



The fate of residual treatment water in gas shale



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ABSTRACT

More than $2 \times 10^4 \text{ m}^3$ of water containing additives is commonly injected into a typical horizontal well in gas shale to open fractures and allow gas recovery. Less than half of this treatment water is recovered as flowback or later production brine, and in many cases recovery is <30%. While recovered treatment water is safely managed at the surface, the water left in place, called residual treatment water (RTW), slips beyond the control of engineers. Some have suggested that this RTW poses a long term and serious risk to shallow aquifers by virtue of being free water that can flow upward along natural pathways, mainly fractures and faults. These concerns are based on single phase Darcy Law physics which is not appropriate when gas and water are both present. In addition, the combined volume of the RTW and the initial brine in gas shale is too small to impact near surface aquifers even if it could escape. When capillary and osmotic forces are considered, there are no forces propelling the RTW upward from gas shale along natural pathways. The physics dominating these processes ensure that capillary and osmotic forces both propel the RTW into the matrix of the shale, thus permanently sequestering it. Furthermore, contrary to the suggestion that hydraulic fracturing could accelerate brine escape and make near surface aquifer contamination more likely, hydraulic fracturing and gas recovery will actually reduce this risk. We demonstrate this in a series of STP counter-current imbibition experiments on cuttings recovered from the Union Springs Member of the Marcellus gas shale in Pennsylvania and on core plugs of Haynesville gas shale from NW Louisiana.

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Introduction

Production of gas from shale by horizontal drilling and high volume hydraulic fracturing (HVHF) offers a suite of environmental benefits while raising other environmental concerns (Howarth et al., 2011; Jackson et al., 2013). Concerns involving water quality, the topic we discuss here, arise because as much as $2 \times 10^4 \text{ m}^3$ of treatment water with additives is injected into a typical horizontal well that will tap the gas from ~83 acres of a ~45 m thick shale bed (Table 1). The additives prevent bacterial growth, prevent scaling of steel pipes, aid in rapid flow, prevent swelling of the clay minerals in the shale, and carry sand which props fractures open. This treatment water enters the gas shale via open fractures, but less than half is ever recovered as flowback or later production brine (Pagels et al., 2011; Striolo et al., 2012). In some gas shale plays, clean up and recovery of the fracture fluids prior to bringing gas on stream typically recovers only ~4–8% of the originally

injected volume of water (Richard Newhart, Encana; Oklahoma Geological Survey presentation, July 2011. Norman, Oklahoma).

The water initially injected into the subsurface is fresh, typically with a TDS content of 1–5 kppm (TDS = total dissolved solids). The treatment water that does return to the surface carries back natural components of the gas shale including salt, some metals, and radionuclides. This water tends to be highly saline, often with TDS contents of as much as 200 kppm (Gregory et al., 2011). While recovered treatment water is safely managed at the surface, the water left in place, called residual treatment water (RTW), slips beyond the control of engineers. The environmental concern that we address is whether this RTW, more than 10^4 m^3 per horizontal well, could eventually flow out of the gas shale and contaminate overlying groundwater.

The possibility of such eventual leakage and ground water contamination has been raised (Myers, 2012; Warner et al., 2012). Warner et al. (2012) classified 426 water samples from shallow aquifers in an $80 \times 160 \text{ km}$ area of northeastern Pennsylvania where hydraulic fracturing is currently being done within the Marcellus gas shale. This classification consists of 4 groups based the Br, Cl, Na, Ba, Sr, Li concentration in the samples and isotopic

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Table 1

Examples of treatment fluid volumes. For a typical Marcellus well the approximate size of the stimulated reservoir volume assumes a 6000' (1830 m) lateral with a stimulation reaching 300' (91 m) on each side of the lateral (83 acres) and a vertical dimension of 150' (45.7 m). For a typical Haynesville well single stage data are reported.

<i>Marcellus well</i>		
Volume of Marcellus tapped	15.3 × 10 ⁶ m ³	(83 acres) (4047 m ² /acre) (45.7 m)
Volume of treatment water (83 acre well)	20,000 m ³	5,300,000 gallons
Volume of capillary-bound water	1.5–3 × 10 ⁵ m ³	(1–2% porosity) (15.3 × 10 ⁶ m ³)
Vol. of free water that could leak	8035 m ³	(2.4 m/45.7 m) (1% porosity) (15.3 × 10 ⁶ m ³)
Volume filled with gas	1.2 × 10 ⁶ m ³	(≈8% porosity) (15.3 × 10 ⁶ m ³)
<i>Haynesville well</i>		
Volume of treatment water (single stage)	1309 m ³	346,500 gallons
Total proppant in single stage	159,282 kg	352,257 lbs

ratios ⁸⁷Sr/⁸⁶Sr, ²H/H, ¹⁸O/¹⁶O, and ²²⁸Ra/²²⁶Ra. One group with high Br/Cl and Sr/Ca but low ⁸⁷Sr/⁸⁶Sr, their Type D waters, is interpreted to be diluted residual brine that migrated from the deep formations along cross formational pathways. Warner et al. (2012) imply that this natural migration might be ongoing today. By referring to the source repeatedly as the “Marcellus”, they imply that leakage is from the Marcellus and they suggest the pathways of natural gas leakage might be areas of higher risk for leakage of RTW. The Marcellus is portrayed as leaking now without any human assistance through cross-formational pathways and the concern is raised that hydraulic fracturing in the Marcellus could make this leakage worse. If this happens for the Marcellus it would be of concern for any HVHF gas or oil development globally.

The plausibility of RTW leakage upward to groundwater was amplified in recent models suggesting that high-permeability fractures connect the Marcellus directly to the water table (Myers, 2012). Issues with such models include imbibition of RTW into the Marcellus, the interconnectivity of fractures and faults between the Marcellus and groundwater, the role of multiphase flow, and the lack of a pressure drive (Engelder, 2012; Saiers and Barth, 2012). Modeling a high-permeability pathway to groundwater suggests that RTW might climb upward to drinking water aquifers in less than 10 years. Here we examine the extent to which the Warner et al. (2012) interpretation and the Myers (2012) model, collectively called the Warner–Myers hypothesis, may or may not be plausible. We do not address issues that may arise as a consequence of engineering failures between wellbore and casing.

Brines with low Na/Cl and high Br/Cl are residual brines produced by evaporation of seawater beyond the point where halite precipitates. They are thus distinct from saline waters produced by road salt and from normal low salinity ground waters in the area (Warner et al.'s other ground water types). The heart of Warner et al.'s (2012) argument is that: (1) the low ⁸⁷Sr/⁸⁶Sr ratio of these distinctive brines means that they must have come from formations the same age or older and the same depth or deeper than the Marcellus because pore waters have low strontium ratios only in these strata, (2) the brines are likely coming from the Marcellus to topographically low areas because these areas are more fractured and faulted and the fractures and faults connect to the Marcellus, and (3) hydrofracturing the Marcellus could make brine leakage from the Marcellus worse by increasing the permeability of the fractures and faults.

The possibilities raised by Warner–Myers hypothesis are extremely unlikely for four reasons. First, the near-total lack of free water in gas shale means that it cannot feed a steady upward leakage of the kind proposed (Zagorski et al., 2010). Second, the fact gas shale readily imbibes water, and only a fraction of the hydrofracturing treatment water is returned, shows that the treatment waters are flowing into, not out of, the shale (Engelder, 2012). Third, the high salinities (200–300 kppm) observed in flowback brines (RTW), produce significant osmotic fluid pressure gradi-

ents. Coupled diffusion–osmosis processes and the forces associated with surface tension and adhesion (capillary forces) propel water into the matrix of gas shale and generate the high salinities observed in the recovered RTW (Bryndzia, 2012). Fourth, although there may be other environmental issues worthy of attention during gas production by high volume hydraulic fracturing, the leakage of water and gas along natural pathways from gas-filled shales like both the Marcellus and Haynesville is essentially eliminated by capillary forces which have maintained overpressuring of the gas and brines between >100 My (the Haynesville) and >250 My (the Marcellus) (Cathles, 2001).

The purpose of this paper is to elaborate on these four reasons for the implausibility of the Warner–Myers hypothesis (i.e., that frack fluid migrates out of gas shale to contaminate groundwater). The implausibility becomes apparent with an understanding of how the Marcellus was deposited and evolved, why it imbibes water when it is hydrofractured, and why capillary and osmosis forces lead to the conclusion that the Warner–Myers hypothesis is misguided. New experimental data from imbibition experiments on both Marcellus and Haynesville shale suggest that the Warner–Myers hypothesis should be viewed with great skepticism.

The geological history of the Marcellus

The Marcellus is organic rich black shale of Middle Devonian age with up to 12 wt.% total organic carbon near the maximum flooding surface at the base of the Union Springs Member. The shale was deposited 389 million years ago under euxinic conditions (Engelder et al., 2011; Kohl et al., 2013; Lash and Engelder, 2011). The Marcellus basin was filled from the SE by a river delta system carrying larger volumes of clay and fine silt, whereas a carbonate bank fed the basin at a slower rate from the NW. This difference in source material and sedimentation rate led to differences in composition of sedimentary fill in the Marcellus Basin. The Marcellus is thus more carbonate-rich, has a higher wt.% TOC, has a lower water saturation and is thinner unit on the western side of the basin.

Over the 30 million years following its deposition, the Marcellus and surrounding organic-rich shales (e.g., the overlying Genesee/Burket, Rhinestreet, and Dunkirk/Huron) and other strata were buried to 1–2 km depths or more by sediments from either the Devonian Catskill Delta complex to the SE or the carbonate bank to the NW. During the initial phase of burial, shale porosity collapsed by mechanical compaction. This reduced the shale permeability which resulted in membrane filtration of expelled water and retention of much of the original solute load in the shale matrix. When the shales became sufficiently impermeable, compaction disequilibrium developed (Osborne and Swarbrick, 1997). The pore fluids became overpressured with respect to hydrostatic and came to support some of the overburden (Engelder and Oertel, 1985; Lash and Blood, 2007).

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