, while no data on carbonaceous species was available for Central Asian countries.

In this framework, the present work aims to cover the need for a better understanding of the air quality challenges Central Asian urban centers face. For this purpose, two measuring campaigns were performed during 2015–2016 and 2018–2019 in order to characterize PM_{2.5} concentration levels and major chemical components. The chemical composition database was used for the application of PMF, in order to identify the major PM sources and to quantify their contribution to PM_{2.5} concentrations. The importance of research on Dushanbe's air quality extends beyond the city itself. Central Asian nations share many of the same air quality challenges, and findings from Dushanbe can contribute knowledge to other urban centers in the region. By understanding the unique sources and characteristics of PM_{2.5} in Dushanbe, policymakers can develop more effective strategies for tackling PM air pollution across Central Asia, leading to improved public health outcomes for millions of people. It should be noted that this is the first source apportionment study in Dushanbe that includes data on carbonaceous aerosol, a critical parameter for the identification of combustion sources.

Experimental

Sampling and study area

Two PM_{2.5} sampling campaigns were carried out in Dushanbe, Tajikistan (38° 33'34"N, 68° 51'23"E, elevation 867 m; Fig. 1), specifically at an urban background site situated on a hill within the campus of the Physical-Technical Institute of the Academy of Sciences of Tajikistan (Hofer et al. 2017). The sampling site, located about 7.5 km away from the city center of Dushanbe, is surrounded by residential buildings. It should be noted that the city is characterized by significant industrial activities, including a cement plant OJSC "Tochikcement" (Location C) and a large 400-Mega-watt coal-fired power station (Location B), with the addition of small-sized heating enterprises and construction sites (Fig. 1). Moreover, in close proximity to the sampling area,

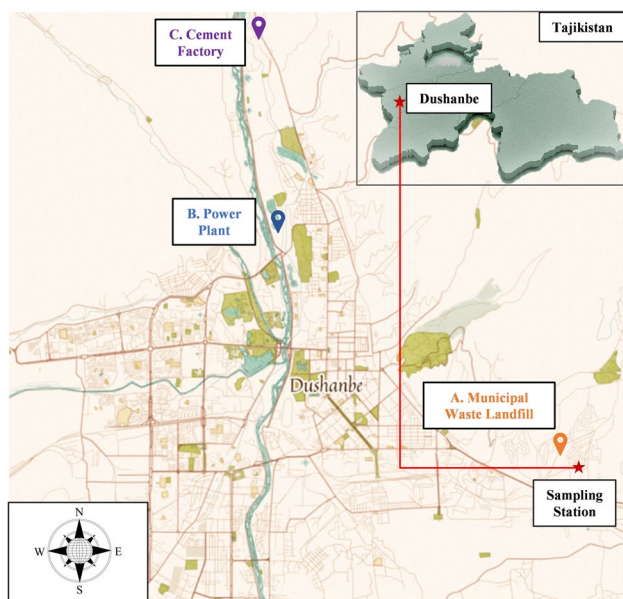


Fig. 1 Study area of Dushanbe

there is a municipal waste landfill (Location A), situated approximately 850 m away.

The campaigns were conducted during two periods, from August 2015 to May 2016, and from October 2018 to February 2019, including both warm and cold seasons. Specifically, in the 2015–2016 period, the warm period extended from August 2015 to Mid-October 2015 and from Mid-April 2016 to the end of May 2016. Conversely, the cold period of 2015–2016 spanned from the second half of October 2015 to mid-April 2016. Similarly, in the 2018–2019 period, the cold period extended from October 2018 to February 2019.

Sampling was performed with the low volume sampler MVS6D D-10623 (Berlin, Germany Kleinfiltergerät). 24 h $PM_{2.5}$ samples were collected onto Teflon membrane filters (PTFE Whatman), 47 mm in diameter, with 1 μm pore size. On average, 9 samples were collected per month, leading to a total of 138 filters, with 88 filters obtained during the 2015–2016 period and 50 filters collected during the 2018–2019 period. The filters were used for the gravimetric determination of $PM_{2.5}$ mass concentration, as well as for PM chemical speciation.

PM chemical characterization

Elemental analysis

$PM_{2.5}$ samples were analyzed for major and trace elements using the high-resolution energy dispersive X-ray fluorescence 3-D optics spectrometer Epsilon 5 (PANanalytical). The spectrometer consists of a side-window low power X-ray tube with a W/Sc anode. The characteristic X-ray radiation emitted by the sample is detected by a Ge detector

Table 1 LOD and uncertainties for each analyzed element with ED-XRF

El	LOD (ng/m ³)	Unc%	El	LOD (ng/m ³)	Unc%
Na	14.3	24	Mn	1.1	16
Mg	11.8	22	Fe	0.9	11
Al	11.8	16	Co	1.2	20
Si	13.5	24	Ni	0.5	12
S	3.0	11	Cu	0.5	11
Cl	1.3	17	Zn	0.6	11
K	0.5	11	Br	1.3	14
Ca	2.3	11	Rb	1.4	14
Ti	0.9	11	Sr	1.0	18
V	0.7	12	Ba	11.9	18
Cr	0.4	12	Pb	2.9	18

with a measured energy resolution of approximately 150 eV FWHM at Mn-K α (5.89 keV). The spectrometer provides a selection of 9 secondary targets (Al, CaF₂, Fe, Ge, Zr, Mo, KBr, Al₂O₃, LaB₆) that can polarize the X-ray beam, and therefore 9 measuring conditions were selected for the optimal analysis of the aerosol samples.

For the calibration of the XRF spectrometer, we utilized infinitely thin, single-element, compound, or multi-elemental standards. Specifically, we employed various reference materials including 27 Micromatter thin reference materials deposited on 6.3 μm Mylar (such as NaCl, MgF₂, GaP, SiO, KCl, CaF₂, V, Fe, Cr, Co, CuS_x, Al, Ni, CsBr, RbI, SrF₂, Ge, Ag, Sn, Sb, Pt, AgHg, CdSe, Pb, Au, BaF₂, and Ce), 7 custom-made on Kapton (including Ti, Cr, Mn \times 2, Co, Cu, and BaF₂), and 8 multi-standards on filter media (such as SRM 2783 and seven custom-made). The custom-made standards on filter media were devised due to the inadequate availability of certified APM materials on membrane filters covering a suitable range of concentrations. Consequently, CRMs 2584 and 2583 (indoor dust) were dispersed and collected on membrane filters (PTFE filters with PMP support ring and PTFE filters without support ring), with the dust particles of the reference material being rendered airborne in dry form using a 220 Topas Aerosol Generator powered by compressed pure N₂. The uncertainty associated with Micromatter standards is specified at 5% while for multi-element standards, is set at 10%.

All measurements were conducted under vacuum, and the total analysis time per sample was about 90 min. The methodology that was used for the elemental characterization is explained in detail in Manousakas et al. (2018). Twenty-two elements were determined by the ED-XRF method, namely Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, Rb, Sr, Ba, and Pb. The detection limits (LOD) (ng/m³) and the corresponding uncertainties (Unc) (%) are given in Table 1.

Black carbon analysis

PM_{2.5} samples were measured by the Multi-wavelength Absorption Black carbon Instrument (MABI) (ANSTO) for the determination of their black carbon (BC) content. MABI measures the light transmission decay caused by the particles collected on a filter sample, at seven different wavelengths (405, 465, 525, 639, 870, 940, and 1050 nm).

While data from all seven wavelengths were considered, the choice of 870 nm was the prevailing choice. At 870 nm, black carbon exhibits significant absorption properties with the additional benefit of minimizing interference from other components such as brown carbon (Olson et al. 2015; Zhang et al. 2019; Liakakou et al. 2020). Secondly, the selection aligns with the operational wavelength (880 nm) of many aethalometers, which are commonly used instruments for BC monitoring (Backman et al. 2017). The detailed methodology for the calculation of BC concentrations using MABI is explained elsewhere (Kebe et al. 2021).

Quality assurance/quality control procedure

The quality assurance/quality control (QA/QC) protocol for ED-XRF analysis incorporates various essential procedures for ensuring data quality and instrument performance. These procedures encompass the following: (a) weekly calibration of the Ge detector, where the XRF software automatically adjusts energy channels to maintain optimal performance; (b) routine assessment of each analyte performance against the multi-elemental reference material SRM 2783, with daily weekly analyses to validate analytical accuracy and precision; and (c) continuous evaluation of each analyte performance through the systematic analysis of selected PTFE blanks on a daily basis, aimed at assessing long-term reproducibility and consistency.

Similarly, for the Multi-wavelength Absorption Black carbon Instrument (MABI), the QA/QC procedures were the following: (a) The system's calibration is checked every four samples; (b) detector calibration is conducted before every batch of measurements to ensure consistent performance; and (c) regarding calibration, a visual representation is provided through a calibration chart. This chart indicates whether all measurements reside within specified tolerance levels. If the measurements meet the correct criteria, the calibration is accepted, and measurements on actual filters can proceed. If not, the calibration is rejected, and the process is repeated (Cohen et al. 2000).

Positive matrix factorization

Positive matrix factorization (PMF) is a receptor model which aims to identify the major PM sources, based on their chemical profiles, and quantify their contribution to

measured PM concentration levels (Paatero and Tapper 1994). The basic mass balance equation that is used by PMF can be expressed as follows:

$$X_{mn} = \sum_{p=1}^k g_{mp} \cdot f_{pn} + e_{mn} \quad (1)$$

where X is the concentrations of each species (n) measured in each sample (m), g is the contribution of each source (p) in each sample (m) generated by the model, f is the contribution of each species (n) to the chemical profile of each source (p) generated also by the model, and e the residual for species (n) in sample (m).

The goal of PMF is to solve Eq. (1) by minimizing the sum of squared residuals (E) between the measured and predicted concentrations until a stable solution (Q) is found (Paatero 1999):

$$Q = \sum_{m=1}^v \sum_{n=1}^{\mu} \frac{e_{mn}^2}{s_{mn}^2} \quad (2)$$

where s_{mn} is the uncertainty of each species (n) in each sample (m).

The EPA PMF 5.0 model was applied to the combined dataset from 2015 to 2016 and 2018 to 2019, which included in total 138 sampling days and 23 species (BC, Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, Rb, Sr, Ba, and Pb).

The expanded uncertainties (including sampling and analytical uncertainties) of all measured concentrations were calculated and included in the input database. Regarding the elemental analysis with X-Ray fluorescence, the current study considered several key factors contributing to measurement uncertainty, including uncertainties related to peak area, calibration, field operations, sampling, sample deposition, attenuation of characteristic X-rays from light elements, and the relative standard deviation of consecutive measurements (with a sample size of 3 for each sample). The total expanded uncertainty was calculated as the square root of the combined variances (Manousakas et al. 2018).

Regarding BC measurement by MABI, we considered specific factors affecting the measurements, including sample handling and instrument sensitivity. Given the challenges in precise BC quantification, we assigned a 15% expanded uncertainty to accommodate potential variability and errors.

Values below the limit of detection (LOD) were substituted with half of the LOD, and their uncertainties were set at 5/6 of the LOD. A 5% modeling uncertainty was introduced to factor in modeling inaccuracies. Three species were categorized as “weak” (Na, Mg and Ni), while PM was included as the total variable.

Results

PM_{2.5} chemical composition

The average PM_{2.5} concentration measured was $90.9 \pm 68.5 \mu\text{g m}^{-3}$ (Table 2), which greatly exceeds the EU annual limit value ($25 \mu\text{g m}^{-3}$) and the stricter WHO annual guideline ($10 \mu\text{g m}^{-3}$). The highest average concentrations were observed during October ($143.7 \mu\text{g m}^{-3}$) and November 2015 ($142.6 \mu\text{g m}^{-3}$). The lowest PM_{2.5} concentrations were recorded during April and May 2016, with average concentrations of 29.4 and $44.3 \mu\text{g m}^{-3}$ respectively. The monthly average PM_{2.5} concentrations are presented in Fig. 2. Higher PM concentration levels were recorded during 2015–2016, in comparison to 2018–2019. Specifically, the average PM_{2.5} concentration during 2015–2016 was $99.5 \pm 77.6 \mu\text{g m}^{-3}$, significantly higher than the average concentration of $72 \pm 41 \mu\text{g m}^{-3}$ observed during 2018–2019. This represents a significant difference of approximately 32% between the two sampling periods (at $p=0.01$). While there is limited data on PM pollution in Central Asian urban areas, it should be noted that Dushanbe ranks among the

Table 2 PM_{2.5} concentration (in $\mu\text{g m}^{-3}$) and composition (in ng m^{-3}) during the entire study

Species	Average	St. Dev	Min	Max
PM _{2.5}	90.0	68.5	18.1	417.1
BC	9100.0	6400.0	1700.0	44000.0
Na	2370.0	2830.0	160.0	16200.0
Mg	490.0	580.0	10.0	2100.0
Al	1930.0	1810.0	130.0	7800.0
Si	6620.0	6560.0	250.0	28100.0
S	1430.0	1070.0	400.0	6700.0
Cl	1370.0	1420.0	40.0	8100.0
K	1380.0	1090.0	290.0	6200.0
Ca	4870.0	6240.0	110.0	21900.0
Ti	150.0	200.0	4.0	870.0
V	4.0	3.0	0.7	30.0
Cr	1.0	2.0	0.4	10.0
Mn	40.0	60.0	1.0	300.0
Fe	1750.0	2740.0	60.0	11200.0
Co	4.0	3.0	0.4	30.0
Ni	3.0	2.0	0.5	10.0
Cu	10.0	20.0	1.1	100.0
Zn	200.0	210.0	1.8	1040.0
Br	10.0	10.0	1.3	70.0
Rb	10.0	10.0	1.5	70.0
Sr	20.0	40.0	1.1	130.0
Ba	40.0	50.0	12.0	220.0
Pb	50.0	40.0	4.0	190.0

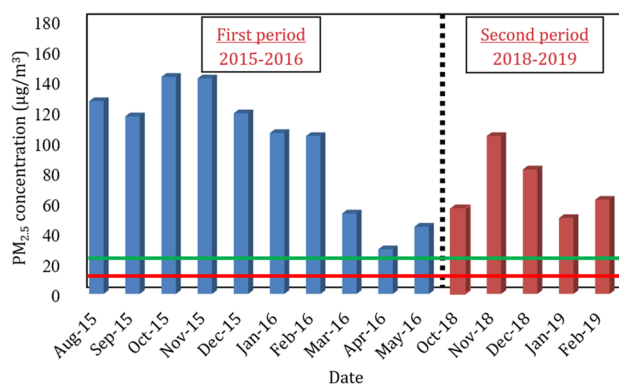


Fig. 2 Monthly average PM_{2.5} concentrations for the whole study period. The red line represents the WHO annual guideline and the green line the EU annual limit value

most polluted cities in the area, according to 2021 data (Tursumbayeva et al. 2023), with an average PM_{2.5} concentration of $59.5 \mu\text{g m}^{-3}$, in agreement with the mean concentration measured in this study during 2018–2019 ($70.8 \mu\text{g m}^{-3}$). Almeida et al. (2020) reported on the PM_{2.5} concentration levels for the same site, during 07/2014–05/2015. The average PM_{2.5} concentration measured was $124 \mu\text{g m}^{-3}$, which is in agreement with our results, especially with respect to the 2015–2016 campaign.

The average BC concentration was measured equal to $9.1 \pm 6.4 \mu\text{g m}^{-3}$. In agreement with PM mass concentrations, the highest BC concentrations were observed during October and December of 2015, with average concentration values of 14.4 and $18.0 \mu\text{g m}^{-3}$ respectively. There was a notable increasing pattern of BC concentrations from August 2015 ($7.4 \mu\text{g m}^{-3}$) to December 2015 ($18.0 \mu\text{g m}^{-3}$) (Figure S2). This increase in BC concentrations is likely due to residential heating. In January and February, these concentrations slightly decreased (12.5 and $9.5 \mu\text{g m}^{-3}$ respectively), reaching the lowest reported values from March to May (5.3 to $3.1 \mu\text{g m}^{-3}$). The same trend appears to be followed during the 2018–2019 period, but with the peaking month during November 2018 ($11.3 \mu\text{g m}^{-3}$). The lowest concentrations were recorded in April and May 2016, with average concentrations of 2.7 and $3.1 \mu\text{g m}^{-3}$ respectively. A significant percentage decrease was observed during these two sampling periods (45%). This variability in BC concentrations underscores the potential impact of residential heating on the local air quality. Chen et al. (2022) similarly noted decreased concentration values of EC during the spring period compared to the winter period of their study conducted between 2018 and 2019. They reported average concentrations of $4.2 \mu\text{g m}^{-3}$ during the spring period, contrasting with $7.3 \mu\text{g m}^{-3}$ observed during the winter period.

The main elements contributing to PM mass were Si and Ca, with maximum concentrations up to 28.1 and

21.9 $\mu\text{g m}^{-3}$ respectively, followed by Na, Fe, Al, and S. The high concentration levels of crustal elements indicate the increased soil dust resuspension and transport, which is frequent in central Asia (Hofer et al. 2017).

Correlation analysis of the different elemental components was utilized to explore the similarity of their time trends. The correlation coefficients observed between various elements offer insights into potential shared sources. The Pearson correlation (PC) matrix is provided in Table S1. Al, Si, Ca, Ti, and Fe exhibited strong correlations with each other ($PC > 0.82$), indicative of a soil dust origin. Additionally, Zn and lead Pb displayed strong correlation ($PC = 0.88$), suggesting potential common sources related to vehicular traffic and waste burning activities. Moreover, V was strongly correlated with Ni, Mg, Ca, Ti, Mn, Fe, and Cr ($PC > 0.85$), hinting at common sources associated with industrial activities in the region.

The elements' enrichment factor (EF) was also calculated and is displayed in Figure S1. EF values greater than 10 indicate dominance by anthropogenic sources, while EF values lower than 5 suggest elements of crustal origin. EF values falling between 5 and 10 denote elements originating from both natural and anthropogenic sources (IAEA 1992). The EF of an element for each PM sample is determined using the formula:

$$EF = \frac{\left(\frac{X}{C}\right)_{PM-Sample}}{\left(\frac{X}{C}\right)_{Crust}} \quad (3)$$

where X/C is the ratio of concentration of each element over the concentration of a reference element of crustal origin, calculated for each PM sample and for the Earth's crust. The reference element is typically chosen from Al, Si, Sc, Mn, Ti, and Fe. For our analysis, Si was selected as the reference element. The elemental composition of the Earth's crust was obtained from Wedepohl (1995). EF analysis revealed that S, Cl, Co, Cu, Zn, Br, and Pb are mainly related to anthropogenic sources. Additionally, Ca and Ni displayed EF values between 5 and 10, suggesting contributions from both natural and anthropogenic sources. Conversely, the remaining elements (Na, Mg, Al, K, Ti, V, Cr, Mn, Fe, Rb, Sr, and Ba) analyzed exhibited EF values lower than five, indicating primarily crustal origin. Emissions of crustal aerosols in the study area may be related to soil dust resuspension and transport but also to industrial activities, i.e., the operation of the cement plant (Fig. 1).

PM_{2.5} source apportionment

The model was run for different numbers of sources. After investigating a range of possible solutions ranging from 4 to 10 sources, the eight-source solution was selected as the

optimum. The best solution was identified by the use of key performance indicators, including Q values, distribution of scaled residuals, fit of measured PM concentrations, as well as by assessing the physical meaning of the obtained source profiles and contributions (Reff et al. 2007). The final solution was selected from 100 runs, while the uncertainty of the solution was assessed by implementing the Bootstrapping and Displacement error estimation tools provided by EPA PMF 5.0 (Manousakas et al. 2017). The differences between Q_{true} and Q_{rob} were less than 1%, while between Q_{true} and Q_{theor} was less than 10%.

The assessment of the agreement between the measured PM_{2.5} mass concentration and the concentration predicted by the model serves as a valuable indicator for evaluating the quality of the fit. The model-predicted concentration and the actual concentration exhibit a strong positive correlation ($y = 0.64x$, $R^2 = 0.85$), demonstrating a robust fit that effectively characterizes the true contribution of PM_{2.5} sources in the studied area.

High rotational ambiguity was revealed for the base run which indicated that the solution had high uncertainty. A few constraints were then introduced to the model to achieve a more stable solution (Manousakas et al. 2017) and obtain chemical profiles that better describe the main aerosol sources identified. Specifically, the following constraints were introduced: BC was pulled up maximally in the factors of waste burning and traffic and set to zero in the soil dust factor. In addition, S was pulled up maximally in the power plant profile. The $dQ\%$ was kept in all cases at the lowest value of 0.5% ensuring that no significant changes with respect to the unconstrained results would occur. The introduction of all four constraints resulted in an overall dQ of 1.9%.

Very low rotational ambiguity was observed after applying the constraints. The bootstrap (BS) error estimation was used to check the reproduction of the factors, and the number of runs was set to 100. BS results were very good, revealing reproducibility above 80% for all factors. In addition, displacement showed no factor swaps for the solution, for all dQ levels.

The seasonal source contributions are presented in Fig. 3. The chemical profiles of the eight-factor solution are presented in Fig. 4, and the average relative contributions of the different sources to the observed PM_{2.5} concentration levels are presented in Fig. 5.

Factor 1 is identified as soil dust and is characterized by the high levels of Al, Si, Ca, Mg, Ti, K, and Fe, elements that are related to mineral dust (Viana et al. 2008; Jain et al. 2020). The calculated ratios of Si/Al (3.31), Fe/Al (0.68), and Ti/Al (0.07) are in agreement with the upper crustal ratio values reported by Wedepohl (1995), namely 3.92, 0.40, and 0.04 respectively, further supporting the identification of this factor as soil dust. Soil dust accounts on

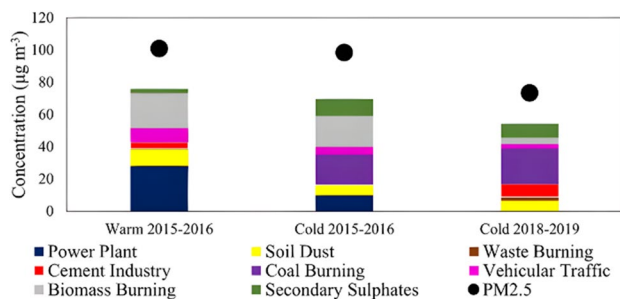


Fig. 3 Average source contribution to PM_{2.5} and measured PM_{2.5} in $\mu\text{g m}^{-3}$ for the warm and cold seasons of the whole study period

average for 11.3% of total PM_{2.5} concentration, contributing slightly more during the warm months (by 14% on average). This profile agrees well with the soil dust profile reported by Almeida et al. (2020), which accounted for 14% of the total PM_{2.5} concentration.

Factor 2 contains mainly S and is thus associated with the production of secondary aerosol, and specifically secondary sulfates. Secondary sulfates are major components of secondary inorganic aerosols and are known to contribute to visual degradation (Kang et al. 2004) and negatively affect respiratory health in humans (Mangia et al. 2015). They may be produced locally or transported over long distances from anthropogenic emissions of their gaseous precursor, SO₂. In general, the main tracers for secondary sulphates include the ionic species SO₄²⁻ and NH₄⁺ (Viana et al. 2008; Fakhri et al. 2024). Given the absence of major ion concentrations in the database, S serves as the primary indicator, which is a methodology employed in numerous studies (Marcazzan et al. 2003; Manousakas et al. 2017; Diapouli et al. 2022). The average contribution of this factor to PM_{2.5} corresponds to 11.5% and accounts for 14% of PM_{2.5} during the cold months and 4% during the warm months.

Factor 3 is characterized by high levels of Ca and Fe and contribution from other crustal elements (such as Ti, Mg, K, and Na), while it accounts for a high percentage of Cr, V, and Ni. The factor is identified as cement industry. Emissions from cement facilities are associated with cement dust (crustal elements with increased presence of Ca) and combustion emissions. The profile was also compared with published cement profiles from the SPECIEUROPE (EC-JRC) repository (Pernigotti et al. 2016). SPECIEUROPE contains particulate chemical profiles, including organic and inorganic species, derived from both measurements of emission sources and source apportionment studies carried out in Europe. Very good agreement was observed with the profiles from Yatkin and Bayram (2008) for the crustal elements but also for elements associated with combustion processes like Ni, V, and Zn. The V/Ni ratio in the profile was calculated at 1.9, which may point towards heavy fuel oil combustion.

High levels of Cr have been also associated with cement industries, as all forms of cement contain a certain amount of chromate (Demir et al. 2003). Furthermore, Cr has gained significant attention in the cement industry due to its potential to cause cement eczema, a type of allergic contact dermatitis (Paone 2008). The average contribution of this source to PM_{2.5} is at 6.9% and displays significant contribution during the cold period of 2018–2019 (16%).

The next factor (Factor 4) is identified as coal burning and is traced by BC, Cl, and Br; more than 40% of the mass of these species is explained by the coal burning factor. Cl has consistently been used as a tracer for coal combustion in various studies (Zong et al. 2016; Deng et al. 2014). Additionally, Rai et al. (2020) have highlighted the use of a Cl-Br factor as a potential indicator for coal burning activities. Moreover, Peng et al. (2021) reported unprecedented levels of bromine chloride (BrCl) at a mid-latitude site in North China during winter, with widespread coal burning in rural households. The contribution of this factor is quite high during the winter periods, specifically between December 2015 and January 2016 and December 2018 and February 2019, indicating residential coal burning for heating purposes. It has been observed that Cl and Br tend to exhibit high volatility during coal combustion, as seen in the partitioning behavior of trace elements (Vassilev et al. 2000; Clarke 1993). The average contribution of coal burning to PM_{2.5} is 21.3% and accounts for 48% of PM_{2.5} during the winter months (December to February). It should be noted that residential coal burning was also found to contribute significantly to PM_{2.5} levels in Kurchatov (Kazakhstan) (Almeida et al. 2020), highlighting the need to mitigate residential heating emissions in Central Asian urban areas.

Factor 5 is identified as waste burning, with main tracers: BC, Zn, Pb, and Cl. Waste burning is a fairly common practice to dispose unwanted materials and takes place in both developing and developed countries. In Dushanbe, municipal waste management strategies have not yet been implemented, and waste separation remains unresolved (Environmental Performance Reviews: Tajikistan 2017). Zn and Pb are trace metals that have been linked with waste incineration (Duan and Tan 2013; Rai et al. 2020; Manousakas et al. 2022; Pant and Harrison 2012), and Cl is also reported to be an important elemental tracer for plastic waste burning (Li et al. 2012; Jayarathne et al. 2018). It is primarily emitted as HCl and results to a large degree from polyvinylchloride (PVC) (Christian et al. 2010). The SPECIEUROPE database was once again used to check for similarities between municipal waste profiles, and good agreement was observed with the published profile of Samara et al. (2003), corresponding to metal scrap incinerator, especially for the elements of Zn (scrap metal incineration) and Pb (lead smelter). The closest municipal waste landfill facility is located North-West approximately 850 m away from the sampling site. This factor accounts for 1.5% of the average PM_{2.5}

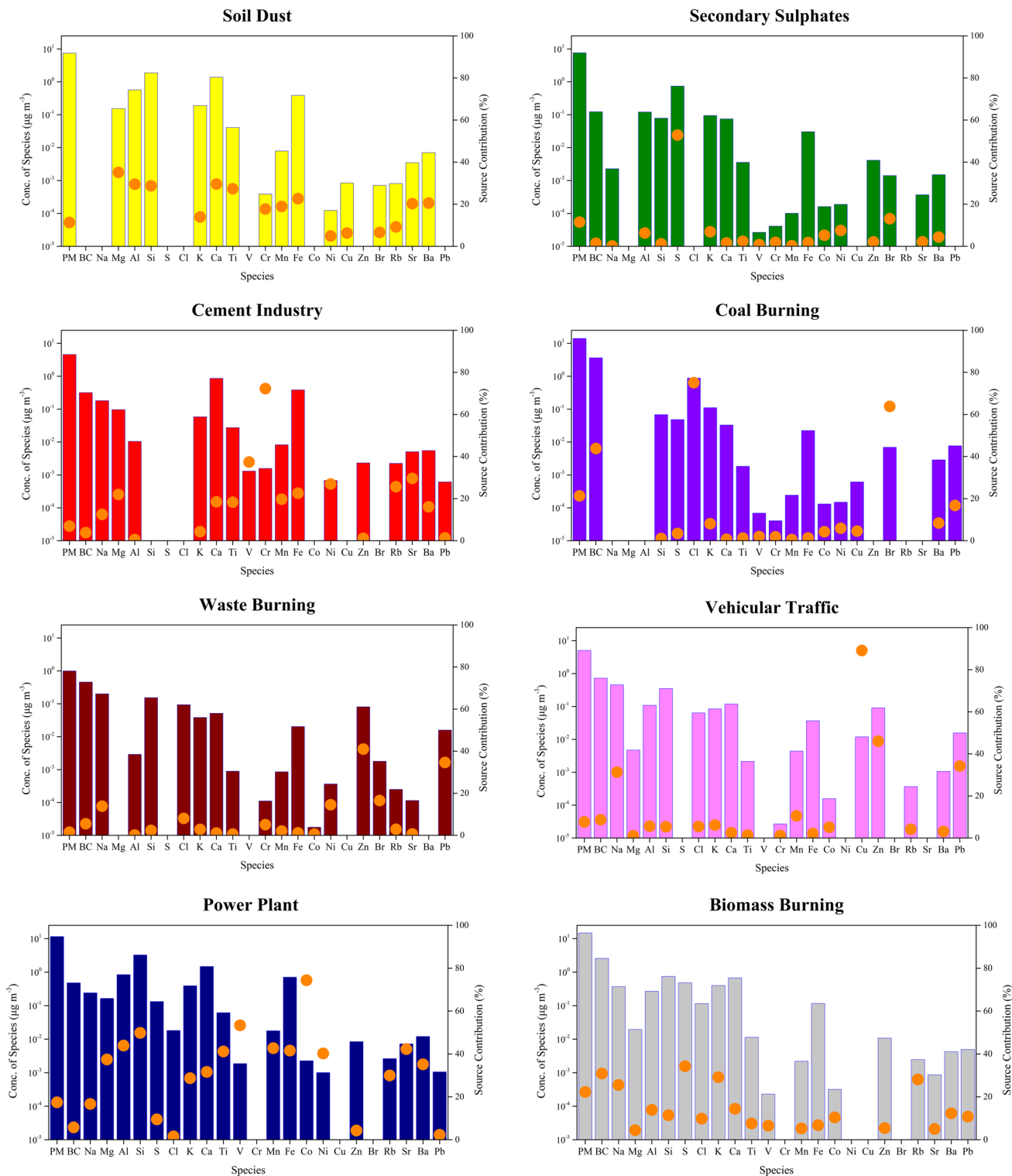


Fig. 4 The PM_{2.5} source profiles have been obtained by positive matrix factorization (PMF) using PM, BC, and elemental concentrations. The bars express the concentrations of species (in µg m⁻³), and

the orange circles display the % contribution of the source to the average species concentration

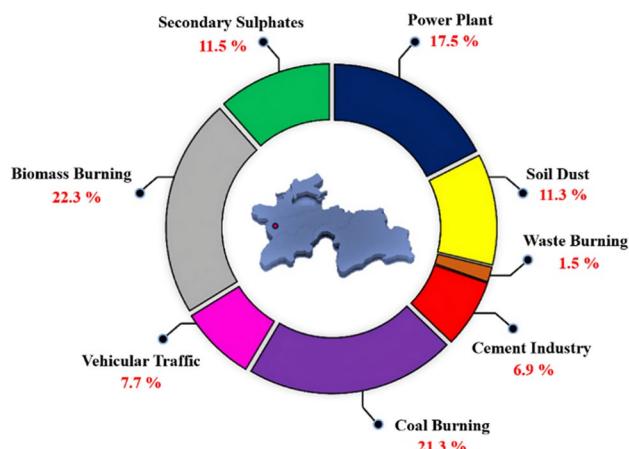


Fig. 5 Relative contribution of $PM_{2.5}$ sources for the whole study period

concentration, displaying the lowest contribution between all identified sources. No seasonal variability was observed for the years 2015–2016, but the contribution of this source was significantly increased during 2018–2019 (on average 4%).

Factor 6 accounts for most of the mass of Cu and Zn (89 and 46%, respectively) and a major part of Pb mass (34%), while it is also characterized by high levels of BC. These markers, commonly associated with vehicular traffic, are identified as including both exhaust and non-exhaust emissions (Pant and Harrison 2012). Non-exhaust emissions are comprised of brake, tire and road wear, and road dust resuspension. Zn is a metal that is generally used in rubber to strengthen the tires and has been suggested for tire wear emissions (Rhodes et al. 2012), while Cu and Pb particles are formed from friction between brake pads and disks (brake wear) (Huang et al. 2018; Thorpe and Harrison 2008). The average contribution of this factor to $PM_{2.5}$ corresponds to 7.7%. It should be noted that the lack of information on organic mass may have led to an underestimation of traffic exhaust emissions. This source contributed more during the warm season of 2015–2016 ($8.8 \mu g m^{-3}$, corresponding to 11% of $PM_{2.5}$ mass). A significant decrease was observed in the more recent years, with the cold period of 2015–2016 displaying almost double the contribution of the cold period of 2018–2019 ($4.6 \mu g m^{-3}$ in 2015–2016 versus $2.6 \mu g m^{-3}$ in 2018–2019, corresponding to 7 and 5% of the total $PM_{2.5}$ mass, respectively).

Factor 7 is associated with most of Co, V, and Ni mass (74, 53, and 40%, respectively) and a significant part of Rb, Ba, and Sr mass (30–42%). It is also characterized by high levels of Al, Si, Ca, Fe, K, BC, and Na. This factor is identified as power plant emissions. In Dushanbe, the main power plant is the Dushanbe-2 power station. It is a 400-megawatt coal-fired facility and is located in the north-west part of the city. Coal is a complex material that is comprised of organic matter and inorganic ash that is created over centuries by multi-layers of fallen vegetation (Vardar and Yumurtaci 2010). Co has been

used as a tracer for coal-fired (lignite) power plants (Vardar and Yumurtaci 2010, Uyar et al. 2016). V, Ni, Mg, Si, Ca, Fe, Al, and S are included in fly ash (Argyropoulos et al. 2013). Coal power plants are obligated to have electrostatic filters that retain the high percentages of fly ash, but a significant amount usually escapes those filters, and is emitted in the air. Finally, the presence of Na in this factor can be also explained by the desulfurization process of the flue gas (Manousakas et al. 2015). Flue gases are a mixture of combustion products, including water vapor, carbon dioxide, particulates, heavy metals, and acidic gases generated from direct (incineration) or indirect (gasification and pyrolysis) oxidation of RDF (refuse-derived fuels) or intermediate syngas (Materazzi and Lettieri 2017). The average contribution of power plant emissions to $PM_{2.5}$ is 17.5% and accounts for 22% during the warm months, indicating the increased activity of the plant, possibly due to high energy demand.

The final factor (Factor 8) is identified as biomass burning and is mainly traced by BC, K, and S (Viana et al. 2008). 29–34% of the total mass of these species is associated with this factor. In addition, this source accounts for 28% of the total mass of Rb, an element that has been also suggested as tracer for Biomass burning emissions (Massimi et al. 2020). The increased contribution of S in the biomass burning profile may be attributed to the aging process of the smoke particles. Li et al. (2003) highlight that with the aging of smoke, potassium chloride (KCl) particles from biomass burning are converted to potassium sulfate (K_2SO_4) and potassium nitrate (KNO_3) through reactions with sulfur- and nitrogen-bearing species from biomass burning as well as other sources. This indicates that while more KCl particles occur in fresh smoke, aged smoke contains more K_2SO_4 and KNO_3 . Almeida et al. (2020) also utilized S as an important marker for aged biomass burning in the profile of Skopje/MKD. The biomass burning impact is more pronounced during 2015–2016, with similar contributions during warm and cold season, at 28% of total $PM_{2.5}$ mass. Very low contribution is observed for the cold period of 2018–2019 (7%). This factor displays the highest average contribution to $PM_{2.5}$ (22.3%). Biomass burning has also been identified by Chen et al. (2022) as a significant contributor to PM air pollution in Dushanbe.

The conditional probability function (CPF) was also used to investigate the effects of different emission sources in different directions by integrating source apportionment information, with wind speed and direction data (Kim et al. 2012; Rai et al. 2016). The implementation of CPF proves advantageous for identifying pollution origins, provided that said origins reside within a specific range from the sampling location. The wind speed and wind direction data utilized in this study were sourced from the local monitoring network of Dushanbe (<https://www.visualcrossing.com/weather/weather-data-services>). Only the highest 10% of the source contributions was utilized (90th percentile) in this analysis. The mathematical representation of CPF is given by the equation:

$$CPF = m_{\Delta\theta} / n_{\Delta\theta} \tag{3}$$

Here, $m_{\Delta\theta}$ represents the count of wind occurrences in sector $\Delta\theta$, that surpasses a specific threshold, while $n_{\Delta\theta}$ denotes the total number of data points within the same wind sector. It is probable that the sources are situated in the direction with relatively high values of conditional probability (Fig. 6).

The results from the CPF analysis revealed distinct emission source patterns in relation to the sampling station's geographical location. Notably, the western-northwestern region was found to be associated with the cement industry, power plant, and waste burning sources, coinciding with the locations of the cement plant, Dushanbe-2 coal power station, and the municipal waste landfill. The southwestern direction was found to be a predominant source of soil dust, attributed to long-range transport from nearby desert regions such as Leili

and Karakum deserts (Abdullaev and Sokolik 2019). Biomass burning was found to originate predominantly from the South and may be also associated with long range transport, as further supported by the high S contribution in the biomass burning profile. Vehicular traffic and secondary sulphate sources predominantly originate from the western area, pointing towards the city center. The coal burning source demonstrates a wider spatial distribution, pointing towards residential heating emissions from various locations around the study area.

Conclusions

This is the first work effort to quantify pollution sources in Dushanbe, Tajikistan, an area with significant air quality issues, particularly concerning PM pollution. The measurement campaign, conducted during 2015–2016 and

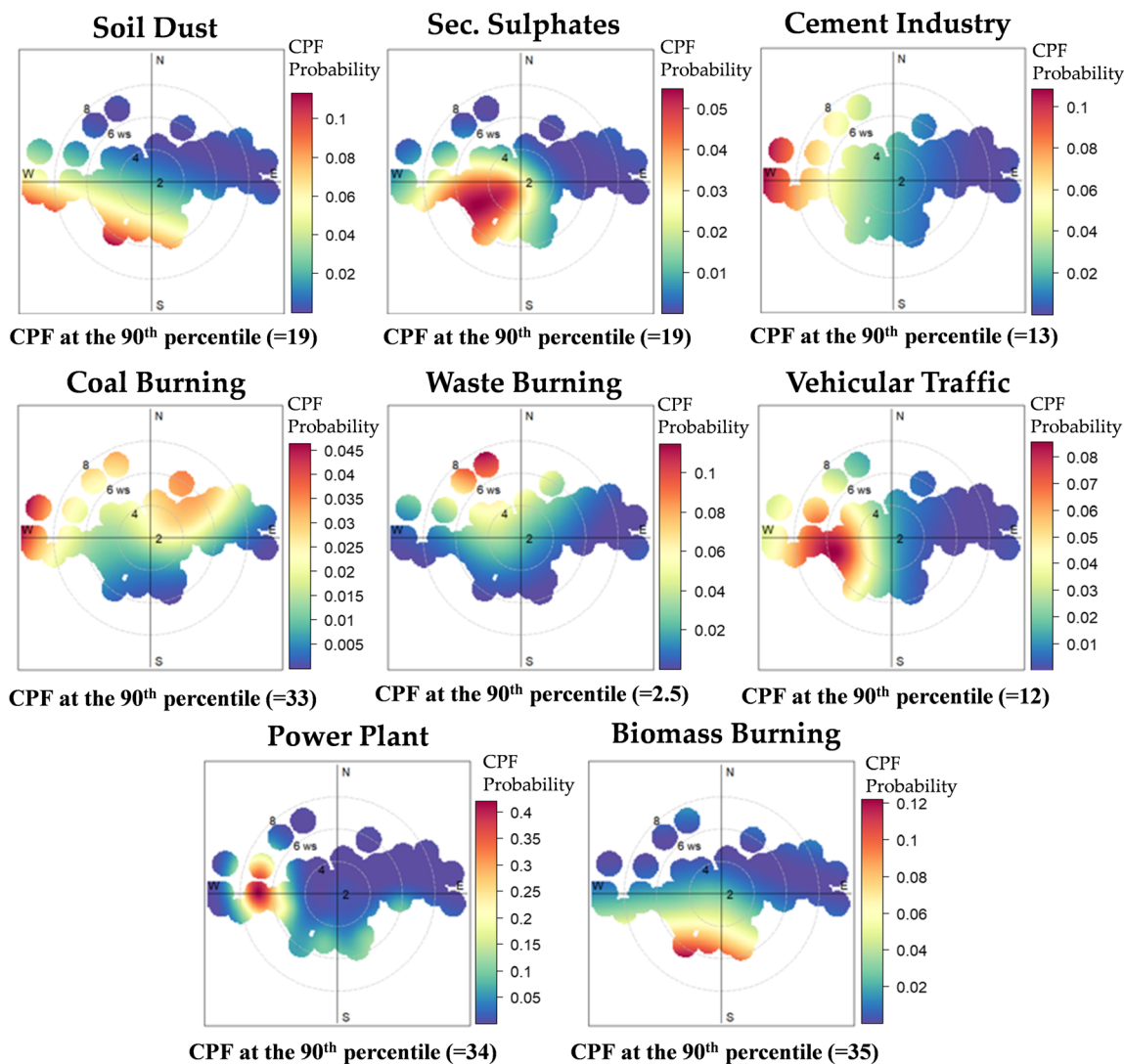


Fig. 6 CPF polar plots for source contribution

2018–2019, provided valuable insights into the composition and sources of PM_{2.5} in the region.

Our findings reveal alarmingly high PM_{2.5} concentrations that surpass the established limits set by both European standards and WHO guidelines. The identification of eight primary sources of pollution, including coal burning, biomass burning, and cement industrial emissions, underscores the complicated nature of the PM pollution problem in Dushanbe. Furthermore, seasonal variations in source contributions highlight the dynamic nature of pollution sources, with coal burning dominating during winter months and power plant emissions peaking during warmer seasons.

Notably, this work addresses a critical gap in research by focusing on Central Asia, an often-overlooked region in air quality studies. By incorporating carbonaceous species (BC) into source apportionment analysis, we aimed to provide a better understanding of combustion sources in the area.

However, future studies should aim to delve deeper into the understanding of PM pollution in Central Asian cities. By expanding research efforts and incorporating additional source tracers, such as organic aerosols and secondary inorganic species, more effective strategies for mitigating air pollution can be developed. We hope that this study may serve as a starting point for a comprehensive characterization and source apportionment of PM pollution in central Asia, where large cities face significant environmental problems due to increasing population and economic activities.

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Author contribution S.P.: writing-original draft, conceptualization, methodology, software, formal analysis, investigation, visualization. S.F.A.: resources, conceptualization, supervision, formal analysis, data curation, investigation, writing-review and editing. V.V.: conceptualization, supervision, formal analysis, data curation, investigation, writing-review and editing. M.I.M.: conceptualization, methodology, investigation, visualization, supervision, writing-review and editing. K.E.: conceptualization, visualization, investigation, supervision, project administration, writing-review and editing. E.D.: conceptualization, methodology, software, formal analysis, investigation, visualization, supervision, project administration, writing-review and editing.

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Declarations

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Consent to participate Not applicable.

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