



Urban Environmental Pollution 2013 – Creating Healthy, Livable Cities

## Atmospheric methane mixing ratio in a south Indian coastal city interlaced by wetlands

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### Abstract

Industrialisation and urbanization leads to an increase in concentration of greenhouse gases, which eventually alters the radiation balance of the climate system. Urban regions are hotspots of greenhouse gas emissions which include CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, etc. Methane emitting sources hosted by cities include fossil fuel combustion, municipal waste and sewage management, blocked drains and pools etc. Waste discharges from the residences, food wastes, market places etc., contribute to the methane production. Urban heat island causing warm nights in the city is also a suitable condition for the generation of methane.

Ground level mixing ratio of methane in the tropical coastal city of Cochin in South India, during calm early morning periods was measured in this study. A mobile traverse method was employed from January 2011 to March 2013. Measurements were taken during both winter and summer seasons. It was observed that the ground level methane concentrations were significantly higher than the global average value. Intra-city variation in ground level mixing ratio was also significant. The maximum value of ground level methane in winter and summer were 3.85 ppm and 3.21 ppm respectively. The study reveals that the city acts as a source of atmospheric methane.

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## 1. Introduction

Greenhouse gases play a crucial role in the thermodynamics of the atmosphere. They trap radiation emitted by the earth, thereby producing the greenhouse effect. The global mean concentration of the three most important greenhouse gases, CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O in 2012 stood at 393 ppm, 1.82 ppm and 325.1 ppb respectively [1]. Methane (CH<sub>4</sub>) is the second largest contributor among greenhouse gases to global warming through radiative forcing of the Earth's climate system [3,4]. The global mean concentration of methane is determined by a variety of terrestrial and atmospheric processes. Methane is released into the atmosphere by a wide number of sources, both natural and anthropogenic [5,6]. About 40% of atmospheric methane originates naturally from wetlands and termites and the remaining 60% comes from anthropogenic sources like burning of fossil fuels, rice cultivation, ruminant animals, landfills and urban regions. Urban regions are now considered as a major source of methane [7, 8, 9]. The World Meteorological Organisation (WMO) Greenhouse Gas Bulletin states that the increase in global average methane levels was associated with increased emissions in the tropics and mid-latitudes [1,2]. The atmospheric methane concentration has increased from 0.7 ppmV to a global average of 1.82 ppmV, over the last 300 years. The 157% increase in the atmospheric methane concentration since 1750 to the present is attributable mainly to increasing emissions from anthropogenic sources. Though the current atmospheric concentration of CH<sub>4</sub> is significantly lower than that of CO<sub>2</sub>, its large Global Warming Potential, estimated as 21 times as that of carbon dioxide, gives it a significant climatic importance. Methane contributes 18.5% of direct radiative forcing due to the long lived greenhouse gases affected by human activities.

It is reported that land-use changes have decreased natural emissions by only approximately 10 per cent [10]. It is also reported in 2006 that CH<sub>4</sub> emissions to the atmosphere from wetlands are approximately 150 Tg CH<sub>4</sub> yr<sup>-1</sup> [11]. Emissions of CH<sub>4</sub> from wetlands vary significantly from year to year depending on temperature and precipitation. Other natural sources include termites, oceans (approx. 20 Tg CH<sub>4</sub> yr<sup>-1</sup> each), wild fires and wild ruminant animals (approx. 5–10 Tg CH<sub>4</sub> yr<sup>-1</sup> each). Based on a pre-industrial CH<sub>4</sub> atmospheric mole fraction of approximately 700 ppb, total pre-industrial emissions were approximately 215 Tg CH<sub>4</sub> yr<sup>-1</sup>[12]. The increase in the atmospheric CH<sub>4</sub> from pre-industrial to modern times is the result of increased emissions from fossil fuel exploitation, agriculture, waste management and anthropogenic biomass burning. Total emissions from fossil fuel are estimated at approximately 110 Tg CH<sub>4</sub> yr<sup>-1</sup>. Major agricultural sources are rice cultivation (approx. 30–40 Tg CH<sub>4</sub> yr<sup>-1</sup>) and ruminant animals (approx. 80–90 Tg CH<sub>4</sub> yr<sup>-1</sup>). The emission from biomass burning is initiated by humans for agriculture are estimated to be 45–55 Tg CH<sub>4</sub> yr<sup>-1</sup>. Waste related sources are decomposition of biodegradable municipal solid waste in landfills and animal and human waste streams (approx. 50–60 Tg CH<sub>4</sub> yr<sup>-1</sup>) [13]. Emission from municipal waste and biomass burning causes rise in atmospheric methane, even though the emission from natural sources decreased.

There has been a growing interest in greenhouse gas emissions from urban regions across the globe. Reports of methane concentrations within different urban areas around the globe show a significant variability in ground level methane concentrations, both in terms of inter-city and intra-city scales. There have also been a few measurements

from aircraft platforms. The present study is focused on investigating the seasonal variation of ground level methane mixing ratio in an urban centre interlaced by wetlands located on the southwest coast of India. The present paper also investigated the spatial variation of ground level methane concentration in different parts of the city including industrial areas, wetlands, markets, and the city centre.

## 2. Study Area

Kochi, one of the fast growing urban centres located on the southwest coast of India between  $09^{\circ} 45' N$  and  $10^{\circ} 20' N$  latitude and between  $76^{\circ} 10' E$  and  $76^{\circ} 35' E$  longitude, hosts a number of industries and a population of 2.2 million [14]. The city is interlaced by estuaries fed by perennial rivers. Major industrial units include an oil refinery, a fertilizer factory, and a number of chemical industries. Though some parts of the city are served by a centralized sewage treatment system, most of the city has independent domestic units. Solid waste disposal is mostly through open dumps. The water table is shallow resulting in high soil moisture. Soil in the coastal regions is mostly 'loamy sand'. Soil in the eastern regions of the study area is 'gravelly clay' on the top layers, and 'laterite' underneath. The average altitude towards the eastern fringes is about 7.5 m above MSL, and towards the west the altitude is less than one metre on an average. The whole of the land slopes gradually from east to west.

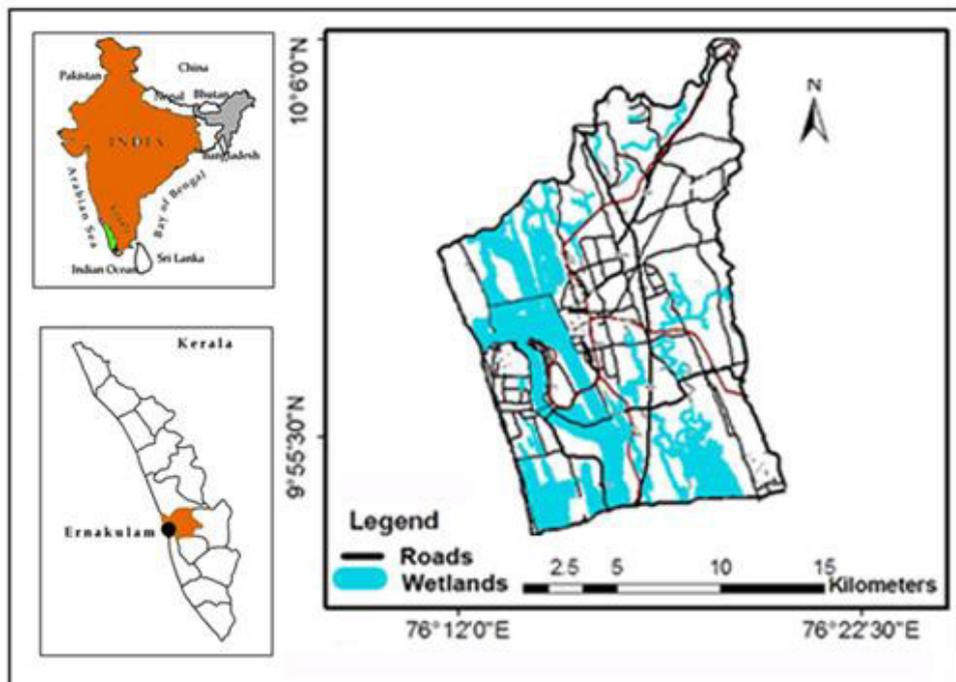


Fig.1. Sketch map of the study area.

Kochi features a tropical monsoon climate. Its proximity to the equator along with its coastal location results in little seasonal temperature variation, with moderate to high levels of humidity. Air temperatures range between 20 – 35 °C (68–95 °F). The highest temperature recorded in Kochi is 38 °C and the lowest is 17 °C. The average annual rainfall is about 3500 mm with an average 132 rainy days annually; the bulk of the rainfall is from the South-West monsoon. The winds are moderate, with slight increase during summer and the monsoon seasons. A sketch map of the study area is shown in the Fig.I.

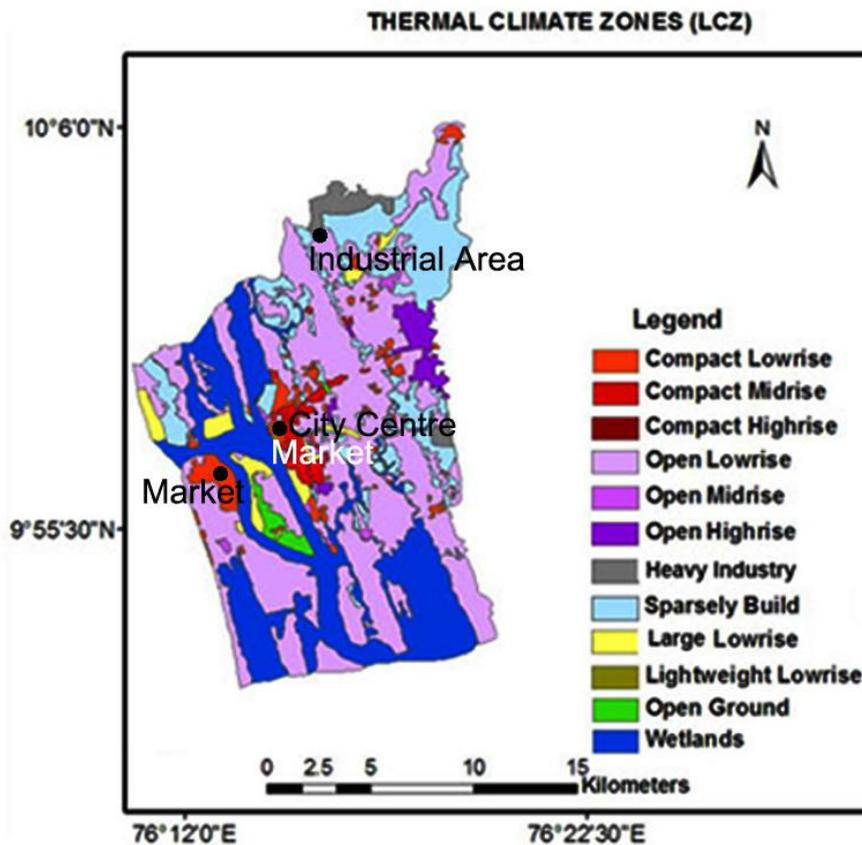


Fig.2. Thermal climate zone classification of Kochi

Local climate zones (LCZ) are defined as regions of uniform surface-air temperature distribution at horizontal scales of  $10^2$  to  $10^4$  metres [15]. Each LCZ exhibits a characteristic geometry and land cover that generates a unique surface-temperature climate under calm, clear skies. The classification is mainly based on proportion of sky hemisphere visible from the ground, building mean height, street width, vegetative fraction, anthropogenic heat flux etc. Thermal differentiation between the classes is mainly controlled by these factors. Based on these, Stewart and Oke (2012) classifies the urban –rural build up into a hierarchy of 10 climate zones, namely Compact High - rise (CHR), Compact Midrise (CMR), Compact Low - rise (CLR), Open High - rise (OHR), Open Midrise (OMR), Open

Low - rise (OLR), Large Low - rise (LLR), Lightweight Low - rise (LWLR), Heavy Industry (HI), Sparsely Built (SB) and land cover types to dense trees, scattered trees bush, scrub, low plants, bare rock and paved, bare soil and sand etc [16]. Large, open water bodies such as seas and lakes, or small bodies such as rivers, reservoirs, and lagoons also come under land cover system. Compact zones in the classifications have compact and dense buildings with high rate of human activities. Open set and sparsely build zones are characterized by detached buildings separated by natural surfaces. Classified image of the study area is shown in the Fig.2.

### 3. Methodology

Ground level mixing ratio of methane in the tropical coastal city of Kochi in South India, during calm early morning periods was measured. Sampling was carried out before sunrise to avoid the effects of ground heating and vertical mixing of air, as well as disturbance from vehicular traffic. Air samples were drawn into gas tight syringes, at screen height (1.2 m above ground), and sampling location and time recorded. A handheld GPS (Garmin 76S) was used to obtain the position of the sampling location. The traverses were mostly along roads chosen out of the city network to obtain uniform spacing between sampling locations. Samples were taken from approx. 40 locations in each survey. The distance between sampling locations were of the order of 0.5 to 1.0 km, and selected to cover different land cover and land use categories in terms of residences, markets, building height, etc. The syringes were closed and slightly pressurized by compressing the piston of the syringe to ensure a small positive pressure inside to prevent any unexpected leakage of ambient air into the syringe. Air temperature was also recorded with a high resolution RTD probe (MadgTech USA). Automatic temperature recorders with 0.01 K resolutions and 0.1 K accuracy were used for reading air temperature. These measurements were taken during both winter and summer seasons, during 2011 to 2013.

Samples stored as above were brought to the laboratory and analysed on a Flame Ionisation Detector (FID) in Gas Chromatograph (NUCON 5765). Isothermal separation in a packed 2 m long (PORAPAK Q) 80/100 mesh column was done at 70° C, with 5N purity Nitrogen gas as carrier. FID was calibrated with standard gas mixtures. The systematic uncertainty in methane estimation was  $\pm 0.357$  ppm. The use of gas chromatography gives higher accuracy to the measurements, especially since the range of methane concentration expected to encounter is the range 1.0 to 3.0 ppmV only. Contour plot of ground level methane mixing ratios were prepared. The measurements were repeated during different seasons.

### 4. Results and discussion

It is observed that the ground level methane concentrations were significantly higher than global average value. Intra-city variation in ground level mixing ratio was also significant. The maximum value of ground level methane in winter and summer during the entire survey were 3.89 ppmV and 3.21ppmV respectively. It is observed that the maximum value of ground level methane was always higher than the global average of 1.9 ppmV. Fig.3 and Fig.4 show the spacial distribution of ground level methane during calm, dry winter and summer mornings respectively.

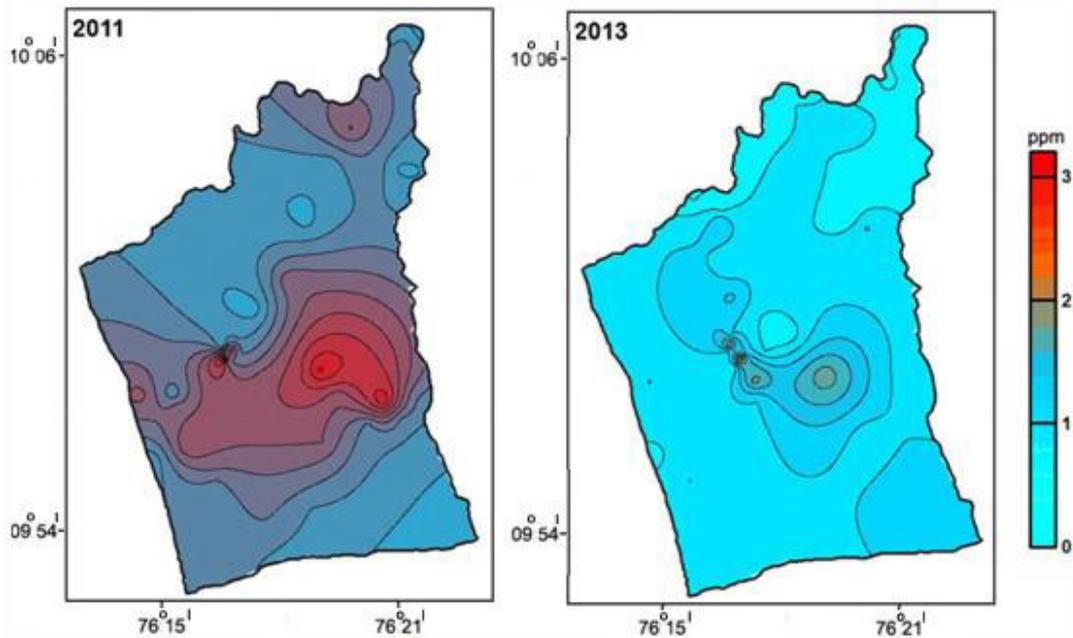


Fig.3. spatial distribution of ground level methane during winter mornings during the years 2011 and 2013.

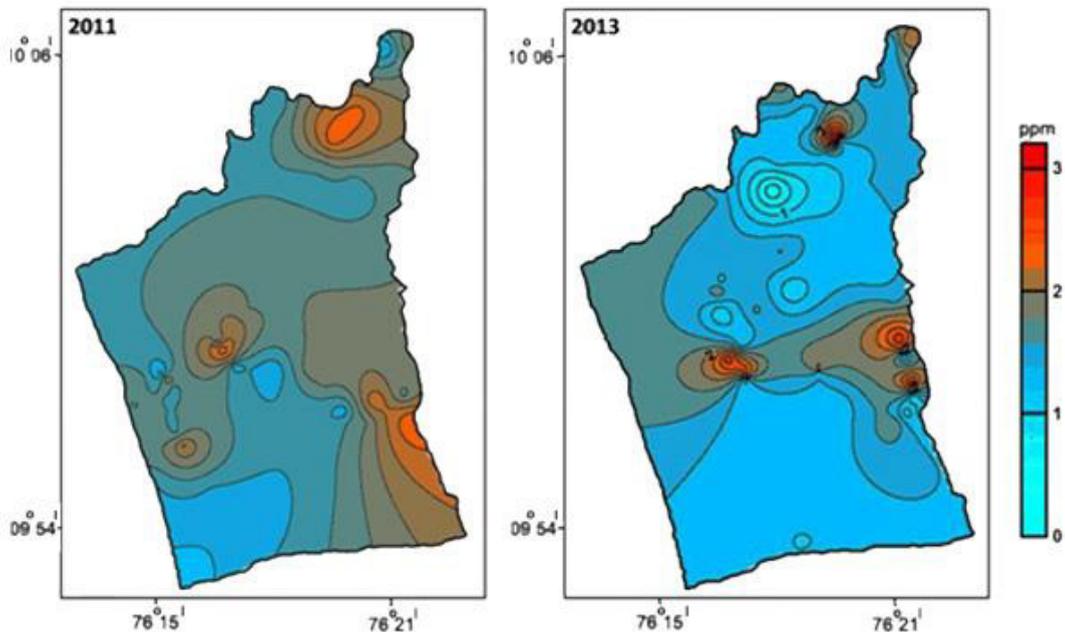


Fig 4. Spatial distribution of ground level methane during summer mornings during the years 2011 and 2013.

Ground level methane concentration varies across different parts of the city. Industrial areas, wetlands, markets, and city centre are regions where higher methane concentration is observed. Wetlands and canal networks here are significant sources of methane emission. City centre comes under compact mid rise zone in the Local Climate Zone classification showed higher concentration of methane in all observations. Higher concentration of methane is also

observed near two markets which fall under compact low rise zone in the classification. These two major markets are potential methane emitting sources in the region. Stagnant water bodies adjoining to the market area play an important role in the formation of methane. The water here is nearly stagnant since the entire city is close to sea level. Discharges from residences, eateries, market places, etc., are rich in biological waste are also contribute to methane production in this regions. Wetlands play a major role in the production of methane in sparsely build zone and open set zones. Table.1 gives a summary of measured ground level methane concentrations over entire area during early morning stable atmospheric conditions.

**Table.1** Ground level methane concentration (ppmV) during summer and winter in three consecutive years

	WINTER 2011	SUMMER 2011	WINTER 2012	SUMMER 2012	WINTER 2013	SUMMER 2013
<b>MAX</b>	3.89	3.21	2.45	2.57	2.67	3.43
<b>MIN</b>	2.09	1.71	1.61	0.86	1.17	0.41
<b>AVERAGE</b>	2.67	2.33	1.87	1.30	1.54	1.60
<b>MEDIAN</b>	2.62	2.30	1.78	1.17	1.49	1.45
<b>STDEV</b>	0.44	0.41	0.22	0.43	0.29	0.57

The first half of the year 2013 during which the above observations were made experienced lower rainfall than the corresponding period in 2011 and hence was relatively drier. The drier soil and waste dumps could be the reason for the generally lower CH<sub>4</sub> concentrations in 2013 compared to that in 2011.

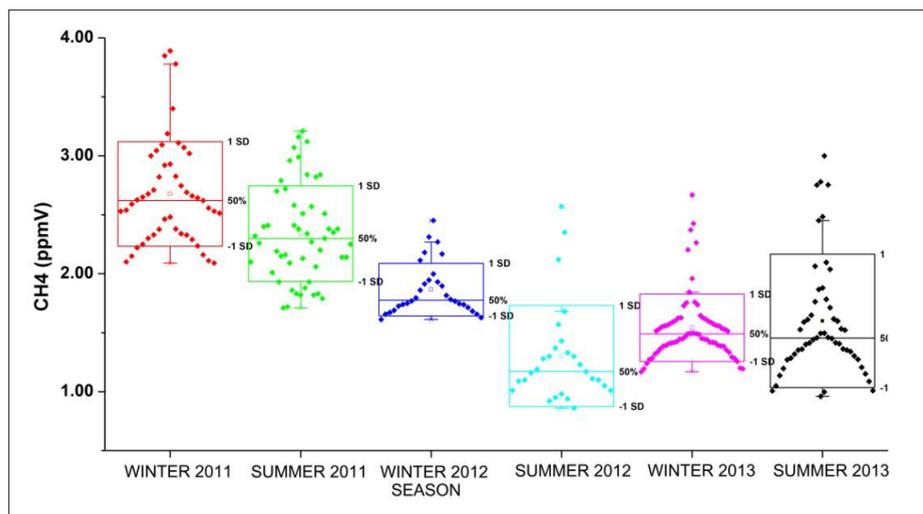


Fig. 5. Ground level methane concentrations during different seasons.

The spread diagram of methane concentration values is shown in the Figure 5. Box plot indicates that the average concentration in summer is slightly lower compared to respective winter in 2011 and 2012. Summer season in the region precedes the monsoon rainfall. Occasional pre-monsoon showers modified this pattern in 2013. However, methane concentration in the city centre remain relatively constant during both seasons. Thomas and Zachariah [17] have, in a study of the urban heat island in Kochi, plotted the proportion of water cover in this study

area. It is reported here that the water cover is as high as 70 percent in some parts of the area. These wetlands receive the storm water drains from the city which could also be carrying biological matter. This also produces a conducive environment for methanogenesis and could be one reason for the relatively high methane mixing ratio observed. It is also reported that the winter Urban Heat Island effect is higher than summer intensity. High soil moisture and higher temperature in the city centre serve as a suitable situation for the generation of methane in winter. Wastes originating from urban, industrial and rural areas have different methanogenic potential, and are categorised [18]. The major sources of methane in cities are industries, sewages, vehicular emissions, solid waste etc. Discharges from residences, eateries, market places etc., are rich in biological waste and hence contribute to methane production. The handling and disposal of municipal waste is a growing concern as the global volume of waste increases continuously [19, 20]. Increased urbanization, accelerated industrial growth, and increased pressure on waste disposal systems are important factors needing attention in this context [21].

Major factors that influence the production of methane are soil moisture, soil temperature and air temperature etc. Air temperature values for the survey days (during the mobile traverse time) in both seasons are shown in the Table 2. However no correlation was observed between the concentration of methane and the instantaneous temperature of the measuring point. Correlation coefficient varies from 0.21, -0.35, -0.48 during winter season and -0.34, -0.71 and 0.07 during summer season in 2011, 2012 and 2013 respectively. The average temperature for both seasons lies between 24 °C and 28 °C in early morning. Meanwhile the temperature in the urban area is comparatively higher than this in the night time, which is considered to be the optimum temperature for the methane production.

**Table.2** Air temperature (°C) for the survey day (during the mobile traverse time) in three consecutive years.

	WINTER 2011	SUMMER 2011	WINTER 2012	SUMMER 2012	WINTER 2013	SUMMER 2013
<b>MAX</b>	25.90	28.50	27.50	29.50	26.30	29.10
<b>MIN</b>	20.70	26.00	24.70	26.60	23.70	24.90
<b>AVERAGE</b>	23.75	27.00	25.83	28.13	24.70	26.45

The spatial and temporal behavior of atmospheric methane (CH<sub>4</sub>) in the Nagoya metropolitan area reported a higher value in the urban area [22]. The large and active landfills are the major anthropogenic CH<sub>4</sub> sources and are located at the hill sites in the northeast region is the major source of methane in Nagoya. It was considered that the air mass with the high concentration of CH<sub>4</sub> flowed from the landfill sites into the urban area, and exerted substantial influences on the spatial and temporal variations of atmospheric CH<sub>4</sub> concentrations in the central city area. Ground level methane concentration at Thiruvananthapuram, another coastal city in India about 200 km from the present study area Kochi, were reported with a maximum value of 3.16 ppmV, and was higher than the background values. It was seen in this study that the vertical profile of CH<sub>4</sub> concentration within the urban canopy layer was neutral during stable pre-dawn conditions. It is also observed that the spatial distribution of the ambient

methane exhibits sensitivity to urban environment [23]. Urban influences in the atmospheric concentration of other trace gases have also been detected. Urban signatures in tropical ozone column products derived from satellite measurements has been reported [24]. Methane concentration in the urban environment over Delhi has been reported as varying from 1.75 to 9.5 ppmV, with an average value of  $4.121 \pm 0.354$  ppmV [25].

High atmospheric concentrations of methane in air samples from urban locations compared to those from non-urban locations at the same latitude have been reported by BlakeVan et al [26]. This “urban excess”, i.e. urban concentration minus remote concentration, was 1000 to 2000 times larger on a molar basis. Continuous measurement of methane made in Cincinnati and Los Angeles (USA) showed diurnal patterns of concentrations of methane and non-methane hydrocarbons in the atmosphere. Average hourly values for methane were 2.6 and 2.4 ppm, in Los Angeles and in Cincinnati respectively [27]. Shorter et al [28] have carried out methane concentration measurements in Germany and have reported values between 1800 and 2600 ppb in Wurzen and 1990 ppb in Leipzig. Kuc et al [29] showed that the CH<sub>4</sub> level in Krakow (Poland) urban areas is between 1650 ppb and 4200 ppb. Ito and co-workers [30] compared the atmospheric CH<sub>4</sub> concentrations recorded in Nagoya with the values measured at Mauna Loa Observatory in Hawaii (USA) and estimated that the excess concentration of CH<sub>4</sub> in the urban atmosphere of Nagoya was 170 ppb in 1988 and 150 ppb in 1997. Nguyen et al [31] have reported CH<sub>4</sub> concentrations of  $2.24 \pm 0.42$  ppm by urban road-side and  $2.06 \pm 0.31$  ppm in the urban background during 1996 to 2006 in Seoul (South Korea). Wang et al [32] reported 1.9 - 3.7 ppm methane concentration in sub-urban area, Taiwan. Smith et al [33] have reported 7.971 ppm methane concentration in Mexico City in the morning. The corresponding values for night and mid-day were 1.8 ppm and 2.001 - 2.999 ppm.

Waste water treatment plants in urban areas are an important source of methane emissions. The methane emission related to the anaerobic digestion of primary and secondary sludge counts for about three quarters with respect to the waste water treatment plants overall methane emission and causes a slightly larger greenhouse gas footprint than the carbon dioxide emission that is avoided by using the resulting biogas for energy generation [34]. It is reported [35] that the methane uptake capacity of urban forests and lawns is significantly lower than that of their rural counterparts. While rural forests had a high capacity for CH<sub>4</sub> uptake ( $1.68 \text{ mg m}^{-2} \text{ day}^{-1}$ ), this capacity reduced to  $0.23 \text{ mg m}^{-2} \text{ day}^{-1}$  in urban forests and almost completely disappeared in lawns. Possible mechanisms suggested for these reductions include increases in atmospheric N deposition and CO<sub>2</sub> levels, fertilization of lawns, and alteration of soil physical conditions that influence diffusion.

The observed mixing ratios for CH<sub>4</sub> near the Mt. Wilson (MW) Observatory in southern California (USA) ranged from 1.76 to 2.16 ppm [36]. Wunch et al [37] have carried out measurements in the Los Angeles region which show that urban emissions are a significant source of CH<sub>4</sub>, substantially higher than current estimates. These findings suggest that urban emissions could contribute 7–15% to the global anthropogenic budget of methane.

## 5. Conclusion

Urban regions are considered to be a major source of methane. Waste originated from urban areas and industrial areas under suitable conditions emit large amounts of methane to the atmosphere. The maximum value of ground level methane concentration in Kochi (India) urban area during winter was recorded at 3.89 ppmV and the corresponding value in summer was 3.21ppmV. The maximum value of ground level methane was always higher than the global average of 1.9 ppmV. Considering that this values was observed in free air conditions and that methane is a lighter than air gas, this high value indicates strong local emissions. A higher concentration of methane was observed in the city centre, market areas and industrial areas. Open waste dumps, sewers, stagnant water in canals, emission from vehicles, etc., are sources of methane in the urban environment.

It was observed that the methane concentration in the free air in the urban area was significantly higher than the global average values reported as well as the background values observed in adjoining suburban regions of the study area. This indicates strong local methane sources. It was also observed that the ground level concentration is higher in localities like industrial area, market places, waste disposal sites, wetlands, etc.

A major portion of the study area was covered by wetlands, which play a major role in the generation of methane in this region. Correlation between methane concentration and temperature was not evident in the measured data, even though temperature is generally understood to be an influencing parameter. Two factors that could be responsible for this are the variability in other influencing parameters mentioned above, and the already warm tropical climate keeping methanogenesis high. This should be the focus of future studies.

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