Geochemical assessment of light gaseous hydrocarbons in near-surface soils of Kutch–Saurashtra: Implication for hydrocarbon prospects

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Light hydrocarbons in soil have been used as direct indicators in geochemical hydrocarbon exploration, which remains an unconventional path in the petroleum industry. The occurrence of adsorbed soil gases, methane and heavier homologues were recorded in the near-surface soil samples collected from Kutch–Saurashtra, India. Soil gas alkanes were interpreted to be derived from deep-seated hydrocarbon sources and have migrated to the surface through structural discontinuities. The source of hydrocarbons is assessed to be thermogenic and could have been primarily derived from humic organic matter with partial contribution from sapropelic matter. Gas chromatographic analyses of hydrocarbons desorbed from soil samples through acid extraction technique showed the presence of methane through *n*-butane and the observed concentrations (in ppb) vary from: methane (C_1) from 4–291, ethane (C_2) from 0–84, propane (C_3) from 0–37, i-butane (iC_4) from 0–5 and *n*-butane (nC_4) from 0–4. Carbon isotopes measured for methane and ethane by GC-C-IRMS, range between −42.9‰ to −13.3‰ (Pee Dee Belemnite – PDB) and −21.2‰ to −12.4‰ (PDB), respectively. The increased occurrence of hydrocarbons in the areas near Anjar of Kutch and the area south to Rajkot of Saurashtra signifies the area potential for oil and gas.

1. Introduction

Hydrocarbon gases are the important constituents of petroleum and can be used as geochemical markers for hydrocarbon migration and accumulation in the earth's crust. The light gaseous hydrocarbons (methane through pentane) migrate to the surface from the subsurface through microseepage and get adsorbed in the near-surface soil matrix, which on further quantification gives an inference on the potential of the area (Madhavi et al. 2009; Kalpana et al. 2010; Prasanna et al. 2010; Madhavi et al. 2011). Tectonic features such as faults and fractures provide a pathway for hydrocarbon migration. These hydrocarbons may be produced by thermogenic or biogenic action on the source organic matter at depth under pressure, which is reflected in its carbon isotopic composition that is helpful in finding out the maturity and type of organic matter that acted as precursor for the hydrocarbons (Stahl 1979). The isotope ratios are characteristic of genesis of the gas even if the gas has migrated over a long distance since the carbon isotope fractionation caused by migration of natural gas is small in contrast to possible large changes in hydrocarbon compositions (Stahl 1974; Bernard *et al.* 1976; Schoell 1983a, 1983b; Clayton *et al.* 1990; Abrams 1996a, 1996b). All

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economically important oil accumulations are the result of thermal maturation processes and methane is a product that originates during all stages of the maturation of organic matter (Fuex 1977).

Surface geochemical techniques are instrumental in assessing the area's potential of hydrocarbons in frontier basins and highly effective in regions where geophysical studies are not useful due to geological or environmental factors. In the present study, we have evaluated the hydrocarbon potential of Kutch–Saurashtra using surface geochemical studies.

2. Geological setting of the study area

The Kutch and Saurashtra regions are tropical monsoonic and can be ecoclimatically classified as arid and semi-arid, respectively. Alluvial and black soils, respectively, predominate in the areas of Kutch and Saurashtra and clay minerals such as illite and smectite are present. Vegetation, in general, is associated with the rainfall pattern of the region. Dang forests with perennial type of vegetations are found in southern region. Crops such as sugarcane and cotton; and orchards such as banana, mango, sapota, etc. are the prominent crops in this region. Gir forests are found in less rainfall area of Saurashtra region. The crops grown are of short duration. Xerophytic type of vegetations are found in minimum rainfall area of northern part and Kutch (Hardikar and Pandey 2008).

The Saurashtra peninsula is located in the western periphery of India in the state of Gujarat (figure 1). Saurashtra is a pericratonic rifted passive margin basin comprising an onshore area of 52,000 km^2 and a 20,000 km^2 offshore extension into the Arabian Sea (Biswas 1982; Merh 1995). The Kutch basin is a pericratonic rift basin situated in the western margin of India. Nagar Parkar uplift in the north and Kathiawar uplift (Saurashtra horst) in the south, respectively, along Nagar Parkar and North Kathiawar faults delimit the E–W rift (Biswas 2005). The Kutch rift was initiated during the Late Triassic break-up of the Gondwanaland by the reactivation of primordial faults in the Precambrian Delhi fold belt. The rifting was aborted during Late Cretaceous pre-collision stage of the Indian plate. During post-collision compressive regime of the Indian plate, the Kutch rift basin became a shear zone with strike-slip movements along sub-parallel rift faults. The Kutch Mainland Fault along the rift axis became the active principal fault.

The peninsula developed by rifting along the three intersecting Precambrian orogenic trends, the ENE–WSW Narmada Son lineament, the west coast fault with the NNW–SSE Dharwar trend and the NE–SW Delhi Aravalli trend (Biswas 1980, 1987; Rao and Tewari 2005), at different stages

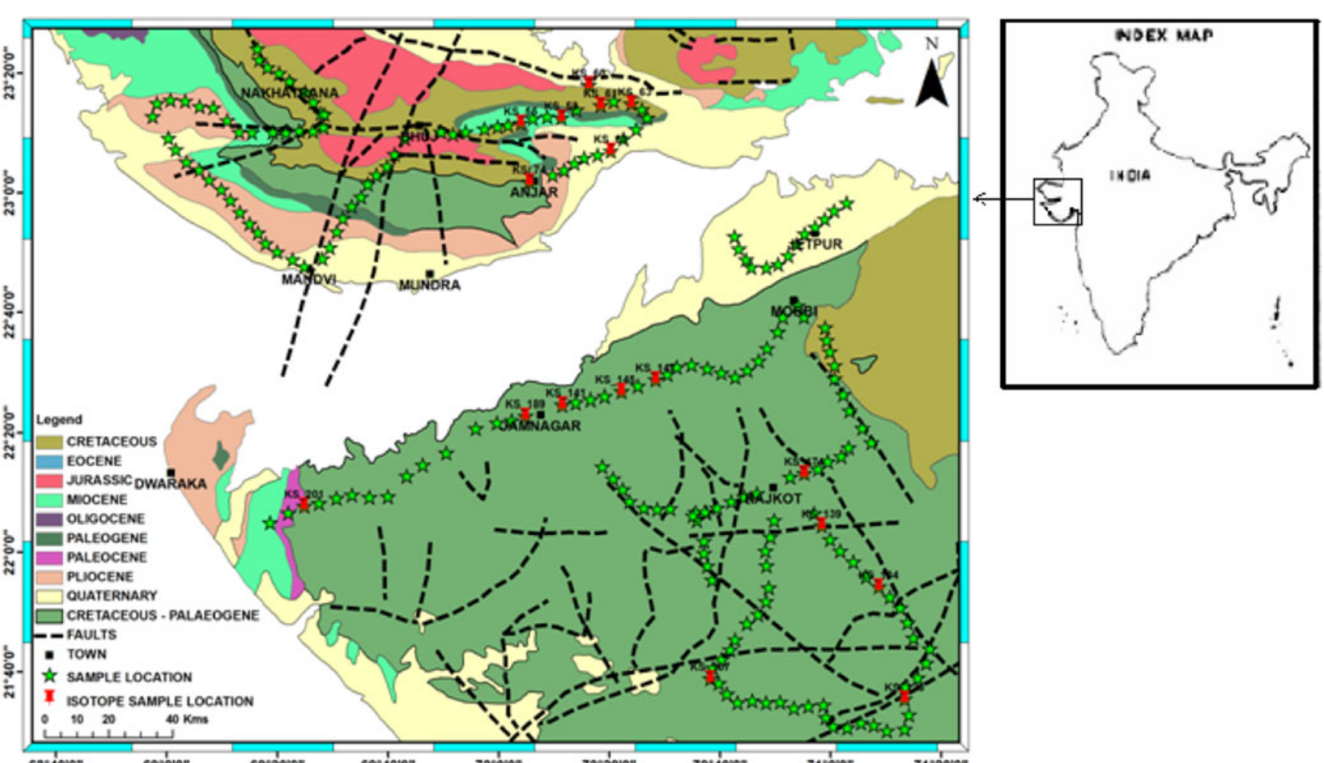


Figure 1. Geological map of Kutch–Saurashtra basin with sample location points.

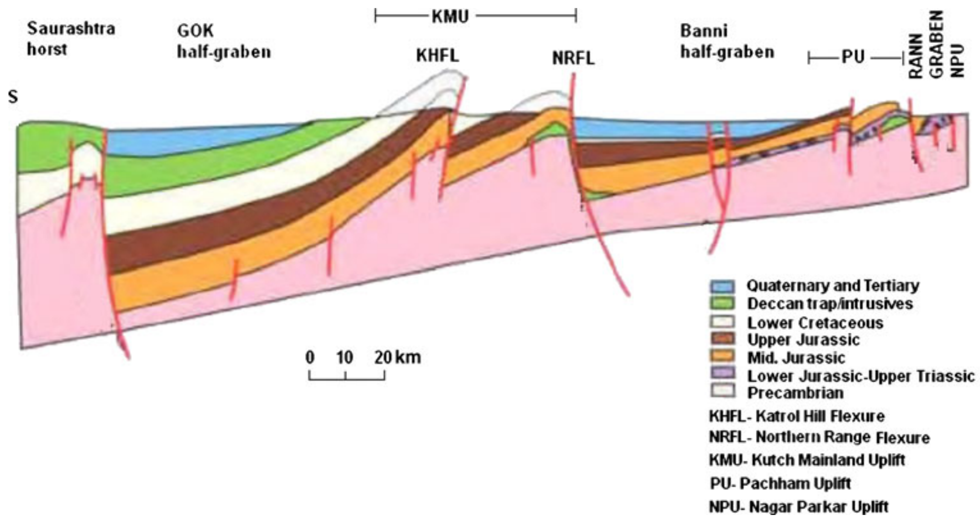


Figure 2. Geological cross-section across Kutch basin.

of evolution of the Indian sub-continent and their subsequent reactivation during the Mesozoic and Tertiary times. Bounded by major faults and rift basins, the northern limit of Saurashtra is marked by the faulted margin of Kutch and the southern coast runs parallel to the extension of Narmada geofracture. The eastern side is flanked by the Western Cambay basin bounding fault. Stratigraphically, the basin contains Mesozoic and Cenozoic rocks and Lower Cretaceous and Tertiary sediments along with the Deccan Trap Formations overlying the Precambrian basement. It is overlain by the Wadhwan Formation of marine origin comprising limestone bands within sand and shale sequences (Biswas and Deshpande 1983). The Wadhwan Formation occurs as a marine tongue within an overall deltaic system. The Deccan Trap Formations occupy most of the exposed area of Saurashtra, consisting of alternating basaltic flows and intervening sedimentary depositions.

Sedimentation under deltaic conditions with marine interfingering environment is a favourable phenomenon for the generation of hydrocarbons. The depositional environment of the Dhrangdhra Formation is similar to the Bhuj Formation of the Kutch and the Serau and the Viramgam Formations of the Cambay Basin, which are highly prospective and producing in terms of hydrocarbon reserves. The burial and thermal effect of the Deccan Volcanics may have enhanced the maturation process of hydrocarbons, besides proving to be a constructive factor in preserving the underlying Mesozoic strata and hydrocarbons from exposure and erosion (Zutshi *et al.* 1989; Zutshi 1991).

The stratigraphy of Kutch basin comprises strata ranging in age from the middle Jurassic to Holocene (figure 2). The middle Jurassic sediments comprise both clastic and carbonate facies followed by mainly sandstone and shales of upper Jurassic. The entire sequence is characterized by a major transgressive cycle followed by a regression. The sedimentary thickness of the upper Jurassic and Cretaceous age greatly increases towards the west. The grey and khaki shales of Jhurio and older Kaladonger Formation might form source rocks in the deeper basinal parts of the west. The Upper Jurassic shales of Jhuran Formation contain sufficient organic matter required for hydrocarbon generation. The bedded and the massive sandstones of the Upper member and Katesar Member of Jhuran Formation overlying these shales are good and readily available reservoirs (Biswas and Deshpande 1983).

The Cretaceous sequence representing deltaic deposits is made up of sandstones with minor shales. This prograding deltaic sequence characterized by rapid sedimentation of subsiding basin marine intertonguing beds, represents the Ukra member. This in turn formed favourable tectono-sedimentary environments for hydrocarbon generation and entrapment. The requisite heat generation due to Deccan trap volcanism during Late Cretaceous greatly aided the thermal maturation processing of the organic content, and thus further increasing the Cretaceous prospects of western Kutch.

Table 1. *Statistical summary of adsorbed soil gas data (in ppb).*

	C1	C2	C3	iC4	nC4
Minimum	4	0	0	0	0
Maximum	291	84	37	5	4
Mean	51	6	2	0.04	0.02
Standard deviation	50	13	6	0.39	0.30
Mean+Standard deviation	101	19	8	0.43	0.32

Table 2. *Correlation coefficients of hydrocarbon components.*

r	C_1	C_2	C_3	C_{2+}
C_1	1			
C_2	0.91	1		
C_3	0.88	0.98	1	
C_{2+}	0.9	0.99	0.99	1

3. Materials and methods

A total of 203 soil core samples were collected with a sample spacing of 4–5 km along the existing roads in reconnaissance pattern, by manual hammering of hollow metal pipes to 1–2 m depth with their global positioning system locations marked. Samples were wrapped in aluminum foils and sealed in poly-metal packs for analysis in the laboratory. The sample locations are marked on the geological map of Kutch–Saurashtra basin (figure 1). The analysis of adsorbed soil gases and determination of their carbon isotopic ratio is based on acid extractable hydrocarbons occluded in soil particles (Horvitz 1981) and carried out by gas chromatography and GC-C-IRMS, respectively. A sample of 1 gm of 63 μm wet sieved soil is treated with ortho-phosphoric acid for the desorption of light gaseous hydrocarbons and the evolved CO_2 by the decomposition of carbonates present in the soil is trapped in KOH provided in the side limb of the degasification apparatus. The gaseous hydrocarbons desorbed from the soil sample are collected in the graduated tube fitted with rubber septa through water displacement. The analyses were carried out on Varian 3380 Gas Chromatograph equipped with a flame ionization detector. The GC-C-IRMS comprises a Agilent 6890 Gas Chromatograph coupled to a Finnigan-Delta PlusXP Isotope Ratio Mass Spectrometer via a GC combustion III interface (Madhavi *et al.* 2009).

4. Results

The gas chromatographic determination of adsorbed light gaseous hydrocarbons in near-surface soil samples of Kutch–Saurashtra area has shown the presence of methane through butanes. The concentrations (ppb) of methane (C_1) varied from 4–291, ethane (C_2) from 0–84, propane (C_3) from 0–37, *i*-butane (iC_4) from 0–5 and *n*-butane (nC_4) from 0–4. The statistical parameters of the adsorbed soil gas hydrocarbon data is given in table 1.

Histograms of hydrocarbon components are helpful in evaluation of frequency distribution of variables in the obtained dataset. In near-normal mode of frequency distribution, mean represents the background and the samples with one or two standard deviations are considered to be anomalous (Abrams 2005; Mani *et al.* 2011a). Histograms

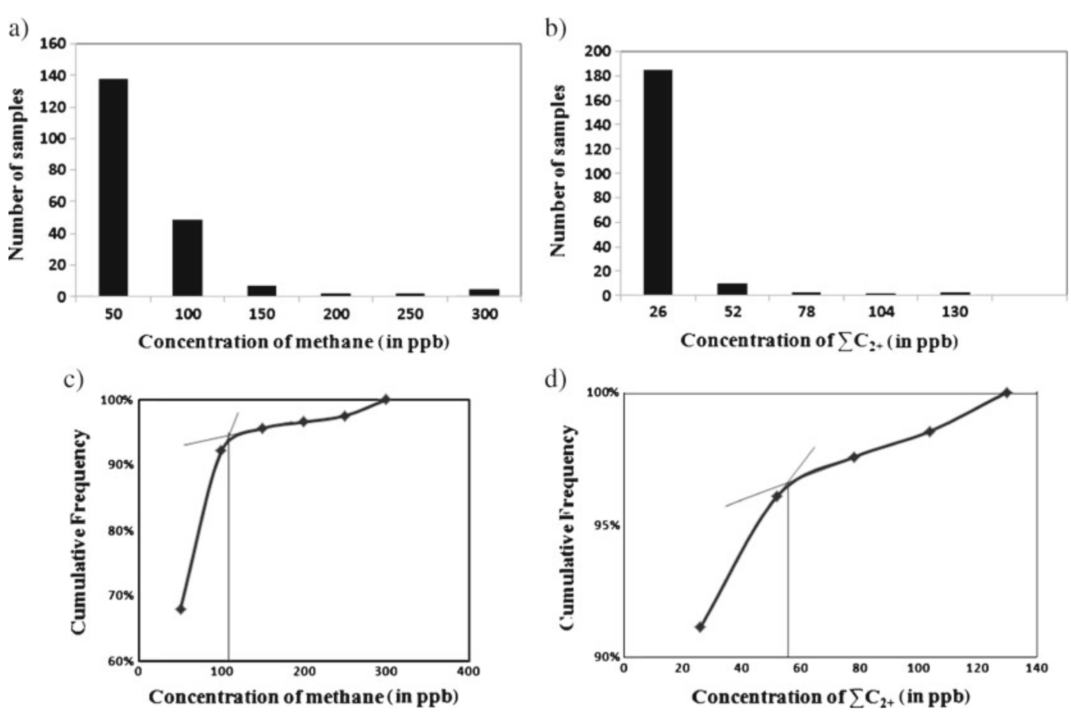


Figure 3. (**a**) Frequency distribution pattern of methane. (**b**) Frequency distribution pattern of $\sum C_{2+}$. (**c**) Cumulative frequency plot of methane. (**d**) Cumulative frequency plot of $\sum C_{2+}$.

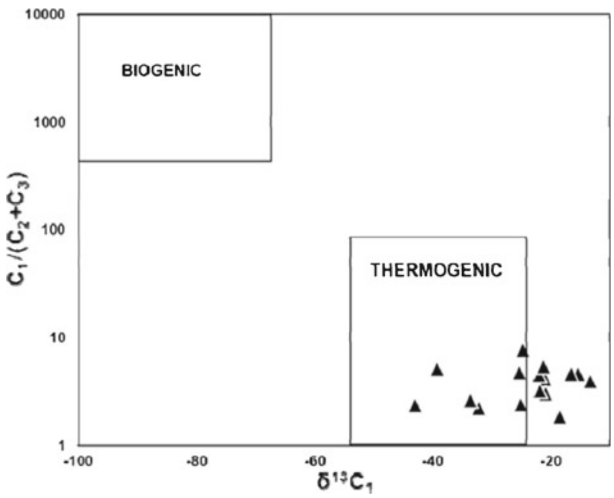


Figure 4. Bernard plot differentiating thermogenic and biogenic sources of hydrocarbons.

of methane and ΣC_{2+} have shown an asymmetrical curve with positive skewness.

The cross plots among the hydrocarbon components help in determining the genetic relationship among the hydrocarbons and their origin through the linearity existing between them (Madhavi *et al.* 2009; Kalpana *et al.* 2010; Prasanna *et al.* 2010). The correlation coefficients for hydrocarbon components are given in table 2, which shows good linear correlation (r).

In the present study, 16 soil samples having high concentrations of hydrocarbons have been analysed for carbon isotopic signatures of methane and ethane. The carbon isotopic values for methane and ethane ranged between −42.9‰ to −13.3‰ and −21.2‰ to −12.4‰ with respect to Vienna Pee Dee Belemnite (VPDB), respectively, indicating a thermogenic source for the hydrocarbons.

5. Discussion

5.1 *Adsorbed soil gas data*

Histograms of methane and ΣC_{2+} given in figure 3(a, b) with one standard deviation and mean plus one standard deviation, respectively (C_1: 50 ppb and ΣC_{2+}: 26 ppb), as class interval shows an asymmetrical curve with positive skewness and the values skewed to the right are considered to be anomalous.

A cumulative frequency scale in probability plot is a linear transformation of normal distribution and the normally distributed population is shown as a straight line and any change in slope reflects dataset deviation from statistical normality (Rice *et al.* 2002). The probability plots of methane and ΣC_{2+} given in figure 3(c, d) show change in linearity at 100 and 60 ppb, respectively, differentiating the background population from the anomalous ones represented by an upward deflection.

The correlation coefficient for C_1–C_2, C_1–C_3, C_2–C_3, C_1–ΣC_{2+}, C_2–ΣC_{2+}, C_3–ΣC_{2+} (table 2) shows good linear correlation (r) and indicates that (i) these hydrocarbons are genetically related, (ii) they are not affected by secondary alteration during their migration from the subsurface, and (iii) they may have been generated thermogenically.

Table 3. *Concentration of light gaseous hydrocarbons desorbed from selected soil samples of Kutch–Saurashtra (in ppb) with carbon isotope values of methane and ethane.*

Sl. no.	Sample Id	C_1	C_2	C_3	$C_1/(C_2+C_3)$	$\delta^{13}C_1$	$\delta^{13}C_2$
1	KS-55	243	74	36	2	−32.20	−19.23
2	KS-58	45	12	7	2	−42.99	−21.06
3	KS-60	147	22	9	5	−25.41	−20.21
4	KS-61	79	20	10	3	−33.66	−21.27
5	KS-63	69	11	4	5	−16.57	–
6	KS-74	193	19	6	8	−24.77	–
7	KS-107	273	48	18	4	−21.02	–
8	KS-125	281	49	22	4	−13.34	–
9	KS-134	291	84	37	2	−25.16	−16.64
10	KS-139	210	47	22	3	−20.94	−15.78
11	KS-141	73	11	5	5	−21.98	−16.04
12	KS-145	69	11	4	5	−15.45	–
13	KS-147	93	14	4	5	−39.31	–
14	KS-174	145	53	26	2	−18.55	–
15	KS-189	61	13	6	3	−21.90	−12.49
16	KS-201	65	8	4	5	−21.35	−16.10

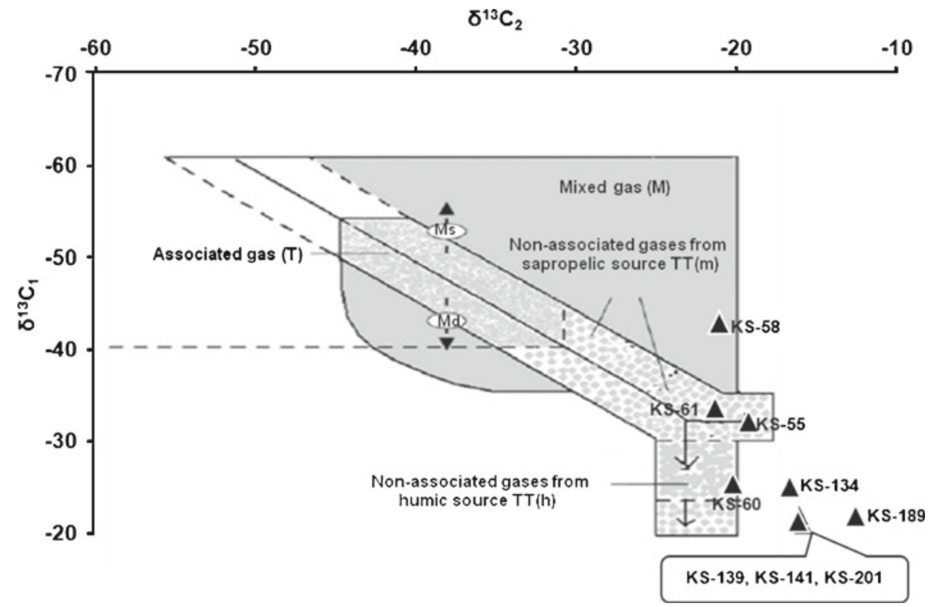


Figure 5. Schoell diagram for genetic characterization of hydrocarbons.

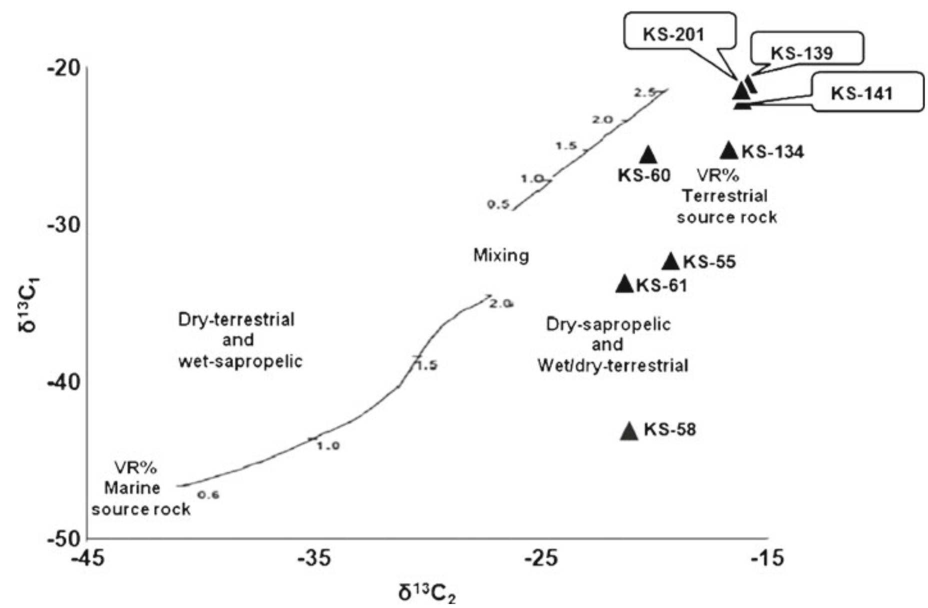
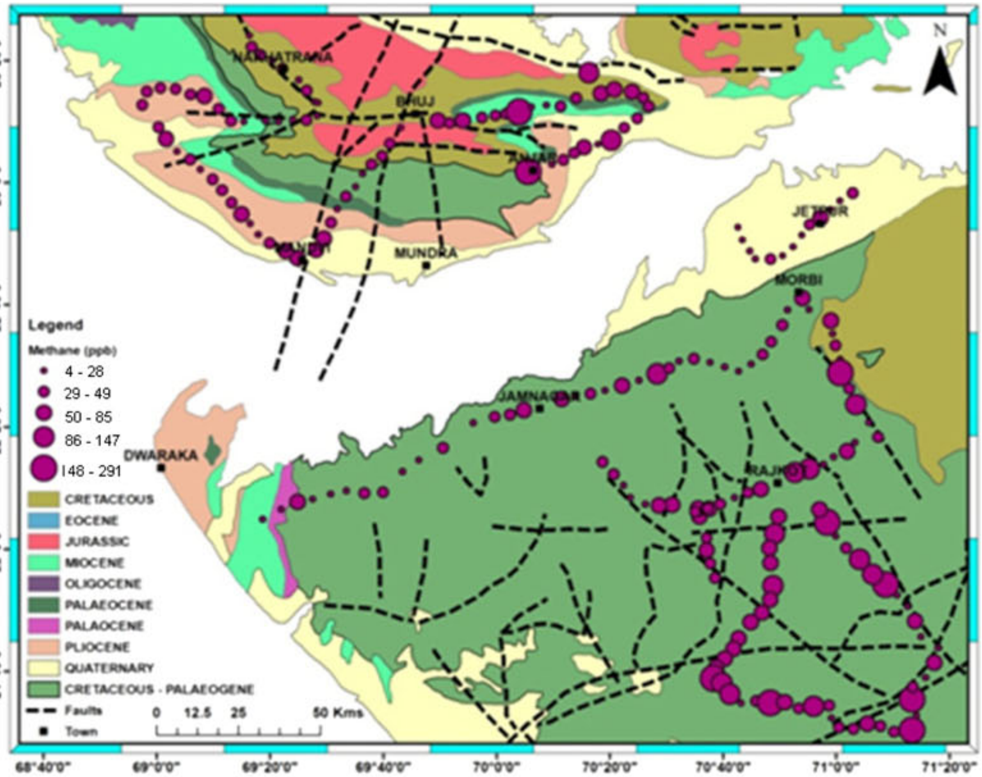


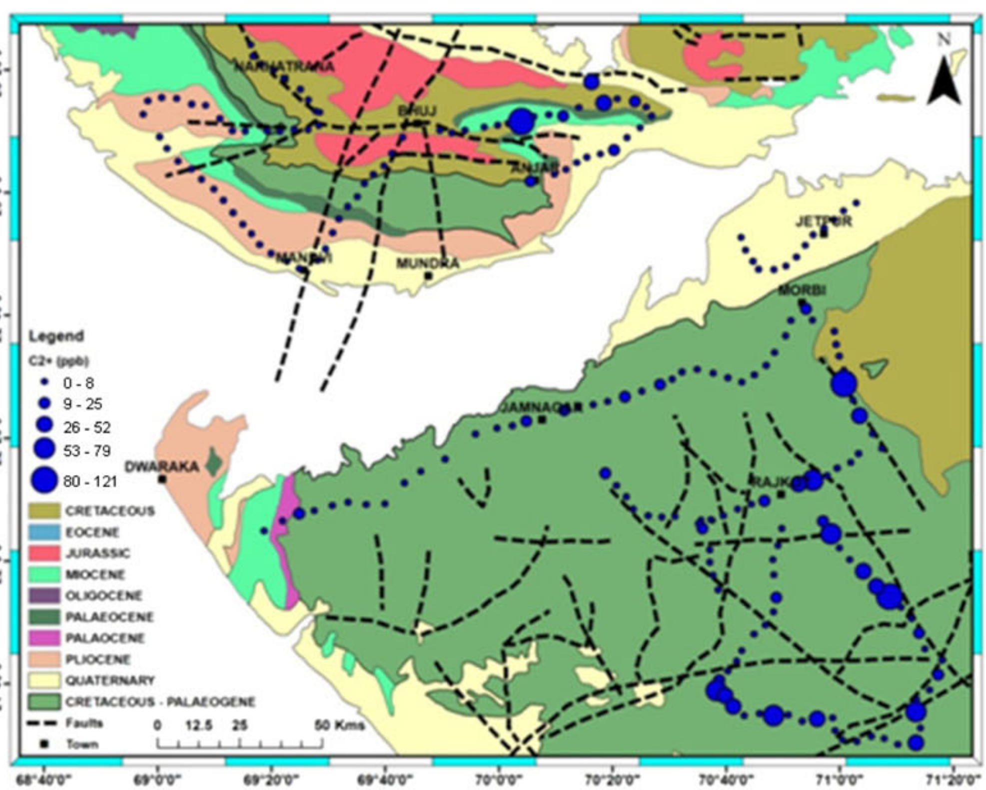
Figure 6. Plot of carbon isotope ratios of methane *versus* ethane along with maturity relationships for adsorbed hydrocarbon gases.

5.2 *Carbon isotopic data*

Using the compositional ratio of C_1/C_2+C_3 and $\delta^{13}C$ value of methane, two major distinct sources of hydrocarbons were categorized, thermogenic and biogenic by Bernard (1978). Microbial degradation produces hydrocarbon gas with $C_1/(C_2+C_3)$ ratios greater than 1000 and contains almost exclusively methane with very low concentrations of ethane, propane and butane (C_{2+}). The hydrocarbons generated by thermal alteration of organic matter under high temperatures and pressures produces methane along with correspondingly high concentrations of ethane, propane and butanes showing the compositional ratio $C_1/C_2+C_3 < 100$ and methane usually having a quite isotopically enriched ^{13}C ratio of greater than −60‰ (Mani *et al.* 2011b). Methane is a common soil gas found in the near-surface, produced by the action of bacteria on organic matter. Biogenically produced methane may mix with the petroleum-generated gases seeping to the surface. Mixing of two gases



a)



b)

Figure 7. Concentration distribution map of (**a**) methane in Kutch–Saurashtra basin and (**b**) C_{2+} hydrocarbons in Kutch–Saurashtra basin.

of different origins results in the variation of isotopic composition and indicates that the constituents of the gas are not cogenetic. If bacterial methane is added to thermal methane, δ^{13}C value of methane varies and the δ^{13}C value of ethane remains constant.

The methane desorbed from soil samples of Kutch–Saurashtra when plotted on Bernard plot has shown a ratio of C_1/C_2+C_3 from 2 to 8 and $\delta^{13}C_1$ ranging between −42.9‰ and −13.3‰ (figure 4) and suggests a thermogenic source for hydrocarbons. Few samples show enriched isotopic values which might be due to microbial oxidation (methanotrophic oxidation) of methane. The compositional ratios, carbon isotopic values of methane and ethane are given in table 3.

Certain standard models of carbon isotopic ratios of methane and ethane which determine the nature of source material and the maturity of the source rock (vitrinite reflectance, Ro) have been considered and applied to the gases desorbed from soil samples of Kutch–Saurashtra. Genetic characterization of hydrocarbon gases with regard to their specific origin (terrestrial or marine) proposed by Schoell (1983a), when applied for the gases of Kutch–Saurashtra have shown to be of non-associated gases of humic source TT(h) (figure 5). The samples KS-55 and KS-61 are representative of non-associated gas (gas derived from cracking reactions of kerogen) derived from sapropelic source. The sample KS-58 in the mixed zone represents non-associated gas derived from sapropelic source, but small amount of shallow gas might have been added to the thermogenic gas resulting in depletion of the isotopic value and the remaining samples are supposed to be non-associated gases derived from humic source where δ^{13}C enrichment is seen in methane and ethane. δ^{13}C enrichment is found in thermal methane derived from humic source rocks compared to sapropelic source rocks which may be accountable for the chemical differences in kerogen structure. Oil producing type-I is rich in aliphatics (sapropelic), whereas gas producing type III (humic) rich in aromatic structures is rich in δ^{13}C isotopes (Stahl 1977). Enrichment of δ^{13}C in methane might also occur due to the bacterial oxidation of hydrocarbons during the migration of gas across the sediment to the near-surface, as the partial consumption of methane by oxidation can significantly shift carbon isotopic ratios positively in the residual methane (Barker and Fritz 1981), thereby decreasing the concentration of methane compared to ethane and propane.

Berner and Faber (1996) proposed a maturity relationship plot considering the carbon isotopic signatures of methane and ethane that demonstrates the source organic material of the gases. The above plot when applied to the gases desorbed from the soil samples of Kutch–Saurashtra showed that the source of these gases could be terrestrial organic material with the maturity level of 1.5 to 2.5 vitrinite reflectance, with partial contribution from marine organic material. One sample KS-58 is characteristic of marine source rock, two samples – KS-55 and KS-61 are characteristic of a mixed source (figure 6) and remaining samples are representative of terrestrial source rock.

Concentration distribution maps of methane and $\sum C_{2+}$ are helpful in finding out the areas with high hydrocarbon potential. High concentrations of hydrocarbons were found to be characteristic of areas near Anjar of Kutch and the area south of Rajkot of Saurashtra as shown in the concentration distribution maps of methane and $\sum C_{2+}$ hydrocarbons (figure 7a, b) signifying the area's potential for hydrocarbons. Geological and magnetotelluric surveys in the Saurashtra peninsula have shown the presence of thick Mesozoic sedimentary sequences in the western part (Harinarayana 2008; Rao and Reddy 2005), and led to the drilling of the Lodhika-1 well near Rajkot (Mani *et al.* 2012) which confirmed the presence of a Mesozoic section beneath the Deccan Traps which could have acted as source rock for the generation of hydrocarbons.

The faults present over the southeastern area of Saurashtra could have provided way for the migration of hydrocarbons from subsurface to the surface, thereby depicting high concentration of hydrocarbons. Considering all the probabilities, the area seems to show positive prospects in terms of hydrocarbon reserves and needs detailed studies along with other geophysical data.

6. Conclusions

The study area in Kutch–Saurashtra has shown the presence of methane through butanes in the soil samples and suggests thermogenic generation of hydrocarbons. The linearity between hydrocarbon components shows genetic relation among them and that they are derived from a single source. The thermogenic origin of the hydrocarbons is further exhibited by the carbon isotopic signatures of methane and ethane. The source material could be terrestrial organic material as indicated by the standard isotope models. The areas near Anjar of Kutch and the area south to Rajkot of Saurashtra are characteristic of high concentrations of hydrocarbons indicating the area to be prospective for hydrocarbons.

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