

Methane Emission and Its Variability in Different Land-uses of Semi-arid Region, Rajasthan

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Abstract: For the first time an attempt was made to quantify the methane fluxes in the different land-uses of the semi-arid region in India. Measurement of methane fluxes was carried out using static chamber in a highly saline wetland – Sambhar Lake, an urban lake – Mansagar Lake, a small reservoir – Kanota Dam and a landfill site in Jaipur district of Rajasthan, India. The results showed that semi-arid region has a significant impact on the methane budget. The parameters for water and soil were also studied and a positive correlation of soil organic matter and total organic carbon with methane fluxes were obtained. In the presence of high sulphate, methane production was observed at Kanota Dam which may be due to the symbiotic relationship between methanogens and sulphur reducing bacteria. The study revealed that the role of the semi-arid region is important for global methane budgeting and Sambhar Lake may play a significant role in methane budget with suitable conservation measures.

Keywords: Greenhouse gases; Sambhar Lake; Methane; Semi-arid region; Land use.

Introduction

In the light of a climate change and global warming, the concentration of greenhouse gases in the atmosphere become important due to their unique property of absorbing the longwave terrestrial emissions emitted from Earth surface and changes induced by them into the Earth-Atmosphere radiation balance. The global research community have considered methane as the second most important greenhouse gas after carbon dioxide with greenhouse warming potential (GWP) around 28 times greater than the GWP of carbon dioxide for 100-year time horizon and so, quantification is a subject of great interest because accurate information is required to determine its contribution to global

greenhouse gas fluxes (Allen et al., 2007; Myhre et al., 2013). The major contributors to the methane budget have been identified but large uncertainty is associated with the inventory processes (IPCC, 2001).

The soil-atmosphere CH₄ flux is the result of the balance between the two offsetting processes of methanogenesis and methanotrophy (Conrad, 1989). Several factors are known to contribute to the spatial and temporal variability of observed fluxes including soil carbon, substrate quality, temperature, moisture, soil diffusivity, microbial activity, pH, and N availability (Verchot et al., 2000). Production of methane is carried out by methanogens in anoxic environment in abundance of organic matter by using relatively simple substrates i.e. H₂+CO₂, acetate, formate, methylated

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compounds and primary and secondary alcohols whereas oxidation/consumption/uptake of methane takes place by the matrotrophs by a process methanotrophy (Le Mer and Roger, 2001). Oxygen and some electron acceptors like nitrate, sulphate etc. act as an inhibitor to the methanogenic activity (Conrad, 1989).

The production of methane is highly variable in the different climatic regions and land-use patterns. The methane fluxes were well studied and quantified in some major climatic regions/ecosystems. In the savanna ecosystem of Venezuela, the cultivated pasture and woodland acted as a weak sink, and the herbaceous and tree savanna have positive fluxes that produces methane (Castaldi et al., 2004). Mosier et al. (1997) quantified the methane fluxes in steppes of Colorado, the USA during 1992-1995 and found that the atmospheric consumption of soil is decreased due to the conversion of grassland to cropland. With significantly higher summer and autumn methane fluxes, a seasonal variability was found in the subtropical river estuary sediment dominated by gray mangrove in South East Queensland, Australia (Allen et al., 2007). The pH and temperature of soil along with the depth of the water table and the net primary productivity of the ecosystem combined with the length of dry season had an important role in the regulation of methane fluxes in the wetland ecosystem (Chang and

Yang, 2003; Sha et al., 2011). A high flux in summer and low in winter with significant correlation with soil temperature existed in wetlands of Taiwan whereas in the alpine wetland vegetation of Southwest China, a diurnal variation pattern was found with minor peak in morning after sunrise and major peak in afternoon (Chang and Yang, 2003; Chen et al., 2010). The contribution of natural wetlands were estimated in China and found that the swamp and salt marsh had very small contribution whereas the freshwater marshes, which had only 25.60% of the total wetland area and contributed to nearly 66.5% of the total estimated budget and peatland wetlands, contributed 1.7 times higher with total area half of the freshwater marshes (Ding et al., 2004). High water-filled pore spaces (WFPS) may result in enhancement of anaerobic conditions which lead to the production of methane and vice-versa, was reported by Palm et al. (2002) for Peruvian Amazon.

Here an important point is that the role of different land-uses in methane budget was studied very well around the world extensively, which include landfill (Mosher et al., 1999), peatland (Inubushi et al., 2003), urban (Zhang et al., 2016), water bodies/wetlands (Bastviken et al., 2004; Chang and Yang, 2003; Chen et al., 2010; Ding et al., 2004; Supparattanapan et al., 2009; Yang et al., 2012) and different soils (Boeckx

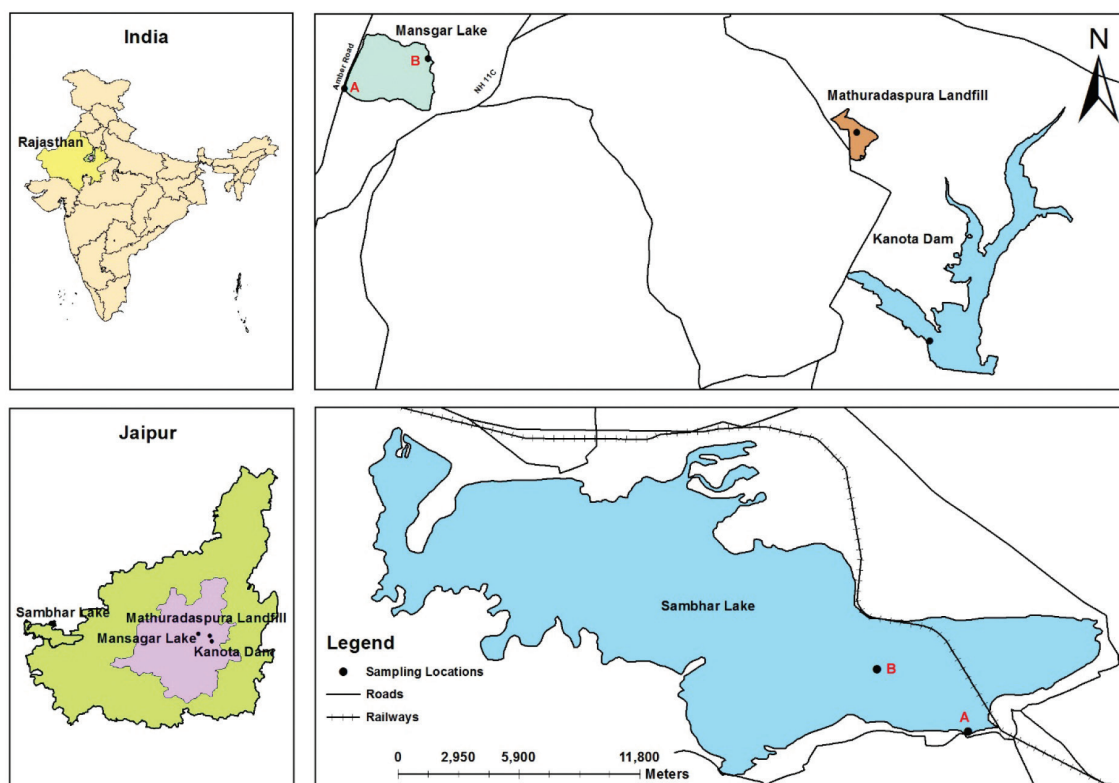


Figure 1: Map showing the study area.

et al., 1997; Castaldi et al., 2004; Ishizuka et al., 2002; Schaufler et al., 2010) but there are few studies conducted in semi-arid region (like Kaye et al., 2004; Wang et al., 2005; Wu et al., 2010) because it was considered that the role of the semi-arid region in the global budget of methane is not so important (Wang et al., 2005). In India, measurement of methane fluxes was carried out mostly in landfills (Chakraborty et al., 2011; Jha et al., 2008; Rawat et al., 2008; Rawat and Ramanathan, 2011) and few studies are available for different land-uses like coastal region (Purvaja and Ramesh, 2001) and urban wetland (Khoiyangbam et al., 2008). The role of the semi-arid region in the budget of methane is never investigated/reported in India.

So, to investigate the behaviour of the semi-arid region towards methane fluxes we carried out this study in which our objectives were to quantify the methane fluxes in different land-uses and identify the major factors which are controlling it.

Materials and Method

Study Area

Measurement of methane fluxes was carried out in the Jaipur district of Rajasthan State in India. Three sampling sites, which are Mansagar Lake, Kanota dam and Mathuradaspora landfill were located within the urban boundary of Jaipur city and the fourth site, Sambhar Lake was located at a distance of 80 km northwest from district headquarter.

The urban boundary of Jaipur (26°55'N, 75°50'E) extends from 26°38'13" North latitude to 27°15'48" North latitude and 75°27'12" East longitude to 76°58'21" East longitude. It is surrounded by the Nahargarh hills in the north and Jhalana hills in the east, parts of the Aravalli mountain chain. The elevation is highest at the northern end and covered by flat-topped hills of Nahargarh, Jaigarh, Amber, and Amargarh which are now extremely dissected and eroded. The southern end of the city is open to plains and have gentle slopes. The Jaipur urban block falls under Proterozoic Delhi Supergroup with quartzite outcrops along northern and eastern margins with the intrusion of younger granite and pegmatites. In the subsurface, the basement of the Delhi Supergroup rocks is formed by Archaean gneisses and schists. Most of the area is peneplained and covered by Quaternary alluvial sand (Pandit et al., 2009). Jaipur falls under semi-arid zone which is characterized by high temperature, low rainfall, and a mild winter. The mean temperature of Jaipur is 36° C, varying from about 14° C in December-January to about 41°C in

May-June, the coldest and hottest months respectively. However, the full temperature range of Jaipur is 45°C to 5°C. Jaipur's winter season begins in November and temperature decreases until January (generally with a minimum temperature of 8°C). The monsoon season decreases the temperature in late June or July. The average annual rainfall of Jaipur is slightly less than 600 millimetres. Ninety percent of the rainfall in Jaipur occurs during the summer monsoon season, which lasts from June to September. Ten percent of the rainfall of Jaipur is a result of winter cyclones. Overall, rainfall in Jaipur is highly variable from year to year. July and August are the rainiest months for Jaipur, as well as for the state of Rajasthan, with a relative humidity of 84% for Jaipur. The higher rainfall, which occurs during July and August, occurs in downpours that cause excessive runoff and are not too useful in recharging groundwater aquifers.

The measurement of methane and collection of soil and water samples were carried out at three locations within the urban boundary of Jaipur city. The description of all the three sites are given below.

Mansagar Lake

The Mansagar Lake (26°56'45"N, 75°51'45"E), locally known as Jalmahal is a large manmade lake on the northern fringe of Jaipur city. The lake came into existence when a dam was constructed by impounding the Darbhavati River and Nagtalai nala on the north side of the Khilangarh fortress by Maharaja Man Singh I in 1610. The total area of the lake was ~130 hectares and maximum depth of the lake was greater than 5.0 m at the time of its construction. At present, due to siltation, the depth of the lake is decreasing to 1.5-2.0 m. The catchment area of the lake is ~23.5 km² and nearly 40% of which falls in the dense urban area. The major source of water in the lake is storm water runoff of the city during the rainy season and sewage from two main wastewater drains namely Brahampuri and Nagtalai in the dry season. The remaining 60% watershed in the form of denuded and forested hills of Aravalli mountain range, virtually contributes very little on account of impediments such as human settlements and hotels in the foothills on western side and construction of roads on three sides of the lake (Raina, 2008; Sharma et al., 2008; Singh et al., 2010).

Kanota Dam

A dam was constructed on the river Dhoond/Dhundh located on the eastern outskirts of Jaipur city. The river is seasonal, a tributary of river Banas and falls under Ganga basin.

• Mathuradaspora Landfill

A landfill site, located 17 km from Jaipur city by the Delhi bypass in the north-eastern outskirts. It is a lowland area adjacent to the flood plain of a seasonal river Dhoond/Dhundh. It is one of the oldest landfill used by the Jaipur Municipal Corporation having a total area more than 46 hectare.

Sambhar Lake

Sambhar Lake (26°52'-27°2'N, 74°53'-75°13'E) is a large saline lake of Na-Cl alkaline type located about 80 km northwest of Jaipur in the Thar Desert of Rajasthan, India. The lake is situated in a closed sedimentary basin with a catchment area of ~7560 km², bounded by 500 m high Aravalli hills from northwest to west of Early and Middle Proterozoic age (Sinha and Raymahashay, 2004). The lake is elliptical in shape and occupies an area of ~225 km² at an elevation of 360 m above mean sea level, with elongation in NW-SE direction, maximum 22.5 km in length and 3.2 to 11.2 km in width (Roy et al., 2006). The bed is flat, the slope is less than 10 cm per km and shallow having average depth ~1 m and the maximum depth ~3 m (eastern part) (Sinha and Raymahashay, 2000). Geologically the lake is underlain by Pre-Cambrian basement rocks, predominantly consisting of schists, phyllites, and quartzites. The crystalline basement is overlain by Quaternary clays and silts (Aggarwal, 1951). The lake is bordered by the vegetated, stabilized sand dunes and rocks of Delhi Super Group and the tectonic origin has been linked to the pull apart structural depression due to the strike-slip faulting along curvilinear planes (Sinha-Roy, 1986). The climate of the lake is transitional in nature with sub-humid in East and semi-arid in the West. The average annual temperature is 23°C with a maximum of 45°C (Sinha and Raymahashay, 2000). It falls in the rain shadow for the southwest monsoon and receives ~550-650 mm average rainfall annually (Sinha et al., 2006). The principal source of water to the lake basin is atmospheric precipitation and inputs from seasonal streams, namely Roopangarh and Mendha from NE and SW respectively (Yadav and Sarin, 2009). Detrital minerals like analcime (NaAlSi₂O₆•H₂O) which is a product of the chemical reaction between the hypersaline brine and detrital feldspars reflects that igneous and metamorphic rocks are dominated in the catchment. The analysis of sediment reflects that the clastic fraction consists of quartz, alkali feldspar, mica, chlorite, amphibole with weathering products like kaolinite and goethite. In the non-clastic evaporite fraction, major minerals are calcite and halite. Gypsum

that is found as the major sulphate mineral below 5 m and thenardite, kieserite and polyhalite are found at the shallower horizon which indicates that the evaporite mineralogy takes a break at this depth. A change in brine chemistry observed to K-Na-CO₃-SO₄-Cl from K-Na-Ca-Mg-SO₄-Cl type gives an idea about the change in evaporative conditions of the lake. In the earlier stage, the brine underwent evaporation under the condition Ca>alkalinity and in more recent times, the evaporite mineralogy has developed with alkalinity>Ca. Mg is removed due to dolomitization of calcite and formation of Mg-clay. The presence of K-bearing evaporites in the core sediments suggests that the evaporation of brine exceeded the halite saturation stage (Roy et al., 2006; Sinha et al., 2006; Sinha and Raymahashay, 2004; Yadav, 1997).

Methane Flux Measurement

Closed chamber method is a well-established and simplified technique for the methane flux measurement which is applied successfully in different environments around the world (Chakraborty et al., 2011; Chang and Yang, 2003; Jha et al., 2008; Khoiyangbam et al., 2008; Purvaja and Ramesh, 2001; Rawat et al., 2008; Supparattanapan et al., 2009). The method involves the trapping of emitted gas from soil surface into the chamber by which the concentration of gas keeps increasing with respect to time and fluxes are determined by drawing the air samples from the chamber at defined intervals and analysis with a gas chromatograph equipped with flame ionization detector (Chakraborty et al., 2011). To maintain the homogeneity inside the chamber the air was mixed continuously with a small DC fan fitted in it. In this study, the chamber was made from 6 mm thick polyacrylic material



Figure 2: Chamber set-up at Kanota Dam site.

(having closed top and open bottom) with dimensions $31\text{ cm} \times 31\text{ cm} \times 46\text{ cm}$. The open end of the chamber was inserted 3–5 cm into the soil depending upon the site feature. At each site, the chamber was embedded a few hours in advance to ensure that the ambient soil environment was maintained. The deployment time of the set-up was 60 minutes at each site and triplicates of samples were collected at each 0, 30 and 60 minutes with 10 mL plastic syringe through the air sampling port at the top. The collected samples were injected into glass gas vials of 5 mL by displacement of deionized water. The samples were stored in a box containing ice and transported to the laboratory. The temperature of the chamber was also continuously monitored with a pre-calibrated thermometer. The gas samples were analyzed using Shimadzu GC-2010 Plus gas chromatograph equipped with a flame ionization detector. The measured methane content from the gas chromatograph is converted into methane flux ($\text{mg m}^{-2}\text{ h}^{-1}$) by using the method described by Chakraborty et al. (2011).

Soil and Water Parameters

Soil and water samples were collected adjacent to the chamber set-up in plastic zip-locks and 500 mL polypropylene sampling bottles respectively. Nitrate in the water samples was analyzed by the methodology described by Zhang and Fischer (2006). Analysis of

sulphate and chloride were carried out by Metrohm 883 Basic IC plus ion-chromatograph and salinity were calculated by the relationship $\text{salinity (ppt)} = 0.0018066 \times \text{Cl}^- (\text{mg/L})$. In soil samples, the moisture content was determined with Precisa XM60 moisture analyzer and total organic carbon was determined by using the method described by Walkley and Black (1934).

Results and Discussion

The in-situ measurement of methane which was conducted at six locations in four different land-uses revealed some interesting results. Out of the six sites, we found negative fluxes at three sites, means consumption of methane and positive fluxes at rest at three sites, i.e. production of methane. The values of methane fluxes for each site were -37.01 , -2.54 , -44.47 , 0.03 , 5898.03 and $83.89\text{ mg m}^{-2}\text{ h}^{-1}$ at Sambhar-A, Sambhar-B, Mansagar-A, Mansagar-B, Landfill and Kanota Dam respectively. It is evident from the results that the Landfill site dominated the other sites in terms of production and Sambhar-A site for consumption.

The flux rates that were obtained during different studies conducted in India for landfills are presented in the table and it shows that the value for methane flux that we found is higher than all of them. The production of methane is highly dependent on the characteristic of waste in case of landfills and the landfill at which the

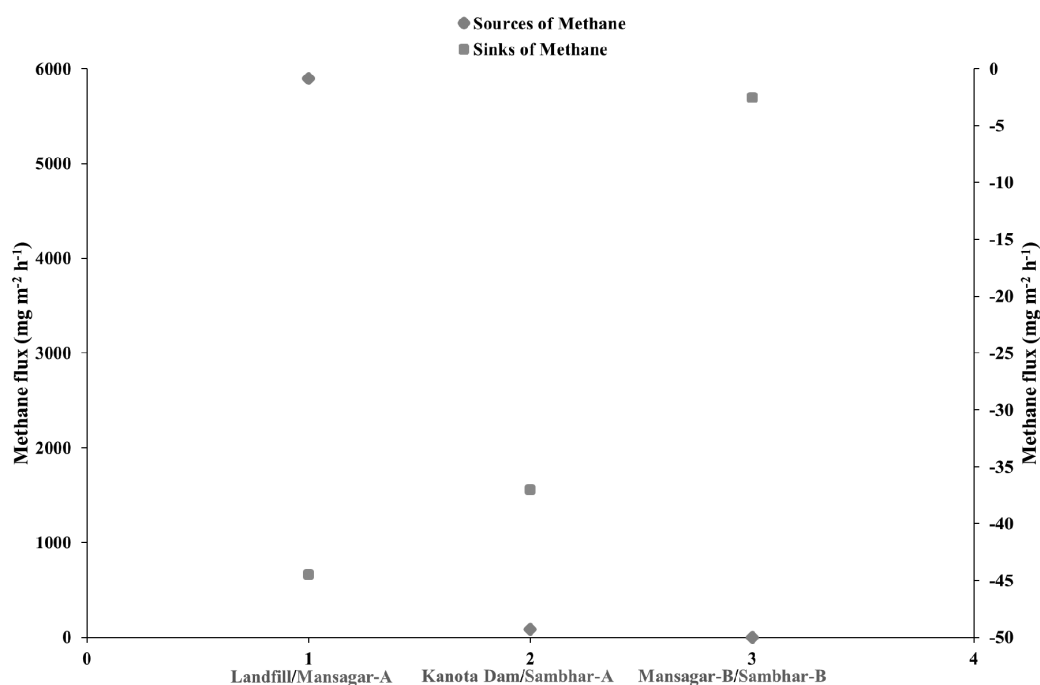


Figure 3: Results of methane flux measurement from all sampling locations.

sampling was carried out, had received a large amount of organic waste from the nearby meat industry during past few days. The high organic content was reflected in the methane flux obtained at that site.

The negative fluxes of methane were observed at two sampling locations: one of Sambhar Lake and one sampling location in Mansagar Lake. The oxidation/consumption/uptake of methane were reported in different studies in different parts of the world and major factors which were responsible for it were pH, salinity, nitrate, sulphur, soil texture and moisture etc. (Ishizuka et al., 2002; Joye et al., 1999; Kaye et al., 2004; Mosier et al., 1997; Palm et al., 2002; Sha et al., 2011; Verchot et al., 2000; Wang et al., 2005; Wu et al., 2010; Yang et al., 2012; Zhang et al., 2016). In Sambhar Lake, we found a high concentration of sulphate and nitrate and a possible reason for the enrichment of sulphur is geochemistry of the region, whereas for nitrate, the contribution of runoff from various human activities like agricultural, is more dominated over the other factors like synthesis by algae, due to the large catchment area.

According to Conrad (1989), the activity of methanogens are inhibited in presence of electron acceptors like sulphate, nitrate etc. and negative relationships with salinity and methane flux have been reported (Poffenbarger et al., 2011; Purvaja and Ramesh, 2001; Supparattanapan et al., 2009). Due to the presence of huge amount of sulphate, nitrate along with the high salinity, the balance was shifted towards the consumption and fluxes became negative. The Sambhar-B site was completely dry while Sambhar-A site was located adjunct to the shallow saline water which accelerated the methanotrophs activities and methane flux becomes more pronounced than Sambhar-B. In Mansagar Lake, we were expecting positive flux for the methane at Mansagar-A, but were surprised with the results. The sediment generated from the eroded hills that surround the lake from three sides played a major role in siltation and resulted into loss of the depth (Raina, 2008). When

the lake was deep and stratified the anaerobic condition prevailed but with decreased depth, stratification of lake was lost and the interaction of atmosphere-water-soil increased which resulted in oxygen rich environment or oxic conditions (Bastviken et al., 2004). The interaction process was accelerated by the topography and climatic condition of the region. Another factor which had important contribution was the location of sampling point. The sampling was performed near a sewage inlet and with an increase in turbulence, the aeration of water increased significantly.

The two different mechanism which are mentioned above contributed significantly to more consumption than the production of methane in oxic environment and negative flux was observed (Le Mer and Roger, 2001; Topp and Hanson, 1991). At the second sampling location on the same lake (Mansagar-B), a very low positive flux was observed. The reasons behind the phenomenon were the prevailing aerobic conditions because the circulation, as well as mixing of water, was hindered by a human-made ridge-like structure. Another possible factor was the contribution of organic matter by the dense vegetation cover present at the east side of the lake. The behaviour of Kanota Dam was slightly different from Mansagar Lake. The reservoir was deeper and organic matter content, sulphate concentration was also higher than Mansagar Lake. A positive flux means the production of methane was observed despite the presence of high concentration of sulphate. So, Kanota Dam emerges as a typical example of the coexistence of anaerobic and sulphur reducing bacteria (Oremland and King, 1988; Purvaja and Ramesh, 2001). King et al. (1983) explained that for compounds like methanol, methylated amines (trimethylamine), and dimethyl sulphide (DMS), the sulphate reducers do not have a strong affinity as with H_2 and acetate, and termed as non-competitive substrates which act as an important precursor for methane. Thus, the emission characteristics

Table 1: Methane flux measurement conducted in India

<i>Methane Flux ($mg\ m^2\ h^{-1}$)</i>	<i>Place</i>	<i>Reference</i>
12.94-293.33	Landfills of New Delhi	Rewat and Ramanathan, 2011
0.9-433.0	Landfill of Chennai	Jha et al., 2008
22.00-637.00	Landfill of Ahmedabad, Bangalore, Chennai, Dehradun, Kolkata and Delhi	Rewat et al., 2008
734.0-2609.6	Landfills in New Delhi	Chakraborty et al., 2011
3.10-21.56	Coastal Wetlands of South India	Purvaja and Ramesh, 2001
12.17-21.25	Urban Wetlands in Jhansi, Uttar Pradesh	Khoiyangbam et al., 2008

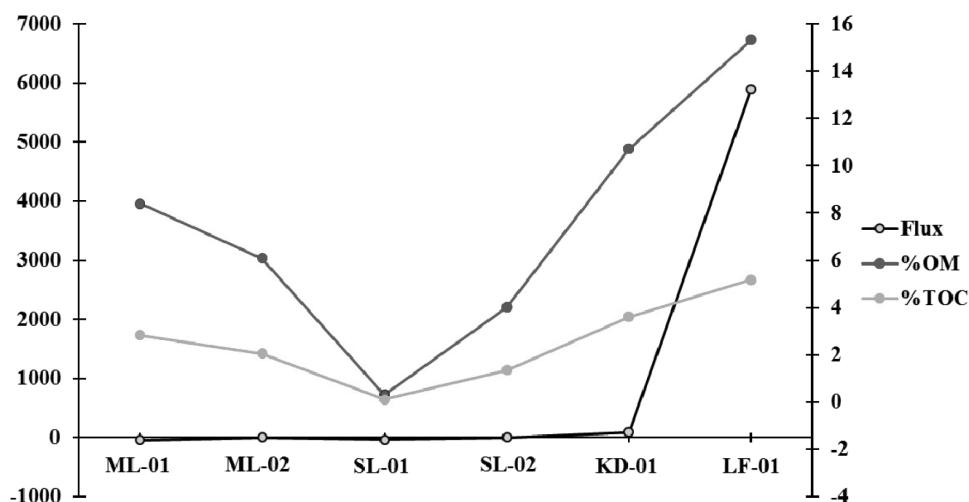


Figure 4: Relation between total organic carbon and organic matter with methane flux.

observed at Kanota Dam seem to be supported by the symbiotic relationship between methanogens and sulphur reducing bacteria, instead of the competitive relationship.

In this study, the number of samples was less so the trend between various parameters of soil and water with methane fluxes were not clearly obtained. A positive correlation between organic matter and total organic carbon with methane flux was observed in general for all the land-uses, when taken together and justified the relationship given in the various studies (Poffenbarger et al., 2011; Purvaja and Ramesh, 2001).

Conclusion

It is a preliminary study conducted to check the role of semi-arid in the methane budget and we found some interesting results i.e. the consumption of methane at Sambhar Lake and production of methane in presence of high sulphate concentration at Kanota Dam site. The behaviour of Mansagar Lake added more surprise because we found two opposite characteristics at two different sampling locations in the same water body. The result obtained at the landfill site is very important to fill the gaps in the inventories for methane because no study was conducted in this region. The most important finding of this study is that a large variation in the behaviour of three water bodies was found based on their characteristics which are different from each other and these findings can be used to improve the inventory process by filling the gaps in the categorization of land-uses for identification. After these initial findings, in next stage, we need more spatial data by adding more

sampling points and temporal data, by excising the sampling tasks in each month of a year or at least in each season, to draw a clear picture about the behaviour of different land-use patterns in this region.

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