Assessment of Greenhouse Gas Emissions in Delhi Landfills: Analyzing Factors Affecting Emissions and Environmental Implications

Abstract

Landfills are a primary method of waste disposal in developing nations despite their environmental impact. The decomposition of municipal organic waste in landfills generates potent greenhouse gases (GHGs) that contribute to the effects of urban climate change. In Delhi, India, which generates 11144 tons per day (TPD) of municipal solid waste (MSW), three major landfill sites (Ghazipur, Bhalswa, and Okhla) were examined using the well-established in-situ static chamber method to measure emissions of carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). . Thishe study highlights the need to address these uncertainties by comprehensively capturing GHG emissions from the diverse dynamics within the landfill through rigorous field experiments that account for spatial and temporal variability. The average CH_4 emission fluxes from three years of extensive field studies exhibited high variability, measured at 1494±893 (CV=59.8%), 1576±746 (CV=47.3%), and 961±322 (CV=33.5%) mg m^{-2} h^{-1} for Ghazipur (GL), Bhalswa (BL), and Okhla (OL), respectively. This resulted in CH_4 emission factors (EFs) of 5.6±3.5, 4.4±1.9, and 4.2±1.4 g kg^{-1} for GL, BL, and OL, respectively. The CO_2 emission fluxes were 7520±3401 (CV=45.2%), 8005±3907 (CV=48.8%), and 5066±1985 (CV=39.2%) mg m^{-2} h^{-1} with corresponding EFs of 20.0±7, 23.3±9, and 16.3±4.7 g kg^{-1}. The N_2O emission fluxes were 1210±329 (CV=27.2%), 998±298 (CV=30%) and 944±339 (CV=36%) µg m^{-2} h^{-1} with EFs of 3.8±0.1, 2.5±0.2, and 3.1±0.3 mg kg^{-1} for GL, BL, and OL, respectively. Total GHG emissions from Delhi's landfills were estimated as 328.6±91.9, 231.0±109.5, and 241.1±112.2 Gg CO_2 equivalent for 2009-10, 2010-11, and 2011-12. . Investigating waste management practices such as spreading, covering, and compaction is essential for understanding their impact on GHG emissions and advancing climate change mitigation through waste-to-energy solutions for sustainable solid waste management and energy production. While the findings offer valuable understandings into emission patterns, the limited sample size introduces some uncertainty, and the EFs should be considered as a preliminary estimation of major GHG in three consecutive years. Future research is necessary to validate these factors with more extensive datasets that capture spatial and seasonal variations in emissions.

Key Words: Greenhouse gas, Municipal Solid Waste, landfill-specific emission factor, Limiting factors, in-situ measurement.

1. Introduction

Municipal solid waste (MSW) management is a critical issue worldwide [1]. The changing lifestyles and consumption patterns observed, especially in urban areas, have been attributed to increasing industrial growth, economic prosperity, and expanding transportation networks in rapidly developing nations like India [2]. This phenomenon has resulted in a notable surge in waste generation due to heightened levels of consumption and production associated with urbanisation and economic development [2], [3]. As a result, these factors contribute to the emission of GHGs from human activities, posing a significant threat to exacerbating climate change [4]. With the world's largest population and undergoing rapid industrialization alongside shifting consumption patterns, India is experiencing a steep increase in MSW generation. In response to these environmental challenges, India actively participates in international negotiations as part of the United Nations Framework Convention on Climate Change (UNFCCC) to address these pressing environmental issues. Understanding the gravity of climate change, India has initiated comprehensive measures to combat it, including the implementation of a National Action Plan [5]. As part of this action plan, India has launched the National Mission on Sustainable Habitat, a promising initiative that aims to promote sustainable living environments. This initiative focuses on various aspects such as developing energy-efficient buildings and effective solid waste management practices [6]. One of the key challenges this mission addresses is the proper disposal of MSW, offering a hopeful solution to the current waste management crisis.

MSW has historically been disposed of in landfills in India, which led to the emission of greenhouse gases (GHGs) like methane (CH_4), carbon dioxide (CO_2), nitrous oxide (N_2O), and volatile organic compounds (VOCs) in the absence of proper scientific management [7]. These GHGs have high global warming potential (GWP) of CH_4 and N_2O, which are 28 and 298 times higher than that of CO_2 for a 100-year time scale [8]. In India, landfilling is one of the most common, cost-effective, but poorly managed ways of managing an MSW. Therefore, landfill sites are susceptible to groundwater contamination through leachate percolation [9]. Landfills, like burning agricultural crop residue, are poorly managed, particularly in Punjab and Haryana. This deteriorates air quality, which ultimately affects individuals' health [10-11],

making the issue of MSW management not just an environmental concern but a personal one. The potential health risks from poorly managed landfills are a cause for concern and underline the need for immediate action. Landfills are estimated to contribute approximately 754 Gg of CH_4 emissions, as reported in India's third Biennial Update Report submitted to the UNFCCC by the government of India in 2016. Only a few experiments have been carried out so far to estimate CO_2 and N_2O from landfills [12] – [18]. A study on Swedish landfills has shown that N_2O emissions are influenced by the CH_4 content of the soil, which, with concentrations above 5%, stimulates N_2O formation [12]. This growth rate highlights the escalating importance of addressing emissions from this sector within India's national GHG mitigation strategies.

However, it is crucial to note that detailed data on limiting factors such as types of waste, quantity of waste, waste composition, moisture levels, thickness of soil cover, and soil characteristics within landfills are not readily available in India. This lack of comprehensive data leads to significant uncertainties in estimating GHG emissions from MSW. The study outlined in this paper aims to address these uncertainties by comprehensively capturing GHG emissions resulting from the diverse dynamics within the landfill through rigorous field experiments that account for spatial and temporal variability. Understanding the emission behaviour of different GHGs is crucial, as these gases play a significant role in changing local climate patterns. In this study, an exhaustive field measurement (in-situ) was undertaken to measure total GHG (viz. CH_4, CO_2, and N_2O) emissions and tried to build a relationship with key limiting factors that influenced GHG emissions from the three landfills of Delhi during the period from 2009 to 2012. The GHG emissions data were used to develop landfill-specific GHG emission factors (EFs) for Delhi. These EFs (emission factors) are critical values that can be used to estimate total emissions from a landfill when conducting in-situ measurements is difficult or not feasible. Additionally, these EFs can serve as reliable alternatives in situations where model-based estimations may lead to overestimation, as highlighted by Chakraborty et al., 2011 & 2013 [13], [19].

2. Materials and Methods

The emitted GHGs (mainly CH_4, CO_2, and N_2O) from the Delhi landfills have been captured by landfill-specific in-situ GHG flux measurements using the Static Chamber Method. Extensive GHG flux measurements have been carried out from 2009 to 2012, covering

different seasons (viz., winter, summer, and monsoon) to reduce the uncertainties in the landfill GHG emission estimations.

2.1. Study Sites

Three landfill sites, namely Ghazipur (commissioned in 1984;28°37′22.4″N, 77°19′25.7″E); Bhalswa (commissioned in 1994; 28°44′27.16″ N, 77°9′27.92″ E) and Okhla (commissioned in 1996; 28°30′42″ N, 77°16′59″ E) situated in Delhi. The region is characterized by a hot, semi-arid subtropical climate with dry winters and significant temperature variations between summer and winter. The monsoon season, lasting from late June to mid-October, brings about 797.3 mm of rain. During the monsoon, rainfall recorded was 529 mm in 2009, 558 mm in 2010, and 590 mm in 2011. The types of MSW waste generally received at GL include waste from households, poultry, fish markets and slaughterhouses. BL receives waste from households and vegetable markets besides construction and demolition (C&D) waste. OL receives household and street sweeping waste along with C&D waste. Total MSW generation in Delhi is about 11144 TPD, where daily landfilling of MSW was estimated to be about 2300, 1800, and 1600 t from Ghazipur (GL), Bhalswa (BL), and Okhla (OL), respectively. Since 2018, approximately 4000 tons per day (TPD) MSW have been directed to Delhi waste-to-energy generation plants, a significant step towards sustainable energy production, yielding 24 megawatts (MW) daily. The remaining waste undergoes recycling processes, along with the management of construction and demolition (C&D) waste, as outlined on the official website of the Delhi Government. For further information on landfill sites, waste management practices, waste composition, and related details, readers are referred to the previous publication by Chakraborty et al., 2011 [13], [19].

2.2. GHG emission estimations

Our study aimed to develop landfill-specific emission factors of GHGs (viz.CH_4, CO_2, and N_2O) from Delhi's landfills. To achieve this, we applied an in-situ measurement method, using the well-established static chamber method to measure GHG emissions in landfills. The GHG flux samples collected from the chamber were analyzed by gas chromatography (GC; Model: 6890 N, Agilent Technologies, USA) using FID for determination of CH_4, CO_2, and ECD for N_2O and fitted with 25' x 1/16" stainless steel Hayesep-D and Porapac-Q columns. In each sampling, the air samples were collected from the static chamber using syringes at intervals of 15 minutes until 1 hr. Inside the chamber, two DC fans were fitted for homogeneous air mixing.

The GC injector, column oven, and detector temperatures have been maintained at 160°C, 50°C, and 250°C, respectively. The carrier gas as nitrogen (with 20 m1 min^{-1} flow rate at a constant pressure of 6 bar) was used to analyze CH_4, CO_2, and N_2O.

For the calibration of GC, the standard gases of 10 and 100 ppm CH_4 were used from NIST (National Institute of Standards and Technology, USA). For CO_2 standardization, MAINZ (Germany) provided standards of 394±0.2 ppm, and 1% CO_2 NIST standards were used. For the N_2O calibration, 500±0.2 ppb and 5.17±0.3 ppm of NIST standards were used. Thus, the output CH_4, CO_2, and N_2O concentration values were used for CH_4, CO_2, and N_2O emission flux calculations. The change of gases' concentration over time in the chamber as emission flux is calculated using equation 1.

The field sampling protocol was followed to capture GHG emissions in different seasons, covering the winter, summer, and monsoon in Delhi's three landfills for three consecutive years from November 2009 to December 2012. This comprehensive approach allowed us to capture the entire variation in the emission flux. The sampling was done twice every other month from three landfills during the period described above. The whole landfill area was divided into six zones of about 4-5 ha each based on the ages (1 month to 1.5 years of dumping) of the dumped MSW. In every sampling event, 8-12 sampling points were randomly chosen in the targeted zones in the landfill. Subsequently, the sampling was conducted in the forenoon and afternoon to capture the temperature influenced GHG flux from the landfill. The aluminium bases were fixed several hours before sampling, even though sometimes these were installed on the previous day of sampling to ensure that the best equilibrium state arrived at that point before putting chambers on the base. The base and the top were sealed using water, as discussed elsewhere [13], [18], [20], [22].

The number of sampling points across the entire landfill area varied seasonally, with 24-32 points in winter, 48-54 points in summer, and another 24-32 points in the monsoon season each year. The observed seasonal variations in GHG fluxes were analyzed to calculate average GHG emission fluxes specific to the landfill. Outlier data points were identified and excluded using the interquartile statistical method to refine the estimation of landfill-specific annual GHG emissions.

$$\text{GHG emission flux (mg m}^{-2}\text{ h}^{-1}) = \frac{\Delta X * EBV_{(STP)}}{10^6} * \frac{M \times 10^3}{22400} * \frac{60}{T} * \frac{1}{A} \ldots\ldots\ldots\ldots \quad (1)$$

Where,

154 Δ X = Change in concentration for each time interval (15, 30, 45 and 60 min) concerning that
155 at 0.
156 $EBV_{(STP)}$ = Effective box volume at standard temperature and pressure
157 T = Flux time in min. (15, 30…, 60)
158 A = landfill area covered by the box in m^2
159 M = Molecular weight of GHG (*viz.*CH_4, CO_2 and N_2O)

160

161 EBV = [(H + h) * L * B]…….. (2)
162
163 Where,
164 EBV = Effective box volume
165 H = Box height (cm)
166 h = Height of aluminum channel above the ground level (cm)
167 L = Box length (cm)
168 B = Box breadth (cm)
169

170 $EBV_{(STP)}$ correction equation

171 $$\frac{P_1V_1}{T_1} = \frac{P_2V_2}{T_2} \quad \text{……(3)}$$

172 Where,
173 P_1= Barometric pressure at the time of sampling in mm Hg
174 V_1= EBV (Effective box volume)
175 T_1= Temperature inside the box at the time of sampling in K (Kelvin)
176 P_2= Standard barometric pressure (760) in mm Hg
177 V_2= $EBV_{(STP)}$
178 T_2= 273 K

179 **2.3. Physico-chemical parameters of landfill soils**

180 In addition to in-situ GHG measurements, a detailed investigation of the physicochemical
181 properties of landfill soil has also been undertaken. The parameters studied, and corresponding
182 standard methodologies adopted for analyzing physicochemical parameters are summarized in
183 Table 1. The range of observed MSW temperature, moisture content, pH, etc. values during
184 the three seasons are 28 to 30 ^{O}C, 36 to 39 %, and 7.0 to 7.3, respectively, of different Landfills,
185 as shown in Table 1.

186 **3. Results and Discussion**

187 **3.1. Physico-chemical parameters**
188 **3.1.1. pH**

189 The samples were collected from the subsurface soil at 6 to 8 inches depth where the pH was
190 7.3±0.6, 7.2±0.8, and 7.0±0.8 from GL, BL, and OL, respectively. The methanogenic bacteria

operate only within the pH range of 6-8 [23], [24]. The accumulation of H_2 and CH_3COOH decreases the pH value, inhibiting the activity of methanogenic bacteria [25]. The pH values of MSW showed a little seasonal variability. During winter, the pH ranged from 6.9 to 7.2; in summer, it fluctuated between 7.2 and 7.8, and in the monsoon season, it varied between 6.8 and 7.1 (see Table 1). According to Li et al. (2007), organic carbon mineralization increases with pH increase, attributed to elevated CO_2 production alongside soil microbial biomass [25]. The pH of the soil surface, where aluminum bases were fixed, presumed to be the surface-emitting GHG, remained consistent across Delhi's landfills. In most instances, the pH levels were observed below neutrality, possibly indicating the fermentation stage. It was reported that the fermentation process strictly follows the pH ranges of <5, enabling ethanol production, and >5 initiates volatile fatty acids production. In this phase, acetate, H_2, and CO_2 are produced. This phase continues after dumping for 100 days, followed by the next anaerobic process, i.e. methanogenesis [26]. The pH levels of the MSW exhibited a weak correlation with GHG emissions in GL, BL, and OL (r = 0.68, 0.59, and 0.67 for GL, BL, and OL, respectively) across all seasons. This could be attributed to pH's crucial role in affecting GHG emissions in a controlled or protective environment. However, field experiments present a different scenario, likely influenced by various other factors. Additionally, leakages through pores, cracks, etc., could alter the statistics and contribute to these differing observations.

3.1.2. Temperature

The ambient temperature in Delhi was around 17 - 25°C in the winter, while in the summer, the ambient temperature ranged between 38 - 47°C. During the monsoon season, the temperature was recorded in the 27 - 41°C range during the sampling events. The humidity during the sampling events in winter varied from 29 - 58%. In the summer, the humidity dropped from 5% to 20%, while in the monsoon season, it remained at 55 to 98%. In summer, the MSW temperature (up to 6 inches depth) was observed to be 5 to 7°C higher than ambient temperature. However, in the winter season, this difference was nearly zero to one degree C. Still, in the monsoon season, sometimes, MSW temperature was lower than the ambient temperature by about 2 to 3°C. The active temperature for methanogenic microorganisms was reported to be 30-50°C [27]. The optimum temperature range of gas generation has been proposed to be between 30-45°C during the landfill gas generation phase. Consequently, it was thought that mesophilic and thermophilic bacteria were the cause of the increased greenhouse gas emissions that occurred throughout the summer. While due to the comparatively low

temperature, the mesophilic bacteria were dominant in winter and monsoon seasons. The impact of temperature change on the growth of biomass and the activity of the microorganisms, as well as GHG (CH_4, CO_2 and N_2O) emissions, was also observed [28].

3.1.3. Moisture content

Wastes have varying capabilities to hold water depending on the type of waste. Therefore, the moisture content in the garbage was found to be diversified. Maximum moisture content was found in monsoon season, reaching up to 80%, but on average, it ranged between 43-57%. Moisture content in the summer was as low as 28%, and the maximum was found to be 35%. In the winter season, the moisture content was found to be between 31 to 48%. Dach and Jager reported that the kinetics of degradation of organic matter was dependent on the water content of that material [29]The moisture below 25% would significantly affect and even inhibit the biodegradation process. Due to the unscientific covering of the top layer, rainwater is likely to enter Delhi's landfills through it, contributing to the hydrolytic process by dissolving readily degradable organic matter, as described by Hernández-Berriel et al. [30]. It was also reported that the moisture content in a typical landfill is lying down in a range of 15 to 40%, with a typical average of 30% [31]. Gurijala et al., 1997 claimed that the samples that contained more than 55% (wt/wt) moisture content produced significant amounts of CH_4 and CO_2 [32], [34]. Other researchers also reported how the moisture content in biodegradable waste influences the CH_4 and CO_2 generation rate in landfill sites [30], [31], [33], [34]. Hence, it was considered that the moisture content in MSW propelled the degradation processes by exchanging the substrate, nutrients, buffer, etc., and spread microorganisms into the different stages of degradation [35], [36]The moisture contents of dumped MSW were 30-42%, 25-35%, and 50-75% in the winter, summer, and monsoon seasons. The three landfill sites' annual average moisture contents were found to be 39±12%, 40±11 %, and 36±9% (Table 1).

3.1.4. Volatile Solids

In addition, the volatile solid is an important parameter, often used to measure the biodegradability of the organic fraction of MSW [37]. Some organic components of MSW, such as newspaper, exhibited high volatility but low biodegradability primarily due to its lignin content [37]. Employing a Muffle furnace set at a temperature of 550°C, the volatile solids (VS) component in the wet waste was determined to be 31.2±9, 32±11 and 29±9% in GL, BL, and OL, respectively (Table 1). The ash content was measured in three landfills and was

discovered to be 7±4%, reflecting the presence of inorganic or metallic constituents within the MSW. It was observed that there was a significant correlation between VS and GHG (CH4 and CO2) emissions, having correlation coefficients of CH_4 (r) of 0.89, 0.83 and 0.79 ($p<0.05$) for GL, BL and OL, respectively.

3.1.5. Organic carbon

The total organic carbon (OC) contents were recorded as 15±6%, 16±6%, and 14±4% in the different landfill sites (GL, BL, and OL, respectively, Table 1). The correlation analysis between OC content with CH_4 and CO_2 emissions revealed significant relationships, with correlation coefficients (*r*) of 0.671 and 0.892 ($p<0.05$), respectively. These findings suggested the active involvement of aerobic and anaerobic degradation processes within the landfills. Furthermore, the OC/VS ratio was determined to be 0.39±0.3, lower than the default value of 0.5 specified by the Intergovernmental Panel on Climate Change (IPCC) in 2006. This indicates a higher proportion of volatile solids relative to organic carbon content in the waste, suggesting potentially more significant anaerobic degradation activity.

3.1.6. Total Nitrogen

The total nitrogen (TN) in the MSW's samples from three landfill sites, GL, BL, and OL, was estimated as 1.7±0.6%, 1.9±0.6%, and 1.7±0.5 %, whereas organic nitrogen was 0.6±0.3, 0.9±0.4, and 0.7±0.4%, respectively. The NH_4^+, NO_3^-, and NO_2^- have been found to be dominant inorganic N forms. The results showed that N-NH_4^+ was higher than N-NO_2 and N-NO_3^- (Table 1). No correlation was discovered between TN, organic nitrogen (ON), NO_3^- and NO_2^- and surface N_2O emissions except with NH_4^+, which showed a weak correlation (*r* = 0.522). This indicated that N_2O emissions could primarily be from aerobic waste degradation through nitrification. The higher presence of NH_4^+ led to the nitrification process. However, nitrification and denitrification are cyclic processes that could be changed on any occasion by the abundance of NH_4^+ or NO_3^-[38].

Overall, these results provided valuable insights into the physicochemical composition of MSW in the studied landfills, highlighting the presence of inorganic components, the significant roles of organic carbon, nitrogen, and volatile solids in GHG emissions, and the active degradation processes of organic waste within the landfills.

3.2. Greenhouse gas flux estimations

The Compaction activity, surface temperatures, microbiological processes, heterogeneous surfaces, and fluctuations in trash composition all affect emissions and add uncertainty to estimates of greenhouse gas emissions from landfills. Similar challenges were encountered in GL, BL, and OL. Periodically, variations in GHG emission flux reached approximately 100% or even higher, necessitating the exclusion of some outlier data points through the interquartile statistical method. The high uncertainties associated with GHG emission flux estimations, attributed to spatial variations across seasons as reported by Zhang et al., 2019 [39], are further elaborated in the subsequent section, highlighting the seasonal variability in emissions.

3.2.1. Methane flux estimation

Figure 1 shows the seasonal variations in the CH_4 emission fluxes observed during 2009-2012. The GL site in Delhi showed a CH_4 emission flux of 1197.0±325.4 mg m^{-2} $h^{-1,}$ which was lower compared to the CH_4 emission flux in the other two landfill sites, e.g., 2201±472.1 and 1411.3±404.4 mg m^{-2} h^{-1}at BL and OL in the winter season (November to February) during 2009-10. During the summer season (March to June) in 2009-10, the CH_4 flux at GL, BL, and OL sites was observed to be 3617.5±994.4, 3006.2±1021.3 and 1154.3±393.8 mg m^{-2} h^{-1} respectively, with OL showing the lowest values. In the monsoon and post-monsoon season (July to October) in 2009-10, the CH_4 emissions flux at the GL, BL, and OL sites was observed to be 918.6±199.4, 833.7±294.5 and 557.5±122.8 mg m^{-2} h^{-1}respectively.

Likewise, in the winter season 2009-10, the GL site had low CH_4 emission fluxes (i.e., 951.0±321 mg m^{-2} h^{-1}) compared to BL and OL (1303±427 and 995±244 mg m^{-2} h^{-1}, respectively) during 2010-11. In the summer, the CH_4 fluxes followed the trends GL>BL> OL (2603±1025>2152±430>1476±464 mg m^{-2} h^{-1}). In the monsoon and post-monsoon, the variation in the CH_4 emissions fluxes in the landfills was not significantly different from each other, i.e., 833±283, 709±177 and 776±195 mg m^{-2} h^{-1} at all three sites.

During the winter season in 2011-12, the GL showed a similar pattern as found in previous years with lower CH_4 emission fluxes (920±224 mg $m^{-2}h^{-1}$) compared to the other two landfill sites, e.g., 980±346 mg m^{-2} h^{-1} at BL and 1062±394 mg m^{-2} h^{-1} at OL. During the summer, the CH_4 fluxes at GL, BL, and OL sites were detected as 2675±935, 2022±288, and 1632±378 mg m^{-2} $h^{-1,}$ respectively, while OL showed the lowest values other than two landfills. In the monsoon and post-monsoon seasons, the CH_4 emissions fluxes at the GL, BL, and OL sites were observed to be 909±262, 854±282 and 792±192 mg m^{-2} $h^{-1,}$ respectively.

There was considerable variability observed in seasonal CH_4 emission fluxes. Based on the aggregated data measured over the 2009-2012 periods, the seasonal CH_4 emission fluxes and average CH_4 flux from the three landfills of Delhi were calculated. The measured CH_4 emission fluxes were further subjected to statistical treatment for removal of outlier values using the inter-quartile method, which yielded the average seasonal CH_4 emission fluxes as 1027±286, 1485±591 and 1132±351 mg m^{-2} h^{-1} from GL, BL, and OL respectively in the winter season. In the summer, the CH_4 fluxes were detected as 2856±975, 2331±771, and 1312±537 mg m^{-2} h^{-1} from GL, BL, and OL, respectively. In the monsoon season, the CH_4 emission fluxes were 856±227, 755±221, and 660±169 mg m^{-2} h^{-1} from GL, BL, and OL, respectively. The results indicated higher CH_4 emissions during summer, followed by the winter and monsoon seasons. The average annual CH_4 emission fluxes calculations were 1494±893, 1576±746, and 961±322 mg m^{-2} h^{-1} from GL, BL, and OL, respectively (Table 2).

Although no significant correlation coefficient was observed between CH_4 flux and soil temperature, the role of temperature in CH_4 emissions could be ruled out. Afternoon emissions were found to be higher compared to forenoon emissions in the landfills (Fig. 2). This suggested that influencing factors for methane emissions from the landfill could extend beyond temperature alone; parameters such as humidity, atmospheric pressure, and others likely also contribute and deserve further exploration in future studies. The difference in average CH_4 flux values of 326±245, 370±448 and 208±187 mg m^{-2} h^{-1}, respectively, for GL, BL, and OL, was observed between forenoon and afternoon CH_4 emission fluxes indicating the influence of temperature on CH_4 emission processes in the landfills (Fig. 2). Continuous measurements of CH_4 emission in the landfills during the period from 8 AM to 4 PM in different seasons have also been carried out. It was observed that the CH_4 emissions were highest during the afternoon in all three landfills (Fig. 3). The CH_4 emission fluxes derived from three landfills in Delhi align with values reported by various authors [13], [18], [40].

3.2.2. Carbon dioxide flux estimation

Only a few experiments have been carried out to estimate CO_2 from landfills in India [13], [18]. The emission estimated by measurements carried out during the three-year study period revealed temporal and spatial variability in CO_2 emissions from Delhi's landfills (Fig.1). To minimise the error, the statistical inter quartile method was applied to remove outlier data points and in winter the CO_2 emission fluxes were spotted to be 6595±1418, 7754±2309 and

4016±1314 mg m^{-2} h^{-1} whereas, in the summer, the CO_2 fluxes were noticed as 10518±3223, 9956±4168 and 5824±2033 mg m^{-2} h^{-1} from GL, BL, and OL, respectively.

During the monsoon season, the emissions were 4468±1309, 4912±2703, and 4152±1602 mg m^{-2} h^{-1} from GL, BL, and OL, respectively. The lowest CO_2 emission fluxes were exhibited during the monsoon season compared to the other two seasons, except for OL, which showed higher emissions during the monsoon season than the winter season. The average CO_2 emission fluxes for three landfills were 7520± 3401, 8005±3907, and 5066±1985 mg m^{-2} h^{-1} (Table 3). The CO_2 emission fluxes obtained from three landfills in Delhi correspond closely to values reported in prior studies [40].

3.2.3. Nitrous oxide flux estimation

The N_2O emission fluxes were meticulously monitored from the landfills in Delhi over three consecutive years spanning 2009-2012. In GL, N_2O emissions were recorded as 1419.6±588, 1416.7±483, and 1208.3±489 μg m^{-2} h^{-1} in the years 2009-10, 2010-11 and 2011-12, respectively. At the same time, BL was noted to emit 1254.3±642, 1029.4±333 and 1006.6±312 μg N_2O m^{-2} h^{-1}, while the values obtained on the OL site were 1187.1±804, 987.9±342 and 994.9±287 μg m^{-2} h^{-1} in three consecutive years. The average of three years N_2O emission fluxes was found as 1210±329 998±298 and 944±339 μg m^{-2} h^{-1} from GL, BL, and OL, respectively (Table 4). The GL was observed as the highest N_2O emitter compared to BL and OL (Fig 1). However, no significant differences in the seasonal N_2O emission fluxes were noticed in Delhi's landfills. In landfills, the factors influencing GHG emissions are interdependent and reciprocal. As all GHG samples were collected at the same time intervals, it could be presumed that the emissions of all gases occurred simultaneously. When one gas was used as a normalizing factor for the other two gases, a moderate to high correlation (r^2=0.504, correlation= 0.71) was observed, mainly when CH_4 was used as the normalizing factor, and N_2O was plotted against CO_2. The available literature suggested that methane-oxidizing bacteria could also be a source of N_2O [26], [41], [21], indicating that N_2O generation and emission from landfills might depend not only on nitrification and denitrification but also on phenomena related to CH_4 emissions.

The increase of mineral and ammonium concentration inhibited methanotrophic activity [38]. The observation that CO_2 and CH_4 were not well correlated when N_2O was kept constant in landfill emissions suggested a complex interplay among these gases. This finding challenges the simplistic assumption that changes in one gas directly correspond to changes in another

when N_2O levels are steady. Instead, it implied that methane generation and oxidation dynamics could significantly influence the emissions of both N_2O and CO_2 from landfills. Furthermore, methane oxidation to CO_2 is another critical process in landfill environments. Methanotrophic bacteria convert methane to CO_2 as part of their metabolic activity. Therefore, changes in methane oxidation rates could impact the relative proportions of CH_4 and CO_2 released into the atmosphere.

Jha et al. documented N_2O fluxes ranging from 6 to 460 μg m^{-2} h^{-1} in Chennai's landfills during September 2004, consistent with the values observed in Delhi's landfills [18]. Elevated N_2O emissions were documented in landfills that utilised sewage sludge disposal or employed landfill leachate circulation as part of their management practices, as both methods contribute to increased nitrogen levels within the landfills [42]–[44]. However, such practices were not implemented in Delhi's landfills. Bo¨rjesson and Svensson observed N_2O emissions reaching 35.7 mg Nm^{-2} h^{-1} from active landfills covered with sewage sludge [12].

3.3. Factors affecting GHG emissions in landfills

The comprehensive analysis of the physicochemical composition of MSW in the studied landfills revealed several key factors influencing GHG emissions. The pH levels slightly below neutrality supported methanogenic activity, particularly during fermentation. Temperature variations also significantly impact microbial activity and GHG emissions, with higher emissions during the summer due to optimal conditions for mesophilic and thermophilic bacteria. In the seasonal variability assessment, the inferences were drawn from the CH4 and CO2 emissions, which were generally higher in summer compared to other seasons, except N2O emissions were indifferent among the seasons. However, surface temperature and GHG emission fluxes were not significantly correlated. Still, the impact of increased temperature was evident by higher CH_4 fluxes in the summer seasons and likewise in the forenoon compared to the afternoon when the ambient temperature was comparatively higher. Moisture content, crucial for biodegradation, which triggers hydrolysis processes, varies seasonally; the significant correlation between moisture content and CH4 emissions was spotted ($r = 0.75$, 0.7, and 0.69 for GL, BL, and OL, respectively) in summer and where the non-significant relationship was found in the monsoon post monsoon. The Volatile solids and organic carbon showed strong correlations with CH_4 and CO_2 emissions, indicating active degradation processes. A lower OC/VS ratio indicated a higher proportion of volatile solids, which

suggested active anaerobic activity and simultaneous aerobic degradation processes. Nitrogen content, particularly NH_4^+, correlates with N_2O emissions, primarily through nitrification.

During the study, different rainfall amounts were detected in the monsoon season (late June to mid-September) and post-monsoon season (September-October) from 2009 to 2012. Despite varying precipitation levels, CH_4 emission fluxes from the three landfills exhibited a consistent pattern, lower than in the summer months. There could be several reasons for these lower CH_4 fluxes during monsoon season. Hernandez-Berriel et al. claimed that rainwater reduces CH_4 and CO_2 emissions by dissolving mineralized carbon and washing out the nutrients along with microorganisms [30]. However, Visvanathan et al. proposed that CH_4 oxidation increased manifold in tropical regions when adequate moisture content was present in landfill-cover soils in addition to sufficient light and heat exposure [41]. In the three landfills during the monsoon season, the moisture levels of MSW have been estimated to be between 70 and 80 per cent. The monsoon season's CH_4 emissions were reduced due to the development of cracks by drying wet MSW and the diffusion of air oxygen into the underlying layers of MSW. It was also observed that the standard management procedures of MSW stacking, levelling, and compaction were challenging to adhere to adequately due to greater moisture concentrations. During the monsoon season, the available unfilled areas of the landfill were also used for dumping waste, which was later removed and put back on the pile after the end of the monsoon season. As a result, when temperature increased, the decomposition of waste heaps occurred aerobically instead of anaerobically.

The results indicated that CH_4 constituted about 27, 29, and 31%, whereas CO_2 constituted 73, 71, and 69% of the total GHG emissions (based on the total mass) from GL, BL, and OL, respectively, while N_2O emissions constituted only <0.02% in the total GHG emissions. In Delhi's landfills, it was noticed that a sizable amount of MSW decomposed aerobically rather than anaerobically, favouring the release of N_2O through the nitrification process. Some of the N_2O emissions also occurred through the denitrification process. Based on the samples collected from MSW piles dumped at different times in the landfills, it was noticed that 30-60% of carbon contents in organic matter in MSW was degraded within the first 3 to 6 years initially under aerobic conditions followed by an anaerobic process, then slowly taper off, continuing for periods up to 25 years or more [18].

Other causes may be attributed to the management practices employed by municipal authorities, such as dumping and spreading of MSW, compaction, soil cover, and the types of cover materials used. These practices, which were comparatively better in certain seasons,

resulted in lower GHG emissions. Conversely, poor landfill management practices in other seasons led to higher diffusivity through cracks and fissures, resulting in increased landfill gas emissions [42].

3.4. GHG emission estimation from 2009 to 2012

To estimate CH_4, CO_2, and N_2O emissions from Delhi's landfills, the average flux value of each year during the 2009-12 period was used (Table 2,3,4). The cumulative CH_4 emissions were 10.2±2.9, 6.8±3.3, and 7.2±3.5 Gg in 2009-10, 2010-11, and 2011-12, respectively. The CH_4 estimations results were found to be similar to those reported by the other previous [13], [18], [20]. The CO_2 emissions were estimated as 41.5±10.5, 39±17, and 38±14 Gg for 2009-10, 2010-11 and 2011-12, respectively. The N_2O emissions were 5.8±0.7, 6.0±0.5, and 5.8±0.6 Mg in 2009-10, 2010-11 and 2011-12, respectively (Table 5). These variations suggested potential shifts in waste management practices, landfill conditions, environmental factors influencing gas emissions, or climatic conditions affecting microbial activity. Therefore, achieving consistent GHG emissions estimates for landfills could be challenging, highlighting the need for rigorous and comprehensive studies to establish country-specific emission estimations. The total GHG emissions were estimated from the landfills of Delhi for the period from 2009-10 to 2011-12 as 328.6±91.9, 231±109.5, and 241.1±112.2 Gg CO_2 eq. (GWPs were calculated using a 100-year time scale). It was also estimated that the landfill gases composed of CH_4, CO_2, and N_2O in volume by volume (v/v) percentages ranging from 30% to 45%, 50% to 70%, and less than 1%, respectively. Therefore, a significant contribution of GHG emissions from landfills in Delhi underscores the urgent need for targeted interventions in waste management practices to mitigate climate change impacts. The estimated CO_2, CH_4, and N_2O emissions from each of the three Delhi landfills are shown in Table 5.

3.5. GHG emission factors

The landfill-specific CH_4 emission factors (EFs) were developed from the *in-situ* field measurement results in three Delhi landfill sites. The amount of MSW deposited (excluding inert materials) in the three years with total CH_4, CO_2, and N_2O emissions from each of the landfills in respective years were used to assess EFs (i.e. the ratio of *Total Emission* vs. *deposited MSW* in a particular year) of these gases. The CH_4 EFs were 5.6±3.5, 4.4±1.9, and 4.2±1.4 g kg^{-1} for GL, BL, and OL, respectively (Table 6). The CO_2 EFs were calculated to be

20±7, 23.3±9 and 16.3±4.7 g kg^{-1} for GL, BL, and OL, respectively, while the N_2O EFs were 3.8±0.1, 2.5±0.2, and 3.1±0.3 mg $kg^{-1}$for GL, BL and OL, respectively.

It is imperative to consider spatial and temporal variations in GHG emission estimations when developing representative emission factors (EFs). The study highlighted that estimating emissions without considering seasonal variations might lead to underestimating or overestimating total GHG emissions from landfills. For instance, if we consider CH_4 emission fluxes obtained during the monsoon season, the annual CH_4 emissions from GL, BL, and OL would be estimated at 2.1, 1.7, and 0.7 Gg, respectively. In contrast, if we consider emission fluxes obtained during the summer seasons, the results would be 9.3, 7.4, and 1.5 Gg for GL, BL, and OL, respectively.

4. Limitation and Future Scope

The research provides valuable insights into emission patterns from three landfills in Delhi, India, using static/flux chamber methods. However, the development of Emission Factors (EFs) is constrained by the limited sample size collected across the seasons over three consecutive years. Additionally, key factors influencing GHG emissions in landfills were not fully addressed due to limited data availability and a lack of comprehensive insights into these variables.

To strengthen these findings, future studies should address these gaps through extensive sample collection in line with field monitoring guidelines. Capturing both temporal and spatial variations in GHG emissions from landfills will provide a more comprehensive understanding of their patterns. This approach will also help accurately identify and address the key factors most significantly influencing GHG emissions. Foundational static/flux chamber methods provide valuable understandings, but we still understand that the advancement methods strengthen the study. Hence, this is one of the limitations of the present study, and in future studies, more advanced methods will be used for the measurement of gas emissions.

5. Conclusions

With increasing population and urbanization, MSW management has become a pressing issue, primarily addressed through landfill disposal, leading to GHG emissions. This study aimed to provide reliable estimations of GHG emissions from landfills and understand the processes that

influence GHG emissions. Field sampling encompassed winter, summer, and monsoon seasons, revealing significant seasonal variations in GHG emission fluxes. CH_4 emission factors (EFs) were determined as 5.6±3.5, 4.4±1.9, and 4.2±1.4 g kg^{-1} for GL, BL, and OL, respectively. CO_2 EFs were 20±7, 23.3±9, and 16.3±4.7 g kg^{-1} for GL, BL, and OL, respectively, while N_2O EFs were 3.8±0.1, 2.5±0.2, and 3.1±0.3 mg kg^{-1} for GL, BL, and OL, respectively.

The comprehensive analysis of MSW in the studied landfills revealed key factors influencing GHG emissions. Slightly acidic pH levels supported methanogenic activity during fermentation. Temperature variations significantly impacted microbial activity, with higher CH_4 and CO_2 emissions in summer due to optimal conditions for mesophilic and thermophilic bacteria. While surface temperature and GHG emissions were not significantly correlated, higher CH_4 fluxes were observed in summer and forenoon. Moisture content, crucial for biodegradation, significantly correlated with CH_4 emissions in summer but not during the monsoon. Volatile solids and organic carbon strongly correlate with CH_4 and CO_2 emissions, indicating active degradation. The lower OC/VS ratio suggests active anaerobic activity alongside aerobic processes. Nitrogen content, especially NH_4^+, correlates with N_2O emissions primarily through nitrification. The highest CH4, CO2, and N2O emissions occurred from April to July, attributed to increased temperature conditions, optimum moisture concentration, readily degradable organic carbon content, C: N ratio and volatile solids.

Overall, the findings underscore the importance of considering spatial and temporal variations in GHG emissions from landfills to generate accurate estimations. The study highlighted the necessity for further research to develop more reliable data, particularly in harnessing energy from waste sources, which could address solid waste management and power crisis issues. These insights shed light on the physicochemical composition of MSW in landfills, emphasizing the roles of organic carbon, nitrogen, and volatile solids in GHG emissions and the active degradation processes occurring within the landfills. This research offers a preliminary valuable understanding of emission patterns through the creation of EFs However, it's essential to acknowledge the limitations caused by the limited number of samples. Therefore, the presented EFs should be considered as an incentive estimation. To enhance the accuracy and usefulness of these factors, future research should focus on gathering more extensive field data, considering both spatial and seasonal variations in emissions. This will

lead to more reliable emission assessments and validate the findings of this study across various conditions.

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Table Captions:

Table 1. Analysis of physico-chemical parameters of MSW

Table 2. Seasonal and average annual CH_4 emission flux (mg m^{-2} h^{-1} ± SD)

Table 3. Seasonal and annual CO_2 emission flux (mg m^{-2} h^{-1} ± SD)

Table 4. Annual variations in N_2O emission fluxes (2009-12)

Table 5. Emission estimations of CH_4, CO_2 and N_2O in years 2009-12

Table 6. Landfill specific GHG emission factors (EF) for GL, BL, and OL.

Figure Captions:

Fig. 1. Seasonal and annual variations of CH_4, CO_2 and N_2O emission during 2009-2012

Fig. 2. Impact of temperature on CH_4 emission in three landfills

Fig. 3. Temporal variations in CH_4 emission in landfills during the day time