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Research paper

## Geochemical application of tricyclic and tetracyclic terpanes biomarkers in crude oils of NW China





Shizhen Tao <sup>a</sup>, Chuanyuan Wang <sup>b, \*</sup>, Jianguo Du <sup>c</sup>, Lei Liu <sup>c</sup>, Zhi Chen <sup>c</sup>

<sup>a</sup> Research Institute of Exploration and Development, China National Petroleum Corporation, Beijing 100083, China

<sup>b</sup> Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences, Yantai 264003, China

<sup>c</sup> Institute of Earthguake Science, China Seismological Bureau, Beijing 100036, China

#### A R T I C L E I N F O

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#### ABSTRACT

Composition and distribution of tricyclic terpanes, tetracyclic terpanes and hopane in crude oils from Tarim, Turpan, Ordos and Qaidam basins, NW China, were investigated, and some new source dependent parameters were developed and evaluated.  $C_{19}$  and  $C_{20}$  tricyclic terpanes are more abundant in terrigenous oils, with  $C_{24}$  tetracyclic terpane dominant. Relatively higher carbon number tricyclic terpanes, especially  $C_{23}$  tricyclic terpane, are often the dominant homologues in crude oils from saline lacustrine and marine sources. On the whole, the relative content of  $C_{23}$  tricyclic terpane of crude oils follow the order of saline lacustrine oils > marine oils > freshwater lacustrine oils > terrigenous oils for  $C_{19-23}$  tricyclic terpane. Some new parameters based on the tricyclic/hopane and tetracyclic/tricyclic terpane as well as tetracyclic/hopane ratios, could be used to distinguish the oils and source rocks with similar maturity deposited in different sedimentary environments.

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#### 1. Introduction

Tricyclic terpanes are important biomarkers and occur widely in source rock extracts and crude oils (de Grande et al., 1993; Fazeela et al., 2011). Tricyclic terpanes (TT) and tetracyclic terpanes (TeT) from sedimentary organic matter can provide valuable information to assess organic input, maturity, correlation of crude oils (Didyk et al., 1978; Azevedo et al., 1992; Farrimond et al., 1999; Tuo et al., 1999; Nady, 2008; Samuel et al., 2010; Wang et al., 2012; Zhang and Philp, 2012; Jin et al., 2014). The tricyclic terpane series are commonly observed in petroleum samples up to C<sub>29</sub>, but the higher members of the series are often masked by hopanes in the m/z 191 mass chromatogram. In petroleum source rocks, an increase in the ratio of tricyclic terpanes relative to the hopanes as far as the late oil window has been observed and this is thought to reflect earlier thermal degradation of hopanes relative to the tricyclic terpanes (Farrimond et al., 1999). Peters and Moldowan (1993) suggested that C<sub>19</sub> or C<sub>20</sub> tricyclic terpanes are more abundant in terrigenous oils; while C<sub>23</sub> tricyclic terpane is often the dominant homologue in crude oils with a marine source. Moreover, several tricyclic terpane maturity parameters are used to effectively assess the thermal maturities of carbonate source rocks, especially at the immature to early mature stages (Tuo et al., 1999). Although the origin of  $C_{24}$  TeT is still unknown, it is thought to originate from thermal or microbial degradation of hopanes (Peters et al., 2005). Farrimond et al. (1999) observed an increase in the  $C_{23}$  tricyclic terpanes maturity parameters  $C_{23}/C_{21}$ TT,  $C_{23}/C_{24}$ TT and  $C_{23}/C_{26}$ TT. Based on this, it can be concluded that the maturity of crude oil or source rocks might be difficult to assess in an oil-gas geochemistry study without careful examination of the type of organic constituents (e.g. marine/ freshwater algae or terrigenous land plant).

Although tricyclic and tetracyclic terpanes have been increasingly applied in petroleum exploration, limited studies were reported about the effect of depositional environments on the distribution of tricyclic and tetracyclic terpanes. The Tarim, Qaidam, Turpan and Ordos basins in NW China are important petroleum producing basins in China, which represent different depositional environments, such as marine, freshwater lacustrine, saline lacustrine and terrigenous environments. In this paper, some crude oil samples from Tarim, Qaidam, Turpan and Ordos basins were analyzed by gas chromatography-mass spectrometry (GC–MS) for the tricyclic and tetracyclic terpanes in order to help fill the knowledge gaps. To achieve the goal, the following tasks were carried out: (1) analyses of distribution of tricyclic and tetracyclic terpanes in crude oils from Tarim, Qaidam, Turpan and Ordos basins; (2) investigation of depositional environment effects on the

<sup>\*</sup> Corresponding author. E-mail address: cywang@yic.ac.cn (C. Wang).

distribution of tricyclic and tetracyclic terpanes; and (3) development of a set of source dependent parameters for oil correlation and differentiation.

#### 2. Geological background

The Tarim, Qaidam, Turpan and Ordos basins in NW China are important petroleum producing basins in China. The Qaidam Basin, located in Qinghai Province, NW China, covers an area of 121,000 km<sup>2</sup> and is a Mesozoic-Cenozoic sedimentary basin characterized by an inland saline lake facies. The Qaidam Basin has distinct petroleum systems, namely a Lower-Middle Jurassic freshwater lacustrine petroleum system in the northern margin, a Tertiary saline lacustrine oil system in the western part and Quaternary biogenic gas system in the eastern area (Zhang and Philp, 2012). The south area of western Qaidam Basin is the largest Tertiary saline lacustrine petroleum-producing area in China. The Turpan Basin, located in the Xinjiang Uygur Autonomous Region, NW China, covers an area of 53,500 km<sup>2</sup> and is one of the three large sedimentary basins in Xinjiang Uygur Autonomous Region, NW China. The basin can be described as two depressions and one uplift in terms of its structural framework: the Turpan depression in the west, the Hami depression in the east and the Liaodun uplift in the middle. The Tarim Basin, located in the southern Xinjiang Uygur Autonomous Region, NW China, is one of the world's largest frontier basins, with an area of 560,000 km<sup>2</sup>. Tahe oilfield in Tarim Basin is the largest oilfield found in Paleozoic marine carbonate rocks in China, crude oils have been mainly recovered from the Ordovician carbonate. The Ordos Basin, a large intracontinental sedimentary basin in China with an area of about 37,104 km<sup>2</sup>, contains abundant Mesozoic crude oil resources with a reserve estimate of up to  $10 \times 10^8$  metric tons of oil from Mesozoic reservoirs (Duan et al., 2008). The crude oils have been considered to be derived from non-marine source rocks. Details on the geology are available in Zhu et al. (2005), Duan et al. (2008), Yu et al. (2011), Zhang and Huang (2005), Zhang and Philp (2012) and Mo and Zhang (2012).

#### 3. Samples and experimental

#### 3.1. Samples

The crude oils from Tarim, Qaidam, Turpan and Ordos basins in NW China (Fig. 1) were separated into four kinds: marine oils,



freshwater lacustrine oils, saline lacustrine and terrigenous oils. A

#### 3.2. Experimental

field in Turpan Basin.

The oils were deasphaltened using hexane before fractionation and were separated into saturate hydrocarbons (Sat.), aromatic hydrocarbons (Aro.) and non-hydrocarbons (Non.) by column chromatography using 10 g of neutral alumina and 20 g of silica gel. Sat., Aro. and Non. were obtained by successively eluting with *n*hexane, toluene and chloroform/methanol (98:2), respectively.

The saturated hydrocarbons were analyzed with a 6890N GC-5973N mass spectrometer. Sample extracts were injected in a splitless mode onto a HP-5 capillary column (50 m  $\times$  0.32 mm  $\times$  0.25 µm, Agilent Technologies, USA) at an initial temperature of 80 °C. The GC oven temperature was programmed to 300 °C at 4 °C/min and was held at the final temperature for 30 min. Helium was used as a carrier gas. Mass spectrometer conditions were electron ionization at 70 eV with an ion source temperature at 250 °C.

The GC–MS system was operated in full scan mode. Individual *n*-alkanes were identified based on the retention time of the authentic standards (n-C<sub>10-40</sub>, Sigma). The terpanes and steranes were detected in their key mass chromatograms (m/z 191, 217) based on the relative retention times and by comparing their mass spectra with published data. The standard deviation of the calculations of the peak areas was better than 0.5%.

#### 4. Results and discussion

## 4.1. Source of organic matter and depositional environments of crude oils

Pristane/phytane (Pr/Ph) ratios have been used to assess the redox potential of the depositional environment. It is generally accepted that very low values of the pristane/phytane ratio ( $\leq$ 1) indicate anoxic conditions (Didky Zhang and Huang, 2005) while high values (Pr/Ph > 3) are related to terrigenous organic matter input under less restricted conditions (Peters et al., 2005; Duan et al., 2008). Gammacerane was first described in a study of the Green River shale in Colorado (Hills et al., 1966). The gammacerane precursor is tetrahymanol (XV) found in hypersaline palae-oenvironments in Green River shale (Henderson and Steel, 1971). More recently, gammacerane is commonly considered to be of tetrahymanol origin which occurs in saline waters (Fu and Sheng, 1989; Sinninghe Damsté et al., 1995; Yu et al., 2011).

The distribution of Pr/Ph ratios and gammacerane index ( $G/C_{30}$   $\alpha\beta$  hopane) from crude oils in the Tarim, Qaidam, Ordos and Turpan basins show distinct differences (Fig. 2). The Pr/Ph ratios of crude oils from Tarim Basin and Qaidam Basin are <1 indicating anoxic conditions whereas higher values (1–3) of the crude oils from



Fig. 1. Location map indicating the studied basins in China.

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