Comprehensive assessment of heavy metal contaminations in agricultural soil through Pollution Indices from a rapidly developing city of India

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Abstract
This study has been conducted for the comprehensive evaluation of heavy metals (HMs) contamination such as Cr, Mn, Fe, Ni, Cu, Zn, Cd and Pb in agricultural soils via pollution indices i.e. geoaccumulation index (Igeo), contamination degree (C_D), pollution load index (PLI), potential ecological risk (PRI) etc. Assessment for the determination of ecological damage was caused by these HMs in different agro-ecosystems around metropolitan city Lucknow. For this, data has been taken from our previous study[8] depicting significant enrichment of above mentioned HMs in agricultural soil present near riparian areas, brick kilns, waste dump sites etc. Our results indicated that soil samples collected from Gaughat (GAF) and Sitapur (SAF) showed very high degree of contamination (C_D>24). Further, pollution load index also reflected "very high" and "moderate to high" contamination with values 6.9075 and 3.3214 in these two sites respectively. Overall, Igeo values indicated moderate contamination with Mn and Ni metals and low contamination with Cr, Cu, Zn and Cd metals.

However, Igeo with value 4.923 indicated extremely high degree of contamination due to Cd in Gaughat (GAF) soil. In the view of potential ecological risk index, GAF site poses a "very high" potential ecological risk while maximum sites pose "moderate" ecological risk. The ecological risk factor in agricultural soil was in the following descending order: Cd > Ni > Cu > Pb > Cr > Mn > Zn > Fe. Cadmium predominantly contributes significant amount to the PRI of soil while Ni, Cu, Pb, Cr, Mn and Zn showed low ecological risk.

Keywords: Heavy metals, Pollution indices, Contamination factor, Ecological risk.

Introduction
Soil contamination with heavy metals (HMs) in urban areas or typical towns has continued to capture a great attention from the environmental explorers and public around the world because of their toxic nature and close association with human health[1,18]. Increasing industries, rapid development of towns and expanding vehicular emissions have largely surpassed HMs contamination in the soil of urban areas[1]. Recently, some researchers revealed that certain HMs such as As, Pb, Cd and Ni are non- vital to different metabolic and biological functions[6,31] and have been added in the category of 20 top dangerous substances by United State Environmental Protection Agency (USEPA) and Agency of Toxic Substances and Disease Registry (ATSDR)[19,31,37].

Higher occurrence of HMs in agricultural soil is problematic and creates a universal environmental problem due to its crucial importance for the crop production and food security[17,22]. Increasing awareness of ever-spreading industrialization and intensive usage of agricultural soil and their impact on the level of HMs in the soil demand the proper estimation as well as assessment of ecological risks[1,4,21].

Pollution indices (PI) are widely recognized emphatic tools for the comprehensive evaluation and assessment of environmental quality, degree of contamination and forecasting of future environmental sustainability[22,35], especially in the agroecosystems. Basically, pollution indices used for the evaluation of HMs are classified into two categories: Single (individual metal) and multi or integrated (sum of all studied metals together) pollution index[11,35]. Single metal PI includes contamination factor (CF), geoaccumulation index (Igeo) and ecological risk factor (ERF) and integrated PI involves contamination degree (C_D), modified contamination degree (mC_D), pollution load index (PLI) and potential ecological risk index (PRI).

Lucknow, the largest city of Uttar Pradesh with 3.76 million populations is facing contamination problems due to expanding industrialization and urbanization. Previously, Kumar et al[23] reported that 32% of vegetables cumulated from different areas of Lucknow city consisted of HMs, above FAO/WHO prescribed permissible limit. Kumar et al[24] also observed that vegetables sampled from different locations of this city contained 2-240 times higher concentration of Pb than its maximum allowable concentration, which indicated contamination of agricultural soils. The comprehensive evaluation of agricultural soil of peri-urban areas is necessary for sustainable development of this city. However, some former studies carried out in this area have mainly focused on the level of HMs in the agricultural soil, spatial distribution, correlations and sources[8,33]. The estimated total HMs concentration, the interrelationships between the HMs content and properties of soil such as regression or correlation analysis and statistical mechanisms do not provide extensive knowledge on the degree of soil's contamination[21,28]. Earlier, a geochemical investigation has created a pervasive database of HMs background values of Lucknow[10] that can now be explored for the estimation of environmental quality.

Recently, the bioaccumulation, ecological risk and health hazards caused by carcinogenic metals such as As, Cr, Ni, Pb and Cd in the agricultural ecosystem were demonstrated[25]. The objective of present study is to assess the degree of intensity of HMs contamination in agricultural soils near different contamination sources such as riparian areas, municipal waste dumping sites, brick kilns etc. through pollution indices.

Material and Methods

Lucknow, the capital of state Uttar Pradesh and a big city of northern India, has an area of about 310 km^2 in the centric (or pivotal or middlemost) plain of Indian subcontinent. It lies between 23°52′–31°28′N and 77°3′– 84°39′E and 128 m above sea level. Agriculture is the main occupation of people inhabiting in rural areas of this city. Crop and vegetable namely *Chenopodium album* (bathua), *Spinach oleracea* (spinach), *Trigonella foenum-graecum* (meethi), *Triticum aestivum* (wheat), *Raphanus sativus* (radish), *Brassica juncea* (mustard), *Solanum tuberosum* (potato), *Brassica oleracea var. Botrytis* (cauliflower), *Brassica oleracea var. capitata* (cabbage) etc. are commonly cultivated in these areas.

However, this study involves quantification of soil contamination of formerly monitored agricultural soil[8] on the basis of various pollution indices including ecological risks and demarcation of contaminated agricultural lands. A brief description of the studied sites and the levels of reported HMs in agricultural soil is mentioned in the supplementary table 1 and 2.

Contamination and ecological risk assessment: For comprehensive evaluation of the contamination levels of HMs in the agricultural soil, different calculated pollution indices are mentioned below:

Single - metal pollution indices (SMPI): It gives us intellect about how a single metal or element is concentrated at the concerned site relative to its background value and thus it can be utilized to estimate metal contamination level. It includes:

Contamination factor (CF): Soil contamination can be assessed using both the contamination factors (CF) and degree (C_D). CF value was calculated through formula given by Hakanson[12]:

$$CF = \frac{Ca}{Cb} \quad (1)$$

where C_a and C_b denote the concentrations of measured metal at the site (agricultural soil) and background concentration of same metal respectively. However, the background values of Lucknow for Cr, Mn, Fe, Ni, Cu, Zn, Cd and Pb were opted from Gupta and Arya[10] and are mentioned in supplementary table 2. Hakanson[12] classified four classes of degree of contamination present in soil on the basis of CF.

Geoaccumulation index (Igeo): Igeo techniques measure the intensity of HMs contamination in the contaminated environment. Technique evaluates contamination level on the basis of geochemical criteria[2,7]. Igeo value can be calculated using formula given by Muller[29]:

$$Igeo = \log_2\left(\frac{Cn}{1.5\times Bn}\right) \quad (2)$$

The background matrix correlation factor 1.5 was used to lessen the lithogenic variability effect. Muller[29] suggested seven Igeo index classes for contamination assessment.

Ecological risks factor (ERF): The ecological risk factor (ERF) value of a given contaminant was calculated using formula given by Hakanson[12]:

$$ERF = T_r * CF \quad (3)$$

where T_r is "toxic-response" factor for a given metals, Cd = 30, Cr = 2, Pb=Cu = Ni= 5 and Zn =Mn= Fe=1[3,12].

Supplementary Table 1
Description of sampling locations selected for estimation of different Pollution indices

Sites	Area (Code)	Location description
S1	Gaughat (GAF)	Agricultural lands present on Gomti River riparian areas
S2	Mohanlalganj (MAF)	Agricultural lands present on Sai River riparian areas
S3	Sitapur road (SAF)	Agricultural lands present near city's major municipal waste dumping site
S4	Bijnaur (BAF)	Agricultural lands near to brick-kiln industry
S5	Devaroad (DAF)	Agricultural lands present on the roadside
S6	Barabanki (BbAF)	Agricultural lands lying away from any kind of direct contamination source like contaminated river or industries

GAF Gaughat agricultural field; MAF Mohanlalganj agricultural field; SAF Sitapur agricultural field; BAF Bijnaur agricultural field; DAF Devaroad agricultural field; BbAF Barabanki agricultural field.[8]

Table 1
Classification criteria of the CF, Igeo, ERF, C_D, mC_D, PLI and PRI of Heavy metals in the agricultural soil of the study area[2,5,12,13,29,35]

CF	Degree of pollution (Individual metal)	Igeo	Soil quality	ERF	Soil Ecological risk factor classification
CF < 1	Low contamination	Igeo ≤ 0	Practically uncontaminated	ERF < 40	Low Potential ER
1 ≤ CF ≤ 3	Moderate contamination	0 < Igeo ≤ 1	Uncontaminated to moderate	40 ≤ ERF < 80	Moderate potential ER
3 ≤ CF ≤ 6	Considerable	1 < Igeo ≤ 2	Moderate	80 ≤ ERF < 160	Considerable potential ER
CF > 6	Very high contamination	2 < Igeo ≤ 3	Moderate to heavy	160 ≤ ERF < 320	High potential ER
-	-	3 < Igeo ≤ 4	Heavy	ERF ≥ 320	Very high potential ER
-	-	4 < Igeo ≤ 5	Heavy to extreme	-	
-	-	Igeo > 5	Extreme contamination	-	

MULTI-ELEMENT POLLUTION INDEX

C_D	Contamination degree (C_D) classification	mC_D	Modified contamination degree (mC_D) classification	PLI	Classification of soil pollution	PRI	Potential ecological risk classification
$C_D < 6$	Low contamination	$mC_D < 1.5$	Unpolluted	< 1	Unpolluted	PRI < 65	Low risk
$6 \leq C_D < 12$	Moderate	$1.5 \leq mC_D < 2$	Slightly polluted	1 to 2	Unpolluted to moderate	65 ≤ PRI < 130	Moderate risk
$12 \leq C_D < 24$	Considerable	$2 \leq mC_D < 4$	Moderately polluted	2 to 3	Moderate polluted	130 ≤ PRI < 260	Considerable risk
$C_D \geq 24$	Very high contamination	$4 \leq mC_D < 8$	Considerably polluted	3 to 4	Moderate to high	PRI ≥ 260	High risk
-		$8 \leq mC_D < 16$	Highly polluted	4 to 5	High polluted	-	
-		$16 \leq mC_D < 32$	Strongly polluted	> 5	Very high polluted	-	
		$mC_D \geq 32$	Extremely polluted				

CF Contamination factor, EF Enrichment factor, Igeo Geoaccumulation index, ERF Ecological risk factor, C_D Contamination degree, mC_D Modified Contamination degree, PLI Pollution load index, PRI Potential ecological risk index.

Multi- metal pollution indices (MPI): MPI techniques were applied during contamination assessment of soil to combat the limitations of single-metal pollution indices[9]. Most commonly and extensively employed techniques include:

Contamination degree (C_D) and modified contamination degree (mC_D): C_D of soil can be calculated by the addition of contamination factors of all metals at each point[7] while mC_D can be calculated by dividing the value of contamination degree with n number of analysed elements.

$$C_D = \sum_1^n CF \quad (4)$$
$$mC_D = \sum_1^n CF/n \quad (5)$$

where n is the number of studied HMs. The categorization prescribed by Hakanson[12] and Brady et al[5] for assessing contamination degree (C_D) and modified contamination (mC_D) degree was used for soil contamination assessment.

Pollution load index (PLI): PLI explicits the overall toxicity level of contaminants in the sample and PLI is the n^{th} (number of elements) root of multiplied contamination factor (CF) values. It was calculated with the formula given by Tomilson et al[36]:

$$PLI = \sqrt[n]{CF1 * CF2 * CF3 * CF4 * \dots\dots\dots\dots\dots\dots * CFn} \quad (6)$$

Potential ecological risk index (PRI): PRI is extensively used to estimate the potential ecological deficit caused due to HMs[12]. This approach integrates various interdisciplinary fields like ecology, bio-toxicology and environmental chemistry to reflect the impacts of different contaminants on the environment and their comprehensive effects too[27]. PRI is the sum of all calculated ERF for each individual soil[12]:

$$PRI = \sum_{1}^{n} ERF \quad (7)$$

A detailed classification criterion of various pollution indices is summarized in table 1.

Statistical Analysis: The data was calculated by the Excel 2010 (Ms Office, USA) and graphs were created using Prism 5.0 software.

Results and Discussion

Various pollution indices were calculated in respect of Cr, Fe, Ni, Cu, Zn, Cd, Pb and Mn due to availability of local BGV and toxic factor value of selected metals in the studied area. Our previous investigation[8] highlighted a higher concentration of Pb and Cu beyond USEPA guidelines.

Single and integrated metal pollution indices

Geoaccumulation index: Igeo is a powerful numerical model, which has been vastly explored to estimate the contamination caused by HMs in both urban and agricultural soils[2,26,29]. In general, Cr, Cu, Zn and Cd emerged out as less to moderate contaminated (figure 1); Mn and Ni as moderately high whereas Fe and Pb were least contaminated among the studied sites. However, Cd had exceptionally high Igeo value at GAF site. The obtained Igeo values of each individual metal among the studied sites varied greatly, exhibiting the variability of edaphic properties and HMs contamination sources[16].

The Igeo of Fe showed practically no contamination in 66.66 % of total studied sites (MAF, BAF, DAF and BbAF) whereas GAF (Igeo =1.132) showed moderate and SAF (Igeo = 0.916) showed uncontaminated to moderate contamination. Igeo value below 0 indicates no contamination of agricultural soil through different sources. Further, Igeo value of Cr showed moderate level pollution in GAF (Igeo =1.942) and SAF (Igeo =1.593) sites while 49.99 % of studied sites (MAF, BAF and BbAF) reflected low to moderate contamination. Similarly, Igeo of Zn also reflected moderate contamination in GAF and SAF sites and no pollution in MAF while nearly 50% sites showed uncontaminated to moderate contamination.

Additionally, Igeo of Mn exhibited moderate to heavy contamination in GAF (Igeo = 2.177) and SAF (Igeo = 2.141) sites whereas BAF and BbAF soil were moderately contaminated. The Igeo value of Ni exhibited moderate contamination in 66.66 % of monitored sites, except for GAF (Igeo =2.102) showing moderate to heavy contamination and MAF (Igeo = 0.517) showing non-contaminated to moderate contamination. However, the greatest Igeo value of Cu metal was measured from the GAF site (Igeo = 2.153) depicting moderate to heavy pollution in soil and negative impact on the agricultural soil while no pollution was observed in MAF (Igeo = -0.081) and DAF (Igeo = -0.077) sites.

Supplementary Table 2
Concentrations of various heavy metals reported in the agricultural soil[8,10]

Sites	Statistics	Cr (mg kg^{-1})	Mn (mg kg^{-1})	Fe (mg kg^{-1})	Ni (mg kg^{-1})	Cu (mg kg^{-1})	Zn (mg kg^{-1})	Cd (mg kg^{-1})	Pb (mg kg^{-1})
GAF	Mean	16.669	301.797	10873.894	19.633	30.545	92.581	12.700	16.932
	Range	(12.659-24.950)	(285.683-316.343)	(8954.994-14097.381)	(17.066-23.801)	(17.947-59.545)	(50.963-171.868)	(0.604-48.887)	(7.481-27.823)
MAF	Mean	5.848	96.386	3981.451	6.725	6.046	17.638	0.104	4.645
	Range	(4.146-8.565)	(52.693-132.789)	(2537.812-5692.208)	(4.212-9.197)	(4.501-8.774)	(11.820-28.277)	(0.027-0.196)	(2.807-5.840)
SAF	Mean	12.948	296.676	9371.392	15.993	12.374	48.610	0.368	8.321
	Range	(9.461-16.943)	(242.602-341.604)	(7148.399-11802.738)	(12.371-17.564)	(9.285-15.477)	(36.884-59.677)	(0.100-1.091)	(6.329-9.792)
BAF	Mean	8.978	173.531	4975.085	11.157	8.469	25.399	0.151	5.745
	Range	(4.907-14.499)	(133.165-236.181)	(2655.592-7923.977)	(7.865-12.950)	(5.022-11.969)	(18.567-31.011)	(0.017-0.577)	(3.023-9.228)
DAF	Mean	2.956	126.952	2153.118	13.195	5.947	33.204	0.169	2.652
	Range	(0.974-6.255)	(82.441-153.004)	(1155.084-3462.873)	(8.529-17.689)	(4.585-6.809)	(21.137-39.575)	(0.088-0.316)	(1.392-4.144)
BbAF	Mean	6.288	207.311	3287.758	11.787	11.043	31.638	0.109	5.590
	Range	(1.296-12.116)	(176.091-252.582)	(1394.96-6463.849)	(8.048-16.833)	(5.972-17.429)	(21.651-44.717)	(0.069-0.241)	(1.249-7.843)
Background values of Lucknow		2.80	44.45	3259.68	3.03	4.12	15.25	0.09	5.68

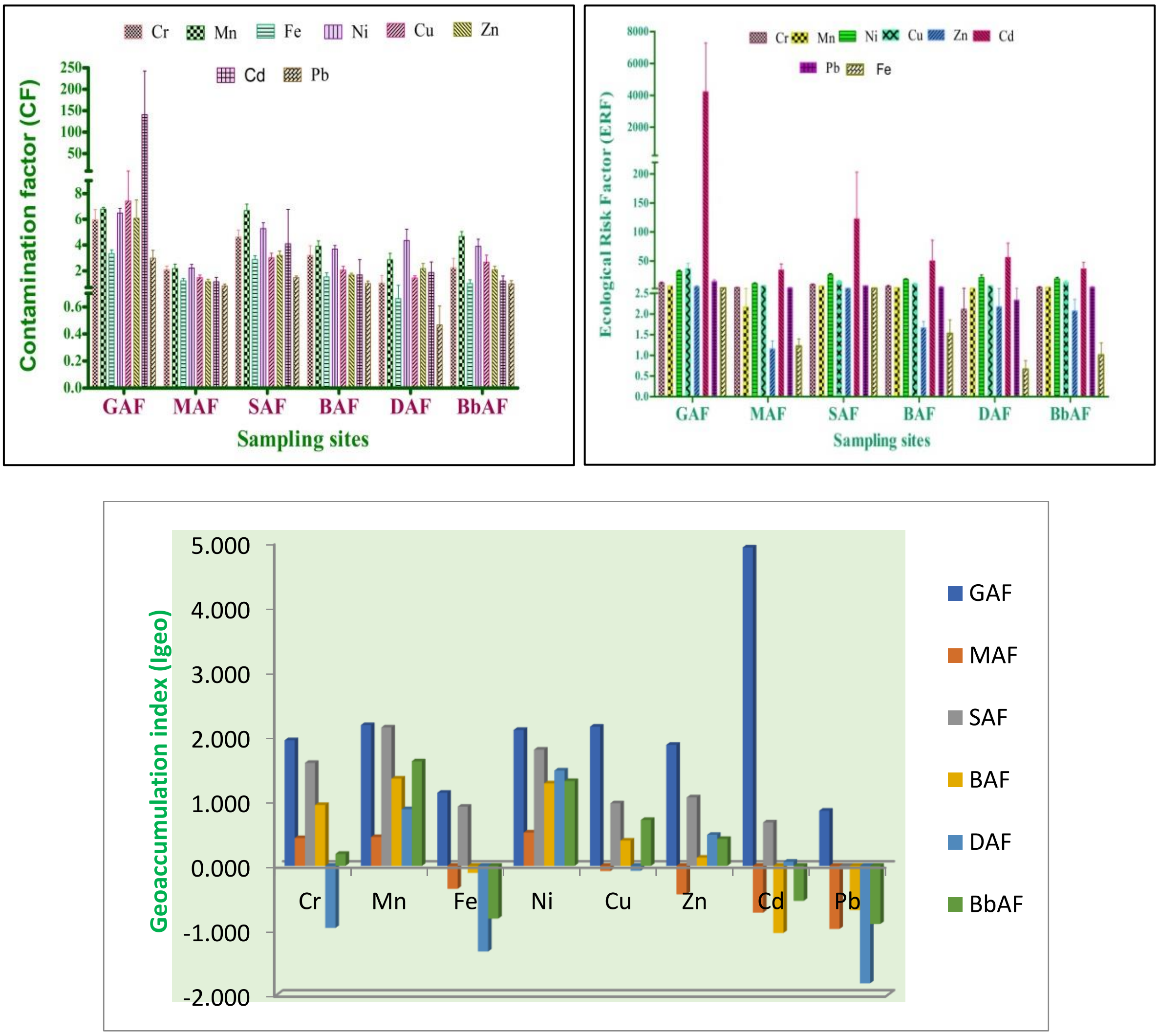

Fig. 1: Mean values of various single element pollution indices CF, Igeo and ERF of the studied sites

Among all studied metals, Igeo of Cd exhibited heavy to extreme level contamination in GAF (Igeo = 4.923) whereas nearly 50 % sites (MAF, BAF and BbAF) were practically non-polluted. Contrarily, Igeo of Pb in agricultural soil showed practically no contamination in 83.33 % of total investigated sites. Overall, the descending order of Igeo among the investigated sites was as GAF > SAF > BAF > BbAF > MAF > DAF.

Contamination factor: The calculated CF of Cd indicates very high (CF >6) level of contamination in the GAF site while 66.66 % of total selected sites consisted of moderate contamination (figure 1). The CF of Cr indicates nearly 50% of studied sites including GAF, SAF and BAF with values 5.953, 4.624 and 3.206 which were considerably contaminated with Cr, while rest 50% sites had moderate level contamination. The CF of Mn showed very high contamination in GAF and SAF sites, whereas rest of the sites showed moderate (MAF and DAF) to considerable (BAF and BbAF) contamination. Further, the CF of Fe showed moderate level of contamination in 66.66 % of monitored sites, except for GAF (3.336) and DAF (0.660) showing considerable and low contamination respectively.

The CF of Pb reflected low to moderate level pollution among the sites. The CF of Ni reflected very high contamination in GAF (6.480) and moderate contamination in MAF (2.220) site while remaining 66.66 % of total sites showed considerable level of Ni contamination in the agricultural soils. The CF value of Cu and Zn showed similar results where GAF showed very high and SAF showed considerable level of contamination with these two metals and remaining sites were moderately contaminated.

Overall, the decreasing trend of CF of the agricultural soil through various HMs was observed as: Cd > Cu > Mn > Ni > Zn > Cr > Fe > Pb in GAF; Ni > Mn > Cr > Cu > Fe > Zn > Cd > Pb in MAF; Mn > Ni > Cr > Cd > Zn > Cu > Fe > Pb in SAF; Mn > Ni > Cr > Cu > Cd > Zn > Fe > Pb in BAF; Ni > Mn > Zn > Cd > Cu > Cr > Fe > Pb in DAF and Mn > Ni > Cu > Cr > Zn > Cd > Fe > Pb in BbAF. This trend of CF value shows dominance of Mn and Ni in mostly studied sites whereas Fe and Pb were least dominated.

Among all monitored agricultural sites, GAF revealed very high contamination (CF > 6) of Mn, Ni, Cu, Zn and Cd in the soil signifying heterogenous anthropogenic pollution sources, exhibiting that these toxic and trace metals can cause potential carcinogenic and non-carcinogenic health risks to surrounding manmade ecosystem[13,32]. Previously, Kumar et al[25] revealed very high contamination of Cd and low contamination of Ni, Cr and Pb in all the soil samples collected from the Biswan, Khairabad, Sindhuali, Pasonda and Mohanlalganj areas of Lucknow city.

Pollution load index, degree and modified degree of contamination: The overall contamination assessment of studied sites was done on the basis of contamination degree (C_D) and modified contamination degree (mC_D), which are illustrated in figure 2. Results depicted that two sites GAF and SAF had very high degree of contamination ($C_d > 24$) whereas 66.66% of sites had considerable contamination of HMs. However, mC_D revealed strong pollution in GAF site, moderate pollution at SAF, BAF, BbAF and slight pollution in MAF and DAF.

PLI value equal to zero signifies perfection, 1 represents only occurrence of baseline level of the various contaminants whereas value more than 1 reflects progressive soil contamination by the trace metals[14,32]. As per these grades, in this study, the agricultural soils collected from various sites were considerably contaminated with the HMs (figure 2) as the PLI value was greater than 1[13].

The highest PLI value was perceived at GAF (6.9075) followed by SAF (3.3214) and least value was obtained in the DAF (1.3990). Both GAF and SAF indicated very high pollution and moderately to high pollution with the HMs respectively. An elevated level of PLI in the studied sites suggested that use of wastewater for irrigation, agrochemicals, municipal waste disposal sites and brick-kiln industries near the agricultural fields, to some extent might have caused ecological risk to the peri-urban agricultural lands[3,34,38].

Potential ecological risk factor and index: According to the potential ecological risk factor (ERF) and potential ecological risk index (PRI) of the monitored agro-ecosystem (figure 1 and 2), soil from GAF site posed very high potential ecological risk ($PRI \geq 260$) and SAF showed considerable potential ecological risk ($130 \leq PRI < 260$). The ERF in the agricultural soils was in the following descending order: Cd > Ni > Cu > Pb > Cr > Mn > Zn > Fe.

Conceivable variations were noticed for ERF of individual elements, signifying that ecological risk of these metals varied with different agricultural sites. Almost all agricultural soils showed low ecological risk ($ER < 40$) with respect to Cr which varied from 2.111- 11.907, Mn (2.168 - 6.790), Ni (11.098 - 32.398), Cu (7.218 - 37.069), Zn (1.157 – 6.071), Fe (0.661 - 3.336) and Pb (2.335 – 14.904), except cadmium which represented varied results. Cd showed very high ecological risk ($ERF \geq 320$) in GAF site and a considerable risk ($80 \leq ERF \leq 160$) in SAF while 66.66% agricultural sites posed low to moderate ecological risk. Cd significantly contributes to the PRI of the agricultural soil which might be due to use of phosphate fertilizers and wastewater for irrigation[20], use of municipal sewage sludge and manure etc.[15,30]

PRI indicates sensitivity of the biological communities to numerous toxic substances and demonstrates the probable ecological risks resulted by trace metals[13]. The PRI of the agricultural soil for the different sites can be arranged in the following order: GAF > SAF > BAF > DAF > BbAF > MAF. Overall, the PRI value for all agricultural sites varied from 65.801 – 4345.744, reflecting moderate to very high ecological risks. The highest value of PRI (4345.744 in GAF site) denotes very high potential ecological risk for agricultural land due to usage of polluted river water for irrigational purposes and flooding.

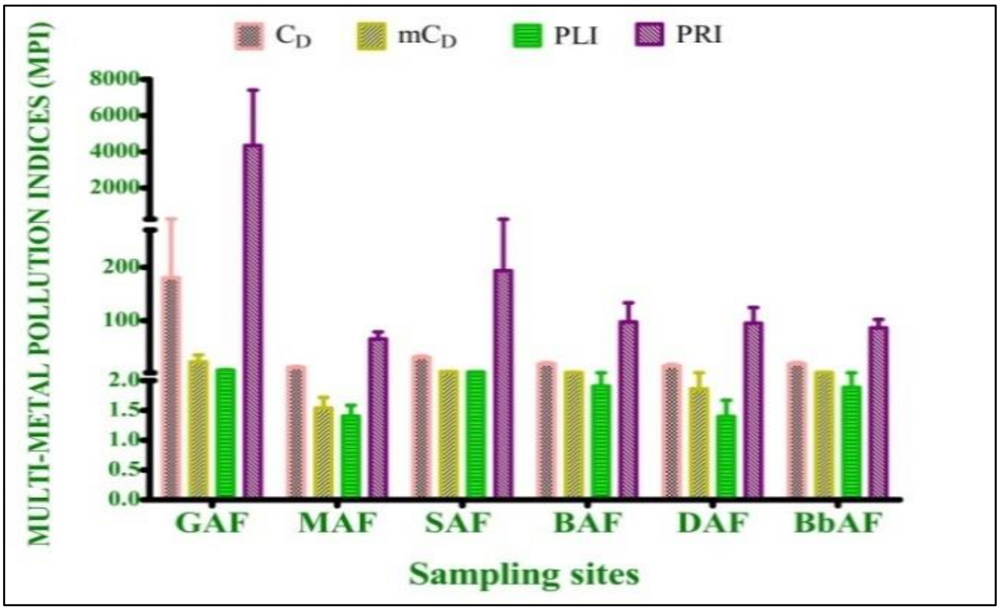

Fig. 2: Mean values of various multi- element pollution indices C_D, mC_D, PLI and PRI of the studied sites

https://doi.org/10.25303/2708rjce1150122

Conclusion

This study confirmed that HMs from the different sites showed low to high degree of contamination on the basis of calculated values of different pollution indices. However, the agricultural soil around riverside Gaughat (GAF) and Municipal waste dump sites Sitapur (SAF) were highly contaminated than other studied sites respectively. The toxic situation of these agricultural soil is evident from the PRI value of two sites, GAF and SAF falling in the "very high risk" and "considerable risk" category respectively whereas 66.66% of the sites are in the "moderate risk" category.

Similarly, PLI also signifies "very high" and "moderately to high" pollution in GAF and SAF sites respectively. However, Igeo value of Mn and Ni showed moderately high whereas Cr, Cu, Zn and Cd depicted low to moderate level contamination due to accumulation of these metals in the agricultural soils. Flourishing soil substrata is vital for the healthy production of food crops; hence continuous monitoring of contaminants at a regular time interval is necessary and it is also important to study the impact of HMs on the microbial community residing in such type of contaminated soil as microbes play nutrient recycling and soil fertility.

Acknowledgement

Dipti is grateful to University Grants Commission (UGC) for a financial support in the form of National Fellowship for Schedule Caste Candidates 2019-20 (NFSC 2019-20).

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(Received 27th September 2022, accepted 31st October 2022)