Methane in soil gas and its transfer to the atmosphere in the Yakela condensed gas field in the Tarim Basin, Northwest China

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Abstract: In this study, by analyzing CH_4 concentration and $\delta^{13}C_{CH_4}$ in soil-gas profiles, the potentials of CH_4 gas transfer from ground to atmosphere were studied at four representative sectors in the Yakela condensed gas field in the Tarim Basin, Xinjiang, China. These are: 1) the oil–gas interface sector, 2) fault sector, 3) oil–water interface sector, 4) an external area. Variation in CH_4 in soil-gas profiles showed that CH_4 microseepage resulted from the migration of subsurface hydrocarbon from deep-buried reservoirs to the earth's surface. It was found that CH_4 from deep-buried reservoirs could migrate upwards to the surface through faults, fissures and permeable rocks, during which some CH_4 was oxidized and the unoxidized methane remained in the soil or was emitted into the atmosphere. The lowest level of CH_4 at the soil-gas profile was found at the CH_4 gas-phase equilibrium point at which the CH_4 migration upwards from deep-buried reservoirs and the CH_4 diffusion downwards from the atmosphere met. The $\delta^{13}C_{CH_4}$ and ethane, propane in soil gas exhibited thermogenic characteristics, suggesting the occurrence of CH_4 microseepage from deep-buried reservoirs. A linear correlation analysis between CH_4 concentrations in soil gas and temperature, moisture, pH, Eh, Ec and particle size of soil indicated that both soil Eh and soil temperature could affect CH_4 concentration in soil gas while soil pH could indirectly influence soil methanotrophic oxidation via impacting soil Eh.

Key words: Soil gas, CH_4 concentration, carbon isotope, microseepage, oil reservoir

1 Introduction

During the past two decades, geologic CH_4 emission has always been considered as a negligible contributor to CH_4 concentration in the atmosphere. According to the Second and Third IPCC Assessment Report (Lelieveld et al, 1998), methane hydrate was found to be a minor source of natural geologic methane, only accounting for about 0.9% of the total atmospheric methane budget. However, recent studies have shown that natural geologic emissions of CH_4 could play an important role in the atmospheric methane budget, mainly due to CH_4 emissions from petroleum seepage through faults, fissures and permeable rocks, mud volcanism, marine seeps and geothermal manifestations. Meanwhile, these geologic CH_4 emissions may represent an important component of the 'missing' source of fossil CH_4 (radiocarbon-free), as recently recognized in the atmospheric budget (Etiope et al, 2008). In the Fourth Assessment Report of IPCC, geological CH_4 sources have been considered as the second highest natural

source for CH_4 emissions after wetlands, while geological seepage has been recognized as a new category in the UNECE/EMEP Task Force Emission Inventory Guidebook (Etiope, 2009; Etiope et al, 2008; Etiope and Ciccioli, 2009; Etiope and Klusman, 2010).

During the 1920s and 1930s, studies suggested that a close correlation existed between concentration anomalies of hydrocarbon gases near the surface of the earth and deep-buried oil and gas reservoirs. Soil gas methane has been an important indicator of deep-buried reservoirs (Laubmeyer, 1933; Klusman, 1993; Hunt, 1996; Abrams, 2005). It was not until recently that numerous field surveys have been conducted on CH_4 flux from petroleum-bearing sedimentary basins by researchers in the USA, Europe (Italy, Germany, Greece), and Asia (Azerbaijan, China) (Klusman and Jakel, 1998; Thielemann et al, 2000; Etiope, 2004; 2009; Etiope and Milkov, 2004; Yang et al, 2004; Tang et al, 2007; 2008; 2009; 2010; Etiope and Ciccioli, 2009; Etiope and Klusman, 2010). Now, it has become an international research focus that microseepage of hydrocarbon gas throughout the area related to petroleum-bearing sedimentary basins is an important source of atmospheric CH_4. Microseepage capacity of hydrocarbon gas from deep-buried reservoirs is influenced

by the complexity of geological conditions, instability and regionality of hydrocarbon gas distribution, difference in concentration and pressure of hydrocarbon gas in reservoirs, which is more obvious at faults or broken caprocks. The leakages can lead to particular traces underground and in the surface, such as anomalies of hydrocarbon concentration of soil gas or adsorbed gas in the surface. CH_4 which failed to be oxidized or decomposed may eventually be released into the atmosphere (Klusman, 1993). Our previous CH_4 flux surveys, conducted at different geologic sectors in the Yakela condensed gas field in the Tarim Basin, Northwest China, have shown a spatial variability of microseepage flux in relation to the subsurface geo-structural features and gas-oil setting. The CH_4 flux is higher at faults and areas associated with gas-oil interfaces, and lower over oil-water interfaces (where the gas pressure is lower) and outside the petroleum field boundary (Tang et al, 2007; 2008; 2009; 2010).

As a follow-up work of our previous CH_4 flux surveys conducted in the Yakela condensed gas field (Fig. 1), this study aims to further identify the "extra-soil" CH_4 sources and the potentials of CH_4 gas transfer from ground to the atmosphere by analyzing soil CH_4 concentration and $\delta^{13}C_{CH_4}$ at soil-gas profiles, and by comparing soil CH_4 in soil gas with our previous CH_4 fluxes survey data.
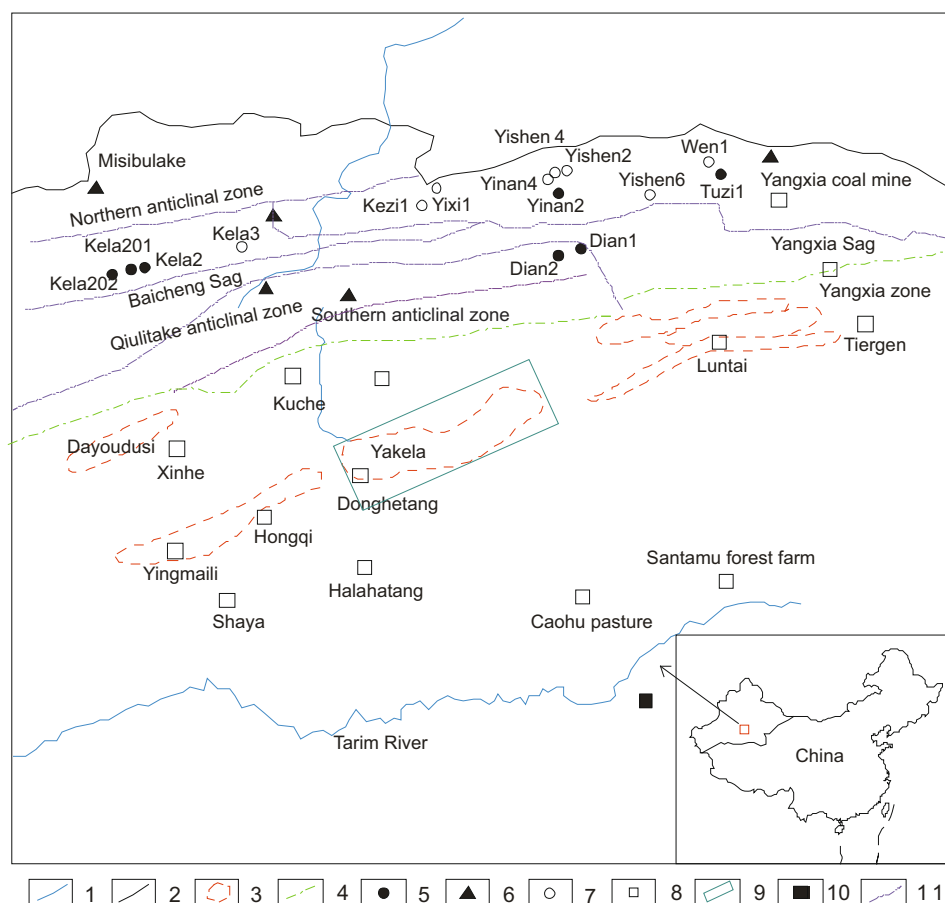
Fig. 1 Sketch geological map showing distribution of the Kuche Depression and main exploratory wells and oil fields (Liang et al, 2004). 1. River, 2. Kuche Depression border, 3. Trap structure, 4. Boundary of onshore (continental) and marine facies areas, 5. Oil/gas well, 6. Oil sand outcrop, 7. Exploratory well, 8. Place name, 9. Study area, 10. Background area, 11. Second-order tectonic units

2 Geologic setting

The Yakela condensed gas field (Fig. 1) lies in the Kuche Depression, northern Tarim Basin in the arid northwest of China which is a distinctive oil accumulation area (Song et al, 1998). This area is characterized by arid climate, high evaporation and infiltration, strong salt-base reaction and low land productivity, thereby resulting in limited biogenetic production of CH_4 near the earth surface. Therefore, the Yakela condensed gas field is an ideal site for investigating geologic CH_4 emission behaviors.

The Yakela condensed gas field is a large high pressure condensed field, which is located tectonically in the Luntai-Yakela faulted uplift zone of the eastern part of the Shaya Upwarping in the northern Tarim Basin (Fig. 1). The zone is a faulted block lying between the southern Luntai Fault and the northern Yaha Fault. There are three types of buried hill condensate reservoirs in the field, namely 1) condensate reservoirs trapped in an anticline structure in the Cretaceous Kabushaliang Group, 2) lithological-structural composite condensate reservoirs in the middle to lower Jurassic, and 3) buried-hill type condensate reservoirs in the upper

Proterozoic, upper Cambrian and lower Ordovician. All these oil/gas traps were formed from carbonate and clastic rock types. The caprocks are mainly of mudstone lithology (mudstone, shaly siltstone, etc) formed in low sedimentation, with evaporites. There are multi-set caprocks from Palaeozoic to Meso-Cainozoic, which are dominated by the Jidike Formation, Kapushaliang Group and lower Jurassic (Chen et al, 2012). Based on the geologic and petroleum reservoir features, soil gas borehole drilling was undertaken during a dry spring season in the following four areas: 1) oil-gas sector, 2) fault sector in correspondence with Luntai fault-linked crossing the deep reservoirs (Deng et al, 2001; Zhu et al, 2012), 3) oil-water sector, 4) an external area, i.e., outside the northern gas field boundaries. Meanwhile, in order to make a comparative study, an area was selected near the Taklamakan Desert for a control study. It is located about 110 kilometers southeast of the Yakela condensed gas field.

3 Methodology

3.1 Field methods

A portable copper probe with an interior diameter of 0.08 m was used for collecting soil gas. At each site, soil gas at depths of 0.3, 0.6, 0.9, 1.2 and 1.5 m were collected for two continuous days at 8:30–12:30 a.m., 1:30–5:30 p.m., 6:30–10:30 p.m. and 0:00–4:00 a.m., respectively. However, at the faults, soil gas samples were only collected during the former three time periods. Two samples of soil gas were collected at each depth during the same mean time period. Meanwhile two air samples were also taken as soil gas samples (0 m at the ground surface).

The detailed sampling procedure was as follows. The lower end of the probe was closed with a steel ball of diameter 0.08 m in order to avoid the probe being blocked by soil when it was inserted into the ground. The probe was inserted into the ground with a sliding hammer and pulled again upwards about 0.10 m to the required depth, so that the steel ball seal opened letting the soil gas diffuse into the probe. The top end of the copper probe was closed with Shimadzu rubber mats. In order for the probe to be fully sealed, tape was used to seal the junction between the copper probe and the rubber mat. Soil gas was extracted by inserting a syringe. Before the soil gas was collected, the air inside the probe was purged to avoid contamination and after about 4-hour's equilibration the soil gas was collected by extracting it with a syringe.

Meanwhile, air temperature and barometric pressure was monitored in order to check potential 'false anomalies' due to atmospheric pumping. Soil properties such as temperature, moisture, porosity, pH, Eh and salinity were also monitored in order to address their potential influence on CH_4 concentration in soil gas.

3.2 Laboratory measurement

Concentrations of CH_4, C_2H_6 and C_3H_8 in soil gases were analyzed by using an HP-5890 gas chromatograph equipped with a flame ionization detector (FID). Standard gas of 2.04×10^{-6} CH_4, 1.01×10^{-6} C_2H_6 and 1.05×10^{-6} C_3H_8, mixed with 99.999% pure He were used for making external standard curves. The working condition was set as: stainless steel column (2 mm–2 m) filled with a 13X molecular sieve of 60/80 meshes, N_2 (99.999%) as load gas with a flow of 30 mL/min, at column temperature of 55 °C, FID detector temperature of 200 °C, and CH_4 measure error of 0.11%–0.25%. To ensure the comparability of data, all concentrations were converted to standard temperature and pressure conditions (STP: 0 °C, 101.325 kPa).

The stable carbon isotope atmospheric CH_4 were measured by using an on-line CH_4 extraction system as described in detail by Tang et al (2006). The system was interfaced with a Thermo Quest GC/TC (Thermo Finnigan, Bremen, Germany) and an isotope ratio mass spectrometer (Thermo Quest Delta plus XP, Thermo Finnigan, Bremen, Germany). The GC–pyrolysis interface (Precon) contains a combustion furnace and is connected to isotope-ratio mass spectrometer (IRMS). Isotope values are given in the d-notation relative to the internationally adopted VPDB standard. The minimum detectable concentration of CH_4 was 5 nmol/L. The $\delta^{13}C$ values were determined with a precision of 0.4‰ (*n*=10).

4 Results and discussion

4.1 Variation characteristics of CH_4 concentrations in soil gas
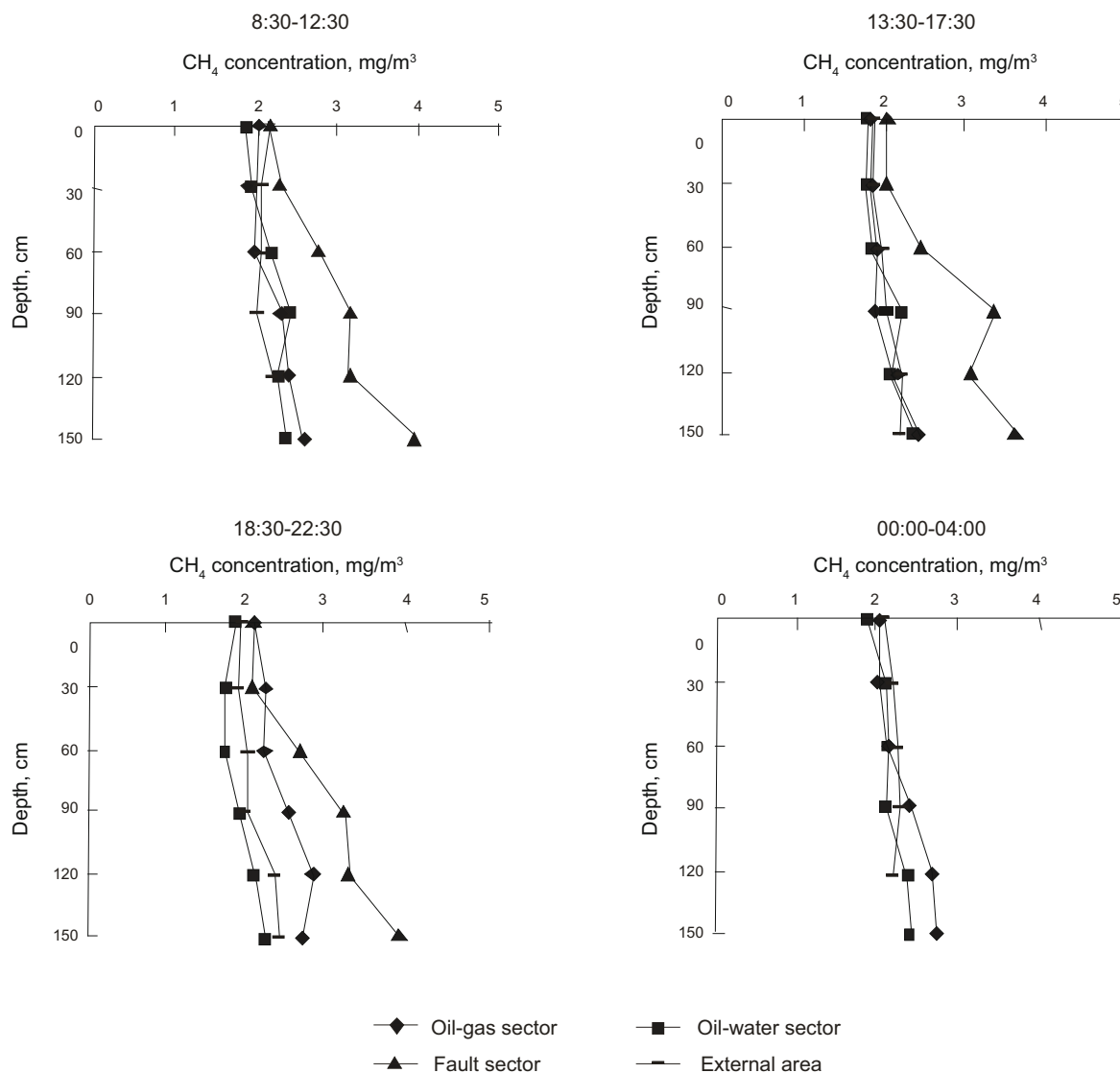
In the Yakela condensed gas field, thick sand strata are widely distributed, predominantly consisting of light loam and sandy loam, while only a slight change in soil textures, which is mainly consisted of light brown fine sandy loam to light loam changing with depth into brown light loam or sandy clay loam. There is only a little difference in thickness of soil layers among the four sites. Therefore, to a certain degree, the CH_4 sources can be easily identified by examining the variation characteristics of CH_4 concentrations at soil gas profiles among the four sites.

Fig. 2 shows the variation in CH_4 concentrations in soil gas for different time periods at the four sites in the Yakela condensed gas field. It can be seen overall that CH_4 concentrations increased significantly with an increase of depth in soil gas profiles for each time period at all the four sites, similar to the results reported by Klusman et al (2000) in four semiarid sedimentary basins in the American state of Colorado. The phenomena might suggest that there was CH_4 microseepage from deep-buried reservoirs. At 30 cm to 60 cm depth, CH_4 concentrations at soil gas profiles dropped slowly with increasing depth and reduced to a value less than the atmospheric concentration in the afternoon at all the four sites. Similar phenomena were also observed at some other sites studied. The phenomenon above might indicate that the soil has sufficient permeability and porosity to allow CH_4 from the atmosphere to diffuse downward into the soil gas. A part of the CH_4 which had diffused downwards was oxidized by free oxygen or methanotrophic bacteria in soil. Then CH_4 concentrations of soil gas increased progressively to concentrations above atmospheric concentration with an

increase of depth at the 90 cm and deeper at all the four sites. These increasing gradients were higher than sampling and analytical error (0.11%–0.25%), especially at the faults. Thus it can be seen that the lowest point of CH_4 concentration at the depth of 30 cm or 60 cm was just the CH_4 equilibrium point of gas-phase formed by the gas migrating upwards from the deep-buried reservoirs and the gas diffusing downwards

from the atmosphere (Fig. 2).

In addition, Fig. 3 indicated that the CH_4 concentration at the same depth in the soil gas profile was obviously higher at the fault than that at the other three sites while CH_4 concentrations at the other three sites showed little difference. CH_4 concentrations at 90 cm depth were slightly higher than or close to those measured at 120 cm depth at the
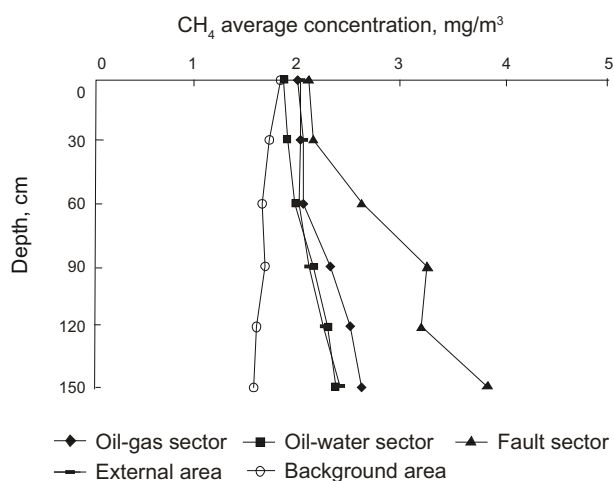

Fig. 2 Variation of CH_4 concentrations in soil gas in representative areas of the Yakela condensed gas field (Notes: CH_4 concentration of atmosphere at 0 cm)

fault, presumably due to CH_4 microseepage arising from the existence of a 10-20 cm fine sandy layer at the fault. Thus it can be concluded that light hydrocarbon gases, i.e. CH_4 from deep-buried reservoirs in the Yakela condensed gas field could relatively easily migrate upwards to the earth's surface along the fault.

Moreover, average CH_4 concentrations at the depth of 90-150 cm at the oil-gas sector were only slightly higher than those found at the oil-water sector and the external area. Although the external area is located outside the Yakela

condensed gas field, the existence of a subfault named Ya-III would be able to result in a small amount of CH_4 seepage to the surface from deep oil/gas traps. The phenomenon above is consistent with those observed in our previous study (Tang et al, 2010), in which CH_4 fluxes at the fault sector were found to be higher than those at the other three sites in the Yakela condensed gas field, suggesting that CH_4 from deep-buried reservoirs could migrate upwards to surface through permeable rocks, fissures and faults, of which part was oxidized or decomposed during the migrating process.

In contrast, CH$_4$ concentrations decreased gradually with an increase of depth in the soil-gas profile at the control study area, indicating the possible methanotrophic oxidation of atmospheric CH$_4$ diffusing downwards (Fig. 3).
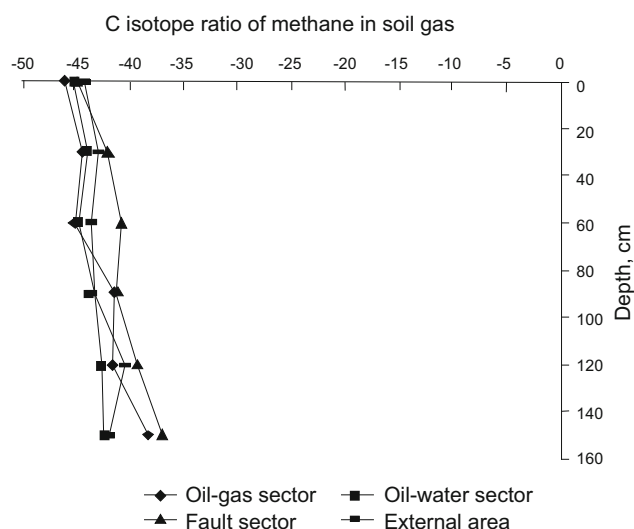

Fig. 3 Variation characteristics of CH$_4$ average concentrations in soil gas in representative areas of the Yakela condensed gas field (Notes: CH$_4$ concentration of atmosphere at 0 cm)

4.2 Variation of carbon isotope of CH$_4$ in soil gas

With the development of isotope measurement techniques, C and H isotope systematics (^{13}C/^{12}C, D/H) and radioactive carbon isotope (^{14}C) have been widely applied for tracing the sinks/sources of atmospheric CH$_4$, for estimating global or regional CH$_4$ budgets and source strength, and for interpreting environmental mechanisms of production, transport and release of CH$_4$ (Rust, 1981; Stevens and Engelkemeir, 1988; Wahlen et al, 1989; Hilkert et al, 1999; Rice et al, 2001). The origin of CH$_4$ can be divided into two major categories, namely "bacterial" and "thermogenic" CH$_4$. Thermogenic CH$_4$ is generally, but not exclusively enriched in ^{13}C compared with bacterial CH$_4$, and has a range of δ^{13}C extending roughly from -50‰ to -20‰. The term "bacterial CH$_4$" is preferred over "biogenic" because the carbon in both bacterial and thermogenic natural gases was originally from biological material. Bacterial CH$_4$ has a wide range of δ^{13}C, varying from -110‰ to -50‰. Petroleum geological studies in the Yakela condensed gas field have demonstrated that δ^{13}C$_{CH_4}$ in deep natural gases is typical of pyrogenic gases, ranging from -42‰ to -31‰ (Tang and Liu, 2002).

Variation characteristics of δ^{13}C$_{CH_4}$ in soil gas at the four sites are showed in Fig. 4. It can be seen that the δ^{13}C$_{CH_4}$ of the soil gases taken from air at the surface (0 cm) ranged from -44.2‰ to -45.8‰, which is close to the global atmospheric δ^{13}C$_{CH_4}$ level (-47.3‰ to -46.2‰) (Stevens, 1993; Tyler, 1999). It is obvious that δ^{13}C$_{CH_4}$ in soil gas became gradually heavier with increasing depth (Fig. 4). The increasing gradients were higher relative to sampling and analytical precision (the precision of δ^{13}C$_{CH_4}$ is 0.4‰). δ^{13}C$_{CH_4}$ at the depth from 30 cm to 60 cm in the Yakela condensed gas field, except at the fault became slightly heavier with

Fig. 4 Variation characteristics of δ^{13}C$_{CH_4}$ in soil gas in representative areas of the Yakela condensed gas field (Notes: δ^{13}C$_{CH_4}$ of atmosphere at 0 cm)

an increase in depth, which was accompanied by a decrease of CH$_4$ concentrations in soil gas to a value less than the atmospheric concentration (Fig. 3). The case above shows that CH$_4$ from atmosphere diffused downwards into soil gas, where the microbial processes of methanotrophic bacteria in soil preferentially consumes the lighter isotope while leaving the residual CH$_4$ isotopically heavier. Moreover, δ^{13}C$_{CH_4}$ at the depth from 90 cm to 150 cm became gradually heavier with the increase of depth and fell into the range characterized as thermogenic CH$_4$, in concert with our previous petroleum geological findings in the Yakela condensed gas field (Tang and Liu, 2002). Meanwhile, the processes above were accompanied by an increase of CH$_4$ concentrations in soil gas to a value more than the atmospheric concentration. All these suggested that there was a thermogenic source. Specifically, occurrence of C$_2$H$_6$ and C$_3$H$_8$ in soil gas further proved that there was hydrocarbon microseepage from deep-buried reservoirs in the Yakela condensed gas field (Whiticar, 1999), especially at the fault, while there were no C$_2$H$_6$ and C$_3$H$_8$ detected at the background area studied (Table 1).

4.3 Influencing factors on CH$_4$ concentrations in soil gas

CH$_4$ concentrations in soil gas have been reported to be greatly affected by temperature, moisture, particle size of soil and seasonal changes, etc. A linear correlation analysis (Table 2) indicated that CH$_4$ concentration of soil gas was negatively correlated with soil temperature and soil Eh at a significant level of α =0.05.

Soil Eh is a measure of the oxidation-reduction status in soil. Methanotrophic bacteria are obligate aerobic bacterium. Higher soil Eh is helpful for the activities of methanotrophic bacteria. Therefore, CH$_4$ migrating upwards from deep-buried reservoirs is easily oxidized when soil Eh is high, leading to lower CH$_4$ concentration in soil gas. Conversely, when activity of methanotrophic bacteria is subdued, this

Table 1 Hydrocarbon compositions in soil-gas profiles in representative sites of the Yakla condensed gas field

Site	Depth, cm	CH_4	C_2H_4	C_2H_6	C_3H_6	C_3H_8	nC_4H_{10}	iC_4H_{10}
Oil-gas sector	0	2.01	0.04	nd	nd	nd	nd	nd
	30	2.01	0.05	nd	nd	nd	nd	nd
	60	2.05	0.03	nd	nd	nd	nd	nd
	90	2.28	0.1	0.03	nd	nd	0.01	nd
	120	2.50	0.03	0.04	nd	0.02	nd	nd
	150	2.61	0.09	0.11	nd	0.07	0.03	nd
Oil-water sector	0	1.86	nd	nd	nd	nd	nd	nd
	30	1.90	nd	nd	nd	nd	nd	nd
	60	1.97	0.03	nd	nd	nd	nd	nd
	90	2.16	nd	nd	nd	nd	nd	nd
	120	2.23	0.03	0.06	nd	0.01	nd	nd
	150	2.36	0.07	0.09	nd	0.06	0.02	nd
Fault	0	2.11	0.07	nd	nd	nd	nd	nd
	30	2.14	0.05	nd	nd	nd	nd	nd
	60	2.62	0.11	0.03	nd	nd	nd	nd
	90	3.25	0.1	0.05	nd	0.03	0.06	nd
	120	3.16	0.08	0.11	0.03	0.09	0.08	nd
	150	3.91	0.17	0.19	0.03	0.11	0.07	nd
External area	0	2.01	0.03	nd	nd	nd	nd	nd
	30	1.97	nd	nd	nd	nd	nd	nd
	60	2.06	0.03	nd	nd	nd	nd	nd
	90	2.08	0.05	0.02	nd	nd	nd	nd
	120	2.24	nd	nd	nd	nd	nd	nd
	150	2.33	0.06	0.03	nd	0.01	nd	nd
Background	0	1.82	nd	nd	nd	nd	nd	nd
	30	1.71	nd	nd	nd	nd	nd	nd
	60	1.64	nd	nd	nd	nd	nd	nd
	90	1.67	0.02	nd	nd	nd	nd	nd
	120	1.59	0.03	nd	nd	nd	nd	nd
	150	1.56	nd	nd	nd	nd	nd	nd

Notes: Concentrations are as ppmv, 'nd' indicates 'not detected'

leads to higher CH_4 concentration in the soil gas. The Eh of soil in the Yakela condensed gas field ranged from 97.3 to 190.7, showing weakly reducing conditions, which favored the preservation of CH_4 migrating upwards from deep-buried reservoirs. Relatively speaking, Eh (97.3–123.7) at the fault sector was lower than that at the other three sectors (Eh: 126.5–160.3 at the oil-gas sector, 123–180 at the oil-water sector, 135.9–190.7 at the external area, respectively), resulting in a higher CH_4 concentration at the fault than at the other three sectors, which also verified that the fault was a good conduit for light hydrocarbons to migrate upwards from deep-buried reservoirs.

Similarly, oxidizing activities of methanotrophic bacteria in soil are also affected by soil temperature. Higher temperature is advantageous for methanotrophic oxidation in soil. Thereby a part of CH_4, migrating from the oil/gas reservoirs to the surface, was absorbed and oxidized by soil.

In addition, Table 2 shows that soil pH was significantly and negatively correlated to soil Eh ($\alpha = 0.05$), suggesting that soil pH could indirectly influence soil methanotrophic oxidation by affecting soil Eh. However, no obvious correlation was found between soil CH_4 concentration and other soil factors such as moisture, salinity and porosity.

5 Conclusions

Variation in CH_4 in soil-gas profiles showed that CH_4 microseepage was a quite common phenomenon in the Yakela condensed gas field. Hydrocarbon gases, methane in particular, were liable to migrate upwards by microseepage from the deep-buried reservoirs to the earth's surface through faults, fissures, micro-fracture networks and rock pore networks, during which some unoxidized CH_4 was preserved in soil gas or emitted into the atmosphere. Results showed that CH_4 microseepage was relatively easily emitted along the fault, and the $\delta^{13}C_{CH_4}$, ethane and propane in soil gas exhibited thermogenic characteristics. Moreover, higher soil temperature and Eh could enhance the activities of methanotrophic bacteria, while soil pH, via impacting soil Eh, could indirectly influence soil methanotrophic oxidation, thereby resulting in a reduction in soil gas CH_4 concentration.

Table 2 Correlation between CH_4 concentration in soil gas and environmental factors of soil in the Yakela condensed gas field ($\alpha = 0.05$)

	CH_4 concentration	Temperature	Moisture	pH	Eh	Ec	Porosity
CH_4 concentration	1.000						
Temperature	-0.215*	1.000					
Moisture	-0.092	0.027	1.000				
pH	0.207	-0.199	-0.604*	1.000			
Eh	-0.549*	0.239*	0.053	-0.443*	1.000		
Ec	-0.168	0.045	0.782*	-0.512*	0.100	1.000	
Porosity	-0.062	-0.050	-0.334*	0.048	-0.131	-0.152	1.000
Number of samples	N=88						

*Significant correlation at the level of $\alpha = 0.05$

Acknowledgements

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