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Carbonate and carbon isotopic evolution of groundwater contaminated by produced water brine with hydrocarbons

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ABSTRACT

The major ionic and dissolved inorganic carbon (DIC) concentrations and the stable carbon isotope composition of DIC ($\delta^{13}C_{DIC}$) were measured in a freshwater aquifer contaminated by produced water brine with petroleum hydrocarbons. Our aim was to determine the effects of produced water brine contamination on the carbonate evolution of groundwater. The groundwater was characterized by three distinct anion facies: HCO_3^--rich, SO_4^{2-}-rich and Cl^--rich. The HCO_3^--rich groundwater is undergoing closed system carbonate evolution from soil $CO_{2(g)}$ and weathering of aquifer carbonates. The SO_4^{2-}-rich groundwater evolves from gypsum induced dedolomitization and pyrite oxidation. The Cl^--rich groundwater is contaminated by produced water brine and undergoes common ion induced carbonate precipitation. The $\delta^{13}C_{DIC}$ of the HCO_3^--rich groundwater was controlled by nearly equal contribution of carbon from soil $CO_{2(g)}$ and the aquifer carbonates, such that the $\delta^{13}C$ of carbon added to the groundwater was −11.6‰. In the SO_4^{2-}-rich groundwater, gypsum induced dedolomitization increased the ^{13}C such that the $\delta^{13}C$ of carbon added to the groundwater was −9.4‰. In the produced water brine contaminated Cl^--rich groundwater, common ion induced precipitation of calcite depleted the ^{13}C such that the $\delta^{13}C$ of carbon added to the groundwater was −12.7‰. The results of this study demonstrate that produced water brine contamination of fresh groundwater in carbonate aquifers alters the carbonate and carbon isotopic evolution.

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1. Introduction

Produced water brine containing petroleum hydrocarbons from oil field operations are a major source of groundwater contamination at thousands of sites across the USA and the world (e.g., Kharaka and Otton, 2007; Otton et al., 2007). Produced water brine is an environmental contaminant and investigations of produced water brine contaminated sites focus on defining the spatial extent of contaminant plumes, the magnitude of contamination and the short to long term attenuation of the contamination from major ion chemistry (e.g., Herkelrath et al., 2007; Kharaka et al., 2007; Whittemore, 2007).

Many studies that have investigated the effects of the chemical and biological degradation of petroleum products (e.g., jet fuel or diesel) on carbonate evolution in fresh groundwater have used concentrations of dissolved inorganic carbon (DIC) and stable carbon isotopes ($\delta^{13}C$) of DIC (e.g., Fang et al., 2000; Godsy et al., 2003; Atekwana et al., 2005; Scow and Hicks, 2005; Whittemore, 2007; Parker et al., 2012). Recently, Su et al. (2013) investigated a site contaminated by hydrocarbons from oil field activities. However, produced water brine was not an important component of the contaminants at the site. Most studies investigating highly saline water contamination are focused on salt-water intrusion in coastal aquifers where hydrocarbon contamination is usually not a concern (e.g., Cates et al., 1996; Nicholson and Fathepure, 2005; Ulrich et al., 2009; Currell and Cartwright, 2011). The produced water brine and the chemical and biological degradation of petroleum hydrocarbons are likely to have a significant impact on the carbonate evolution of contaminated fresh groundwater.

The carbonate evolution of fresh groundwater can be determined from equilibrium and mass balance approaches based on weathering of watershed and aquifer rocks (Drever, 1997). Data on the chemical reactions and their effects on carbonate evolution occurring in fresh groundwater contaminated by produced water brine with hydrocarbons are lacking. In this study we investigate carbonate reactions in fresh groundwater that was contaminated by produced water brine with hydrocarbons. We aim to develop an understanding of the carbonate evolution in a freshwater aquifer contaminated by produced water brine. Our objectives were to (1)

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document the spatial and temporal chemical and stable carbon isotopic characteristics of fresh groundwater contaminated by produced water brine and (2) determine the effect the contamination and subsequent reactions on the carbonate evolution.

2. Study site

2.1. Site history

The study site is located at an abandoned oil production site in Skiatook, Oklahoma, USA (Fig. 1). The study site (site A) is one of two sites in Oklahoma previously investigated by the US Geological Survey (USGS) under the Osage-Skiatook Petroleum Environmental Research (OSPER) project. Between the mid 1910's to the late 1970's, about 100,000 barrels of oil was extracted and the produced water brine associated with the operation was stored in unlined, earthen pits on site (Kharaka et al., 2005). The produced water brine was discharged into an ephemeral creek to the north of the site creating a salt scar (Kharaka et al., 2005; Otton et al., 2007). The salt scar, with erosion up to 2 m below the surface, extends from the pre-1937 pits to the lakeshore of the NE cove of Skiatook Lake (Fig. 1). Skiatook Lake was constructed in 1987, creating a peninsula at the study site. The USGS identified the produced water brine at the study site to be a Na—Ca—Cl brine with a salinity of ~150,000 mg/L total dissolved solids, and high concentrations of Mg^{2+}, Sr^{2+} and NH_4^+ and low concentrations of SO_4^{2-} and H_2S (Kharaka and Otton, 2003). Over time, the produced water brine stored in the earthen pits seeped into the fresh groundwater creating a high salinity hydrocarbon plume (Herkelrath et al., 2007).

2.2. Geology

The study site is nested in a saddle between two hills, with a drainage divide running generally NW—SE through the middle of the site (Fig. 1). Detailed geology of the study site is reported by Otton et al. (2007) and is briefly summarized below. Five units identified from surface mapping and coring are shown in the schematic cross section (Fig. 2) along profile A—A′ shown in Fig. 1. The surficial sediments consist of unconsolidated very fine grain eolian sands and colluvium in a sandy to clayey matrix. Unit 1, which is a weathered sandstone is of limited areal distribution and only found in the northwest portion of the site. Unit 2 occurs throughout the site and consists of weathered, very fine-grained sandstone and clayey sandstone cemented by Fe oxyhydroxide. Unit 3 intertongues with Unit 2 and consists mostly of weathered shale, sandy siltstone, sandy mudstone and sandstone which is exposed within the salt scar. The unweathered shale contains abundant pyrite and gypsum. Unit 4 consists of dolomite cemented unweathered sandstone, clay partings and thin shale beds that are carbonaceous and have fossil detritus. Unit 5 contains interbedded muddy sandstone, shale, siltstone and mudstone, with the sandstone cemented by dolomite.

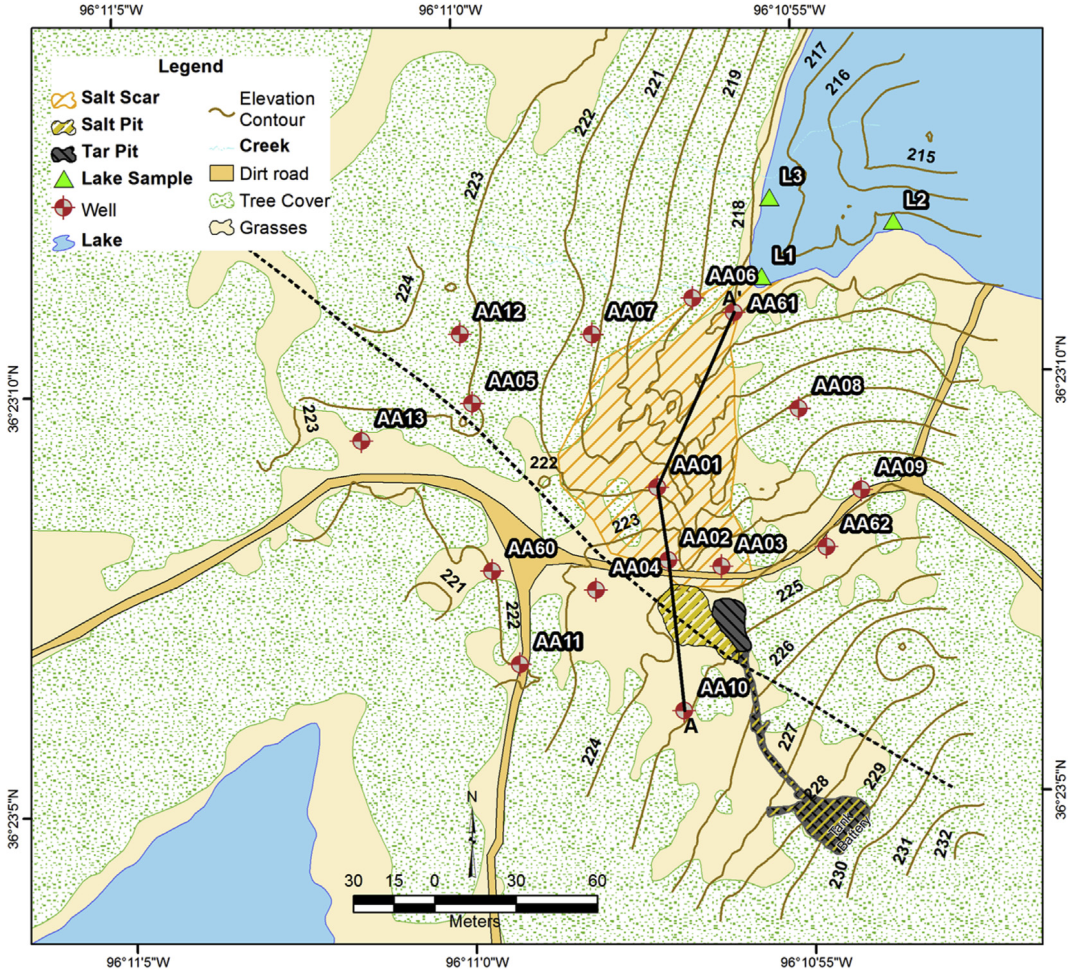

Fig. 1. Map of study site (Site A at Skiatook Lake, Osage County), Oklahoma, USA showing monitoring well locations, salt scar, oil and salt pits and transect A—A′. The dash line depicts the drainage divide at the study site.

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