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Gas geochemistry and methane emission from Dushanzi mud volcanoes in the southern Junggar Basin, NW China

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In memory of Tsanyao Frank Yang from Department of Geosciences, National Taiwan University, Taipei 10699, Taiwan.

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ABSTRACT

There are many mud volcanoes in the southern margin of the Junggar Basin, northwest China, of which the Dushanzi area is the most typical and active one, emitting large amount of greenhouse gases associated with water and mud. The emitted gas is dominated by methane (average 90.1%), together with other gases, such as ethane (4.84–5.46%), propane (0.06–0.90%), CO_2 (0.67–1.0%), and N_2 (2.8–3.3%). The carbon ($\delta^{13}C_1$) and hydrogen (δD) isotopic ratios of methane are in the ranges of −40.6‰ to −45.0‰ and −221‰ to −249‰, respectively, whereas carbon isotope ratios of ethane ($\delta^{13}C_2$) are −25.2‰ to −27.6‰. Based on $\delta^{13}C$ values, the released gas is characterized as a thermogenic coal-type and possibly originated from the middle-low Jurassic coal-bearing sequences according to the gas-source correlation and regional geology. Helium isotopes show a crustal source. The methane flux of Dushanzi mud volcanoes from both macro-seepage (craters/vents) and micro-seepage (ground soil exhalation) ranged over the orders of magnitude, from 0.4–2.7 kg d^{-1} and 4950 mg m^{-2} d^{-1} on average, respectively. Positive CH_4 fluxes from dry soil were widespread throughout the investigated areas. The total CH_4 emission from Dushanzi mud volcanoes is estimated to be at least 22.6 tons a^{-1}, of which about 89% is from micro-seepage surrounding the mud volcano vents.

1. Introduction

Being one of the important geological sources for greenhouse gases, mud volcanoes have become attractive again for geochemical studies, especially regarding their emission of atmospheric methane, which is one of the most powerful greenhouse gases (Etiope and Klusman, 2002; Dimitrov, 2003; Milkov et al., 2003; Etiope and Milkov, 2004). However, quantifying the total emission of greenhouse gases from mud volcano systems remains a huge challenge because of the limited number of direct flux measurements performed so far. The first flux survey of mud volcano gas emission was performed using a closed-chamber method (Klusman et al., 2000), which showed that methane emission from mud volcanoes could be attributed not only to macro-seepage, including visible gas manifestations like bubbling pools, salses and gryphons, and eruptive events, but also to micro-seepage from invisible and diffuse emissions, as described, for example, by Abrams (2005); Etiope and Klusman (2002); and Etiope et al. (2011). To date, greenhouse gases emissions from mud volcanoes to the atmosphere have been measured in Romania (Etiope et al., 2004; Spulber et al., 2010), Azerbaijan (Etiope et al., 2004; Kopf et al., 2009), Italy (Etiope et al., 2007), Taiwan, plus some other counties and regions (Yang et al., 2004; Chang et al., 2010; Chao et al., 2010; Hong et al., 2013). More recently, the ground degassing rate was measured in Japan (Etiope et al., 2011). In the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC), geological methane has been listed separately as an important source of greenhouse gases: however, there is still lack of available data from mud volcanoes and other kinds of geological sources on China (Etiope, 2015; Guo et al., 2017).

There are several mud volcanoes distributed in the southern margin of the Junggar Basin, NW China, which were considered as indicators of some petroleum reservoirs. For example, the Dushanzi oilfield was discovered on the Dushanzi mud volcano area more than 100 years ago and was then developed as one of the earliest commercial petroleum fields in China (ECPGX, 1993). However, hydrocarbon seepage fluxes

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from these mud volcanoes have only been reported sporadically (e.g., Dai et al., 2012; Ma et al., 2014), and gas emission rates were estimated only based on bubbling manifestations from the active craters. In 2011, a preliminary investigation was performed on micro-seepage using a closed-chamber method within distance of 10–15 m from the two main bubbling vents (Ma et al., 2014). Considering the fact that large areas of the central Dushanzi anticline have been bleached by petroleum (Zheng et al., 2010), it is necessary to extend the measurement area to obtain more accurate flux data of gas emission from both macro- and micro-seepages. In order to satisfy the ever-increasing requirements from sustainable development of the domestic economy and societal needs in China, and also the international obligations concerning atmospheric carbon budgets (Edenhofer, 2015), more greenhouse gases flux measurements of mud volcano systems are essential, and these should include active craters and their surrounding ground emission areas. Thus the main topic of this study is to provide basic information about detailed gas flux investigations *in-situ* by using a closed-chamber method for the Dushanzi mud volcanoes in the southern margin of the Junggar Basin. The gas geochemistry and origin is also discussed based on various geochemical parameters and with respect to the geological background.

2. Geological settings and mud volcanoes

2.1. Geological settings

The study area, located in the southwestern margin of the Junggar Basin (Fig. 1), is famous for being the location of the first oil wells drilled in the basin and, indeed, for being the earliest oilfields discovered and developed in China. The Junggar Basin is a Paleozoic and Mesozoic-Cenozoic superimposed basin controlled by the Tianshan Mountains to the south and Altai Mountains to the north. The southern part of the basin is underlain by 16 km of thick sedimentary rocks deposited since the Permian period; the low-middle Jurassic systems were mainly composed of thick-layer coal and mudstone, being regarded as the major source rocks of hydrocarbons in this region (Qiu et al., 2008). The sedimentary rocks are folded and faulted by the northward thrusting of the Tianshan Mountains, which have induced several rows of anticlines in front of the high mountains. The Dushanzi mud volcanoes are developed on the Dushanzi hill, being one of the folds and named as Dushanzi Anticline or Fold (Avouac et al., 1993; Fu et al., 2007).

The southern margin of the Junggar Basin is tectonically a typical foreland depression with well-developed thrust tectonics and fold zones as found at other foreland depressions in central and western parts of China (Jia et al., 2003). A number of mud volcanoes occurred on the folded zones, mainly including four active sites: the Anjihai, Dushanzi, Aiqigou, and Baiyanggou, from east to west (Fig. 1). The Dushanzi area has a typical arid inland climate with high evaporation rate (annual = 3009 mm), abundant sunshine, large diurnal fluctuations, and sharp seasonal changes with hot summers (as high as 45 °C) and cold winters (as low as −35 °C). Most of the region has sparse plant cover excluding some oasis-like areas, which are normally located at the outlets of rivers or streams from the Tianshan Mountains (Zheng et al., 2010). On the mountains, there are many glaciers and heavy snow, supplying massive snow melt water to the dry desert, including formation water for the petroleum reservoirs under the Dushanzi Fold.

The Dushanzi mud volcanoes are developed on the axis of the Dushanzi anticline (Zhang et al., 1999), which is a closed fold structure of oil and gas storage with a normal fault system in the anticline axis with many fractures due to the local extension. The structures of the Paleogene system are the main oil and gas reservoirs, with the sequence of Neogene mud and sand deposits being the confining bed, which represents the effective trap. The abnormally high pressure of strata and tension fractures of the arch core are the principal factors behind the geological conditions for the formation of the Dushanzi mud volcanoes and their activity (Wang et al., 1997).

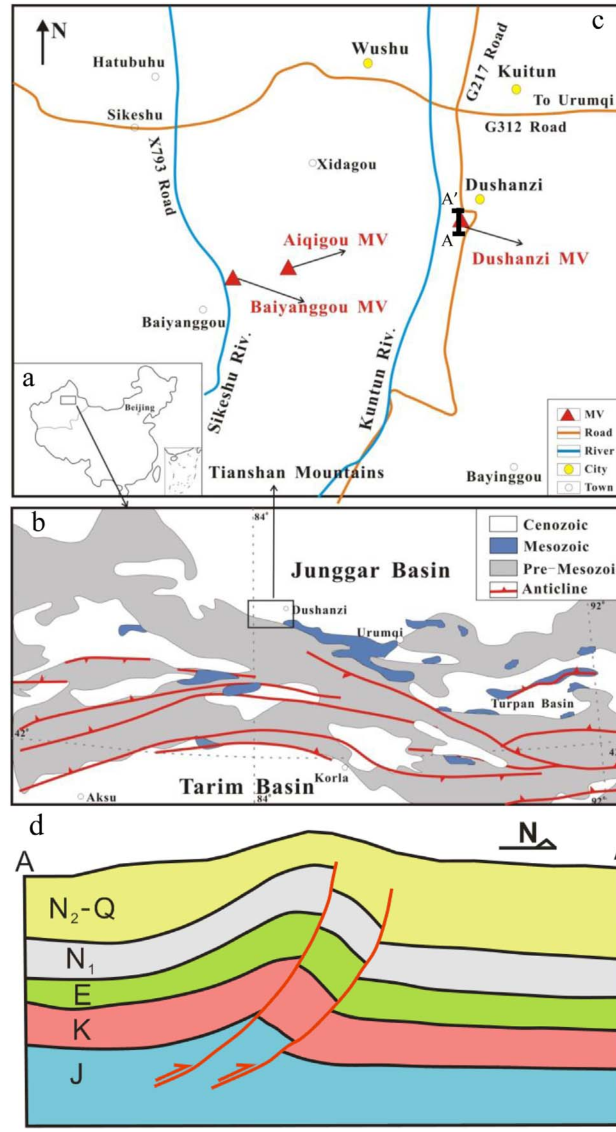

Fig. 1. Maps showing study area, geological settings and mud volcanoes locations (simplified after Wan et al., 2013; Dai et al., 2012). a – location of the study area. b – geological surroundings of the mud volcano area studied, which is located in the transition region between the Tianshan Mountains to the south and the Junggar Basin to the north. c – map of the study area, showing the mud volcanoes of Dushanzi, Aiqigou and Baiyanggou, main rivers and roads are also indicated. d – stratigraphic section map of Dushanzi area.

2.2. Dushanzi mud volcanoes

The Dushanzi mud volcanoes, located at 84°50′E and 44°18′N, are typical petroleum-related site. There are three major vents with eruption activities. (1) DSZ-01 (84°50.355′E and 44°18.234′N) is a mud pool with a diameter of about 1.5 m, in which there are always continuous bubbles with water, mud, gases and even oil (Fig. 2). (2) DSZ-02 (84°50.843′E and 44°18.315′N) is a huge crater lake or pond formed during mud volcanic eruption in 1995. The lake is mostly like a pit nowadays and is about 120 m long and 35 m wide. The crater pit can be divided into two sub-basins. The northern basin is always covered with surface water and some small bubbles are active with the emission of oil, gas, water, and little mud from the pit bottom. The lake water level decreased dramatically as recorded during survey in 2014 compared with former visits in 2011 and 2009. Around the lake, there are also