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PAPER

7 **Revealing the discrepancy in potential**
8 **driving factors of particulate-bound**
9 **mercury between urban and suburban**
10 **sites in a southeast asian megacity using**
11 **generalized additive model**

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14 Particulate-bound mercury (PBM) plays a critical role in atmospheric mercury
15 (Hg) cycling, yet its complex spatiotemporal variability and potential driving
16 factors remain insufficiently understood, particularly in the Southeast Asia
17 (SEA) region. This study reported year-round (May 2022 to April 2023) data of
18 PBM at an urban (Nguyen Van Cu: 59.81 ± 29.15 pg m⁻³) and a suburban site
19 (Can Gio: 26.4 ± 9.59 pg m⁻³) in southern Vietnam. Distinct seasonal trends were
20 observed at both sites, with elevated PBM concentrations in the dry season
21 (November-February), likely driven by changes in source origin and transport
22 paths of air masses. Lower PBM concentrations in the wet season (July-
23 September) may result from enhanced removal by wet deposition, whereas
24 limited rainfall in the dry season reduces this effect, leading to higher
25 concentrations. We employed Generalized Additive Models (GAM), which
26 effectively captured nonlinear relationships between PBM and meteorological-
27 chemical covariates. GAMs explained 87.7% of PBM variance in the urban area
28 and 41.6% in the suburban area, indicating better model performance in urban
29 vs suburban areas. In the urban area, metals (i.e. Cr, Sr, Pb, and V) were the
30 dominant contributors (36.7%), suggesting influence from industrial and traffic-
31 related sources. In contrast, PBM at the suburban site was mainly modulated
32 by temperature (60.8%), Zn (21.3%), and planetary boundary layer height
33 (17.9%), pointing to the significance of atmospheric processes over local

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emissions. Our findings highlight the utility of GAM in resolving complex PBM-attribution and informing targeted mercury mitigation strategies in environment interactions and indicate its potential for advancing source

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Environmental significance

Particulate-bound mercury (PBM) is a highly toxic form of atmospheric mercury, capable of regional-scale transport through complex atmospheric processes, posing substantial risks to ecosystems and public health. However, its behavior in tropical urban-suburban settings remains poorly characterized. This study provides the first year-round observational dataset of PBM in southern Vietnam, revealing distinct seasonal patterns and site-specific drivers. By employing Generalized Additive Models, we indicated the effectiveness of data-driven approaches in disentangling complex PBM-environment relationships. Our findings suggest the contrasting roles of anthropogenic emissions and meteorological processes in shaping PBM levels in urban and suburban areas. These insights are essential for improving source attribution, supporting air quality management, and informing mercury mitigation strategies in rapidly developing regions of Southeast Asia.

1. Introduction

Mercury (Hg) is a persistent heavy metal with high bioaccumulation potential, which has been shown to have significant adverse effects on human health and ecosystems.¹⁻³ Although it comprises less than 5% of total atmospheric Hg, particulate-bound mercury (PBM) plays a pivotal role in environmental processes owing to its relatively high deposition velocity and its function as a crucial vector connecting the atmosphere with other environmental compartments.^{1,4} PBM originates from a mix of anthropogenic and natural sources and shows large differences in source contributions between different regions (e.g., urban vs suburban).^{5,6} Key anthropogenic sources of PBM include combustion processes (e.g., coal combustion), metal production, and various industrial activities, while natural sources involve volcanic eruptions and natural biomass burning.⁶⁻⁹ These emissions are further influenced by atmospheric processes such as oxidation, adsorption, and condensation.¹⁰⁻¹²

East and Southeast Asia (SEA) contribute approximately 38.7% of global anthropogenic Hg emissions, driven by rapid

industrialization and urbanization.^{2,13} Studies indicated the critical role of the East and SEA region in the global Hg cycle.^{6,13,14} While research in East Asia has advanced understanding of PBM source and transport, significant uncertainties remain, particularly regarding PBM variability in SEA region.^{6,15-17} This shortage is mainly due to limited PBM data in SEA and the diverse emission sources, complex atmospheric chemistry, and region-specific meteorological conditions.¹⁸ Improved characterization of PBM behavior in SEA is essential for identifying dominant sources and informing effective regional mitigation policies.

Source apportionment of PBM has traditionally relied on multivariate techniques such as Principal Component Analysis (PCA) and Positive Matrix Factorization (PMF).¹⁸⁻²¹ While PCA reduces data dimensionality, it assumes linearity and permits negative loadings, limiting its interpretability.²² PMF addresses non-negativity but still assumes linearity, requires pre-specifying the number of sources, and is sensitive to outliers.^{20,23,24} To address the nonlinear relationships inherent in environmental data, Generalized Additive Models (GAM) have been increasingly applied in air pollution research, including atmospheric Hg.²⁵⁻²⁸ GAMs offer

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84 a flexible statistical framework that can model complex, nonlinear
85 relationships between pollutant concentrations and
86 environmental/meteorological variables without requiring prior
87 assumptions about the functional form of these relationships.^{25,26}
88 For atmospheric Hg, Wu et al.²⁷ applied GAM to quantify the
89 contribution factors on TGM concentrations in Beijing, attributing
90 47.1% of the observed TGM variability to meteorological drivers. In
91 a subsequent study, Wu et al.²⁸ employed GAM to illustrate the
92 effects of COVID-19 lockdown measures from meteorological
93 influences such as relative humidity and temperature on TGM.
94 These studies indicated the flexibility of GAM in environmental
95 modeling for capturing nonlinear environmental responses.
96 However, the use of GAM for PBM remains limited in the SEA region
97 as well as in the literature. This represents a critical gap in the
98 current understanding of PBM dynamics and their interactions with
99 environmental factors.

100 In this study, we present a year-round observational dataset
101 (May 2022 - April 2023) on PBM concentrations and the elements
102 of total suspended particles (TSP) obtained from two selected
103 monitoring sites (urban and suburban) in Ho Chi Minh City (HCMC)
104 Vietnam. From the perspective of particle size, TSP refers to the
105 total fraction of airborne particles collected by a sampler without a
106 size-selective inlet, typically capturing particles with aerodynamic
107 diameters up to ~100 μm .²⁹ Unlike $\text{PM}_{2.5}$ or PM_{10} , TSP therefore
108 encompasses both coarse and fine particles without a specific cutoff
109 limit. HCMC is a representative megacity of Southeast Asia
110 characterized by rapid urbanization, high population density, and
111 complex emission profiles. For the first time in this region, these
112 data are integrated into a GAM model to explore key atmospheric
113 processes influencing PBM. The primary aims of our study are: (1)
114 To characterize the spatiotemporal variations in PBM

concentrations across contrasting environments; (2) To investigate
the relationships between PBM, chemical constituents of TSP, and
meteorological parameters; and (3) To apportion and quantify the
relative contributions of various anthropogenic and natural
emission sources to PBM levels at both sites. The results from this
study will not only enhance the understanding of PBM dynamics in
SEA but also offer valuable insights for the development of targeted
Hg mitigation strategies in rapidly growing urban centers across the
region.

2. Materials and methods

2.1. Site description and TSP sampling

In this study, TSP samples were collected from two sites in
HCMC (Fig. S1), representing contrasting environmental contexts:
an urban site (Nguyen Van Cu - NVC; 10.762°N, 106.680°E) and a
suburban/coastal site (Can Gio - CG; 10.402°N, 106.947°E). At NVC
site, TSP sampling was conducted on the rooftop of an 11-story
building at the University of Science, VNU-HCM. The site is situated
approximately 10 meters from a major multi-lane roadway and is
influenced by anthropogenic emissions, particularly from vehicular
traffic.³⁰ In contrast, the CG site is positioned on the second floor of
Can Thanh High School in Can Gio District, approximately 50 km
southeast of central HCMC. This site located in a low-density
residential area, about 40 meters from a local road and 400 meters
inland from the coastline and is surrounded by extensive mangrove
forests (Fig. S1). It is relatively isolated from major anthropogenic
emission sources.^{31,32} These monitoring sites have been applied in
numerous atmospheric studies^{18,32,33} and are considered
representative of urban and suburban/coastal atmospheric
environments in HCMC.

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At two monitoring sites, a total of 8 TSP samples were collected each month, with sampling dates properly scheduled between the 10th and 20th of each month, from May 2022 to April 2023. At the CG site, TSP was collected using a high-volume air sampler (Model 120H, Kimoto Electric Co., Japan) operating at 600 L min⁻¹ with glass fiber filters (GFF; Advantec GA-55, 203 × 254 mm). At NVC, a high-volume sampler (Model HV-500R, Sibata Scientific Technology Ltd, Japan) operated at 500 L min⁻¹ using GFFs (Advantec; 110 mm diameter). The flow rate accuracy of both sampling instruments was lower than ±5% of the set value, as specified by the manufacturers. All filters were pre-combusted at 400°C for 8 hours to eliminate residual Hg. Filters were conditioned at 25 ± 2°C and 40 ± 5% RH for 48 hours before and after sampling. Samples were stored at -20°C until analysis. Field blanks were prepared, stored, and analyzed following the same protocol to assess potential contamination during sampling and handling. All procedures followed strict QA/QC protocols, including the use of field and laboratory blanks throughout the campaign. Detailed protocols for filter preparation and sample handling are described in previous studies.^{18,23,33}

2.2. Mercury and elements analysis

The concentrations of Hg in GFF samples collected in this study were analyzed at the laboratory of Dr. Guey-Rong Sheu, National Central University, Taiwan. Total Hg was quantified using a MA-3 Solo thermal decomposition analyzer (Nippon Instruments Corporation, Japan) in accordance with USEPA Method 7473, a widely adopted protocol for Hg determination.^{18,34,35} The thermal protocol involved an initial heating at 350°C to remove moisture and volatile organics, followed by a ramp to 850°C to decompose and release Hg. Gaseous Hg was transported to a gold amalgamation trap, thermally desorbed at 600°C, and quantified by atomic absorption spectrometry (AAS). Monthly blank values were subtracted to obtain PBM concentrations. Method blanks were < MDL (0.05 ng), with liquid standard recoveries of 99 ± 3% (n = 31). Certified reference materials (NIST SRMs 1648a and 2685c) showed recoveries of 99.8 ± 7.2% and 95.7 ± 3.0%, respectively. These results confirm the method's high precision and analytical reliability. The operational principles and standard QA/QC procedures of the analytical method have been thoroughly documented in previous literature.¹⁸

Elemental analysis was conducted via wet acid digestion followed by inductively coupled plasma mass spectrometry (ICP-MS). 20 elements (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Ni, Pb, Sb, Se, Sr, Ti, V, Zn) were quantified according to U.S. EPA Methods 6020B and 200.8 (Rev. 5.4). Samples were digested using a 2:1 (v/v) mixture of HNO₃ and HCl in Teflon vessels, employing microwave-assisted digestion.^{23,36} The digested solutions were filtered through 0.45 µm PTFE membranes and analyzed using an Agilent 7700x ICP-MS system (Agilent Technologies, USA). Analytical accuracy was validated with the certified reference material SRM 1684a, achieving recovery rates between 80% and 120% for all 20 elements. Reagent blanks were included to control contamination, and blank values were subtracted to obtain corrected concentrations. Further instrumentation and analytical details are available in our previous study.²³ In this study, elemental data were mainly applied to incorporate into the GAM framework as predictor variables to identify potential drivers of PBM variability rather than to explore the contribution to TSP sources.

In addition, meteorological variables, including temperature (T) and relative humidity (RH), were recorded at sampling sites. Planetary boundary layer (PBL) height and surface-level PM_{2.5}

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concentrations data were derived from the MERRA-2 (Modern-Era Retrospective analysis for Research and Applications, Version 2) reanalysis, which assimilates satellite observations from infrared, microwave, and GPS radio occultation sources. ^{37,38} A full system description is available in Buchard et al. ³⁹ In addition, this study employs the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model to analyze the backward trajectories of air masses to two study areas. Five-day backward trajectory simulations were conducted for each month within the study period, aiming to identify the origins and movement patterns of the air masses to these locations. ^{40,41}	View Article Online DOI: 10.1039/D5EA00094G Step 1: Data preparation The dataset used in this study was split into two subsets corresponding to the two monitoring locations. Each sample record includes concentrations of PBM and 20 elements (Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Ni, Pb, Sb, Se, Sr, Ti, V, and Zn) in TSP alongside three meteorological variables: T (°C), RH (%), and PBL (m). Precipitation was not included as a predictor in the GAM model due to the lack of high-resolution data for the entire study period and its strongly discontinuous distribution, which reduces model stability. Similarly, other meteorological parameters (i.e., wind speed, wind direction, and atmospheric pressure) were excluded due to data discontinuity and incomplete coverage across the study period. All elemental and meteorological datasets were subjected to standardized quality control procedures. Outliers beyond ±2.5 standard deviations were capped at the respective thresholds. Step 2: Variable selection To evaluate the normality of the input data, the Shapiro-Wilk test was employed to assess the conformity of each variable to a Gaussian distribution. Details of the test outcomes and the corresponding transformations applied to approximate normality prior to inclusion in the GAM model are provided in Table S1. Penalized cubic regression splines were utilized to smooth the continuous predictors, offering a flexible yet controlled means of capturing nonlinear relationships while minimizing the risk of overfitting.^{26,43} Model optimization was guided by generalized cross-validation (GCV), which balances model complexity with predictive performance.²⁸ Predictor significance was evaluated using p-values, serving as diagnostic metrics for model refinement. The initial full model included all candidate variables, followed by
2.3. Building the GAM model	
In this study, GAM was used to investigate the relationships between PBM concentrations and chemical elements, as well as meteorological variables. GAM offers a straightforward and interpretable model for representing the associations between dependent variable and independent variables. ⁴² Specifically, the model is expressed as follows:	
$g(\mu_i) = X_i\theta + f_1(x_{1i}) + f_2(x_{2i}) + \dots + f_n(x_{ni}) + \xi_i \quad (1)$	
where i denote the i th observation, g is the link function, and μ_i represents the expected value of the dependent variable, defining the relationship between the linear predictor on the right-hand side of Equation (1) and the dependent μ_i . The term $X\theta$ corresponds to the parametric component of the model, capturing the relationship with independent variables characterized by linear relationships. The function $f(x)$ denotes the smooth term associated with nonlinear predictors.	
The model construction procedure was carried out in the following steps:	

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iterative removal of non-significant terms based on their statistical relevance.²⁶⁻²⁸

Step 3: Model Evaluation

To rigorously evaluate the predictive performance and generalizability of the GAM constructed for the NVC and CG datasets, a 10-fold cross-validation procedure was implemented. This approach is widely recognized in statistical learning⁴⁴, particularly in GAM applications^{43,45}, and is increasingly employed in environmental and ecological modeling.⁴⁶⁻⁴⁸ Each dataset was randomly divided into 10 equal subsets. In each iteration, 9 subsets were used to train the model, while the remaining subset was used for validation. This process was repeated 10 times, ensuring that each subset served as the validation set once. Model performance in each fold was quantified using the coefficient of determination (R^2), and the mean R^2 across all folds was reported as a robust indicator of the predictive capability of the model.⁴⁶ In addition to assessing the adequacy of model assumptions and supporting the validity of inference, diagnostic checks were conducted, including quantile-quantile (Q-Q) plots to evaluate the normality of residuals and autocorrelation function (ACF) plots to examine residual independence (Wood, 2017).

3. Results and discussion

3.1. Characterization of PBM data

Between May 2022 and April 2023, a total of 192 TSP samples were collected from two monitoring sites in HCMC: 97 samples from NVC (urban) and 95 samples from CG (suburban). Meteorological conditions were generally similar between the two suburban sites (t-test, $p > 0.05$), with mean T of $29.0 \pm 1.2^\circ\text{C}$ at NVC and $30.9 \pm 1.7^\circ\text{C}$ at CG, and average RH values of $73.6 \pm 6.8\%$ and $69.4 \pm 5.3\%$, respectively. In contrast, PBM concentrations displayed marked spatial variation. The mean PBM concentration at NVC was $59.81 \pm 29.15 \text{ pg m}^{-3}$, more than twice the level observed at CG, which averaged $26.4 \pm 9.59 \text{ pg m}^{-3}$. This disparity highlights the stronger influence of urban emissions (i.e. anthropogenic emissions) on PBM levels within the city center.

Table 1 compares the PBM level in HCMC to other sites worldwide. In general, PBM concentrations in HCMC were considerably lower than those reported in several heavily industrialized cities.^{19,49,50} For instance, Beijing, China reported mean levels of $210.1 \pm 146.2 \text{ pg m}^{-3}$, approximately 3.5 times higher than in HCMC.⁵⁰ Similarly, Kathmandu, Nepal recorded concentrations of $850.5 \pm 926.8 \text{ pg m}^{-3}$, exceeding HCMC levels by a factor of 14 (Guo et al., 2021). Urban centers in developed regions such as Detroit, USA ($20.8 \pm 30 \text{ pg m}^{-3}$) and Seoul, South Korea ($23.9 \pm 19.6 \text{ pg m}^{-3}$) also reported higher/comparable values, respectively.^{51,52} Comparable trends were observed in Shanghai, China (560 ± 220 and $330 \pm 90 \text{ pg m}^{-3}$;49), and Xi'an, China ($640 \pm 540 \text{ pg m}^{-3}$;53). These elevated PBM concentrations have been attributed to emissions from coal combustion, metallurgical activities, and other fossil fuel-related sources.^{49,53} Despite its status as a major economic hub, HCMC (particularly NVC) appears to experience relatively limited direct impact from such heavy industrial activities compared to other megacities.

Suburban comparisons also reveal important contrasts. For example, Xiamen, China reported PBM concentrations approximately 6.7 times higher than those at CG.⁵⁴ Dhulikhel, Nepal recorded levels of $108.7 \pm 86.2 \text{ pg m}^{-3}$, about four times those measured at CG.¹⁹ Similarly, PBM levels reported at Zabrze, Poland ($65.5 \pm 53.7 \text{ pg m}^{-3}$) also exceeded the concentrations at CG,

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highlighting regional variation in emission sources and atmospheric processes.^{55,56} In contrast, the concentration observed at CG ($26.3 \pm 9.59 \text{ pg m}^{-3}$) is comparable or higher than those in cleaner or less industrialized coastal settings such as Nova Scotia, Canada ($2.32 \pm 3.09 \text{ pg m}^{-3}$), Beltsville, USA ($8.6 \pm 56.8 \text{ pg m}^{-3}$), high mountain central Taiwan ($3.1 \pm 8.5 \text{ pg m}^{-3}$), Taoyuan City, Taiwan ($18.7 \pm 86 \text{ pg m}^{-3}$), and Okinawa, Japan ($2.6 \pm 3.6 \text{ pg m}^{-3}$) (Table 1).^{3,5,17,56} These comparisons indicate the relatively moderate level of PBM at CG suggesting limited influence from heavy industrial activities and reflecting a suburban emission profile distinct from urban industrialized regions. It is worth noting that the comparisons of PBM concentrations (Table 1) were derived from measurements based on different particle size fractions (i.e., TSP, PM_{10} , and $\text{PM}_{2.5}$). Consequently, certain variability is inevitable due to the size-dependent partitioning behavior of mercury.⁵⁸ Moreover, distinct physical and chemical characteristics associated with each particle size fraction may further contribute to the observed variations in PBM levels.

Fig. 1 illustrates the relative distribution of element composition in TSP collected from urban and suburban environments. In urban site, Ca was the most dominant element accounting for 41.12%, followed by Fe (21.98%), Al (13.26%), Ti (10.07%), and Mg (7.96%) (Fig. 1a). This compositional profile indicates a predominant influence from crustal and anthropogenic contributions, particularly construction, vehicular emissions, and resuspended road dust, characteristic of densely populated urban areas.^{18,30,36} The elevated levels of Fe and Al indicate mineral dust inputs, while K and Mg may reflect inputs from biomass burning and soil-derived particles.^{8,23,59} Additionally, trace elements such as Zn (2.97%) and Cu (10.51%), found within the category of other elements also suggest the influence of industrial activities or traffic related sources.⁶⁰⁻⁶² Conversely, in the suburban area, although Ca remained the dominant element (45%), Mg emerged as the second most abundant at 24.87%, surpassing Fe (6.68%) (Fig. 1c). This shift suggests a different source profile, potentially related to greater vegetation cover, proximity to the coast, lower human activity intensity, and reduced construction emissions.^{32,36,63} Al (9.69%) and K (7.86%) also contributed significantly, along with Cu (3.72%) and Zn (1.3%), which may originate from local anthropogenic activities such as agricultural practices, domestic combustion, or long-range atmospheric transport.^{8,23,64}

The distribution of trace elements in TSP at the urban and suburban areas shows distinct differences in the distribution and relative abundance of elements between urban and suburban areas. In the urban area, elements such as Pb (18.09%), Mn (15.66%), and Cu (10.51%) dominate (Fig. 1b), reflecting the influence of anthropogenic activities, particularly emissions from traffic, industrial activities, and construction.⁶⁵⁻⁶⁷ The correlation analysis among the measured elements was conducted to explore potential associations and shared origins (Tables S2 and S3). Meanwhile, strong correlations among transition metals (i.e., Cr, Mn, and Co) further confirm their common anthropogenic origin ($r = 0.82-0.91$; $p < 0.05$). Likewise, the significant associations between Pb-Cd ($r = 0.84$), As-Se ($r = 0.79$), As-Cd ($r = 0.79$), and Cd-Sb ($r = 0.77$) suggest concurrent emissions from combustion sources and industries using heavy metals.^{23,32,36} In contrast, in the suburban area, elements such as Ti (23.18%) and Mn (18.75%) are more prominent (Fig. 1d), indicating a greater influence from natural sources such as soil dust and vegetation.^{23,32} The moderate correlations among crustal elements, for instance, Al-K ($r = 0.55$), Al-Ti ($r = 0.53$), and Al-Ca ($r = 0.52$), further confirm the predominance of soil-derived sources. Meanwhile, the significant correlations

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among Fe-Mn ($r = 0.69$), Ti-Mn ($r = 0.74$), Zn-Fe ($r = 0.60$), and Zn-Mn ($r = 0.51$) likely indicate the influence of anthropogenic activities, possibly traffic-related emissions (Table S3). Additionally, Pb (13.02%) is present at lower levels, indicating less contribution from industrial emissions compared to the urban area.^{23,32,36} These differences illustrate spatial variability in elemental composition between the two environments, suggesting differences in population density and surrounding land use characteristics.^{32,63,68} These observations are crucial for understanding the behavior of PBM, as the chemical environment in which PBM exists can influence its transformation, transport, and potential health impacts.^{11,18,69} Therefore, characterizing elemental distribution patterns is a key foundation for interpreting PBM dynamics in complex urban-suburban transitions.

3.2. Seasonal variation and potential driving factors

Fig. 2 reveals a pronounced seasonal pattern of PBM concentrations at both urban and suburban sites in HCMC. During the major rainy season (July-September), mean PBM concentrations were lower, averaging $38.5 \pm 12.4 \text{ pg m}^{-3}$ at NVC and $19.8 \pm 10.7 \text{ pg m}^{-3}$ at CG. In contrast, the major dry season (November-February) exhibited elevated levels at the urban site ($109.0 \pm 50.9 \text{ pg m}^{-3}$), and suburban ($40.6 \pm 22.2 \text{ pg m}^{-3}$). The seasonal variation in PBM concentrations observed in both urban and suburban sites in HCMC reflects the combined influence of rainfall-driven removal and seasonal air mass transport dynamics.^{18,23,32} During the rainy season, frequent and intense rainfall events associated with the Southwest monsoon promote efficient wet scavenging of PBM, resulting in generally lower concentrations. Rainfall facilitates the removal of PBM, especially in tropical environments where convective systems are prevalent.^{4,14,18} In contrast, during the dry season, limited rainfall reduces atmospheric cleansing, allowing PBM to accumulate in the boundary layer, resulting in an elevation of PBM levels. In addition, meteorological parameters such as T and RH remained relatively constant across seasons ($p > 0.1$, ANOVA), suggesting that local meteorological variability had limited direct influence on PBM seasonal dynamics.

Furthermore, seasonal variations in prevailing air mass transport patterns, governed by the East Asian monsoon system, play a critical role in shaping atmospheric composition in HCMC.^{32,33,70} Results from backward trajectory analyses (Fig. S2, S3) indicate that during the dry season, air masses predominantly originate from the northeast, possibly originating from highly industrialized regions across continental East Asia.^{14,71,72} These air masses could carry high PBM levels, enhancing background PBM levels across both urban and suburban sites in HCMC.^{5,17,70} In contrast, during the rainy season, the sampling sites are primarily influenced by southwesterly air masses originating over the ocean (Fig. S2, S3), which transport relatively clean maritime air to HCMC. Similar seasonal atmospheric Hg trends linked to continental outflow have been reported across East Asia, including Taiwan, Japan, South Korea, and Vietnam.^{5,17,70,73}

Notably, during the rainy season, a pronounced difference in PBM concentrations was observed between the urban and suburban sites in June, with the NVC site recording a seasonal maximum of $76.5 \pm 29.5 \text{ pg m}^{-3}$, approximately 64% higher than its rainy season mean value (Fig. 2). In contrast, no corresponding peak was detected at the CG site (Fig. 2). Analysis of backward trajectories and surface $\text{PM}_{2.5}$ revealed comparable air mass origins for both sites during this period, suggesting that long-range atmospheric transport was not the primary contributor to the

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443 elevated PBM levels at NVC (Fig. S2, S3). Results from surface PM_{2.5} 444 from MERRA-2 further support this conclusion (Fig. S4). The 445 absence of a similar increase at the suburban site supports the 446 interpretation that local accumulation processes, likely driven by 447 stagnant meteorological conditions, predominated at the urban 448 site.^{32,33} This interpretation is further supported by meteorological 449 data, which show that June experienced the lowest mean wind 450 speed of the season (1.1 m s⁻¹), considerably lower than values 451 recorded in other months (1.5–1.7 m s⁻¹), favoring the buildup of air 452 pollutants. In summary, the seasonal variability of PBM in HCMC is 453 governed by a complex interplay of local accumulation processes, 454 monsoonal shifts in prevailing air mass origin, and regional-scale 455 pollutant transport. While local emissions dominate during 456 stagnant conditions in the wet season, transboundary transport 457 becomes increasingly significant during the dry season, highlighting 458 the importance of considering both local and regional scales in 459 atmospheric Hg management strategies. 460 3.3. Analysis and evaluation of GAM performance 461 Based on the variable selection results presented in Table S4, a 462 total of 19 independent variables (PBL height and 18 elements) 463 were retained for the NVC dataset, while only 3 variables (PBL, T, 464 and Zn) were retained for CG ($p < 0.05$, summary function). Model 465 performance metrics are summarized in Table S5. The GAM 466 indicated strong correlations between the selected predictors and 467 PBM concentrations, with R^2 reaching 90.8% for NVC and 94.5% for 468 CG, indicating robust initial fits.^{28,43,74} However, the explanatory 469 power of the models, as measured by adjusted R^2, differed 470 markedly: 0.73 at NVC and only 0.32 at CG (Table S5). The adjusted 471 R^2 of 0.73 at NVC indicates a strong and reliable model 472 performance, suggesting that the selected predictors captured	473 most of the variability in PBM concentrations at this site. In contrast, 474 the much lower adjusted R^2 at CG implies a weaker model fit, 475 potentially due to unaccounted site-specific factors or greater 476 variability in local conditions. This contrast suggests a reduced 477 ability to generalize the influencing factors of PBM concentrations 478 in suburban environments.^{75,76} 479 The higher explanatory capacity of the NVC model can be 480 attributed to the broader spectrum of input variables, which likely 481 captured key emission sources and meteorological influences. In 482 contrast, the limited predictor set for CG constrained the model's 483 ability to represent PBM variability. Comparative assessment with 484 prior studies further highlights the improved performance of the 485 current urban model. The statistical model explained 87.7% of the 486 variance in PBM concentrations, with the adjusted R^2 reaching 0.73 487 at the NVC site, indicating superior performance compared to the 488 model of TGM in the Beijing-Tianjin-Hebei region, which achieved 489 an adjusted $R^2 = 0.62$ and explained 63.2% of the variability.²⁸ In 490 contrast, at the CG site, the statistical model showed a considerably 491 lower predictive capacity, explaining 41.6% of the variance in PBM 492 concentrations, with the adjusted R^2 reaching 0.32, consistent with 493 the findings of Wu et al.²⁷, where the model explained 56.7% of the 494 TGM variability (adjusted $R^2 = 0.551$). It should be noted that our 495 model was developed for PBM, while the referenced studies 496 modeled TGM, which may inherently differ in their atmospheric 497 behavior, influencing model sensitivity and explanatory capacity. 498 These results emphasize the critical importance of 499 incorporating a diverse array of explanatory variables. The limited 500 inclusion of local environmental indicators (e.g., land use, 501 vegetation cover, potential emission sources) may have further 502 hindered model performance at CG.^{25,68,77} Diagnostic analyses from

Fig. S5 and S6 indicate that both GAM models at the NVC and CG sites satisfy key statistical assumptions and are well-suited to the environmental characteristics of each location. At NVC, the residuals follow a normal distribution, are homogeneous and dispersed around zero, and the predicted values align closely with observations, demonstrating high model accuracy and strong predictive capacity. At CG, the residuals also exhibit a normal distribution, with a symmetric histogram and Q-Q plot, and no systematic bias across fitted values. Overall, both models are statistically reliable and highlight the flexibility of GAM in analyzing PBM across contrasting environmental settings.^{43,78,79}

3.4. Relationship analysis results

Fig. 3 and S8 show the nonlinear relationships between PBM and other parameters (i.e. elements and meteorology) at the NVC site, revealing complex interactions between emission sources and atmospheric processes. Several crustal (e.g., Al, Ti; Fig. S8) and anthropogenic elements (e.g., Cd, Co, Cr, Cu, Pb, Sb) exhibited statistically significant positive associations with PBM levels, suggesting their potential role in PBM-enriched source sectors such as traffic-related emissions, coal combustion, and industrial activities at the sampling site.^{18,23,32} This finding aligns with previous GAM-based studies, where key emission sectors (e.g., cement power plants) exhibited significant relationships with Hg levels, emphasizing the dominance of anthropogenic drivers on atmospheric Hg variation.^{27,28} By contrast, the PBL showed an inverse relationship with PBM (Fig. 3), consistent with enhanced vertical mixing and dilution under higher PBL conditions.^{30,54} Similar findings were reported by Wu et al.²⁸ in Beijing, China, indicating that elevated PBL contributed to reductions in atmospheric Hg concentrations due to improved atmospheric dispersion.

Some elements (i.e., As, Ba, Fe, Mn, Ni, and Se) showed threshold-dependent nonlinearities. For instance, As and Ba displayed negative relationships with PBM at lower concentrations ($<0.4 \text{ ng m}^{-3}$ and $<9 \text{ ng m}^{-3}$, respectively; Fig. S8) but transitioned to positive relationships above these thresholds, indicating potential shifts in dominant sources or physicochemical partitioning behavior at higher loadings. These turning points may reflect saturation kinetics or source mixing, where low concentrations are dominated by background or natural sources, whereas elevated levels originate from combustion-related emissions.^{10,12,80,81} Moreover, elements such as K, Sr, V, and Zn exhibited highly nonlinear trends with multiple inflection points (Fig. S8), suggesting their involvement in complex multiphase reactions or simultaneous contributions from diverse sources (e.g., biomass burning, traffic, and re-suspension processes). For instance, the GAM-based spline revealed that Zn exhibited a positive association with PBM concentrations at lower levels (up to $\sim 125 \text{ ng m}^{-3}$), after which the relationship plateaued or slightly declined (Fig. 3). This trend may reflect atmospheric saturation effects, particle-phase condensation limitations, or increased partitioning into coarse-mode particles at higher Zn concentrations, which may be less efficient at binding PBM.¹⁶ Additionally, in humid atmospheric conditions, Zn may facilitate heterogeneous reactions on particle surfaces or act as a catalyst in redox transformations of oxidized Hg.^{82,83} These findings imply the dual role of Zn as both a source tracer and a potential participant in secondary atmospheric processes influencing PBM formation and stability. Given the ubiquitous presence of traffic emissions in urban environments, the role of Zn as a proxy for this source category highlights the importance of traffic-related activities as a critical and persistent contributor to atmospheric Hg cycling.

At the CG site, meteorological variables, including T and PBL showed similarly complex effects on PBM (Fig. 4). T showed nonlinear association with PBM (Fig. 4a), characterized by a negative response between 26–30 °C and a slight increase above 30 °C. The PBM–T relationship is inherently complex, as T controls the gas–particle partitioning of Hg(II), lower temperatures favor particle-bound fractions, whereas higher temperatures enhance gaseous forms.^{9,12} Moreover, temperature variations may reflect shifts in air mass origins (e.g., land-sea breeze), further influencing PBM levels.^{12,84,85} The inverse relationship between PBL and PBM became evident above 330 m, reinforcing the role of boundary layer dynamics in pollutant dilution. Notably, Zn at CG revealed a concentration-dependent relationship with PBM, positively correlated below 125 ng m⁻³ but negatively correlated beyond this threshold. This may indicate distinct PM binding capacities and interactions under varying atmospheric chemical regimes. Similar concentration-reversal effects of RH on TGM were reported by Wu et al.²⁸, where RH >80% enhanced aqueous phase reduction processes and TGM re-emission, further illustrating the nonlinear behavior of Hg species in atmospheric systems.

In general, these results indicate the multifactorial nature of PBM variability, governed by a combination of potential emission source profiles and meteorological controls at both urban and suburban sites in HCMC. However, it should be acknowledged that several nonlinear relationships revealed in the GAM remain partially unexplained, particularly those involving threshold-dependent behaviors and inflection points. These complexities may stem from unmeasured variables, insufficient temporal resolution, or limitations in chemical speciation data. Consequently, our ability to fully interpret the integrated influence of chemical and meteorological predictors on PBM remains constrained.

3.5. Contribution rates of independent variables, implications for potential driving factors on PBM variabilities

Table S6 shows the proportional contributions of factors influencing PBM concentration variability at NVC and CG sites, as determined by the VarImp function in the GAM model.^{25,27,28} The relative importance of independent variables reveals striking contrasts between the two monitoring environments, highlighting the distinct roles of emission sources and meteorological controls in shaping PBM dynamics.

At the CG suburban site, meteorological conditions overwhelmingly dominated PBM variability, collectively accounting for 78.7% of the total variance (Fig. 5). Among these, T alone explained 60.8%, consistent with a strong positive, though nonlinear, association observed in the GAM spline. In addition, the PBL contributed 17.9% to PBM variation, reflecting the dispersive capacity of elevated mixing layers to dilute PBM concentrations, as noted in previous studies.^{32,33} These results indicate that in areas with limited direct emissions, PBM levels are primarily modulated by atmospheric dynamics.

In contrast, meteorological factors played a minor role at the urban NVC site, explaining only 1.2% of the variance (Fig. 5). This suggests that urban PBM levels are largely dominated by localized anthropogenic emissions. Indeed, anthropogenic sources contributed over 87.7% of the PBM variability at NVC, reflecting the complex contribution of combustion, industrial, and traffic-related emissions in a densely populated setting. Among combustion-related sources, several elements showed high contributions to PBM at NVC, including Pb (8.8%), V (8.3%), Cd (5.5%), and As (5.6%), all commonly associated with fossil fuel burning.^{86,87} Pb and V are particularly indicative of the use of heavy oils and fossil fuels in both

transportation and industrial processes.^{36,59,65} K (4.6%), Se (4.6%),
and Sb (3.6%) also played significant roles within the combustion
related source group, although they are often considered to
originate from mixed sources. K is a well-known tracer of biomass
burning, commonly found in plant ash and produced from the
combustion of wood, crop residues, or other organic materials.^{88,89}
The prominent presence of K suggests the possible influence of local
open burning or resuspension of ash-derived dust within the urban
environment. In contrast, Se is typically found in emissions from
heavy oil and coal combustion, particularly from power plants and
industrial facilities using fossil fuels.⁹⁰⁻⁹²

These elements also exhibited strong and significant positive
associations in GAM splines, suggesting their diagnostic importance
in PBM variation. In addition, industrial-related elements, including
Sr (8.9%); Ni (5.8%), Co (3.9%), and Ba (0.6%), were also important
predictors at NVC. These originate from diverse industrial activities
such as steel production, alloy and battery manufacturing, pigment
and flame-retardant use.^{36,61,93,94} Although anthropogenic
influences were predominant, natural sources contributed a non-
negligible 11.1% to the PBM variation at the NVC site. Specifically,
elements such as Ti (5.4%), Fe (4.2%), and Al (1.5%), commonly
associated with crustal material, re-suspended soil dust, or biogenic
emissions, were linked to natural origins.^{36,63} These elements likely
reflect background processes such as wind-driven dust
resuspension, long-range transport of mineral aerosols, and
interactions with natural surfaces. While their contributions were
smaller than those from combustion or industrial sources, they
provide a stable background signal and may influence PBM behavior
through surface reactivity or chemical partitioning mechanisms.
Therefore, the combined effect of both anthropogenic and natural

inputs should be considered in assessing PBM dynamics and
developing targeted control strategies.

Furthermore, non-combustion traffic-related elements (i.e. Cr,
Mn, Cu, and Zn) accounted for 27.5% of PBM variability at NVC.
These metals are predominantly released from brake and tire wear,
engine oil leakage, and road dust re-suspension.^{18,62,95} At the CG
site, Zn was the only elemental contributor to PBM variability
(21.3%), suggesting that localized emissions from traffic-related
sources remain relevant even in suburban settings. This is
consistent with previous findings indicating that Zn can persist
regionally due to its semi-volatile behavior and association with
fine-mode aerosols.^{96,97}

In general, the distribution of PBM drivers across the two sites
reflects fundamental environmental contrasts. While the urban
NVC site is characterized by intensive anthropogenic emissions from
multiple sectors, the suburban CG site is primarily governed by
meteorological processes, with localized Zn emissions playing a
secondary role. These insights support the development of site-
specific air quality control strategies: urban areas should prioritize
emission mitigation (especially from traffic and fuel combustion),
whereas regional management must consider atmospheric mixing
and temperature-driven re-emission processes.

4. Conclusion

This study presents the first year-long dataset of PBM at urban
and suburban sites in HCMC, providing novel insights into PBM
dynamics in a tropical SEA megacity. Distinct spatial and temporal
variations were observed, with higher mean PBM concentrations at
the urban site ($59.81 \pm 29.15 \text{ pg m}^{-3}$) compared to the suburban site
($26.4 \pm 9.59 \text{ pg m}^{-3}$). Seasonal trends revealed elevated levels during

the dry season. By applying GAM, this study successfully quantified the nonlinear influences of chemical and meteorological parameters on PBM. The GAM explained 87.7% of PBM variance at the urban site, compared to 41.6% at the suburban site, indicating its greater predictive power in emission-rich environments. At the urban site, anthropogenic emissions dominated PBM variability, with industrial and combustion-related elements such as As, Cd, Pb, V, K, Se, and Sb contributing 41%. In contrast, PBM at the suburban site was primarily modulated by temperature (60.8%), Zn (21.3%) and planetary boundary layer height (17.9%), reflecting the stronger role of atmospheric processes in low-emission areas. The findings implied the efficacy of GAM in unraveling complex PBM-environment interactions, offering a robust framework for source apportionment. Importantly, the observed site-specific drivers suggest that urban areas should prioritize emission controls targeting traffic and industrial sectors, while suburban management strategies should consider meteorological influences and regional transport mechanisms. Overall, this work provides critical evidence for tailoring Hg mitigation policies to local environmental contexts and emphasizes the utility of flexible, data-driven modeling approaches in advancing atmospheric Hg research and air quality management. On the other hand, we acknowledge certain analytical limitations specific to the current study. Upcoming studies should incorporate expanded datasets that integrate higher time-resolution measurements, detailed chemical speciation of PM and more complete meteorological parameters, together with advanced analytical approaches such as integrated PMF-GAM modeling.^{98,99} Such developments will enable a more robust identification of source contributions, thereby improving the mechanistic understanding of PBM dynamics in complex atmospheric environments.

Author contributions

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Conflicts of interest

The authors declare no conflicts.

Data availability

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

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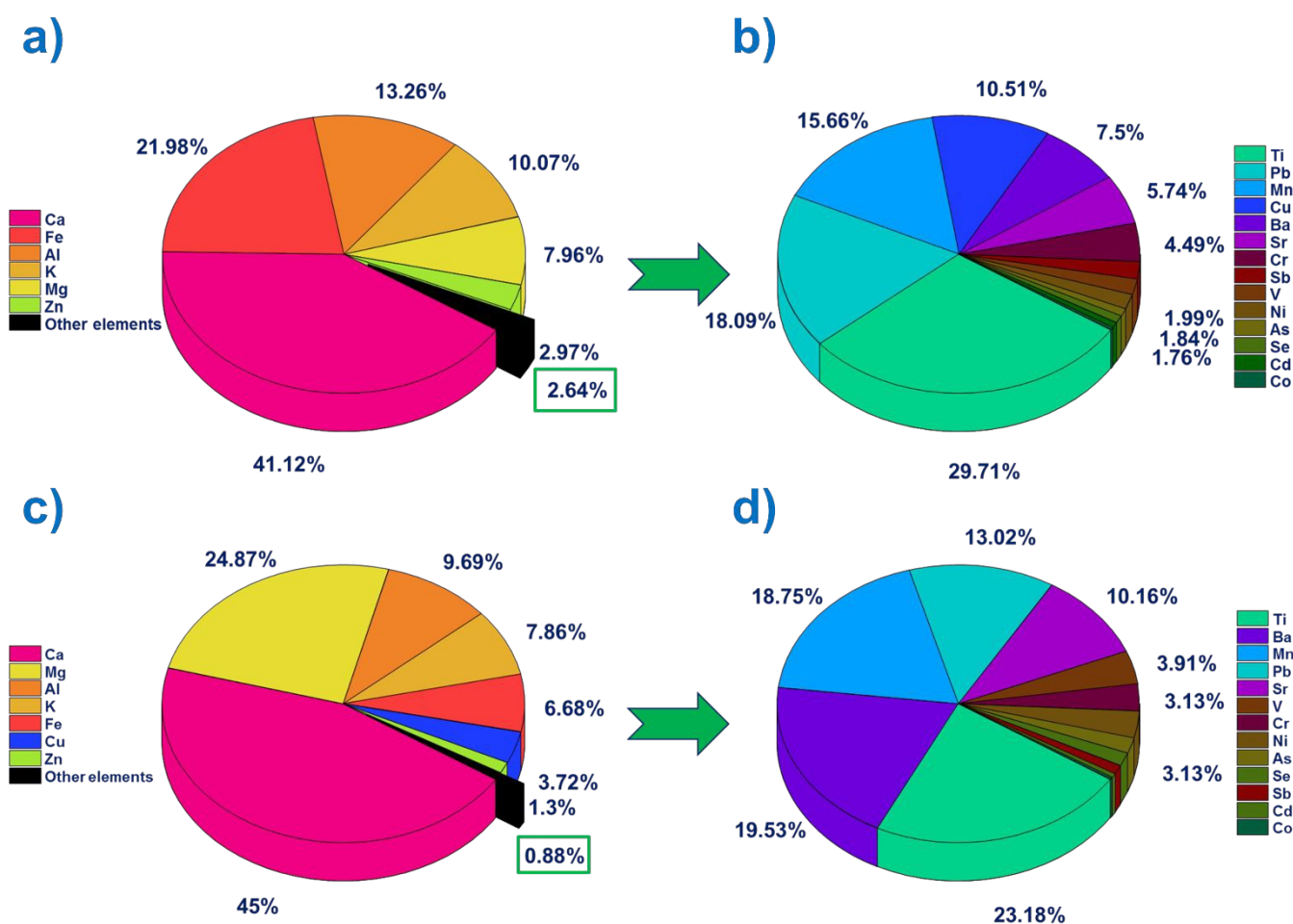


Fig. 1 Pie charts illustrating the percentage (%) of elements TSP samples collected from urban and suburban areas: (a) and (c) show major elements in urban and suburban areas, respectively; (b) and (d) show trace elements in urban and suburban areas, respectively.



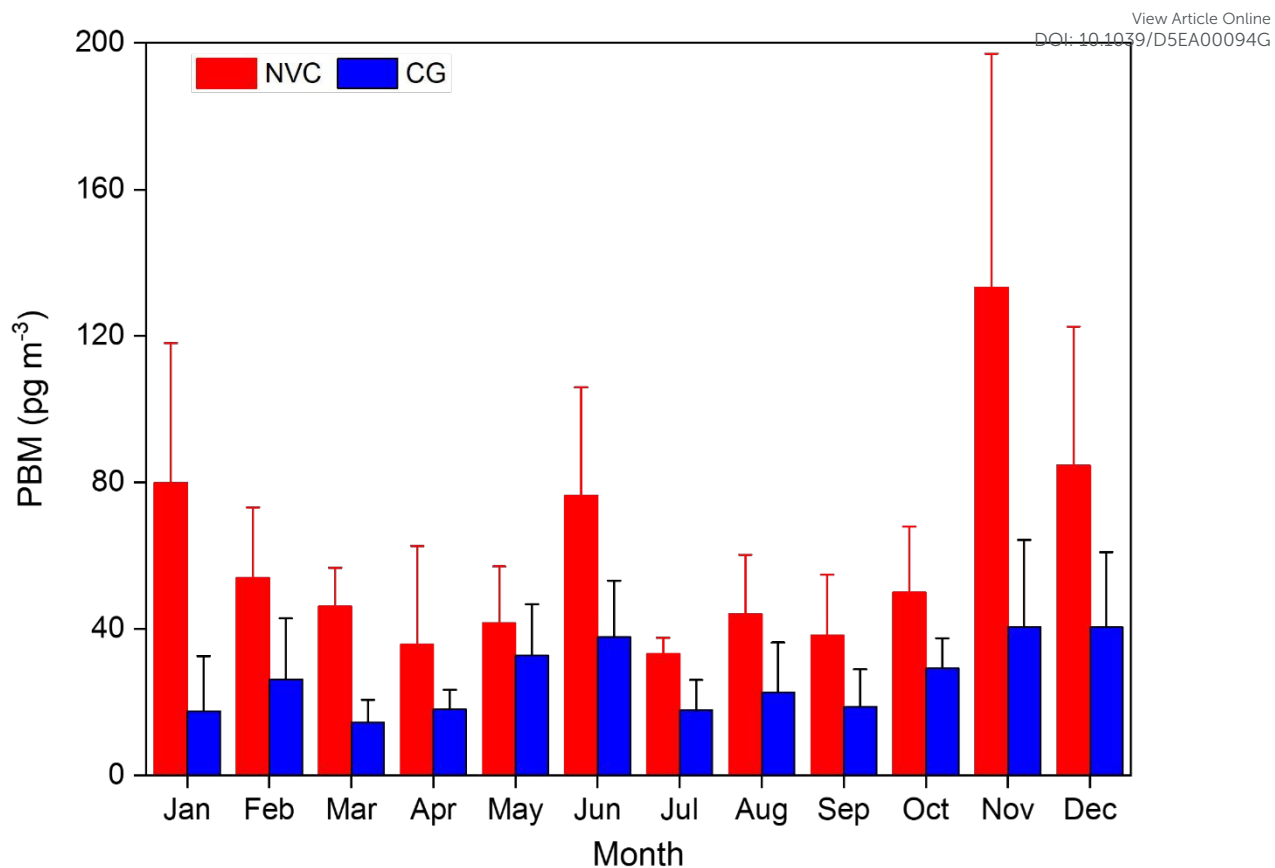


Fig. 2 Monthly variation of PBM concentration (pg m⁻³) at two sites, Nguyen Van Cu and Can Gio, from May 2022 to April 2023



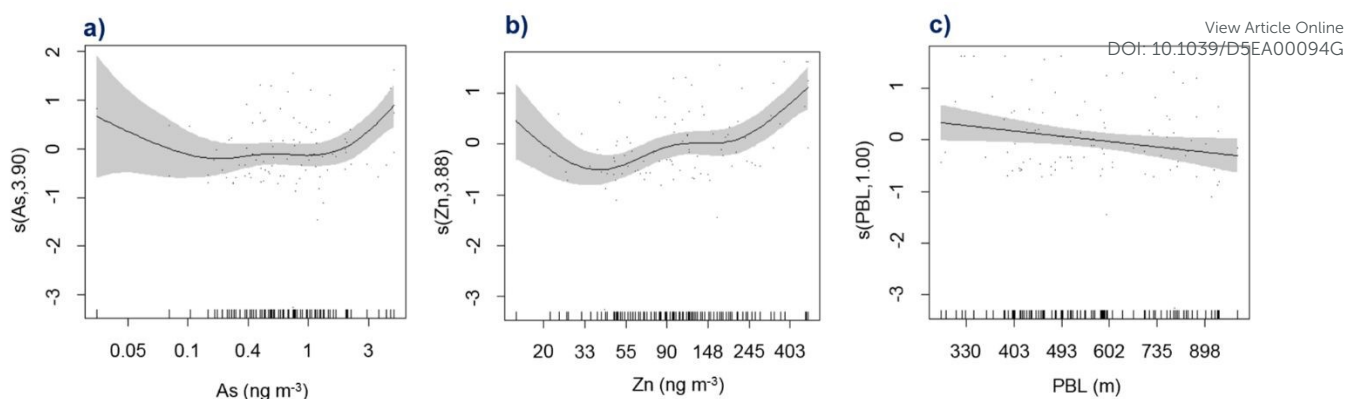


Fig. 3 Spline plots illustrate the relationships between PBM concentration and elemental concentrations at the NVC monitoring site: (a) As, (b) Zn, and (c) PBL - the meteorological parameter. The y-axis represents the spline values of PBM as a function of each independent variable, and the x-axis corresponds to the observed values of each independent variable. The solid line indicates the fitted spline, while the shaded area around it represents the 95% confidence interval for the predicted response. Tick marks on the x-axis show the distribution of the data points. The number in parentheses in each y-axis label denotes the estimated degrees of freedom (EDF). Dots in the plots represent residual values.

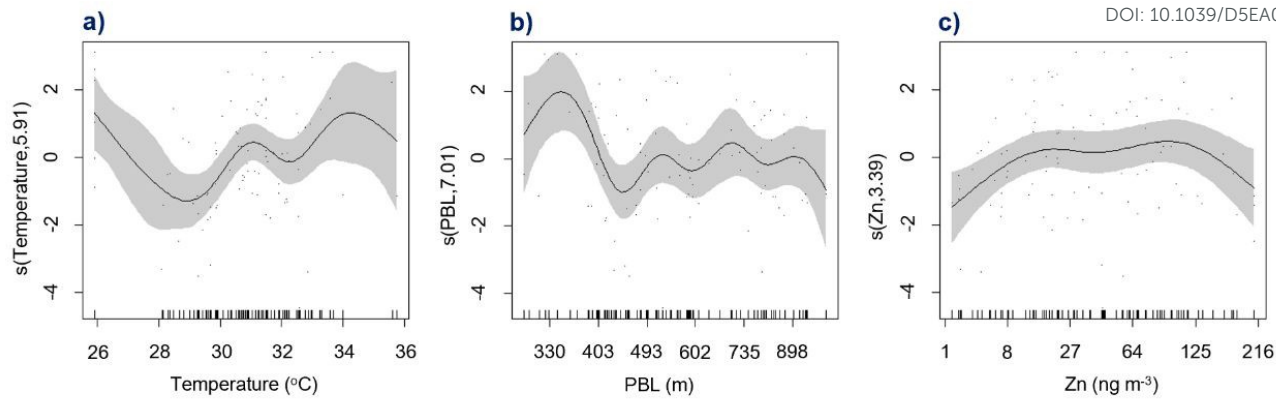


Fig. 4 Spline plots illustrate the relationships between PBM concentration and elemental concentrations at the CG monitoring site: (a) Temperature, (b) PBL, and elemental concentration: (c) Zn.

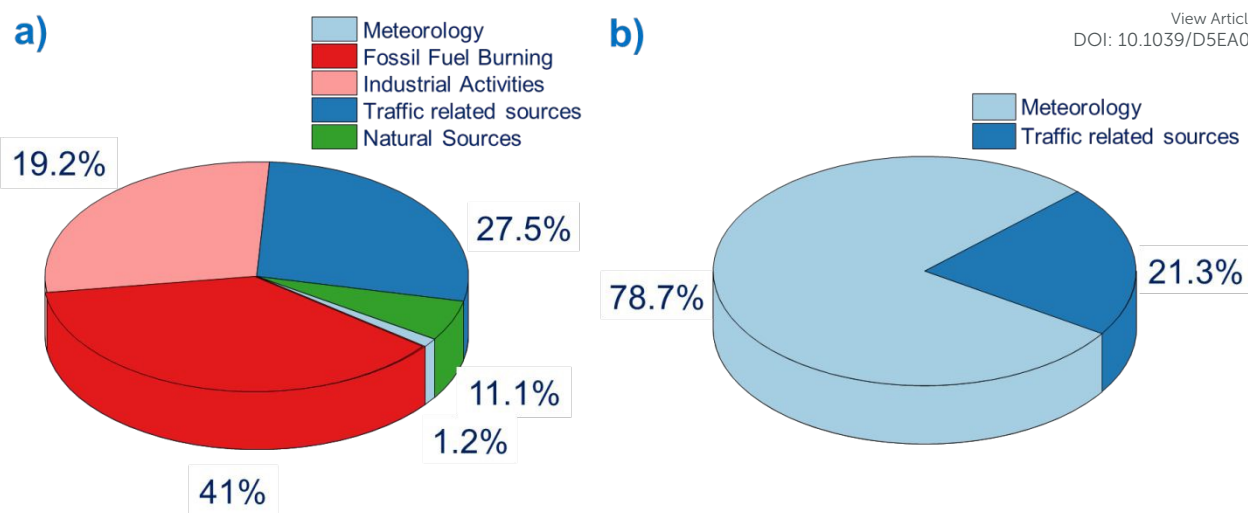


Fig. 5 Source category contributions to PBM variability at the two sampling sites (a) urban and (b) suburban

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Table 1. Comparison of PBM concentrations (pg m⁻³) in samples with values reported in other countries worldwide.

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Site	Size	Type	Year	PBM (pg m ⁻³)	References
NVC, Vietnam	TSP	Urban	2022-2023	59.81 ± 29.15	This study
CG, Vietnam	TSP	Suburban	2022-2023	26.4 ± 9.59	This study
Beijing, China	TSP	Urban	2016 - 2017	210.1 ± 146.2	50
Kathmandu Valley, Nepal	TSP	Urban	2013-2014	850.5 ± 926.8	19
Shanghai, China	TSP	Urban	2004-2006	560 ± 220, 330 ± 90 (2 sites)	49
Xi'an, China	TSP	Urban	2010-2013	640 ± 540	53
Detroit, USA	PM _{2.5}	Urban	2006	20.8 ± 30	51
Seoul, Korea	PM _{2.5}	Urban	2005-2006	23.9 ± 19.6	52
Xiamen, China	TSP	Suburban	2012-2013	174.1 ± 160.9	54
Dhulikhel, Nepal	TSP	Suburban	2018	108.7 ± 86.2	19
Okinawa, Japan	PM _{2.5}	Suburban	2009-2018	2.6 ± 3.6	17
Zabrze, Poland	TSP	Suburban	2013	65.5 ± 53.7	55
Zabrze, Poland	PM ₁₀	Suburban	2013	63.6 ± 53.0	55
Nova Scotia, Canada	PM _{2.5}	Suburban	2010-2011	2.32 ± 3.09	100
Beltsville, Maryland, USA	PM _{2.5}	Suburban	2007-2015	8.6 ± 56.8	56
High mountain in central Taiwan, Taiwan	PM _{2.5}	Suburban	2009-2016	3.1 ± 8.5	3
Taoyuan City, Taiwan	PM _{2.5}	Suburban	2017-2018	18.7 ± 86.8	5

Data availability

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

