



## Design and Implementation of an Instrument for Methane (CH<sub>4</sub>) and Nitrous Oxide (N<sub>2</sub>O) Measurements

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### Article Information

#### Keywords:

Greenhouse gases, semiconductor sensors, dump site, control site

Received 15 August 2019

Revised 30 August 2019

Accepted 4 September 2019

Available online 16 Sept. 2019

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### Abstract

Methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) are noxious greenhouse gases. They are the two most important greenhouse gases after carbon dioxide. Their production and release to the atmosphere are largely due to human activities. To curtail the buildup and the inadvertent greenhouse effects of these gases, their concentration contribution from the different compartments needs to be ascertained and well understood. Versatile instruments that can track the atmospheric concentration of these gases are needed before any meaningful investigation can take place. To fill this gap, an instrument for measuring methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O) gas concentrations as well as ambient temperature and relative humidity have been constructed. Semiconductor sensors with high sensitivity to their respective target gases were utilized. The specification of the instrument includes immediate result display and storage for subsequent data analysis. The instrument was tested at different locations with the propensity for the emission of the target gases. Contrary to expectation, the obtained results showed that CH<sub>4</sub> emission was much more at the market location than at a gas filling station. The trend between the obtained results at other locations compared with the one from the control site showed that the data obtained with the instrument is reliable.

## 1. Introduction

Without the existence of some natural “greenhouse effects”, temperature would have been about 33°C (60°F) lower than they are now and existence on earth would have been impossible [1]. Unfortunately, the continuous increase of greenhouse gases (CH<sub>4</sub>, N<sub>2</sub>O, CO<sub>2</sub> e.t.c) in the atmosphere has led to increase in the absorption of energy by the Earth and this has advertently led to climate change such as the warming of the earth’s surface beyond what is expected [2]. Atmospheric CH<sub>4</sub> has increased by about 1,000 ppb since the beginning of the industrial era, representing the fastest change in the atmospheric concentration of the gas over the last 80 years. The global average abundance of CH<sub>4</sub> at the hemispheres has reached about  $1,774.62 \pm 1.22$  ppb, which has doubled its pre-industrial value. Since 1998, the atmospheric N<sub>2</sub>O levels have risen steadily, reaching about  $319 \pm 0.12$  ppb in 2005 [3].

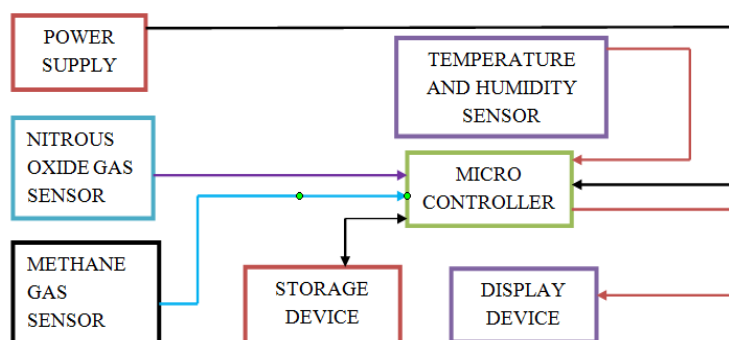
Various human activities such as incomplete combustion of fossil fuels [4], microbial activities at refuse dumps [5] and agricultural practices have substantially added to the amount of greenhouse gases in the atmosphere. The added gases have enhanced the natural greenhouse effect, thereby leading to increase in global average temperature and its related climate changes [6]. In order to address any greenhouse gas emissions issue, it is important to identify all emissions sources. The effect and environmental consequence of methane and nitrous oxide is immense. The residence times of CH<sub>4</sub> and N<sub>2</sub>O in the

atmosphere are 12yrs and 120yrs respectively. Methane is a precursor to CO<sub>2</sub> (another potent greenhouse gas) formation. It can reduce the oxygen content available for respiration in the atmosphere. High concentration of N<sub>2</sub>O on its parts can contribute to heart and lung problems.

Several international collaborations are on-going to intensify efforts at reducing Green House Gas (GHG) emissions. One of such was the United Nation Framework Convention on Climate Condition (UNFCCC) inaugurated on June 4, 1992 and ratified on March 21, 1994. While the developed nations are deploying huge funds and conducting the necessary research geared at mitigating the problem, most developing and under-developed nations are not doing well due to their meager resources. The in-situ GHG data collection and analysis in Nigeria is scarce. This is largely due to lack of the requisite instruments and the political will on the part of government. To control or mitigate GHG emission problems, useful data need to be collected from potential high-risk areas. Unfortunately, the unavailability of affordable measuring equipment in this regard is a deterrent factor regarding this laudable concern. Thus, the focus of this work is to develop a low-cost instrument for in-situ measurement of CH<sub>4</sub> and N<sub>2</sub>O concentration in the atmosphere.

## 2. Materials and Method

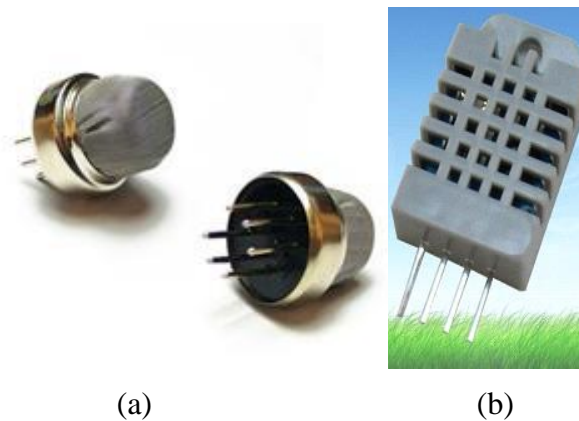
Many efforts have been made at GHG emission measurements. Wojtas *et al.* [7] developed a nitrous oxide detection system that uses the cavity enhanced absorption spectroscopy. The instrument was able to measure N<sub>2</sub>O concentration in ppb (part per billion), with the best sensitivity at a wave length of 4.53 $\mu$ m. A microcontroller based Liquefied Petroleum Gas (LPG) detection instrument that uses the GSM module for measuring gas leakages have also been described [8]. Available information showed that the available commercial GHG detection instruments do not measure meteorological parameters alongside their target gases. Meteorological parameters such as temperature, humidity, pressure, precipitation etc could impact on GHG emission sources/sinks and transport. The functional block diagram of the designed instrument is shown in Figure 1. The instrument contains the following circuits/units: two gas sensors; the MQ-4 (for methane), MQ-135 (for nitrous oxide) and one temperature/humidity sensor, an SD storage device, a Liquid Crystal Display (LCD) and an Arduino board. The Arduino board has the ATMEGA 2560 microcontroller, sample and hold circuits, a multiplexer and the Analogue-to-Digital Converter (ADC) required for data conversion. The MQ-4 (for methane) and MQ-135 (for nitrous oxide) are semiconductor sensors with low conductivity in clean air. The compositions of the sensors are micro Al<sub>2</sub>O<sub>3</sub> ceramic tube, Tin IV oxide (SnO<sub>2</sub>) sensitive layers, measuring electrodes and a heater. These parts are fixed into a crust made by plastic and stainless-steel net. The heater provides the necessary working condition for the sensitive components. The MQ-4 and MQ-135 are 6 pins devices (see Fig. 2 (a)) with 4 of the pins used to generate signals while the other 2 are used for providing the heating current that ensures that the sensors work properly. The sensors are cheap and believed to give stable operation over its quoted long-life span [9].



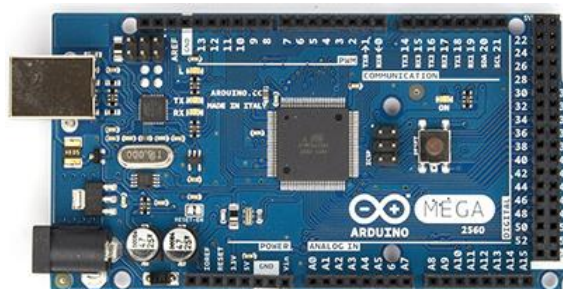
**Fig. 1: Functional block diagram for the designed methane and nitrous oxide measurement instrument**

The RHT04 (humidity and temperature sensor) is a product of maxdetect technology (see Fig. 2 (b)). Its operational voltage is between 3.3 and 5V direct current. It takes about two seconds for the sensor to attain signal stability. It can drive a long signal transmission line up to 100m. With a well synchronized control signal, RHT04 can be easily operated between the standby mode and the active mode, making it highly suitable for devices where low power consumption is desired. The Seeduino SD card shield was used to equip the instrument for real time data storage. The Seeduino was newly-released and it is able to provide storage space for the Arduino microcontroller. The advantage of the Seeduino is that it combines the standard SD slot and the Micro SD slot on a single board with an adaptor that enables a switch between the two options.

The microcontroller used was the Arduino ATmega 2560. Its recommended operational voltage range is 7 to 12V. The ATmega 2560 has 248KB of flash memory for code storage. It has 16 analog input pins and 54 digital pins. The microcontroller has sixteen Analog to Digital Converters (ADCs). The output of the ADCs are multiplexed by a 16-channel multiplexer. The input of each ADC has a sample and holds circuit which ensures that the signal at the input of the ADC is held at a constant level during conversion. The Arduino is equipped for serial communication on any of its digital pins with full library support for Inter Integrated Circuit bus (I<sup>2</sup>C) and Serial Peripheral Interface (SPI) communications. The Arduino board (see Fig. 3) comes with a bootloader, which allows uploading of new codes without the use of any external hardware programming tools. Since the instrument is meant for in-situ measurement, it becomes necessary to power it with sources that can be readily available on-site. The power source used was 4A 12V battery. This voltage was scaled down to the voltage need by each of the units in the instrument with appropriate voltage regulators.



**Fig. 2: (a) MQ-4 and MQ-135 gas sensors (Source: [9]) and (b) RHT04 relative humidity and temperature sensor (Source: [10])**



**Fig. 3: The Arduino ATmega 2560 board (Source: [11])**

## 2.1 The circuit

The complete circuit diagram is shown in Figure 4. The output of the gas sensors was connected to the analog input pins 5 (for MQ-4) and pin 2 (for MQ-135) respectively on the Arduino board. The relative humidity and temperature sensor (RHT04) were connected to the analog input pin 4 on the Arduino board. The LCD was also interfaced with the microcontroller as shown in Fig. 4. The control firmware in the ATmega 2560 synchronized and directed the entire process of the instrument. All parameters from the sensors were processed and the atmospheric concentration of methane and nitrous oxide were computed alongside the relative humidity and atmospheric temperature. The results are immediately displayed on the LCD screen and also stored on a micro SD storage device for subsequent analysis on the computer.

## 2.2 Calibration

Due to unavailability of standard equipment, all calibration procedure relied on the understanding of the functionality of the various used hardware and the manufacturer supplied methane and nitrous oxide calibration guidelines [9]. For the hardware calibration, all *offset* and *drift* voltages that could arise from signal amplifiers, the ADC, the sample and hold and the microcontroller itself were nullified. This was achieved by applying known voltages (0V and 5V) independently into the inputs. These inputs were read and the discrepancy noticed was corrected for in the program code. The second stage involves the use of the apparatus shown in Fig. 5. The gas sensor was inserted into the gas sensor inlet of the tube, while pure oxygen was connected to the gas inlet tube. A control valve was fitted on the outlet side. After completely filling the tube with pure oxygen, the valve was then released to allow oxygen to flush out any gas that might have accumulated in the tube. The valve was closed after about 5 minutes after which  $R_s$  and  $R_L$  (10k $\Omega$ ) were adjusted so as to have zero  $CH_4$  and  $N_2O$  concentrations respectively. Moreover, the sensors were unable to settle down to zero concentrations, but 323ppb and 0.423ppb for methane and nitrous oxide at room temperature. These values were thereafter programmed into the firmware controlling each sensor so as to adjust any further reading to take care of the zero-point shifts.

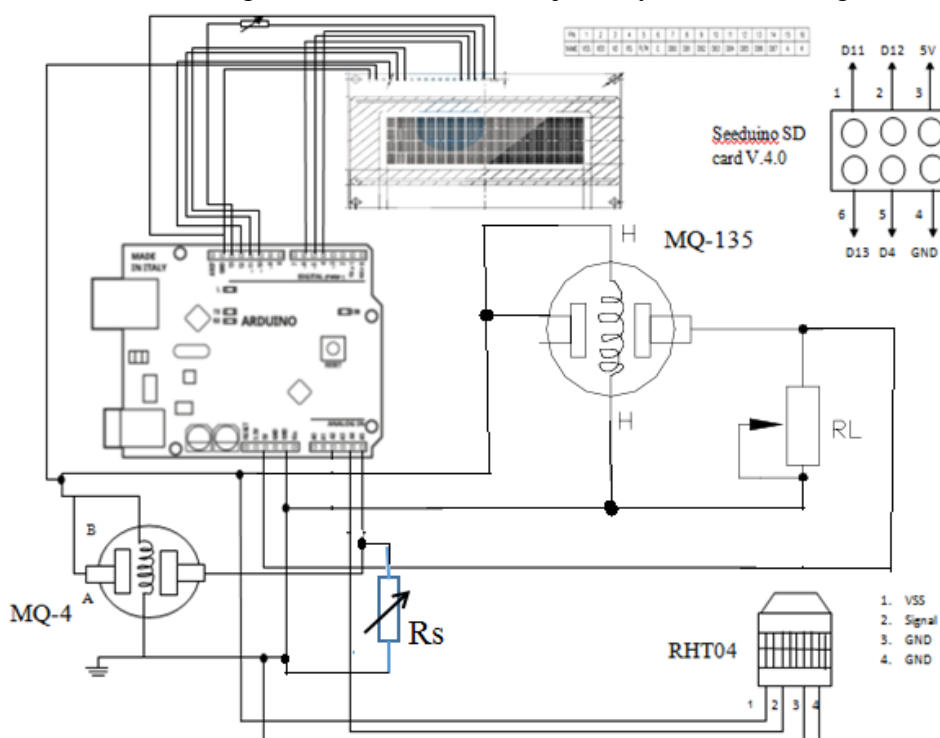
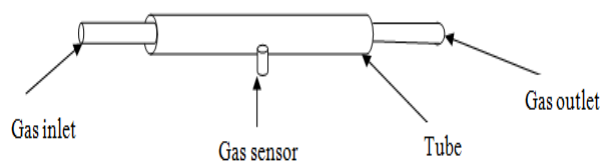


Fig. 4: Connection between the components and the Arduino board



**Fig. 5: Schematic of the calibration apparatus**

## 2.3 Data Collection

To test the usefulness of the instrument, it was used to collect data at different locations between October and November 2018. The choice of each location was dictated by their propensity towards the emission of the target gases. The first location was a local market. The used market is the biggest and the most patronized market in Akure, south western Nigeria. The market is un-kept and many of the activities around the place which included meat processing and decomposition from livestock excreta and perishable food items e.t.c were capable of subjecting the market to more CH<sub>4</sub> and N<sub>2</sub>O emissions. The second location was a gas filling station, owned by the Nigeria National Petroleum Corporation (NNPC). The gas filling station is the biggest in Akure with about thirty dispensing points and quite a large number of consumers patronize the station daily. The third location was a dump site, operated by the Ondo State Waste Management Authority. Dump sites have high potential for gas emission due to microbial processes. The fourth location was a busy arterial road in Akure. Vehicular movements in this area during the busy hours of the day are enormous and for this, the location is popularly called “road-block”. The fifth location was a control site. The control site is located inside the premises of the Federal University of Technology Akure and the area chosen could be adjudged to be near nature due to the vegetation and low human activities in the area.

## 3. Results and Discussion

### 3.1 The Market Location

The hourly mean temperature and relative humidity variation, the hourly mean methane and nitrous oxide variation as well as the hourly mean variation of temperature on methane and nitrous oxide results in the market area are shown in Fig. 6(a – d). The trend shown in Fig. 6(a) reveal that the minimum mean temperature was 28.9°C at 9:00 Local Time (LT) before increasing steadily towards a maximum value of 33°C at 14:00LT. It then decreases to around 32°C at 15:00LT. Humidity reaches its highest value of 79.5% around 9:00LT and attains its lowest value of 68.6% around 14:00LT. The variation of methane and Nitrous oxide as seen in Fig.6(b) shows that Methane had its highest value of 8812.13ppb at 9:00LT before a gradual decrease of the gas at 10:00LT which later increases toward the afternoon at 14:00LT and then drop toward the evening period. The concentration of nitrous oxide increases to a value of 0.72ppb at 10:00LT before reaching a peak value of 1.61ppb and thereafter drops tremendously around the afternoon period. The trend in the atmospheric temperature result is not surprising because lower temperature values are expected during early hours due to turbidity. After sunrise, the temperature is expected to increase steadily which was why the peak value was attained at 14:00 LT and then declined gradually to around 32°C at 15:00 LT. The reduction can be a result of decrease in the incoming solar radiation received at the earth surface. The trend shows that the surface temperature obeys solar cycle. Humidity reaches its highest value around 9:00 LT and its lowest value at around 14:00 LT showing a trend similar to that of the temperature. The concentration of methane was noticed to be high before 9:00LT. At 10:00 LT, a gradual decrease in the concentration of the gas was observed. This may

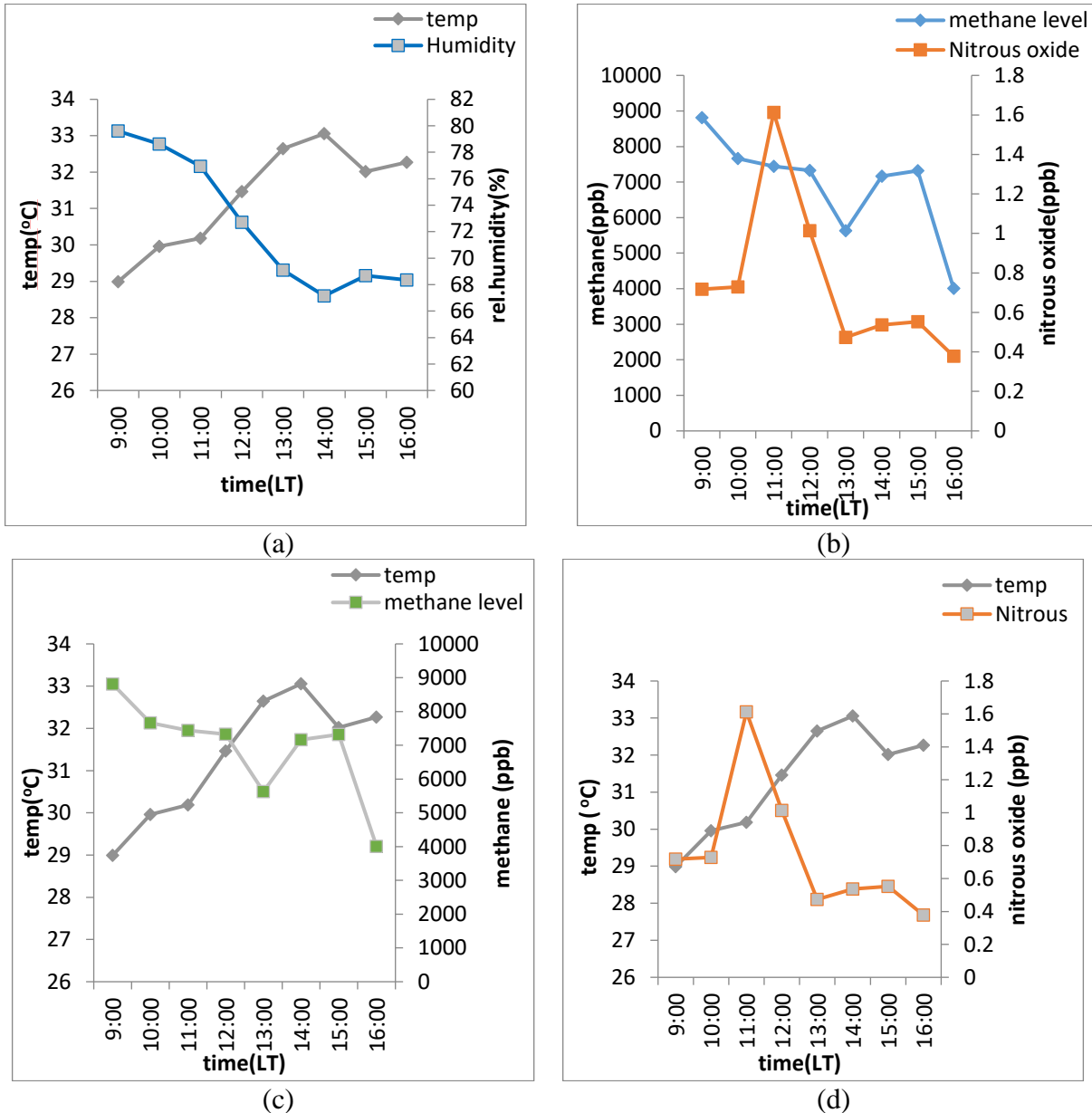
be due to the prevailing market activities at that time of measurement, which could include increased activities leading to the wind dispersion of the gas. Slight increase in Methane concentration was accompanied with a sharp increase in temperature at 14:00 LT as observed in Figure 6(c). This observation is in line with the discussion in [12], where such an increase can be as a result of microbial decomposition of animal wastes. Under anaerobic and non-anaerobic conditions, any decomposition would lead to increase in the gas particularly at elevated temperatures. It has also been shown [12] that methane is a by-product of enteric fermentation in ruminant livestock (e.g cattle), therefore, meat processing in the abattoir and also meat distribution process in the market could have resulted in the sharp increase in methane under the high temperature.

The peak result for nitrous oxide was observed before noon (1.61 ppb at 11:00LT) as shown in Fig. 6(d). The trend shown in Fig. 6 for nitrous oxide may be attributed to emission from decomposing crops like pepper, tomatoes under lower temperature and this agrees with the work cited in [13]. An insignificant relationship between N<sub>2</sub>O emission and temperature has been shown from the work discussed in [14] and [15]. Similar trend (as shown in Fig 6(d) is seen from the measurement given by the designed instrument. Precipitation is the only climatic parameter that has been shown to have significant effect on N<sub>2</sub>O concentration variation [14] in the atmosphere. Because animal activities do not contribute significantly to nitrous oxide emission, its diurnal variation is expected to be relatively constant [16].

### 3.2 The Gas Station

According to the results in Fig. 7(a), temperature has its lowest value of 26.5°C at 9:00 LT before increasing gradually to attain its peak value of 29.3°C at 16:00LT. Humidity has its highest value of 80.6% at 11:00LT in the morning period before dropping to 73.43% at 14:00LT. In Figure 7(b), the concentration of methane was 3876.5ppb at 9:00LT before dropping steadily to a value of 3040.89ppb at 12:00LT and later increases steadily in the afternoon period to a value of 5628.38ppb at 15:00LT. Nitrous oxide attains its maximum value of 0.24ppb at 9:00LT and later drop sharply to its minimum value of 0.024ppb at 12:00LT. The high concentrations of methane and nitrous oxide as measured in the morning hours might be attributed to high level of gas purchase at such hours from refueling processes. The same result is also noticed between 12:00LT to 14:00LT, which happens to be another time frame within which refueling is also common. Leakages from the fuel pumps can also contribute to the concentrations at these hours. The trend in the concentration variation of methane and nitrous oxide at the gas station can be largely attributed to refueling activities. The concentration of nitrous oxide in fossil fuel is low, which may explain the reasons for obtaining low nitrous oxide concentration values.



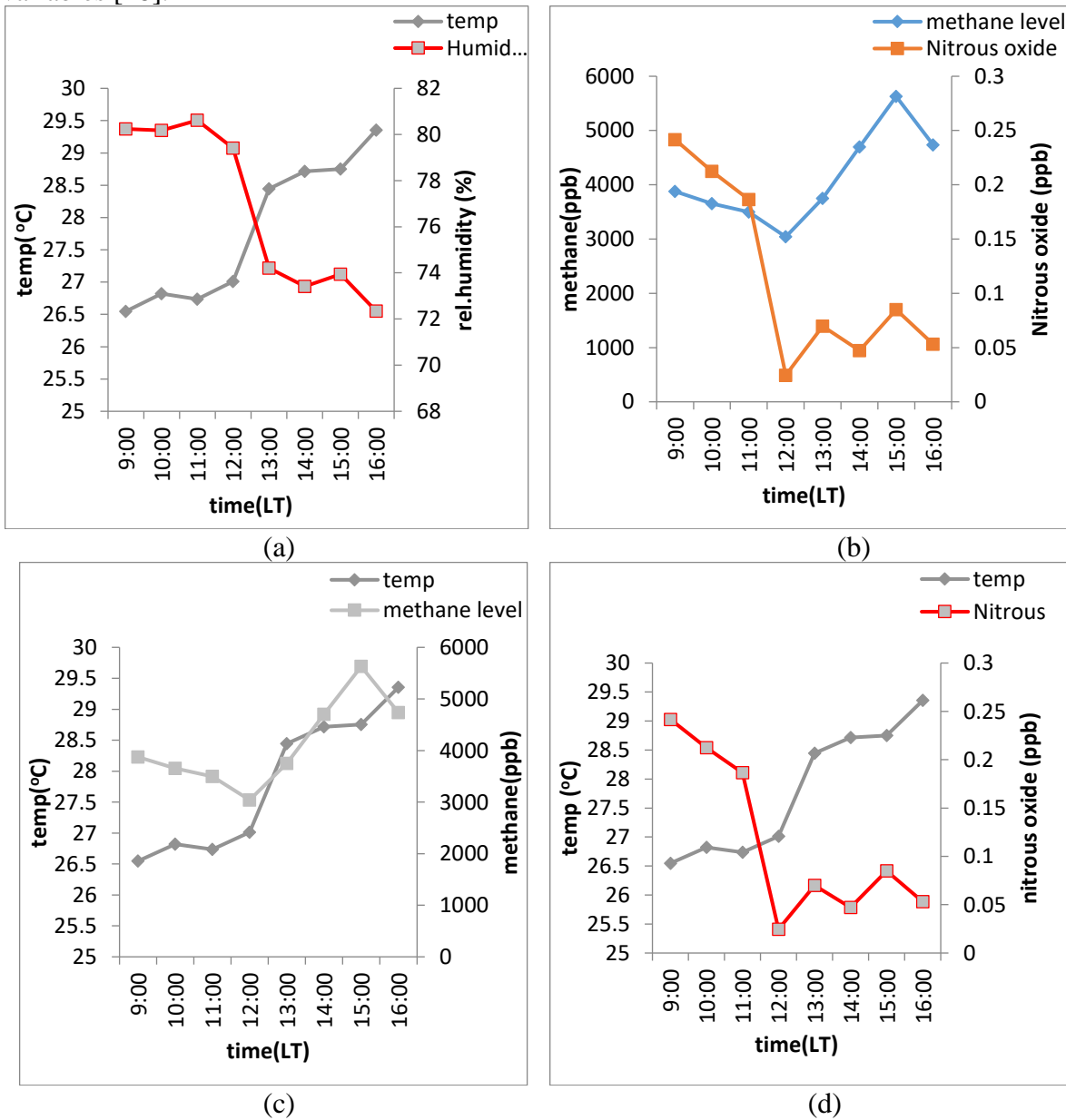


**Fig.6: Hourly mean variation of (a) temperature and relative humidity in the market area (b) Methane and Nitrous oxide in the Market Area (c) temperature on methane in the market area (d) temperature on Nitrous oxide in the market Area**

### 3.3 The Dump Site

According to the results in Fig. 8(a-d), an average minimum temperature of 30.8°C was observed at about 10:00LT which was afterwards maintained at a value of 31°C and later increases to a maximum value of 40.3°C at 15:00LT. Humidity has a maximum value of 72.9% at 9:00LT before decreasing slowly to constant value of 66% between 10:00LT and 12:00LT before a minimum value of 55.9% was observed at 15:00LT. Methane had its maximum value of 6322ppb at 9:00LT in the morning period before dropping to 874ppb at 15:00LT toward the evening period while nitrous oxide attained a value of 0.63ppb which later reduce to 0.25ppb at 11:00LT before increasing to its maximum value of 0.78ppb at 15:00LT. These trends are being expected as anaerobic bacteria activities easily lead to methane and carbon dioxide production, particularly at low temperatures [17]. These bacterial (called methanogens),

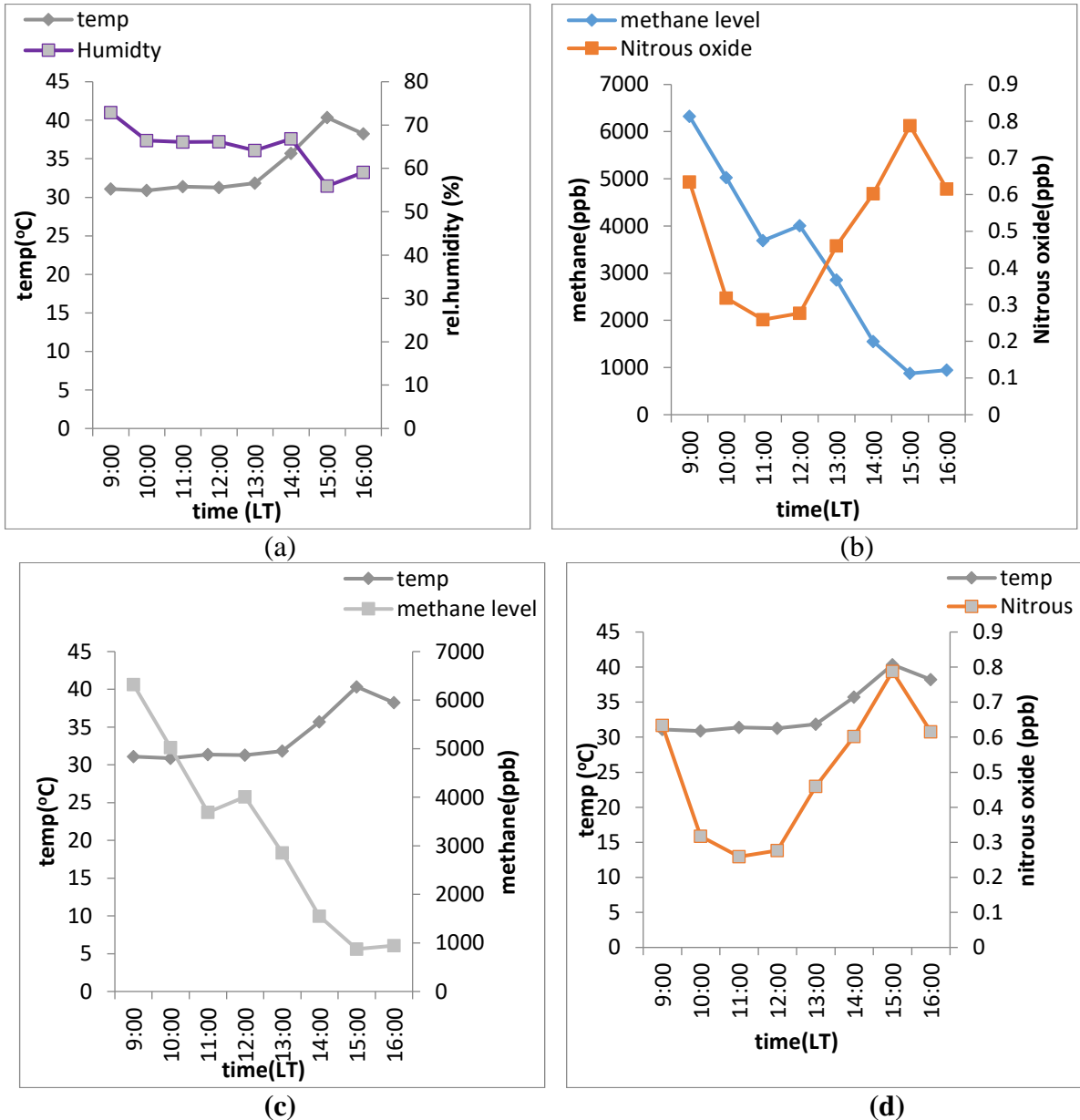
are special group of microbes that break down certain molecules such as hydrogen and carbon dioxide to produce methane through the process of methanogenesis [5]. Methane concentration values at dump sites is a function of the waste composition, cover materials, soil moisture, temperature and other variables [18].



**Fig.7: Hourly Mean variation of (a) Temperature and relative humidity in petrol station (b) Methane and Nitrous oxide in petrol station (c) temperature on methane at the petrol station (d) temperature on nitrous oxide in petrol station.**

Therefore, the decrease in the measured concentration of methane as observed in Figures 8(b) and 8(c) can be attributed to high oxidation at the surface. The moisture content and the temperature of the cover soils directly affect oxidation of methane [18,19,20,21,22]. This may further explain why high concentration of methane was obtained during low temperature but high humidity times. Additionally, the vertical transportation of methane from anaerobic zones is slow and therefore, a percentage of the methane emitted could possibly have been oxidized and converted to nitrous oxide by methanotrophs (methane oxidizing bacteria) [23,24] which could have been responsible for the reduction in methane concentration, leading to high nitrous oxide concentration between the hours of 12:00LT to 15:00LT.





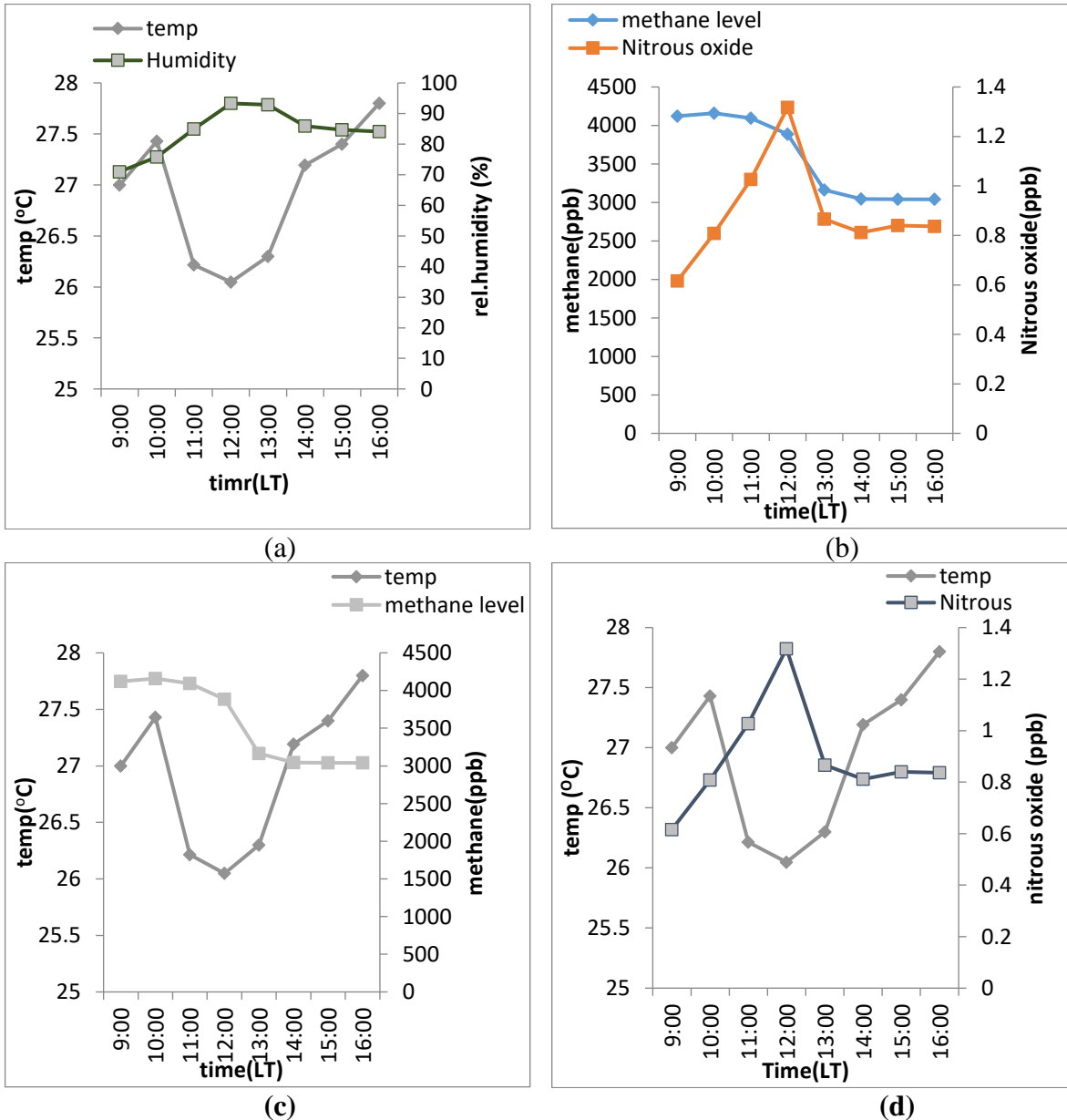
**Fig.8: Hourly Mean variation of (a) Temperature and relative humidity in refuse dump site (b) methane and nitrous oxide in refuse dump site (c) temperature on methane in refuse dump site (d) temperature on nitrous oxide in refuse dump site.**

### 3.4 The Road Location

The temperature and relative humidity results for this location are shown in Figure 9(a). Due to the morning rain that occurred on the day of measurement, the temperature profile was somehow unstable. The temperature was 27.4°C at 10:00LT before decreasing steadily to 26°C at 12:00LT. The humidity increased from a minimum value of 70% at 9:00LT to a maximum value of 93.3% at 12:00LT before dropping steadily at 16:00LT to a value of 84%. High concentration of methane was observed between 9:00LT and 12:00LT before it attained a constant trend from 12:00LT to 13:00LT. The nitrous oxide rose around 9:00LT to reach a maximum value of 1.31ppb at 12:00LT before the concentration reduced to a value of 0.86ppb at 13:00LT and maintained an unstable trend from 13:00LT to 16:00LT.

The concentration of methane was high between 9:00 and 12:00 LT and incidentally within these periods, the temperature was increasing and the humidity decreasing (see Fig. 9(b) and 9(c)). These

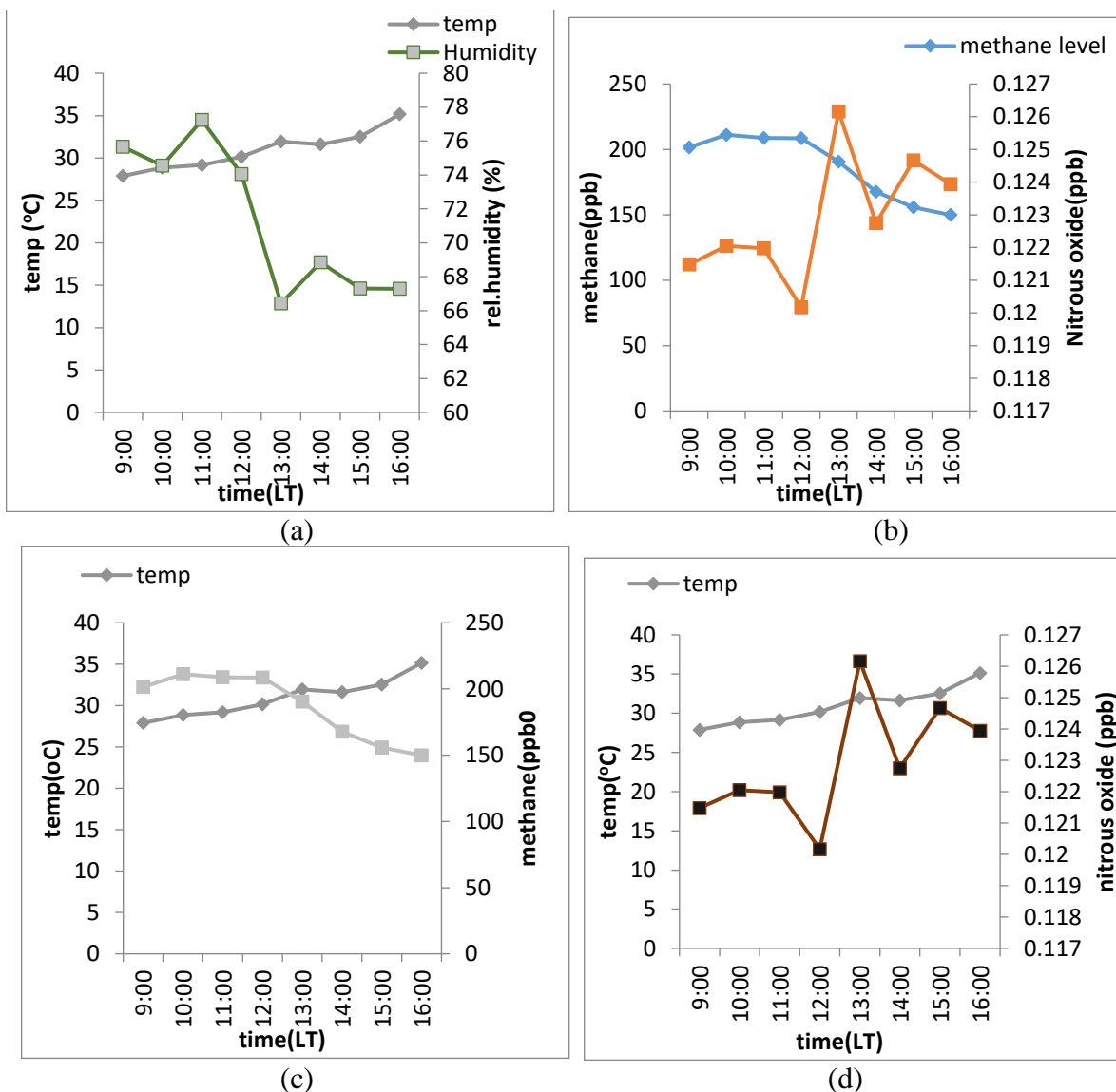
trends can be attributed to traffic congestion at these periods, which by extension will lead to incomplete combustion of fossil fuel in the car engines and the incomplete oxidation of methane at the exhaust, thereby leading to higher methane emission [4]. Methane emission is higher at lower temperature [25,26]. At lower temperatures, liquid fuel does not vaporize/burn completely due to lower combustion. It was noticed that methane later attained a constant value after slight drop between 12:00LT to 13:00LT as shown in Figure 9(b) or 9(c). This can be adduced to low vehicular activity at the hours. Nitrous oxide rose around 9:00 LT to attain its maximum value of 1.31ppb at 12:00 LT. It has been shown that N<sub>2</sub>O is formed at relatively low temperature [27,28,29] but at higher temperatures, the N<sub>2</sub>O itself is destroyed and this may explain the trend between atmospheric temperature and N<sub>2</sub>O as observed in Fig 9(d).



**Fig.9: Hourly Mean variation of (a) Temperature and relative humidity at road side (b) methane and nitrous oxide at road site (c) temperature on methane at road side (d) temperature on nitrous oxide at road side.**

### 3.5 The Control Site

The maximum methane concentration level measured at the control location was 208.63ppb and it took place in the early hours (8:00LT to 10:00LT) as seen in fig 10(b). The maximum nitrous oxide concentration value was 0.126ppb at 13.00LT as seen in figure 11(b) and 11(d). As indicated in fig 10(a), the measured temperature variation was between 27.81°C and 35.14°C at 16:00LT and the humidity was high in the morning at a value of 75.6% before reaching its highest peak of 77.23% and thereafter decreasing steadily to a value of 66.92% at 13:00LT. The control site measurement indicates that the hourly value of methane is extremely low compared to the values of the methane measurement from other sites. This may be as a result of the lower level of emission resulting from low human activities within the region. However, it was discovered that the value of nitrous oxide at the control site was only slightly lower compared to the value from other site. This might be as result of microbial activities in soil under vegetation. According to the IPCC, about 60% of emitted Nitrous oxide are from uncultivated soils, under natural vegetation [30,31]. This could account for the large percentage of Nitrous oxide recorded at the control site which is predominantly a vast area with natural vegetation.



**Fig.10. Hourly Mean variation of (a) temperature and relative humidity at control site (b) Methane and nitrous oxide at control site (c) temperature on methane at control site (d) temperature on nitrous oxide at control site.**

#### 4. Conclusion

The emission of the Green House Gases (GHGs) and its adverse effects on the entire ecosystem have become subject of international debate. To address a challenge of this nature, realistic data have to be collected and analyzed before an effective policy or mitigating plan can be effected. For this to happen, widespread deployment of instruments that can capture GHGs emission data is expedient. The under-developed and developing countries might not be able to contribute meaningfully towards any emission reduction campaign or strategy due to the meager resources available to them. To circumvent these challenges, there is the need to develop local content and infrastructure towards GHG emission source identification, data collection and analysis. This paper has been able to clearly demonstrate an attempt in this regard. An instrument that can measure, display and store data on methane, N<sub>2</sub>O, atmospheric temperature and relative humidity has been implemented. Within available resources, the instrument was calibrated and thereafter used to collect data in some locations noted as high GHG emission prone areas. Compared with the results from the control location (208.63ppb), the average concentration of methane was highest at the market place (8812.13ppb), followed by the emission at the dump site (6322ppb), the gas station (5628.38ppb) and then the road location (4159ppb). Likewise, the average concentration of nitrous oxide was highest at the market place (1.61ppb), followed by the emission at the road location (1.31ppb), the dump site (0.78ppb) and then the gas filling station (0.24ppb) as against the value of 0.126ppb at the control location. Surprisingly however, the methane level at the market and the dump site locations were much higher than that at the gas filling location. Going forward, the figure recorded for the concentration of these gases are high compared to the global mixing ratios stated in different literatures [32,3]. It will be interesting to probe this observation further in order to gain more insight into the mechanism responsible for this. Also, probing these observations relative to a larger number of similarly selected sites will provide more useful data for the study of the environmental and health effects on the society, as these sites will be well distributed in their large numbers within cities. Based on the various results and the trends thereof, it can be concluded that the instrument worked satisfactory to a reasonable extent. The instrument can be easily modified for additional sensors.

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