



## Hydrogeochemistry and coal-associated bacterial populations from a methanogenic coal bed



Elliott P. Barnhart<sup>a,f,g,\*</sup>, Edwin P. Weeks<sup>b</sup>, Elizabeth J.P. Jones<sup>c</sup>, Daniel J. Ritter<sup>d</sup>, Jennifer C. McIntosh<sup>d</sup>, Arthur C. Clark<sup>e</sup>, Leslie F. Ruppert<sup>c</sup>, Alfred B. Cunningham<sup>f,h</sup>, David S. Vinson<sup>i</sup>, William Orem<sup>c</sup>, Matthew W. Fields<sup>f,g,j</sup>

<sup>a</sup> U.S. Geological Survey, 3162 Bozeman Ave, Helena, MT 59601, United States

<sup>b</sup> U.S. Geological Survey, National Research Program, Denver Federal Center, Lakewood, CO 80225, United States

<sup>c</sup> US Geological Survey, 12201 Sunrise Valley Drive, Reston, VA 20192, United States

<sup>d</sup> Department of Hydrology and Water Resources, University of Arizona, Tucson, AZ 85721, United States

<sup>e</sup> U.S. Geological Survey, Denver Federal Center, Lakewood, CO 80225, United States

<sup>f</sup> Center for Biofilm Engineering, Montana State University, Bozeman, MT 59717, United States

<sup>g</sup> Department of Microbiology and Immunology, Montana State University, Bozeman, MT, 59717, United States

<sup>h</sup> Department of Civil Engineering, Montana State University, Bozeman, MT, 59717, United States

<sup>i</sup> Department of Geography & Earth Sciences, University of North Carolina at Charlotte, Charlotte, NC, 28223, United States

<sup>j</sup> National Center for Genome Resources, Santa Fe, NM, 87505, United States

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### ABSTRACT

Biogenic coalbed methane (CBM), a microbially-generated source of natural gas trapped within coal beds, is an important energy resource in many countries. Specific bacterial populations and enzymes involved in coal degradation, the potential rate-limiting step of CBM formation, are relatively unknown. The U.S. Geological Survey (USGS) has established a field site, (Birney test site), in an undeveloped area of the Powder River Basin (PRB), with four wells completed in the Flowers-Goodale coal bed, one in the overlying sandstone formation, and four in overlying and underlying coal beds (Knoblach, Nance, and Terret). The nine wells were positioned to characterize the hydraulic conductivity of the Flowers-Goodale coal bed and were selectively cored to investigate the hydrogeochemistry and microbiology associated with CBM production at the Birney test site. Aquifer-test results indicated the Flowers-Goodale coal bed, in a zone from about 112 to 120 m below land surface at the test site, had very low hydraulic conductivity (0.005 m/d) compared to other PRB coal beds examined. Consistent with microbial methanogenesis, groundwater in the coal bed and overlying sandstone contain dissolved methane (46 mg/L average) with low  $\delta^{13}\text{C}$  values ( $-67\%$  average), high alkalinity values (22 meq/kg average), relatively positive  $\delta^{13}\text{C}$ -DIC values (4‰ average), and no detectable higher chain hydrocarbons,  $\text{NO}_3^-$ , or  $\text{SO}_4^{2-}$ . Bioassay methane production was greatest at the upper interface of the Flowers-Goodale coal bed near the overlying sandstone. Pyrotag analysis identified *Aeribacillus* as a dominant *in situ* bacterial community member in the coal near the sandstone and statistical analysis indicated *Actinobacteria* predominated coal core samples compared to claystone or sandstone cores. These bacteria, which previously have been correlated with hydrocarbon-containing environments such as oil reservoirs, have demonstrated the ability to produce biosurfactants to break down hydrocarbons. Identifying microorganisms involved in coal degradation and the hydrogeochemical conditions that promote their activity is crucial to understanding and improving *in situ* CBM production.

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## 1. Introduction

### 1.1. Biogenic coal bed methane

Biogenic coalbed methane (CBM) has become an important natural gas resource in the United States, Canada, Australia, India, and China

(Palmer, 2010). Accumulation of CBM in deep coal beds is primarily a by-product of thermal coalification processes, while in shallower coal beds, such as those in the Powder River Basin (PRB) in southeastern Montana and northeastern Wyoming, biogenic CBM accumulates from the activity of *in situ* microbial communities (Barnhart et al., 2013; Faiz and Hendry, 2006; Ritter et al., 2015; Strąpoc et al., 2011). The microbial communities in the PRB have produced an estimated 14.26 trillion cubic feet (TCF) of biogenic CBM that has been developed commercially over the past few decades (Strąpoc et al., 2011; U.S.

\* Corresponding author at: USGS Wyoming-Montana Water Science Center, 3162 Bozeman Ave., Helena, MT 59601, United States.

Geological Survey National Assessment of Oil and Gas Resources Team et al., 2014). There were approximately 17,500 active CBM production wells in the PRB in 2008, but production was short-lived with an average productive well life of <12 years (Meredith et al., 2012; Sando et al., 2014). Push-pull tests indicate the microbial communities within PRB coal beds are active and have generated biogenic CBM in the recent geologic past (Ulrich and Bower, 2008). Several hypothetical models for active microbial CBM formation have been proposed that involve the anaerobic degradation of coal by bacteria, which results in the formation of precursor metabolites that cross-feed methane-producing archaea (methanogens) (Jones et al., 2010; Meslé et al., 2013; Strapoć et al., 2008). The rate-limiting step of microbial CBM formation appears to be the initial bacterial degradation of coal organic matter constituents (Wawrik et al., 2011). Currently, neither the specific bacterial communities and enzymes nor the optimal *in situ* conditions for bacterial coal degradation are known.

Increasing coal bioavailability and/or stimulating specific bacterial populations involved in coal degradation with microbial enhanced CBM (MECoM) technology could increase *in situ* CBM production and sustain the life of wells in the PRB and other basins. Private companies have implemented pilot scale MECoM tests in the PRB, but have not been able to predict or document the associated changes to the bacterial community, partly due to the current lack of understanding of the *in situ* bacterial community dynamics within the coal beds (Ritter et al., 2015). Recent advances in DNA sequencing technology coupled with improved sampling of the subsurface matrix have allowed for better characterization of microbial communities in environmental samples (Barnhart et al., 2013). Next-generation sequencing provides the coverage required to establish correlations between the microbial community composition and environmental conditions that drive the microbial community dynamics. These techniques were utilized in this study along with a multidisciplinary approach to investigate the hydrogeochemical characteristics of several major PRB coal beds along a vertical transect.

## 1.2. Microbial CBM formation in the PRB

The PRB contains one of the most significant, low-rank (subbituminous) coal deposits in the world (Molnia and Pierce, 1992). Low-rank coals are thought to contain a higher proportion of bioavailable compounds and macerals richer in heteroatoms than higher-rank coals (Strapoć et al., 2011). Production of CBM from the PRB mostly occurs from coal beds in the Paleocene Tongue River Member of the Fort Union Formation (Fig. 1) (Ellis et al., 2002; Flores et al., 2008; Rice et al., 2008; Scott et al., 2011). Several coal beds in the Tongue River Member, which are investigated here, include the Knobloch, Nance, Flowers-Goodale, and Terret coal beds (Fig. 2). It has been suggested that groundwater recharge originally inoculated these and other PRB coal beds with the active CBM-producing microbial communities (Strapoć et al., 2011). Shallow groundwater in recharge areas of these coal beds, which commonly contains sulfate, has low methane concentrations. Sulfate-reducing bacteria can outcompete methane-producing methanogens for substrates (Meredith et al., 2012; Raskin et al., 1996). As groundwater moves deeper in the coal beds, bacterially-mediated sulfate reduction lowers the aqueous sulfate levels and increases the bicarbonate concentrations so methanogenesis may proceed (Brinck et al., 2008).

Microbial methanogenesis in the PRB is identifiable by distinctive gas and groundwater geochemical signatures. PRB gas is dry (very low in ethane or  $\geq C_3$  hydrocarbons) and exhibits fairly negative  $\delta^{13}C\text{-CH}_4$  values typical of microbial gas ( $-83\%$  to  $-51\%$ ; Bates et al., 2011; Flores et al., 2008; Gorody, 1999). The isotope fractionation that generates negative  $\delta^{13}C\text{-CH}_4$  values also yields positive values of  $\delta^{13}C\text{-CO}_2$ . Biodegradation that includes sulfate reduction and methanogenesis generates  $CO_2$ , although with different (opposite) isotopic values for sulfate reduction and methanogenesis, respectively. The  $\delta^{13}C\text{-CO}_2$  values for sulfate reduction tend to be lower, closer to the value of organic matter (coal), while  $\delta^{13}C\text{-CO}_2$  values for methanogenesis become

AGE	Stratigraphic Units Powder River Basin, Montana		
QUATERNARY	White River Formation		
PLIOCENE-OLIGOCENE	surficial deposits		
EOCENE	Wasatch Formation		
PALEOCENE	Fort Union Formation	Tongue River Member	
		Lebo Member	
		Tulloch Member	
LATE CRETACEOUS	Hells Creek Formation		
	Fox Hills Sandstone		
	Pierre Shale		
	Niobrara Formation		
	Carlile Shale		
	Greenhorn Formation		
	Belle Fourche Formation		
	Mowry Shale		
	EARLY CRETACEOUS	Muddy Sandstone	
		Thermopolis Shale	
Fall River Formation			
Lakota Formation			

Fig. 1. Generalized stratigraphic column of the Montana part of the Powder River Basin. Coal beds examined in this study occur in the Paleocene Fort Union Formation, Tongue River Member. (Modified from Scott et al., 2011, after Ellis et al., 2002).

increasingly more positive with greater extents of methanogenesis (Osborn and McIntosh, 2010). The  $\delta^{13}C\text{-CO}_2$  values of producing gas wells in the PRB range from  $-25\%$  to  $22\%$  ( $\%$ ; Bates et al., 2011; Flores et al., 2008; Gorody, 1999). Due to microbial respiration, PRB coal bed groundwater exhibits high alkalinity concentrations (6–50 meq/kg; Bates et al., 2011).

Regionally, groundwater in the PRB is believed to move northward to northeastward but the relative importance (or contribution) of regional versus local flow systems and vertical versus horizontal flow components are not well understood (Bates et al., 2011; Rice et al., 2008). Understanding the flow velocity and direction can help interpret the extent of methanogenesis and the environment within which methanogenesis occurs. A carefully designed aquifer test with specific well placement and sophisticated analytical techniques can be used to determine local groundwater flow and hydraulic conductivity within a

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