Light gaseous hydrocarbon studies in the near-surface soils of Tatipaka, Pasarlapudi and Kaza areas of Krishna–Godavari Basin, India

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One hundred and thirty-five soil samples were collected from a depth of 2–2.5 m near oil and gas fields of Tatipaka, Pasarlapudi, Ponnamanda, Kaza and Palakollu areas of Krishna–Godavari Basin, Andhra Pradesh, India and were analysed for light hydrocarbon gases. The adsorbed soil gas analysis showed the presence of moderate to low concentrations of methane (CH_4; 2–83 ppb), ethane (C_2H_6; 1–92 ppb), propane (C_3H_8; 1–134 ppb), butane (n-C_4H_{10}; 1–187 ppb), pentane (n-C_5H_{12}; 1–316 ppb) and $\sum C_{2+}$ sum of C_2H_6, C_3H_8 and n-C_4H_{10} (1–729 ppb) in the soil samples of the study area. The carbon isotopic composition $\delta^{13}C_1$ of the samples ranged between –36.6‰ and –22.7‰, which indicates thermogenic origin of methane. Observed concentrations of adsorbed light hydrocarbon gases in soils have shown an interesting correlation of surface light hydrocarbon anomalies with the subsurface oil and gas accumulation in the study area, demonstrating the usefulness of adsorbed soil gas technique in petroleum exploration.

Keywords: Adsorbed soil gas, hydrocarbons, microseepage, oil and gas fields, petroleum exploration.

SURFACE geochemical prospecting is an exploration method based on the premise that the light hydrocarbons, namely methane (C_1), ethane (C_2), propane (C_3), butane (C_4) and pentane (C_5) migrate upward from subsurface petroleum accumulations by diffusion and effusion. The light hydrocarbons in the soil are the most important indicators in geochemical exploration of hydrocarbons. The analysis of light gaseous hydrocarbons (C_1–C_5) in soils represents the earliest surface geochemical methods used and is one of the most researched and tested geochemical survey approaches. Surface geochemical prospecting involves the search for the presence of surface manifestations of hydrocarbons, which are indicative of deep-seated petroleum reservoirs in onshore and offshore regions. Adsorbed soil gas survey is a direct technique in which the trace amount of light hydrocarbons occurring in the pore space of the soil and adsorbed on the fine-grained portion of the soil, or those that are incorporated in soil cements are analysed. Trace amounts of light hydrocarbons collected near the Earth's surface provide clues to present-day subsurface fluid composition and migration from deep-seated oil and gas reservoirs[1]. Micro-seeps are invisible and can be recognized by the presence of anomalous concentrations of light hydrocarbons (C_1–C_5) in the near-surface soils/sediments along with other surface manifestations of hydrocarbon seepage which can be in the form of microbial and trace element anomalies, mineralogical changes, and altered electrical, magnetic and seismic properties. These near-surface anomalies provide clues on the nature and composition of subsurface petroleum occurrences and help demarcate the anomalous hydrocarbon zones and grade the frontier basins[2–5]. Migration mechanisms generally proposed for microseepage include buoyancy for subsurface microbial flora and diffusion for subsurface adsorbed gases[6].

Hydrocarbons reside in the near surface as free and bound gases; however, only the free gases migrate from depth[7]. Free gases occur as either vapour in pore spaces or as a gas dissolved in an aqueous solution. If the gas is attached to the sediment matrix or contained within the interstices of rocks or certain minerals, such as calcite or oxide coatings[7,8], it is considered to be bound. Bound gases include adsorbed and chemi-adsorbed gases. Gases that have reached the soil horizon may also contain biogenic, thermogenic, and/or abiogenic gases that migrated to the surface from deep sources[9]. Near-surface free gases are dominated by gases from deep sources, but may also contain gases formed during diagenesis, such as biogenic methane[10]. To minimize the influence of biogenic or another source of C_1, some other hydrocarbon constituent (for example, ethane, propane and butane) should be measured[7,11].

This article reports the adsorbed light gaseous hydrocarbon concentrations in soils, and the outcome of carbon isotopic analysis in parts of the oil and gas-bearing Krishna–Godavari (K–G) Basin.

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Geological setting

The K–G Basin is a pericratonic rift margin system with an Archean crystalline rock basement on the east coast of the Indian peninsula[12]. It covers an area of 28,000 sq. km on land and 24,000 sq. km off land up to 200 m depth[13]. The basin lies between 15°30′–17°N lat. and 80°–82°30′E long. between Kakinada in the northeast and Ongole in the southwest. Archean crystalline basement and Upper Cretaceous sedimentary outcrops occur at the basin margin on land. A significant part of the on-land basinal area is covered by Recent alluvium. Outcrops along the basin margin include Permean Chintalapudi sandstone, Cretaceous to Jurassic Gollapalli sandstone, Raghavapuram shale and Tirupati sandstone exposed around Dwaraka Tirumala area of West Godavari District. The Rajahmundry sandstone occurs near Rajahmundry and Dowleswaram areas, which are feldspathic, ferruginous and laterised sandstone of Miocene age, equivalent to that of the Ravva Formation of offshore area.

Basin architecture

Based on Bouguer gravity data, Murty and Ramakrishna[14] have identified three sub-basins separated by two basement horsts. From the southwest, these are the Krishna, West Godavari and East Godavari sub-basins separated by the Bapatla and Tanuku horsts respectively. The West Godavari sub-basin is further subdivided into Gudiwada and Bantumilli grabens, which are separated by Kaza-Kaikalur horst[15]. The Krishna sub-basin contains 1560 m of Cretaceous and older sediments above the Archean basement. Bapatla horst lies between the Krishna and the West Godavari sub-basins. Many lower Mesozoic sequences are thin over the Bapatla horst and lie disconformably with erosional contact, which suggests that the Bapatla horst was uplifted during the Cretaceous. In the West Godavari sub-basin, Cretaceous sediments are thin over the Kaza-Kaikalur horst, compared to the thick section on either side of the horst in the Bantumilli and Gudiwada grabens. The data suggest that the Kaza-Kaikalur horst remained uplifted during early Cretaceous. The sedimentary cover over the Kaza-Kaikalur horst is about 1900 m, whereas in the Gudiwada graben it is around 3500 m. In the Bantumilli graben the sediment thickness has increased to 4500 m.

Tectonic history

In the East Godavari sub-basin, the sediments ranged from 2900 m over the pre-existing basement horsts to more than 5000 m in the deep basin area in the southeast[16]. The 2000 m of argillaceous sediments of the Cretaceous in the southeast created a series of en-echelon faults and also a steep step-fault zone in early Palaeocene Basalts. The fault zone is known as the Matsyapuri–Palakollu fault zone. Location map of the study area of the K–G Basin is shown in Figure 1.

Lithostratigraphy

A brief description of various lithounits is given below and a stratigraphic sketch of the basin under study is given in Figure 2.

Basement: In Lingala–Kaikalur area, the depth to basement varies from 1900 m (on the Kaikalur horst) to more than 3000 m in the Lingala field fault block. This is the oldest formation; it varies in composition from granite gneiss to Khondalite.

Kanukollu sandstone: This is the oldest sedimentary unit; it overlies the basement and is overlain unconformably by the Raghavapuram Shale. This is primarily composed of sandstone with thin beds of shale and coal at places. This formation is deposited in a marginal environment during early Cretaceous (Albian) time.

Raghavapuram shale: This formation unconformably overlies the rift fill sequence of Kanukollu Sandstone and in turn is overlain by Tirupathi Sandstone formation. This formation is mainly argillaceous and consists of shale with minor interbedded sandstone and thick sandstone units at the top and bottom. It was deposited in a marginal marine to shallow inner shelf environment during early Cretaceous and represents transgressive phase of depositional cycle.

Tirupathi sandstone: It unconformably overlies Raghavapuram Shale and is unconformably overlain by Razole Formation. It comprises of mainly sandstone with thin

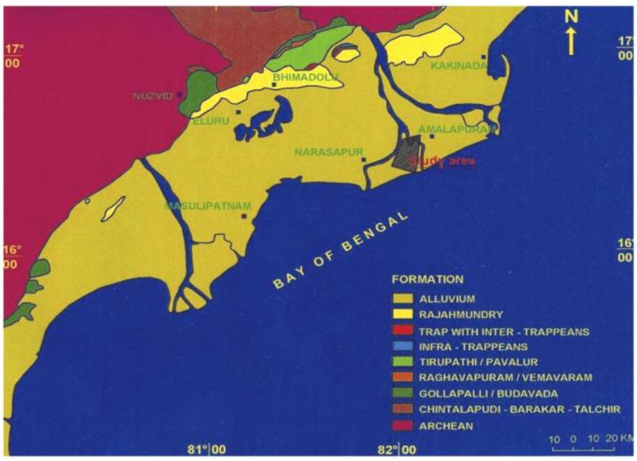

Figure 1. Geological location map of the study area in the Krishna–Godavari (K–G) Basin (Source: Directorate General of Hydrocarbons, India).

intercalations of clay stone and belongs to late Cretaceous. It was deposited in Paralic environment.

Razole Formation: This formation comprises of basalt and intertrappean claystone, sandstone and limestone. Three distinct flows of basalt are recognized. The thickness of this formation is around 100 m and occurs generally around 900 m in the study area. It belongs to the Palaeocene. The study area with oil/gas wells shows this formation.

Nimmakuru Sandstone: This formation about 3500 m is non-conformably underlain by the Trap and unconformably overlain by the Narsapur Claystone. This extends up to the Kaza horst. This was deposited in marginal marine environment and belongs to Palaeocene to Oligocene age. In the study area, Palakollu region shows this sandstone. The Pasarlapudi Sandstone formation of the Vadaparru shale showing oil/gas wells of the study area lies above Nimmakuru sandstone.

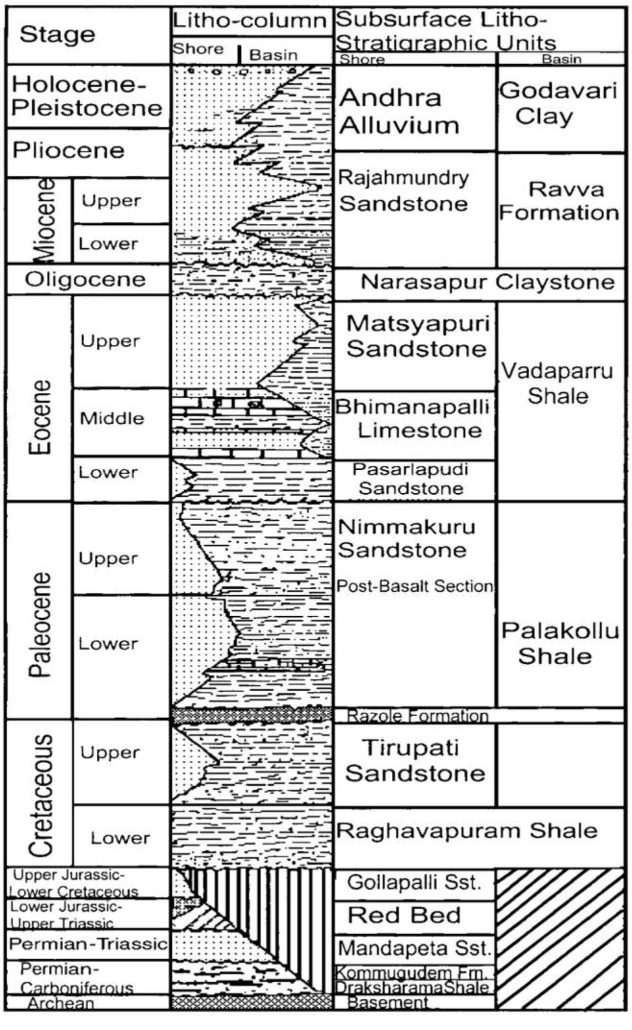

Figure 2. Lithostratigraphic nomenclature of sedimentary sequences in the K–G Basin.

Narsapur Claystone: This argillaceous Formation unconformably overlies the Nimmakuru sandstone and is covered by Recent alluvium. It comprises of mainly claystone with few thin beds of sand. This unit was deposited in a shallow inner shelf to middle shelf environment and belongs to Oligocene to Pliocene.

The source, reservoir, cap rocks, entrapment and fault system

Raghavapuram shale – a regional source rock: The majority of good source rock intervals lies within the Raghavapuram Shale sequences with proclivity to generate gas and liquid hydrocarbons. The maximum total thickness of this sequence encountered in the so-far drilled wells is 997 m in West Godavari sub-basin. The Raghavapuram Shale sequence was the result of post-rift marine transgression into the NE inclined rift grabens (half grabens), probably forming a restricted and shallow water environment. The sedimentation took place at a very slow rate, probably with no classical delta build-up, except for minor bar sands along the shore deposited during both transgressive and regressive phases. The stagnant water conditions and slow rate of sedimentation led to the deposition of organic-rich shales[17].

Reservoir rock: The reservoir rocks are mostly multi-storied sand bodies of both the transgressive and regressive marine bars deposited along the palaeo shoreline above and below Raghavapuram Shale and also lenticular sand bodies within Raghavapuram shale. The rift fill sandstones of Kanukollu Formation underlying Raghavapuram shale are also good hydrocarbon reservoirs. The occurrence of reservoir sands in the sub-surface ranges between 1800 and 2400 m in the Lingala–Kaikalur area.

Cap rock: The shale sequences within Raghavapuram Shale act as cap rocks for the reservoir sandstones.

Entrapment style: The study of the reservoirs in the Lingala–Kaikalur area proved that the entrapment style is strati-structural in nature. These include pinch outs, wedge outs against basement highs, unconformity and fault seals. Fault system plays the role of hydrocarbon migration and accumulation.

Fault system: Raghavapuram Shale reveals six major faults present in the Lingala–Kaikalur area. These represent the NE–SW basement faults. These faults help in establishing communication from the subsurface reservoir to the near surface by microseepages.

Manmohan *et al.*[17] correlated the Lower Cretaceous shale sequence of West Godavari sub-basin with the Raghavapuram Shales of Mandapeta area. The overall temperature gradient is 4.2°C/100 m. The hydrocarbon occurrences associated with the Razole Formation have their origin solely related to their short deposition sequence probably involving short-distance migration. Restricted shallow water environment for the deposition of the sequence in this basin favoured the richness of organic matter and resulted in a good regional source rock. The depositional environment varies from continental to lagoonal, marine, littoral, infraneritic and deltaic conditions. The sediments yield rich faunal assemblages like arenaceous foraminifera (*Ammobaculites* sp., *Ammodiscoides* sp., etc.), *Trigonia*, *Inoceramus*, *Lima*, *Pecten*, *Belemnites*, *Helicoceras*, *Cardita*, lamellibranches, gastropods, etc.[18].

Materials and methods

Soil sampling

The reconnaissance survey along the existing roads can help evaluate a large tract of land for its hydrocarbon resource potential and prioritize the areas for further exploratory studies. The soil samples have been collected in reconnaissance pattern away from the existing roads as they may have anthropogenic contamination, possible petroleum spills or coal tar. Some of the samples were collected in the areas where oil/gas wells were found. A total of 135 samples were collected at an interval of 3 km and from a depth of top 2–6 m using a hollow metal pipe by manual hammering to the required depth. A sample location map is shown in Figure 3. About 500 g of core soil samples collected were wrapped in aluminum foil and sealed in poly-metal packs. The samples were sealed in the re-sealable plastic bags with their sample number and global positioning system (GPS) locations marked. Disturbed or excavated areas, soils contaminated with hydrocarbons, chemicals or animal wastes, swamps and areas under waterlogged conditions were avoided for sampling. While collecting the samples, rocks, coarse materials, plant residues and animal debris have been excluded[19,20].

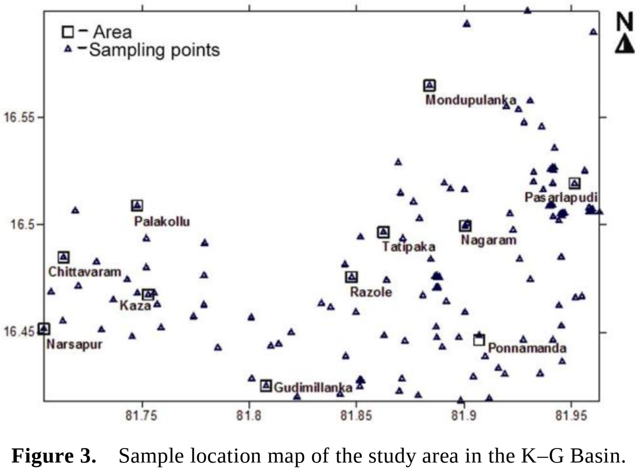

Figure 3. Sample location map of the study area in the K–G Basin.

Analysis of light gaseous hydrocarbons

The light gaseous hydrocarbons were extracted from the soil sample using a gas extraction system developed by Horvitz in 1981 (ref. 21). One gram of 63 µm fraction of wet sieved soil sample was used for extracting light gaseous hydrocarbons after acid treatment in glass degasification apparatus. The gas was analysed on a gas chromatograph (GC) for all samples, and gas chromatograph-combustion-isotope ratio mass spectrometer (GC-C-IRMS) analysis was carried out for samples that had higher concentrations of hydrocarbons. During acid treatment, the dominant gas released was CO_2 which was trapped in KOH solution. The light gaseous hydrocarbons were collected by water displacement in a graduated tube fitted with rubber septa. The volume of desorbed gas was recorded and 500 µl of desorbed gas sample was injected into Varian CP 3380 GC fitted with Porapak Q column, equipped with flame ionization detector. The GC was calibrated using external standards with known concentrations of methane, ethane, propane and *n*-butane. The quantitative estimation of light gaseous hydrocarbon constituents in each sample was made using peak area measurements and the correction for moisture content on wet basis was applied. The hydrocarbon concentration values of individual hydrocarbons from methane through pentane are expressed in parts per billion (ppb)[22].

Analysis for carbon isotopes of light hydrocarbons

Carbon isotopic composition of light hydrocarbons ($\delta^{13}C_1$) in soil samples is determined using GC-C-IRMS[23], which comprises of Agilent 6890 GC coupled to a Finnigan–Delta Plus^{XP} Isotope Ratio Mass Spectrometer via a GC combustion III interface. One millilitre of desorbed gas is injected to GC in splitless mode with helium as carrier gas at fixed oven temperature of 28°C. The light hydrocarbon gases eluting from the GC column enter the combustion reactor maintained at 960°C, where they get converted to CO_2 and water Nafion membrane tube is used to remove water prior to the entry of CO_2 into the mass spectrometer. Reference standards were injected along with the samples to calibrate and check instrumental performance. The carbon isotope ratio in the sample was determined by comparing isotope ratios with that of standard NIST RM 8560 (IAEA NGS2) using ISODAT software. The carbon isotopic composition is reported in per mil relative to the Pee Dee Belemnite (PDB). The precision of the isotopic analysis is ± 0.5‰. The $\delta^{13}C$ is calculated for methane gas (CH_4) in a sample using the following equation

$$\delta^{13}C_1 = \{(^{13}C_1/^{12}C_1)_{sample}/(^{13}C_1/^{12}C_1)_{PDB} - 1\} \times 1000.$$

Table 1. Statistical evaluation of hydrocarbons by adsorbed soil gas technique in the study area of the Krishna–Godavari Basin

Parameters	C_1 (CH_4)	C_2 (C_2H_6)	C_3 (C_3H_8)	C_4 (C_4H_{10})	C_5 (C_5H_{12})	$\sum C_{2+}$
Total no. of samples	135	135	135	135	135	135
Minimum (ppb)	2	1	1	1	1	1
Maximum (ppb)	83	92	134	187	316	729
Arithmetic mean	23.037	1.977	1.918	3.007	6.507	13.362
Standard deviation	16.218	8.581	12.617	18.407	35.542	74.045
Positive samples (%)	100	25.93	5.93	5.19	5.19	26.67
Samples showing nil count (%)	0	73.33	94.07	94.81	94.81	73.33
Samples above arithmetic mean (%)	37.78	14.07	4.44	5.19	5.19	5.19
Samples above standard deviation (%)	60.74	4.44	2.96	3.70	4.44	3.70

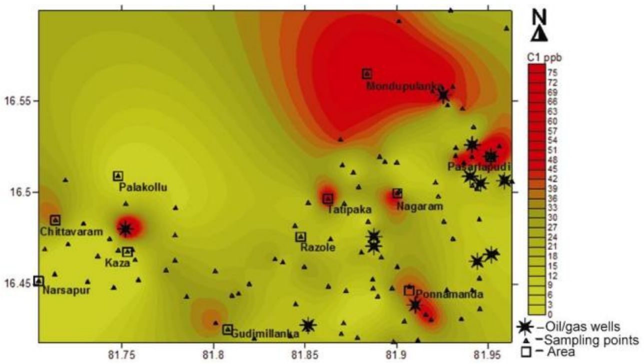

Figure 4. Adsorbed methane concentration map of soil samples from the study area.

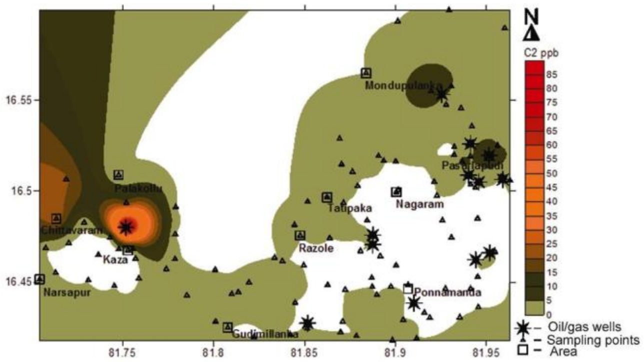

Figure 5. Adsorbed ethane concentration map of soil samples from the study area.

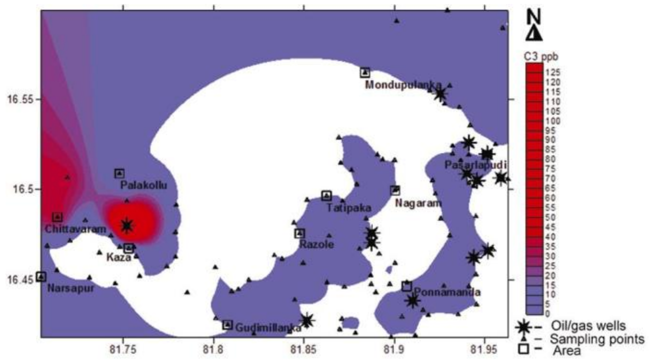

Figure 6. Adsorbed propane concentration map of soil samples from the study area.

Results and discussion

The GC analysis of 135 soil samples collected near oil/gas wells and their surrounding areas and the isotopic abundance of each of the organic constituents (CH_4, C_2H_6, C_3H_8 and n-C_4H_{10}) were measured and are expressed in ppb and parts per mil respectively. The number of samples, range of values, mean, standard deviation, percentage of samples above mean and percentage of the samples falling above standard deviation (depicting anomaly) for each analysed soil gas constituent are summarized in Table 1. Evaluation of adsorbed soil gas data comprises of statistical analysis to identify the light gaseous hydrocarbon concentrations in the data from the samples with background abundances of half their standard deviation values[24]. C_1–C_5 data are interpreted using hydrocarbon cross-plots. Their correlation coefficients are excellent statistical tools for determining how well the grouped hydrocarbon pairs correlate with each other and also help in determining their sources[25]. The light hydrocarbon ratios are used to predict the oil/gas potential of the basin.

The concentrations of light gaseous hydrocarbons, i.e. C_1 to C_4 (CH_4, C_2H_6, C_3H_8 and n-C_4H_{10}) in the near-surface soils shows that CH_4 is present in 100% of the samples, whereas C_2H_6 in 25.93% of the samples, C_3H_8 in 5.93%, n-C_4H_{10} in 5.19%, n-C_5H_{12} in 5.19% and 26.67% of the samples show $\sum C_{2+}$ content. The concentrations of adsorbed soil gases ranged for methane-C_1 from 2 to 83 ppb, ethane-C_2 from 1 to 92 ppb, propane-C_3 from 1 to 134 ppb, butane-C_4 from 1 to 187 ppb, and sum of hydrocarbon constituents of C_2H_6, C_3H_8 and n-C_4H_{10} and n-C_5H_{12}, excluding CH_4 i.e. $\sum C_{2+}$ from 1 to 729 ppb. The percentages of the samples falling above half of the standard deviation values depict the anomaly for each analysed soil gas constituent. The concentration distribution maps of the individual light gaseous hydrocarbons and also $\sum C_{2+}$ are prepared to define the hydrocarbon potential of the study area (Figures 4–9). Oil/gas wells are also marked on all the maps. The maps reveal that the soils collected near oil/gas wells and the surrounding areas of the K–G basin showed good concentration of adsorbed light hydrocarbon gases near Kaza, Palakollu

Table 2. Pearson correlation coefficient for adsorbed soil gas in the study area

	C_1 (CH_4)	C_2 (C_2H_6)	C_3 (C_3H_8)	C_4 (C_4H_{10})	C_5 (C_5H_{12})	ΣC_{2+}
C_1	1.00					
C_2	0.40	1.00				
C_3	0.27	0.97	1.00			
C_4	0.25	0.96	0.99	1.00		
C_5	0.22	0.91	0.95	0.98	1.00	
ΣC_{2+}	0.26	0.95	0.99	0.99	0.99	1.00

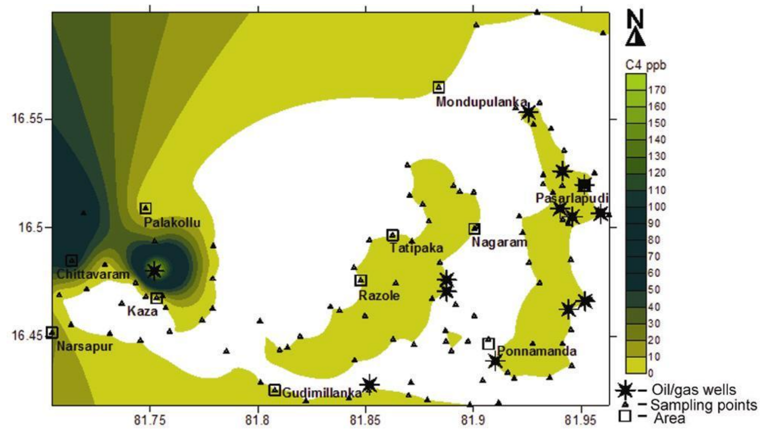

Figure 7. Adsorbed butane concentration map of soil samples from the study area.

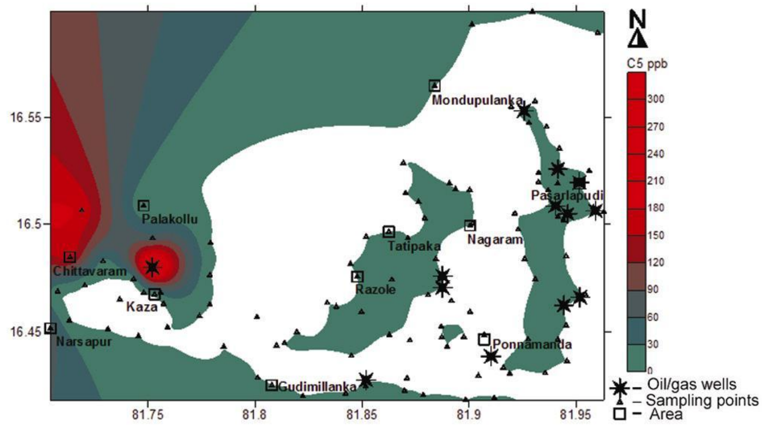

Figure 8. Adsorbed pentane concentration map of soil samples from the study area.

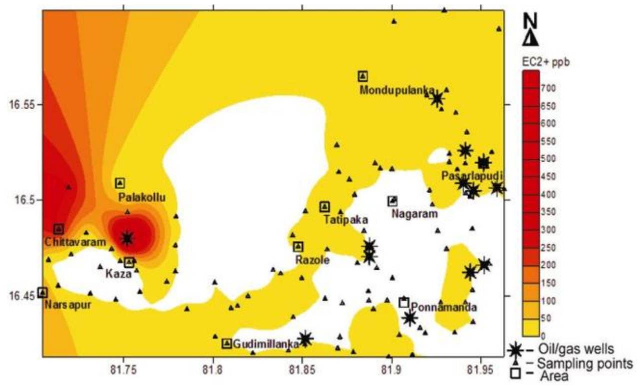

Figure 9. Adsorbed ΣC_{2+} concentration map of soil samples from the study area.

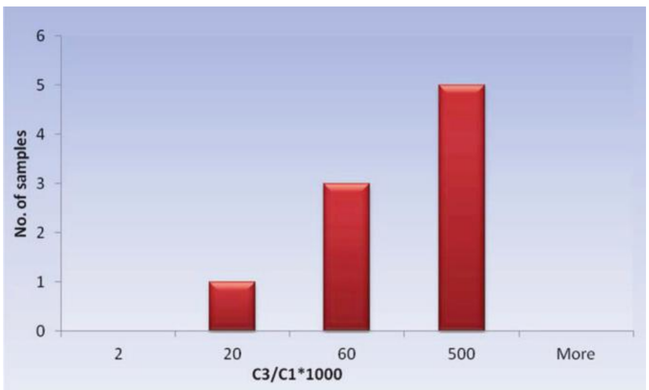

Figure 10. ($C_3/C_1 \times 1000$) ratio to indicate whether the samples fall in oil, oil + gas or gas zones in the study area.

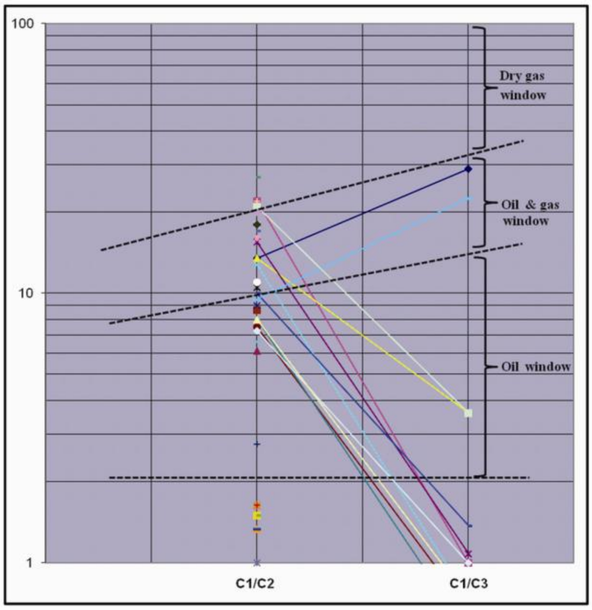

Figure 11. Pixler's plot for discriminating oil, oil and gas and gas windows using C_1/C_2 and C_1/C_3 ratios.

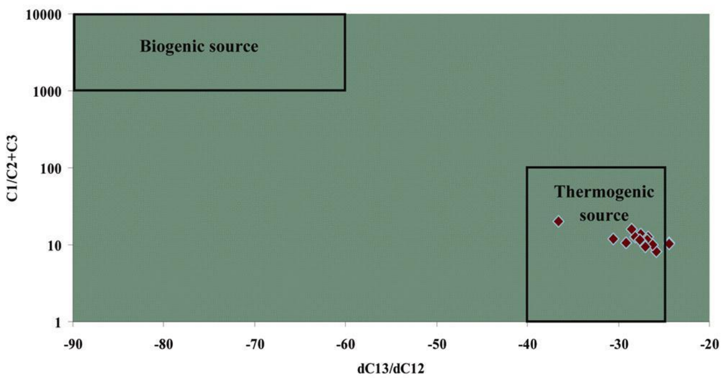

Figure 12. Bernard plot indicating the possible source for the light gaseous hydrocarbons in the study area.

and Chittavaram areas away from the oil/gas wells, indicating that the microseepage occurred laterally and not vertically due to the existing vertical pressure-driven active wells of the study area.

The classification of compositional ratio of $C_3/C_1 \times 1000$ by Jones and Drozd[2] predicts that the $(C_3/C_1 \times 1000)$ ratio of 60–500 is characteristic of oil zone, 20–60 of gas-condensate zone, and 2–20 of dry gas zone. The $(C_3/C_1) \times 1000$ ratios obtained for 135 samples showed that 16.66% of the soil samples fall in dry gas zone, 50% fall in oil and gas condensate zone and 83.33% fall in oil zone (Figure 10). Pixler's[26] plot (Figure 11) is indicative of the zone of occurrence of the accumulated hydrocarbons. Here, the samples fall in oil and oil-gas zone. The Pearson correlation coefficients of various light hydrocarbon components are presented in Table 2. It is clear that excellent correlation exists among ethane, propane, butane and pentane, but there is poor correlation with methane. Such correlations based upon a large number of samples indicate that these hydrocarbons most probably share a single source. The light hydrocarbon ratios are generally used to discriminate biogenic hydrocarbons from thermogenic hydrocarbons and can also predict the oil/gas potential of the basin. The cross-plots between C_2, C_3, nC_4, nC_5 and $\sum C_{2+}$ show excellent correlation ($r = > 0.9$) indicating that (i) these hydrocarbons are genetically related; (ii) they are not affected by secondary alteration during migration from subsurface and subsequent adsorption in the surface soil, and (iii) they might have been generated from a thermogenic source because of the presence of C_3 through nC_5 components.

Bernard[27] suggested a genetic diagram for isotopic abundances of light gaseous hydrocarbons by correlating gas wetness, i.e. $C_1/(C_2 + C_3)$ ratios with the $\delta^{13}C$ of methane to classify natural gas into biogenic or thermogenic types. Molecular ratios of $C_1/(C_2 + C_3)$ less than 100 and $\delta^{13}C_1$ values between –25 and –50‰ (PDB) are typical for thermogenic hydrocarbon gases, while $C_1/(C_2 + C_3)$ above 1000 with $\delta^{13}C_1$ values between –60 and –85‰ (PDB) are indicative of biogenic origin. Biogenic methane usually has an isotopically depleted $\delta^{13}C$ ratio of more than –60‰. The carbon isotopic composition of $\delta^{13}C_1$ (methane) in samples of the study area ranged between –36.6‰ and –22.7‰ (PDB), indicating thermogenic origin. Evidence is in favour of considering the adsorbed soil gases as catagenetic. The isotopic values of the samples also are characteristic of thermogenic origin as seen in Figure 12.

Conclusions

The K–G Basin possesses oil and gas-prone source rocks ranging in age from Permo-Carboniferous to Plio-Pleistocene, some of which are proven and others are not directly linked to discoveries and remain as defined potential sources. The maps reveal that the soils collected near oil/gas wells and the surrounding areas of the K–G Basin showed good concentrations of adsorbed light hydrocarbon gases near Kaza, Palakollu and Chittavaram areas. The carbon isotopic composition of $\delta^{13}C_1$ (methane) in samples ranged between –36.6‰ and –22.7‰ (PDB), indicating thermogenic origin, and that light gaseous hydrocarbons migrated from the subsurface hydrocarbon pools and not any other source. The $C_3/C_1 \times 1000$ values of the adsorbed gases in the light of studies by Jones and Drodz[2] indicate that they are from a gas condensate zone (which has both oil and gas) and also oil zone; also Pixler's and Bernard diagrams for light hydrocarbons and isotopic carbon ratio ($\delta^{13}C$) respectively, indicate that the samples fall in the oil and gas-condensate zone and are of thermogenic nature. Thus, an interesting correlation of the observed surface light hydrocarbon concentrations with the migrated oil and gas from the subsurface oil and gas accumulations in the study area demonstrates the usefulness of adsorbed soil gas technique in petroleum exploration.

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