Microbial Ecology of Hydraulic Fracturing: Implications for Sustainable Resource Development

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The economics of oil and gas production from deep, tight, hydrocarbon-rich shale formations has been considered unfavorable until recently. Advancements in drilling and stimulation technologies have greatly improved the economics of production from deep from these reservoirs. The exploitation of these natural resources has led to greatly increased domestic production volumes of natural gas and oil in the United States. Similar unconventional resources in Australia, Europe, South America and China are poised transform global fossil energy markets through localized production of fuels for transportation, heating, and electricity generation. However, as the industries associated with unconventional oil and gas production migrate into regions without a history of production, new challenges arise and in particular with regard to the management of wastewater that is coproduced at the surface and the microbial communities that drive geochemical processes and ultimately management strategies.

The focus of this article is on the microbial ecology and biogeochemical processes that impact production of oil and gas, management of wastewater, and product quality from hydraulically fractured wells. Hydraulic fracturing is discussed from the perspective of water management, including the volume and make-up of fracturing fluids that give rise to produced water microbiology. Recent studies on the chemistry and microbiology of produced water from the Marcellus and Barnett Shale regions are discussed. Microbial ecology present at wellheads is described as well as that of mature communities in stratified impoundments for produced water from hydraulic fracturing. A prospectus on the implications and control of these communities is presented.

Hydraulic Fracturing Overview

The recent surge in unconventional oil and gas development is the result of greatly improved economics, enabled by an amalgamation of horizontal drilling and hydraulic fracturing. Horizontal drilling allows access to a far greater portion of a formation than vertical wells by following the horizontal contour of the formation for thousands of meters. In this manner, horizontal wells also greatly reduce the surface impacts my minimizing the number of wells required to develop a particular area. That said, horizontal drilling, a technology that has been broadly applied since the 1980s, was insufficient for the favorable economics of unconventional development. Hydraulic fracturing, (aka fracking) is used in conjunction with horizontal wells to increase the permeability of the formation and extend the radius of influence of the well-bore for an overall increase the productive surface are of the reservoir.

Hydraulic fracturing is the process of pumping fracturing fluid into the wellbore at a rate that exceeds the ability of the formation to accept without fracturing. Resistance to the injection increases pressure in the well-bore above the fracture pressure of the formation rock, breaking the formation. Although a number of fracturing fluids exist, the most common is known as "slickwater" and consists of water as a solvent for chemical modifiers and carrying fluid for proppant. The mixture of water, sand, and chemicals is pumped into the well-bore at pressures of 500-900 atm, depending on the needs of the target formation. The proppant, most commonly sand, flows into the preexisting and newly initiated fractures in the formation and holds them open after pumping has stopped and pressure decreases. In the Marcellus Shale region, hydraulic fracturing may require 6-15 million liters of water, depending on the depth to the target formation and length of the horizontal leg of the well (Gregory 2011). The chemical additives used in fracturing fluid vary by the needs of the target formation for optimal product recovery, but may include those described in Table 1.

Flowback and Produced Water

Following pumping for hydraulic fracturing but prior to the production of hydrocarbons, the pressure is allowed to dissipate and fluid returns to the surface through the wellhead and is collected and stored. This initial water that is produced to the surface prior to product recovery is referred to as “flowback”. Although water continues to be produced to the surface along with hydrocarbon products and is expected over the lifetime of the well, this wastewater is managed differently than flowback and is referred to as “produced water”. In essence, the difference flowback and produced water is merely operational because both are produced (to the surface) from the fractured formation. However, because the downhole pressure is greatest immediately following hydraulic fracturing, the rate at which flowback is returned to the surface is greater than that of produced water. As a result, flowback is managed differently that produced water.

Flowback and produced waters are a mixture of hydraulic fracturing fluid and water that was originally present in the formation. Therefore, the quantity and quality of water that returns to the surface during flowback is variable among the unconventional oil and gas plays and is also dependent on the hydraulic fracturing procedure used. For example, flowback from hydraulic fracturing of the Marcellus Shale results in 9-53% (Average 10%) of the injected volume returning (Vidic 2013). The concentration of total dissolved solids (TDS, including salts and metals) in the water that returns to the surface is variable in time; the earliest flowback from a well resembles the hydraulic fracturing fluid itself and has the lowest concentration of TDS, while the later flowback has much higher TDS, more resembling the chemistry of the formation water. Generally speaking, the quality of produced water from the same formation is also highly variable (Barbot 2013). The study by Barbot et al. reveals that the TDS in produced from the Marcellus varied from ~1.0 to 345g/L with an average of 106g/L. Additionally revealed was that the TDS in the produced water was predominantly from the ions, Cl^-, Na^+, and Ca^{2+} and further characterized by high concentrations of magnesium, barium and strontium. Organic compounds in produced water include those introduced with the fracturing fluid as well as polycyclic aromatic hydrocarbons (PAHs), heterocyclic compounds, alkyl phenols, aromatic amines, alkyl aromatics (alkyl benzenes, alkyl biphenyls), long-chain fatty acids, and aliphatic hydrocarbons (Orem 2014), all of which may be used as carbon sources and electron donors for bacterial growth.

The majority of produced water in the United States is disposed through deep-well injection (Clark 2009). However, because there are few deep-well injection options in Pennsylvania, reuse of flowback as the make-up water for subsequent hydraulic fracturing has become the preferred management option in the Marcellus region. Additionally, reuse of flowback reduces the need for freshwater withdrawals and reduces the costs incurred for transportation and treatment or disposal. Wastewater brines used for subsequent hydraulic fracturing are used directly and diluted with a freshwater source, treated to remove solids and divalent cations prior to reuse. The reuse of flowback (and produced) water for hydraulic fracturing is a novel technology and management solution for oil and gas wastewater brines but a recent analysis of data from the Pennsylvania DEP, revealed a ~90% reuse rate of oil and gas brines from the Marcellus (Maloney 2012).

Flowback is typically impounded at the surface prior to subsequent disposal, treatment or reuse. While treatment or disposal may take place immediately following the flowback period, reuse of flowback for subsequent fracturing requires storage of a variable volume of wastewater for variable periods of time prior to the next hydraulic fracturing that is suitable and economical for the source of brine. Flowback is either transported by truck or pumped to centralized impoundments that serve multiple wells or multiple pads with multiple wells. The volume of impoundments may vary from ~10,000 to 60,000 m^3, depending on the intended use and the number of wells served. The flowback and produced water brines in impoundments may be stored for periods of weeks or months. During this storage, microbes introduced to the impoundment in the flowback and produced fluids evolve and give rise to water management challenges such as the production of malodorous compounds, biofouling of the formation and production equipment and infrastructure, biocorrosion, and the alteration of the solubility of metals including radionuclides. Although not well documented for unconventional reservoir development, these problems are well understood by the oil and gas industry through experience with development of conventional onshore and offshore reservoirs. The issues associated with the proliferation of bacteria during oil and gas production are ubiquitous and manifest themselves throughout the production infrastructure and are costly to manage (Ollivier 2005).

Microbial Communities in Produced Fluids

Microbial processes that impact oil production from conventionally developed reservoirs are well documented (Orem 2005; Van Hamme 2003). In these reservoirs, the stimulation of bacteria may result in detrimental issues such as reservoir fouling, biocorrosion and product souring (sulfidization) while it may conversely provide benefit through enhancing product recovery, removal of soured product and paraffins. The microbial ecology of these processes in conventional reservoirs is well understood. In contrast, there are scant few studies on the detrimental or beneficial impacts of bacteria or the microbial ecology of unconventional oil and gas development. This knowledge gap exists despite similar concerns over well souring and biofouling and the knowledge that the microbiology is costly to control.

A recent study of the microbial community in wellhead samples from the Marcellus during flowback and produced water phases reveal that the microbial community changes in time with the geochemistry (Murali Mohan 2013b). The community that was present on day 1 of the flowback closely resembled that which was present in the fracturing fluid itself and the source water (Figure 1). The microbes in the early flowback were most similar to species associated with non-halophilic, aerobic and phototrophic metabolisms; all of which would be expected in a make-up water obtained from a freshwater surface source. However, later into the flowback period, the TDS increases as do the number of bacterial species that are most closely related to halophilic, thermophilic anaerobes. The rise in the abundance of these halophilic anaerobes occurs at the expense of the non-halophilic aerobes and phototrophs and at the expense of diversity (Murali Mohan 2013b). For example, note the gradual decrease in *Rhodobacterales* (freshwater phototrophs) and increase in *Halanaerobiales* (anaerobic halophiles) during hydraulic fracturing of a well in the Marcellus shale (Figure 1). This emergence of halophilic anaerobes was concomitant with the emergence of anaerobic geochemistry (e.g. Fe^{2+} and HS^-) in the water. The decreased diversity with time is the result of the loss of virtually all species present in the initial flowback except the *Halanaerobiales* which eventually represents >99% of the community, as recent studies of produced water from the Marcellus confirm (Cluff 2014). During the late, produced water phase, all of the bacteria identified in the sample were closely associated known, anaerobic, fermentative and sulfur-reducing bacteria in the *Halanaerobiales*. A study of flowback from the hydraulic fracturing of the Barnett Shale revealed a community that was similarly changing with time evolving towards one that was well-adapted to anoxic and saline conditions (Struchtemeyer 2011b).

It is important to note that the anaerobic and halophilic species present in great abundance in the produced water were revealed by next generation sequencing to also be present in very low abundance in hydraulic fracturing fluid. This suggests that the organisms in the produced water may be originating at the surface and introduced by hydraulic fracturing, rather than native to the connate water in the deep subsurface reservoir. But there is a chicken and egg problem revealed by the fact that the trucks and equipment for handling the water likely have prior exposure to brines from hydraulic fracturing. Therefore the source of the organisms could be any equipment that was used for hauling the water. As a result, the source of the organisms in flowback and produced water is poorly understood. While bacteria are well known to inhabit deep subsurface environments (Fredrickson 2006), their presence in natural, connate brine from Marcellus and Barnett formations for example has not been documented. Obtaining representative samples that are assured to be free of bacterial contamination from drilling or sampling is difficult and costly. Moreover, these formations are the source-rock for in-place hydrocarbons in extremely low permeability rock. The formations and any bacteria that were present when the formation sealed were at temperatures and pressures sufficient to produced natural gas from biosolids; >120 ºC for millions of years. With those temperatures, durations, and limited permeability for nutrients delivery robust and dynamic microbial communities that are observed to evolve rapidly in wellhead samples are not likely. However, shallow and lower temperature hydrocarbon-rich formations and those that contain natural fractures that may serve as pathways for exchange of fluids, such as the Antrim basin in Michigan are more likely to have a native community (Martini 1998). Regardless of the source of these organisms at the wellhead, they come in contact with equipment for handing and transporting fluids and eventually are in an impoundment where new geochemistry, impacted by management strategy and the surface environment drives further changes in these adaptive bacterial communities.

Microbial Communities in Impoundments

Impoundments for storage of flowback and produced water have robust and dynamic microbial communities that similarly correspond with the geochemistry of the impoundment water. Impoundments receive water and bacteria from a variety of sources, including the wells, source water, drilling muds (Struchtemeyer 2011a), equipment, and the environment (rain, dust, animals, runoff, etc). Which species are capable of surviving depends on their ability to adapt to the brine concentrations above 100g/L and the spatially and temporally dynamic geochemistry that occurs as a result of environmental processes and human intervention during management of impoundments.

Only a single study of the microbiology in flowback water impoundments has been performed (Murali Mohan 2013a). Most importantly, the study finds that microbial communities in impoundments will stratify with geochemistry and be impacted by the management strategy. The onset of anaerobic conditions in an impoundment may be observed by the production of malodorous compounds from bacterial activity such as volatile fermentation products and sulfide gas. This condition is an aesthetic issue from the production standpoint and is commonly controlled by the addition of biocides or aeration of the impoundment. Samples collected at depths ranging from the surface to the bottom of aerated and unaerated impoundments revealed that the geochemistry and microbiology in the aerated impoundment was homogenous with depth (Figure 2). Moreover, findings show that the microbial community at all depths contained species that were most similar to known aerobes and phototrophs. This follows from the expected vertical mixing of the community between the bottom and surface depths from vigorous aeration. In contrast, the untreated impoundment was stratified from a geochemical and microbial perspective. Bacteria that were most similar to aerobes and phototrophs were confined to the surface layer of the impoundment while anaerobes such as sulfidogenic and methanogenic bacteria were confined to the anoxic middle and bottom layers. In all samples, regardless of treatment or depth, the species present were most similar to taxa that are known halophiles, showing that the brine conditions of the water are an overarching driver of the ecology. While this study does not reveal the activity of the organisms, it does suggest that the bacteria in impoundments will adapt to the conditions imposed and this, despite the addition of biocides to the initial fracturing water (Murali Mohan 2013a), as reported from studies of flowback from the Barnett Shale as well (Struchtemeyer 2011b).

Implications for Unconventional Oil and Gas Production

Research into the microbiology of unconventional oil and gas development is a nascent focus area for engineers and scientists. While analogies with conventional petroleum microbiology may be highlighted, on the water management side of the development equation, many research questions exist; the answers to which will impact the practice, as well as the economic and environmental sustainability of hydraulic fracturing for oil and gas production. Most importantly, a unifying theme from the existing studies is that the communities present in wellheads and impoundments appear to be dynamic in time and space, indicating that biocides may not be working or used as intended. Moreover that diversity drops sharply during flowback while it becomes enriched with survivor species. The implication is that recycling of flowback and produced water for subsequent hydraulic fracturing may be introducing deleterious bacteria that are well-adapted to the harsh environment of the well (and biocides) to new wells. These well adapted bacteria will grow quickly, advance the onset of sulfide production, and be more recalcitrant to treatment options. Another unifying theme from the existing studies is the presence and great abundance of species that are similar to *Halanaerobium congolense*, a sulfidogen. Sulfide production in wells leads to human and environmental health risks, leads to corrosion, and the costly degradation of product quality. This organism has been identified in produced waters form conventional development, but is of significance here because it cannot reduce sulfate and instead uses thiosulfate and sulfur as electron acceptors for sulfide production. Standardized tests employed by the oil and gas industry for assessing sulfidogenic potential in produced water, rely on assessing the numbers of sulfate reducing bacteria. Therefore, these conventional tests will yield false negative reports for sulfide production potential, a risk for the industry. New tests that enable assessment of sulfide production from sulfur-reducing bacteria should be employed for unconventional wells.

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Additive Class	Example	Purpose
Acid	Hydrochloric acid	Dissolve minerals, initiate fractures
Biocide	Glutaraldehyde, DBNPA, Quaternary Ammonium Chloride	prevent bacterial growth, minimize biocorrosion and sour-gas formation
Breaker	Ammonium persulfate, sodium chloride	control breakdown of gel polymers
Corrosion inhibitor	Isopropanol, methanol, acetaldehyde	Prevent pipe corrosion by acid additive
Crosslinker	Borate salts, ethylene glycol	Maximize viscosity as fluid temperature rises
Friction reducer	Polyacrylamide	Minimize friction and reduce pumping costs
Gelling agent	Guar gum, hydroxyethyl cellulose, polysaccharides	Suspend proppant
Iron Control	Citric acid, thioglycolic acid	Chelate iron, prevent scale formation
Oxygen scavenger	Ammonium bisulfide	Reduce pipe corrosion
Non-emulsifier	Lauryl Sulfate	Break oil-water mixtures to improve recovery
pH adjustment	Sodium hydroxide, carbonate	Maintain efficacy of other additives
Scale inhibitor	Acrylamide, phosphonic acid salt	Reduce scale formation in pipes
Surfactant	Lauryl Sulfate, Ethanol, isopropyl alcohol, 2-butoxyethanol	Decrease surface tension for better water recovery

Table 1. Some common classes of hydraulic fracturing fluid additives. Information was sourced from (Gregory 2011) and (Vidic 2013).

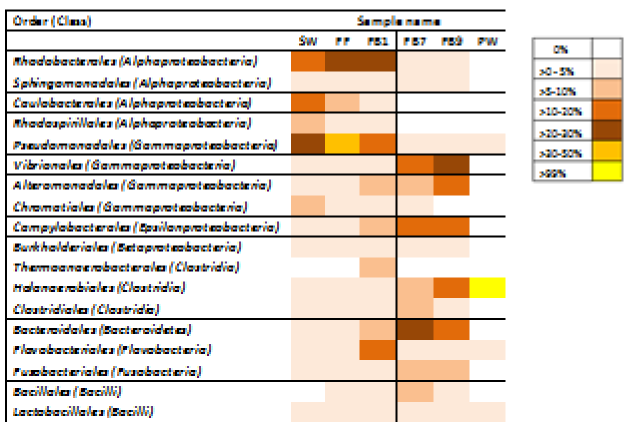

Figure 1. Heat map revealing the relative abundance of bacterial taxa in water samples associated with hydraulic fracturing of the Marcellus Shale as revealed by 454 pyrosequencing. SW = Source Water, FF = Fracturing Fluid, FB1-9 = Flowback on day 1-9, and PW = Produced water on day 187.

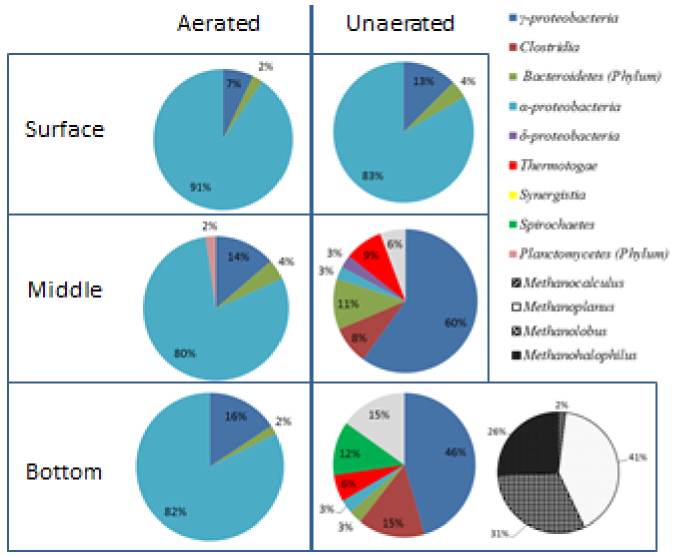

Figure 2. Relative abundance of bacterial taxa in flowback water impoundments at various depths. Data was generated by clone libraries and subsequent sequencing of 16s rRNA genes recovered from samples. Sequences with similarity to Archaea were only recovered from the bottom of the unaerated impoundment and are shown in black and white. Data adapted from (Murali Mohan 2013a)