



# Characterization and ecotoxicological risk assessment of sewage sludge from industrial and non-industrial cities

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## Abstract

The present study highlights the occurrence and the temporal variations of physicochemical properties, and heavy metals in the sludge from sewage treatment plants (STPs) located in industrial (two sites) and non-industrial (one site) cities of Haryana, India. The sludge was acidic (5.59) to neutral (7.21) with a mean EC of 7.4 dS m<sup>-1</sup>. Prominent heavy metals present in the sewage sludge from industrial sites were Cd, Ni, and Cr with maximum values of 2.83, 1449.0, and 3918.5 mg kg<sup>-1</sup>, respectively. The contamination and enrichment factor better explained the buildup of Ni, Cr, and Cu in the sewage sludge from industrial sites. The pH, total carbon, phosphorus, and other water-soluble anions, viz. SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, HCO<sub>3</sub><sup>-</sup>, and PO<sub>4</sub><sup>3-</sup>, were the most important attributes of sludge controlling the binding and removal of the metals with particulate matters during the phase separation in STPs. These attributes explained about 90% of the variation in Cd, Ni, Cr, Cu, Mn, and Zn content of the sludge from different STPs. Sludge from the non-industrial site had a low potential ecological risk index of 74.0 compared to a very high-risk index of 2186.5 associated with the industrial sites. This study concludes that besides the concentration of the heavy metals, the enrichment factor coupled with geo-accumulation or ecological risk index can effectively categorize the sludge. However, these indices need to be linked with bioaccumulation, bioaccessibility, and biomass quality under different agroecologies for guiding the safer use of sewage sludge in agriculture.

**Keywords** Ecotoxicological risk · Sewage sludge · Contamination · Heavy metals · Risk index · Enrichment factor · Heavy metals

## Introduction

The global rise in population has increased the demand for food and simultaneously the generation of urban and industrial waste and wastewater. The direct disposal of wastewater in the rivers and other surface water bodies had led to severe pollution and threat to aquatic and human life (Skelly et al. 2010; Wear et al. 2021). Almost every country worldwide suffers from the contamination of surface water bodies because of the entry of widespread

and excessive untreated waste (Kiulia et al. 2015). The volume of wastewater discharged into the water bodies has crossed the self-purification limit (Saito et al. 2002). Now, wastewater treatment before disposal to water bodies is mandatory to reduce the environmental impact. The ever-increasing wastewater generation and increased capacity of the municipal wastewater treatment facilities (UN Habitat and WHO 2021) have led to the manifold increase in the sewage sludge (SS) generation. These treatment plants produce about 0.2–0.6 kg sludge dry matter per kg of the removal of chemical oxygen demand depending upon the technology used (Ginestet and Camacho 2007; Taşeli 2020). An estimate showed the annual per capita generation of 20–25 kg sludge dry matter in European countries (EUREAU 2021). In China, it was slightly on the higher side (28.1 kg year<sup>-1</sup> person<sup>-1</sup>) (Wei et al. 2020). Appropriate management of waste is important to prevent contamination of natural resources (Fijalkowski et al. 2017; Wei et al.

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2019). Thus, the dramatic increase in sludge generation also needs suitable treatments for safe disposal in a sustained manner. Currently, agricultural use, composting, and landfills are the most practiced method of sludge disposal in many countries (Karagiannidis et al. 2011; Wei et al. 2020; EUREAU 2021). Very little proportion is disposed of by incineration, forestry, and within production site containment (Karagiannidis et al. 2011). Since the last decade, a worldwide attempt has been made to develop a common strategy for waste handling with the objective of reuse for different purposes. Many countries including the European Union had banned the landfill disposal of SS. The landfill sites had a major challenge to environmental safety because of the leaching of metals from SS which may contaminate the soil, groundwater, and surface water bodies (Fang et al. 2016; Li et al. 2017). Land application for agricultural practices is now the most preferred option in many countries because of the presence of organic matter and plant nutrients, viz. nitrogen, phosphorus, and other micronutrients in plenty (de Barros et al. 2021).

India had the potential to generate about 39.6 million tons SS annually on a dry weight basis upon complete treatment of wastewater generated (Singh et al. 2020). At present, only 26.6% of the household wastewater is safely treated in India (UN Habitat and WHO 2021). The estimated SS production is expected to increase gradually with population growth, rapid urbanization, and continuous improvement in sewage drainage systems and wastewater treatment facilities. The treated wastewater is generally discharged into the water bodies spread across the country. The Government of India has mandated the discharge of only properly treated domestic as well as industrial wastewater into the flowing rivers/canal following the guidelines of the Central Pollution Control Board (CPCB). In Haryana state, major cities are located along the Western Yamuna Canal (WYC) and its one branch also connects to the national capital territory Delhi in a later course and finally drained out into the Yamuna River. Most of the municipal wastewater from these cities and industrial discharges from the paper mill, sugar mill, distilleries, starch mill, textile industries, and metal industries located along WYC are let out into this canal. The available reports highlight the deterioration of water quality of WYC because of the mixing of industrial wastewater with domestic discharges (Sakhuja and Sharma 2016). The SS produced by sewage treatment plants (STP) in these cities may also contain a high amount of toxic heavy metals (Dai et al. 2006; Verma et al. 2021). During wastewater treatment, the metals accumulate in the sludge because of sorption, inner- and outer-sphere complexation, and precipitation reactions (Suanon et al. 2017). The composition of SS may vary with season, rainfall, the status of the segregation of effluents at the source, and technology used in STPs (Suanon et al. 2017; Sundha et al. 2021). Reports showed spatio-temporal variation in

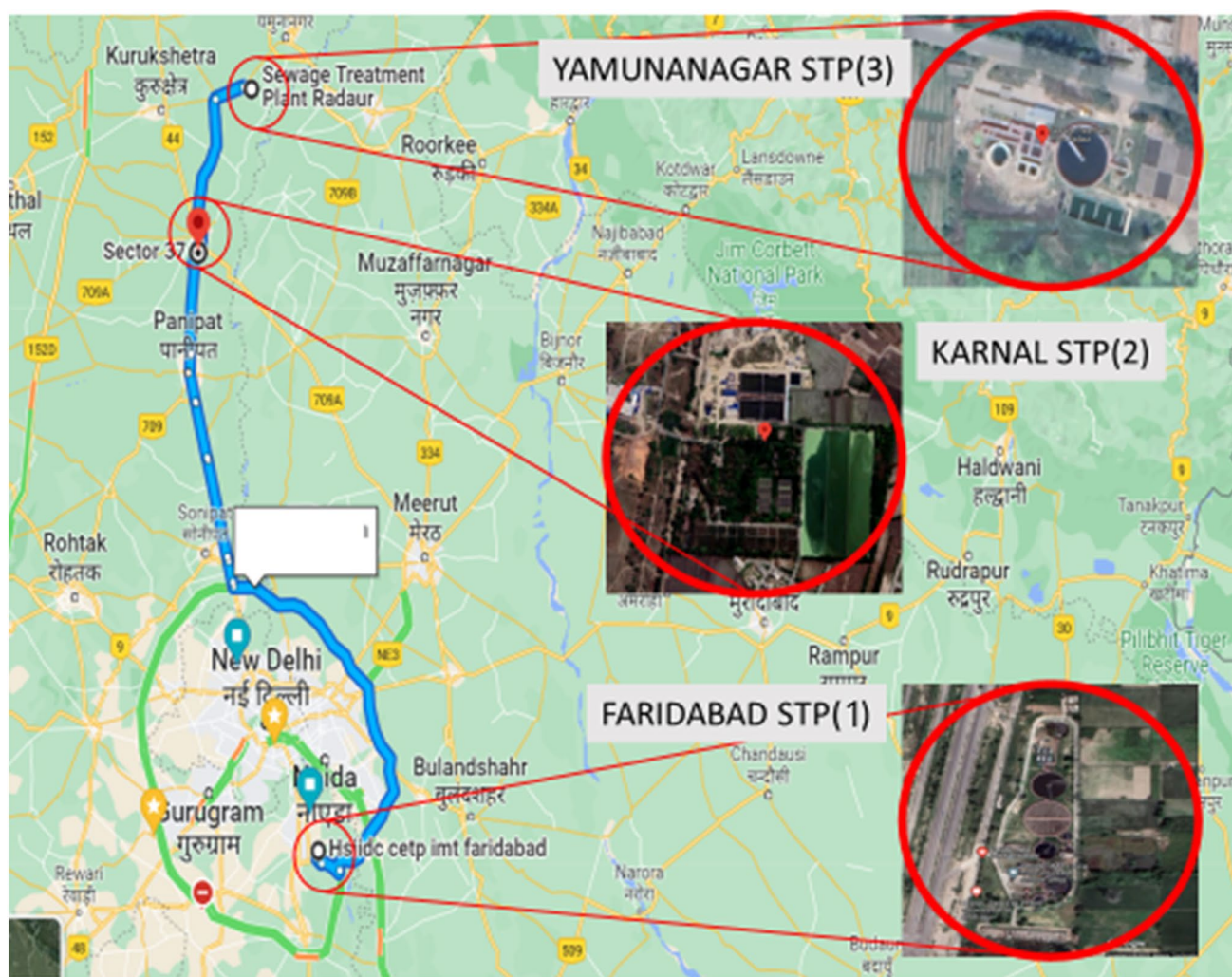
nutritional elements and the presence of hazardous metals in SS (Suanon et al. 2017; Chu and He 2021). STPs in industries-dominated cities generally show the presence of heavy metals (Islam et al. 2017). Appropriate disposal of this huge quantity of SS is a major challenge in India as well as other populous countries (Singh et al. 2020). Although soil application for agricultural usage is considered a better option with relatively lower environmental risk compared to other disposal techniques (Collivignarelli et al. 2020; Campo et al. 2021; EUROSTAT 2021), reduced availability of land, climate, cost of application, the nutrient value of sludge, and acceptability of the produce from SS treated soils are the important consideration in practicing soil application of SS for agricultural production (Foladori et al. 2010; Collivignarelli et al. 2020).

The increased concerns for the safety of the food system also necessitates the prior evaluation of associated risk with soil application of SS. Available reports showed the build-up of toxic metals because of SS application, which subsequently transfers into the food chain (Al-Najar et al. 2005; Bouriouq et al. 2015; Obiora et al. 2019; Olowoyo and Mugivhisa 2019). Contrarily, many studies report the counter view with the application of SS (Karami et al. 2009; Tesfahun et al. 2021). The extent of food chain contamination is reported to vary with the nature and composition of the SS, source characteristics, its application rate, soil properties, crop species, and management practices (Nag et al. 2022). Therefore, source-dependent variation in the composition of SS and associated ecotoxicological risk in their agricultural usage need to be assessed to develop an effective utilization strategy. Pollution indices, associated with SS of different origins, can serve as effective tools in assessing the environmental risk related to their utilization (Kowalska et al. 2016; Mazurek et al. 2017). These indices do offer an opportunity to predict the environmental risk and soil degradation caused by successive applications of SS, because of metal accumulation or leaching to the deeper depths (Caeiro et al. 2005). These indices can be applied to better understand the overall ecological impact of SS application in soils. Therefore, the present study was planned to (1) study the temporal occurrence and concentration of heavy metals in SS from industrial and non-industrial cities and (2) assess the ecological risks associated with the usage of SS.

## Materials and methods

### Sewage sludge sampling and site description

The sewage sludge samples were collected during summer, monsoon, and winter seasons from industrial (Faridabad and Yamunanagar) and non-industrial (Karnal) cities in Haryana located along the Western Yamuna Canal (Fig. 1).



**Fig. 1** Location map of studied sewage treatment plants (STP)

The municipal wastewater of these cities is treated in the sewage treatment plants (STPs) before being discharge into the Yamuna Canal. All the STPs carry out a pre-treatment of the SS to remove coarse materials, sand, and grits. The details of the methods used in wastewater treatment plants of different cities are mentioned in Table S1. SS samples were oven-dried (65 °C) and ground to pass through a 2-mm sieve to maintain the homogeneity of the samples for further analysis.

### Physicochemical analysis

The 1:5 (w/v) aqueous suspension of oven-dried sample was used to determine the pH and electrical conductivity (EC) using a digital pH meter (Systronics <sup>h</sup>pH system 362) and EC meter (Systronics conductivity meter 306<sup>mc</sup>), respectively (Page et al. 1985). Total C and N content was determined by a CHNS analyzer (Elementar Vario EL III,

Germany). Calcium carbonate in the sludge was determined by neutralization with HCl (Allison and Moodie 1965). For total elemental composition, diacid digestion (9 mL of HNO<sub>3</sub>; 4 mL of HClO<sub>4</sub>) was carried out followed by ICP-OES used for detection of the elemental composition of SS (ICPE-9000, Shimadzu), while total phosphorus in the acid digest was determined spectrophotometrically (UV–Vis spectrophotometer, Analytik Jena) using the ascorbic acid reduction method as described by Murphy and Riley (1962). The water-extractable elements were estimated in 1:5:: SS: water extract obtained after shaking at 120 RPM for 1 h. Flame photometer (Flame photometer 128<sup>mc</sup> Systronics) and atomic absorption spectrophotometer (Spectrum instruments Z–xpress–8000) were used for the estimation of Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>, respectively. The CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, and Cl<sup>-</sup> concentrations were estimated by methyl red, phenolphthalein, and argentometric titration (Jackson 1967). SO<sub>4</sub><sup>2-</sup> was estimated using

Nephelometer (SI98713; Hanna, Romania) as described by Chesnin and Yien (1951).

## Pollution indices

### Contamination factor ( $C_f$ )

The  $C_f$  was calculated to evaluate the pollution level associated with the individual element (Table S2). It was calculated using Eq. 1 (Taylor and McLennan 1995; Maanan et al. 2015).

$$Cf_x = \frac{[X]_{\text{sludge}}}{[X]_{\text{crust}}} \quad (1)$$

where  $[X]_{\text{sludge}}$  and  $[X]_{\text{crust}}$  are the concentration of the element in the SS and earth crust, respectively. The values of different elements in the earth's crust reported by Taylor and McLennan (1995) were used for the calculation of  $C_f$ .

### Enrichment factor ( $E_f$ )

The  $E_f$  was calculated with reference to the concentration of Fe used for geochemical normalization using Eq. 2 (Rudnick and Gao 2003; Taylor 1964).

$$E_f = \frac{(C_i/C_{\text{Fe}})_{\text{sludge}}}{(C_i/C_{\text{Fe}})_{\text{reference}}} \quad (2)$$

where  $C_i$  and  $C_{\text{Fe}}$  are the concentration of an element and Fe at each sampling point, respectively, in sludge and reference (earth crust). Categories of enrichment factor ( $E_f$ ) are given in Table S3.

### Pollution loading index (PLI)

The PLI was used to estimate the pollution level of multiple contaminants using Eq. 3 (Håkanson 1980) with respect to the product of  $n$ th root of the contamination factor ( $C_{fx}$ ) of contaminants. The categories of PLI are given in Table S4.

$$PLI = \left( \prod_{x=1}^n Cf_x \right)^{1/n} \quad (3)$$

where  $C_{fx}$  is the contamination factor of element  $x$ , and  $n$  is the number of elements.

### Polymetallic contamination index (IPD)

This index evaluates the level of pollution based on the sum of the contamination factors (Luo et al. 2007). The categories of IPD are given in Table S5.

$$\sum_{x=1}^n Cf_x \quad (4)$$

### Geoaccumulation index ( $I_{geo}$ )

The geoaccumulation index was computed with respect to concentration in the earth crust using Eq. 5 (Müller 1979).

$$I_{geo} = \log_2 \left[ \frac{C_n}{1.5B_n} \right] \quad (5)$$

where  $C_n$  and  $B_n$  represent the concentration of elements in the sludge and earth crust respectively; factor 1.5 used to compensate the possible variations which may be attributed to lithologic variations in earth crust. The categories of geoaccumulation index are given in Table S6.

### Ecological risk index ( $E_i$ ) and potential ecological risk (PERI)

The  $E_i$  and PERI associated with Cd, Cr, Ni, Cu, Mn, and Zn were calculated to assess the toxicity risk of the pollutants and their overall ecological sensitivity using the Eqs. 6 and 7, respectively (Håkanson 1980; Singh et al. 2010; Douay et al. 2013).  $E_i$  and PERI were evaluated for six heavy metals having great environmental concern.  $E_i$  and PERI were calculated using the following equations:

$$Ei_x = Tr_x \times Cf_x \quad (6)$$

where  $Tr_x$  and  $Cf_x$  are the toxic response factor and contamination factor for element  $x$ .

$$PERI = \sum_{x=1}^n Ei_x \quad (7)$$

where  $Tr_x$  and  $Cf_x$  are the toxic response factor ( $Tr$  for Cd, Cr, Cu, Mn, Ni, and Zn are 30, 2, 5, 1, 6, and 1, respectively) and contamination factors of element  $x$ , respectively. Categories of  $E_i$  and PERI are given in Table S7 and S8, respectively.

### Statistical analyses

Data generated from the experiments were analyzed with SPSS. The Kruskal–Wallis test was performed for the analysis of variance. All pair-wise comparisons were made using  $P$  values ( $P < 0.05$ ) adjusted by the Bonferroni correction for multiple tests. The relationship of physicochemical parameters and water-soluble constituents of sewage sludge on the concentration of heavy metals of environmental concern was evaluated using redundancy analysis (RDA) using R (V4.0.4). The significance of sludge parameters affecting the binding of the heavy metals with the particulate matter was evaluated using the Monte Carlo permutation test ( $P < 0.05$ ). A Pearson correlation matrix was also developed for different response variables.

## Results

### Physicochemical characterization of the sludge

The pH<sub>1:5</sub> of the sewage sludge (SS) of STP1 was lower in the summer season compared to monsoon and winter (Table 1). The SS of STP2 and STP3 had acidic pH<sub>1:5</sub> (5.36–6.81). The electrical conductivity (EC<sub>1:5</sub>) of STP1 was higher compared to STP2 and STP3. The EC<sub>1:5</sub> of the STP1, STP2, and STP3 was higher in the monsoon, winter, and summer seasons, respectively ( $P < 0.05$ ). Total carbon (TC) and N (TN) content ranged from 5.9–23.8% and 0.57–1.90%, respectively. The TC and TN content in STP2 and STP3 were similar ( $P > 0.05$ ) but higher than the STP1 ( $P < 0.05$ ). Total phosphorus (TP) was higher in SS collected from STP3 (3918.9 mg kg<sup>-1</sup>). The TC, TN, and TP content were decreased in monsoon compared to the summer and winter seasons. The CaCO<sub>3</sub> content in all the seasons was higher in STP1 followed by STP2 and STP3. Seasonal variation in the CaCO<sub>3</sub>

content was uniform on all sites and sludge collected in monsoon seasons showed a lower value of CaCO<sub>3</sub> compared to other seasons. Except for EC<sub>1:5</sub>, all other parameters were better compared to the FCO (1985) standard prescribed for organic amendments for agricultural applications. Except for a few observations in the monsoon season, the mean TC, TN, and TP content of SS from all sites were higher than the C, N, and P found in farmyard manure. The cations (Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>) and anions (HCO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, and SO<sub>4</sub><sup>2-</sup>) in the 1:5::SS: water extract were higher in STP1 (Table 2). All the water-soluble elements in SS of STP1 and STP2 were higher in summer and winter compared to the monsoon season ( $P < 0.05$ ), while in STP1, the Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, and Cl<sup>-</sup> content were higher in monsoon compared to other seasons. The water-extractable P (PO<sub>4</sub><sup>3-</sup>) was maximum in STP2 followed by STP1 and STP3. The PO<sub>4</sub><sup>3-</sup> content was higher in STP2 and STP3, but lower in STP1 in the monsoon season compared to the summer and winter seasons.

**Table 1** Physicochemical characterization of sewage sludge collected from different sites

Sites	Season	pH <sub>1:5</sub>	Electrical conductivity <sub>1:5</sub> (dS m <sup>-1</sup> )	Total C (%)	Total N (%)	Total P (mg kg <sup>-1</sup> )	CaCO <sub>3</sub> (%)
Faridabad (STP1)	Summer	6.88 <sup>bA</sup>	8.26 <sup>bA</sup>	18.8 <sup>aB</sup>	1.17 <sup>abB</sup>	2810.6 <sup>B</sup>	5.55 <sup>aA</sup>
	Monsoon	7.66 <sup>aA</sup>	19.56 <sup>aA</sup>	5.9 <sup>bB</sup>	0.57 <sup>bB</sup>	2601.7 <sup>A</sup>	4.68 <sup>bA</sup>
	Winter	7.11 <sup>abA</sup>	7.91 <sup>bA</sup>	17.7 <sup>aB</sup>	1.30 <sup>aB</sup>	2583.6 <sup>B</sup>	5.10 <sup>aA</sup>
	Mean	7.21 <sup>P</sup>	11.91 <sup>P</sup>	14.25 <sup>Q</sup>	1.01 <sup>Q</sup>	2665.3 <sup>Q</sup>	5.11 <sup>P</sup>
Karnal (STP2)	Summer	6.06 <sup>bAB</sup>	4.79 <sup>bB</sup>	22.51 <sup>aA</sup>	1.51 <sup>AB</sup>	4501.5 <sup>aA</sup>	2.96 <sup>aB</sup>
	Monsoon	6.82 <sup>aAB</sup>	4.98 <sup>bB</sup>	18.93 <sup>bA</sup>	1.33 <sup>A</sup>	1640.5 <sup>bB</sup>	2.38 <sup>bB</sup>
	Winter	6.17 <sup>bAB</sup>	6.13 <sup>aAB</sup>	22.94 <sup>aA</sup>	1.55 <sup>AB</sup>	2871.6 <sup>abB</sup>	3.34 <sup>aAB</sup>
	Mean	6.35 <sup>Q</sup>	5.30 <sup>Q</sup>	21.46 <sup>P</sup>	1.46 <sup>P</sup>	3004.5 <sup>Q</sup>	2.89 <sup>Q</sup>
Yamunanagar (STP3)	Summer	5.46 <sup>bB</sup>	8.64 <sup>aA</sup>	23.79 <sup>A</sup>	1.90 <sup>aA</sup>	1.36 <sup>B</sup>	1.36 <sup>B</sup>
	Monsoon	5.94 <sup>ab</sup>	2.60 <sup>bB</sup>	16.21 <sup>A</sup>	1.22 <sup>bA</sup>	0.94 <sup>B</sup>	0.94 <sup>B</sup>
	Winter	5.36 <sup>bB</sup>	3.52 <sup>bB</sup>	22.97 <sup>A</sup>	1.87 <sup>aA</sup>	1.28 <sup>B</sup>	1.28 <sup>B</sup>
	Mean	5.59 <sup>r</sup>	4.92 <sup>Q</sup>	20.99 <sup>P</sup>	1.66 <sup>P</sup>	3918.9 <sup>P</sup>	1.19 <sup>r</sup>
Summer		6.13	7.23	21.71 <sup>P</sup>	1.53 <sup>P</sup>	3914.3 <sup>P</sup>	3.29
Monsoon		6.81	9.05	13.68 <sup>Q</sup>	1.04 <sup>Q</sup>	2208.5 <sup>Q</sup>	2.66
Winter		6.21	5.85	21.21 <sup>P</sup>	1.57 <sup>P</sup>	3465.9 <sup>P</sup>	3.24
FCO 1985 Standards <sup>#</sup>		6.5–7.5	< 4.0	> 16.0	> 0.5	-	-
Mean concentration in FYM (Sundha et al. 2022)		8.69 ± 0.03	9.86 ± 0.70	10.58 ± 0.04	1.02 ± 0.05	3500.0 ± 0.02 (unpublished)	2.48 ± 0.6 (unpublished)

Values with different uppercase letters (A–B) in columns are significantly different ( $P < 0.05$ ) for the season among different sites; values with different lowercase letters (a–b) in columns are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) in columns are significantly different ( $P < 0.05$ ) for comparison of the site across the seasons; values with different uppercase letters (P–Q) in columns are significantly different ( $P < 0.05$ ) for comparison of the season across the sites

<sup>#</sup><https://companydemo.in/apps/nocf/uploads/pdf/FCOpdf-a34603742462f8064bc7b534fd433db2.pdf>

**Table 2** Water-soluble cations-anions ( $\text{mg kg}^{-1}$ ) in sewage sludge collected from different sites

Site	Season	$\text{Na}^+$	$\text{K}^+$	$\text{SO}_4^{2-}$	$\text{Ca}^{2+}$	$\text{Mg}^{2+}$	$\text{HCO}_3^-$	$\text{Cl}^-$	$\text{PO}_4^{3-}$
Faridabad (STP1)	Summer	4.14 <sup>ba</sup>	0.43 <sup>ba</sup>	0.23 <sup>aa</sup>	2.55 <sup>abA</sup>	1.33 <sup>aA</sup>	0.72 <sup>aA</sup>	861.3 <sup>ba</sup>	1.81 <sup>aA</sup>
	Monsoon	8.16 <sup>aa</sup>	7.12 <sup>aa</sup>	0.20 <sup>ba</sup>	2.98 <sup>aA</sup>	1.02 <sup>ba</sup>	0.38 <sup>bbB</sup>	2398.9 <sup>aa</sup>	0.23 <sup>bbB</sup>
	Winter	4.36 <sup>ba</sup>	0.32 <sup>ba</sup>	0.21 <sup>ba</sup>	2.34 <sup>bbB</sup>	1.42 <sup>aA</sup>	0.51 <sup>a</sup>	822.7 <sup>ba</sup>	1.07 <sup>aA</sup>
	Mean	5.56 <sup>p</sup>	2.63 <sup>p</sup>	0.21 <sup>p</sup>	2.62 <sup>p</sup>	1.26 <sup>p</sup>	0.54 <sup>p</sup>	1361.0 <sup>p</sup>	1.04 <sup>q</sup>
Karnal (STP2)	Summer	0.18 <sup>abB</sup>	0.10 <sup>aB</sup>	0.08 <sup>aB</sup>	1.69 <sup>aB</sup>	0.53 <sup>aB</sup>	0.50 <sup>B</sup>	130.5 <sup>B</sup>	1.86 <sup>ba</sup>
	Monsoon	0.09 <sup>bbB</sup>	0.04 <sup>bbB</sup>	0.01 <sup>bbB</sup>	0.36 <sup>bbB</sup>	0.26 <sup>bbB</sup>	0.63 <sup>A</sup>	127.8 <sup>B</sup>	2.57 <sup>aa</sup>
	Winter	0.26 <sup>aB</sup>	0.09 <sup>aB</sup>	0.12 <sup>aB</sup>	2.38 <sup>aB</sup>	0.60 <sup>aB</sup>	0.47	121.1 <sup>B</sup>	1.43 <sup>ba</sup>
	Mean	0.17 <sup>q</sup>	0.08 <sup>q</sup>	0.07 <sup>q</sup>	1.47 <sup>q</sup>	0.46 <sup>r</sup>	0.53 <sup>p</sup>	126.5 <sup>q</sup>	1.95 <sup>p</sup>
Yamunanagar (STP3)	Summer	0.13 <sup>bbB</sup>	0.08 <sup>abB</sup>	0.11 <sup>aB</sup>	2.66 <sup>aA</sup>	0.87 <sup>B</sup>	0.27 <sup>B</sup>	101.2 <sup>bbB</sup>	0.15 <sup>B</sup>
	Monsoon	0.18 <sup>bbB</sup>	0.07 <sup>bbB</sup>	0.08 <sup>bbB</sup>	1.82 <sup>ba</sup>	0.54 <sup>AB</sup>	0.31 <sup>B</sup>	114.5 <sup>bbB</sup>	0.33 <sup>B</sup>
	Winter	0.25 <sup>aB</sup>	0.10 <sup>aB</sup>	0.11 <sup>aB</sup>	2.83 <sup>aA</sup>	0.74 <sup>B</sup>	0.29	142.4 <sup>aB</sup>	0.17 <sup>B</sup>
	Mean	0.19 <sup>q</sup>	0.08 <sup>q</sup>	0.10 <sup>q</sup>	2.44 <sup>p</sup>	0.72 <sup>q</sup>	0.29 <sup>q</sup>	119.4 <sup>q</sup>	0.22 <sup>r</sup>
Summer		1.48	0.21	0.14 <sup>P</sup>	2.30	0.91 <sup>P</sup>	0.50	364.3	1.27
Monsoon		2.81	2.41	0.10 <sup>Q</sup>	1.72	0.61 <sup>Q</sup>	0.44	880.4	1.04
Winter		1.62	0.17	0.14 <sup>P</sup>	2.52	0.92 <sup>P</sup>	0.42	362.1	0.89

Values with different uppercase letters (A–B) in columns are significantly different ( $P < 0.05$ ) for the season among different sites; values with different lowercase letters (a–b) in columns are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) in columns are significantly different ( $P < 0.05$ ) for comparison of the site across the seasons; values with different uppercase letters (P–Q) in columns are significantly different ( $P < 0.05$ ) for comparison of the season across the sites

## Metals in sewage sludge

The SS of the STP3 contained a higher concentration of Ni, Cr, Mn, Cu, Zn, and Fe compared to STP1 and STP2 in all the seasons ( $P < 0.05$ ; Table 3). The metal content observed in STP3 was much higher than the values estimated in manure and the values permitted in organics. The STP1 recorded the presence of Cd in SS only in the summer season, whereas STP3 had a notable amount of Cd in all the seasons. The Cr was high in all the samples collected from three STPs compared to permissible limits of FCO standards for agricultural usage. Except for monsoon and winter season, SS from the STP1, all samples from three STPs recorded Ni content higher than the permissible limits. The Cu was lower in summer for STP1 and in monsoon for STP2. Zn was higher in SS collected in the summer season for all the STPs. Fe was higher in summer and lower in the winter season for STP1; contrarily, STP2 and STP3 recorded high Fe content in monsoon. The Ca and Mg content were similar in the SS from STP1 and STP2 ( $P > 0.05$ ) but higher than STP3 ( $P < 0.05$ ), while within sites, the contents of Ca and Mg were higher in monsoon for STP1 and their concentration was higher in winters for STP3.

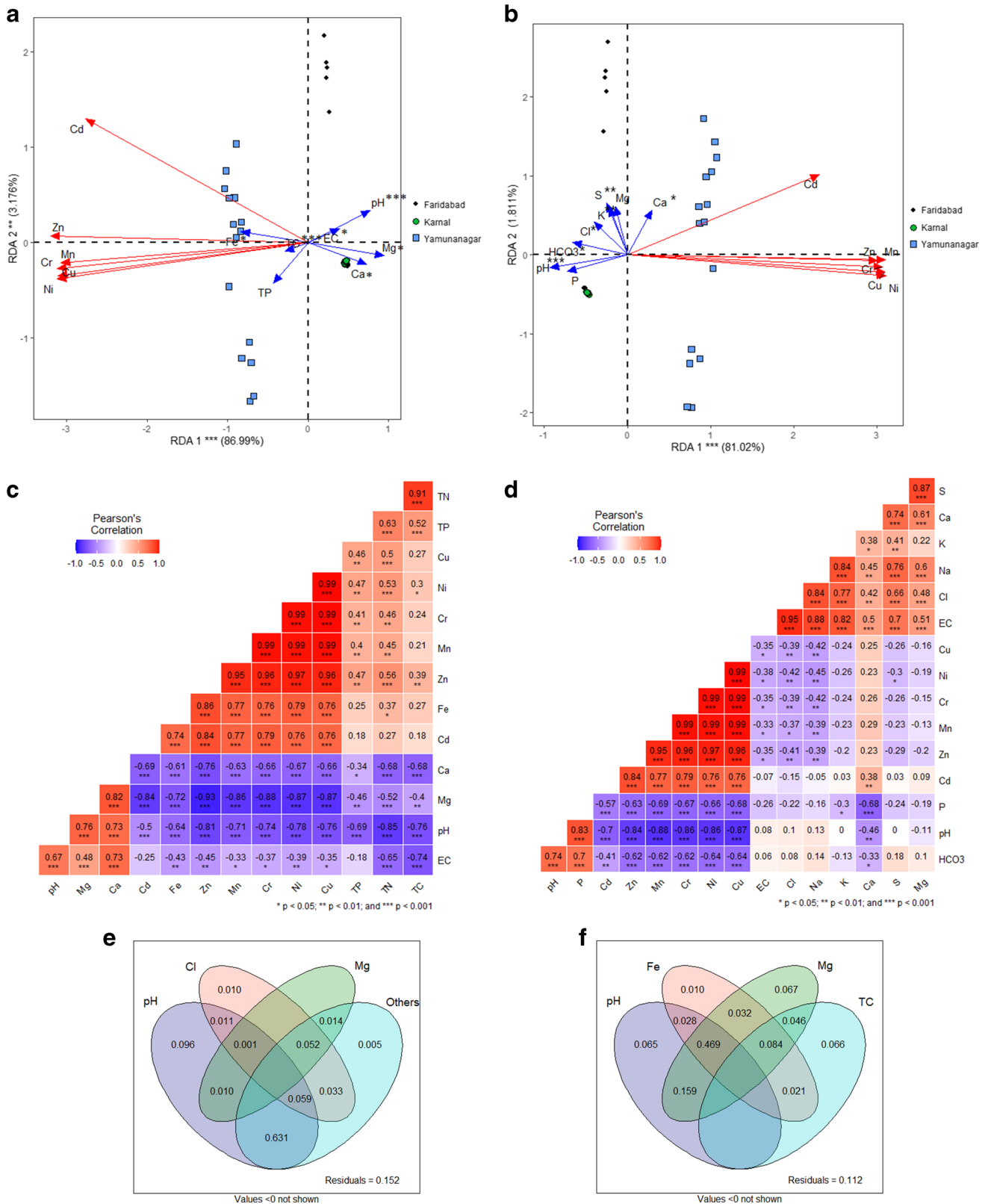
## Influence of physicochemical characteristics on heavy metals content

The redundancy analysis showed the influence of sludge characteristics, viz. pH, EC, TC, TP, Ca, and Mg on the concentration of different heavy metals (Cd, Ni, Cr, Cu, Mn, and Zn) (Fig. 2). Total contents were better in explaining the variability in heavy metals associated with sludges compared to water-soluble constituents (Fig. 2a and b). The pH, TC, TP, Ca, and Mg explained ~90% of the total variability of heavy elements (Cd, Ni, Cr, Cu, Mn, and Zn) in sludge. pH, EC, TC, Ca, and Mg were having a significant conditional effect on the heavy metal content of the sludge (Fig. 2a). The TC, Fe, and TP were positively correlated, while pH, EC, Ca, and Mg showed a strong negative correlation with heavy metals of SS (Fig. 2c). Among water-soluble constituents,  $\text{Ca}^{2+}$  had a positive, while  $\text{Na}^+$ , TP,  $\text{K}^+$ ,  $\text{Cl}^-$ , and  $\text{HCO}_3^-$  had a strong negative correlation with heavy metals. Water-soluble  $\text{PO}_4^{3-}$  showed a stronger correlation with heavy metals compared to TP (Fig. 2d). The variance partitioning analysis depicted that the sludge pH has a higher effect on the heavy metal content compared to Fe, Mg, and TC (Fig. 2e). The interaction effect of these

**Table 3** Total heavy metals content (mg kg<sup>-1</sup>) in sewage sludge collected from different sites

Site	Season	Cd	Ni	Cr	Mn	Ca	Mg	Cu	Zn	Fe <sup>#</sup>
Faridabad (STP1)	Summer	2.43	75.3 <sup>ab</sup>	167.7 <sup>ab</sup>	203.2 <sup>bb</sup>	15.7 <sup>bb</sup>	2.99 <sup>baB</sup>	192.0 <sup>bb</sup>	1360.9 <sup>aAB</sup>	1.85 <sup>aA</sup>
	Monsoon	nd	20.4 <sup>bb</sup>	92.8 <sup>bb</sup>	218.7 <sup>ab</sup>	44.1 <sup>aA</sup>	5.26 <sup>aA</sup>	219.8 <sup>abB</sup>	374.3 <sup>bb</sup>	1.40 <sup>abB</sup>
	Winter	nd	17.1 <sup>bb</sup>	136.9 <sup>abb</sup>	210.5 <sup>abb</sup>	22.0 <sup>aba</sup>	4.48 <sup>abA</sup>	222.1 <sup>ab</sup>	394.9 <sup>bb</sup>	1.13 <sup>bb</sup>
	Mean	2.43	37.6 <sup>t</sup>	132.5 <sup>q</sup>	210.8 <sup>q</sup>	27.2 <sup>p</sup>	4.25 <sup>p</sup>	211.3 <sup>q</sup>	710.0 <sup>q</sup>	1.46 <sup>q</sup>
Karnal (STP12)	Summer	nd	97.6 <sup>ab</sup>	86.5 <sup>B</sup>	188.3 <sup>abb</sup>	26.3 <sup>aA</sup>	3.84 <sup>bA</sup>	147.5 <sup>bb</sup>	875.5 <sup>ab</sup>	1.46 <sup>bb</sup>
	Monsoon	nd	89.3 <sup>abb</sup>	80.1 <sup>B</sup>	208.4 <sup>ab</sup>	26.4 <sup>aAB</sup>	4.81 <sup>aA</sup>	131.1 <sup>bb</sup>	781.4 <sup>bb</sup>	1.69 <sup>abB</sup>
	Winter	nd	80.8 <sup>bb</sup>	81.8 <sup>B</sup>	172.6 <sup>bb</sup>	23.4 <sup>bA</sup>	4.51 <sup>bA</sup>	182.1 <sup>ab</sup>	786.0 <sup>bb</sup>	1.54 <sup>abAB</sup>
	Mean	nd	89.2 <sup>q</sup>	82.8 <sup>t</sup>	189.8 <sup>t</sup>	25.4 <sup>p</sup>	4.39 <sup>p</sup>	153.6 <sup>t</sup>	814.3 <sup>q</sup>	1.56 <sup>q</sup>
Yamunanagar (STP3)	Summer	3.44 <sup>a</sup>	1416.6 <sup>A</sup>	3864.7 <sup>bA</sup>	652.6 <sup>A</sup>	13.1 <sup>B</sup>	1.49 <sup>ab</sup>	3082.8 <sup>A</sup>	2913.0 <sup>A</sup>	1.95 <sup>A</sup>
	Monsoon	3.66 <sup>a</sup>	1459.9 <sup>A</sup>	4345.6 <sup>aA</sup>	689.4 <sup>A</sup>	11.7 <sup>B</sup>	1.19 <sup>bb</sup>	3125.3 <sup>A</sup>	2865.9 <sup>A</sup>	2.16 <sup>A</sup>
	Winter	1.38 <sup>b</sup>	1470.6 <sup>A</sup>	3545.3 <sup>bA</sup>	628.3 <sup>A</sup>	12.0 <sup>B</sup>	1.67 <sup>ab</sup>	3113.8 <sup>A</sup>	2785.9 <sup>A</sup>	2.12 <sup>A</sup>
	Mean	2.83	1449.0 <sup>p</sup>	3918.5 <sup>p</sup>	656.8 <sup>p</sup>	12.3 <sup>q</sup>	1.45 <sup>q</sup>	3107.3 <sup>p</sup>	2854.9 <sup>p</sup>	2.08 <sup>p</sup>
Summer	Summer	2.93 <sup>p</sup>	529.8 <sup>p</sup>	1373.0 <sup>p</sup>	348.0	18.4 <sup>p</sup>	2.77 <sup>p</sup>	1140.8 <sup>p</sup>	1716.5 <sup>p</sup>	1.75 <sup>p</sup>
	Monsoon	3.66 <sup>p</sup>	523.2 <sup>p</sup>	1506.2 <sup>p</sup>	372.2 <sup>p</sup>	27.4 <sup>p</sup>	3.76 <sup>p</sup>	1158.7 <sup>p</sup>	1340.5 <sup>q</sup>	1.75 <sup>p</sup>
	Winter	1.38 <sup>q</sup>	522.8 <sup>p</sup>	1254.7 <sup>p</sup>	337.2 <sup>p</sup>	19.1 <sup>p</sup>	3.55 <sup>p</sup>	1172.7 <sup>p</sup>	1322.3 <sup>q</sup>	1.60 <sup>p</sup>
	FCO 1985 Standards <sup>#</sup>	<5.0	<50.0	<50.0	-	-	-	<300.0	<1000.0	-
Mean concentration in FYM (Sundha et al. 2022)		-	34.2±19.8	-	257.6±82.3	8.83±0.39	86.2±1.50	9.3±5.4	108.9±27.2	1.62±0.0003
Permissible total content in SS for 30.0 soil application for grain crop (Delibacak et al. 2020)		-	400.0	1000.0	-	-	-	1500.0	3500.0	-

<sup>#</sup>The unit of Fe is percent (%); values with different uppercase letters (A–B) in columns are significantly different ( $P < 0.05$ ) for the season among different sites; values with different lowercase letters (a–b) in columns are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–t) in columns are significantly different ( $P < 0.05$ ) for comparison of the site across the seasons; values with different uppercase letters (P–Q) in columns are significantly different ( $P < 0.05$ ) for comparison of the season across the sites; *nd*, below the detection limit (1.25 mg kg<sup>-1</sup>)



parameters had a higher effect on the heavy metal content compared to the individual parameters. The interaction effect of the pH and other soluble constituents in sludge

( $K^+$ ,  $SO_4^{2-}$ ,  $Ca^{2+}$ ,  $HCO_3^-$ , and  $PO_4^{3-}$ ) depicted about 63.1% variability in the heavy metal contents in sludge (Fig. 2f).



**Fig. 2** Biplot from redundancy analysis (RDA) showing the influence of physicochemical parameters (a) and water-soluble constituents of sludge (b) on the phasing out of heavy metals in sewage sludge samples; correlations among the physicochemical and total heavy metals content (c); correlations among the water-soluble constituents and total heavy metals content (d); variation partitioning analysis depicting the effect of pH, TC, total Fe, and Mg content in the sludge on phasing out of heavy metals in sludge (e); variation partitioning analysis depicting the effect of pH,  $\text{Cl}^-$ ,  $\text{Mg}^{2+}$ , and others water-soluble ions ( $\text{K}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{HCO}_3^-$ , and  $\text{PO}_4^{3-}$ ) on phasing out of heavy metals in sludge (f)

### Contamination factor ( $C_f$ )

The  $C_f$  was very high for Cd in SS, during summer (24.8) in STP1 and all seasons (14.1–37.3) for STP3 (Fig. 3; Table S9). The STP3 recorded very high  $C_f$  values for Ni, Cr, Cu, and Zn in all the seasons. The  $C_f$  for Zn and Cu was considerable to very high, while Cr was moderate to considerable in STP1, and STP2. The  $C_f$  for Ni was lower for STP1 in monsoon and winter seasons, whereas higher in summer. STP2 showed a considerable contamination factor of Ni. Overall, STP1 and STP2 showed a low to moderate degree of contamination for all the elements. The contamination factor was low ( $C_f < 1$ ) for Ca in all the STPs in all the seasons except in the monsoon season in STP1 (1.47).

### Enrichment factor ( $E_f$ )

The enrichment factor ( $E_f$ ) for Cd in SS collected from STP3 was very severe to extremely severe (Fig. 4; Table S10).  $E_f$  for Cd was absent in STP2 and STP1 (except summer season). Summer season SS at STP1 showed very severe  $E_f$  for Cd. The  $E_f$  for other elements like Ni, Cr, Cu, and Zn were extremely severe for SS from STP3. The  $E_f$  for Cu and Zn were also severe to very severe in SS from STP1 and STP2. Except for Fe and Mg, the  $E_f$  for all the elements was higher in STP3 in all three seasons. Except for Cu and Cr in STP1, within sites, all the elements showed higher  $E_f$  in the summer season. Severe enrichment of Cr (12.0) was noticed in winters for STP1 and moderate to severe for STP2 (4.85–5.9) and highly severe for STP3 (167.2–202.1). There was minor or no enrichment for Mn at all the sites. Ca and Mg showed no enrichment.

### Geoaccumulation index ( $I_{geo}$ )

A high to extremely high contamination of Cd was noticed in SS collected in all the seasons from STP3 and the summer season from STP1 (Table 4). The Mn, Fe, Ca, and Mg did not show any contamination in the SS samples ( $< 1$ ) from any of the STPs. Contamination of Cr, Cu, and Zn in STP1 and Ni, Cr, Cu, and Zn in STP2 was within moderate

to high levels, while in STP3 the Cd, Ni, Cr, Zn, and Cu contamination was in the range of high to the extreme level. STP3 showed relatively higher contamination in all seasons compared to other sites and the seasonal effect was apparent only for Cr showed relatively lower contamination in winter. At STP1 and STP2, summer season recorded higher contamination of Ni, Cr, and Zn.

### Pollution loading index (PLI) and polymetallic contamination index (IPD)

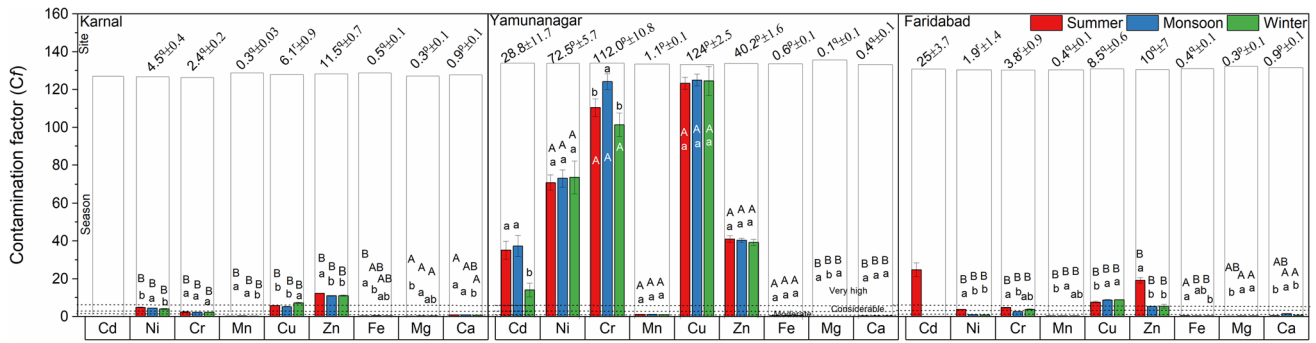
The value of the pollution loading index was moderate for SS collected from STP1 (1.20–2.23) and STP2 (1.50–1.54), whereas the pollution index was very high for STP3 (6.30–7.01) in all the seasons (Fig. 5). The PLI index was higher in summer compared to monsoon and winter seasons. The polymetallic contamination index (IPD) was considered too high for STP1 and STP2 (Fig. 6). This index was very high for STP3 (354.9–402.1) in all the seasons.

### Ecological risk index and potential ecological risk

A higher ecological risk index ( $E_i$ ) of Cd was noticed in SS collected in the all-seasons from STP3 and summer season from STP1 (Fig. 7; Table S11). The  $E_i$  of all the elements except Cd (summer season) was within low to the reasonable limit at STP1 and STP2, while in STP3, it was high to very high for Cd, Ni, Cr, and Cu. The ecological risk for Mn and Zn was low for all the STPs. The overall potential ecological risk (PERI) associated with SS was low for STP2 and STP1 (monsoon and winter season). The potential ecological risk was very high for STP3 (1728.7–2358.2). PERI for summer season SS was higher than the monsoon season in STP1 and STP2; it was considerably low in the winter season compared to summer and monsoon season in STP3 (Fig. 8).

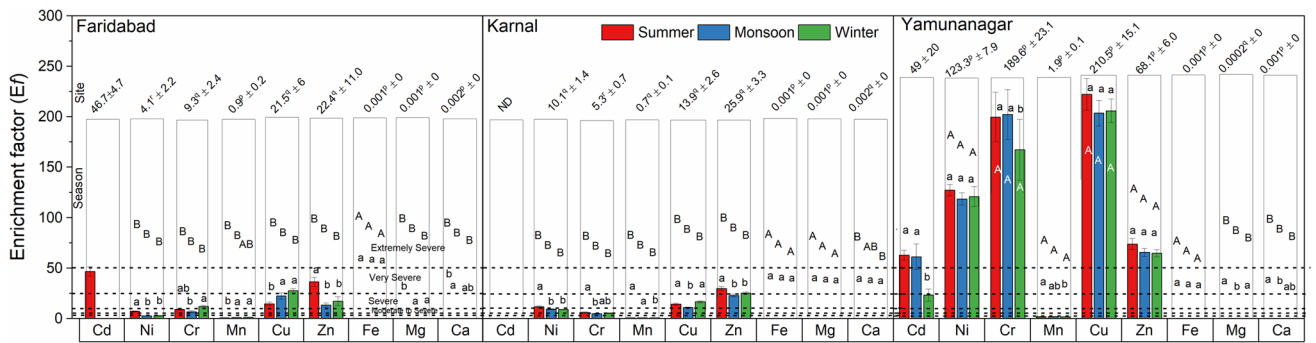
### Discussion

Worldwide a safe disposal of the ever-increasing SS is the major concern of urban settlements. After the ban on landfill disposal in many countries, land application for agricultural usage is the major pathway for safe disposal (Eurostat 2021; Eureau 2021). STPs needs to adopt strict operational standards for quality assurance of the SS intended for agricultural usage to address the increased concern for the safety of the food chain. Metals are the important group of pollutants likely to be present in SS. Accidental mixing of the stormwater and industrial and domestic effluents is the major cause of metals in the sludge (Foladori et al. 2010; Ahuja 2014; Kumar et al. 2020). The observed higher values of the heavy



**Fig. 3** The contamination factor ( $C_p$ ) of heavy metals in sewage sludge collected from Faridabad (STP1), Karnal (STP2), and Yamunanagar (STP3); values with different uppercase letters (A–B) on the bars are significantly different ( $P < 0.05$ ) for the seasons among different sites; values with different lowercase letters (a–b) in on the bars

are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) on the bars are significantly different ( $P < 0.05$ ) for comparison of sites across the seasons;  $\pm$  represents standard deviations from mean; whisker on the bars represent the standard deviations ( $\pm$ )



**Fig. 4** The enrichment factor ( $E_f$ ) of heavy metals in sewage sludge collected from Faridabad (STP1), Karnal (STP2), and Yamunanagar (STP3); values with different uppercase letters (A–B) on the bars are significantly different ( $P < 0.05$ ) for the seasons among different sites; values with different lowercase letters (a–b) in on the bars are

significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) on the bars are significantly different ( $P < 0.05$ ) for comparison of sites across the seasons;  $\pm$  represents standard deviations from mean; whisker on the bars represent the standard deviations ( $\pm$ )

metals in SS from the STP3 may be because of the mixing of the industrial effluents in the domestic sewage system. Yamunanagar (STP3) is having a big cluster of food product, leather, wood, and wood product industries besides many metal and mineral products and electrical, electronic, and machinery manufacturing units (Table S12). These industries are commonly using salts of Cu, Cr, Cd, and Ni in various processes. The effluents of these industries might be directly entering the domestic sewerage lines and spiking a very high concentration of these elements in SS generated from STP. Several reports showed the evidence of the release of untreated effluents from these industries into the wastewater of Yamunanagar city (<https://yamuna-revival.nic.in/wp-content/uploads/2021/01/CPCB-NEERI-Report-on-pollution-on-Haryana-drains-received-by-YMC-on-6.2.19.pdf>). Huge enrichment of SS with Cd, Ni, Cr, and Cu from STP3 also indicated the mixing of the effluents from iron and steel electroplating, electronics, electrical, leather, and wood processing industries. Mixing of effluents from food

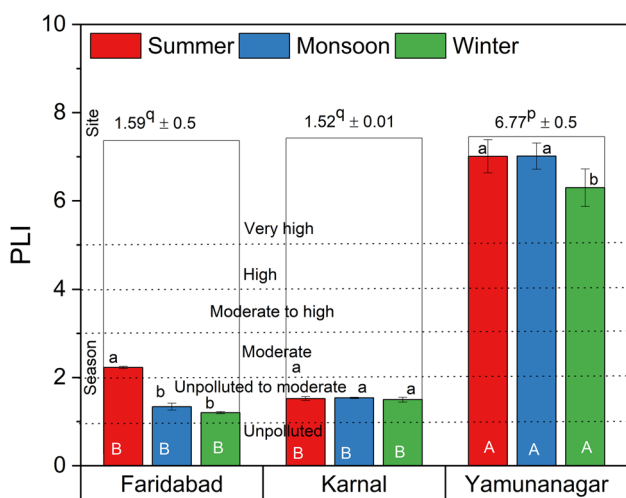
processing industries, leather, and wood processing may be the primary cause of Ni, Cr, and Cu enrichment in SS of the STP3 in Yamunanagar, respectively. Besides manufacturing units, the allied sector dealing with battery reconditioning, electroplating, and paint shops also generates effluents bearing an appreciable amount of metals into sewage (Oghenrobor et al. 2014). Similarly, industrial effluents discharge from the tannery, motor vehicle exhaust, storage batteries, and agrochemicals are the major source of Cd contamination in soils (Farooq et al. 2016; Table S1). Although STP1 situated in Faridabad had a higher number of different industries except for food products and wood processing, the concentration of the studied heavy metals was many times lower than Yamunanagar (STP3). This indicated the regulatory compliance and reduced mixing of the effluents in domestic wastewater reaching the STPs.

Heavy metal distribution in the sludge depends on the metal content, sludge characteristics, wastewater treatment process, amount of organic carbon, humic matter content,

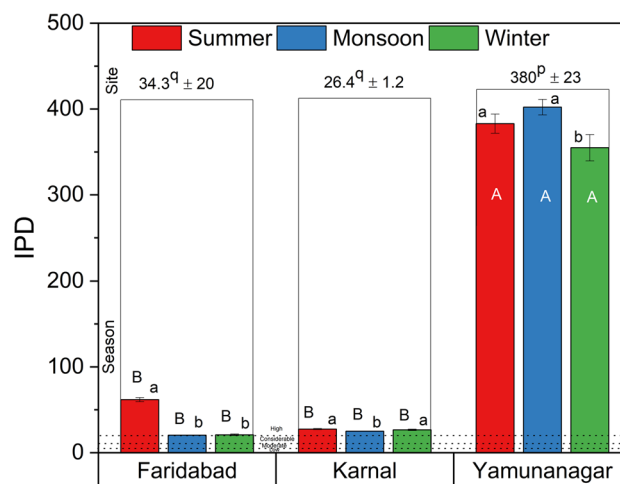
**Table 4** Geoaccumulation index ( $I_{geo}$ ) values of elements in the sludge samples collected from different sites

Site	Season	Cd	Ni	Cr	Mn	Cu	Zn	Fe	Ca	Mg
Faridabad (STP1)	Summer	4.1	1.33 <sup>ab</sup>	1.67 <sup>ab</sup>	-2.15 <sup>baB</sup>	0.62 <sup>bb</sup>	3.67 <sup>ab</sup>	-1.51 <sup>aA</sup>	-1.52 <sup>bb</sup>	-2.74 <sup>ba</sup>
	Monsoon	-	-0.56 <sup>bb</sup>	0.82 <sup>bb</sup>	-2.04 <sup>ab</sup>	0.82 <sup>ab</sup>	1.81 <sup>bb</sup>	-1.92 <sup>abB</sup>	-0.04 <sup>aA</sup>	-1.94 <sup>aA</sup>
	Winter	-	-0.81 <sup>bb</sup>	1.36 <sup>abB</sup>	-2.10 <sup>abB</sup>	0.83 <sup>ab</sup>	1.85 <sup>bb</sup>	-2.22 <sup>bb</sup>	-1.03 <sup>abA</sup>	-2.16 <sup>aA</sup>
	Mean		-0.02 <sup>r</sup>	1.29 <sup>q</sup>	-2.10 <sup>q</sup>	0.76 <sup>q</sup>	2.44 <sup>q</sup>	-1.88 <sup>q</sup>	-0.86 <sup>p</sup>	-2.28 <sup>p</sup>
Karnal (STP2)	Summer	-	1.70 <sup>ab</sup>	0.71 <sup>B</sup>	-2.26 <sup>abA</sup>	0.24 <sup>abB</sup>	3.04 <sup>ab</sup>	-1.85 <sup>bb</sup>	-0.77 <sup>aA</sup>	-2.38 <sup>ba</sup>
	Monsoon	-	1.57 <sup>abB</sup>	0.61 <sup>B</sup>	-2.11 <sup>ab</sup>	0.07 <sup>bb</sup>	2.88 <sup>bb</sup>	-1.64 <sup>abA</sup>	-0.77 <sup>aAB</sup>	-2.05 <sup>aAB</sup>
	Winter	-	1.43 <sup>bb</sup>	0.64 <sup>B</sup>	-2.38 <sup>bb</sup>	0.55 <sup>ab</sup>	2.88 <sup>bb</sup>	-1.77 <sup>abB</sup>	-0.95 <sup>ba</sup>	-2.16 <sup>abA</sup>
	Mean		1.57 <sup>q</sup>	0.65 <sup>r</sup>	-2.25 <sup>q</sup>	0.29 <sup>q</sup>	2.93 <sup>q</sup>	-1.75 <sup>q</sup>	-0.83 <sup>p</sup>	-2.20 <sup>p</sup>
Yamunanagar (STP3)	Summer	4.54 <sup>a</sup>	5.56 <sup>A</sup>	6.20 <sup>abA</sup>	-0.47 <sup>B</sup>	4.63 <sup>A</sup>	4.77 <sup>A</sup>	-1.43 <sup>A</sup>	-1.78 <sup>B</sup>	-3.75 <sup>ab</sup>
	Monsoon	4.62 <sup>a</sup>	5.60 <sup>A</sup>	6.37 <sup>abA</sup>	-0.39 <sup>A</sup>	4.65 <sup>A</sup>	4.75 <sup>A</sup>	-1.29 <sup>A</sup>	-1.96 <sup>B</sup>	-4.08 <sup>bb</sup>
	Winter	3.19 <sup>b</sup>	5.61 <sup>A</sup>	6.08 <sup>ba</sup>	-0.52 <sup>A</sup>	4.64 <sup>A</sup>	4.71 <sup>A</sup>	-1.31 <sup>A</sup>	-1.91 <sup>B</sup>	-3.61 <sup>ab</sup>
	Mean	4.12	5.59 <sup>p</sup>	6.22 <sup>p</sup>	-0.46 <sup>p</sup>	4.64 <sup>p</sup>	4.74 <sup>p</sup>	-1.34 <sup>p</sup>	-1.88 <sup>q</sup>	-3.81 <sup>q</sup>
Summer		4.29 <sup>p</sup>	2.86 <sup>p</sup>	2.86 <sup>p</sup>	-1.62 <sup>p</sup>	1.83 <sup>p</sup>	3.83 <sup>p</sup>	-1.60 <sup>p</sup>	-1.36 <sup>p</sup>	-2.95 <sup>p</sup>
Monsoon		4.62 <sup>p</sup>	2.20 <sup>p</sup>	2.60 <sup>p</sup>	-1.52 <sup>p</sup>	1.85 <sup>p</sup>	3.15 <sup>q</sup>	-1.61 <sup>p</sup>	-0.92 <sup>p</sup>	-2.69 <sup>p</sup>
Winter		3.19 <sup>q</sup>	2.07 <sup>p</sup>	2.69 <sup>p</sup>	-1.67 <sup>p</sup>	2.01 <sup>p</sup>	3.15 <sup>q</sup>	-1.76 <sup>p</sup>	-1.30 <sup>p</sup>	-2.64 <sup>p</sup>

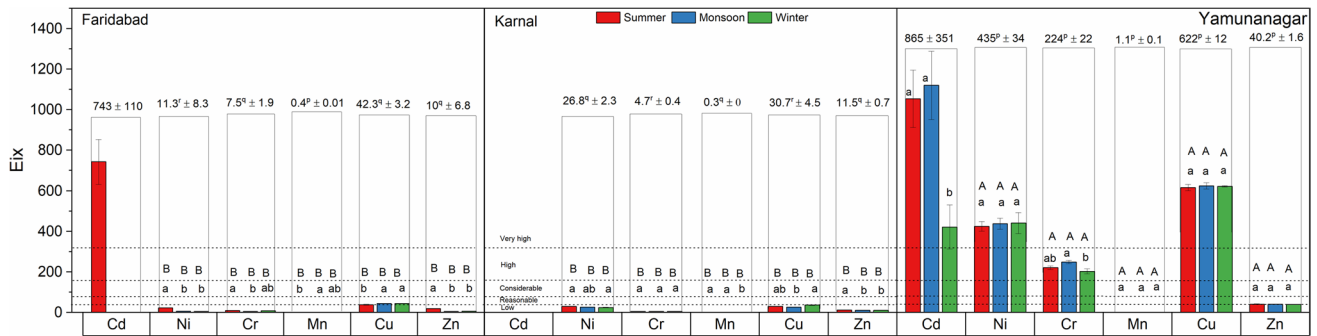
Values with different uppercase letters (A–B) in columns are significantly different ( $P < 0.05$ ) for the season among different sites; values with different lowercase letters (a–b) in columns are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) in columns are significantly different ( $P < 0.05$ ) for comparison of the site across the seasons; values with different uppercase letters (P–Q) in columns are significantly different ( $P < 0.05$ ) for comparison of the season across the sites; “-” represent that Cd was below the detection limit;  $I_{geo}$  categories: uncontaminated ( $\leq 0$ ), uncontaminated to moderate (0–1.0), moderate (1.0–2.0), moderate to high (2.0–3.0), high to extreme (3.0–4.0), highly to extremely high polluted (4.0–5.0) and extreme ( $> 5.0$ )



**Fig. 5** The pollution loading index (PLI) of heavy metals in sewage sludge collected from Faridabad (STP1), Karnal (STP2), and Yamunanagar (STP3); values with different uppercase letters (A–B) on the bars are significantly different ( $P < 0.05$ ) for the season among different sites; values with different lowercase letters (a–b) in on the bars are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) on the bars are significantly different ( $P < 0.05$ ) for comparison of the sites across the seasons; ± represents standard deviations from mean; whisker on the bars represent the standard deviations (±)



**Fig. 6** The polymetallic contamination index (IPD) of heavy metals in sewage sludge collected from Faridabad (STP1), Karnal (STP2), and Yamunanagar (STP3); values with different uppercase letters (A–B) on the bars are significantly different ( $P < 0.05$ ) for the season among different sites; values with different lowercase letters (a–b) in on the bars are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) on the bars are significantly different ( $P < 0.05$ ) for comparison of the sites across the seasons; ± represents standard deviations from mean; whisker on the bars represent the standard deviations (±)

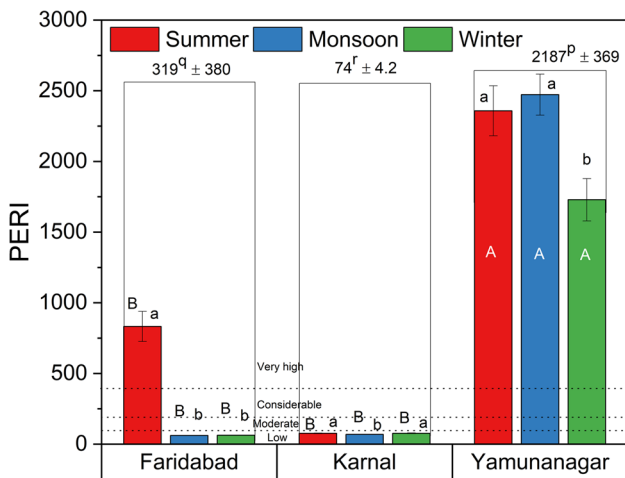


**Fig. 7** The ecological risk index ( $E_i$ ) of heavy metals in sewage sludge collected from Faridabad (STP1), Karnal (STP2), and Yamunanagar (STP3); values with different uppercase letters (A–B) on the bars are significantly different ( $P < 0.05$ ) for the season among different sites; values with different lowercase letters (a–b) in on the bars

are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) on the bars are significantly different ( $P < 0.05$ ) for comparison of the sites across the seasons;  $\pm$  represents standard deviations from mean; whisker on the bars represent the standard deviations ( $\pm$ )

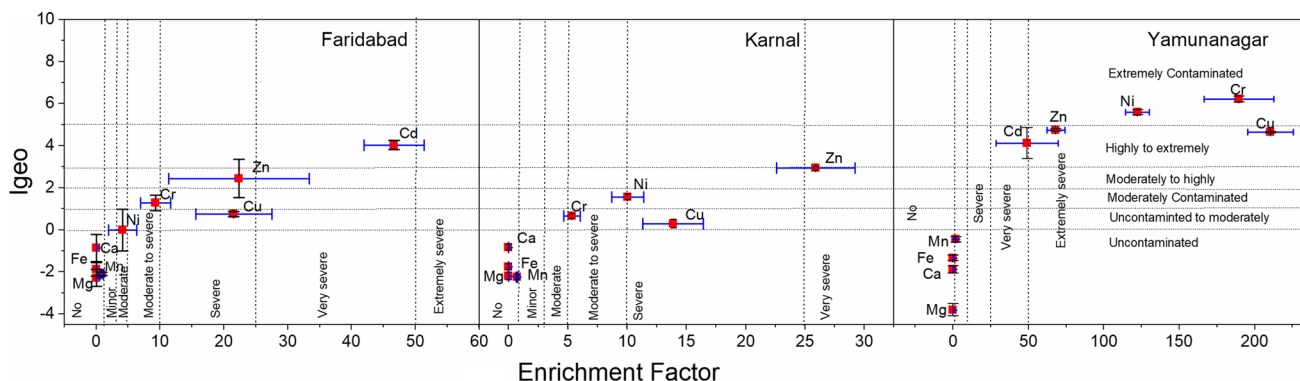
pH, site, and type of locality (Wang et al. 2005; Cai et al. 2007). The wastewater fugitively mixed with industrial waste or stormwater under tropical and subtropical regions undergoes rapid anaerobic digestion resulting in the precipitation of metals as sulfides (Rao et al. 2021). This makes treated water free from heavy metals but increased the loading of metals in SS. The physicochemical properties of the sludge govern the phase separation of the heavy metals. Reports showed higher binding and removal potential of heavy metals with particulate matter and coexisting ions like carbonates, hydroxyls, and bicarbonates (Kumar et al.

2020). Our results also corroborate this observation. The pH, TC, TP, Ca, and Mg explained ~90% of the total variability of heavy metals (Cd, Ni, Cr, Cu, Mn, and Zn) in sludge from different sites. Total carbon showed a strong positive correlation with heavy metal content in sludge. Positive role of the organic component of sludge in the sorption of heavy metals is well reported (Kumar et al. 2020; Rao et al. 2021). Metal ions form ion pair or complexes with co-existing anions like  $\text{CO}_3^{2-}$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$ , and organic matter (Kumar et al. 2020; Rao et al. 2021). The RDA analysis in the present study also showed a strong negative correlation with soluble  $\text{SO}_4^{2-}$ ,  $\text{Ca}^{2+}$ ,  $\text{HCO}_3^-$ , and  $\text{PO}_4^{3-}$  with heavy metals in SS (Fig. 2). Besides coexisting ions, the relative proportion of the heavy metals in solution and complexation and ion pair forms also depend upon the pH of the wastewater (Barker and Stuckey 1999; Kumar et al. 2020). A decrease in pH causes solubilization of the metals from the solid phase (Kumar et al. 2020); therefore, negative correlation was observed in the present study between pH and heavy metal content in SS. The increasing proportion of hydrolyzed metal species tends to increase pH favoring the immobilization of heavy metals with oxides, hydroxides, carbonates, and phosphates present in SS (Méndez et al. 2012; Zhang et al. 2017). Total nitrogen, carbon, and phosphorus content were found low in monsoon sampling because of dilution effect.



**Fig. 8** The potential ecological risk (PERI) of heavy metals in sewage sludge collected from Faridabad (STP1), Karnal (STP2), and Yamunanagar (STP3); values with different uppercase letters (A–B) on the bars are significantly different ( $P < 0.05$ ) for the season among different sites; values with different lowercase letters (a–b) in on the bars are significantly different ( $P < 0.05$ ) for seasons within site; values with different lowercase letters (p–r) on the bars are significantly different ( $P < 0.05$ ) for comparison of the sites across the seasons;  $\pm$  represents standard deviations from mean; whisker on the bars represent the standard deviations ( $\pm$ )

Different indices were applied to study the contamination, enrichment, accumulation, and ecotoxicological risk assessment of the different metals present in the SS. Although the concentration of some metal was lower in the SS, but their indices for ecotoxicological risk assessment were quite high. It was quite evident from Cd, Cr, Cu, and Zn content in STP1 and STP2 that in spite of concentration being in the permissible range (Delibacak et al. 2020), the contamination risks associated with their application was very high (Table 3; Figs. 3 and 4). Therefore, the calculation

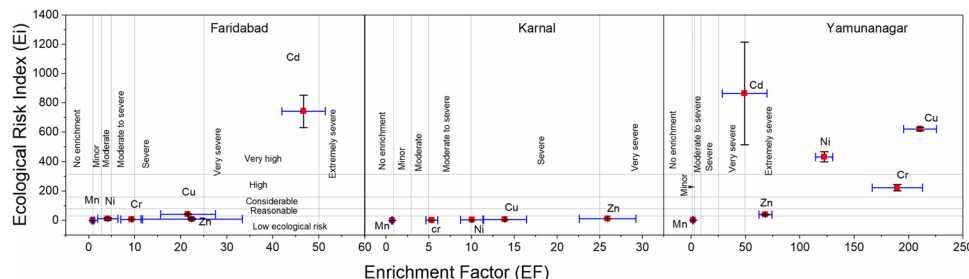


**Fig. 9** Bivariate plot of enrichment factors ( $E_f$ ) vs geoaccumulation index ( $I_{geo}$ ) for the metals present in sewage sludge samples from Faridabad (STP1), Karnal (STP2), and Yamunanagar (STP3); vertical

and horizontal whiskers on the coordinates represents standard deviations ( $\pm$ ) of  $I_{geo}$  and  $E_f$ , respectively

of toxicity indices is very important to estimate the risk of particular heavy metals on ecology (Ali et al. 2015). Cd, Ni, and Cr were major toxic metals present in the sludge samples. Similarly, high contamination factors were reported for Ni, Cu, Zn, and Cd in SS of Xiamen city, southeast China (Nkinahamira et al. 2019). Contamination and enrichment factors presented a far more precise and realistic picture of the ecotoxicological impacts of metal contents in SS. On the other hand,  $I_{geo}$  showed low metal accumulation in contrast to the results of  $C_f$  and  $E_f$ .  $I_{geo}$  classified all samples of SS as non-polluted to moderately polluted for elements present in high concentrations in the earth’s crust like Cu and Zn (Fig. 9). For this reason,  $I_{geo}$  remains inappropriate in explaining the metal toxicity as also reported earlier by Abraham and Parker (2008). The results obtained from  $I_{geo}$  better explained the presence of heavy metals in the sewage sludge (Cd, Ni, and Cr). Generally, levels of toxicities vary for individual metals. The sum or product of contamination factors of different metals did not produce the true picture of the extent of risk of toxicity. Pollution load index and polymetallic contamination indices presented the wholesome risk associated with the metals in SS. However, the individual metals have their respective toxicity levels, so this warrants

the strong need for rectification in the calculation of the toxicity effects of metals present in SS. It was observed that the enrichment factor coupled with geoaccumulation indices represents better results. Values of  $C_f$  above 6 and  $E_f$  above 10 are considered severe metal contamination/enrichment ( $C_{fCu}=8.5$  and  $6.1$  and  $E_{fCu}=21.5$  and  $13.9$  for STP1 and STP2, respectively), whereas geoaccumulation indices for the same were uncontaminated (Ali et al. 2015; Suanon et al. 2017). These indices are somewhat misleading and cause difficulty in categorizing the values under different levels of threat. PLI values for all SS were moderately polluted (PLI; 1–2 for STP1 and STP2) reflecting not much concern for soil applications. However, few metals like Ni, Cr, Zn, and Cu were present to varying degrees in SS and could pose a significant risk during soil application. Hence, PLI did not provide the risk associated with the individual metal pollution with the application of SS. The potential ecological risk index identified STP2 under low risk (Fig. 10) but metals like Ni, Cu, and Zn in SS samples of STP2 showed medium to severe risk for other indices. These indices can only be judged effectively after the application of SS in soils and assessing the fate of contamination, enrichment, accumulation, and further ecological risks through on-site studies.



**Fig. 10** Bivariate plot of enrichment factors ( $E_f$ ) vs ecological risk index ( $E_i$ ) for the metals present in sewage sludge samples from Faridabad (STP1), Karnal (STP2), and Yamunanagar (STP3); verti-

cal and horizontal whiskers on the coordinates represent the standard deviations ( $\pm$ ) of  $E_i$  and  $E_f$ , respectively

## Ecological threats and future perspectives

The source of wastewater and nearby locality influences the spatial variation of the elements in SS collected from the different STPs. Industrial wastewater contains several toxic components like metals, oil, grease, and recalcitrant compounds. However, standard rules and regulations ensure their proper treatment before discharge into the public water bodies. But, somehow this wastewater if mixed with the municipal wastewater may result in sludge contamination. Industrial areas are often associated with the flow of their wastewater to wastewater treatment plants (WWTPs) along with domestic wastewater. STP2 accounts for mainly residential wastewater, while STP1 and STP3 wastewater were composed of residential wastewater as well as discharge from manufacturing and industrial units located in the cities. This was the reason for the higher concentration of metals observed in STP1 and STP3. SS from different STPs showed the presence of an appreciable amount of C, N, P, Ca, Mg, and other nutrient elements. The N, P, and Ca values were lower than the earlier reported values for SS but these were higher than those of the farmyard manure (Nandakumar et al. 1998; Mercl et al. 2020; Sundha et al. 2022). However, certain environmental indices were very high indicating potential ecological risks associated with the land application. The higher enrichment of metals in SS indicated the unsafe handling of the domestic and industrial effluents. Other indices indicated potential ecotoxicological risk associated with its land application for agricultural purposes. Prolonged accumulation of such SS may result in bioaccumulation and biomagnification of heavy metals in soil and crops (Sciubba et al. 2015; Gattullo et al. 2017). Low soil microbial activities may deteriorate the soil health in the long run and ultimately may affect human health (Khwaitrakpam and Bhargava 2009; Rihani et al. 2010). Therefore, before focusing on the application of SS in agriculture, the incoming wastewater should be critically examined and the sludge must be properly stabilized for avoiding the environmental contamination and buildup of heavy metals in SS amended soil beyond the prescribed permissible limit (Table S13). Besides, the heavy metal accumulation, bioavailability, and biotoxicity associated with agricultural usage need to be further evaluated under different soil and climatic conditions to identify the suitable ecotoxicological indices for guiding agricultural usage of the ever-increasing SS generated in different wastewater treatment facilities.

## Conclusion

The present study concluded the presence of essential plant nutrients such as N (0.57–1.90%), P (0.30–0.40%), Ca (12.3–27.2 mg kg<sup>-1</sup>), and Mg (1.42–4.40 mg kg<sup>-1</sup>) in

the sewage sludge (SS) collected from different sites. The spatiotemporal variation in the quality of sludge implies the prior characterization of sludge for agricultural use. Mixing municipal wastewater with industrial discharges vitiates the beneficial properties of sludge. SS produced from municipal wastewater from nonindustrial cities posed a low potential ecological risk (PERI, 74.0), while industrial cities were associated with high heavy metals content with high ecological risk (319.0–2187.0). Heavy metal partitioning in sludge was highly dependent (~90%) upon the source, carbon content, and other associated anions like sulfate, chloride, bicarbonate, and phosphate. The SS generated in monsoon seasons had lower values for different pollution indices and can be used for agricultural purposes depending upon the soil type and nature of the crops. However, SS generated in summer and winter seasons had higher values for different indices; therefore, more precaution is needed for agricultural application. These findings highlighted the need for proper segregation of industrial and domestic effluents to avoid serious issues of heavy metals associated with SS application for agricultural purposes in the future. The enrichment factor coupled with geoaccumulation or ecological risk well explained the extent of the risk of heavy metals in the SS. This study further showed that the critical limits of the metals did not explain the degree of environmental risk; therefore, risk indexing must be carried out before agricultural usage of SS.

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**Availability of data and material** Data are available within the article and supplementary materials.

## Declarations

**Ethical approval** Ethical approval (Research article 49/2021) was obtained from the Project Monitoring and Evaluation Cell headed by the Director, CSSRI, Karnal (India).

**Consent to participate** Not applicable.

**Consent to publish** All the researchers participated in the study had approved this manuscript and given their consent for publication in this journal.

**Competing interest** The authors declare no competing interests.

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