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Occurrence and profiles of methylsiloxanes and their hydrolysis product in aqueous matrices from the Daqing oilfield in China

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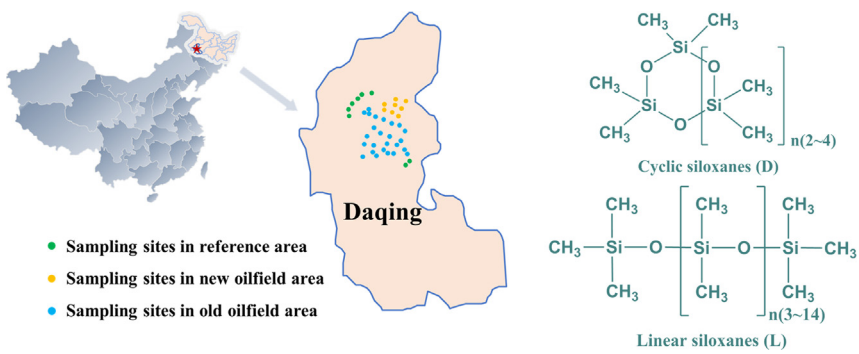
HIGHLIGHTS

- Wastewater samples from oilfield combination station have been analyzed.
- Methylsiloxanes were detected in the aqueous environment matrix of Daqing oilfield.
- Dimethylsilanediol was quantitated in surface water of Daqing oilfield.

GRAPHICAL ABSTRACT



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ABSTRACT

In the present study, the occurrence and distribution of methylsiloxanes (D4-D6, L5-L16) in the surface water and sediment collected from the Daqing oilfield were explored. For wastewater samples from an oilfield combination station, the concentrations of total cyclic methylsiloxanes (∑CMS, D4-D6) and total linear methylsiloxanes (∑LMS, L5-L16) ranged from 137–1911 ng/L to 49.4–190 ng/L, respectively. Cyclic methylsiloxanes were the predominant methylsiloxanes in the analyzed wastewater samples from the oilfield combination station. In addition, methylsiloxanes were detected in the surface water from three sampling areas, and the total concentrations varied from <LOQ-785 ng/L (mean = 219 ± 277 ng/L, median = 120 ng/L) for the new oilfield area, 15.1–1808 ng/L (mean = 270 ± 409 ng/L, median = 105 ng/L) for the old oilfield area and 18.0–252 ng/L (mean = 103 ± 90.4 ng/L, median = 52.3 ng/L) for the reference area. In the sediment, the ∑CMS and ∑LMS values in the samples collected from the old oilfield area were almost four to nine times higher than those in the samples from the new oilfield area. In addition, elevated concentrations of dimethylsilanediol were detected in the three sampling areas, especially in the old oilfield area (mean = 85.6 ± 98.3 ng/L), which confirmed that hydrolysis occurred.

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1. Introduction

Crude oil is crucial for the economic development of the world, and its demand is expected to continually grow in the future. However, after several decades of development, many oilfields have already entered

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the middle or past development period, and the difficulties of crude oil production have continuously increased. Particularly, most of the oilfields in China are located in continental basins and are characterized by serious heterogeneity and high oil viscosity. In addition, water breakthrough occurs in these fields at an early stage, leading to lower recovery, even when water injection is increased (Li et al., 2000). Therefore, enhanced oil recovery (EOR) techniques are widely used in the Chinese oil industry. Among all the EOR techniques, chemical EOR methods are one of the most promising (Buckley and Liu, 1998; Rosen et al., 2005; Hirasaki et al., 2011) and have been applied to many oilfield chemical auxiliaries. These chemical auxiliaries primarily include polymers and surfactants that are used for enhancing oil recovery by reducing the interfacial tension between the crude oil and the water present within the reservoir (Li et al., 2000).

The Daqing oilfield, one of the largest oilfields in China, was discovered in 1956 and is located in Daqing City, Heilongjiang Province. The annual oil production volume of the Daqing oilfield reaches approximately 40 million tons, with >10 million tons of oil obtained from EOR techniques (Wang et al., 2005). In EOR, organosilicon compounds are widely used in oilfield development because of their superior properties, including low surface tension, high thermal stability, and lubricating properties (Owen, 2005; Kochetkov et al., 2001; Tragoonwichian et al., 2007; Horii and Kannan, 2008; Lu et al., 2011; Zhang et al., 2011). For example, polydimethylsiloxane (PDMS) can be utilized as a de-emulsifier, defoamer, and drag reducer in crude oil production (Shaban, 1995; Daniel-David et al., 2005).

Generally, PDMS results from the polymerization of low-molecular-weight methylsiloxanes. Methylsiloxanes are composed of a backbone of alternating silicon oxygen (Si-O) units with a methyl group attached to each silicon atom, including cyclic methylsiloxanes (D4-D6) and linear methylsiloxanes (L5-L16). Normally, the synthesis of PDMS involves equilibrium reactions, causing the presence of low-molecular-weight methylsiloxanes (D3, D4) and their rearrangement products (D5 and D6) as impurities in the silicone products (Xu et al., 2014).

In recent years, as a high production volume chemical categorized by the US Environmental Protection Agency and the Organization for Economic Cooperation and Development (Dudzina et al., 2014), methylsiloxanes have been ubiquitously found in various environmental media (Horii et al., 2017; Homem et al., 2017; Wang et al., 2017; Tran et al., 2015; Tran and Kannan, 2015; Wang et al., 2015; Pieri et al., 2013; Gouin et al., 2013; Yucuis et al., 2013; Buser et al., 2013; Wang et al., 2013a; Zhang et al., 2011; Lu et al., 2010; McLachlan et al., 2010; Warner et al., 2010; Sparham et al., 2008), industrialized regions (Xu et al., 2012; Shi et al., 2015) and in some remote areas (Sanchís et al., 2015). Additionally, evidence has suggested that these chemicals can lead to adverse effects, including endocrine disruption, connective tissue disorders, adverse immunological responses, and liver and lung damage, in organisms (Capela et al., 2017; Rücker and Kümmerer, 2015; Wang et al., 2013b). Considering that the Daqing oilfield has already entered the high water-cut stage, the demand for organosilicon compounds in oil production has continuously increased. Methylsiloxanes are likely to be released into environmental matrices in oilfields during the application of silicone de-emulsifiers, defoamers and drag reducers in oil production.

To the best of our knowledge, only a few studies have focused on the occurrence and distribution of methylsiloxanes in the ambient environment of oilfields. One study reported the distribution of cyclic methylsiloxanes in soil collected from the Shengli oilfield (Shi et al., 2015). However, the sources of methylsiloxanes in soil are different from those in an aqueous environment. For example, the leakage of oil sludge containing methylsiloxanes during transfer could release these chemicals into the soil environment. Unlike the soil environment, in an aqueous environment, the discharge of wastewater is an important source of methylsiloxanes. In oilfield combination stations, a large volume of wastewater generated from the oil/water separation process may enter the aqueous environment. Therefore, it is important to investigate the occurrence and profile of methylsiloxanes in the aqueous environment of the Daqing oilfield. To this end, the levels of 15 methylsiloxanes (D4-D6, L5-L16) in the wastewater from the oilfield combination station and in the surface water and sediment collected from the Daqing oilfield were studied. Additionally, the distribution of dimethylsilanediol was discussed.

2. Materials and methods

2.1. Chemicals

Octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), dodecamethylcyclohexasiloxane (D6), linear siloxanes [polydimethylsiloxane mixture (PDMS)] and tetrakis(trimethylsilyloxy)silane (M4Q) were obtained from Sigma-Aldrich (St. Louis, MO, USA). The compositions of the linear siloxanes (L5 to L16) in the PDMS mixture have been reported previously and are described in the supporting materials (Table S1). ^{13}C-labeled D4, D5, and D6 were obtained from Cambridge Isotope Laboratories (Andover, MA, USA). Dimethylsilanediol was purchased from J&K Chemical Co. Ltd. (Beijing, China). Ethyl acetate and n-hexane were purchased from Fisher Scientific (Fair Lawn, New Jersey, USA).

2.2. Sampling

The present study was carried out in the crude oil production areas in Daqing, Heilongjiang Province, China. There are numerous lakes in Daqing, which is located in a sub-temperate zone with a sub-humid continental monsoon climate. Paired surface water and sediment samples were collected from 41 lakes in Daqing (November 2015).

The sample sites are shown in Fig. 1. Eight samples were collected from a new oilfield area, which is a sparse and newly developed oilfield area with only a few oil-producing wells. Twenty-five samples were collected from an old oilfield area with a longer oil production history and a very dense network of oil-producing wells. All the sample sites in the new and old oilfield areas were located away from residential areas and any other industrial sources except those related to oil production that might lead to additional potential inputs of methylsiloxanes into the lakes from which the samples were collected. Eight samples were collected from the reference area located far away from the dense oilfield area. Considering that methylsiloxanes are volatile and have low solubility in water, Knoerr et al. (2017) developed a method that it utilizes low density polyethylene (LDPE) to inhibit loss of methylsiloxanes during sampling and transport to the laboratory. Due to lack of LDPE, other precautions have been taken during our sampling event. Briefly, surface water samples (0–0.5 m) were collected using a clean stainless steel bucket, and the samples were stored in 1 L glass bottles sealed with no headspace until sample pretreatment. All the glass bottles and the stainless steel bucket had been previously rinsed several times with the intended sample matrix. Surface sediment (approximately 3–5 cm deep) was collected, and the sample consisted of a composite of five grab samples using a stainless steel shovel. All the samples were stored at 4 °C until sample pretreatment, which occurred within 48 h of the sample collection. To investigate the loss of methylsiloxanes, field spikes were prepared in triplicate at the sampling site by adding an acetone solution (50 μL, 100 μg/L) of D4, D5 and D6 to ultrapure water. And the loss of D4, D5 and D6 in field spikes was about 4–5% before sample pretreatment.

In an oilfield, the oilfield combination station is an important component of the crude oil gathering and transportation system. Crude oil from oil wells is gathered and transported to the oilfield combination station and then preliminarily processed before being transported to a long-distance oil delivery pipe. Sometimes, due to emergency situations and operation reasons, the wastewater discharged by the oilfield combination station becomes an important pollution source to surface water. In the present study, wastewater samples ($n = 6$) were collected from

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