RESEARCH ARTICLE

Temporal variation of leachate pollution index of Indian landfill sites and associated human health risk

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Abstract

The present study investigated the variation in leachate pollution index (LPI) of four municipal solid waste (MSW) dumping sites: nonengineered (Okhla, Ghazipur, Bhalswa) and engineered (Narela-Bawana) of Delhi, India. A review of 142 screened studies from Google Scholar database was done for synthesis of information on LPI parameters. Further, the rate constant determination and human health risk assessment for various leachate parameters was done. Results showed the following LPI trends: Okhla landfill: irregular with exceedance to threshold value; Bhalswa landfill: exponential increase; and Narela-Bawana landfill: linear increase. Parameters such as pH, dissolved solids, copper, nickel, zinc, and chromium of Bhalswa landfill, exhibited an exponential decay with LPI variation. Whereas, for Narela-Bawana's leachate BOD and COD parameters, an exponential decay in LPI vs zinc and linear increase for LPI vs lead was observed. For all dumping sites, a positive correlation was observed between heavy metals and LPI. In case of human health risk assessment, order of oral risk posed by Okhla's metals was cadmium > chromium > nickel > lead, with maximum hazard quotient (HQ) of 1.61 for cadmium. For Ghazipur and Bhalswa landfills, cancer risk values for both adult and child subpopulations were observed to be maximum for cadmium followed by nickel, chromium, and minimum for lead. For Narela-Bawana landfill, the order of cancer risk was as follows: chromium > nickel > lead. HQ for Pb-contaminated groundwater exceeded the threshold limit in Ghazipur and Bhalswa landfills. For dermal groundwater exposure, cadmium for Okhla (adult 2.3×10^{-4} and child 1.4×10^{-4}), Ghazipur (adult 9×10^{-5} and child 5.2×10^{-5}), and Bhalswa (adult 1.5×10^{-4} and child 8.6×10^{-5}) was observed to have maximum cancer risk. The analyzed year-wise LPI trend, calculated rate constants, and human health risk values from present study provide a basis to waste managers and regulators for understanding various waste sources.

Highlights

• Effect of landfill type on leachate pollution index's rate constant and its parameters.

- For Okhla and Bhalswa landfills, positive correlation was observed between metals (Cr, Zn, Pb) and leachate pollution index.
- Cadmium accounts for the highest cancer risk as compared with other metals for all landfills.
- Lead and cadmium observed to have significant risk for ingestion of leachate-contaminated groundwater.

The authors declare that the manuscript is not submitted to other journals for simultaneous consideration. The work is original and has not been published elsewhere in any form or language (partially or in full) that can be published in the present form on acceptance. Further, no data, text, or theories by others are presented and proper acknowledgements to other works have been given if referred.

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[•] Temporal trend suggests the continuous increase in Leachate Pollution Index of Bhalswa landfill.

Keywords Leachate pollution index \cdot Heavy metals \cdot Correlation \cdot Rate constant \cdot Landfill \cdot Human health risk

Introduction

Landfills are the most reliant and cheaper alternatives adopted for waste disposal purposes. Solid waste disposal in an unrestricted and non-systematic manner is a common practice prevalent in developing countries like India (Vijayalakshmi and Abraham [2017](#page-15-0)). Anthropogenic activities such as disposal of mixed wastes having hazardous characteristics in MSW landfill can have serious adverse effect. However, improper construction of these landfills without performing integrated impact assessment can become a major threat to both environment as well as human health. Leachate, a highly concentrated liquid generated through these landfills contains high concentration of organic, inorganic, and heavy metals (Christensen et al. [2001;](#page-14-0) Kumar and Alappat [2005;](#page-15-0) Afsar et al. [2015\)](#page-14-0). The leachate generated from the landfill sites depends upon various factors such as type of waste dumped, age of landfill, waste composition, climate, and hydrology of the site (Kumar and Alappat [2003](#page-15-0); Afsar et al. [2015\)](#page-14-0). These leachate constituents emanating from non-engineered/open-dumping sites have an immense potential to contaminate nearby groundwater (Mor et al. [2006](#page-15-0); Singh and Mittal [2009;](#page-15-0) Ahamad et al. [2019](#page-14-0)). This makes groundwater to be unsuitable for drinking and other purposes. However, to estimate the leachate pollution potential of different landfills, a quantitative hazard identification tool known as leachate pollution index (LPI) was developed (Kumar and Alappat [2005\)](#page-15-0). LPI is based on 18 leachate parameters, pH, total dissolved solids (TDS), biological oxygen demand (BOD), chemical oxygen demand (COD), total kjeldhal nitrogen (TKN), ammoniacal nitrogen (NH3-N), phenols, chloride, total coliform bacteria (TCB), cyanide, arsenic (As), nickel (Ni), mercury (Hg), lead (Pb), zinc (Zn), chromium (Cr), iron (Fe), and copper (Cu). Based on these parameters' concentration, the pollution potential of different landfills is compared. The higher the LPI value, the higher the risk of environmental deterioration (Arunbabu et al. [2017](#page-14-0)). Landfills having LPI surpassing the standard limit of 7.5 tend to be unsafe for leachate disposal in environment (Gupta and Arora [2016\)](#page-14-0). In literature, landfill leachate was also characterized for parameters different from LPI parameters such as sulfates, cadmium, cobalt, electrical conductivity (EC), salts (calcium, magnesium, potassium and sodium), aluminum (Al), and manganese (Mn) (Bhalla et al. [2014\)](#page-14-0). These studies assess the toxicity (Gupta and Paulraj [2017\)](#page-14-0) public health aspects (Singh and Mittal [2009\)](#page-15-0), chemical and toxicological evaluation of leachate (Ghosh et al. [2015](#page-14-0)), leachate quality assessment of different dumping sites (Somani et al. [2019](#page-15-0)), characterization of leachate and assessment of groundwater pollution near landfill site (Mor et al. [2006\)](#page-15-0), leachate transport and groundwater

contamination (Kumari et al. [2019;](#page-15-0) Srivastava and Ramanathan [2008](#page-15-0)), and potential risk to human health (Ahamad et al. [2019](#page-14-0)). Further literature review indicated the following knowledge gaps at present: (i) lack of information on comparison of leachate rate constant of engineered and non-engineered landfill, (ii) yearly variation of leachate pollution index of these two types of landfills, (iii) level of contribution of leachate parameters towards LPI in successive years, and (iv) human health risk (carcinogenic and noncarcinogenic) due to heavy metals leaching through both types of landfills.

Indian landfills such as Okhla, Ghazipur, and Bhalswa are non-engineered and open dumping sites (Guleria and Chakma [2019\)](#page-14-0). Due to absence of bottom liner system, leachate generated through these dumpsites can enter the unconfined groundwater aquifers, posing a significant risk to environment as well as to human health residing in the nearby landfill area. Delhi (India) generates approximately 10,000 TPD of solid waste, out of which Ghazipur, Bhalswa, and Okhla landfill receives 1200, 1650, and 1700 t of waste per day, respectively (Srivastava and Chakma [2020](#page-15-0)). Due to industrial waste dumping, leachate may also contain high levels of toxic elements such as Cd, Cr, Cu, Pb, Ni, and Zn (Christensen et al. [2001\)](#page-14-0). Heavy metal leaching from a dump site can be a potential source of groundwater contamination (Mor et al. [2006](#page-15-0)). Population residing nearby these dump sites is exposed to contaminated groundwater (Guleria and Chakma [2019;](#page-14-0) Adelekan and Alawode [2011\)](#page-14-0). Contamination source of chromium in landfills can be pigment or leather waste, and its exposure can result in lung cancer and dermatitis (Kim et al. [2015;](#page-15-0) Majolagbe et al. [2017](#page-15-0)) whereas lead release can be due to improper disposal of lead acid batteries resulting in neuron degeneration (Cohen et al. [1996](#page-14-0)). Groundwater samples from the handpumps within 0 to 1.53 km of the Narela-Bawana landfill have high concentration of TDS, chloride, Fe, Cu, and Cr (Gupta and Arora [2016](#page-14-0)). Ingestion of groundwater contaminated with metals can result in carcinogenic or noncarcinogenic risk to human health (Alidadi et al. [2019\)](#page-14-0).

An attempt to analyze the conditions of Delhi landfills and variation in their emanating leachate have been assessed in the present study. Time series analysis has been done to study the change in LPI values with the increase in successive years (from 2003 to 2018). Also, the variation in the metal concentration with time (years) has been studied. The study investigated that which metal is observed to have highest correlation with the change in LPI with time. Further, the rate constants were estimated by curve fitting between calculated vs reported LPI values with time, leachate parameters vs time, and LPI vs leachate parameters. Further, human health risk assessment (HHRA) for leachate's heavy metal–contaminated

groundwater was done for ingestion and dermal routes. The findings from this analysis can aid in policy formulation focusing on in situ impact analysis of leachate to groundwater and its associated risk. The present studies aimed to understand the following aspects:

- 1. Variation of LPI for non-engineered and engineered landfills over time.
- 2. Growth or decay in leachate parameters with time through exponential and linear model fitting.
- 3. Variation in heavy metals between engineered and nonengineered landfills.
- 4. Correlation between LPI and its constituents over the years.
- 5. Carcinogenic and non-carcinogenic risk posed to adult and children due to cadmium, chromium, nickel, and lead exposure.

Methodology

Study area

Delhi, India, was selected for present study as three out of the four landfills are non-engineered or open dumpsites and are still in working condition instead of reaching its end-of-life period (Afsar et al. [2015\)](#page-14-0) shown in Fig. S1 (supporting information document). Okhla, Ghazipur, and Bhalswa dumping sites are not designed as per the schedule 3 of the MSW Rules [2000](#page-15-0) (DPCC [2016](#page-14-0)). The information of all the dumpsites of Delhi is given in Table [1](#page-3-0).

Search criteria

In present study, the secondary data has been compiled from the review of existing available literature on four MSW landfills of Delhi, reported in last 20 years. The total number of studies obtained by keyword "Leachate Pollution Index" search in Google Scholar were 384 (on 20 October 2019). The keyword was selected in order to cover all possible studies related to temporal analysis of LPI for MSW landfills. After initial screening by removing duplicate or incomplete or irrelevant articles, significantly relevant articles obtained were 142. The overall methodology followed in present study depicting each step with its plausible explanation is shown in Fig. [1](#page-4-0). The available number of studies on the four selected landfills (Okhla, Ghazipur, Bhalswa, and Narela-Bawana) was divided into two categories: studies included qualitative analysis and studies included quantitative analysis. The quantitative studies were categorized in four sections: leachate pollution index, groundwater, temporal trend, and human health risk. Further, the studies under LPI section were used to compile reported LPI values and calculated LPI value from given data, and its variation against time was studied. The leached heavy metal concentrations were summarized and compared with their threshold values. Furthermore, linear and exponential models were fitted to investigate the leaching rate of various parameters and correlation analysis using Minitab 18.0, to determine the most critical parameters effecting LPI. Also, HHRA was done for heavy metal– contaminating groundwater.

Estimation of leachate pollution index

LPI is a hazard identification tool, formulated using Rand Corporation Delphi technique (Kumar and Alappat [2003\)](#page-15-0). LPI can be calculated by using the Eq. 1:

$$
LPI = \sum_{i=1}^{n} wipi \tag{1}
$$

where w_i is weight of the *i*th is pollutant variable, p_i is sub-index value of the *i*th pollutant variable, and n is the number of leachate pollutant variables. Different parameters have different w_i values which were derived by Kumar and Alappat [\(2003\)](#page-15-0) on the basis of expert opinion. When the number of leachate pollutant variables < 18, then the following modified equation is used:

$$
LPI = \frac{\sum_{i=1}^{m} w_i p_i}{\sum_{i=1}^{m} w_i}
$$
 (2)

where m is number of leachate pollutant variables, for which data is provided.

For most of the reviewed studies with respect to four Delhi landfills, data was reported for less than 18 leachate parameters resulting in application of Eq. 2. Most of the parameters obtained for Okhla, Ghazipur, Bhalswa and Narela-Bawana landfills were pH, TDS, BOD, COD, NH₃-N, Fe, Cu, Ni, Zn, Pb, Cr, and chloride. Very few studies have reported values for TKN, Hg, As, phenol, cyanide, and TCB, despite of being included in standard LPI equation. From the screened studies, data available on 18 physio-chemical parameters were compiled and analyzed, i.e., year and month of sampling, location of sampling, value exceeding threshold limit, and reported groundwater contamination information. Further, studies lacking LPI values but reporting the data on physicochemical parameters were used to calculate the LPI value. Moreover, details of sampling, i.e., pre-monsoon condition, monsoon condition, winter condition, and active dumping site, were clearly considered while calculating the LPI score (Fig. [2](#page-4-0)). It should be noted that only one article was considered from the screened articles reporting the leachate data for same sampling site and same sampling year. The sub-index (p_i) value against each leachate parameter were obtained from the sub-index rating curves (Kumar and Alappat [2005](#page-15-0)). To estimate the temporal LPI variation, firstly, the calculated LPI values for four

Table 1 Information on Delhi landfill sites

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NA not available

Fig. 1 Overview of literature review process for LPI studies

landfills were plotted against time (years). With the help of time series analysis, the change in LPI values (increasing or decreasing) over the years was estimated. Also, the yearly variation of each leachate parameter with time

and its correlation with the respective LPI was analyzed. Heavy metals, such as Fe, Zn, Pb, Cr, and Cu, were compared with their standard disposal values for every studies year for each landfill/dumpsite.

Fig. 2 Outline of methodology used in this study

Determination of rate constant correlation coefficient of LPI with its constituents

Rate constants (K_d) were calculated by plotting reported or calculated values versus time, and linear and exponential models were applied to obtain the best fit. Similar approach was also applied to investigate the correlation between variation in LPI and its constituents with one parameter at a time. The correlation of various leachate parameters, such as organic, inorganic, and heavy metal concentration of Okhla, Ghazipur, Bhalswa, and Narela-Bawana dumpsites, were determined against their respective LPI values. The slope of the fit was used as rate constant and K_d with coefficient of determination $(R^2) \ge 0.75$ were compiled and selected for further analysis (Nain and Kumar [2020a](#page-15-0)). Exponential model was found to be the best fit model for various parameters in maximum number of studies. Spearman correlation coefficient $(p < 0.05)$ was determined by using Minitab 18.0 software. Higher the correlation coefficient between the LPI and its constituents, the higher is the probability of variation in LPI value with change in the respective parameter value.

Human health risk assessment

HHRA consists of four main steps: hazard identification, exposure assessment, toxicity assessment, and risk characterization (US EPA [2001;](#page-15-0) Rajasekhar et al. [2018](#page-15-0)).The methodology adopted for risk assessment is described in Fig. S2 (SI). This study estimated the point-based risk posed on general (adult) and sensitive (children) population exposed to contaminated groundwater due to leachate percolation. It is assumed that when leachate is percolated through soil in groundwater, its constituents get diluted in the ratio of 1:100 (Christensen et al. [2001](#page-14-0)). To estimate the adverse effects of leachate on human health, metal concentration in leachate was diluted in the ratio of 1:100. Similar approach with same dilution factor has been previously proposed by Ghosh et al. ([2015\)](#page-14-0) and Guleria and Chakma ([2019](#page-14-0)). As leachate production shows seasonal variation, thus ratio of 1:100 dilution holds to be a good estimate.

Further, non-carcinogenic probabilistic human risk assessment (PHRA) was also done for Pb, Cd, Cr, and Ni. The methodology and all the parameter values used for probabilistic risk were followed from a recent study by Nain and Kumar [\(2020b\)](#page-15-0). Monte Carlo simulation was used to generate ten thousand random numbers for leachate metal concentration and dilution factor (range 1 to 1000) were used to estimate metal concentration in groundwater.

Hazard identification

Hazard identification is the first step to identify the elements that contribute to risk near the landfill site. On the basis of

literature review, Cd, Cr, Ni, and Pb were identified as possible hazards for human population. Reported highest observed concentration of these metals in leachate and groundwater is summarized in Table S2 (supporting information).

Exposure assessment

The primary exposure routes considered were oral consumption of leachate-contaminated groundwater and dermal contact through bathing. The landfill sites are surrounded by many residential areas; therefore, the exposure assessment was performed for two age groups: adults (21–70) years and children (0–6) years. The input parameters used for quantifying oral and dermal dosage exposure are shown in Table S3 (supporting information). Daily dose for oral and dermal pathways were estimated by using Eqs. 3 and 4, respectively, given by (US EPA [2004\)](#page-15-0).

$$
CDI = \frac{C_{\text{water}} \times WI \times EF \times ED}{BW \times AT}
$$
 (3)

where CDI: chronic daily intake (mg/kg day), C_{water} concentration of metal in groundwater (mg/L), WI: water intake (L/day), EF: exposure frequency (days/year), ED: exposure duration (years), BW: body weight (kg), and AT: exposure average time (days).

$$
DAD = \frac{DA_{event} \times SA \times EV \times EF \times ED}{BW \times AT}
$$
 (4)

where DA_{event} is absorbed dose per event (mg/cm² event) $(K_p \times C_W \times t_{event})$, DAD: dermal absorbed dose (mg/kg) day), SA: skin surface area available for contact $(cm²)$, EV: event frequency (events/days), K_p : dermal permeability coefficient of heavy metal in water (cm/h), and t_{event} : event duration (hour/event). K_p values for Cd, Ni, Cr, and Pb are 1.00E −03, 2.00E−04, 1.00E−03, and 1.00E−04 cm/h respectively (US EPA [2001](#page-15-0)).

Toxicity assessment

Information regarding toxicity of concerned heavy metals with their toxicity values used in risk estimation is summarized in Table S4 (supporting information). The oral cancer slope factor with administered dose is used to estimate the probability of increased cancer incidence over a lifetime excess cancer risk (RAIS [2020](#page-15-0)). To estimate the risk from dermal exposure, oral toxicity factor is adjusted to represent an absorbed dose (US EPA [2004](#page-15-0)). To analyze dermal toxicity, oral slope factor was extrapolated to dermal absorption using gastro-intestinal absorption factor (ABS_{GI}). This adjustment factor is based on absorption of element into gastrointestinal tract (Rajasekhar et al. [2018\)](#page-15-0). Dermal cancer slope (SF_{ABS}) SF_O / ABS_{GI}) factor can only be quantified for only Cd and Ni because the absorption factor value is not available for Cr and Pb (US EPA 2004). ABS_{GI} is the fraction of metal absorbed in gastrointestinal tract (dimensionless with Cd and Ni values as 0.05 and 0.04, respectively) (Elakhovskaya [1972;](#page-14-0) US EPA [2016\)](#page-15-0).

Risk characterization

Carcinogenic risk and non-carcinogenic risk assessment were performed by application of SF and RfD, respectively, using equation from 5 to 8 (USEPA 1989). This risk exposure via oral pathway was estimated for both the age groups (adult and child). Chronic daily intake (CDI) and cancer risk (CR) for adults and children were calculated for estimating carcinogenic risk. Dermal absorbed dose (DAD) and dermal cancer risk (DCR) for adults and children were calculated for estimating carcinogenic risk. While for estimating non-carcinogenic risk, hazard quotient (HQ) was evaluated.

$$
CR = CDI \times SF \tag{5}
$$

$$
HQ_{\text{oral}} = \frac{CDI}{RfD} \tag{6}
$$

 $DCR = DAD \times SF_{ABS}$ (7)

$$
HQ_{\text{dermal}} = \frac{\text{DAD}}{\text{RfD}}\tag{8}
$$

where CR: oral cancer risk, CDI: chronic daily intake (mg/kg day), DAD: dermal absorbed dose, SF: slope factor (mg/kg day)⁻¹, DCR: dermal cancer risk, SF_{ABS}: absorbed slope factor $(mg/kg day)^{-1}$, and HQ: hazard quotient.

Results and discussions

Time-dependent LPI estimation

The summary of selected screened studies reviewed on keyword search is presented in Table S1 (supporting information). The yearly trend of studies providing information on LPI and nearby groundwater contamination across various world landfill sites is presented in Fig. S3. With time, there has been increase in number of studies from 2003 to 2019 with a small drop in 2017. These studies were reviewed to investigate the aspects such as landfill type, year of leachate sampling, characterization of leachate parameters, its estimation over the years, and groundwater contamination due to leachate percolation over the years. The selected studies after final screening based on groundwater contamination were comparatively very less with respect to LPI estimation-based studies. Further, the screened LPI studies conducted on Delhi landfills are summarized in Table [2.](#page-7-0) Screened studies on Okhla (2003–

2018), Ghazipur (2003–2017), Bhalswa (2004–2014), and Narela-Bawana (2014–2017) were reviewed to estimate the respective LPIs and analyze its variation over the years.

Temporal trend

The LPI values for Okhla and Ghazipur landfills showed an irregular trend, but there is a significant decrease in LPI in 2017 and 2018, Table S5 (supporting information). For Bhalswa landfill, LPI showed an increasing trend from 2004 to 2014. These above stated landfills are non-engineered landfills. For Narela-Bawana despite of being an engineered landfill, LPI showed an irregular trend, with highest value observed in 2014, probably due to low-grade quality liner and improper leachate collection system, and thus, leachate has high potential to contaminate groundwater (Fig. [3](#page-10-0)). LPI trend of landfills with time can give an estimate of how the nature of the waste is changing in year wise manner. The quantity (concentration) and the quality (organic or inorganic) of waste affects the LPI of a particular landfill. The concentration of a component in leachate depends on waste type that is being dumped in these landfills. Therefore, variation in leachate composition and waste type can be one of the reason of these irregular trends. Due to non-availability of data on physiochemical parameters of these dumping sites, LPI was not calculated for some years. Based on LPI studies done on Narela-Bawana landfill, the index value is very high and have the potential to contaminate groundwater (Gupta and Arora [2016\)](#page-14-0). The Etueffont new cell landfill of France have LPI value of 13.3 on the basis of data reported in 2000–2010 (Grisey and Aleya [2016\)](#page-14-0). However, this value is much lower than LPI value of Narela-Bawana site. Through this comparison, it can be interpreted that there is a need to formulate stringent rules to determine the actual waste type that should be dumped in MSW landfill. The higher LPI values denote that the landfill may have received large amount of waste than its carrying capacity along with hazardous industrial waste because many studies reported high heavy metals such as Pb, Cr, Zn etc. in leachate (Singh et al. [2017](#page-15-0); Purwar [2018;](#page-15-0) Gupta and Paulraj 2016; Gupta and Arora [2016;](#page-14-0) Bag et al. [2016\)](#page-14-0). Further, this question the authenticity of the bottom lining system made to prevent leachate ingression in groundwater aquifers. The compiled concentration of heavy metals present in the leachate of engineered vs non-engineered landfills from reviewed studies has been summarized in Table S6 (supporting information). It can be seen from Table S6 that two landfill sites of India and France differ in pollution potential irrespective of having proper liner system. Several studies reported high heavy metal concentration in Narela-Bawana leachate and groundwater samples of the Narela-Bawana site (Gupta and Arora [2016](#page-14-0) and Gupta and Rani [2014](#page-14-0)). The yearly variations of Fe, Pb, Cr, and Cu is shown in Fig. [4.](#page-10-0) In case of Okhla landfill, iron concentration in the leachate decreased

Reference Year of	sampling	LPI	Outcome of the study	Information on groundwater and soil Knowledge gap pollution	
Okhla Kumar and Alappat (2003)	NA		42.18 1. All parameters except pH and Zn exceed permissible limits. 2. High metal concentration in leachate indicates possibility of industrial waste getting mixed with MSW. 3. Calculated LPI for Okhla landfill is	Unlined landfills and their leachate can contaminate the groundwater and can pose a danger to human health.	1. Year of sampling not known. 2. Only 13 parameters data are given.
Sharma et al. (2008)	NA		significantly high. 37.91 LPI for Okhla landfill 37.91. Reasons for high LPI: landfill not isolated from its environment, does not have liner or leachate treatment system, leachate flows freely and affect the water quality, with no gas collection system.	Every landfill has a localized groundwater aquifer and damage due to landfill leachate is the greatest threat to the surrounding environment.	1. Year of sampling is not known.
Singh and Mittal (2009)	June and Dec 2007		30.73 1. Leachate collected in December was found more toxic with 96-h LC50 less than 3% for L1 and less than 4% for L2, compared with June leachate with LC50 less than 13% for L1 and less than 12% for L2. 2. High Fe, Ni, and Mn concentrations.3. High concen- trations of ammonical nitrogen was also detected in leachate.	The leachate percolated down the ground surface and excess quantity gets collected in some low-lying areas and sometimes gets mixed up with sewer and drainage systems, thereby polluting groundwater and surface water sources.	1. Ecotoxicological evaluation and continuous monitoring of leachate is required. 2. Deleterious effects of leachate need to be assessed.
Ghosh et al. (2015)	May, 2012		22.11 1. Low metal concentration while organic compounds exceeded permissible limit by up to 145 times. 2. Low cancer risk. 3. Cytotoxic and genotoxic effects of leachate on HepG2 cell line was observed. 4. Leachate sample has alkaline pH, high conductivity and high organic matter. High Cd, Cr, Fe, and Cu concentration in Okhla landfill. 4. Percentage of naphthalene, pyrene, and anthracene were highest.	Accidental leachate leakage percolation into groundwater/surface run-off into water bodies can take place and it can pose a severe impact on human as well as can pose an effect on aquatic species	1. No standard maximum allowable discharge limit for landfill leachate is there in India, 2. HHRA needs to be done on the physio-chemical param- eters for the people living in the vi- cinity of landfill.
Afsar et al. Oct, 2014 (2015)			16.25 1. Comparison of the leachate from uncontrolled landfills of Delhi, i.e., Okhla, Bhalswa, and Ghazipur has been done in this paper. 2.Okhla landfill has higher concentration of P, organic nitrogen, Mg, K in comparison with Ghazipur and Bhalswa landfill site.	1. Major potential environmental impact Only 13 physico-chemical parameters, on landfill leachate are pollution of groundwater and surface water. 2. Potential for leachate migration to groundwater is very high.	included in Leachate pollution index are mentioned.
Purwar (2018)	2015		44.14 1. Seasonal variation of Okhla landfill's LPI has been evaluated. 2. Calculated LPI was 62.32 and 44.14 in summer and winter respectively.	Areas near landfills have greater potential of groundwater contamination	Only 7 parameters data are given.
Samal and NA Madgu- ni.		23.8	The LPI for pre-monsoon season is 27.01, monsoon is 23.8 and post-monsoon is 43.13.	Leachate has the potential to leach slowly into the soil and contaminate groundwater.	Only 9 parameters data are given.
(2018) Somani et al. (2019)	Jan, 2017		26.82 All physico-chemical parameters were higher than threshold value in six landfills of Delhi, Hyderabad and Kadapa. 2. Leachate from fresh waste was found to be more hazardous in comparison with leachate accumulat- ed in pond.	When leachate approaches the ground water table after migrating from unsaturated zone, it forms a plume that spreads in the flow direction and ultimately contaminates the groundwater.	Only 10 parameters data are given.
Ghazipur Mor et al. (2006)	Sep, 2003		25.95 High EC (24,500 μS/cm), TDS (27,956 mg/L), BOD (19,000 mg/L), and COD (27,200 mg/L), ammonical nitrogen (2675 mg L) 2. Heavy	1. High concentrations of CI^- , NO_3^- , SO_4^2 ⁻ , NH ₄ ⁺ , phenol, Fe, Zn, and COD in groundwater, indicating contamination by leachate. 2.	Only 11 parameters data are given.

Table 2 Summary of selected studies used for compiling calculated and non-calculated LPI values for Delhi dumping sites

Table 2 (continued)

Table 2 (continued)

Reference Year of	sampling	LPI	Outcome of the study	Information on groundwater and soil Knowledge gap pollution	
Ahmad et al. (2018)	2013		31.51 1. LPI is higher in case of pre-monsoon as compared with post-monsoon. 2. The reported values of Chlorides are much higher (9150 mg/L in pre-monsoon period). 3. Fe, Cr, Zn, Mg, Cu, Ni, Co, Pb, and Cd are found to be more common in leachate	Non-engineered landfill or open dumping of MSw leads to degradation of groundwater by generating leachate and its seepage into the ground. Highest value of Electrical conductivity, i.e., $10870 \mu S/cm$ in pre- and $10,450 \mu S/cm$ in post-monsoon sea- sons was found in sample number 29, located in landfill premises.	Only 11 parameters data are given.
Narela Bhawana					
Gupta and 2014 Paulraj. (2016)			34.34 1. Lower concentration of leachate stimulate germination and growth of Triticum aestivum while higher concentrations inhibited the process 2. High concentration of Fe, BOD, COD.	A large volume of ground water can be Only 10 parameters data are given. contaminated by even a small amount of landfill leachate.	
Gupta and Rani. (2014)	2014		34.08 High Cl concentration (4466.10 mg/L) and high Fe concentration	Landfill may lead to potential sources of Only 9 parameters data are given. groundwater contamination; pH 8.93, EC 4620 µS/cm, TDS 2220, Cl 1693.98, SO_4^{2-} 186.4, PO_4^{3-} 0.572, Ca^{2+} 113.58, Na ⁺ 85.62, K ⁺ 28.67, Mg ²⁺ 58.64, Fe 0.660, Cr 0.116, Cu 0.291 , Zn 0.86 .	
Gupta and Arora. (2016)	2016		25.03 1. High value of TDS as well as Chlorides were observed.2. Among heavy metals the conc. of Iron was the highest.	Landfill leachate can reach the groundwater aquifers and has the potential to contaminate the groundwater. pH 7.89, TDS 2209.6, Cl ⁻ 1702.21, total Fe 0.602, Pb 0.13, Zn 0.861, Cu 0.291, total Cr 0.116, Ni 0.4	15 out of 18 parameters were analyzed. Lack of study based on leachate variation with time.
Somani et al. (2019)	2017		25.96 1. Leachate was found to be highly alkaline.2. Cr, Pb, and Zn were also found higher in the leachate of Narela-Bawana landfill.	When leachate approaches groundwater Only 10 parameters data are given after migrating from unsaturated zone, it forms a plume that spreads and contaminate groundwater	

LPI leachate pollution index, TDS total dissolved solids, BOD biological oxygen demand, LC lethal concentration, COD chemical oxygen demand, Fe iron, Mn manganese, Ni nickel, Cd cadmium, Cu copper, Cr chromium, Pb lead, Cd cadmium, Si silicon, HCO3 bicarbonate ion, PAH polyaromatic hydrocarbons, EC electrical conductivity, Cl[−] chloride, NO₃ nitrate, NO₂[−] nitrite, SO₄^{2−} sulfate, NH₄⁺ ammonium, Zn zinc, TC total coliform, FC fecal coliform, F− fluoride, PO₄ phosphate, CaCO₃ calcium carbonate, Ca²⁺ calcium ion, Mg²⁺ magnesium ion, Na⁺ sodium ion, K⁺ potassium ion, TKN total Kjeldhal nitrogen, ULBs urban local bodies

*All leachate values in mg/L except pH and EC, TC, and FC

from 2003 to 2012 and further increased from 2014 to 2017. Further, the iron concentration was observed higher in 2017 in Narela-Bawana landfill. At present, there are no threshold values for iron in landfill leachate. In case of lead, the concentration in 2007 was much lower than the Narela-Bawana lead concentration (Singh et al. 2009). Okhla landfill shows the highest increase in lead concentration in the year 2015 and, thus exceeding the standard threshold value of 0.1 mg/L. Despite of having a liner system and leachate treatment facility, Narela-Bawana landfill have also exceeded the standard value of lead in leachate, except in the year 2016 (Leachate disposal standards, Municipal Solid Waste (Management and Handling) Rules [2000\)](#page-15-0). Chromium concentration showed a declining trend from 2003 to 2014 and from 2017 to 2018 for Okhla landfill with, the concentration much lower than Narela-Bawana's Cr concentration in 2017. The Cr concentration for both the landfills were within the standard limits (2 mg/L; CPCB [2017\)](#page-14-0) except for Okhla landfill in year 2003 and 2005 (Kumar and Alappat [2003;](#page-15-0) Sharma et al. [2008\)](#page-15-0). For Okhla landfill, the Cu concentration showed a declining trend from 2003 to 2017. In 2014, the concentration of Cu was much higher (exceeded the threshold of 3 mg/L) in Narela-Bawana landfill leachate than the Okhla landfill leachate. Zinc showed an irregular trend for Okhla dumpsite with time, i.e., first it decreases then increases. In 2016 and 2017, the zinc concentration in the leachate was much higher (exceeded the threshold of 5 mg/L) in case of Narela-Bawana landfill as compared with Okhla landfill. The presence of heavy metals suggests that the type of waste dumped in this landfill is mostly of industrial type ultimately affecting the LPI of the landfill.

Fig. 3 LPI variation of Delhi landfills with time (years)

Rate constant

Two models (linear and exponential) were applied to estimate the relationship between LPI vs time (years), time vs leachate parameters and LPI vs leachate parameters. Table S7 and Table S8 (Supporting Information) represents the rate constant K_d and R^2 values for all the LPI parameters respectively. The model giving rate constant values with $R^2 > 0.75$ was considered as best fit. The model explains the type of increase or decrease, i.e., linear growth or decay in terms of LPI or exponential growth or decay in terms of LPI vs time and leachate parameters. Out of all the landfills studied so far, only

Fig. 4 Metal concentration variation of Okhla and Narela-Bawana landfills with time

Bhalswa landfill LPI showed an exponential increment with time. Also, pH, TDS, Cu, Ni, Zn, and Cr showed an exponential fit model with time. However, BOD and COD (organic estimates of leachate) showed an exponential decay with time. The concentration of BOD and COD decreased with each successive year. With respect to comparison of K_d values with LPI and leachate parameters, TKN showed an exponential increment with LPI in case of Okhla and Ghazipur landfill. pH, TDS, Ni, Zn, and Cr also showed an exponential increase with LPI in Bhalswa landfill. This indicates that inorganic components in leachate are major contributors of LPI index. While in case of Narela-Bawana landfill, only Zn and Pb showed an exponential increase with their LPI values. Although Bhalswa landfill showed an exponential decay for BOD values with change in LPI and Narela Bawana showed an exponential increase for BOD with LPI. Rate constant values accounts for all processes that affects BOD and COD concentration including microbial degradation, dilution and dissolution (Bhatt et al. [2016](#page-14-0)). The K_d and R^2 values highlighted the significance of waste composition in MSW of landfills.

Correlation between LPI and leachate parameters

Correlation determines the strength of relation between two parameters (Gupta and Paulraj 2016). The spearman coefficient of correlation was calculated for LPI with its parameters (Table S9). The positive correlation was observed mostly between various heavy metals and LPI for all the landfills/open dumping sites. Figure 5 depicts the correlation of LPI with pH, TDS and chlorides. The higher correlation between inorganic constituents and LPI denotes that these parameters contributes more towards overall LPI variation. In case of Ghazipur landfill, positive correlation was observed between LPI and chlorides. Chloride is one of the inorganic macro components whose sorption, complexation and precipitation reactions are negligible. Thus, it can act as a conservative pollutant (Bag et al. [2016\)](#page-14-0). In Bhalswa landfill, a strong negative correlation is observed which is insignificant, whereas in case of Narela-Bawana landfill, correlative is positive but again insignificant one.

In case of Okhla landfill, the highest correlation is observed between LPI and Cu, Zn, Pb, and Cr (Fig. [6](#page-12-0)). This depicts that the heavy metal contribution is significantly more to landfill pollution (Singh and Mittal [2009](#page-15-0)). Presence of lead denotes that the contamination source can be disposal of lead acid batteries and it has a potential of posing cancer risk. (Cohen et al. [1996\)](#page-14-0). Similarly, in case of Bhalswa landfill, the highest correlation is shown between LPI and Ni, Zn, and Pb. The presence of nickel denotes that the contamination source can be discharge of rechargeable batteries and can pose an adverse effect on human health if it gets percolated through

Fig. 6 Correlation of LPI with

heavy metals

groundwater. Presence of chromium in landfills can cause adverse effect on human health such as lung cancer and its potent source is disposal of pigment, dyes etc. (Kim et al. [2015\)](#page-15-0).

HHRA for leachate contaminated groundwater

Carcinogenic and non-carcinogenic risk due to ingestion pathway

Landfills without bottom liner facility can become a source of groundwater contamination which can ultimately pose a risk to human health. Henceforth, health risk assessment becomes a major tool to determine the extent of risk due to unlined landfill leachate with excess amount of heavy metals. Both carcinogenic and non-carcinogenic risk (oral and dermal pathways) were evaluated for both the age groups (adult and child) for consumption of groundwater contaminated with leachate constituting heavy metals (Cd, Cr, Ni, and Pb) (Table [3\)](#page-13-0). The chronic daily intake and dermal absorbed dose estimated for ingestion and dermal pathways respectively is shown in Table S10. The Okhla, Ghazipur, and Bhalswa landfill contaminated groundwater, the risk posed by cadmium was maximum in comparison with other metals. For Okhla, Ghazipur, and Bhalswa landfills, cancer risk exposure of Cd in adults (Okhla 2.22 × 10^{-3} , Ghazipur 8.37 × 10−⁴ , Bhalswa 1.39 × 10−³) was greater as compared with children (Okhla 1.04×10^{-3} , Ghazipur 3.90×10^{-4} , Bhalswa 6.49×10^{-4}). For Narela-Bawana, Cd concentration was not reported in any literature, therefore, Cr metal exposure risk was compared with other metals, and it was found that Cr holds the maximum cancer risk in adults (1.26×10^{-4}) than children (5.89×10^{-5}) . However, these values were found to be greater than one in one million

individual's chance of getting cancer via consumption of metal contaminated groundwater. The safe threshold value for carcinogenic risk is $CR < 10^{-6}$ (Health Canada [2004,](#page-15-0) Alidadi et al. [2019](#page-14-0)). Although, the range of risk threshold suggested by EPA is 1×10^{-4} to 1×10^{-6} and unacceptable if the risk will exceed this threshold. A carcinogenic risk of 1×10^{-4} possess potential health hazard (Guleria and Chakma [2019](#page-14-0)). For non-cancer risk estimation, the HQ showed higher values for children in comparison with adults. During Cd exposure from consumption of groundwater contaminated with Okhla and Bhalswa leachate, HQ values was calculated to be 1.61and 1.01, respectively whereas during Pb exposure from consumption of groundwater contaminated with Ghazipur and Bhalswa leachate, HQ values was calculated to be 1.24 and 1.29, respectively. Further, on the basis of probabilistic based risk assessment for Bhalswa landfill, only lead showed an average risk of 3.47×10^{-6} and 1.16×10^{-6} for child and adult sub-population, respectively (Table S11). These values were slightly above the acceptable limit of $HQ < 1$. Therefore, Cd and Pb can be referred as a pollutant of concern in Okhla, Ghazipur, and Bhalswa landfill leachate. The LPI studies done by Kumar and Alappat [\(2005\)](#page-15-0) emphasized that heavy metals have a strong preference weighting in LPI evaluation but heavy metals such as cadmium irrespective of its toxic health effect was not considered in LPI formulation.

Carcinogenic and non-carcinogenic risk due to dermal contact

Since the absorption factors for Cr and Pb was unavailable, risk estimate of Cd and Ni only was evaluated for Okhla, Ghazipur, Bhalswa, and Narela-Bawana landfills. It was assessed that the risk posed by Cd was maximum among all

CR cancer risk, DCR dermal cancer risk, HQ hazard quotient

the heavy metals for Okhla, Ghazipur, and Bhalswa landfills. For Okhla, Ghazipur, and Bhalswa lanfills, cancer risk exposure of Cd in adults (Okhla 2.2 × 10⁻⁴, Ghazipur 9×10^{-5} , Bhalswa 1.5×10^{-4}) was greater as compared with children (Okhla 1.04×10^{-4} , Ghazipur 5.2×10^{-5} , Bhalswa $8.6 \times$ 10−⁵). For Narela-Bawana, Ni risk exposure was below the acceptable risk of $CR < 10^{-6}$. For non-cancer risk estimation, all calculated HQ values were below the threshold limit of $HQ < 1$, indicating that there is no possibility of having noncancer risk due to dermal exposure of heavy metals in either of the age groups. However, on the basis of probabilistic based non-cancer risk assessment for Bhalswa landfill, Pb, Cd, Cr, and Ni showed an average risk of 1.29×10^{-4} , 9.5×10^{-4} , 1.31×10^{-6} , and 3.32×10^{-4} for child sub-population and 4.45×10^{-6} 5, 3.76×10^{-4} , 5.29×10^{-7} , and 1.28×10^{-4} for adult sub-population, respectively (Table S11).

Overall, our findings indicate that LPI for various landfills observed to be high in some years. With respect to new solid waste management rules, 2016, concentration of various metals exceeded the standard limit such as cadmium (1.0 mg/L) and lead (1.0 mg/L) (Somani et al. [2019\)](#page-15-0). This further proved by human health risk assessment that there is significant risk for lead and cadmium appropriate action should be taken for their removal.

Conclusions

In present study, following findings were observed for LPI trend analysis with respect to time and other parameters:

- 1. LPI for Bhalswa landfill showed an increasing trend from 2004 to 2014 whereas for other landfills, an irregular trend was observed. In case of LPI vs time, Bhalswa landfill showed an exponential increase whereas the Narela-Bawana showed a linear decay.
- 2. An exponential increase was observed in case of time vs TKN, and LPI vs TKN for both Okhla and Ghazipur landfill. Though, in case of time vs Cu and LPI vs Cu, an exponential and linear decay was observed, respectively, for Okhla landfill site.
- 3. For Bhalswa landfill, an exponential increase was observed in case of time vs pH, TDS, Cu, Ni, Zn, and Cr; and LPI vs pH, TDS, Ni, and Cr whereas an exponential decay was observed in case of time vs BOD and COD; and LPI vs BOD. For all sites, LPI showed a significant positive relation with heavy metals ($p < 0.05$).
- 4. For Narela-Bawana landfill, an exponential decay was observed for time vs BOD and COD whereas for LPI vs BOD an exponential increase was observed.

5. Heavy metal variation from 2003 to 2018 was observed between Okhla (non-engineered) and Narela-Bawana (engineered) landfills. A linear decay model was observed for time vs Cu and an exponential increase for time vs Zn, Pb, and Cr.

With respect to risk assessment due to cadmium, lead, chromium, and nickel, it was found that cadmium is the most potent carcinogen possessing high CR values for all landfills. Also, for non-carcinogenic risk estimation, HQ > 1 was observed for mostly cadmium and lead for all the landfills except Narela-Bawana site. Cadmium, being a toxic metal and not a part of LPI equation posed risk. Therefore, there is a need to reformulate the LPI index with the addition of constituent such as cadmium because according to MSW 2000 rules, the LPI index needs to be 7.378 before discharge into inland surface water, so there is a need to control those leachate constituents also which are not a part of LPI evaluation but can play a major role in causing a health hazard due to its presence in landfill leachate.

From present analysis, it was observed that the heavy metal contribution in LPI value from time to time is significant. This suggests that the some portion of waste being dumped in these municipal landfills includes e-waste. Further, these nonengineered landfills do not have any liner system which increases the possibility of leaching of heavy metals into groundwater aquifers which can ultimately affect human health. Since, the inappropriate waste disposal practices cannot be controlled in a one-way process. The policies and rules made by government should be followed in a stringent manner along with the proper waste segregation and leachate treatment system. Also, relocation of landfill site can be considered as the old landfills have already reached their maturation period and devoid of liner system as well. There is need of regular monitoring of parameters that are not included in LPI formulation such as cadmium and yearly analysis of groundwater near the sites need to be monitored.

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Data availability Raw data could be provided to reviewer on request; however, author is not submitting it while initial submission.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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