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METHANE AND CARBON DIOXIDE EMISSION AND LEACHATE CHARACTERIZATION FROM LANDFILLS ALONG GANGES RIVER AT VARANASI (U.P.), INDIA

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Abstract

Landfills are now considered to be among the largest anthropogenic source of green house gas (GHG) emission in the world. Methane and carbon dioxide emission rates were studied at two landfill sites along the Ganges river at Varanasi (U.P.), India. Chemical characteristics of leachates emerging from both the landfills were also studied. Overall, the emission rates ranged from 10.73 to 60.20 mg CH_4 m^{-2} h^{-1} and from 17.28 to 281.27 mg CO_2 m^{-2} h^{-1}. Emissions increased sharply with rising temperature in summer months. The rates were higher at young landfill site and between site differences in CH_4 and CO_2 emissions were significant. Landfill sites differed also with respect to leachate characteristics. The BOD_5: N: P ratio averaged to 90 : 3 : 1 at Site 1 and 80 : 2 : 0.5 at Site 2 indicating that, for leachate treatment, the aerobic biological systems may operate successfully without nutrient supplement. Although, more exhaustive data sets are needed, the present study, that forms the first report on GHG emission from Varanasi landfills, will help reducing uncertainties in CH_4 and CO_2 emission estimate scenario.

Key words : Climate change, Ganga river, green house gas, landfill, leachate.

Introduction

Among the many environmental issues associated with rapidly growing population and economic development, an important issue is the rising accumulation of municipal solid wastes (MSW) around towns and metro-cities. The management of MSW is a major problem faced by the municipalities of India. Per capita production of MSW in India ranges from 0.2 to 0.5 Kg day^{-1} (Sharholy *et al*., 2005) and the most common method applied to manage over 90% of MSW is open dumping (Ithnin *et al*., 2012). Major environmental problems associated with MSW include availability of space, emission of green house gases (GHGs) and the leachate that drains from landfills. Emission of GHGs from MSW contributes to global warming, while leachate drains from dumping sites contaminate surface and ground waters (Chakroborty *et al*., 2011 and Ithnin *et al*., 2012). Compared to most of the developed countries, the MSW in developing countries contain higher percentage of organic wastes and by implication, may have high potential to emit GHGs per unit weight of MSW. Further, when water percolates through a large mass of MSW, it generates high strength contaminated liquid commonly known as leachate. It varies widely in composition depending upon the age of landfill and the constituents it contains. Untreated leachate generated from MSW may lead serious threat to environment including contamination of surface and ground water resources (Cortez *et al*., 2011).

Among the global sources of methane reaching the atmosphere are wetlands (Matthews and Fung, 1987; Cicerone and Oremland, 1988; Smith *et al*., 2000), contributing to about 88% of all natural methane emission (IPCC, 2007 and Flury *et al*., 2010). Recent studies have shown that landfills are among the largest anthropogenic source of methane (Jha *et al*., 2007; Chang *et al*., 2009; Chakraborty *et al*., 2011 and Vargas *et al*., 2013). On global scale, generation of MSW is about 1200 Tg/yr (1 Tg = 10^{12} g), >70% of which is landfilled. Landfilling of waste contributes about 30-35 Tg methane (CH_4) annually to the world's total CH_4 emission of ~550 Tg/yr. Compared to wetlands and paddy fields, landfills function as a closed system characterized by controlled burial of biodegradable organic materials. The compositions, moisture content, pH and the management practices all influence the methane and carbon dioxide production in landfills. The essential reaction in a landfill is anaerobic

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biodegradation of organic components of MSW. It involves hydrolysis of complex molecules into soluble product by bacteria; conversion of these soluble products by acid forming bacteria to simple organic acids, carbon dioxide and hydrogen gas and finally, production of methane either by breaking down the organic acids or by reducing CO_2 with H_2 by methanogens (Chen *et al*., 2008) under optimal pH (6.82 to 7.4) and moisture content.

Although, the global warming potential (GWP) of methane is about 25 times higher than that of carbon dioxide, the contribution of the latter to overall green house forcing is about four folds higher (Forster *et al*., 2007). On 9th May 2013, the daily mean carbon dioxide concentrations at Mauna Loa, Hawaii crossed the milestone of 400 ppm (Monastersky, 2013). Values much larger than this value have been recorded on local and regional scales depending upon local source- sink relations and atmospheric stability (Bala, 2013). During recent years, anthropogenic emissions and large scale land use changes have been implicated for this rise in CO_2 (Ballantyne *et al*., 2012). However, an understanding of magnitude of anthropogenic impact on composition of radiative gases requires information pertaining to the contribution of other sources to overall emission scenario. The contribution of MSW in total CO_2 through local and regional-scale CO_2 emission has received relatively less attention.

In developing countries such as India, inventory estimates of emission of green house gases from landfills show wide uncertainties due to paucity of data on *in situ* emission estimates. *In situ* measurements of GHGs emission from landfills therefore bears great relevance in not only reducing uncertainties in inventory estimates, but also for local and regional contributions to overall green house forcing. In the present study, we present the results of site specific emission fluxes of CH_4 and CO_2 measured at two landfill sites of Varanasi for six consecutive months. We also attempted to characterize the quality of leachate being generated from the landfills and to compare our results with other observed records. With a population of over 1.6 million, the city witnesses massive generation of MSW annually. The total quantity of MSW generated in Varanasi is about 600 million ton (Municipal Corporation of Varanasi). Being the cultural capital of India, a major part of waste generated in Varanasi city comprises of biodegradable, compostable and recyclable materials. Varanasi is a prominent tourist place and therefore, receives significant floating population that also contribute to massive amount of waste generation. For these reasons, the present study bears scientific and environmental relevance and contributes to national GHGs emission inventory estimates data base. The study, that forms the first report from Varanasi landfills, will help reducing uncertainties in GHG emission scenario.

Materials and Methods

Study area

The present study was conducted during dry months (January to June) of 2013 to measure fluxes of CH_4 and CO_2 emission from two landfill sites of Varanasi city (25° 18' N latitude, 83°1' E longitude and 76.19 m above mean sea level) situated at the west margin of Ganga river. Climate of the region is tropical with three distinct seasons; winter (November to February), summer (March to June) and rainy (July to October). Varanasi region receives 1100 mm average rainfall, about 90% of which occurs during rainy season. During the study period, mean monthly maximum temperature ranged from 27.5 to 45.4°C and minimum temperature from 9.5 to 25.7°C. During winter, night temperature sometimes drops below freezing. Wind direction shifts from predominantly westerly and south-westerly in October through April to easterly and north westerly in remaining months. The alluvium soil of the region is highly fertile light textured sandy loam with pH ranging from 6.8 to 7.9.

Landfill Site 1 is situated at Purana Pul near Kazzakpura (KZP), Varanasi. The site is currently being used for dumping MSW. The waste dumped consist of household waste, animal waste, street sweeping (mainly polythene, plastic material, foam, paper, packing materials, metals, cloths etc.), construction and demolition waste and excavated soil. Site 2 is situated at Bypass (BPS) near Ramnagar, Varanasi (U.P.), India. This site is abandoned from last one year although the composition of the MSW dumped at the Site 2 is almost similar to that of Site 1.

Experimental design

The experimental protocol of the investigation consists of two tiers of studies, including characterization of leachate and emission of methane and carbon dioxide from two landfill sites. The strategy was to evaluate the possible contamination from the leachate draining to Ganga River and to record source emission estimates of two most important green house gases, CH_4 and CO_2. The factorial design of the whole experiment consisted of two landfills x twelve leachate quality variables x two green house gases.

In-situ measurement of CH_4 emission flux

The *in situ* emission flux of CH_4 was measured during dry months (January to June) of 2013 from two MSW

dumping sites namely, Kazzakpura (KZP) and Bypass (BPS). Closed chamber technique (Majumdar *et al*., 2000) was used for measuring CH_4 emission from landfills. Sampled areas were partly covered by excavated soils and/or construction and demolition wastes. Cylindrical steel chambers of 25 cm diameter were inverted and fixed firmly penetrating the soil to 6 cm deep. A battery operated pump was used to circulate air to ensure proper mixing of air inside the chamber. Each chamber was devised with one sampling pot on the top with a non-reactive silicon rubber septum. Samples were collected at 0, 15, 30, and 45 minute intervals in 10 ml air tight syringes fitted with three way top cork to prevent leakage of air samples. During each sampling event eight air samples from a chamber were collected using air tight syringes for gas chromatographic (GC) analysis. Landfill temperature, atmospheric temperature and chamber temperature all were also continuously monitored. Concentration of CH_4 was analysed in a gas chromatograph (Agilent Technologies 7820A, Germany) equipped with Thermal Conductivity Detector (TCD) and a Porapak Q Column (3 m). The carrier gas was N_2 with flow rate of 7 ml/minute. The detector and injector temperatures were set at 250°C and 210°C, respectively. The oven temperature was set at 60°C for 0 min and then ramped to 180°C at the rate of 10°C/min for 17 min. During analysis, standard gas of 10 ppm and 100 ppm of NIST (National Institute for Standards and Technology, USA) were used for calibration. The CH_4 concentration values, thus obtained, were used for CH_4 flux calculation. Concentration of gas was calculated from respective relative peak area from the standard gas of known concentrations obtained by running known volume of standard gas in GC. The methane emission flux (mg m^{-2} h^{-1}) was calculated using the following equation :

$$CH_4 \text{ emission flux } (\text{mg m}^{-2}\text{ h}^{-1}) = \frac{\Delta X \times EBV_{(STP)} \times M \times 10^3 \times 60 \times 1}{10^6 \qquad 22400 \quad T \quad A}$$

Where,

ΔX = change in concentration for each time interval (0, 15, 30, 45 min)

$EBV_{(STP)}$ = effective box volume at standard temperature and pressure

T = flux time in min (0, 15, 30, 45 min)

A = landfill area covered by the box in m^2

M = molecular weight of CH_4

In-situ measurement of CO_2 emission flux

Carbon dioxide emission flux was measured at each experimental site using closed chamber technique. This technique involves trapping of the gas in a closed perspex chamber as it leaves the soil surface and allowing the gas to absorb in KOH within the chamber for a known period of time, which is latter analyzed volumetrically. The perspex chambers were inverted and fixed firmly penetrating the soil up to 6 cm deep. The CO_2 built–up inside the chamber was trapped in 50 ml of 0.5 N KOH kept in a beaker inside the chamber for a known period of time. The KOH with absorbed CO_2 was brought to the laboratory and titrated against 0.5 N HCl. The CO_2 emission flux was measured in terms of mg m^{-2} h^{-1}.

Landfill leachate quality

The samples of landfill leachate were collected from both sites and stored in 1 L polythene containers at 4°C before analysis. Physical and chemical parameters of leachate were analyzed following standard methods. The pH, conductivity, total dissolved solids (TDS) and salinity all were analyzed by multi-parameter tester 35 series (Eutech PCSTESTR 35-01 × 441506/Oakton 35425-10). Biological oxygen demand (BOD) of the leachate was estimated after 5 day incubation following standard method (APHA, 1998). Nitrate-N was quantified using a brucine sulphuric acid method (Voghe, 1971) and phosphate-P using ammonium molybdate method (Mackereth, 1963). Chloride content was measured following Mohr's method (APHA, 1998). Leachate samples were digested in concentrated HNO_3 and the concentrations of Na, K and Ca were determined using Flame Photometer, (Systronics, India).

Statistical analysis

Standard error of difference (SE_d) was used to test the significant differences between sites. Samples were collected in replicates and all the measurements, except a few that were excluded for not following normal distribution pattern were considered in statistical analysis. Data normality was determined using the Kolmogorov-Smirnov test. Coefficient of variation (cv) with least significant difference ($\alpha = 0.05$) was computed for expressing data variability. To give an indication of uncertainty of the mean, arithmetic means are accompanied by ± SE. The SPSS package (version 16) was used for statistical analysis.

Results and Discussion

One of the major problems associated with MSW is the production of massive amount of leachate, which if not properly managed, may lead serious threat to environment. Leachate may enter the underground water and/or mixed with runoff and directly contaminate the surface waters (Munafo *et al*., 2005 and Lee *et al*.,

2006). This has relevance in the present study since the leachate emerging from the study sites could directly contaminate the Ganges, a major river system of India. In the present study, although pH of the leachate did not show a definite trend, other variables such as conductivity, TDS, salinity, BOD and concentration of Cl^-, Na^+, K^+, Ca^{2+}, NO_3^- and PO_4^{3-} all were comparatively higher at Site 1 (KZP) and lower at site 2 (BPS) (table 1). Between- site differences were significant at $p > 0.05$ for Na^+, K^+, Ca^{2+}, NO_3^-, PO_4^{3-} and BOD and at $p > 0.01$ for conductivity, TDS, salinity and Cl^- (table 1). The temporal and between site variations in leachate quality showed a pattern similar to the previous studies (Farquhar, 1989). Our observations showed a change in leachate quality overtime although the rate of change was lower than those reported for changes in the quality of leachate generated from woodwastes (Tao *et al*., 2005).

Landfill leachates are generally characterized as a high strength waste water. Leachate generated from mature landfills has typically low biochemical oxygen demand (BOD_5) indicating resistance to biodegradability (Cortez *et al*., 2011). In a previous study, the young wood waste leachate showed higher oxygen demand than the older leachates (Tao *et al*., 2005). In the present study also, the BOD_5 was higher in leachate generated at Site 1 containing greater percentage of younger wastes. The contribution of easily biodegradable fraction to total oxygen demand decreases rapidly overtime, resulting in a more recalcitrant leachate. This indicates that microbial decomposition within the landfill reduces the availability of labile organic carbon for percolation with the age of the landfill. This has relevance regulating concentrations of dissolved organic carbon (DOC) in leachate runoff generally added to down-stream Ganga River.

As water percolate through the waste, contaminants mobilize into the liquid phase through dissociation and suspension from the stationary phase, thus producing a concentrated leachate. In the present study, the low BOD_5 of older landfill leachate pool suggests that the anaerobic degradation might have predominated the degradation process producing CH_4, CO_2 and metabolic intermediates. Taylor and Carmichael (2003) reported a significant BOD decline due to storing the leachate in a catch basin under anoxic condition. The anaerobic reducing environment within the landfill causes a decrease in the concentration of nitrate as was evident in the present study.

Relatively low concentration of TDS coupled with synchronous trends in the concentrations of ions was clearly reflected through between-site differences in conductivity (table 1). The biological treatment processes rely on the establishment and activity of mixture of microorganisms and the nutrients that support their growth. Among the many factors regulating leachate treatment efficiency, aerobic biological system operate efficiently with a BOD_5 : N : P ratio of 100: 5: 1 (Forgie, 1988). In the present study, the mean BOD_5: N: P ratio was 90: 3: 1 at Site 1 and 80: 2: 0.5 at Site 2 indicating that for the treatment of leachate draining from both the study landfills, the aerobic biological systems may operate efficiently without nutrient supplement (Tao *et al*., 2005).

With respect to leachate temperature, higher temperature in summer months would improve microbial metabolism in the landfill to produce concentrated leachate. Further, the leachate quality showed a mixed effect of moisture and temperature. Month-wise trends observed in the present study did reflect such possibility as the concentrations of ion and nutrients were generally high in summer months (table 1). On the other hand, relatively higher moisture content of MSW in winter months could enhance microbial activity and leaching of ions and soluble compounds. In present study, the leachate quality showed a mixed effect of moisture and temperature. The effect of wet-winter however, was found sub-ordinate to the effect of summer temperature. A marked synchrony between temperature and concentrations of ions and nutrients in leachate confirmed the effect of climate variables on landfill leachate quality (Trankler *et al*., 2005).

Our observations showed methane emission fluxes of 16.91 to 20.11 mg m^{-2} h^{-1} at Site 1 and 10.73 to 11.14 mg m^{-2} h^{-1} at Site 2 during winter months (January-February). During summer (March-June), the CH_4 flux ranged between 20.27 and 60.20 mg m^{-2} h^{-1} at Site 1 and between 13.89 and 40.20 mg m^{-2} h^{-1} at Site 2. This suggests that Site 1 with relatively higher proportion of new waste dominated the overall CH_4 emission flux during the study period. Despite range of variabilities, the values of CH_4 emission flux obtained in the present study are lower than those reported in previous studies. For instance (Börjesson and Svensson, 1997) reported methane emission flux ranging from 0.54 to 320 mg m^{-2} h^{-1} from landfill areas in Sweden. Chen *et al*. (2008) estimated 8.8 to 163 mg m^{-2} h^{-1} from landfills of Taiwan. Jha *et al.* (2007) and Ankolkar *et al*. (2008) quantified methane emission fluxes of 1 to 433 mg m^{-2} h^{-1} and 982.8 to 5972.4 mg m^{-2} h^{-1} from landfills of Chennai and Pune respectively. Similarly, Chakraborty *et al.* (2011) estimated methane emission fluxes of 1154.3 to 3617.5 mg m^{-2} h^{-1} from three landfills of Delhi. Similar ranges of methane emission flux were reported by Rawat *et al.* (2008), Kumar *et al*. (2004) and Mor *et al*. (2006) from different landfills of India.

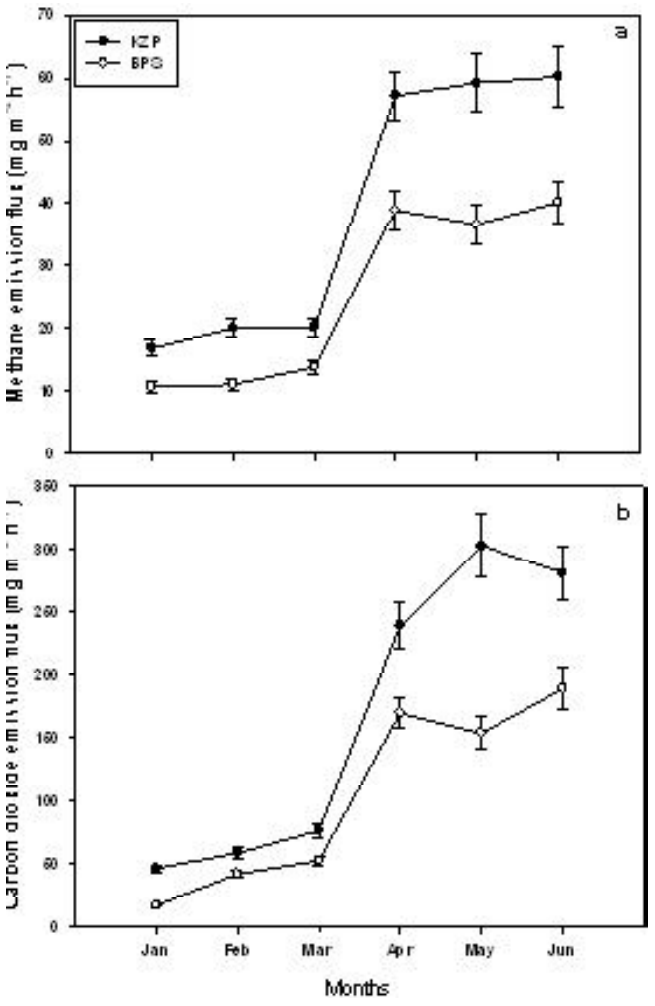

Fig. 1 : Methane (a) and carbon dioxide (b) emission flux at two landfill sites of Varanasi. Value are mean (n=5) ± 1 SE.

The decomposition of MSW is regulated by factors such as temperature, moisture, composition and age of landfill. The most conducive range of temperature for methane emission is 30 °C- 40 °C. This has relevance since this temperature range prevails in tropical countries including India for a major part of the year. However, CO_2 emission is relatively rapid for easily biodegradable materials where the rate of decomposition is faster especially at high temperature and moisture content. Accordingly, a major fraction of carbon is emitted as CO_2 rather than CH_4 (Jha *et al*., 2007). Being the cultural capital of India, a major part of the waste generated in Varanasi city comprises of biodegradable and compostable materials. Due possibly to this reason CH_4 emission flux were lower in present study compared to other observations. Although the CO_2 emission flux did override CH_4 emission flux, the CO_2 emission was relatively lower than those reported in previous studies conducted at other landfills of India (Jha *et al.,* 2007).

Table 1 : Characteristics of leachate from two landfill sites at Varanasi. Values are in mgL^{-1} except temperature (°C), pH and conductivity (iS). Between-site values not following the same letter are significantly different at $^{b}p > 0.05$; $^{c}p > 0.01$; a : not significant.

Variables	KZP						BPS					
	Jan	Feb	Mar	Apr	May	Jun	Jan	Feb	Mar	Apr	May	Jun
Temp(^{0}C)	21.4^a	25.1^a	29.6^a	39.9^a	43.6^a	48.7^a	20.8^a	23.5^a	27.9^a	38.3^a	39.4^a	42.6^b
pH	6.98^a	7.1^a	7.06^a	6.03^a	6.12^a	6.09^a	5.35^b	6.92^a	6.48^a	7.21^b	7.32^b	7.93^b
Cond (iS)	2678^a	2652^a	2870^a	4961^a	5248^a	5510^a	1294^c	1286^c	1320^c	1792^c	1731^c	1883^c
TDS (ppm)	1728^a	1794^a	1910^a	2994^a	2968^a	3810^a	784^c	732^c	820^c	1432^c	1569^c	1620^c
Sal (ppm)	994^a	986^a	1280^a	2748^a	2756^a	2872^a	598^c	608^c	632^c	1132^c	1094^c	1190^c
Cl^-(mg/l)	483.95^a	496.35^a	571.55^a	703.55^a	698.95^a	763.25^a	173.5^c	186.3^c	195.25^c	276.3^c	298.5^c	308.8^c
Na^+(ppm	81.67^a	77.49^a	83.81^a	29.46^a	31.48^a	32.48^a	54.38^c	49.02^b	56.31^b	18.34^b	18.96^b	19.56^b
K^+(ppm)	21.84^a	21.63^a	22.66^a	28.32^a	33.48^a	40.04^a	13.22^b	13.36^b	14.81^b	24.48^b	26.36^b	27.96^b
Ca^{2+}(ppm)	97.36^a	105.73^a	112.7^a	27.62^a	30.84^a	32.23^a	76.32^b	76.81^c	81.12^b	19.41^b	19.94^c	21.8^b
NO_3^-(mg/l)	2.68^a	2.77^a	2.80^a	3.98^a	3.86^a	4.08^a	1.53^b	1.55^b	1.69^b	1.98^b	1.16^b	2.44^b
PO_4^{3-}(mg/l)	0.12^a	0.07^a	0.06^a	1.69^a	1.63^a	1.76^a	0.04^b	0.05^a	0.05^a	0.65^b	0.69^b	0.75^b
BOD (mg/l)	86.32^a	73.28^a	78.92^a	102.38^a	102.46^a	136.22^a	62.91^b	64.48^b	73.52^a	86.38^b	92.94^a	96.48^c

Methane emission fluxes were relatively lower in winter months and increased sharply in summer months. Similar trends were reported from landfill sites of Chennai (Jha *et al*., 2007), Pune (Ankokar *et al.,* 2008) and Delhi (Chakraborty *et al*., 2011). Although no direct correlation between CH_4 emission flux and moisture content could be found, yet the impact of moisture content was evident during winter month. Trankler *et al*. (2005) showed that methane oxidation in tropical landfills is enhanced with a rise in the moisture content. In the present study, moisture content of both the landfills were 10-20% higher in winter months relative to summer. However, despite the influence of moisture content, the CH_4 emission flux was higher in summer months indicating the predominant influence of temperature and other factors. The CO_2 emission is relatively rapid for easily biodegradable materials where rate of decomposition is faster especially at high temperature and moisture content. Accordingly, a major fraction of carbon is emitted as CO_2 rather than CH_4. (Jha *et al*., 2007).

In addition to the between-site differences, our in-situ measurement data showed intra-site variations as reflected by high value of coefficient of variation (cv). Intra-site variations could be attributed to the age and composition of waste as well as the percentage of degradable organic carbon (Chakraborty *et al*., 2011). Although not well understood, the landfill management practices such as the type of covering materials, degree of compaction etc., could also influence within-site methane and carbon dioxide emission fluxes. In addition to other factors, wide variations in CH_4 and CO_2 emission fluxes reported from different landfills could be linked to management practices and incomparable frequencies. For instance, landfills covered with excavated soil or construction and demolition wastes generate relatively smaller quantity of methane due probably to the soil being a strong sink of atmospheric methane. Under such condition the process of oxidation transfers methane to carbon dioxide (Houghton *et al*., 1992 and Chakraborty *et al*., 2011). This has scientific relevance and could be suggested as a management practice to be initiated for mitigating methane emission unless otherwise it is translated in a usable energy source. Although more exhaustive data sets are needed, the present study that forms the first report from Varanasi landfills, will help reducing uncertainties in CH_4 and CO_2 emission scenario.

Conclusion

The landfills are becoming an increasing cause of concern pertaining to green house gas emission and release of concentrated leachate. In the present study, the BOD_5: N: P ratios of landfill leachate indicated that the aerobic biological degradation may operate successfully without nutrient supplement for leachate treatment. From human perspectives, it is arguably the Ganga River that is of greatest concern as the river receives leachate draining from the landfills. Under the prevailing state of climatic drivers, the emission of carbon dioxide from both the landfills was over five times higher than that of the methane emission. The rates were higher at young landfill site. The methane emission flux observed in the present study was lower than those reported from other landfills in India. Emission of both the green house gases (CH_4 and CO_2) appeared for more sensitive to temperature than to moisture content. Although more exhaustive data sets are needed, the present study that forms the first report from Varanasi landfills, will help reducing uncertainties in CH_4 and CO_2 emission scenario.

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