Applied Geochemistry 65 (2016) 14–21

Contents lists available at ScienceDirect

Applied Geochemistry

journal homepage: www.elsevier.com/locate/apgeochem

Iodine-129 chronological study of brines from an Ordovician paleokarst reservoir in the Lunnan oilfield, Tarim Basin

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ARTICLE INFO

Article history:
Received 10 April 2015
Received in revised form
17 October 2015
Accepted 31 October 2015
Available online 4 November 2015

Keywords:
Iodine
Iodine 129
Oilfield water
Tarim basin

ABSTRACT

Previous studies have shown that brines in an Ordovician paleokarst reservoir of the Lunnan oilfield in the Tarim Basin, China, are the product of mixing of paleo-evaporated seawater in the east with paleometeoric waters in the west. In order to put time constraints on the brine and related hydrocarbons in this field, 10 brine samples were collected, for which the iodine concentrations and $^{129}I/I$ ratios were measured and discussed. The iodine concentration (3.70–31.2 mg/L) and the $^{129}I/I$ ratio (189–897 $\times 10^{-15}$) show that the iodine in the paleoseawater and meteoric water (MW) had different origins and ^{129}I characteristics. The paleoseawater has a high iodine content (~31 mg/L), indicating that iodine was introduced into the reservoir along with thermally generated hydrocarbons, possibly in the Cretaceous, from the Caohu Sag in the eastern area. Based on consideration of all possible origins of iodine and ^{129}I in the brines, it is suggested that the meteoric water maintained its initial iodine content (0.01 mg/L) and $^{129}I/I$ ratio (1500×10^{-15}), whereas the iodine-enriched paloseawater (IPSW) exhibited a secular ^{129}I equilibrium ($N_{sq} = 39$ atom/μL) as a result of fissiogenic ^{129}I input in the reservoir over a long period of time. The model of brine evolution developed on that basis confirmed that meteoric water entered the reservoir in the Miocene at about 10 Ma, and partially mixed with the iodine-enriched paleoseawater. The movement of meteoric water was facilitated by faults created during the Himalayan orogeny, then became more dense after dissolving Paleogene halite and infiltrated into the reservoir at high pressure. The iodine and ^{129}I concentration in the brine contains information about the path and history of the fluid in the reservoir. This may be useful in oil exploration, since the movement of water was, to some extent, related to hydrocarbon migration.

1. Introduction

Iodine is a liquid-dominated species since it is water soluble and its large ionic radius (133 pm) precludes it from being readily incorporated into minerals (Osborn et al., 2012). It is also strongly biophilic, and often accumulates in marine organic matter (10–490 ppm: Muramatsu and Wedepohl, 1998; 4.8–320 ppm: Muramatsu et al., 2004) by more than 100 times than that in sea water (0.05 ppm). As crude oils have a low iodine content (1 ppm: Fehn et al., 1987; 0–50 ppb: Moran et al., 1995b; 0.1–10 ppm: Worden, 1996), it is assumed that iodine is released into adjacent groundwater during maturation of organic matter in the source rock (Fehn, 2012); thus iodine enrichment in brines serves as a proxy for hydrocarbon migration.

Iodine-129 (^{129}I, $t_{1/2} = 15.7$ Myr) is the only long-lived radioisotope of iodine. Generally, there are three main sources of ^{129}I: (i) cosmogenic ^{129}I produced by the spallation of Xe isotopes into the atmosphere; (ii) fissiogenic ^{129}I produced by spontaneous fission of ^{238}U in the Earth's crust; and (iii) anthropogenic ^{129}I produced by nuclear weapons testing and fuel processing since the 1950s (Fehn,

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http://dx.doi.org/10.1016/j.apgeochem.2015.10.012
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2012). In terms of these three sources, ^{129}I analysis has been successfully used for tracing and/or dating the fluids in a variety of geological settings, such as surface water and groundwater (Schwehr et al., 2005), hydrothermal fluids (Fehn et al., 1992), basinal brine (Osborn et al., 2012) and deep crustal fluids (Fehn and Snyder, 2005). Analysis of ^{129}I has also been used to trace waters associated with hydrocarbons in order to fix the age boundaries of organic accumulations (e.g., brines in oilfields: Birkle, 2006; gas hydrate: Fehn et al., 2000, 2003; forearc methane fields: Muramatsu et al., 2001; coalbed methane: Snyder and Fabryka-Martin, 2007; Snyder et al., 2003). While in these cases, the identification of the potential source formations was the main goal, iodine dating might help to determine mixing patterns and timing of brines in the Lunnan oilfield.

The target of the present study was the Ordovician paleokarst reservoir in the Lunnan oilfield located in the northern Tarim Basin. It is one of the major petroleum producers in that area (Fig. 1). The reservoir is deeper than 5000 m below sea level (mbsl) and is well known for its range of hydrocarbons, its weak intrareservoir connectivity and the tilted oil–water contact. These characteristics may be attributed to the multiple episodes of oil migration, evolution of the karst system, blending of different fluids and complex tectonic activity (Lu et al., 2004; Pang et al., 2007; Yang and Han, 2008; Zhang et al., 2011b).

Previous work on water chemistry and isotopes reached the following conclusions: (i) the brines in the reservoir are mixtures of infiltrated meteoric water at the top of the Lunnan Uplift with rising paleoseawater from the underlying marine strata in the Caohu Sag in the eastern area; (ii) the eastern fluid regimes are chemically separate from the western fluid regimes due to the larger contribution of the meteoric waters (Fig. 1); and (iii) two tentative hypothetical models of brine evolution were proposed (Chen et al., 2013).

Nevertheless, the geological periods in which these waters invaded remain unresolved. It is unclear whether the meteoric water is related to the period of karst development, or was intruded in a late basin uplift period. Furthermore, since the paleoseawater was derived from Cambrian and Lower Ordovician strata in the Caohu Sag, which itself is overlain by Middle–Upper Ordovician source rock, the important question remains as to whether paleoseawater migrated with the generated petroleum. The aims of our study are putting the time constraints on the brine and related hydrocarbons using ^{129}I isotopes as well as iodine concentrations.

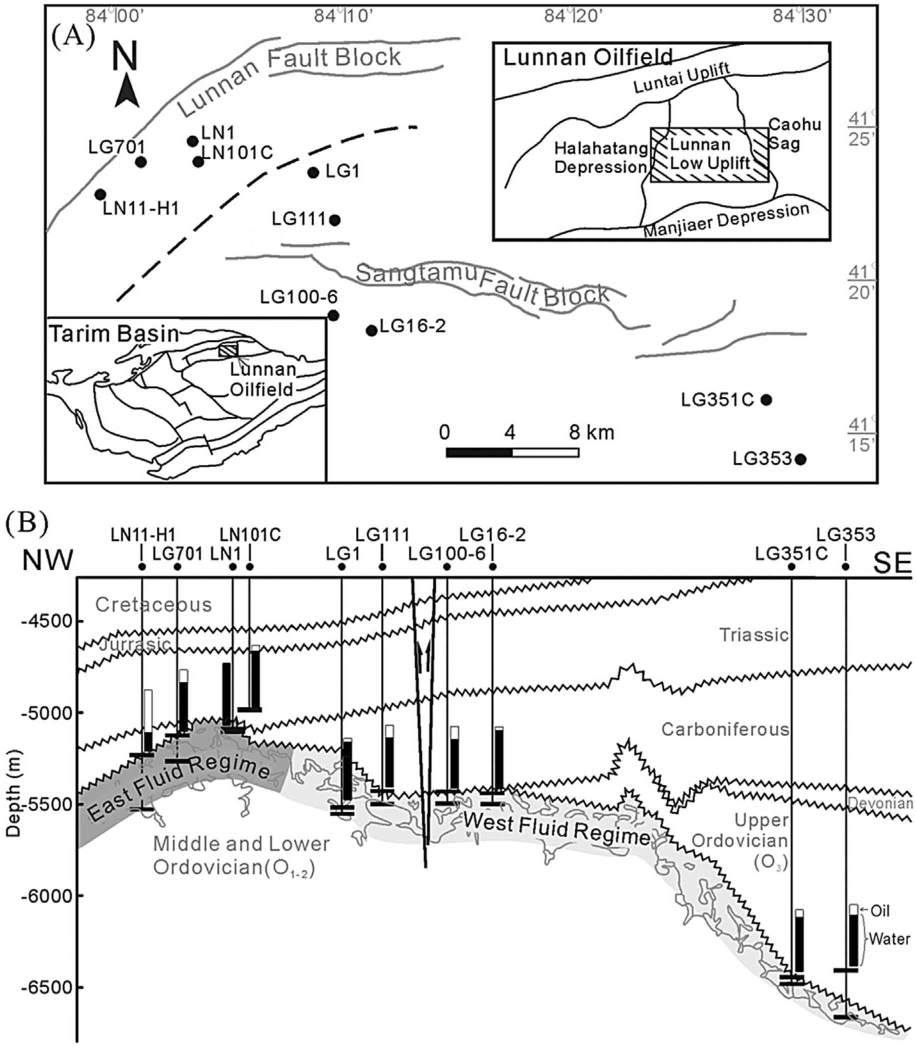

Fig. 1. (A) Study area and location of sampling wells in Lunnan oilfield, Tarim Basin, China. Insets in Fig. 1A are the structural maps of Tarim Basin and Lunnan oilfield. (B) Strata profile from Well LN11-H1 to Well LG353. Note: the oil/water volume ratios are shown in the short columns. Two fluid regimes are divided by dashed line in Fig. 1A and by different colors in Fig. 1B. Reprinted and modified from Chen et al. (2013) by permission from John Wiley and Sons.