



Biomass burning drives urban PM_{2.5} across the Western Balkans: evidence from a six-city source apportionment study in North Macedonia

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ABSTRACT

Biomass burning is widely reported as a driver of wintertime PM_{2.5} pollution across the Western Balkans, but its residential and open-burning components have rarely been resolved using multi-city source apportionment. We applied EPA Positive Matrix Factorization (PMF) 5.0 to 1195 daily filter samples collected at six urban background sites in North Macedonia during two campaigns (October 2020–October 2021 and March 2023–March 2024).

PMF consistently resolved residential wood combustion (RWC) and open/waste biomass burning (OBB) as distinct factors at all sites. RWC profiles were consistent across all 15 city pairs (Pearson distance 0.027–0.236). Campaign-period mean PM_{2.5} ranged from 29.1 to 62.2 µg/m³, corresponding to 1.2–2.5 times the currently applicable annual limit value of 25 µg/m³. Combined biomass burning contributed 21–35% of campaign-period PM_{2.5}, while heating-season RWC concentrations were up to 33 times higher than non-heating values. An undocumented winter OBB signal in Strumica (12.0 µg/m³) was most consistent with greenhouse boiler combustion and supported by stronger heating-season associations with SO₂ ($r = 0.587$ versus 0.185; $p < 0.001$).

In the three cities with continuous and filter-based monitoring, biomass burning exceeded 50% of PM_{2.5} on 83% of evaluable episode days in Skopje (Cliff's $\delta = 0.905$) and 71% in Strumica ($\delta = 0.752$). Episode-day wind speeds were up to 52% lower than non-episode means. Bimodal heating-season diurnal patterns, evening peaks reaching 93 µg/m³, and conditional probability function (CPF) analysis provided convergent evidence that RWC and OBB dominate acute pollution episodes.

The findings support accelerated EU Ecodesign transposition and stronger regulation of open and waste biomass burning, including greenhouse heating in agricultural areas.

1. Introduction

Biomass burning, including residential wood combustion, agricultural residue burning, and open waste burning, significantly contributes to the increasing levels of ambient air pollution globally. Residential wood combustion (RWC) is distinguished from other biomass burning sub-types by its concentration in the cold season and its association with limited-oxygen, low-temperature combustion in ageing residential stoves and during start-up/shutdown phases. It is also characterised by its direct proximity to densely populated areas (Cincinelli et al., 2019; Kukkonen et al., 2020; Zielinska and Samburova, 2011). Biomass smoke is generally dominated by fine particles (García-López et al., 2025; Simões Amaral et al., 2016). It comprises a chemically diverse mixture that includes carcinogenic polycyclic aromatic hydrocarbons, making it

a complex multi-pollutant public-health hazard (Balmes, 2019; Kim et al., 2011; Silva et al., 2015; Yao et al., 2023). PM_{2.5} from biomass combustion is consistently linked to respiratory and cardiovascular morbidity and mortality. Acute, high-concentration episodes drive disproportionate short-term health impacts independent of chronic annual mean exposure (Karanasiou et al., 2021; Macintyre et al., 2016; Torres et al., 2021).

Residential wood and coal heating is recognised as a major winter-time source of urban particulate pollution in Europe. Residential activities are the dominant source in just over half of the 708 European urban areas, with biomass combustion accounting for nearly all primary PM_{2.5} emissions from this sector (Cincinelli et al., 2019; Zauli-Sajani et al., 2024). The EU Ecodesign Regulation (EU) 2015/1185 has imposed binding PM, CO, and efficiency standards for new solid-fuel heaters sold

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in the EU market since January 2022 (Kubica et al., 2022), but no equivalent regulatory framework exists in the Western Balkans.

The six Western Balkan countries, Albania, Bosnia and Herzegovina, Kosovo, Montenegro, North Macedonia, and Serbia—have a combined population of approximately 17 million and experience some of the highest PM_{2.5} concentrations in Europe. Sarajevo and Skopje demonstrate severe annual and episodic concentrations, exacerbated by winter stagnation, domestic heating, and the combustion of solid fuels, leading to recurrent pollution episodes (Asker et al., 2025; Stanojević et al., 2024; Via et al., 2026). Biomass fulfils an estimated 42% of the overall regional heat demand; approximately 88% of houses depend on individual stoves or small boilers, experiencing energy losses of 40–50% (World Bank, 2017). The actual extent of biomass reliance is significantly underestimated by official statistics. WISDOM methodology surveys routinely reveal that actual firewood use surpasses official statistics by a factor of 1.7 in North Macedonia, up to 4.9 in Serbia, and between 4.1 and 4.9 in Bosnia and Herzegovina (Čomić et al., 2021; Glavonjić and Oblak, 2012). Nationally, 38.2% of households utilise firewood as their principal heating fuel, almost 60% employ inefficient wood combustion technology, and space heating constitutes 62% of total household energy usage (Simovski, 2019; State Statistical Office, 2026).

PMF-based source apportionment studies conducted across South-East Europe consistently identify biomass burning as a recurring major PM_{2.5} contributor (Almeida et al., 2020; Belis et al., 2019; Mirakovski et al., 2024). A large multi-city comparison including Skopje, Belgrade, Banja Luka, and Tirana found biomass burning accounted for an average of 16% of total PM_{2.5} mass, with a characteristic wintertime increase attributable to domestic heating (Almeida et al., 2020). A complementary Danube and Western Balkans synthesis concluded that biomass burning is the dominant residential heating sector source and likely underestimated in existing inventories (Belis et al., 2019). Within North Macedonia, a PMF-based source apportionment study of the Skopje urban background established residential wood combustion and open/waste biomass burning as the dominant PM_{2.5} sources during the heating season (Mirakovski et al., 2024). That study resolved their chemical fingerprints using the potassium–elemental carbon–chlorine–rubidium (K–EC–Cl–Rb) tracer framework, on which the present six-city study builds and extends. Further ambient evidence for the role of residential wood burning in driving severe winter pollution comes from Southeastern Europe. In Ioannina, a topographically comparable mountain-basin city in northwestern Greece, a severe residential-wood-burning episode produced winter PM_{2.5} concentrations exceeding 250 µg/m³ at times, with biomass burning contributing approximately 92% of organic carbon and 64% of elemental carbon during the heating season (Kaskaoutis et al., 2022), a magnitude of biomass-burning dominance consistent with the episode-level contributions reported in the present study. Meteorological stagnation has similarly been identified as a key modulator of wood-burning-driven PM₁₀ episodes when comparing high- and low-altitude Greek cities (Dimitriou and Kassomenos, 2018), while a multi-city low-cost sensor network across the Balkan Peninsula has demonstrated strong spatial covariance of PM_{2.5} concentrations during winter anticyclonic conditions, consistent with regional-scale synoptic stagnation driving simultaneous pollution episodes across multiple urban centres (Dimitriou et al., 2023), findings that respectively parallel the emissions–meteorology interaction and cross-city episode coherence identified in the present six-city dataset (see Discussion). Comparable ambient evidence of biomass-burning episodes has also been reported outside Europe, including size-resolved characterisation of episodic biomass smoke in Southeast Asia (Tial et al., 2024a, 2024b), further supporting the wider relevance of ambient biomass-smoke characterisation approaches such as those applied in the present study.

Despite the severity of the air quality problem across the Western Balkans, the relative importance of the principal sources of urban PM_{2.5} pollution remains contested, particularly during winter, when concentrations reach their highest levels. Public discourse on Western Balkan

air quality has been substantially shaped by advocacy reports, policy analyses, and civil-society publications that emphasise coal combustion in large power plants and industrial facilities as the primary regional driver of air pollution. Coal-fired power generation and industrial point sources are unquestionably major contributors to national and transboundary budgets of SO₂, NO_x, primary particles, and secondary PM_{2.5} precursors. However, their influence on ground-level PM_{2.5} concentrations within urban residential areas differs from their importance in regional emission inventories. Tall-stack dispersion and the frequent distance of major facilities from city centres generally reduce their direct contribution to urban ground-level concentrations relative to diffuse sources emitted within populated areas. Modelling studies indicate that coal-based energy production is primarily associated with national and transboundary-scale PM_{2.5}, whereas residential combustion contributes at both local and regional scales (Belis et al., 2019). Consistently, residential emissions have been identified as the dominant source of urban PM_{2.5} in 84% of Western Balkan cities (Joint Research Centre European Commission, 2025). By contrast, residential biomass combustion occurs near ground level, within or immediately adjacent to densely populated areas, and intensifies during stable winter conditions when atmospheric dispersion is limited. The distinction between regional emission budgets and local population exposure is therefore critical. A source that dominates national emissions may contribute less to urban PM_{2.5} exposure and acute pollution episodes than smaller, spatially distributed sources emitting directly within inhabited areas under low-dispersion conditions.

Receptor-based source apportionment using chemically speciated PM_{2.5} measurements provides a direct framework for evaluating source contributions at the urban scale. Nevertheless, three important knowledge gaps remain. First, most regional studies resolve a single undifferentiated biomass-burning factor, obscuring the relative roles of residential wood combustion and open or waste biomass burning, which differ in chemical signature, seasonality, and policy relevance. Second, available studies are concentrated in major urban centres, while smaller cities, peri-urban areas, and agriculturally intensive regions remain poorly characterised. Third, although biomass burning is widely associated with severe winter pollution, no regional study has quantitatively connected PMF-resolved source contributions with episode occurrence using multiple complementary analytical approaches.

This study addresses these three gaps by applying EPA PMF 5.0 to 1195 daily PM_{2.5} filter samples collected across six North Macedonian cities spanning diverse geographic, demographic, and agricultural settings. We resolve and chemically characterise residential wood combustion and open/waste biomass burning as distinct source factors. We quantify their seasonal and spatial variability, including atypical patterns in agriculturally intensive areas. We then evaluate their contribution to acute pollution episodes using convergent evidence from receptor modelling, continuous PM_{2.5} monitoring, meteorological analysis, chemical-profile similarity assessment, and gaseous tracer co-variation. The principal methodological novelty of this study lies in integrating these complementary approaches within a common multi-city framework, combining chemically resolved receptor modelling with inter-city source-profile similarity analysis, continuous temporal monitoring, meteorological stratification, directional CPF analysis, and gaseous combustion tracers. The study demonstrates that RWC and OBB are major contributors to wintertime urban PM_{2.5} pollution and severe pollution episodes in the investigated North Macedonian cities, with broader implications for air-quality policy across the Western Balkans. This regional framing reflects shared structural conditions across the Western Balkans, high biomass reliance, ageing residential heating infrastructure, and valley topography prone to winter atmospheric stagnation, documented in the literature cited above. The source apportionment evidence presented here, however, is drawn exclusively from six North Macedonian cities; its extension to other Western Balkan countries remains an inference pending the confirmatory multi-country source apportionment recommended in the Discussion.

2. Study area

The Republic of North Macedonia is a landlocked country in the central Western Balkans (Fig. 1), with a total area of 25,713 km² and a population of approximately 1.83 million. The terrain is predominantly mountainous, with major river valleys that strongly influence local meteorology. The climate is continental, with cold winters frequently characterised by prolonged temperature inversions and atmospheric stagnation in enclosed valley basins. Six cities were selected to represent the geographic, demographic, and socioeconomic diversity of North Macedonia's urban landscape (Table 1, Fig. 1). Together they span the full range of biomass fuel dependence and heating infrastructure conditions nationally.

Skopje (population ~ 540,000) is in the Skopje basin, an enclosed valley frequently subject to wintertime temperature inversions. It is the only city in North Macedonia with district heating (58,207 connected households); despite this fact, 40,159 Skopje households rely on fuelwood as their primary heating fuel. With 171,171 households, Skopje accounts for 88.8% of the total households in the Skopje Region, making it highly representative of the region's urban environment (Simovski, 2019; State Statistical Office, 2022; 2026).

Gostivar (~35,000) is located in the Polog valley, where 50.4% of households use fuelwood. The municipality has 20,054 households, representing 27.7% of all households in the Polog Region, thus serving as a significant urban centre within this region (Simovski, 2019; State Statistical Office, 2022). Kavadarci (~29,000) lies in the Tikves basin at approximately 270 m elevation and includes 12,375 households, or 26.4% of the total in the Vardar Region, highlighting its regional significance. It should be noted that road reconstruction works affected the 2023–2024 campaign period. Additionally, the area's major industrial source, the FENI nickel smelting facility located 5 km to the west, has been halted since before the 2023–2024 campaign and remains non-operational.

Kumanovo (~70,000) is in the north-eastern Kumanovo valley near the Serbian border, where 56.3% of households rely on fuelwood. The city's 30,450 households make up 62.9% of the Northeast Region's households, underscoring its prominence in the region (Simovski, 2019;

Table 1

PM_{2.5} monitoring site characteristics. All sites classified as urban background per Directive 2008/50/EC. Coordinates: Gauss-Krüger (EPSG:6316).

City	Region	Site description	Coordinates (Y; X)	Campaign	n
Skopje	Skopski	School yard, western residential area; nearest road 20–120 m	7,532,044; 4,650,585	Oct 2020 – Oct 2021	305
Gostivar	Polog	“Goce Delcev” Primary School, residential, nearest road 100 m	7,492,502; 4,627,539	Mar 2023 – Mar 2024	174
Kavadarci	Vardarski	Junction of two major roads, NE outskirts; FENI 5 km W (halted)	7,588,000; 4,588,164	Mar 2023 – Apr 2024	182
Kumanovo	Northeast	Near city hospital; nearest road 35 m; highway 600 m	7,558,623; 4,665,140	Mar 2023 – Mar 2024	173
Struga	Southwest	“Niko Nestor” High School; nearest road 5 m; city centre 900 m	7,472,806; 4,558,995	Mar 2023 – Mar 2024	180
Strumica	Southeast	General Hospital courtyard; nearest road 5 m; hospital boiler 25–30 m	7,637,270; 4,588,810	Mar 2023 – Mar 2024	181
Total					1195

State Statistical Office, 2022). Struga (~19,000) is on the southern shore of Lake Ohrid, in the south-western region with the highest fuelwood dependence of any study city (70.8%). Struga's 14,606 households

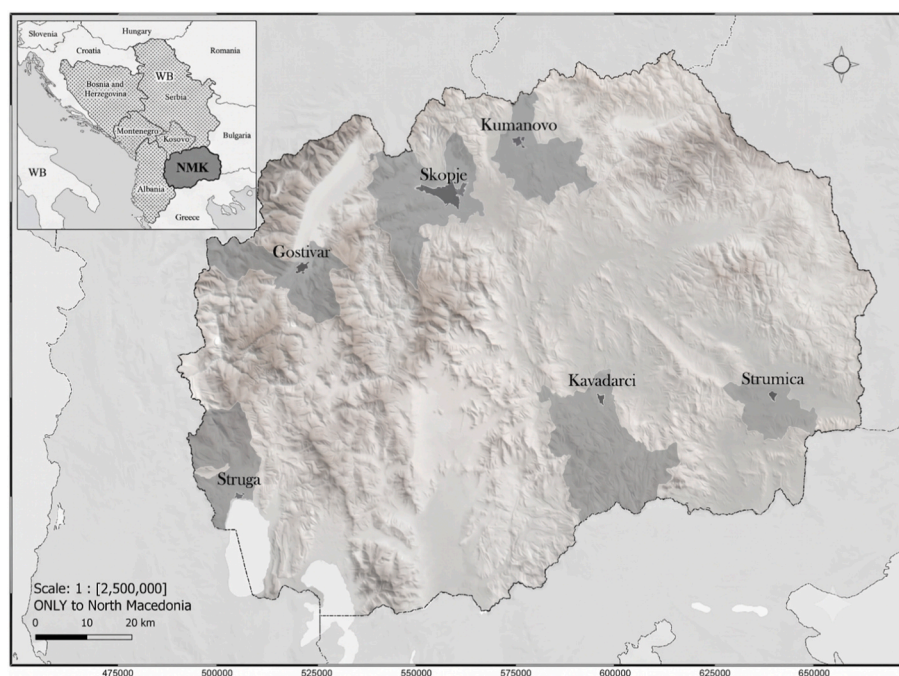


Fig. 1. Locations of 6 p.m.-2.5 monitoring sites (filled circles) across North Macedonia. Sites span the country's main heating-season pollution hotspots, from valley-floor cities prone to inversions to smaller regional centres. Darker shading = study municipality boundaries. Inset: North Macedonia (MK) within the Western Balkans context. Hill shade relief from SRTM 30 m DEM; coordinate system Gauss-Krüger (EPSG:6316).

account for 25.4% of all households in the Southwest Region (Simovski, 2019; State Statistical Office, 2022).

Strumica (~35,000) lies in the intensively cultivated Strumica valley in the south-eastern corner of North Macedonia, specialising in green-house vegetable production, fruit orchards, and viticulture.

The Southeast region reports the highest plant-residue wood consumption of any study region (4052 m³/year or 3.85% of total biomass; Simovski, 2019), and 64.7% of households rely on fuelwood as their primary heating fuel (State Statistical Office, 2026). No district heating is available.

3. Materials and methods

Sampling was performed in full accordance with EN 12341:2014 using low/medium volume sequential sampling systems (PNS 18T-DM-6.1, Comde Derenda, Germany) on 47 mm PTFE membrane filters (Advantec PF 020/040). 24-hour integrated samples were collected daily at Skopje and every other day at the remaining five sites. Filters were conditioned at 19–21°C and 45–50% RH for ≥48 h and weighed twice on a Radwag MYA5. 3Y. F microbalance ($d = 1 \mu\text{g}$) before and after sampling (acceptance criteria: pre-sampling difference <40 μg ; post-sampling ≤60 μg). Samples with measured concentrations flagged as invalid were excluded prior to analysis. Twenty samples from the Skopje, six from Gostivar, four each from Kumanovo and Kavadarci campaigns were removed on this basis, yielding a final dataset of 1195 valid samples across all six cities. All procedures were conducted under the ISO 17025-accredited Standard Operating Procedures of the UGD AMBICON Laboratory (accreditation no. LT-052).

Twenty-eight elements (Na, Mg, Al, Si, P, S, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Zr, Mo, Cd, Ba, Pb, Cl, Sb, W) were quantified by energy dispersive X-ray fluorescence spectrometry (NEX CG, Rigaku; EPA Compendium Method IO-3.3), using multi-element certified reference materials from the UC Davis Air Quality Research Center and SRM 2783 (NIST) for calibration. Three water-soluble ions (SO_4^{2-} , NO_3^- , and NH_4^+) were extracted from half-filters by sonication and determined spectrophotometrically using a Spectroquant® Prove 600 instrument. For the water-soluble ion analysis, quality-control recoveries ranged from 96.4% to 103.1%; CV values were below 7% for sulfate and nitrate and approximately 20% for ammonium. XRF quality-control and inter-laboratory comparison data are reported separately in Supplementary Table S1–S2. Elemental carbon (EC) was determined by optical transmissometry (Magee Scientific SootScan™ OT21, 880/370 nm). Full QC data are provided in Supplementary Material (Tables S1–S3).

Source apportionment was performed using EPA PMF 5.0. Species with $S/N < 0.5$ were excluded from the PMF input, whereas species with S/N between 0.5 and 1.0 were classified as weak and their uncertainties were increased by a factor of 3. Missing values and values below the detection limit were replaced with one-half of the detection limit. Optimal factor numbers were selected by evaluating $Q_{\text{true}}/Q_{\text{robust}}$ behaviour, scaled residuals, factor interpretability, bootstrap mapping, and DISP diagnostics. The same input preparation, uncertainty treatment, factor-selection procedure, and solution-acceptance criteria previously described in detail for the Skopje dataset were applied consistently to all six sites (Mirakovski et al., 2024). Summary

diagnostics for the final solutions are provided in Table 2. Small negative contributions, with absolute values ≤ 5% of the species mean, were set to zero before calculating percentage contributions. The resolved factors were grouped into four categories: residential wood combustion (RWC), open/waste biomass burning (OBB), secondary inorganic aerosol (SIA), and other sources (OS). Inter-city profile consistency was assessed using Pearson distance (PD) and standardised identity distance (SID), with acceptance criteria of $PD < 0.4$ and $SID < 1.0$.

High-pollution episode days were identified using a dual threshold: daily $\text{PM}_{2.5} > 50 \mu\text{g}/\text{m}^3$ and simultaneously $>$ site-specific 90th percentile concentration. The dual criterion ensures that episode days represent both absolute health-relevant exceedances and locally extreme events, enabling consistent comparison across cities with different pollution climates. A sensitivity analysis using only the $50 \mu\text{g}/\text{m}^3$ threshold confirmed consistent direction and statistical significance of results. Episode analysis was performed for three cities with co-located continuous $\text{PM}_{2.5}$ and meteorological monitoring: Skopje, Kavadarci, and Strumica. Statistical differences in RWC, OBB, and combined BB (= RWC + OBB) between episode and non-episode days were assessed using the two-sided Wilcoxon rank-sum test with continuity correction. Effect sizes were quantified using Cliff's delta (δ): negligible $|\delta| < 0.147$; small 0.147–0.330; medium 0.330–0.474; large ≥ 0.474 . A complementary season-controlled filter-day comparison was performed across all six cities, restricting both episode and non-episode observations to the heating season (October–March). All analyses: R version 4.5.3.

Hourly continuous $\text{PM}_{2.5}$ data were collected from Kavadarci, Skopje, and Strumica through the State Automatic Air Quality Monitoring Network (BAM 1020 or EDM optical particle monitors, Met One and GRIMM; EN 16450:2017). Wind speed and direction measurements were obtained from co-located WMO-standard meteorological stations. In Strumica, data for SO_2 ($\mu\text{g}/\text{m}^3$) and CO (mg/m^3) were also sourced from the State Automatic Air Quality Monitoring Network. SO_2 levels are quantified using automatic ultraviolet fluorescence analyzers according to MKS EN 14212:2013, while CO levels are measured using automatic non-dispersive infrared spectroscopy analyzers per MKS EN 14626:2013. The data capture rates were as follows: 94.4% for Kavadarci, 85.4% for Skopje, and 99.8% for Strumica. Seasonal diurnal profiles were computed as hourly averages for the heating season (October–March) and non-heating season (April–September), accompanied by a 95% confidence interval. The Conditional Probability Function (CPF) and time-variation studies utilized the openair package in R version 4.5.3.

To evaluate whether the episode-associated biomass-burning enhancement was consistent across cities, a pooled analysis was additionally performed using heating-season filter days from all six sites. Because absolute BB contributions were right-skewed and included zero values, log1p-transformed BB contributions were modelled using fixed effects for episode status and city. An interaction model additionally included city $\times$ episode terms and was compared with the additive model using an F-test and information criteria (AIC and BIC). As a sensitivity analysis, the season-controlled episode comparison was repeated after excluding samples for which PMF-reconstructed mass exceeded 150% of measured $\text{PM}_{2.5}$.

Table 2
PMF model diagnostics by city.

City	Factors	Selected run	Q_{true} at global min	$Q_{\text{true}}/Q_{\text{expected}}$	F_{peak} (4 directions)	Bootstrap mapping	DISP swaps
Skopje	7	89	6324.86	1.47	Stable	90–100%*	0/7
Kumanovo	6	41	4342.88	1.26	Stable	98–100%	0/6
Kavadarci	6	24	3604.57	0.94	Stable	98–100%	0/6
Strumica	5	41	5687.61	1.56	Stable	94–100%	0/5
Gostivar	5	64	6409.28	1.74	Stable	100%	0/5
Struga	5	33	5628.12	1.55	Stable	99–100%	0/5

Note: Skopje's Industry 1 factor showed 90% bootstrap mapping (8% attributable to the chemically similar Oil Fuel factor); all other factors mapped at 97–100%. For Strumica, the RWC factor showed 94% bootstrap mapping (6% attributable to the chemically similar OBB factor); all other factors mapped at 100%.

4. Results

The results presented in this section draw on convergent evidence from PMF source apportionment, inter-city chemical profile similarity testing, continuous real-time PM_{2.5} monitoring with meteorological analysis, and gaseous tracer co-variation, together supporting the interpretation that RWC and OBB are major contributors to urban PM_{2.5}, particularly during high-pollution episodes.

4.1. PMF model diagnostics

Table 2 summarises PMF model diagnostics across all six cities, including factor number, Q(true) at the selected base run, rotational stability (Fpeak), bootstrap factor-mapping stability, and displacement (DISP) results.

F_{peak} stability was assessed across all four standard rotational test directions (± 0.5 , ± 1.0). Bootstrap mapping percentages and DISP diagnostics were obtained from a supplementary verification model run using identical species, uncertainty, and factor-count configuration to the accepted base solution for each city, undertaken during the revision process; both confirm the rotational stability of the reported factor solutions. Zero factor swaps at every city under DISP indicate that each PMF solution is rotationally stable; this supports confidence in the reported factor structure but does not by itself establish that RWC and OBB represent uniquely identifiable physical sources (see Discussion). No residual pattern specifically indicating instability of the RWC or OBB factors was identified. SO₄²⁻ and EC showed the highest incidence of elevated scaled residuals, particularly at Gostivar and Struga, whereas

all other measured species showed elevated residuals on fewer than 2% of sampling days at every city (Supplementary Fig. S1).

4.2. Particulate matter concentrations and source contributions

Campaign-period mean PM_{2.5} concentrations ranged from 29.1 to 62.2 $\mu\text{g}/\text{m}^3$ and exceeded the currently applicable annual limit value of 25 $\mu\text{g}/\text{m}^3$ at all six sites. Peak daily concentrations reached 239.6 $\mu\text{g}/\text{m}^3$ in Strumica. A pronounced heating-season elevation was evident at all sites: most marked at Skopje (48.0 vs. 24.8 $\mu\text{g}/\text{m}^3$), Kumanovo (51.5 vs. 26.7 $\mu\text{g}/\text{m}^3$), and Strumica (58.0 vs. 27.9 $\mu\text{g}/\text{m}^3$). Monthly PM_{2.5} patterns with source contributions are shown in Fig. 2.

The RWC factor was resolved at all six sites with a consistent chemical signature. Across sites, 16.4–53.1% of measured K, 2.6–23.6% of EC, 2.3–40.9% of Cl, and 7.1–24.2% of Rb were apportioned to the RWC factor, consistent with the established chemical fingerprint of residential wood combustion. K/EC mass ratios of 0.03–0.23 were also consistent with published values for residential wood combustion (García-López et al., 2025; Fadel et al., 2022). At five of the six sites, the OBB factor accounted for 83.2–90.8% of measured Cl, with Pb, As, Rb, and S providing additional source-specific information. This Cl-rich signature supports the interpretation of combustion involving crop residues, treated biomass, or mixed waste, although elemental chlorine is not uniquely source-specific. The Skopje OBB factor was chemically distinct: it accounted for 78.8% of measured As, 63.0% of Rb, 34.3% of Cu, and 16.6% of EC, but only 5.6% of Cl. This profile is more consistent with urban waste combustion than with agricultural-residue burning (Fu et al., 2022; Peter et al., 2018; Saggiu and Mittal, 2020). The elevated

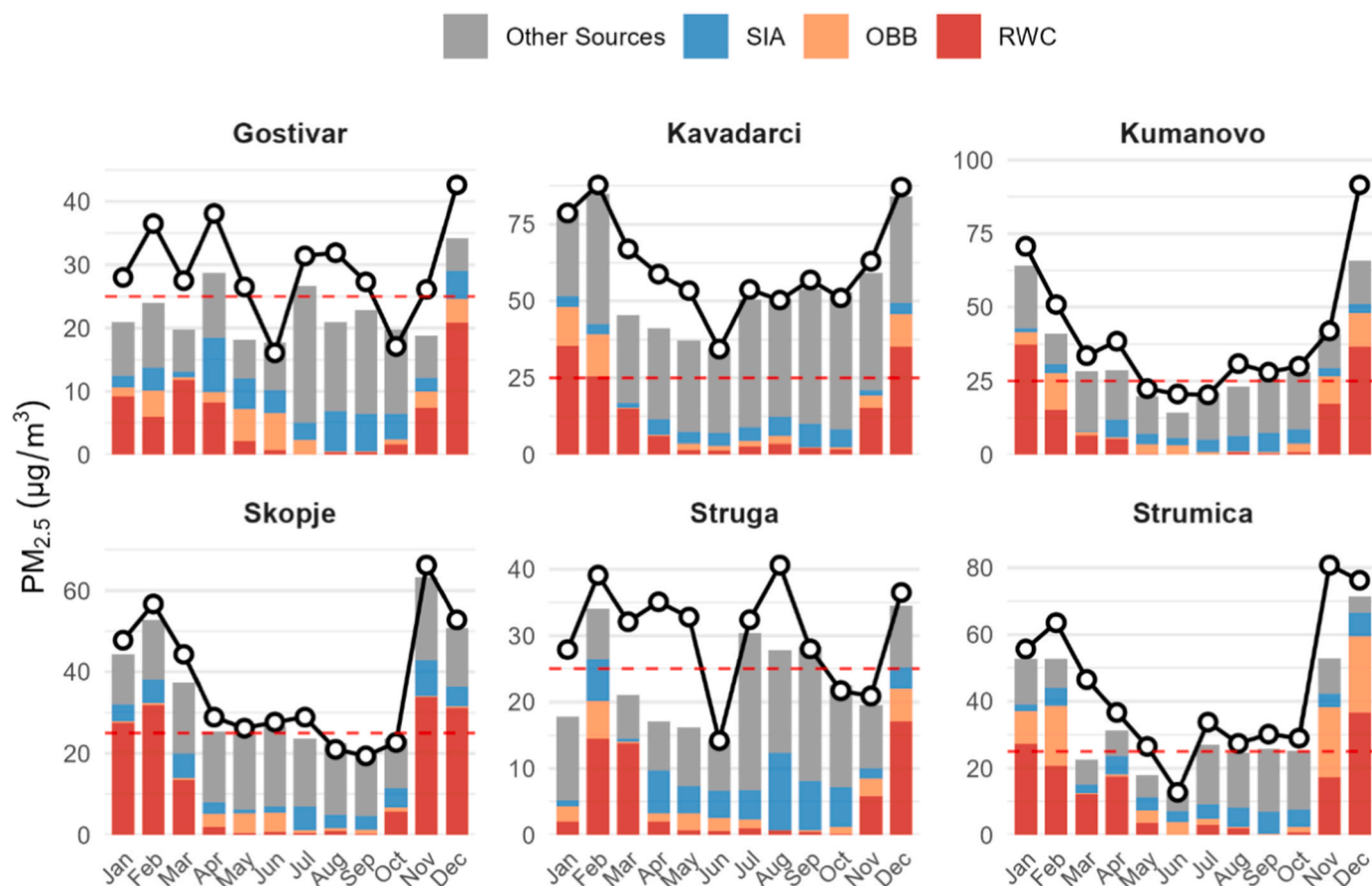


Fig. 2. Monthly mean PM_{2.5} source contributions (stacked bars: RWC = red, OBB = orange, SIA = blue, OS = grey) and measured PM_{2.5} filter mass (black dots and line) at six monitoring sites. RWC and OBB together account for most of the winter PM_{2.5} elevation at all six sites. The dashed red line indicates the currently applicable annual PM_{2.5} limit value of 25 $\mu\text{g}/\text{m}^3$ in North Macedonia and is shown as a reference benchmark rather than a monthly compliance criterion. Dots above bar tops indicate unresolved PM_{2.5} mass within normal PMF model uncertainty.

copper signature is notably consistent with the open burning of insulated electrical cabling, a practice commonly employed by informal metal scrappers in peri-urban areas of Skopje to recover copper wire. Complete profiles for all species and cities are provided in [Supplementary Fig. S2](#).

Inter-city RWC profile consistency was confirmed across all 15 city pairs: PD range 0.027–0.236 (mean 0.128; all within PD < 0.4 acceptance criterion) and SID range 0.150–0.568 (mean 0.410; all within SID < 1.0). This confirms that PMF consistently resolved the same RWC source type across all six cities. OBB profile consistency was confirmed for five cities. Skopje was an outlier for OBB (PD > 0.83 for all Skopje pairs; SID within criterion), consistent with its chemically distinct urban-waste OBB sub-type. The full PD/SID matrix is provided in [Supplementary Fig. S3](#).

Combined biomass burning (BB = RWC + OBB) contributed 21.2–34.6% of campaign-period mean PM_{2.5} across the six cities. RWC was the largest single identified source at Skopje (12.5 µg/m³, 22.6%), Strumica (11.4 µg/m³, 21.6%), and Kavadarci (11.9 µg/m³, 16.2%). OBB was highest at Strumica (6.8 µg/m³, 13.0%) or more than double any other city, consistent with the area's intensive greenhouse agricultural economy. At Kavadarci, Other Sources accounted for an anomalously high proportion of PM_{2.5} mass, largely associated with elevated mineral, road, and construction-related dust during the 2023–2024 road-reconstruction campaign. The FENI nickel-smelting facility was non-operational during the campaign, making it an unlikely contributor to this factor. In the absence of the temporary construction-related source, the relative percentage contribution of RWC would probably have been higher.

Heating-season RWC absolute contributions exceeded non-heating season values by factors of 4.3 (Strumica: 18.7 vs. 4.3 µg/m³) to 33 (Skopje: 23.4 vs. 0.7 µg/m³; [Figs. 2 and 3](#)). During the heating season, RWC alone accounted for 24.9 – 41.2% of PM_{2.5} mass.

Monthly peak RWC values occurred in November–January: Kavadarci (35.5 µg/m³ January), Strumica (36.6 µg/m³ December), Skopje (33.8 µg/m³ November) ([Fig. 4](#)). OBB showed more complex seasonality, with spring–autumn peaks at most cities consistent with agricultural burning cycles, but a markedly anomalous heating-season peak at Strumica (OBB 12.0 µg/m³ heating vs. 1.8 µg/m³ non-heating season) consistent with greenhouse boiler combustion and supported by complementary gaseous-tracer evidence.

4.3. Biomass burning and high-pollution episodes

The episode attribution analysis provides the most direct evidence that biomass burning is not merely an elevated background contributor but the major contributor to the most severe acute pollution events. Episode analysis was performed for three cities with co-located continuous PM_{2.5} and meteorological monitoring, Skopje, Kavadarci, and Strumica, where the complete convergent evidence chain is available.

Using the dual threshold criterion, 31 episode days were identified at Skopje (p90: 74.1 µg/m³), 19 at Kavadarci (p90: 104.7 µg/m³), and 18 at Strumica (p90: 87.4 µg/m³), each representing approximately 10% of total sampling days. For the heating-season episode days with valid BB source-contribution data, combined BB contributions were significantly elevated relative to heating-season non-episode days at all three cities ([Table 3](#); [Supplementary Fig. S4](#)).

The statistical effect was largest at Skopje: BB accounted for 59.8% of PM_{2.5} mass on episode days (mean: 56.7 µg/m³) vs. 39.4% on non-episode heating-season days (16.5 µg/m³; Wilcoxon $p = 2.97 \times 10^{-14}$; Cliff's $\delta = 0.905$, large effect). RWC alone contributed 56.1 µg/m³ on Skopje episode days, more than 3.5 times its non-episode heating-season mean. At Strumica, BB reached 62.8% on episode days (73.6 µg/m³) vs. 40.3% on non-episode heating-season days (20.5 µg/m³; $p = 1.60 \times 10^{-6}$; $\delta = 0.752$, large), with OBB contributing 33.7 µg/m³, approximately 4.9 times the non-episode heating-season mean. At Kavadarci, BB reached 42.8% on episode days (53.4 µg/m³) vs. 31.7% on non-episode heating-season days (20.9 µg/m³; $p = 1.52 \times 10^{-5}$; $\delta = 0.658$, large). At Skopje, BB accounted for more than 50% of PM_{2.5} mass on 82.8% of episode days (24 of 29) and more than 70% on 31.0% (9 of 29). At Strumica, BB exceeded 50% on 70.6% of episode days (12 of 17) and 70% on 52.9% (9 of 17) — the highest episode-day BB dominance of the three cities. At Kavadarci, BB exceeded 50% on 33.3% of episode days (6 of 18) and 70% on 5.6% (1 of 18).

All 31 identified Skopje episode days occurred during November–March, with none during April–October. Biomass-burning source contributions were available for 29 of these 31 episode days; two episode days (1 January and 23 February 2021) lacked valid BB contribution data. One episode day at Kavadarci and one at Strumica fell outside the heating season and were similarly excluded from this comparison; BB source-contribution data were otherwise complete. A complementary filter-based analysis across all six cities, restricting both episode and non-episode comparisons to heating-season days to control for seasonal confounding, is provided in [Supplementary Table S4](#). When all six cities were analysed jointly using heating-season filter days,

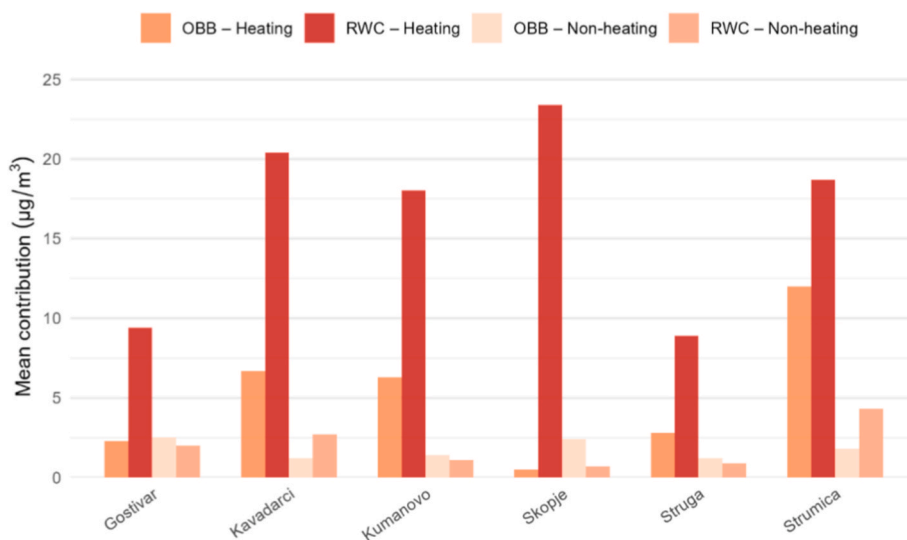


Fig. 3. Mean absolute contributions (µg/m³) of RWC and OBB during the heating season (October–March, dark colours) and non-heating season (April–September, pale colours) at six monitoring sites. RWC shows a stronger heating-season enhancement than OBB at most sites.

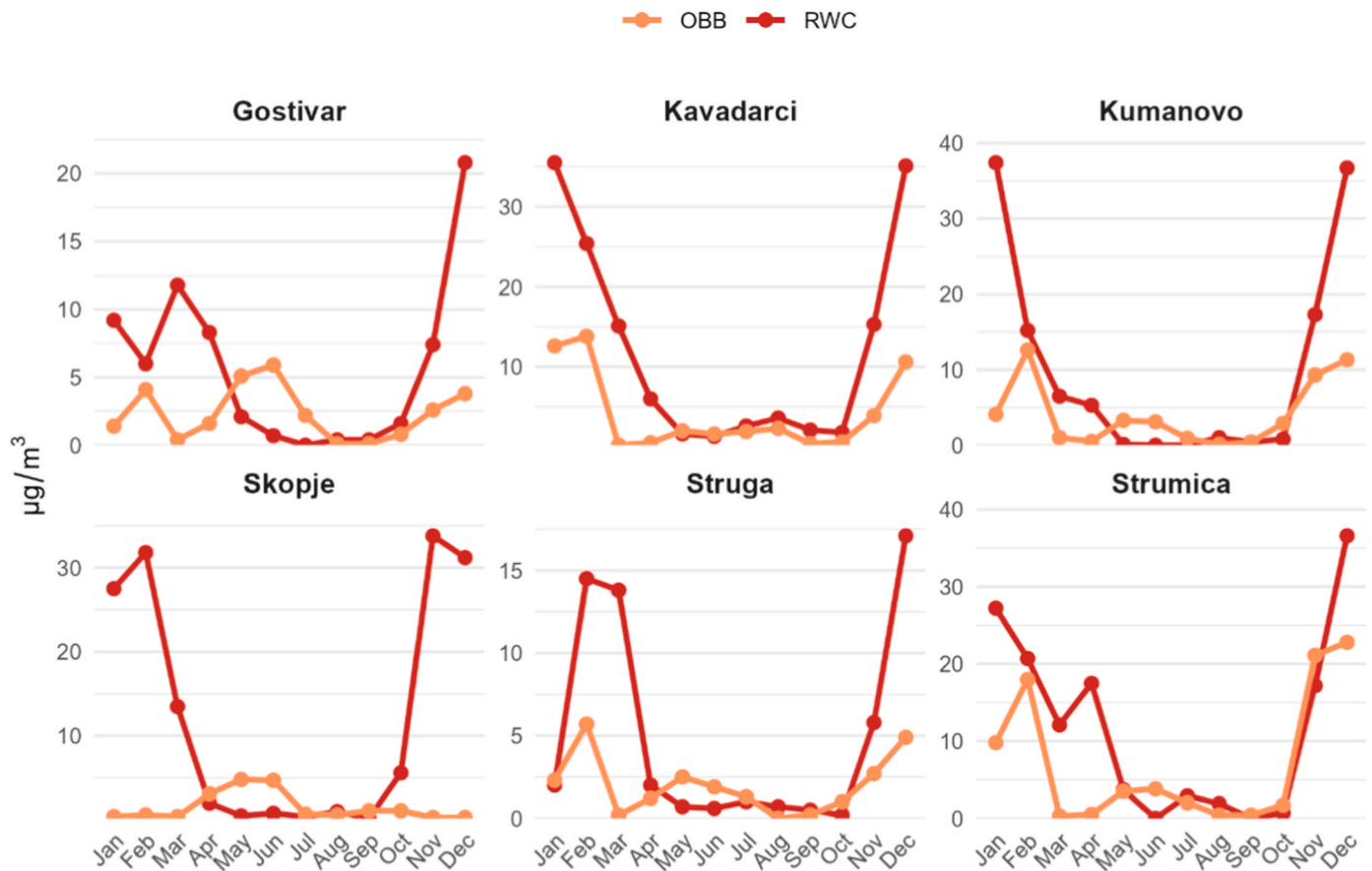


Fig. 4. Monthly mean RWC (dark red) and OBB (orange) contributions ($\mu\text{g}/\text{m}^3$) at six monitoring sites. Y-axis scales are independent per city. Peak-month timing and magnitude vary by city, reflecting local heating and agricultural-burning practices.

Table 3
Episode vs. non-episode $\text{PM}_{2.5}$ source contributions and statistical results.

Parameter	Skopje	Kavadarci	Strumica
Episode days (n)	29	18	17
Non-episode days, heating season (n)	129	77	72
BB - episode days (%)	59.8%	42.8%	62.8%
BB - non-episode days (%)	39.4%	31.7%	40.3%
BB episode mean ($\mu\text{g}/\text{m}^3$)	56.7	53.4	73.6
BB non-episode heating-season mean ($\mu\text{g}/\text{m}^3$)	16.5	20.9	20.5
Cliff's δ	0.905	0.658	0.752
	(large)	(large)	(large)
p	2.97×10^{-14}	1.52×10^{-5}	1.60×10^{-6}

Note: Statistical comparisons use heating-season episode days with valid BB source-contribution data (n = 29, 18, and 17 for Skopje, Kavadarci, and Strumica, respectively).

episode status remained strongly associated with higher biomass-burning contributions after adjustment for city ($p < 0.001$). Allowing the episode effect to vary by city did not improve model fit ($F = 1.179$, $p = 0.318$), indicating no evidence of significant between-city heterogeneity in the episode-associated BB enhancement. The same conclusion was obtained for BB expressed as a percentage of $\text{PM}_{2.5}$ (adjusted increase: 16.5 percentage points, $p < 0.001$; city $\times$ episode interaction: $F = 1.55$, $p = 0.172$; [Supplementary Table S5](#)).

To complement the PMF-derived source-apportionment results, continuous hourly monitoring of $\text{PM}_{2.5}$, wind speed and direction, and SO_2/CO (Strumica only) was analysed alongside the 24-h filter-based PMF dataset.

4.4. Diurnal patterns and meteorological context

All three cities exhibited markedly different diurnal concentration profiles between the heating and non-heating seasons, with heating season concentrations 3–8 times higher at every hour ([Fig. 5](#)).

During the non-heating season, $\text{PM}_{2.5}$ followed a comparatively flat diurnal cycle of $9\text{--}18 \mu\text{g}/\text{m}^3$, with a modest afternoon increase consistent with diffuse background sources and substantially lower residential-heating emissions. During the heating season, a pronounced bimodal structure emerged at all three cities: an early-morning peak corresponding to stove lighting for overnight temperature recovery, and a stronger evening peak corresponding to stove loading after work hours. At Kavadarci, evening peaks reached $74.3 \mu\text{g}/\text{m}^3$ at 19:00, more than 6 times the non-heating season value. At Strumica, the evening peak reached $93.0 \mu\text{g}/\text{m}^3$ at 20:00, with the overnight period remaining substantially elevated, consistent with near-continuous greenhouse boiler operation. At Skopje, a broader nocturnal elevation ($38\text{--}46 \mu\text{g}/\text{m}^3$, 00:00–04:00) reflects combined residential heating emissions and pollutant accumulation under Vardar basin temperature inversions. The limited weekday–weekend contrast provides additional evidence that residential combustion strongly influenced the heating-season diurnal pattern ([Supplementary Fig. S5–S7](#)).

The lower wind speeds ([Table 3](#)) support an important role for reduced atmospheric dispersion in the development and persistence of episode conditions. The effect was most pronounced at Skopje (episode: 3.25 m/s vs. non-episode: 6.72 m/s ; 52% reduction), consistent with the Skopje basin's susceptibility to wintertime temperature inversions. At Strumica (0.61 vs. 1.00 m/s ; 39% reduction) stagnation was similarly strong. At Kavadarci (0.95 vs. 1.08 m/s) the chronically low wind speed indicates persistent poor ventilation independent of episode status.

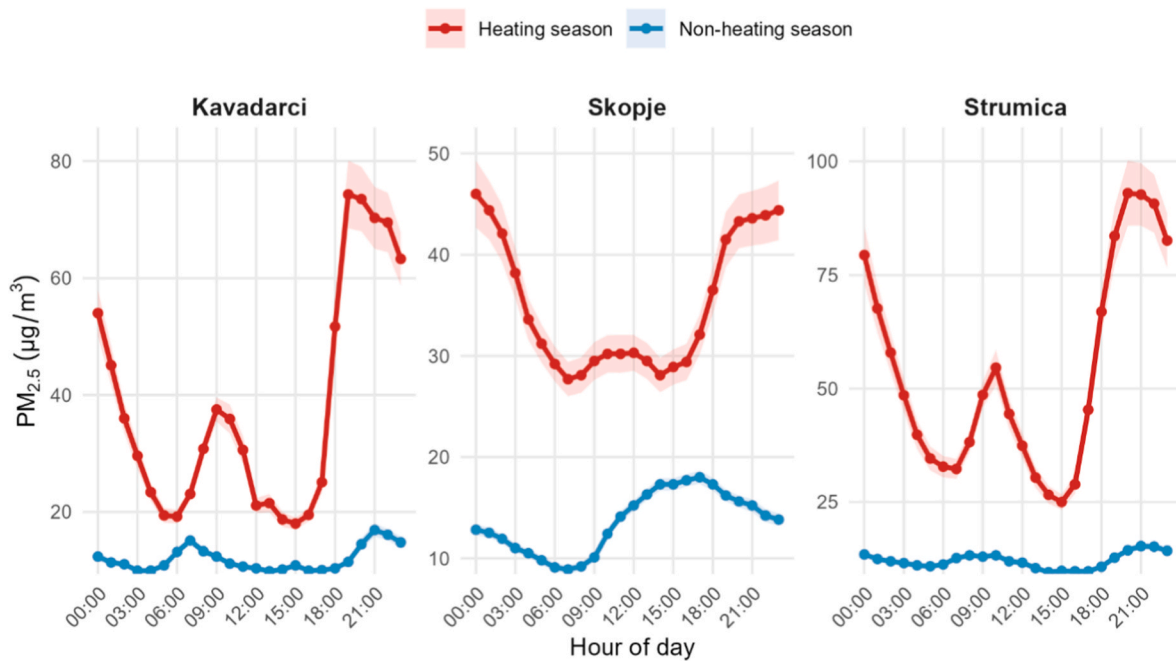


Fig. 5. Mean hourly $PM_{2.5}$ ($\mu\text{g}/\text{m}^3 \pm 1$ SE, shaded band) during the heating season (red, October–March) and non-heating season (blue, April–September) at Kavadarci, Skopje, and Strumica. The heating-season diurnal pattern shows a distinct evening peak consistent with residential stove use. Data from continuous national network monitoring (EN 16450:2017).

CPF analysis (Fig. 6) characterised the spatial origin of high-concentration events. At Skopje, the highest $PM_{2.5}$ percentiles (90th–95th) were associated with a pronounced north–south axis, consistent with channelled flow along the Vardar valley.

At Kavadarci, highest concentrations were associated with southerly to south-westerly winds, directly corresponding to the main urban residential area lying south of the monitoring station, providing complementary spatial support of the RWC-dominated source apportionment result. At Strumica, the near-omnidirectional CPF pattern is the spatial fingerprint of a distributed, valley-wide source consistent with widespread residential and greenhouse heating operations across the entire Strumica agricultural basin.

4.5. SO_2 and CO as complementary combustion tracers — Strumica

Monthly mean SO_2 exhibited a pronounced winter maximum closely tracking $PM_{2.5}$ (Fig. 7): SO_2 rose from a September–October minimum of

$2.74\text{--}2.99 \mu\text{g}/\text{m}^3$ to a December peak of $9.22 \mu\text{g}/\text{m}^3$, more than 3 times the annual minimum. $PM_{2.5}$ similarly reached a peak of $119.0 \mu\text{g}/\text{m}^3$ in December.

CO followed an identical trajectory, rising from a summer minimum of $0.247\text{--}0.286 \text{ mg}/\text{m}^3$ to a December maximum of $2.045 \text{ mg}/\text{m}^3$ (eightfold increase). The heating season SO_2 difference was statistically significant (heating mean 5.32 vs. non-heating $3.41 \mu\text{g}/\text{m}^3$; Wilcoxon $p < 0.001$). The Spearman correlation between hourly SO_2 and $PM_{2.5}$ was more than three times stronger during the heating season ($r = 0.587$) than the non-heating season ($r = 0.185$). This demonstrates substantial wintertime co-variation consistent with an additional sulfur-containing combustion source, potentially associated with biomass or mixed-waste combustion used for greenhouse heating (Supplementary Fig. S8). Together with the PMF evidence, the gaseous measurements support the interpretation of seasonally distinct OBB regimes at Strumica, while the specific source attribution remains uncertain.

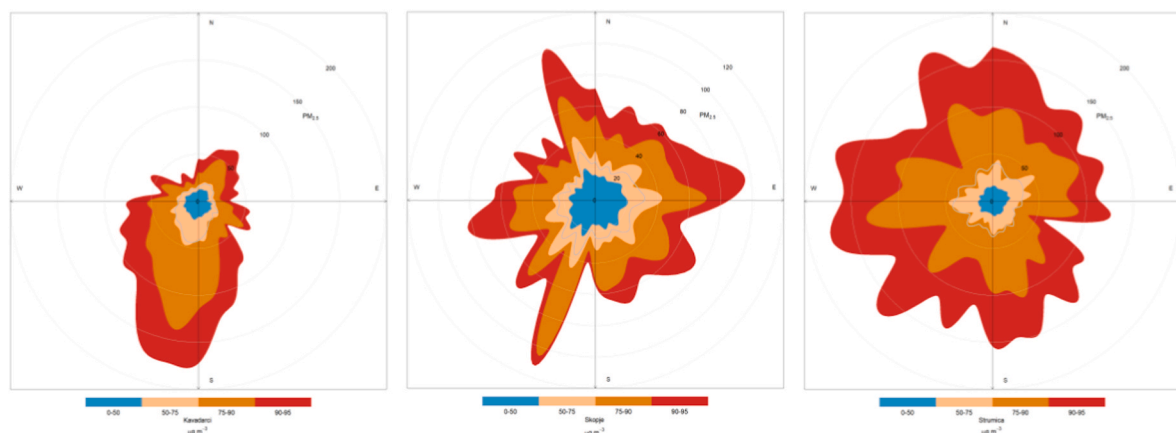


Fig. 6. Conditional Probability Function (CPF) analysis of $PM_{2.5}$ at Kavadarci (left), Skopje (centre), and Strumica (right). CPF patterns range from a directional residential signal at Kavadarci to a diffuse, valley-wide source distribution at Strumica. Colour scale = $PM_{2.5}$ percentile thresholds ($\mu\text{g}/\text{m}^3$): blue = 50th, peach = 75th, orange = 90th, red = 95th. Analysis performed using the openair package (Carslaw and Ropkins, 2012) in R version 4.5.3.

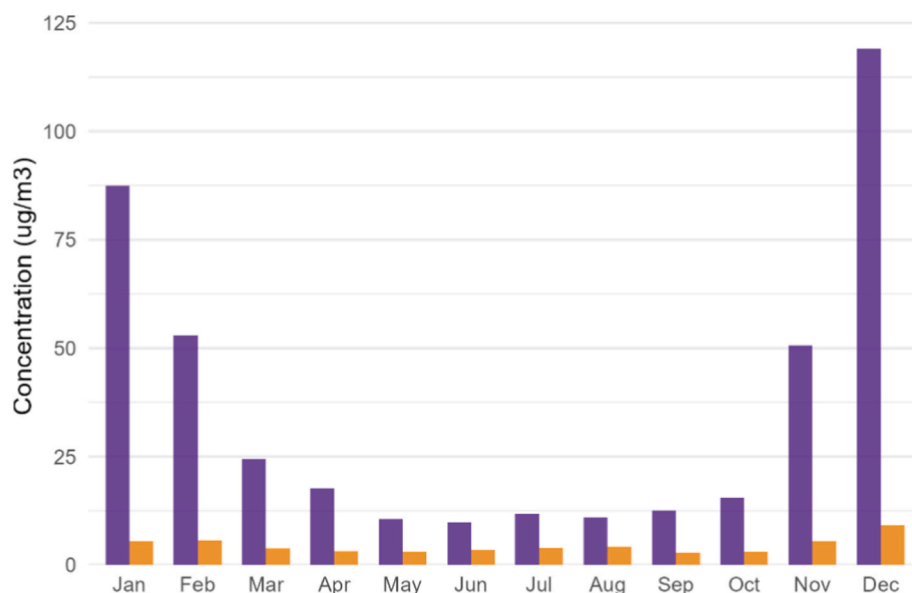


Fig. 7. Monthly mean SO₂ (orange bars) and PM_{2.5} (purple bars) concentrations (µg/m³) at Strumica from continuous hourly monitoring, March 2023–February 2024. SO₂ tracks PM_{2.5} through the heating season, supporting a shared combustion source. SO₂ measured by UV fluorescence analyser (MKS EN 14212:2013); PM_{2.5} by optical particle counter (EN 16450:2017). See [Supplementary Fig. S8](#) for seasonal SO₂–PM_{2.5} regression.

4.6. Convergence of complementary evidence

Four separate analytical pathways were used; PMF chemical source apportionment, inter-city profile similarity testing (PD/SID), real-time diurnal PM_{2.5} patterns with meteorological context, and gaseous tracer co-variation (SO₂, CO). These converge on the same conclusion: RWC and OBB are the major contributors to both the heating-season PM_{2.5} elevation and the most acute pollution episodes at all three monitored cities. Each pathway draws on different instruments, measurement principles, and analytical approaches; their convergence substantially strengthens the reliability of the findings and reduces the likelihood that the overall interpretation reflects limitations of any single analytical method.

5. Discussion

This study demonstrates that residential wood combustion and open/waste biomass burning are major contributors to wintertime urban PM_{2.5} pollution across the six investigated North Macedonian cities. Their influence becomes particularly pronounced during severe pollution episodes, when combined biomass-burning contributions frequently account for more than half of measured PM_{2.5} mass. By integrating receptor modelling with continuous air-quality monitoring, meteorological analysis, chemical-profile similarity assessment, and gaseous combustion tracers, the study provides convergent evidence from four complementary analytical frameworks. This convergence substantially strengthens confidence in the interpretation and reduces the likelihood that the observed source contributions reflect limitations inherent to any single analytical approach.

Although receptor modelling has become a standard tool for PM_{2.5} source apportionment, relatively few studies have corroborated PMF-derived source contributions using multiple complementary datasets. In the present study, chemical source profiles, inter-city profile similarity, continuous diurnal PM_{2.5} behaviour, meteorological conditions, CPF analysis, and combustion-related gaseous tracers show strong mutual consistency. Together, they provide a coherent and internally consistent interpretation of biomass burning as a major contributor to wintertime urban PM_{2.5} pollution. Rather than relying on a single analytical technique, the study combines chemically resolved source apportionment with complementary atmospheric observations, thereby

providing a more robust assessment of source attribution than receptor modelling alone. The evidence can therefore be interpreted hierarchically. Source identification, chemical characterisation, and seasonal behaviour are evaluated across all six cities using the filter-based PMF dataset. Episode-associated changes in biomass-burning contributions are also assessed across all six cities using season-controlled filter-day comparisons. A deeper mechanistic layer of evidence is available for Skopje, Kavadarci, and Strumica, where continuous PM_{2.5} monitoring, meteorological observations, directional CPF analysis, and, at Strumica, gaseous combustion tracers provide additional temporal and process-level corroboration. The robustness of the episode attribution was further evaluated through complementary sensitivity analyses. The main conclusions were preserved when episode identification was based on the absolute PM_{2.5} threshold alone, while restricting both episode and non-episode comparisons to the heating season reduced potential seasonal confounding. In addition, the significant heating-season × wind-speed interaction at all three continuously monitored cities indicates that the observed episode enhancement is not explained by meteorological stagnation alone but reflects the combined influence of elevated biomass-burning emissions and reduced dispersion. A pooled six-city analysis of heating-season filter days further supported this interpretation. Episode status was strongly associated with higher biomass-burning contributions after adjustment for city, while allowing the episode effect to vary by city did not significantly improve model fit ($F = 1.179$, $p = 0.318$). This provides no evidence of detectable between-city heterogeneity in the episode-associated enhancement of biomass-burning contributions. A sensitivity analysis using biomass burning expressed as a percentage of PM_{2.5} produced the same conclusion: episode days showed an adjusted increase of 16.5 percentage points ($p < 0.001$), while the city × episode interaction remained non-significant ($F = 1.55$, $p = 0.172$).

The widespread importance of RWC and OBB reflects long-standing structural characteristics of the residential energy sector in North Macedonia, including the high reliance on firewood and inefficient combustion technology detailed in the Introduction. Official firewood consumption statistics are also substantially understated across the region, as noted above. Under the stable atmospheric conditions that characterise winter in many Western Balkan valleys, these emissions accumulate rapidly, producing the severe PM_{2.5} episodes observed across all study cities.

Skopje is the only city with district heating in North Macedonia (58,207 households; [Simovski, 2019](#); [State Statistical Office, 2026](#)). Yet 40,159 households in the city rely on fuelwood as their primary heating fuel, and RWC contributions during the heating season reached $23.4 \mu\text{g}/\text{m}^3$ (41.2% of $\text{PM}_{2.5}$ mass). This persistence of residential wood burning even where district heating exists demonstrates that fuel switching is constrained not merely by infrastructure availability but by economic barriers. 10.6% of households always have difficulty paying energy bills ([State Statistical Office, 2026](#)), making firewood's low cost a binding constraint. Compounding the volume of biomass burned is the technology through which it is combusted: approximately 60% of households use inefficient appliances, burning wood at 40–50% energy efficiency losses ([World Bank, 2017](#)). Unlike EU member states, where the Ecodesign Regulation (EU) 2015/1185 has imposed binding efficiency and PM standards since January 2022, neither North Macedonia nor any other Western Balkan country currently has an equivalent mandatory certification framework. Even where such frameworks exist, their real-world impact is limited: the EU Regulation applies only to newly manufactured or marketed heaters, leaving the majority of older uncertified stoves in active use, and may be insufficient to effect substantial sector-wide reductions in particulate matter ([Hopan et al., 2025](#)).

The novelty of this study is threefold. Methodologically, it integrates chemically resolved receptor modelling with cross-city profile comparison, continuous temporal monitoring, meteorological analysis, CPF, and gaseous tracers within a common framework. Spatially, it applies this harmonised approach across six cities spanning contrasting urban, topographic, and agricultural settings. Scientifically, it resolves RWC and OBB as distinct source classes while demonstrating that RWC is highly consistent across cities, whereas OBB exhibits greater local heterogeneity in chemical composition and seasonal behaviour. The K–EC–Cl–Rb tracer combination of the RWC factor, with K/EC ratios of 0.03–0.23, is consistent with the established chemical fingerprint of incomplete wood combustion in conventional residential stoves. The Cl-dominated OBB factor (83–91%) at five of the six cities is consistent with agricultural crop-residue combustion. In contrast, the Skopje OBB profile, characterised by high As and Rb and low Cl, illustrates the compositional heterogeneity within the OBB category: agricultural-residue burning and urban-waste burning may both contribute to an OBB factor yet require entirely different regulatory interventions.

This contrast suggests that RWC represents a comparatively reproducible regional source type, whereas OBB is better understood as a heterogeneous class of locally specific combustion activities whose chemical composition and seasonal behaviour depend on agricultural practices, waste-burning patterns, and other local conditions.

The two regimes differ fundamentally in seasonal behaviour. RWC follows a sharp heating-season pattern at all six cities (factors of 4–33 times heating vs. non-heating). OBB shows more complex, city-specific seasonality, including a previously undocumented winter OBB pattern at Strumica that is most consistent with biomass-waste combustion in greenhouse-heating boilers and is supported by SO_2/CO co-variation and by the region's high orchard and plant-residue wood consumption (3.85% of total biomass; [Simovski, 2019](#)). The identification of a previously undocumented winter OBB pattern at Strumica, most consistent with greenhouse-heating biomass combustion, extends the inventory-underestimation concern of [Belis et al. \(2019\)](#) to combustion practices that may fall outside conventional residential-heating inventories.

Annual and seasonal mean contributions understate the true public health significance of biomass burning. The episode-level analysis reveals that RWC and OBB are not merely persistent background contributors but major contributors to the most extreme, health-relevant pollution events. This finding rests on four converging lines of evidence: PMF source apportionment confirms disproportionate BB on episode days; real-time diurnal signatures confirm the residential combustion temporal pattern; wind-stagnation analysis confirms the meteorological

compounding mechanism; and SO_2/CO co-variation at Strumica provides complementary gas-phase evidence for the combustion source. This convergence substantially reduces the likelihood that the episode-driving conclusion reflects an analytical artefact.

At Skopje, biomass burning was responsible for the majority of $\text{PM}_{2.5}$ mass on 83% of evaluable episode days, and for more than 70% on 31.0% of evaluable episode days. At Strumica, these values were 70.6% and 52.9% respectively. To quantify the relative and joint contributions of emissions and meteorology to episode occurrence, we stratified daily biomass-burning contributions by heating-season status and wind speed at the three continuously and filter-based monitored cities. Mean BB contribution was 1.3–2.3 times higher on low-wind heating-season days than on high-wind heating-season days across the three cities (Skopje: 31.9 vs. $14.1 \mu\text{g}/\text{m}^3$; Kavadarci: 35.4 vs. $17.1 \mu\text{g}/\text{m}^3$; Strumica: 36.3 vs. $27.1 \mu\text{g}/\text{m}^3$), while wind speed had little effect outside the heating season (non-heating BB ranged narrowly from 3.0 to $7.2 \mu\text{g}/\text{m}^3$ regardless of wind conditions). A regression of log-transformed BB contribution against heating-season status, wind speed, and their interaction confirmed a significant heating-season $\times$ wind-speed interaction at all three cities ($p = 0.0001$, 0.0024 , and 0.003 , respectively), indicating that atmospheric stagnation amplifies biomass-burning $\text{PM}_{2.5}$ specifically when combined with elevated heating-season emissions, rather than acting as an independent driver. These results support an interaction-based interpretation of severe winter pollution: elevated biomass-burning emissions establish the source burden, while atmospheric stagnation amplifies that burden sufficiently to produce the most severe episodes. Episode occurrence was correspondingly concentrated in the heating-season, low-wind stratum (26–29% of days) compared to the heating-season, high-wind stratum (6–11%) and the non-heating season (0–3% regardless of wind). This has a direct policy implication: emission reduction strategies framed solely around annual average compliance will systematically underweight the episode-driving sources that carry the greatest acute health burden.

Episode timing showed strong regional coherence both within and across campaigns. Among the five cities sampled during 2023–2024, 86% of episode days coincided with an episode at another city within ± 2 days, with pronounced multi-city clusters in mid-December 2023 and late January–early February 2024. Skopje, sampled in a non-overlapping 2020–2021 campaign, could not be directly compared by date, but its episode days aligned closely with the same calendar-day windows as the other cities' episodes in different years: 71% of Skopje's episode days fell within ± 2 calendar days (month/day, irrespective of year) of episodes recorded elsewhere, including a single date (3 February) matching the episode window at all five other cities. This cross-year calendar coherence indicates that severe $\text{PM}_{2.5}$ episodes recur at consistent times of the heating season across North Macedonia, most plausibly reflecting recurring regional-scale synoptic stagnation patterns, rather than occurring at city-specific, independent times.

We also directly compared meteorological conditions between the two sampling periods using continuous hourly meteorological records. Average heating-season conditions were broadly similar: mean temperature (4.5°C vs. 5.8°C for Skopje's 2020–2021 campaign and the other five cities' 2023–2024 campaign, respectively), mean relative humidity (80% vs. 76%), total precipitation (139 mm vs. 152 mm over 120–121 days, with 21 precipitation days in both periods), and calm-wind hours (708 vs. 577 h of the ~ 2900 total heating-season hours in each period). No large systematic differences were found that would be expected to bias cross-campaign comparability.

The results extend and substantially refine the prior North Macedonian source apportionment evidence ([Mirakovski et al., 2024](#)). The six-city mean combined BB contribution of 25.5% (range 21.2–34.6%) exceeds the [Almeida et al. \(2020\)](#) cross-regional average of 16%, suggesting that North Macedonia's peri-urban and agriculturally influenced cities experience a more severe biomass burning burden than the regional mean. The episode-attribution evidence provides the specific, city-level confirmation that the qualitative “elevated on high-pollution

days" finding of Almeida et al. (2020) translates, in the North Macedonian context, into BB accounting for the majority of PM_{2.5} mass on a substantial fraction of the worst pollution days. The identification of a previously undocumented winter OBB sub-type at Strumica extends the inventory underestimation concern of Belis et al. (2019) to entire combustion practices outside the scope of conventional inventories.

The PMF-resolved RWC and OBB factors capture primary combustion-related source signatures; they do not explicitly account for secondary organic or inorganic aerosol formed from the atmospheric ageing of biomass-burning precursor gases (VOCs, NO_x, NH₃). Some of this secondarily formed material may instead load onto the SIA factor rather than the RWC or OBB factors, meaning the reported RWC/OBB contributions likely represent a lower bound on biomass burning's total PM_{2.5} impact, particularly during multi-day winter episodes when atmospheric ageing is more extensive. An additional consideration concerns the size distribution of biomass-burning aerosol. Ambient studies of residential wood combustion in Alpine European towns, where, as in the present study cities, wood burning is a major domestic heating source in winter-stagnation-prone valleys, indicate that biomass-burning aerosols are strongly size-dependent, with ultrafine particles (<100 nm) carrying distinct chemical compositions and toxicological properties compared with the broader PM_{2.5} fraction, including elevated pro-inflammatory and genotoxic activity in winter samples (Corsini et al., 2017). Similar size-dependent chemical signatures, extending to the ultrafine range, have been documented in ambient biomass-burning aerosol more broadly (Phairuang et al., 2026). Since ultrafine particles fall within a toxicologically distinct size range not resolved by bulk PM_{2.5} measurement, the present study's characterisation of biomass-burning contributions at the PM_{2.5} bulk level may not fully capture the size-resolved composition of RWC and OBB emissions. This represents an important direction for future work in the study region.

Large power plants and industrial facilities remain important contributors to national and transboundary emissions of SO₂, NO_x, primary particles, and secondary aerosol precursors. However, the present receptor-based evidence indicates that diffuse, near-ground residential and open-burning emissions are more direct and important drivers of wintertime PM_{2.5} concentrations and acute exposure within the investigated urban areas. Consequently, reducing RWC and OBB emissions is likely to deliver more immediate urban air-quality benefits than strategies focused exclusively on distant large stationary sources. This does not diminish the need to control power-plant and industrial emissions, but it establishes household solid-fuel combustion and uncontrolled biomass and waste burning as priority sources for urban PM_{2.5} management.

These findings also have direct implications for future air quality policy. North Macedonia's current legally binding annual PM_{2.5} limit value is 25 µg/m³. The revised EU Ambient Air Quality Directive establishes a substantially more stringent annual limit value of 10 µg/m³, to be attained by 2030; we use this as an aspirational benchmark for comparison rather than a currently applicable compliance threshold, since North Macedonia is not yet bound by EU ambient air quality legislation. Alignment with this benchmark would require structural reductions in emissions from residential solid-fuel combustion and open or waste biomass burning, rather than measures focused only on short-term episode management. The findings point to a clear policy hierarchy. Because RWC and OBB differ in equipment, ownership structure, and regulatory pathway, we separate policy recommendations by source category below.

Residential stove replacement and accelerated transposition of the EU Ecodesign Directive (2009/125/EC), targeted for 2027 under the North Macedonia Energy Strategy to 2040, represent the most tractable near-term intervention for RWC. Even partial replacement of the oldest, least efficient stove stock could yield health benefits disproportionate to the fraction of appliances replaced, given the emissions–meteorology interaction demonstrated above, whereby stagnant conditions amplify the PM_{2.5} impact of residential combustion.

Open and waste biomass burning currently falls entirely outside North Macedonia's regulatory framework. Three specific interventions follow from the evidence: open burning restrictions during meteorologically unfavourable conditions; combustion efficiency standards for greenhouse heating boilers; and an agricultural residue burning monitoring framework. Regionally, these North Macedonia-specific findings support the case for a coordinated multi-country source apportionment network to establish, rather than assume, comparable RWC/OBB contributions elsewhere in the Western Balkans, sustaining the evidence base needed for effective EU accession-aligned air quality policy (Joint Research Centre European Commission, 2025).

Taken together, these results provide a transferable multi-method framework for evaluating biomass-burning source contributions and their role in severe pollution episodes across heterogeneous urban environments.

5.1. Limitations

This study does not include organic molecular tracers such as levoglucosan, which would enable more precise OBB sub-type differentiation; this may limit the precision of distinguishing among OBB sub-types (e.g., agricultural-residue vs. urban-waste combustion) but does not undermine the RWC/OBB factor separation itself, which is supported by convergent chemical, temporal, bootstrap, and rotational evidence (Table 2; Supplementary Fig. S2–S3).

The two sampling campaigns do not overlap in time, and Kavadarci was additionally affected by road reconstruction during part of the 2023–2024 campaign; cross-campaign comparisons should therefore be interpreted with this timing difference in mind, though a meteorological comparison between the two campaign periods found no large systematic differences expected to bias comparability (see Discussion).

Filter-based episode comparisons could be performed at all six cities, but continuous PM_{2.5} and meteorological corroboration was available only for Skopje, Kavadarci and Strumica. Episode attribution at these three cities can therefore be evaluated using both filter-based source apportionment and continuous temporal/meteorological evidence, whereas conclusions for Kumanovo, Gostivar and Struga rely on the filter-based PMF dataset and seasonal source patterns.

Inherent PMF modelling uncertainties — rotational ambiguity and the influence of measurement uncertainty — should be considered when interpreting precise percentage contributions; DISP diagnostics showing zero factor swaps at all six cities (Table 2) indicate that the resolved factor structure is rotationally stable; source assignments were supported by convergent chemical-tracer, temporal, and seasonal evidence (Methods), with DISP providing complementary, not independent, support for solution robustness. Daily-level source contributions should still be interpreted as approximate rather than exact.

This study is a chemical source apportionment analysis based on elemental, ionic, and carbonaceous speciation with PMF, corroborated by CPF, gaseous co-tracers, and meteorological analysis. Linking source contributions to health outcomes, resolving size-fractionated or ultrafine particle signatures through direct measurement, and incorporating isotopic, radiocarbon, or molecular-tracer validation would each require different sampling and instrumentation than were deployed here and fall outside this study's defined scope.

6. Conclusions

This study presents the first simultaneous, chemically resolved PM_{2.5} source apportionment across six North Macedonian cities, resolving RWC and OBB as distinct PMF factors and demonstrating their major contribution to high-pollution episodes through convergent multi-method evidence. The principal conclusions are:

- Campaign-period mean PM_{2.5} concentrations ranged from 29.1 to 62.2 µg/m³, substantially exceeding the currently applicable North

Macedonian annual limit value of $25 \mu\text{g}/\text{m}^3$ and the WHO annual air-quality guideline. Peak daily concentrations reached $239.6 \mu\text{g}/\text{m}^3$ at Strumica.

- RWC and OBB were resolved as chemically and temporally distinct PMF factors at all six cities, a solution confirmed as rotationally stable by zero factor swaps under DISP. RWC profile consistency was confirmed across all 15 city pairs (PD 0.027–0.236). The chemically distinct Skopje OBB profile is most consistent with an urban-waste-related combustion source rather than the Cl-rich agricultural-residue signature observed at the other five sites.
- Combined biomass burning contributed 21–35% of campaign-period $\text{PM}_{2.5}$. Heating-season RWC concentrations ($9.4\text{--}23.4 \mu\text{g}/\text{m}^3$) exceeded non-heating values by factors of 4–33.
- A previously undocumented winter OBB signal was identified at Strumica ($12.0 \mu\text{g}/\text{m}^3$; 14.9% of heating-season $\text{PM}_{2.5}$; 6.7 times the non-heating mean). Its chemical and seasonal characteristics are most consistent with biomass-waste combustion associated with greenhouse heating, supported by stronger winter $\text{SO}_2\text{--PM}_{2.5}$ co-variation, while some uncertainty in this specific sub-type attribution remains.
- Season-controlled filter-day analysis showed significantly greater absolute biomass-burning contributions on episode days at five of six cities, with Struga showing a borderline difference ($p = 0.052$). A pooled six-city analysis confirmed a significant episode-associated increase after adjustment for city ($p < 0.001$), with no evidence of significant between-city heterogeneity in the effect (city $\times$ episode interaction: $p = 0.318$). At the three cities with both filter-based and continuous monitoring, complementary temporal, meteorological and gaseous-tracer evidence further supports the role of biomass combustion in severe winter pollution events.
- Atmospheric stagnation acts together with elevated heating-season emissions: biomass-burning concentrations were 1.3–2.3 times higher on low-wind than high-wind heating-season days, and significant heating-season $\times$ wind-speed interactions were observed at all three continuously monitored cities.
- The findings support source-specific mitigation: accelerated replacement of inefficient residential solid-fuel appliances and Ecodesign-aligned standards for RWC, alongside dedicated regulation and monitoring of open/waste biomass burning, including agricultural-residue and greenhouse heating practices.

Future research should prioritise organic molecular tracers (particularly levoglucosan) to further differentiate RWC and OBB sub-types; extension of real-time monitoring to all six cities; and quantitative assessment of WISDOM-corrected biomass consumption inventories against PMF-derived source contributions.

CRedit authorship contribution statement

Dejan Mirakovski: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Afrodita Zendelska:** Writing – review & editing, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Ana Mihailovska:** Writing – review & editing, Investigation, Formal analysis, Data curation. **Marija Hadzi-Nikolova:** Writing – review & editing, Validation, Investigation, Data curation. **Gorgi Dimov:** Writing – review & editing, Resources, Investigation. **Boban Samardziski:** Resources, Investigation. **Toni Panov:** Resources, Investigation.

Declaration of generative AI and AI-assisted technologies in the writing process

During the preparation of this manuscript, the authors used AI-assisted writing tools for language editing, and manuscript preparation support. All scientific content, data analysis, calculations, source

apportionment modelling, statistical analysis, and conclusions are exclusively the work of the authors. The authors take full responsibility for the integrity and accuracy of all content presented in this manuscript. AI tools were not used in any part of the data collection, processing, or analysis workflow.

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Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Dejan Mirakovski reports financial support was provided by United Nations Development Programme. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2026.129339>.

Data availability

Data will be made available on request.

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