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Spatial and Seasonal Distribution of PM_{10}-Bound Heavy Metals in Built-up and Agricultural Areas of Rawalpindi, Pakistan

Umair Rashad[1,2], Aansa Rukya Saleem[1,2], and Tahir Hayat Malik[3]

[1]*Department of Earth and Environmental Sciences, Bahria School of Engineering and Applied Sciences, Bahria University, Islamabad, Pakistan*

[2]*Center of Research Excellence-Earth, Climate and Environment, Bahria University Islamabad, 44000, Pakistan*

[3]*Department of Environmental Sciences, COMSATS University Islamabad, Abbottabad Campus, Pakistan*

Corresponding author: arukya.buic@bahria.edu.pk

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Abstract

Atmospheric particulate matter PM_{10} is an important carrier of toxic heavy metals and is associated with significant risks to environmental quality and human health. This Study describes the spatial and seasonal distribution of lead (Pb), chromium (Cr), cadmium (Cd), and copper (Cu) bound to PM_{10} in the built-up and agricultural areas of Rawalpindi, Pakistan. This is, to the best of our knowledge, one of the first studies comparing the PM_{10} -bound heavy metal concentrations of built-up and agricultural environments of Rawalpindi using year-round monitoring. Out of these, six monitoring sites were chosen to collect monthly samples of PM_{10} from May 2022 to April 2023 for further analysis of Pb, Cr, Cd, and Cu by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). Clear seasonal variation was observed in all the studied metals with increased concentrations in winter months and decreased concentrations in summer months. Lead was the most dominant heavy metal in both land use categories, followed by Cu, Cr, and Cd. The mean concentrations at the built-up sites were 0.61 ± 0.19 μg m^{-3} (Pb), 0.08 ± 0.04 μg m^{-3} (Cu), 0.02 ± 0.02 μg m^{-3} (Cr), and 0.008 ± 0.006 μg m^{-3} (Cd), while the corresponding concentrations at the agricultural sites were 0.64 ± 0.16 μg m^{-3}, 0.09 ± 0.04 μg m^{-3}, 0.02 ± 0.02 μg m^{-3}, and 0.012 ± 0.015 μg m^{-3}, respectively. The results indicate that the average concentrations of Pb, Cu, and Cd were slightly higher in agricultural sites, while the concentrations of Cr were comparable between the land use categories. The observed seasonal variation is probably related to reduced atmospheric dispersion and increased accumulation of pollutants in winter, while lower concentrations in summer are indicative of better atmospheric mixing and wet deposition. Land-use characteristics and previous studies suggest possible emission sources including vehicle traffic, re-suspension of road dust, biomass burning, agricultural activities and industrial emissions. However, these sources are inferred from land-use characteristics and previous studies and not confirmed by source apportionment analysis. The current study provides baseline information about the spatial and seasonal distribution of heavy metals associated with PM_{10} in Rawalpindi and underscores the necessity for further monitoring and effective emission control techniques to reduce particle pollution and protect human health.

***Keywords*:** PM_{10}; Heavy metals; Lead; Chromium; Cadmium; Copper; Seasonal variation; Built-up area; Agricultural area; ICP-OES; Rawalpindi; Pakistan.

1. Introduction

Atmospheric particulate matter (PM) is one of the most important air pollutants worldwide for environmental quality, ecosystem sustainability, and human health. Increased vehicular traffic, rapid urbanization, industrialization, and population growth have led to a significant increase in the concentration of airborne particulate matter in many developing

countries, particularly in South Asia (WHO, 2021; HEI, 2024). Among different particle fractions, particulate matter with an aerodynamic diameter ≤10 μm (PM_{10}) is of particular concern because it can remain suspended in the atmosphere for extended periods, penetrate the upper respiratory tract, and transport toxic inorganic constituents over considerable distances. PM_{10} is also important for atmospheric chemistry, visibility, climate forcing, and ecosystem functioning, besides its direct health impacts.

The environmental significance of PM_{10} is not only related to its mass concentration, but also to its chemical composition that is strongly associated with its toxicity. Complex mixtures of crustal minerals, organic compounds, elemental carbon, secondary inorganic aerosols, and trace metals from both natural and anthropogenic sources are airborne particles. Trace metals in PM_{10} such as Fe, Zn, Cu, Pb, Cr, Mn, Ni and Cd are extensively investigated by scientists due to their persistence, non-biodegradability and possible toxicity (Taiwo et al., 2014; Manousakas et al., 2021). Several of these elements are accumulated in biological systems and have been associated with oxidative stress, pulmonary inflammation, cardiovascular diseases, neurological disorders and carcinogenic effects in the long term of exposure (Kelly and Fussell, 2015; Lelieveld et al., 2020).

The elemental composition of particulate matter depends on local sources of emission, meteorological conditions, land use and atmospheric transport processes. Anthropogenic sources of trace metals in the built-up environment are mainly vehicular exhaust, brake and tire wear, industrial emissions, construction activities, biomass burning, coal combustion and re-suspension of road dust. Conversely, agricultural areas can be further enriched from soil erosion, agricultural machinery, biomass burning, fertilizer application and long-range atmospheric transport (Amato et al., 2014; Belis et al., 2013). Hence, the chemical composition of PM_{10} is very useful to identify the emission sources, understand the atmospheric processes, and to develop effective strategies for pollution control.

The elemental composition has been shown to vary significantly between regions, with several studies examining the chemical characteristics of particulate matter in Europe, North America, China, and India. High concentrations of Pb, Zn, Cu and Ni in highly industrialized cities are usually related to automotive emissions and industrial activity. Crustal elements such as Fe, Al, Ca and Mg are usually associated with soil resuspension and construction dust (Querol et al., 2007; Huang et al., 2014; Manousakas et al., 2021). Seasonal variability is an additional important feature of the chemistry of PM_{10}. Concentrations of particulate-bound metals tend to increase due to less atmospheric mixing, reduced boundary layer heights, greater fossil fuel combustion, and temperature inversion events. The spatial distribution of Cu concentrations is presented in. Concentrations are observed to be lower in the summer and monsoon periods, attributed to rain and better atmospheric dispersion (Taiwo et al., 2014; Zhang et al., 2022).

Pakistan is among the countries most affected by severe air pollution. The ambient air quality in the major metropolitan areas has been deteriorating due to rapid urbanization, increased traffic density, expanded industrial activities, and poor emission control measures (HEI, 2024). Numerous studies have been carried out to measure the mass concentrations of $PM_{2.5}$ and PM_{10} in cities like Lahore, Karachi, Islamabad, and Rawalpindi, but there is little information on the elemental composition of PM_{10} and the relative contribution of various emission sources. Most of the existing investigations have been focused mainly on particulate mass concentration rather than detailed chemical characterization. This limits the understanding of pollutant toxicity and source identification.

Rawalpindi represents one of Pakistan's fastest-developing city centers, characterized with the aid of using dense visitors' networks, industrial activities,

residential expansion, and business development. Simultaneously, the city's surrounding agricultural regions offer a possibility to examine anthropogenic and rural emission environments beneath Neath comparable climatic conditions. Evaluating the chemical composition of PM_{10} throughout those contrasting land-use classes can offer precious perception into the impact of various emission reassessments on ambient air quality. Such records are also important for designing effective mitigation plans and supporting evidence-based environmental management policies (Shahid et al., 2019; Shi et al., 2020; Health Effects Institute, 2024).

Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) has greatly enhanced the chemical analysis of particulate matter. The method allows a rapid, accurate, and simultaneous determination of several elements with high accuracy. ICP-OES is now one of the most widely used techniques to study atmospheric aerosols due to its sensitivity, reproducibility, and ability to measure concentrations of both major and trace elements over a large concentration range (USEPA, 2016). As a result, elemental analysis using ICP-OES has become a crucial method for environmental monitoring and source apportionment studies.

Despite the rising challenge of ambient air pollutants in Pakistan, there is a lack of statistics on the spatial and seasonal variation of PM_{10} elemental composition in built-up and agricultural environments. Only a few studies have simultaneously compared these contrasting land-use settings using the same sampling protocols and analytical techniques. This knowledge gap limits the development of efficient air quality management methods and limits knowledge of pollutant sources in rapidly urbanizing cities.

Therefore, the present study aimed to characterize the chemical composition of PM_{10} collected from built-up and agricultural environments of Rawalpindi using ICP-OES analysis. The specific objectives were to (i) determine the concentrations of selected trace elements associated with PM_{10}, (ii) evaluate seasonal and spatial variations in elemental composition, (iii) identify the dominant emission sources responsible for PM_{10} contamination, and (iv) provide baseline information for future air quality management and source apportionment studies in Pakistan.

2. Materials and Methods

2.1 Study Area

The research was carried out in Rawalpindi, Punjab, a swiftly growing metropolitan city in Punjab, Pakistan (Figure 1, Table 1). Rawalpindi is facing rapid urbanization, rising traffic levels, growing commercial activities, industrial advancements, and ongoing infrastructure development, all of which significantly contribute to air pollution (Shahid et al., 2019; Health Effects Institute, 2024). The city is situated on the Potohar Plateau between 33°31′-33°39′ N latitude and 72°53′-73°04′ E longitude at an average elevation of about 508 m above sea level (Pakistan Bureau of Statistics, 2023). In the Köppen climate classification, it is classified as humid subtropical (Cwa), with warm summers, monsoon precipitation, and cold winters (Peel et al., 2007). The dispersion of pollutants into the atmosphere, the deposition of particles, and the accumulation of pollutants are greatly impacted by the changes in weather conditions across different seasons (Seinfeld and Pandis, 2016; Zhang et al., 2022).

To evaluate spatial variability, the study area was classified into two contrasting land-use categories: Built-up: Satellite Town, Saddar, and DHA Phase IV, and Agricultural: Dhok Mianna, Dhoke Atta, and Dhok Patwaran.

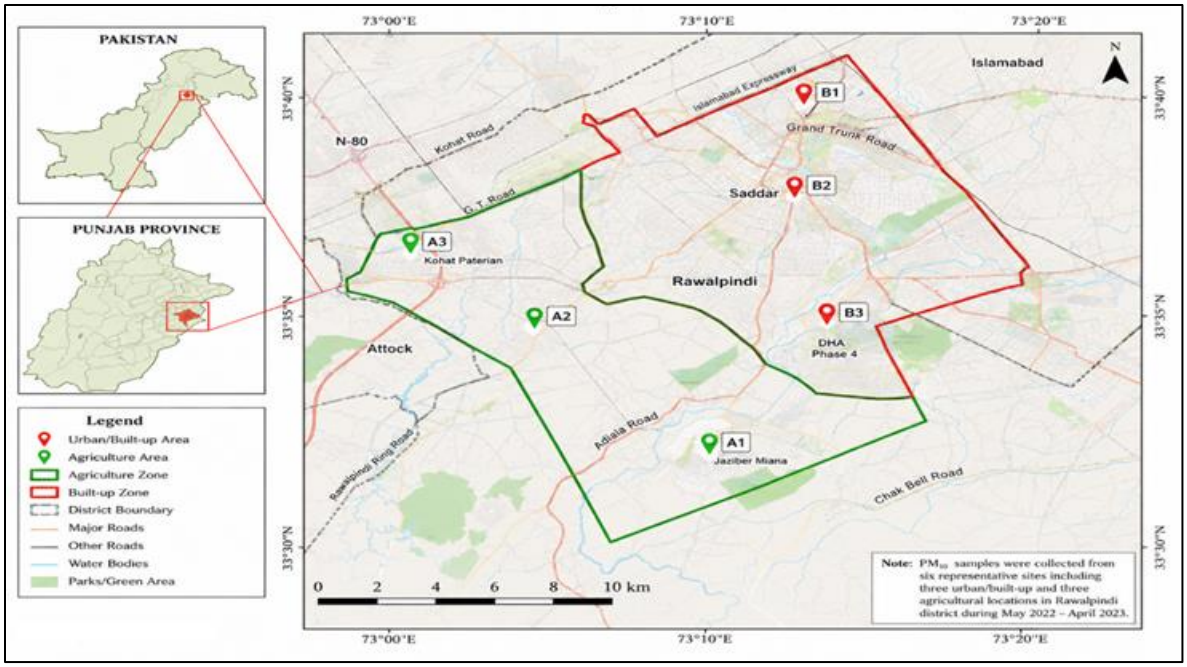

Fig. 1: Location of PM_{10} sampling sites in built-up and agricultural areas of Rawalpindi, Pakistan

Table 1: Geographic coordinates, land-use characteristics, and major nearby emission sources of the PM_{10} sampling sites in Rawalpindi, Pakistan.

Site Code	Sampling Site	Coordinates	Land-use Characteristics	Major Nearby Emission Sources
BU1	Satellite Town	33°38′31.55″N, 73°03′51.77″E	Built-up residential and commercial area	Heavy traffic, commercial activities, diesel generators, road dust resuspension, nearby landfill site
BU2	Saddar	33°35′13.56″N, 73°03′56.29″E	Commercial, educational, and industrial area	Vehicular traffic, commercial activities, industrial emissions, airport-related activities, biomass burning, diesel generators
BU3	DHA Phase IV	33°31′07.33″N, 73°04′41.46″E	Residential built-up area	Vehicular traffic, construction activities, brick kilns, road dust, suburban traffic
AGR1	Dhok Mianna	33°27′01.59″N, 73°01′31.86″E	Agricultural area	Agricultural machinery, soil resuspension, rural traffic, brick kilns
AGR2	Dhoke Atta	33°30′57.16″N, 72°56′22.66″E	Agricultural area	Heavy machinery, agricultural activities, rural traffic, brick kilns
AGR3	Dhok Patwaran	33°33′35.62″N, 72°53′29.69″E	Agricultural area	Agricultural machinery, rural traffic, soil dust, brick kilns

The monitoring locations were selected to represent different land-use scenarios and emission environments. The built-up sites were dominated by dense traffic, commercial activities, residential development and localized industrial emissions, while the agricultural sites were mainly affected by farming activities, soil resuspension, agricultural machinery, rural traffic and brick kiln emissions. Site selection was based on the features of land use, traffic intensity, nearby emission sources, industrial effect, and accessibility to ensure representative spatial coverage of the study area.

2.2 Sampling Design

Ambient PM_{10} sampling was performed from May 2022 to April 2023 to find out the spatial and seasonal fluctuation of PM_{10}-bound heavy metals in Rawalpindi. Six sampling locations were obtained with three monitoring stations located within each land-use group. Standard monitoring protocols were applied to sample three times monthly at each monitoring site. A total of 216 samples of PM_{10} were collected for the study period; 108 samples were taken from the built-up areas and 108 samples from the agricultural sites. The sampling locations were kept constant over the monitoring period to allow for consistent spatial and seasonal comparisons. The geographic coordinates for each monitoring station were obtained using a hand-held Global Positioning System (GPS).

In an effort to compare the seasonal samples, they were divided into summer (April-July) and winter (November-January) on the basis of prevalent meteorological conditions of the study area. The one-year monitoring period (May 2022-April 2023) was chosen to represent the full range of seasonal climatic circumstances affecting the atmospheric PM_{10} concentrations in Rawalpindi. Seasonal variability in response to variations in temperature, precipitation, atmospheric stability and boundary-layer dynamics was assessed by sampling across a full annual cycle, thus eliminating bias associated with short-term monitoring programs.

2.3 PM_{10} Sampling

Ambient PM_{10} samples were collected using an Ecotech HiVol 3000 High-Volume Air Sampler (HVAS). The sampler was equipped with rectangular borosilicate glass fibre filters (250 × 200 mm; approximately 10 × 8 inches) for the collection of particulate matter. The HVAS was set up at a height of approximately 1.8 m above ground level, the average human breathing zone. Samples were collected three times a month from May 2022 to April 2023 at each monitoring site, and each sampling was carried out for 24 hrs.

The sampler was operated at a flow rate of 1.1 $m^3\ min^{-1}$ (66.67 $m^3\ h^{-1}$) using a 110 VAC power supply. During operation, ambient air was continuously drawn through the glass fibre filter, allowing efficient collection of PM_{10} particles for subsequent gravimetric and chemical analyses. The high sampling capacity of the Ecotech HiVol 3000 ensured efficient particle collection under both built-up and agricultural environmental conditions. Before each sampling event, the sampler was inspected to ensure proper operation, and the airflow rate was verified using the instrument's calibrated flow measurement system in accordance with the manufacturer's operating procedures.

Clean borosilicate glass fiber filters were weighed on a calibrated analytical balance before sampling, and the initial filter weight (Wi) was recorded. After each 24 hr sampling event, the exposed filters were removed from the Ecotech HiVol 3000 sampler using clean stainless-steel forceps, placed in sterile polyethylene bags, labeled appropriately, and transported to the Environmental Services Pakistan (ESPAK) laboratory for analysis. The filters were then reweighed to obtain the net particulate matter collected before acid digestion and ICP OES analysis. Precautions were taken to minimize contamination and preserve sample integrity during filter handling and transportation, storage, and laboratory processing.

2.4 Sample Digestion

Before elemental analysis, the whole borosilicate glass fiber filter for each sampling event was cut into small pieces with a clean stainless-steel paper cutter and transferred to a clean digestion vessel. The vessel was filled with approximately 15 mL of freshly prepared aqua regia (HCl: HNO_3; 3:1, v/v) to digest the particulate matter collected on the filter, and the mixture was heated on a hot plate at 120°C for 30 min. Aqua regia digestion was selected because it can effectively extract trace metals from particulate matter retained on borosilicate glass fiber filters and is commonly used in atmospheric aerosol studies before ICP-OES analysis (Alloway, 2013).

After digestion, the solution was cooled at room temperature for about 20 min. Then 10 mL of deionized water was added. The digest was quantitatively transferred to a 25 mL volumetric flask and diluted to volume with deionized water. The final solution was transferred to acid-cleaned polyethylene tubes and stored until elemental analysis by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). The concentrations of Pb, Cr, Cd, and Cu obtained from ICP-OES analysis were converted to atmospheric concentrations (μg m^{-3}) by using the final digest volume and total volume of air sampled during each 24-hour sampling period.

2.5 Elemental Analysis

Elemental concentrations were determined using a Shimadzu ICPE-9000 Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES). Before sample analysis, the instrument was calibrated with the multi-element standard calibration solutions based on the prescribed operating protocols from the manufacturer to ensure analytical accuracy and instrument stability. The ICP-OES technique was chosen for its capacity to simultaneously determine a large number of elements with good sensitivity, precision, and reproducibility, making it suited for the study of trace metals in the air particulate matter.

2.6 Quality Assurance and Quality Control (QA/QC)

Quality assurance and quality control procedures were implemented throughout sample collection, handling, digestion, and elemental analysis to minimize contamination and ensure analytical reliability. All glassware and digestion vessels were cleaned properly prior to use, and analytical-grade reagents along with deionized water were utilized for sample preparation. Instrument calibration was done using standard calibration solutions as per the operating protocols provided by the manufacturer before analysis. All aspects of handling, shipping, storage, and laboratory processing of filters were managed carefully to minimize contamination and maintain sample integrity.

2.7 Statistical Analysis

Descriptive statistics were calculated for all elements analyzed, including mean, standard deviation, minimum and maximum concentrations. The differences between the Built-up and agricultural monitoring stations and the different seasons (summer and winter) were compared through one-way analysis of variance (ANOVA) followed by Tukey's Honest Significant Difference (HSD) test to determine statistically significant differences at a 95% confidence level ($p < 0.05$) (Coelho et al., 2023; Hopke et al., 2023; García-Marlès et al., 2024). Statistical analysis was performed with standard statistical software.

3. Results and Discussion

3.1 Seasonal Variation of PM_{10}-Bound Heavy Metals

Monthly concentrations of PM_{10} - bound lead (Pb), chromium (Cr), cadmium (Cd), and copper (Cu) were measured at three built-up (BU1-BU3) and three agricultural (AGR1-AGR3) sites in Rawalpindi, Pakistan, from May 2022 to April 2023. The time variation of each heavy metal is presented in Figures 2-5. All the metals studied in general show a marked seasonal variation, with concentrations tending to increase during the colder months (November–January) and to

decrease during the warmer months (April–July). Both built-up and agricultural sites recorded a consistent seasonal trend; however, the magnitude of variation differed between sampling locations. The higher winter concentrations are mainly due to lower mixing heights in the atmosphere, lower wind speeds, recurrent temperature inversions, and reduced dispersion of pollutants, favoring the accumulation of heavy metals bound to particulates near the surface. In contrast, lower concentrations during summer are associated with enhanced atmospheric mixing, stronger wind circulation, and occasional precipitation, which promote the dilution and removal of airborne particulate matter. Similar seasonal behavior of atmospheric trace metals has been reported in previous studies from South Asia and other urban environments (Wang et al., 2021; Zhang et al., 2022; Manousakas et al., 2021).

3.1.1 Lead (Pb)

Lead (Pb) was the most common heavy metal associated with PM_{10} at the built-up and agricultural sampling sites during the monitoring period (Figures 2a and 2b). A clear seasonal pattern was observed with Pb concentrations increasing in winter months (November-January) and decreasing in warm months (April-July). This seasonal variation was consistent across all six monitoring sites, suggesting the strong influence of seasonal meteorological conditions on the accumulation of particulate-bound Pb.

Pb concentrations in the built-up locations ranged from 0.30 to 0.89 µg m^{-3}. The greatest concentration of 0.89 µg m^{-3} was recorded at BU2 in December, whereas the lowest concentration of 0.30 µg m^{-3} was recorded at BU1 in April. Similarly, the agricultural sites showed Pb concentrations from 0.41 to 0.91 µg m^{-3}, with the maximum value (0.91 µg m^{-3}) at AGR1 and AGR3 in December and the lowest value (0.41 µg m^{-3}) at AGR2 in July. Overall, agricultural sites exhibited slightly higher average Pb concentrations than built-up sites, consistent with the mean values presented in Figure 6, suggesting that, besides traffic-related emissions, agricultural activities and local biomass burning may also contribute to atmospheric Pb levels.

The higher Pb concentrations recorded during winter are consistent with the seasonal meteorological conditions described above, which favor the accumulation of particulate-bound pollutants. Lead in ambient PM_{10} is likely associated with vehicular emissions, brake and tire wear, road dust resuspension, industrial activities, biomass burning, and the resuspension of historically contaminated roadside soils. Leaded gasoline is no longer used in Pakistan; however, legacy Pb may still contribute to airborne particulate matter through mechanical resuspension of roadside dust. Similar seasonal patterns have been found in various metropolitan areas in South Asia (Wang et al., 2021; Choudhary et al., 2023).

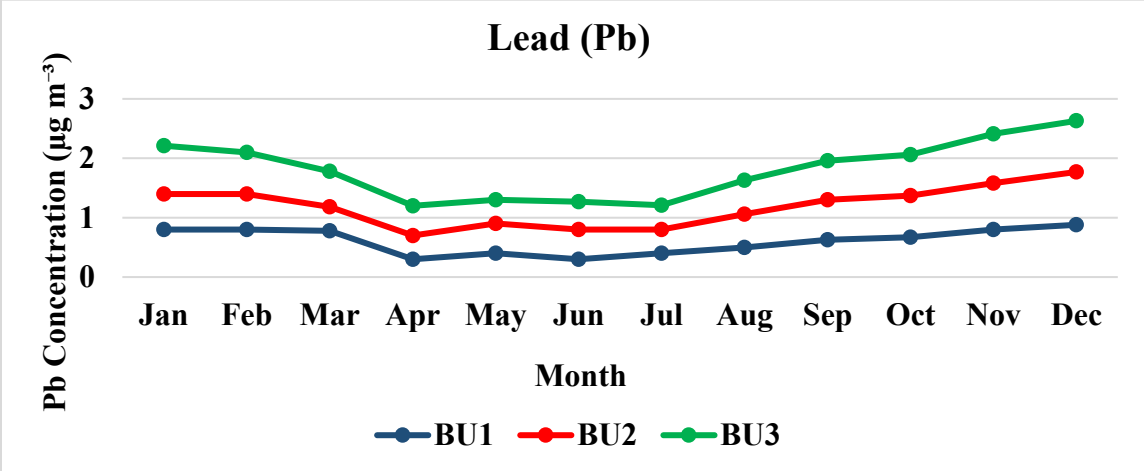

Fig. 2a: Monthly concentration of Pb in the built-up area of Rawalpindi.

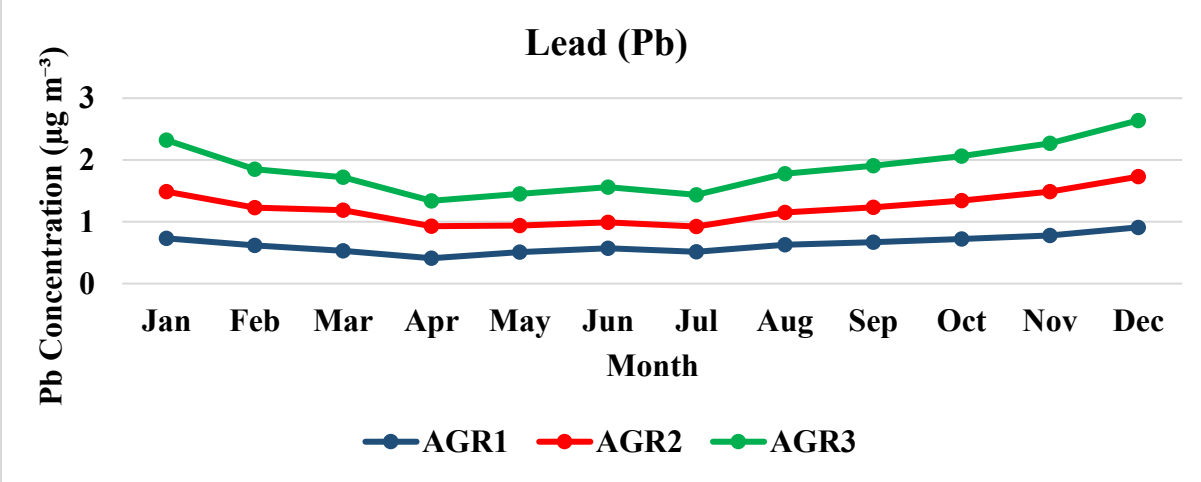

Fig. 2b: Monthly concentration of Pb in the agricultural area of Rawalpindi.

3.1.2 Chromium (Cr)

Chromium (Cr) concentrations in PM_{10} exhibited clear seasonal variations at both the built-up and agricultural sampling sites (Figures 3a and 3b). At the built-up sites, Cr concentrations ranged from 0.008 to 0.040 µg m^{-3}. Concentrations were relatively higher during January and February, gradually decreased from April to July, remained comparatively low during the summer months, and increased again from October, reaching another peak during November. Although slight differences were observed among BU1, BU2, and BU3, all three sites displayed a similar temporal pattern throughout the monitoring period, indicating a strong seasonal influence on atmospheric Cr concentrations.

A similar seasonal pattern was seen in the agricultural areas (Fig. 3b), where Cr concentrations ranged from 0.008 to 0.040 µg m^{-3}. Values were generally higher during the winter months. The lowest amounts were during the summertime. Monthly profiles of AGR1, AGR2 and AGR3 revealed similar trends, indicating that seasonal weather circumstances had a bigger impact on Cr concentrations than local geographical variances.

The seasonal fluctuation of Cr concentrations was comparable to that seen for Pb, with higher values during the winter and lower amounts during the summer. Chromium in atmospheric PM_{10} is most typically associated with vehicle emissions, brake and tire wear, fossil fuel burning, industrial operations, construction dust, and re-suspension of road dust. Contributions from biomass burning, farm machinery, and suspended soil particles may also add to agricultural areas, but were inferred from land-use characteristics and published literature rather than proven by source-apportionment analysis. Such seasonal behavior has been described in prior investigations (Choudhary et al., 2023).

3.1.3 Cadmium (Cd)

Cadmium (Cd) was the lowest concentration among the studied heavy metals, but showed a strong seasonal pattern at the built-up and agricultural sites (Figures 4a and 4b). The values of Cd in the built-up locations ranged from 0.002 to 0.019 µg m^{-3}. Higher concentrations were routinely reported during the winter season (November-January), with the maximum concentration (0.019 µg m^{-3}) at BU1 in January. Compared to the winter period, the summer period (April-July) showed lower concentrations, with minimal values (0.001-0.002 µg m^{-3}) between June and July. Similar seasonal trends were observed at BU2 and BU3, with Cd concentrations increasing in winter and decreasing in summer.

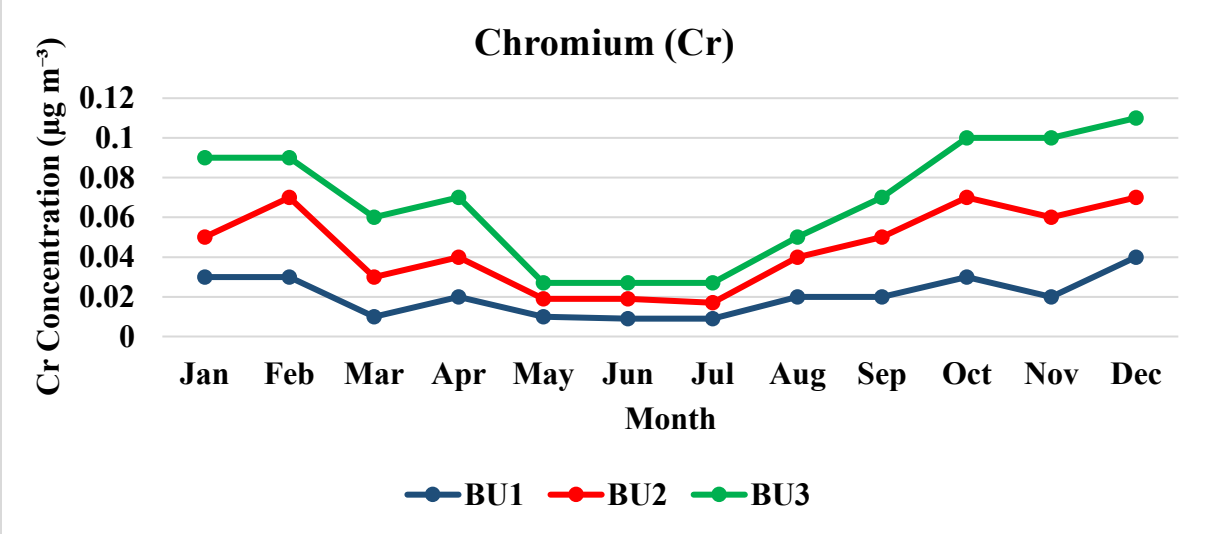

Fig. 3a: Monthly concentration of Cr in the built-up area of Rawalpindi.

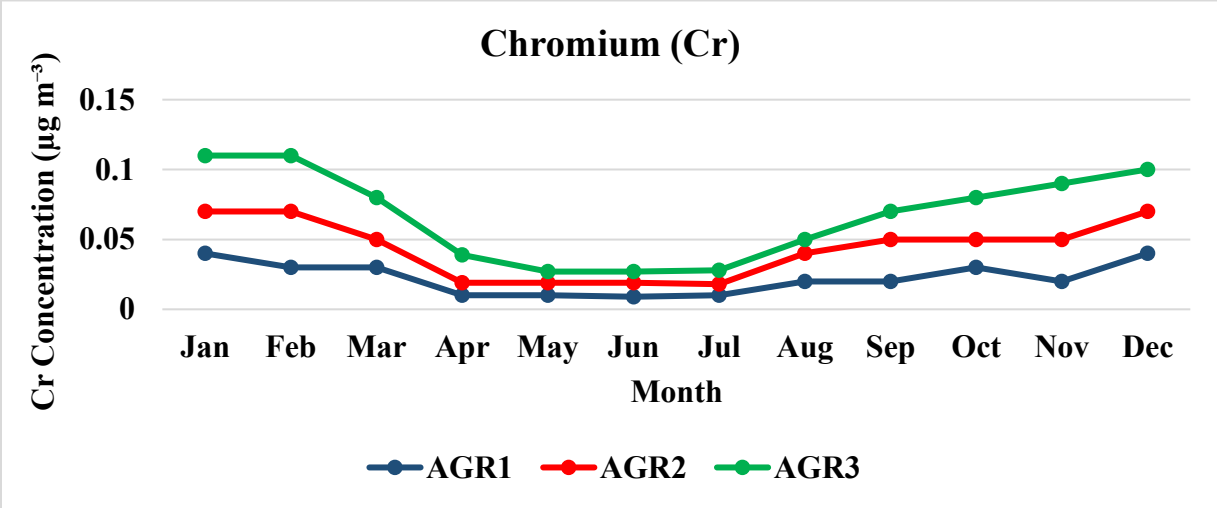

Fig. 3b: Monthly concentration of Cr in the agricultural area of Rawalpindi.

Cd values in agricultural locations were from 0.003 to 0.060 µg m^{-3}. The greatest quantity (0.060 µg m^{-3}) was detected at AGR1 in January, while much lower amounts were recorded during the summer season (April-July). In general, Cd concentrations in agricultural locations were slightly higher than in built-up sites for the whole monitoring period, especially in winter (Figure 4b).

Cadmium also showed a definite seasonal pattern, with higher concentrations in winter and lower in summer, in accordance with the general seasonal tendency reported for all the metals studied. Atmospheric PM_{10} may contain cadmium from fossil fuel combustion, industrial emissions, waste incineration, and biomass burning and agricultural operations such as the application of phosphate fertilizers. However, these possible sources were determined based on land-use features and other studies, rather than directly identified using source-apportionment analysis (Matei et al., 2025).

3.1.4 Copper (Cu)

The concentrations of Cu in PM_{10} at the built-up and agricultural sampling sites showed distinct seasonal variations (Figures 5a and 5b). Cu concentrations were from 0.03 to 0.17 µg m^{3} for the built-up sites. During the winter season (November-January), readings showed a continuous

increase, with the highest concentration (0.17 µg m^{-3}) reported during December at BU1 and BU3. In contrast, Cu concentrations were found to be low throughout the summer period (April-July). The minimal concentration (0.03 µg m^{-3}) was detected in July. The time patterns of the three developing sites were similar, reflecting the effect of seasonal meteorological variables on atmospheric Cu concentrations.

Seasonal changes in Cu concentrations were also seen at the agricultural areas, ranging from 0.04 to 0.17 µg m^{-3}. The highest concentrations were found throughout the winter season, especially in December, and the lower amounts were seen during summer months (April-July). The monthly concentration profiles at AGR1, AGR2 and AGR3 presented similar trends to the built-up sites, with Cu contents decreasing in summer and increasing again from late autumn to winter.

Copper concentrations followed the same seasonal trend as the other metals studied, with high levels in winter and low levels in summer. Copper in ambient PM_{10} is often linked to non-exhaust vehicle emissions (e.g. brake lining wear, tire abrasion), but also industrial operations, resuspension of road dust, biomass burning and the application of copper-containing agrochemicals. These probable emission sources were hypothesized based on land-use characteristics and available literature instead of being proven by source-apportionment analysis (Coelho et al., 2023).

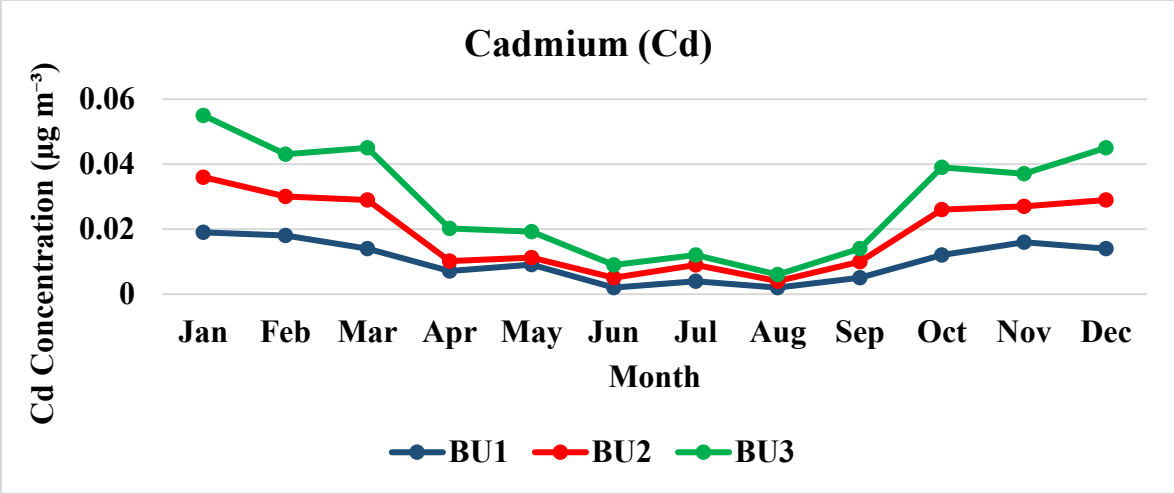

Fig. 4a: Monthly concentration of Cd in the built-up area of Rawalpindi.

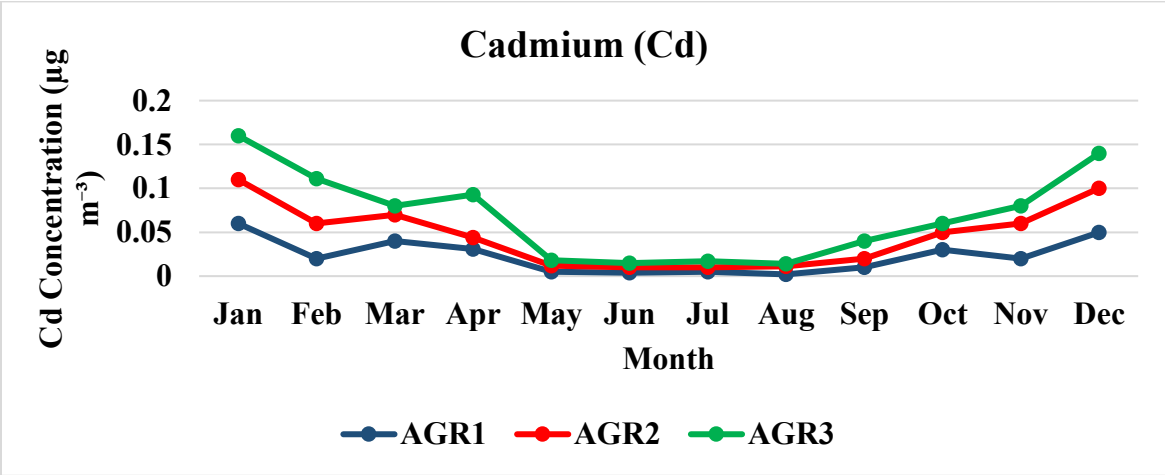

Fig. 4b: Monthly concentration of Cd in the agricultural area of Rawalpindi.

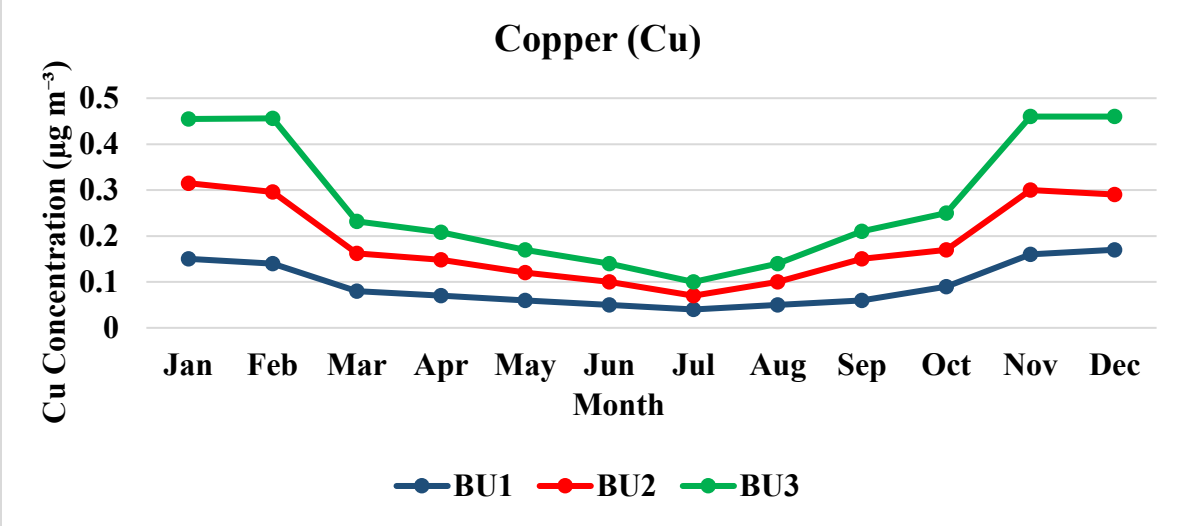

Fig. 5a: Monthly concentration of Cu in the built-up area of Rawalpindi.

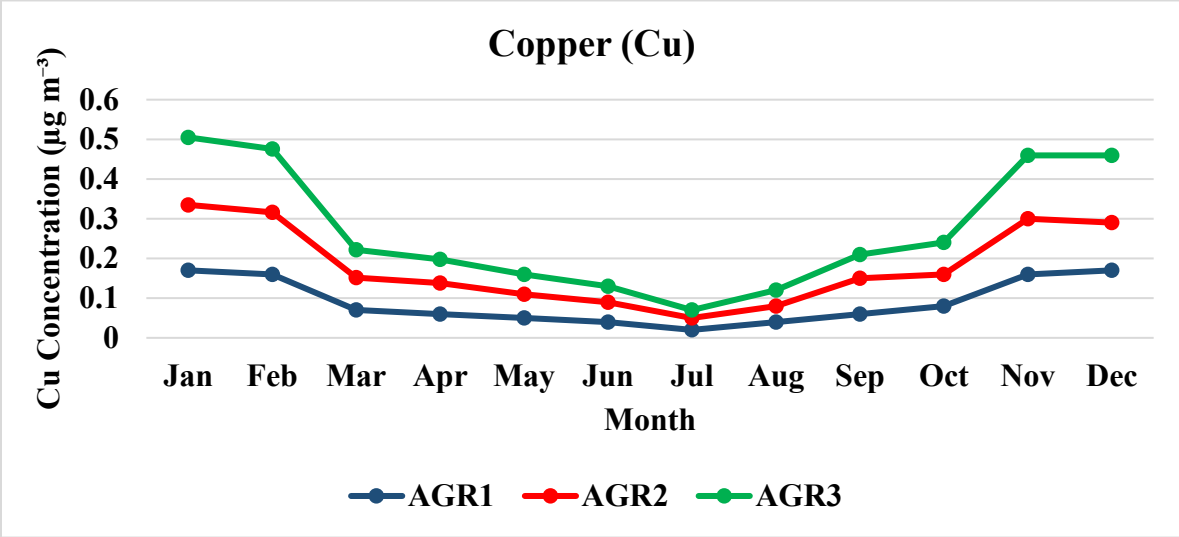

Fig. 5b: Monthly concentration of Cu in the agricultural area of Rawalpindi.

3.2 Comparison between Built-up and Agricultural Sites

Figure 6 presents the mean (±SD) concentrations of Pb, Cr, Cd, and Cu in PM_{10} at the built-up and agricultural sampling sites during the study period. Overall, Pb was the dominant heavy metal at both land-use categories, followed by Cu, Cr, and Cd, indicating the abundance pattern of Pb > Cu > Cr > Cd. The mean concentrations at the built-up sites were 0.61 ± 0.19 µg m^{-3} for Pb, 0.08 ± 0.04 µg m^{-3} for Cu, 0.02 ± 0.02 µg m^{-3} for Cr, and 0.008 ± 0.006 µg m^{-3} for Cd. At the agricultural sites, the corresponding mean concentrations were 0.64 ± 0.16 µg m^{-3}, 0.09 ± 0.04 µg m^{-3}, 0.02 ± 0.02 µg m^{-3}, and 0.012 ± 0.015 µg m^{-3}, respectively.

Mean concentrations of Pb, Cu, and Cd were slightly higher at the agricultural sites, while Cr concentrations were comparable in both land-use categories. The relatively small differences observed in the two land-use settings suggest that both local emission sources and regional atmospheric transport played a role in shaping the ambient heavy metal concentrations. The similarities in concentrations between the two land-use categories further suggest that regional atmospheric transport and seasonal meteorological conditions may have a greater influence on ambient heavy metal

concentrations than the land-use features themselves. Vehicular traffic, commercial activities and built-up areas are the main sources of road dust resuspension, while agricultural areas can also be affected by biomass burning, agricultural machinery, fertilizer application and windblown soil particles. These potential emission sources were inferred from land-use features and published literature and were not confirmed by receptor modeling or source apportionment analyses (Coelho et al., 2023).

3.3 Comparison with Regulatory Guidelines

The annual mean values were compared with available ambient air quality guideline values to assess the environmental significance of the measured heavy metal concentrations. The annual mean concentrations of Pb measured at the built-up (0.61 µg m^{-3}) and agricultural (0.64 µg m^{-3}) sites were higher than the European Union annual target value for ambient air of 0.5 µg m^{-3} (European Commission, 2004). Similarly, the measured annual mean concentrations of Cd (0.008 and 0.012 µg m^{-3}) were higher than the annual target value of 0.005 µg m^{-3} (5 ng m^{-3}) set by the European Union (European Commission, 2004). The guideline values available are only for hexavalent chromium (Cr VI), while the study measured total chromium; hence, chromium concentrations were not directly compared. Similarly, there is currently no internationally agreed ambient air quality guideline for copper. The results indicate the necessity of continuous monitoring and effective strategies for controlling emissions to reduce human exposure to airborne heavy metals.

Given the absence of any ambient air quality standards for individual heavy metals associated with PM_{10} in Pakistan, the European Union target values were used solely as an internationally recognized benchmark for comparison.

3.4 Seasonal Comparison (Winter vs Summer)

Figure 7 shows the seasonal average concentrations of all metals for the six sites combined. Winter (November-January) mean concentrations were significantly higher than summer (April-July) values for all metals: Pb: 0.83 vs 0.45 µg m^{-3}, Cr: 0.03 vs 0.01 µg m^{-3}, Cd: 0.017 vs 0.005 µg m^{-3}, Cu: 0.12 vs 0.05 µg m^{-3}.These results confirm that winter meteorology strongly governs the accumulation of particulate-bound heavy metals in Rawalpindi.

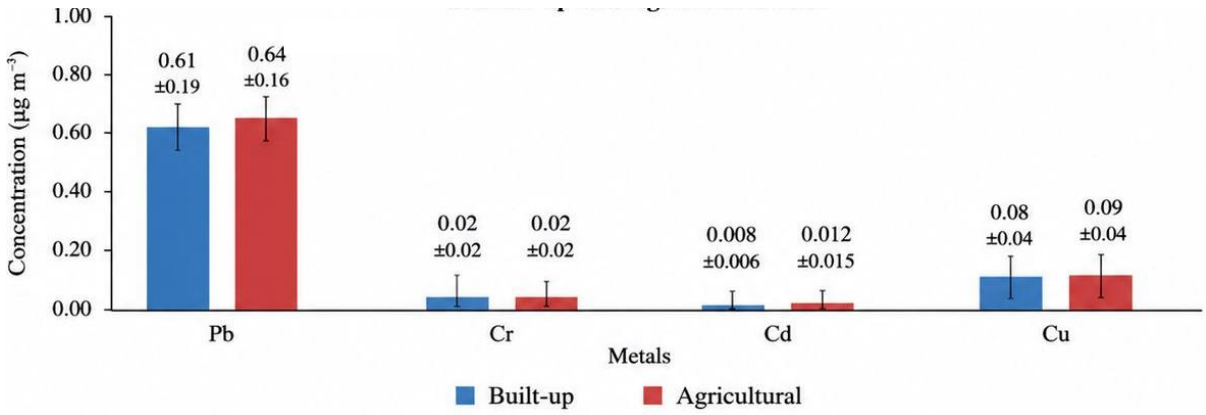

Fig. 6: Mean (±SD) concentrations at Built-up and Agricultural Sites of the study area.

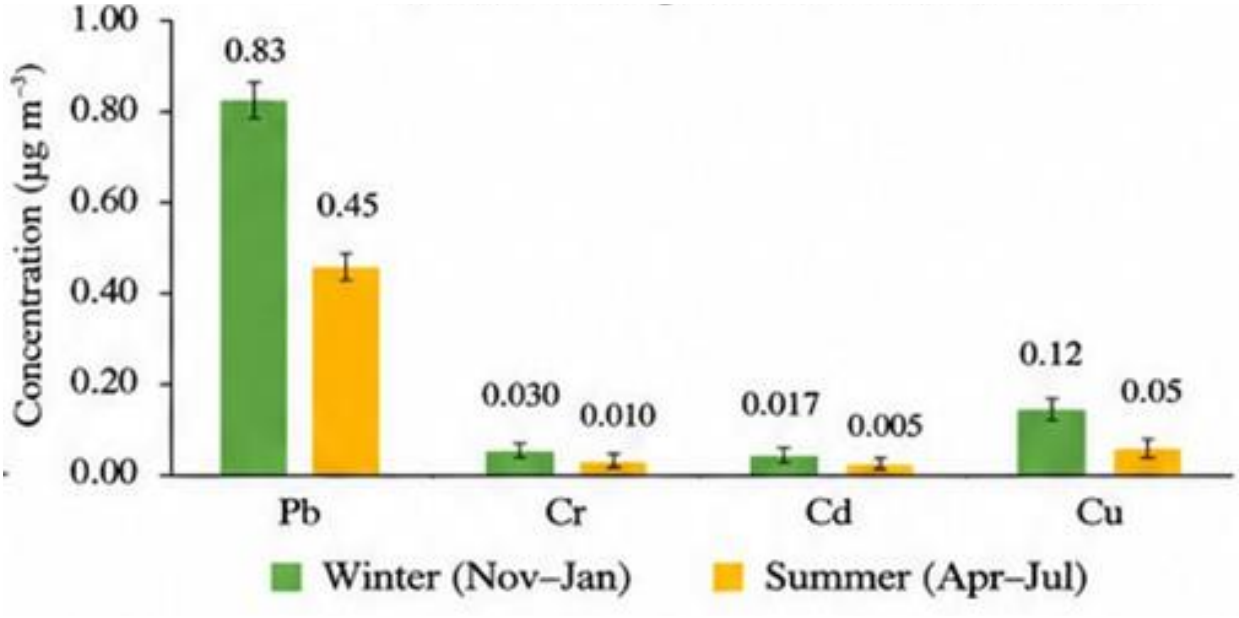

Fig. 7: Seasonal average concentration (All Sites) of the study area.

Higher concentrations of all the investigated heavy metals during the winter season consistently indicated that the seasonal meteorological conditions had more impact on the accumulation of atmospheric heavy metals than local emission variations alone. During winter, lower boundary layer height, lesser atmospheric mixing, and frequent temperature inversion events favor accumulation of particulate-bound pollutants, while during summer, enhanced atmospheric mixing and wet deposition facilitate dispersion and removal of particulate-bound pollutants. This seasonal behavior has also been reported in previous studies in South Asia and other urban environments, confirming the important role of meteorological conditions in controlling atmospheric heavy metal concentrations.

3.5 Spatial Distribution of Heavy Metals in PM_{10}

The spatial distribution of annual average concentrations of Pb, Cr, Cd, and Cu associated with PM_{10} across the six monitoring sites is presented in Figure 8-11. Spatial distribution maps show a high degree of heterogeneity in heavy metal concentrations between built-up (B1-B3) and agricultural (A1-A3) locations, which reflects the combined effect of land use, emission sources, and local meteorological conditions. Generally elevated concentrations were observed at the built-up monitoring sites, but several agricultural locations also showed relatively high concentrations, suggesting the contribution of regional atmospheric transport and agricultural activities.

3.5.1 Lead (Pb)

Among the investigated metals, Pb exhibited the highest spatial variability and the greatest concentrations throughout the study area (Figure 8). The highest concentrations were observed at A3 (0.91 µg m^{-3}) and B2 (0.89 µg m^{-3}), whereas comparatively lower concentrations occurred at B3 (0.63 µg m^{-3}). The high Pb concentrations observed in the built-up area are mainly attributed to heavy vehicular traffic, commercial activities, industrial emissions, and re-suspension of contaminated roadside dust, brake and tire wear. The high concentrations observed at agricultural sites also suggest that Pb is not only locally sourced but also transported from adjacent built-up areas by prevailing winds and atmospheric dispersion. Biomass burning and agricultural residue burning may also contribute to Pb enrichment in rural environments. Comparable spatial trends have been reported in Lahore (Shahid et al., 2019), Delhi (Yadav et al., 2020) and other cities of South Asia where traffic emissions

and road dust are known as major contributors of particulate-bound lead.

3.5.2 Chromium (Cr)

The spatial distribution of Cr shows a lower concentration compared to Pb, with values between 0.030 and 0.040 µg m^{-3} in the monitoring network (Figure 9). The highest concentration was at A3 (0.040 µg m^{-3}), and the built-up sites B1 and B2 showed 0.030 µg m^{-3} and 0.036 µg m^{-3} respectively. The relatively uniform spatial distribution of Cr indicates contributions from multiple diffuse sources rather than a single dominant emission source. Chromium in atmospheric particulate matter is generally associated with fuel combustion, industrial processes, metal processing industries, and vehicular emissions. Similarly, comparable concentrations between built-up and agricultural sites also indicate the influence of regional atmospheric transport. The wide spread of particulate-bound chromium in built-up atmospheres influenced by mixed industrial and traffic emissions has been reported in previous studies (Querol et al., 2007; Wang et al., 2021).

3.5.3 Cadmium (Cd)

Cadmium concentrations were relatively lower than Pb and Cu but with clear spatial variations. The highest concentrations were observed at agricultural sites A1 and A3 (0.017 µg m^{-3}) and the lowest at A2 (0.005 µg m^{-3}). Intermediate concentrations of 0.009-0.012 µg m^{3} were found in built-up locations. The higher concentrations of Cd in agricultural sites suggest that conventional farming methods are a major contributor to atmospheric Cd. Possible sources include application of phosphate fertilizer, biomass burning, pesticide application, agricultural machinery and re-suspended soil particles. Cadmium is present at relatively low concentrations but is of environmental concern because of its persistence, toxicity and potential for bioaccumulation. Similar results have been reported for agricultural areas where the use of fertilizers is an important source of atmospheric Cd (Alloway, 2013; Zhang et al., 2015).

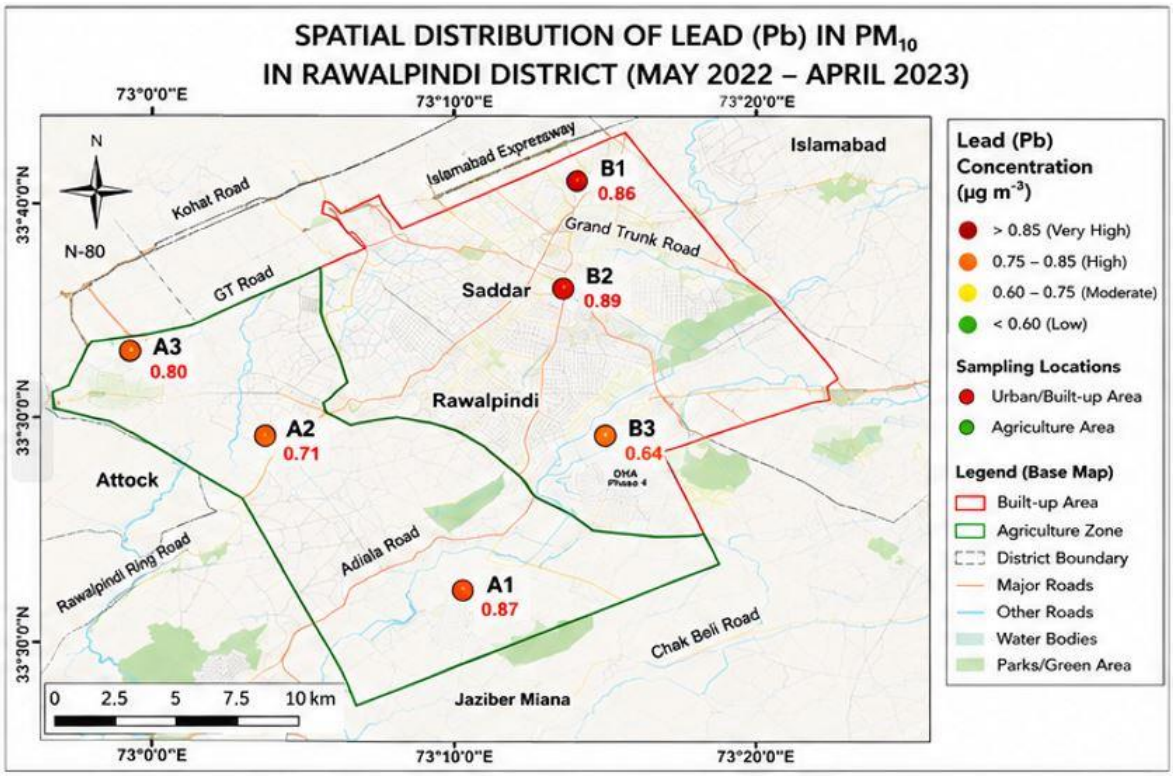

Fig. 8: Spatial Distribution of Lead (Pb) in PM_{10} of Rawalpindi District.

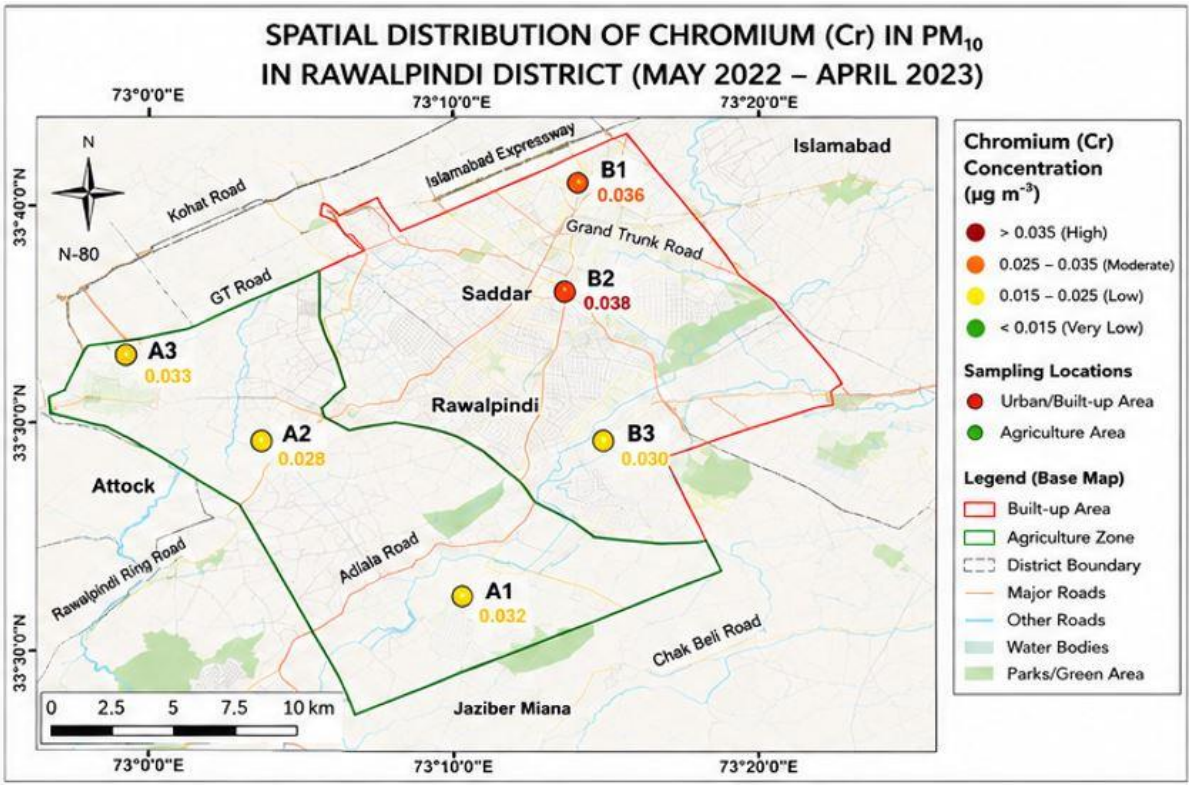

Fig. 9: Spatial Distribution of Chromium (Cr) in PM_{10} of Rawalpindi District.

3.5.4 Copper (Cu)

The spatial distribution of PM_{10}-bound Cu concentrations at the built-up and agricultural sampling sites is presented in Figure 11. Copper concentrations ranged from 0.12 to 0.16 µg m^{-3}, with the highest value recorded at B2 (0.16 µg m^{-3}), while relatively lower concentrations were observed at A2 and B3 (0.12 µg m^{-3}). Cu showed a more homogeneous spatial pattern of the monitoring sites compared to Pb. High Cu levels in urban areas are mainly explained by non-exhaust emissions from vehicles, that is, brake lining wear, tire abrasion and road dust resuspension. Moderate Cu concentrations were also observed in agricultural sites, indicating additional contributions from agricultural machinery, soil particles and long-range transport of built-up emissions. Similar spatial distributions have been reported in urban environments where traffic-related non-exhaust emissions are the principal source of atmospheric copper (Pant and Harrison, 2013; Amato et al., 2014).

Overall, the spatial and seasonal patterns observed in this study are consistent with previous investigations conducted in Pakistan and other South Asian countries, where winter meteorological conditions and mixed anthropogenic emission sources have been identified as the dominant factors controlling PM_{10}-bound heavy metal concentrations.

4. Conclusion

This study indicates clear spatial and seasonal variations of PM_{10}-bound Pb, Cr, Cd, and Cu in built-up and agricultural areas of Rawalpindi, Pakistan. The results show higher concentrations of all the metals investigated during winter compared with summer, indicating the dominant role of seasonal meteorological conditions on the accumulation of particulate-bound heavy metals. Pb was the main metal among the elements studied, followed by Cu, Cr and Cd. The average concentrations of Pb, Cu and Cd were a little bit higher at the agricultural sites compared to the built-up sites, whereas Cr showed similar concentrations for the two land-use categories.

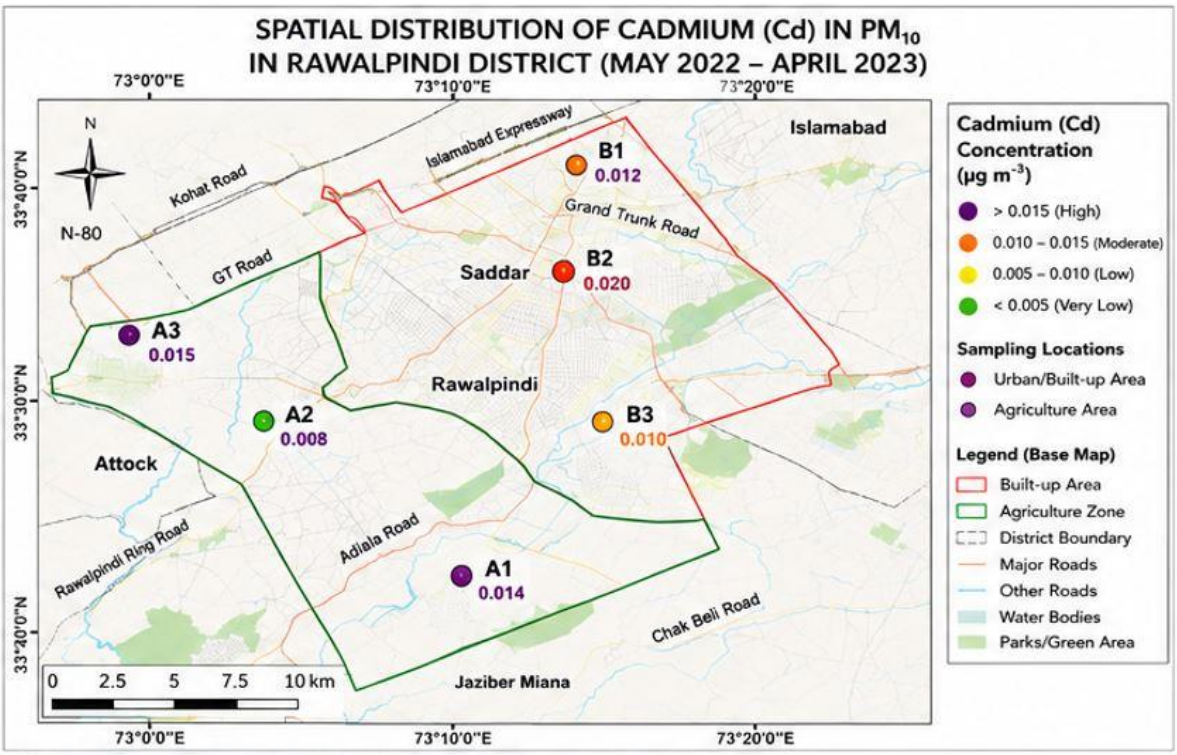

Fig. 10: Spatial Distribution of Cadmium (Cd) in PM_{10} of Rawalpindi District.

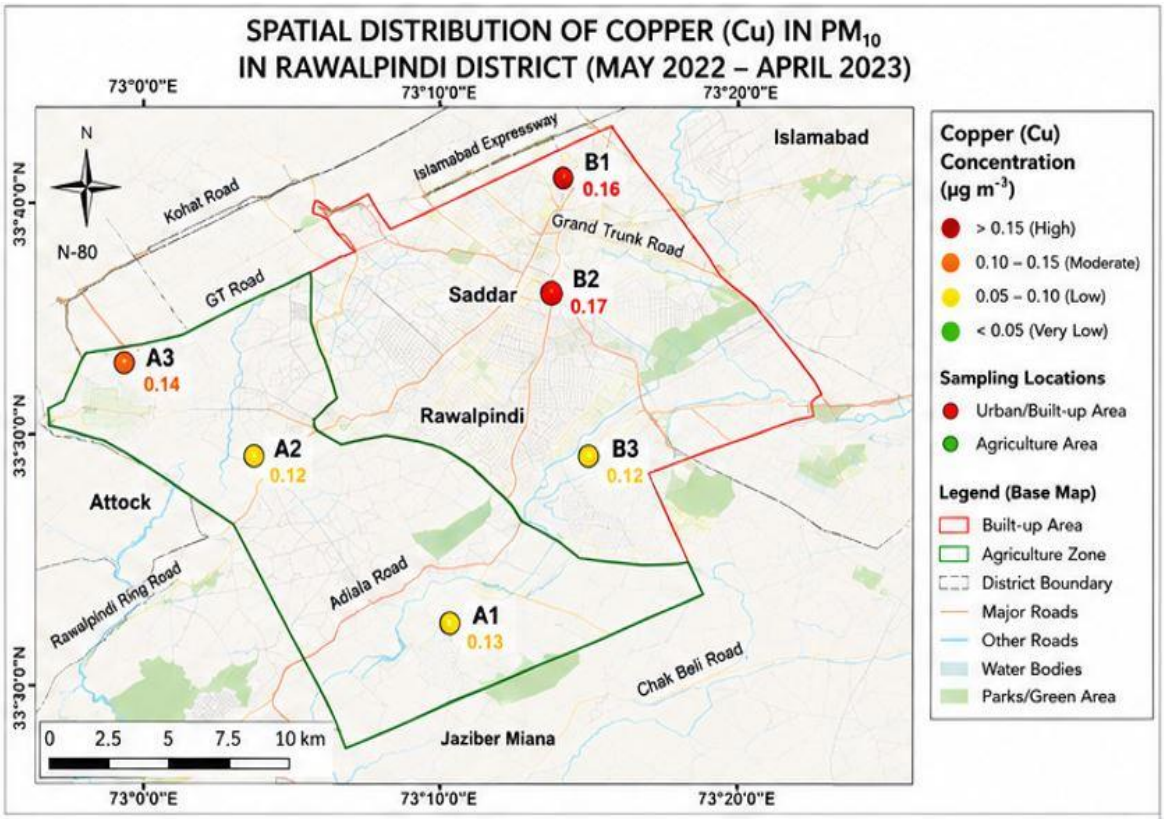

Fig. 11: Spatial Distribution of Copper (Cu) in PM_{10} of Rawalpindi District.

In summary, the relatively small spatial differences imply that the pollution of heavy metals in the atmosphere of the study area is affected by both local activities and regional atmospheric transport. Potential emission sources were inferred from land-use characteristics and published literature; however, the results provide useful baseline information for understanding the spatial and seasonal behavior of PM_{10}-bound heavy metals in Rawalpindi. Future studies should include receptor-based source apportionment techniques (e.g., Positive Matrix Factorization or Principal Component Analysis), a wider variety of trace elements, and human health risk assessments to guide better air quality management and pollution control strategies.

Authors' Contributions

Umair Rashad proposed the main concept of the study, contributed to the writing of the manuscript, and was involved in field data collection. Aansa Rukya Saleem contributed to the review of relevant literature, provided supervision and guidance throughout the study, and critically reviewed the manuscript. Tahir Hayat Malik contributed to the technical review of the manuscript and provided critical comments and revisions before submission. All authors reviewed and approved the final version of the manuscript.

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Data Availability Statement

The original data generated and analyzed during this study are owned by the authors and are available from the corresponding author upon reasonable request. The data have not been published elsewhere and are not subject to any copyright restrictions or third-party ownership. The authors confirm that the data used in this study are original and were collected and generated as part of the present research.

Author's Declaration

We, the authors, declare that the manuscript entitled "Spatial and Seasonal Distribution of PM_{10}-Bound Heavy Metals in Built-up and Agricultural Areas of Rawalpindi, Pakistan" presents original research and has not been published or submitted elsewhere. All authors contributed equally to the research, analysis, and preparation of the manuscript and have approved the final version. The authors declare that there is no conflict of interest, financial or otherwise, that could have influenced the research or its publication. All authors jointly take responsibility for the integrity and content of the manuscript.

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