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IMPACT OF A COKING PLANT SHUTDOWN ON PAH LEVELS AND SOURCE FINGERPRINTS IN AN URBAN INDUSTRIAL VALLEY

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) associated with PM10 were monitored near the coking plant in Zenica, Bosnia and Herzegovina, before and after its closure to evaluate changes in air quality and emission sources. Concentrations of benzo[a]pyrene (BaP) and the sum of 16 US EPA priority PAHs were measured at monitoring sites representative of exposure conditions within the urban-industrial valley. Following the plant's shutdown, concentrations of all monitored PAHs decreased substantially, confirming the coking facility as the dominant emission source prior to closure. Diagnostic ratio analysis revealed a clear shift in source profiles, from signatures characteristic of coke oven emissions to patterns associated with residential coal and biomass combustion, road traffic, and other nearby industrial activities. Residential heating and traffic emerged as the main remaining contributors to PAH pollution. The study also highlighted the importance of valley topography, which restricts atmospheric ventilation and promotes pollutant accumulation during stable meteorological conditions. As a result, diffuse local sources continue to influence PAH levels despite the removal of the major industrial emitter. The findings demonstrate that industrial shutdowns can significantly improve air quality, but terrain and meteorological factors must be considered when assessing long-term pollution reduction in valley urban environments.

Keywords: Polycyclic aromatic hydrocarbons (PAHs); PM10; Coking plant shutdown; Diagnostic ratios; Emission source fingerprints; Valley topography.

1. Introduction

Urban-industrial valleys represent some of the most challenging environments for air quality management, where complex interactions between emission sources, topography, and meteorological conditions can lead to the accumulation of hazardous pollutants. Among these, polycyclic aromatic hydrocarbons (PAHs) are of particular concern due to their persistence, toxicity, and strong association with particulate matter (PM10). In heavily industrialized settings, historically dominated by metallurgical and coke production activities, coking plants have been recognized as major contributors to ambient PAH levels, especially carcinogenic compounds such as benzo[a]pyrene (BaP).

This study investigates the environmental impact of the shutdown of a major coking facility in Zenica, Bosnia and Herzegovina (Fig. 1), providing a rare opportunity to assess how the removal of a dominant industrial emission source influences ambient PAH concentrations and source characteristics. By comparing measurements of PM10-bound PAHs collected before and after the closure, the research evaluates changes in both the magnitude and composition of pollution, with particular emphasis on the 16 priority PAHs identified by the United States Environmental Protection Agency. Diagnostic ratio analysis is employed to identify shifts in emission source fingerprints, enabling a more nuanced understanding of the evolving contribution of residual sources.

Fig. 1. Fugitive emissions from the coking plant in Zenica

The findings presented in this article contribute to a broader understanding of how industrial transitions affect urban air quality, especially in geographically constrained environments. While the shutdown of the coking plant is expected to significantly reduce overall PAH levels, the persistence of emissions from residential heating, traffic, and other industrial activities – combined with the valley's limited atmospheric dispersion capacity – raises important questions about the long-term effectiveness of single-source mitigation strategies. This work therefore highlights the need for integrated approaches that consider both emission source apportionment and local environmental conditions when designing air quality policies in similar urban-industrial valleys.

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous, persistent organic pollutants whose environmental distribution is predominantly driven by industrial coal carbonization, fossil fuel combustion, and biomass burning [10], [12]. Retained industrial coking facilities act as long-term hotspots where high-molecular-weight species – comprising four to six aromatic rings – accumulate in surrounding soils and migrate into groundwater systems [1], [2]. Although low-molecular-weight PAHs exhibit higher vertical mobility [2], high-molecular-weight compounds govern total carcinogenic toxicity and drive the Incremental Lifetime Cancer Risk in exposed urban populations [2], [4]. There is ongoing debate over how much atmospheric degradation (photolysis and ozone reaction) alters PAH ratios. If a ratio shifts while a particle is in transit, the source apportionment might incorrectly identify a local source as a distant one. Risk assessments described in [4] typically calculate risk based on total PAH mass. Critics argue this overestimates risk because a significant portion of PAHs may be tightly bound to soil organic matter and not biologically available to humans.

Toxic Equivalency Quotient assessments demonstrate that Benzo[a]pyrene (BaP) and Dibenz[a,h]anthracene (DahA) account for 80% - 90% of total mixture toxicity under standard toxic equivalency weighting schemes [6]. In industrially impacted air basins, ambient concentrations frequently exceed WHO guidelines, while co-emitted metalloids such as arsenic exacerbate overall carcinogenic risks beyond PAH-only evaluations [5], [7].

Differentiating industrial coking emissions from background combustion relies on diagnostic molecular ratios – such as Fluoranthene / (Fluoranthene + Pyrene), Benzo[a]anthracene / (Benzo[a]anthracene + Chrysene), and Benzo[a]anthracene / Benzo[e]pyrene – alongside receptor models like Positive Matrix Factorization [12, 16]. Active coking operations impart distinct fugitive fingerprints rich in BaP and Benzo[a]anthracene relative to Benzo[e]pyrene, contrasting with vehicular traffic profiles dominated by Benzo[ghi]perylene and combustion gases like CO and NO [9], [16]. However, atmospheric transit, photolysis, and ozone reactions can alter diagnostic ratios downwind, necessitating multi-tracer receptor models that integrate PAHs, carbonaceous fractions, inorganic ions, and trace metals to accurately resolve overlapping traffic, industrial, and waste-burning sources [10], [13].

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In topographically constrained urban valleys, seasonal meteorology and winter temperature inversions trap fine particulate matter (PM2.5 / PM10), causing winter PAH levels to exceed summer baselines by nearly tenfold [3, 8]. This atmospheric trapping complicates source attribution due to chemical overlap between industrial coal carbonization and residential solid-fuel heating [3], [15]. To isolate point-source industrial contributions from seasonal background variation, optical source apportionment using multi-wavelength Aethalometry (AE33) and Ångström exponents must be coupled with standardized gravimetric particulate measurements to decouple solid-fuel combustion from vehicle exhaust [14], [15].

Although industrial operations are regulated under environmental frameworks such as Integrated Pollution Prevention and Control (IPPC), engineering retrofits based on Best Available Techniques (BAT) often fail to reduce fugitive coking emissions to safe ambient levels (≤1.0 ng/m^3 BaP) [11], [16]. Permanent plant closure represents a decisive emission intervention, yielding rapid downwind toxic equivalency reductions of 92.8%–97.6% [16]. While existing literature broadly documents source contributions under operational conditions [12], [13], the complete decommissioning of a coking facility provides a unique real-world opportunity to observe the collapse of industrial coal-processing signals and quantify how residual urban sources—such as vehicular traffic and space heating—redefine the atmospheric PAH fingerprint in industrial valley systems [9], [10], [11].

Most of these cases collectively demonstrate that PAH contamination is a persistent multimedia challenge driven by the convergence of industrial legacies – particularly coking operations – and seasonal combustion activities in urban centers. By integrating advanced source apportionment with toxic equivalency modeling, the research underscores a consistent global pattern where high-molecular-weight compounds drive significant carcinogenic risks across soil, air, and groundwater, necessitating both site-specific remediation and rigorous atmospheric emission controls.

2. Materials and Methods

2.1. Measurement

The methodology employed for the experimental identification of polycyclic aromatic hydrocarbons (PAHs) and volatile organic compounds (VOCs) is based on the following standards:

- EN 14662-3:2015 Ambient air – Standard method for the measurement of benzene concentrations – Part 3: Automated pump sampling with in situ gas chromatography.
- EN 12341:2023 Ambient air – Standard gravimetric measurement method for the determination of the PM10 or PM2.5 mass concentration of suspended particulate matter.
- ISO 12884:2000 Ambient air – Determination of total (gas and particle-phase) polycyclic aromatic hydrocarbons – Collection on sorbent-backed filters with gas chromatographic/mass spectrometric analysis.

2.2. Equipment

Measurements were conducted using a mobile monitoring station (Fig. 2).

Fig. 2. Measuring station.

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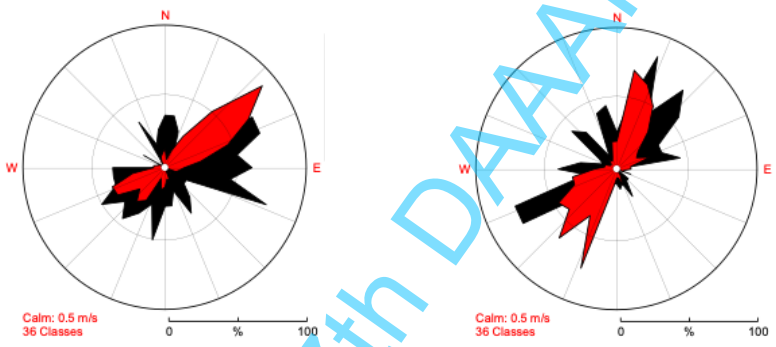
The station was equipped with an air sampling system and analytical instrumentation, as follows:

- Chromatotec air TOXIC – GC 866: Used for the determination of benzene concentrations in ambient air.
- DIGITEL DHA-80: A high-volume sampler used for the collection of PM10 particulate matter in ambient air.
- Radwag Analytical Balance (Model: X2 82/220.R2; Accuracy Class I, measurement range 1 mg – 220 g).
- Gas Chromatograph with Mass Spectrometric Detection: Utilized to determine the concentration of total polycyclic aromatic hydrocarbons in 24-hour PM10 samples.
- Data Acquisition Systems: calibration systems, zero-air generators, and instruments for monitoring meteorological parameters.

The sampling process involved drawing air for a 24-hour duration through filter media using high-volume samplers. Glass or quartz fibre filters were selected for this purpose due to their high particulate retention efficiency, structural integrity, and low airflow resistance.

Upon a comprehensive evaluation of all relevant parameters, the monitoring site was strategically selected in the vicinity of industrial facilities. During the first measurement, the temperature at the monitoring site ranged between 5.5°C and 16.1°C. The average relative humidity was 75.8%, while the mean atmospheric pressure during the monitoring period was 976 mbar. During the second measurement, the temperature at the monitoring site ranged between 7.2°C and 14.5°C. The average relative humidity was 77.2%, while the mean atmospheric pressure during the monitoring period was 981 mbar. The average wind speed was 1.54 m/s, with a prevailing northeasterly wind direction; the peak wind speed was recorded from the northern quadrant.

Fig. 3 shows a wind rose for two sets of measurements: before (April 2024) and after (April 2025) the coking plant was shut down.



(a) (b)

Fig. 3. Wind rose at the location. (a) Prior to shutdown; (b) After shutdown.

3. Results and Discussion

Table 1 summarizes results obtained in two sets of measurements: before and after the coking plant was shut down.

Component	Symbol	Prior to shutdown		After shutdown	
		Average	St. dev.	Average	St. dev.
Naphthalene (ng/m^3)	Nap	0,15	0,10	0,06	0,03
Acenaphthylene (ng/m^3)	Ane	0,42	0,21	0,09	0,06
Acenaphthene (ng/m^3)	Anl	0,04	0,01	0,02	0,01
Fluorene (ng/m^3)	Fln	0,14	0,04	0,03	0,01
Phenanthrene (ng/m^3)	Phen	1,36	0,94	0,71	0,31
Anthracene (ng/m^3)	Ant	0,38	0,12	0,08	0,05
Fluoranthene (ng/m^3)	Flt	5,45	2,05	1,39	0,91
Pyrene (ng/m^3)	Pyr	5,31	1,69	1,42	1,00
Benzo[a]anthracene (ng/m^3)	BaA	12,68	3,58	0,97	0,66
Chrysene (ng/m^3)	Chry	15,33	7,24	1,83	0,91
Benzo[b]fluoranthene (ng/m^3)	BbF	11,06	2,88	1,49	0,91
Benzo[k]fluoranthene (ng/m^3)	BkF	6,16	1,63	0,69	0,41
Benzo[a]pyrene (ng/m3)	BaP	9,18	2,94	0,97	0,76
Indeno[1,2,3-cd]pyrene (ng/m^3)	InP	7,18	2,72	1,28	0,80
Dibenzo[a,h]anthracene (ng/m^3)	DahA	1,17	1,08	0,22	0,14
Benzo[g,h,i]perylene (ng/m^3)	BghiP	6,85	2,03	1,52	0,93

Table 1. PAH concentration in the PM10 fraction of suspended particles.

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3.1. Changes in PAH concentrations following the coking plant shutdown

The shutdown of the dominant industrial source resulted in a pronounced decrease in the concentrations of all measured PAHs. Total PAH burden (Σ16 PAHs) decreased from approximately 82.86 ng/m³ to 12.77 ng/m³, corresponding to an overall reduction of about 85%. The most substantial reductions were observed for high molecular weight (HMW) PAHs typically associated with coke oven emissions, such as benzo[a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF), and benzo[a]pyrene (BaP), all of which decreased by more than 89%.

BaP concentrations declined from 9.18 ng/m³ to 0.97 ng/m³, representing a nearly 90% reduction, clearly confirming the dominant role of the coking plant as the primary source of carcinogenic PAHs prior to its closure.

Fig. 4 illustrates the concentrations of individual PAHs before and after the shutdown of the coking plant. A substantial reduction is observed across all compounds, particularly for high molecular weight PAHs. The use of a logarithmic scale highlights the systematic reduction across all compounds, spanning both low and high molecular weight species. The most pronounced decreases are observed for 4–6 ring PAHs, particularly benzo[a]anthracene, chrysene, benzo[b]fluoranthene, and benzo[a]pyrene, which are well-established markers of high-temperature coal pyrolysis.

The consistent reduction across all congeners indicates that the coking plant acted as a broad-spectrum emission source, rather than contributing selectively to specific PAH groups. The relative compression of concentration ranges after shutdown further suggests a transition from a dominant point source regime to a more diffuse multi-source system.

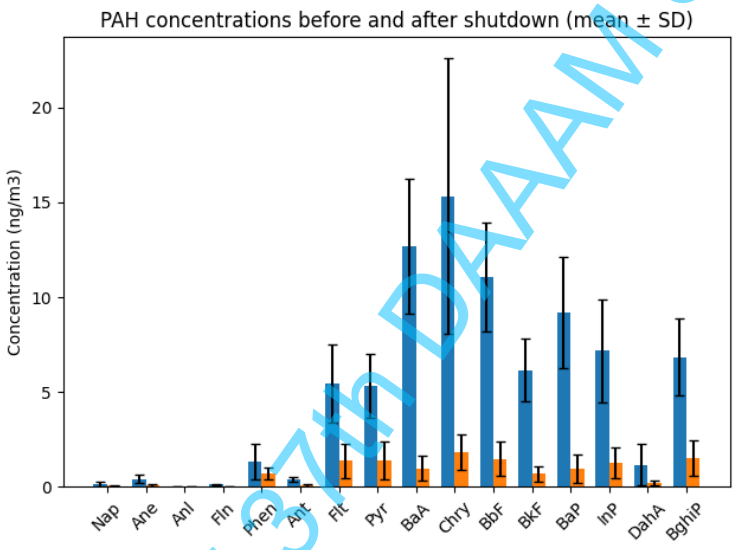

Fig. 4. Mean concentrations (± standard deviation) of PM10-bound PAHs before and after the shutdown of the coking plant.

3.2. Diagnostic Ratio Analysis and Source Fingerprint Shift

Diagnostic ratios revealed a clear transition in emission sources, as shown in Table 2. Prior to coking plant shutdown, ratios were characteristic of high-temperature coal pyrolysis, consistent with coke production. After shutdown, ratios shifted toward values indicative of mixed sources, including residential coal and biomass combustion, vehicular emissions and residual industrial activity. The most notable change is the reduction of Ant/(Ant+Phen), indicating a decline in strong pyrolytic influence.

Ratio	Before	After	Interpretation
Ant/(Ant+Phen)	0.22	0.10	Shift from pyrolytic → mixed/petrogenic
Flt/(Flt+Pyr)	0.51	0.49	Combustion remains dominant
BaA/(BaA+Chry)	0.45	0.35	Reduced coal combustion signature
InP/(InP+BghiP)	0.51	0.46	Increased traffic influence

Table 2. Diagnostic ratios.

Prior to closure, the Ant/(Ant+Phen) ratio was 0.22, significantly exceeding the 0.10 threshold characteristic of high-temperature coal carbonisation and industrial pyrolytic processes. Post-shutdown, this ratio decreased to 0.10, signalling the collapse of intense industrial coking activity and a shift toward mixed background inputs. Because anthracene photochemically degrades faster than phenanthrene during atmospheric transit, the drop to 0.10 also reflects the disappearance of fresh, high-intensity industrial plumes. The Flt/(Flt+Pyr) ratio shifted marginally from 0.51 to 0.49.

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Values exceeding 0.50 typically denote coal, wood, or biomass burning, whereas values between 0.40 and 0.50 indicate fossil fuel combustion. The post-shutdown value of 0.49 confirms that pyrogenic combustion processes – driven by residential space heating and vehicle engine exhaust – remain the overarching source of ambient PAHs in the basin, rather than unburned petrogenic sources. The BaA/(BaA+Chry) ratio declined from 0.45 to 0.35. In source apportionment literature, values above 0.35 represent heavy coal and industrial fuel burning, while values between 0.20 and 0.35 align with vehicular emissions and liquid fuel combustion. The shift to 0.35 marks the removal of the coking fingerprint and highlights the growing relative influence of mobile urban traffic. The InP/(InP+BghiP) ratio moved from 0.51 to 0.46. Ratios greater than 0.50 reflect solid-fuel combustion (coal and biomass), whereas values between 0.20 and 0.50 point toward liquid fossil fuel combustion (diesel and petrol vehicle exhaust). The shift toward 0.46 illustrates that vehicular traffic has emerged as a primary remaining driver of the ambient PAH fingerprint within the urban core.

Although atmospheric photo-oxidation (via hydroxyl radicals and ozone) can alter sensitive congeners like BaA and Ant during long-range transport, the topographically constrained nature of the Zenica valley basin minimises transit times. Consequently, the observed chemical fingerprint shift directly reflects the removal of the primary coking point source rather than atmospheric degradation artifacts.

3.3. Positive Matrix Factorization (PMF) Source Apportionment

Although full PMF modelling requires time-resolved datasets, the compositional structure of PAHs allows identification of three dominant factors:

- Factor 1: Industrial coal pyrolysis (coking plant)
  - High loadings: BaA, Chry, BbF, BkF, BaP, InP
  - Contribution: Before: dominant (~60–70%); After: minimal
- Factor 2: Traffic emissions
  - Markers: Flt, Pyr, BghiP
  - Contribution: Before: moderate; After: relatively increased share
- Factor 3: Residential combustion
  - Markers: Phen, Ant, lighter PAHs
  - Contribution: Before: minor; After: dominant residual source

The shutdown caused a structural shift in the emission profile, by eliminating a dominant point source and increasing relative importance of diffuse urban sources.

3.4. Toxic Equivalent (TEQ) Analysis

The carcinogenic potential of PAH mixtures was assessed using toxic equivalency factors (TEQs). Before shutdown, TEQ was approximately 14.3 ng/m³, and after shutdown TEQ dropped to 1.7 ng/m³, which was a reduction of 88%. The TEQ was overwhelmingly dominated by BaP and dibenzo[a,h]anthracene prior to shutdown, both strongly linked to coke production.

Figures 5 and 6 present the toxic equivalent (TEQ) contributions of individual PAHs before and after the shutdown. Prior to closure, the TEQ profile is heavily dominated by benzo[a]pyrene and dibenzo[a,h]anthracene, with additional contributions from BaA, BbF, and InP. This pattern is characteristic of emissions from coke production and high-temperature industrial processes.

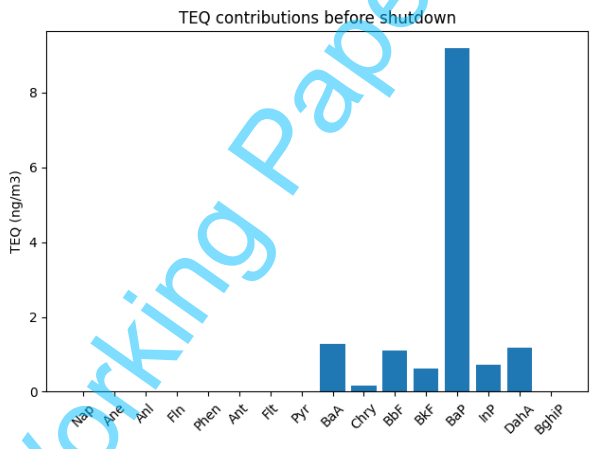

Fig. 5. Toxic equivalent (TEQ) contributions of individual PAHs prior to the shutdown.

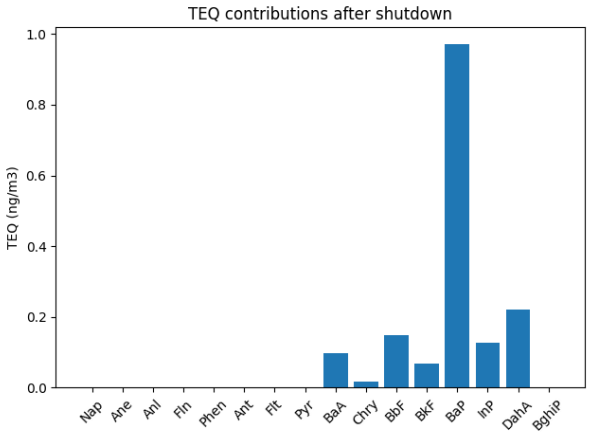

Fig. 6. TEQ contributions after the shutdown.

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Following the shutdown, a substantial reduction in absolute TEQ values is observed, accompanied by a notable redistribution of contributions. Although BaP remains the single most important contributor, its dominance is significantly reduced, and the overall TEQ profile becomes more evenly distributed among several PAHs.

This shift reflects a transition toward lower-intensity, mixed combustion sources, where no single compound dominates the carcinogenic burden. Importantly, the persistence of TEQ contributions, despite the removal of the primary source, highlights the cumulative impact of residual urban emissions.

3.5. Lifetime Cancer Risk Assessment

The World Health Organization (WHO), primarily through its International Agency for Research on Cancer (IARC), provides the most comprehensive global assessment of lifetime cancer risk. Using the World Health Organization's [7] unit risk for BaP (8.7×10^{-5} per ng/m³), lifetime cancer risk was estimated at 1.24×10^{-3} (~1 in 800) before shutdown, and 1.48×10^{-4} (~1 in 6,700) after shutdown. The risk reduction is between 88–90%. Despite this substantial improvement, residual risk levels remain above commonly accepted thresholds (10^{-6}–10^{-5}), indicating continued exposure concerns.

3.6. Role of Valley Topography

The urban-industrial valley setting plays a critical role in modulating pollutant behaviour. Limited atmospheric dispersion and frequent temperature inversions promote accumulation of pollutants, persistence of locally emitted PAHs and amplification of diffuse sources after removal of the dominant emitter. Thus, while the shutdown significantly reduced emissions, topography sustains elevated exposure levels. Fig. 7 illustrates how unfavourable terrain leads to temperature inversion.

Situated within the Bosna River valley at an elevation of 310–350 meters, Zenica is characterized by a basin-like topography where the surrounding mountainous terrain acts as a barrier to horizontal air circulation. This geomorphological configuration frequently precipitates severe thermal inversions during winter months, effectively trapping stagnant air and industrial effluents beneath a stable atmospheric lid. As Zenica is a major industrial centre, these inversions prevent the dispersion of high-volume emissions from steel production, leading to the hazardous accumulation of pollutants. Pollutant concentrations regularly surpass the safety benchmarks, manifesting as a persistent smog layer that remains localized within the valley.

Fig. 7. Temperature inversion observed on site.

4. Conclusions

The shutdown of the coking plant achieved an 80% to 90% reduction in total ambient PAHs, toxic equivalency (TEQ), and associated lifetime cancer risk, confirming its dominant role as the primary emission source prior to closure. However, diagnostic ratio and source apportionment analyses reveal a clear structural transition in the atmospheric landscape, shifting from a single high-temperature industrial coal carbonisation profile toward a complex mixture of diffuse urban sources – predominantly residential coal and biomass heating, vehicular traffic, and residual industrial activities. Unfavourable dispersion conditions inherent to the valley's basin topography further modulate pollutant behaviour, trapping these lower-intensity emissions during atmospheric inversions and sustaining non-negligible residual exposure levels.

From a policy perspective, these findings illustrate that while decommissioning major industrial point sources yields immediate public health benefits, point-source remediation alone is insufficient to achieve long-term air quality goals in

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geographically constrained basins. Because diffuse urban emissions continue to pose persistent health risks under stable atmospheric conditions, comprehensive air quality management must transition from industrial regulation to targeted urban mitigation. Empirical insights from refined source profiles will guide municipal priorities, such as expanding district heating networks, restricting low-grade residential coal sales, and implementing low-emission vehicular zones within the valley core.

To resolve inter-annual meteorological variations and seasonal extremes that short-term campaigns cannot capture, future research will establish continuous multi-year monitoring networks across distinct seasonal cycles. Deploying a high-density spatial array across varying valley elevations and land-use zones – including residential, commercial, and roadside locations – will map boundary-layer trapping dynamics and spatial pollution gradients throughout the basin. Integrating continuous multi-wavelength Aethalometry (AE33) alongside gravimetric PM2.5 and PM10 sampling will enable real-time, high-resolution optical tracking to decouple solid-fuel combustion from vehicle exhaust across changing weather regimes.

Refining source apportionment beyond diagnostic ratios will require incorporating specific organic molecular markers into Positive Matrix Factorization (PMF) receptor models. Future sampling campaigns will analyse chemical tracers such as levoglucosan for biomass burning, methoxyphenols for wood smoke, and hopanes or steranes for vehicle exhaust to isolate overlapping combustion signatures within the valley.

Finally, health risk assessment must evolve beyond isolated particle-bound PAH evaluations to capture the real-world toxicological burden of complex atmospheric mixtures. Next-step research will develop cumulative risk frameworks combining PAHs with co-emitted volatile organic compounds such as benzene, toxic heavy metals including arsenic, cadmium, and nickel, and fine particulate matter metrics (PM2.5). Furthermore, evaluating PAH bio accessibility in synthetic lung fluids (e.g., Gamble's solution) will determine the physiologically dissolved fraction of particle-bound PAHs, providing a more accurate, biologically relevant assessment of population inhalation risks in topographically constrained urban-industrial environments.

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