SHORT COMMUNICATION

Trace Elements and Water-Soluble Ionic Species Associated with Ambient Particulate Matter over Lucknow City, India

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Abstract

Clean air is the foremost essentiality to ensure a good life on our planet. Nonetheless, escalating air pollution levels in urban spaces have caused an increase in morbidity and mortality. The air pollution predicament requires a comprehensive understanding of the local scale. In this perspective, the present study was carried out to assess the load of PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and associated chemical profiles, including trace elements and ionic species of the commercial, industrial and rural areas over Lucknow city during winter months of 2022–23. The average 24-hour $PM_{2.5}$ concentrations (Avg ± SD) in the commercial, industrial, and rural areas were recorded to be 189.1 ± 57.6, 177.7 ± 36.5, and 141.6 ± 23.0 μg/m^3, respectively, with a percentage exceedance over national ambient air quality standards (NAAQS) of 224 to 215, 196, and 136%. Similarly, the concentrations of PM_{10} were 337.9 ± 109.2, 285.6 ± 53.1, and 234.9 ± 43.0 μg/m^3, with 238, 186, and 135% exceedance, respectively. SO_2 level ranged from 8.7 to 23.9 μg/m^3, whereas NO_2 level ranged from 16.8 to 45.7 μg/m^3 for the study period. Based on the air quality index (AQI) values, commercial (353), industrial (344), and rural (317) areas fell in the "very poor" category of AQI. 12 studied elemental species in descending order were Al>Fe>Mg>Zn>Pb>Mn>Cr>Cu>Ni>Co>Cd>Mo while the sequence of 8-ionic species was SO_4^{2-}>Cl^->K^+>NH_4^+>NO_3^->Ca^{2+}>Na^+>F^-. Residents of Lucknow city have been found to be frequently exposed to particulate pollution and related chemical elements, particularly during haze and inversion occurrences in the winter. It is, therefore, imperative to assess and develop pollution abatement strategies to combat urban air pollution.

Keywords: Particulate matter, Urban air pollution, Trace elements, Ionic species, Inversion.

Highlights

- PM_{10} surged by 238, 186, and 135% w.r.t. NAAQ standard of 100 μg/m^3, while $PM_{2.5}$ exceeded NAAQS value of 60 μg/m^3 by 224 to 215, 196, and 136, respectively for a commercial, industrial and rural area, respectively.
- SO_2 and NO_2 levels at selected sites were well within their NAAQ standard of 80 μg/m^3, indicating fewer sources of gaseous pollutant emission than aerosols in the city.
- Elemental concentrations were 41 to 480% higher in commercial locations than in industrial locations and 129 to 850% higher compared to rural locations.
- AQI based on the location was under the 'very poor' category and hence symbolized chances of "Respiratory illness to the people on prolonged exposure".

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Introduction

Urban air pollution is a global human health dilemma, leading to increased morbidity and mortality. Air pollution is the foremost risk factor for human health impairments like enhancement in mortality rate among people with hypertension, tobacco smoking and high glucose levels (WHO, 2023). People are seriously affected and about two million mortalities have been estimated worldwide to be linked with air pollution through diseases like asthma, respiratory diseases, bronchitis, hypertension, chronic obstructive pulmonary disease (COPD), atrial fibrillation and cancer (Abdurrahman *et al*., 2020; Kwon *et al*., 2019). Air pollution has been on the rise in developing countries like India, disproportionately leading to an increased burden of disease on infants, pregnant women and the elderly (Saxena *et al*., 2022; Balakrishnan *et al*., 2019). Fine particulate matter, nitrogen oxides, heavy metals, organic compounds, carbon monoxide and sulphur oxides from vehicles and industry, and home and agriculture contribute the most air pollution in the Asian subcontinent (Saxena *et al*., 2022). The WHO list of worst air quality cities of the world includes several cities of India owing to frequent breaches in air quality guidelines (WHO 2023; Mahato *et al*., 2020). In India, the population is exposed to average concentrations of $PM_{2.5}$ that are much more than the WHO guideline of 15 μg/m^3 (WHO, 2023).

Fine particulates (aerodynamic diameter 2.5μm or lesser) are a threat based on their increasing concentrations in the urban ambient atmosphere. These particles, owing to their re-suspension in the breathing zone, can penetrate deeply into

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various organs of the human body via systemic circulation and cause diseases of the heart, lungs and kidneys (Naqvi *et al.*, 2023; Abdurrahman *et al.*, 2020). Ultrafine particles stay aloft for a longer period of time than larger and heavier particles (aerodynamic diameter 10µm or greater), which tend to sink soon after emission due to gravity (Kumar *et al.*, 2023). Fine particulate matter is injurious to health due to the adsorption/absorption of additional components such as heavy metals and organic compounds.

The majority of Indian cities located in the middle Indo-Gangetic plains (IGP) are subject to ambient pollution from several sources, including burning crop residue, industrial emissions, and other fugitive emissions from vehicles (Singh *et al.*, 2017). Annual average particulate concentrations have been reported to be higher across the Northern states of Uttar Pradesh, Delhi, Uttarakhand, Rajasthan, Jharkhand and Punjab in India (Agarwal *et al.*, 2021; Pant *et al.*, 2019). Delhi emerged as the state with the highest annual population-weighted mean $PM_{2.5}$ concentration, exceeding 125 µg/m^3, followed by Uttar Pradesh, Bihar, and Haryana (Balakrishnan *et al.*, 2019).

Lucknow, the capital city of Uttar Pradesh, is notorious for air pollution, along with many cities that lie in the IGP (Gautam *et al.*, 2021; Srivastava *et al.*, 2020). IGP's air quality is mostly influenced by its geography and meteorology. Another influence is the occurrence of episodic events like the celebration of the Diwali festival, Parali burning or wintertime inversion, which have the potential to raise pollutant concentrations and further degrade air quality. Warmer air traps the pollutant-embedded cooler air near the ground surface, setting a stage for complexation and chelation and forming secondary pollutants. As such, pollutant levels rise manifolds, affecting both humans and the environment disproportionately.

Therefore, it is imperative to understand the environmental concerns associated with ambient air pollutants and their possible impact on air quality. With the above genesis, the present study was carried out at three sampling locations over Lucknow city, representing commercial, industrial and rural areas. The objective of the study was to monitor particulate matter (PM_{10}, $PM_{2.5}$), gaseous pollutants (SO_2, NO_2, O_3), trace elements (Al, Fe, Mg, Zn, Pb, Mn, Cr, Cu, Ni, Co, Cd, and Mo) and water soluble ionic species (Na^+, NH_4^+, K^+, Ca^{2+}, F^-, Cl^-, $NO_3^-$and SO_4^{2-}). Statistical tools and the Central Pollution Control Board (CPCB) AQI calculator were used to evaluate AQI for the sampling locations and to assess correlations among the trace elements and air quality parameters.

Materials And Methods

Study Area

The study area for the present work was Lucknow, the capital city of Uttar Pradesh, which is nearly centered in the state and houses a population of more than 3.5 million. Selected sampling locations were Charbagh (26.50'08°N, 80.55'48°E), Sarojini Nagar (26.43'39°N, 80.50'51°E) and Banthara (26.41'22°N, 80.49'03°E) which were classified as Commercial, Industrial and Rural areas, respectively.

Sampling of Particulates and Gaseous Pollutants

The particulate matter sampling was carried out twice in a month for 24 hr duration from November 2022 to February 2023 at all sampling points using a conditioned 8*10-inch EPM2000 filter paper placed in a Respirable Dust Sampler (RDS) for PM_{10} collection, whereas $PM_{2.5}$ sampling was carried out using a 47 mm diameter quartz filter placed in a Fine Particulate Sampler (FPS). SO_2 and NO_2 gases were sampled using an impinger attached to the RDS, and their concentrations were determined following the methodology indicated in CPCB NAAQS, 2009 manual. Ozone was measured for 1 hr duration during 12:00 noon to 1:00 pm using a real-time ozone analyzer from 2B Tech, USA. AQI was calculated in accordance with the CPCB AQI calculator.

Trace Elements and Ionic Species Analysis

Trace element concentrations were measured using ICP-MS (iCAP RQ, Thermo Scientific). The $PM_{2.5}$ filters were digested using acid digestion method with HNO_3: $HClO_4$ (4:1v/v) until the appearance of dense white fumes. This was followed by syringe filtration and sample makeup to 10 ml with 1% HNO_3. The samples were run on ICP-MS to determine the concentrations of Al, Fe, Mg, Zn, Pb, Mn, Cr, Cu, Ni, Co, Cd, and Mo. Further, ionic constituents were determined using Ion Chromatography-IC (Metrohm, Germany). $PM_{2.5}$ filters were dipped overnight in 10 ml MilliQ water and sonicated for 30 min. Thereafter, the samples were syringe filtered and subjected to IC for determination of anions and cations.

Furthermore, anion-cation (AC) ratio was computed to understand the nature of pollutants by accepted ion balance equations (Saxena *et al.*, 2022; Zhang *et al.*, 2011).

Cation microequivalents =$Ca^{2+}/20+NH_4^+/18+ Na^+/23+K^+/39$
Anion microequivalents = $NO_3^-/62+ Cl^-/35.5+ F^-/19+SO_4^{2-}/48$

Statistical Analysis

To determine the correlation between the trace metals and air pollutants, IBM SPSS software (ver.26) was utilized to estimate the Pearson correlation matrix at both 0.05 and 0.01 levels of significance.

Results And Discussion

Particulate Matter and Gaseous Pollutant concentrations

PM_{10} values ranged from 159.8 to 480.3 µg/m^3 across all sampling locations during the entire campaign. The average PM_{10} level were, for commercial (337.9 ± 94.2), industrial (285.6 ± 53.1) and rural areas (234.9 ± 43.0 µg/m^3), respectively. On the other hand, levels of $PM_{2.5}$ ranged from 107.4 to 262.0 µg/m^3. The average $PM_{2.5}$ levels were 189.1 ± 57.6, 177.7 ± 36.5 and 141.6 ± 23.0 µg/m^3 for commercial, industrial and rural areas, respectively. Thus the average concentration of PM_{10} surged by 238%, 186%, and 135% respectively w.r.t. NAAQS standard of 100 µg/m^3 while $PM_{2.5}$ level surged by 224 to 215%, 196%, and 136%, respectively w.r.t. NAAQS standard of 60 µg/m^3. Temperature dip during the winter season in northern areas of India is mainly due to the infiltration of northern Himalayan cold front winds which leads to the creation of an inversion layer. Furthermore, a dip in air temperature at night, along with a fall in mixing height, enhances the accumulation of aerosol particles and their hazardous components near the ground surface. Such phenomenon might be one of the causative factors behind the increased pollutant concentrations during winter season in the middle Indo Gangetic Plains (Jain *et al.*, 2021; Sen *et al.*, 2017). Highest pollutant concentrations were recorded for the commercial area and least for the rural area. Concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2 and O_3 for the study period have been depicted in Fig. 1.

SO_2 concentrations ranged from 8.7 to 23.9 µg/m^3 across all locations, with average value of 21.2 ± 3.2, 20.9 ± 4.0 and 14.5 ± 2.4 µg/m^3 for commercial, industrial and rural locations, respectively, whereas NO_2 concentrations were ranged from 16.8 to 45.7 µg/m^3 with average value 36.5 ± 5.6, 31.8 ± 5.6 and 24.8 ± 3.0 µg/m^3 for commercial, industrial and rural locations, respectively. Values of both SO_2 and NO_2 were well within their NAAQ standard of 80 µg/m^3 indicating less gaseous emission which was under permissible limit for all selected categories. Also, most of the gaseous pollutants eventually form aerosol particles via complexation and gas to particle conversion (Zhang *et al.*, 2021). Hence, their concentrations in ambient air remain lower than particulate matter. During haze events, chelates like Fe and Mn expedite the process of conversion of secondary sulphates from SO_2 (Bloss *et al.*, 2021).

O_3 concentrations ranged from 26.3 to 116.1µg/m^3 for the selected locations, with the commercial, industrial and rural locations having average concentrations of 102.5 ± 7.6, 64.8 ± 9.8 and 41.9 ± 10.7 µg/m^3, respectively, which was within NAAQS limit (180 µg/m^3) for one-hr. Ozone is a secondary pollutant arising out of the photochemical oxidation reaction between nitrogen oxides and volatile organic compounds in the presence of sunlight (Kumari *et al.*, 2020; Wang *et al.*, 2017). Therefore, tropospheric ozone concentration has two drivers, one sunlight and the other NO_x and VOCs. In the winter season although solar flux is low, locations with higher pollutant concentration tend to register higher ozone concentrations than lesser polluted counterparts as seen in the present study.

AQI was calculated for the three study areas and values were 353, 344, and 317 for commercial, industrial and rural area, respectively which denoted 'very poor' category of AQI and hence symbolized chances of "Respiratory illness to the people on prolonged exposure".

Trace elements associated with $PM_{2.5}$

Twelve elemental constituents associated with $PM_{2.5}$ were assessed, and their concentrations have been detailed in Fig. 2. Order of abundance of elemental species with respect to $PM_{2.5}$ was Al>Fe>Mg>Zn>Pb>Mn>Cr>Cu>Ni>Co>Cd>Mo across all sampling locations. Average elemental concentrations in commercial location were 41-480% higher compared to industrial location and 129-850% higher when compared to rural location. On the other hand, average elemental concentration in industrial location was 51-779% higher as compared to rural location. The elemental concentrations were higher in commercial location as compared to industrial and rural, and this increase spanned across many elements. Metallic species adhered on particles have a negative impact on all organ systems in the human body, producing systemic poisoning and occasionally initiating toxic and carcinogenic consequences (Briffa *et al.*, 2020; Yuan *et al.*, 2019).

Additionally, contribution of particulate bound elements to the total particle mass was calculated in terms of percentage. It was found that percent contribution of sum of elemental constituents to the $PM_{2.5}$ mass loading was 10.3% for commercial area out of which crustal elements like Mg, Al, Fe, Zn and Mn contributed 8.8% whereas trace elements like Pb, Ni, Mo, Cu, Co, Cd and Cr contributed the remaining 1.5%. Similarly, these percentages of total particulate bound elements, total crustal element and total trace element for industrial area were 5.7, 4.5 and 1.2%, and those for rural area were 4.7, 4.2 and 0.5%, respectively. Toxic metals such as Ni, Cr, Cu, Co, which promote the generation of reactive oxygen species in cells, resulting in cell damage and increment in oxidative stress (Hime *et al.*, 2018).

Ionic constituents associated with $PM_{2.5}$

Cations (Na^+, NH_4^+, K^+, Ca^{2+}) and anions (F^-, Cl^-, NO_3^-, SO_4^{2-}) associated with $PM_{2.5}$ were determined by ion chromatography

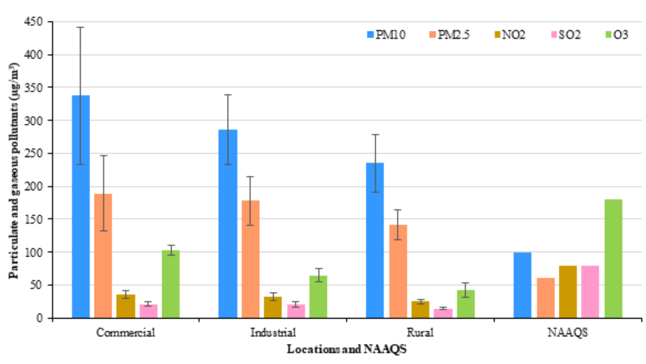

Fig. 1: Concentrations of particulate and gaseous pollutants

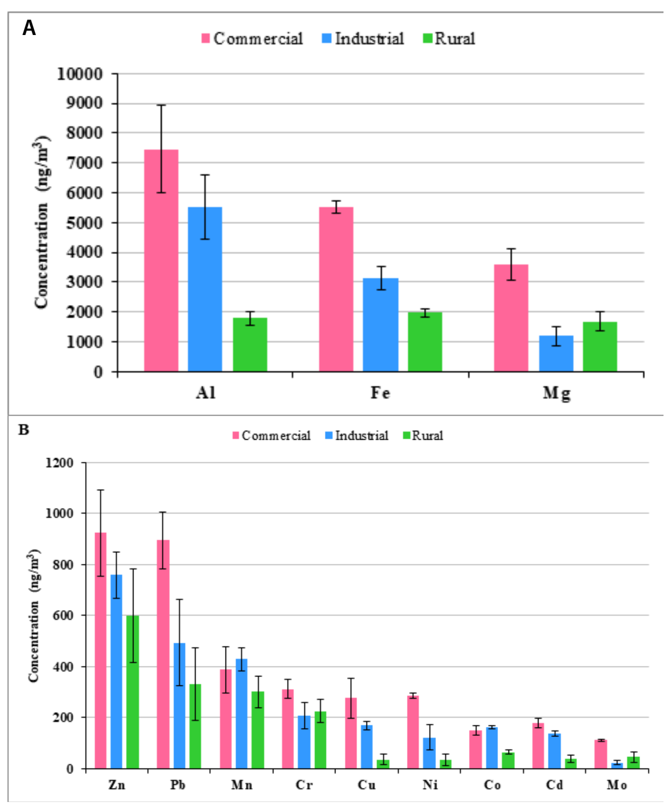

Fig. 2A and B: Concentrations of heavy metals associated with $PM_{2.5}$

and results have been compiled in Table 1. Mean concentrations of ions (in μg/m^3) were in the order F^-(0.27)< Na^+(0.98)< Ca^{2+}(1.67)< NO_3^-(1.85)< NH4$^+$(2.58)< K^+(2.79)< Cl^-(3.25)< SO_4^{2-} (12.02); indicating F^- to be the least and SO_4^{2-} to be the most predominant ion. Percentage contribution of sum of ionic constituents to the $PM_{2.5}$ mass loading was 16.4%, 13.4% and 13.5%, for commercial, industrial and rural area, respectively.

A/C ratio was found to be 1.11 for commercial and 1.43 for rural area whereas it was 0.96 for industrial area. This indicated that the particles for commercial and rural area were acidic in nature, whereas the industrial particle was basic by nature.

Correlation among air pollutants

Pearson's correlation analysis at both 0.05 and 0.01 levels of significance illustrated the correlation between particulate and gaseous pollutants and trace metals. The Pearson's correlation analysis results are shown in Table 2. The results depict that $PM_{2.5}$ is positively correlated with SO_2 at $p<0.01$ whereas with Al, Fe, Ni, Cd and Pb at $p<0.05$ which depict sources of $PM_{2.5}$ to be road dust re-suspension and vehicular exhaust. SO_2 is correlated with NO_2 at $p<0.05$. NO_2 is positively correlated with Mg, Fe, Ni and Mo at $p<0.01$ whereas at $p<0.05$ with Cu and Cr. O_3 is only correlated at $p<0.05$ with Pb showing vehicular engine emissions to be the predominant causatives behind tropospheric ozone. Al with Fe, Co, Ni, Cd, Pb; Fe with Ni, Cu, Mo, Cd, Pb; Co with Cd; Ni with Cu, Cd, Pb; and Cu with Cd are positively correlated at $p<0.01$. However, Al with Cu, Zn; Fe with Cr, Zn; Cr with Mo; Mn with Co; Co with Cu; Ni with Mo; Cu with Pb;Zn with Cd, Pb; Mo with Pb and Cd with Pb are positively correlated at $p<0.05$. As evident, most of the trace metals are highly correlated within themselves showing cumulative impact of group sources like vehicular emission, dust entrainment and biomass burning.

Table 1: Concentrations of anions and cations (in μg/m^3) associated with $PM_{2.5}$

Location/ Ions	*Commercial area*			*Industrial area*			*Rural area*		
	Min	*Max*	*Avg+SD*	*Min*	*Max*	*Avg+SD*	*Min*	*Max*	*Avg+SD*
F^-	0.19	0.32	0.27 ± 0.07	0.3	0.33	0.31 ± 0.02	0.18	0.26	0.23 ± 0.04
Cl^-	3.14	3.46	3.28 ± 0.16	2.79	3.14	3.02 ± 0.19	2.87	4.07	3.44 ± 0.61
NO_3^-	1.83	5.29	3.42 ± 1.71	ND	2.32	1.27 ± 1.17	ND	1.32	0.85 ± 0.74
SO_4^{2-}	10.51	20.52	14.64 ± 5.22	7.58	14.23	11.15 ± 3.35	9.41	11.06	10.27 ± 0.82
Na^+	0.96	1.51	1.18 ± 0.29	0.2	1.15	0.75 ± 0.49	0.8	1.16	1.01 ± 0.18
NH_4^+	1.56	3.5	2.85 ± 1.12	1.04	6.52	3.74 ± 2.74	1.1	1.23	1.15 ± 0.07
K^+	3.86	4.12	4.03 ± 0.15	0.49	3.86	2.27 ± 1.69	1.87	2.46	2.07 ± 0.34
Ca^{2+}	1.63	2.9	2.14 ± 0.67	1.19	1.64	1.41 ± 0.23	0.99	1.72	1.46 ± 0.41

Table 2: Pearson correlation matrix between gaseous pollutants and trace elements associated with $PM_{2.5}$

	$PM_{2.5}$	*SO_2*	*NO_2*	*O_3*	*Mg*	*Al*	*Fe*	*Cr*	*Mn*	*Co*	*Ni*	*Cu*	*Zn*	*Mo*	*Cd*	*Pb*
$PM_{2.5}$	1															
SO_2	.776**	1														
NO_2	0.051	.424*	1													
O_3	-0.577	-0.511	0.499	1												
Mg	-0.508	-0.201	.901**	0.593	1											
Al	0.776*	-0.275	0.439	0.77	0.57	1										
Fe	0.894*	-0.38	.818**	0.53	.837**	.805**	1									
Cr	-0.277	0.079	.732*	0.423	.741*	0.4	.728*	1								
Mn	0.046	-0.31	-0.204	0.325	0.007	0.634	0.313	-0.019	1							
Co	0.01	-0.274	0.15	-0.433	0.233	.825**	0.64	0.111	.723*	1						
Ni	0.765*	-0.461	.802**	0.77	.818**	.839**	.957**	0.629	0.397	0.641	1					
Cu	-0.468	-0.48	.696*	0.105	0.633	.740*	.919**	0.481	0.299	.760*	.861**	1				
Zn	-0.365	-0.38	0.400	0.707	0.38	.707*	.679*	0.593	0.522	0.481	0.661	0.581	1			
Mo	-0.66	-0.308	.885**	0.598	.954**	0.512	.810**	.704*	-0.067	0.175	.762*	0.618	0.357	1		
Cd	0.817*	-0.418	0.478	0.553	0.486	.875**	.875**	0.501	0.601	.866**	.841**	.887**	.775*	0.483	1	
Pb	0.812*	-0.632	0.651	.910*	.737*	.803**	.862**	0.523	0.572	0.575	.919**	.738*	.732*	.713*	.789*	1

**Correlation is significant at the 0.01 level (2-tailed). *Correlation is significant at the 0.05 level (2-tailed).

Conclusion

Ambient air pollutants *viz*. PM_{10}, $PM_{2.5}$, SO_2, NO_2 and O_3 were estimated along with trace metals and water-soluble ions at three locations in Lucknow city in UP, India, during the winter season of the year 2022-23. Results revealed that PM_{10} surged by 135-238% w.r.t. NAAQ standard of 100 μg/m^3 and $PM_{2.5}$ surged over all locations by 136-224% above the standard limit of 60 μg/m^3. Although PM concentrations were found to be the lowest in the rural areas, even those breached NAAQ standards. SO_2, NO_2 and O_3 registered concentrations within the NAAQ standards, implying lesser sources and in turn lesser toxicity. Higher concentrations of Al, Fe, and Mg linked with $PM_{2.5}$ indicated the crustal nature of the particle and hence the predominant sources of the wintertime aerosol may be crustal re-suspension and roadside re-suspension, whereas the presence of ions like K^+ and SO_4^{2-} indicate biomass and coal burning for heating and cooking purposes. Statistics indicated most of the trace metals are highly correlated within themselves showing cumulative impact of group sources like vehicular emission, dust entrainment and biomass burning. This study might be useful in chemical speciation of the ambient aerosol of the urban city, Lucknow. This may be an inevitable source of information to regulatory authorities for implementation of viable rules and standards in the near future.

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Author Contributions

Priya Saxena performed data collection and curation, drew table, graph, and wrote the original draft of manuscript. Ankit Kumar helped in collection of samples and figure preparation, and performed statistical analysis. Komal Sharma helped in sample analysis. Altaf Husain Khan designed study and proofread manuscript. Alka Kumari conceptualized, study designing, proofread and approved manuscript submission.

Conflict Of Interest

The authors declare no conflicting or competing interests. All the authors significantly contributed and approved for submission.

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