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Endocrine disrupting activities of surface water associated with a West Virginia oil and gas industry wastewater disposal site

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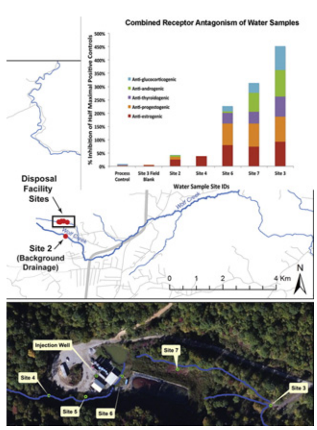
HIGHLIGHTS

- Oil and gas wastewater disposal may increase endocrine disrupting activity in water.
- Tested EDC activity in surface water near oil and gas wastewater injection site.
- Water downstream had significantly more EDC activity than reference water upstream.
- Downstream surface water antagonized five different nuclear hormone receptors.
- EDC activity downstream was above levels known to result in adverse health effects.

GRAPHICAL ABSTRACT



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ABSTRACT

Currently, >95% of end disposal of hydraulic fracturing wastewater from unconventional oil and gas operations in the US occurs via injection wells. Key data gaps exist in understanding the potential impact of underground injection on surface water quality and environmental health. The goal of this study was to assess endocrine disrupting activity in surface water at a West Virginia injection well disposal site. Water samples were collected from a background site in the area and upstream, on, and downstream of the disposal facility. Samples were solid-phase extracted, and extracts assessed for agonist and antagonist hormonal activities for five hormone receptors in mammalian and yeast reporter gene assays. Compared to reference water extracts upstream and distal to the disposal well, samples collected adjacent and downstream exhibited considerably higher antagonist activity for the estrogen, androgen, progesterone, glucocorticoid and thyroid hormone receptors. In contrast, low levels of agonist activity were measured in upstream/distal sites, and were inhibited or absent at downstream sites with significant antagonism. Concurrent analyses by partner laboratories (published separately) describe the

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Injection well
Wastewater disposal

analytical and geochemical profiling of the water; elevated conductivity as well as high sodium, chloride, strontium, and barium concentrations indicate impacts due to handling of unconventional oil and gas wastewater. Notably, antagonist activities in downstream samples were at equivalent authentic standard concentrations known to disrupt reproduction and/or development in aquatic animals. Given the widespread use of injection wells for end-disposal of hydraulic fracturing wastewater, these data raise concerns for human and animal health nearby.

1. Introduction

It has recently been demonstrated that chemicals used in and/or produced by unconventional oil and natural gas (UOG) operations include endocrine disrupting chemicals (EDCs) (Bolden et al., 2015; Kassotis et al., 2014; Webb et al., 2014). EDCs are exogenous chemicals or mixtures of chemicals that can interfere with any aspect of hormone action (Zoeller et al., 2012). As many as one thousand EDCs have been identified (TEDX, 2013), both synthetic and naturally occurring, that can directly interact with hormone receptors as agonists or antagonists (Tyler et al., 1998; Yang et al., 2006), or indirectly interact via modulating responses to endogenous hormones (Chen et al., 2007; Jansen et al., 2004), endogenous hormone levels (Chen et al., 2007; Hayes, 2002), or through other mechanisms (Diamanti-Kandarakis et al., 2009). EDCs can exhibit biological effects at very low environmental concentrations (Roepke et al., 2005), can exhibit non-monotonic response curves (quantitatively and qualitatively different outcomes at low versus high concentrations), and can alter development during critical windows and increase the risk of disease (Vandenberg et al., 2012; Welshons et al., 2003).

UOG extraction involves harvesting oil and natural gas reserves, including shale gas, coal bed methane, and shale oil, trapped in impermeable or low-permeability geologic layers. As such, extraction of these energy resources requires stimulation, routinely via processes such as hydraulic fracturing (high pressure injection of water, chemicals, and suspended solids), to fracture the target layer and release the trapped natural gas and/or oil (Waxman et al., 2011; Wiseman, 2008). While less than fifty chemicals are typically used for the hydraulic fracturing of a single well, there are approximately 1000 different chemicals used by industry across the US (US EPA, 2015; Waxman et al., 2011); of these, >100 are known or suspected EDCs (Colborn et al., 2011; Kassotis et al., 2014; Waxman et al., 2011). A small percentage of injected fluids are recovered as “flow back” over approximately the first two weeks, while “produced water” is then generated over the life of the producing well (Deutch et al., 2011; Engle et al., 2014). These wastewaters can be heavily laden with naturally occurring radioactive compounds, heavy metals, and other compounds from the shale layer (Akob et al., 2015; Rowan et al., 2015), as well as chemicals and compounds used and produced by fracturing operations, and are routinely injected into disposal wells, reused in future fracturing operations, and/or pumped into open evaporation pits for disposal (Deutch et al., 2011; Lee et al., 2011; Lester et al., 2015; Wiseman, 2008).

Economically feasible methods to treat and reuse hydraulic fracturing wastewater are still under development, so injection remains the major disposal method, despite concerns over associations between injection disposal wells and increased seismicity and earthquakes (Ellsworth et al., 2015; Weingarten et al., 2015). More than 95% of produced wastewater in the US is injected for final disposal (US EPA, 2015; Clark and Veil, 2009), though centralized wastewater disposal facilities handle a more significant portion of wastewater in the Marcellus Shale region specifically (US EPA, 2015; Lutz et al., 2013). Spills and/or discharges of wastewater have been shown to increase: 1) fracturing chemical concentrations in local water supplies and sediments (DiGiulio et al., 2011; Rozell and Reaven, 2012; Skalak et al., 2014), 2) heavy metals in drinking water (Fontenot et al., 2013; Jackson et al., 2013; Osborn et al., 2011), and 3) radioactivity, salinity, and total dissolved solids in rivers downstream from treatment plants and/or discharges (Harkness et al., 2015; Hladik et al., 2014; Warner et al., 2013), potentially leading to the production of disinfection byproducts (Harkness et al., 2015; Hladik et al., 2014; Parker et al., 2014). Previous work in our laboratory has reported potential human and animal health concerns via UOG contamination (Kassotis et al., 2014, 2015c; Webb et al., 2014) as well as adverse health outcomes in male C57 mice exposed during gestation to potentially environmentally-relevant concentrations of a hydraulic fracturing chemical mixture (Kassotis et al., 2015b). Because of these health concerns and the many potential contamination pathways (spills during transport to/from sites, improper handling and disposal of wastewater, failure of well casings, etc.), it is important to fill key data gaps in understanding contamination via underground injection activities and potential environmental impacts (US EPA, 2015).

As such, the goals of this study were to characterize the endocrine disrupting activities of water samples collected from a site where the chemical analyses indicated release of UOG wastewater had occurred and to ascertain potential health risks. Due to the high degree of conservation in nuclear receptor pathways (Diamanti-Kandarakis et al., 2009), in vitro screens such as reporter gene assays and yeast receptor screens are commonly used to assess potential health effects in human and wildlife populations (Naylor, 1999; Soto et al., 2006). These in vitro screens can more easily assess potential threats to human and environmental health than more costly and time-consuming animal studies, since the ability of a chemical to interfere with any aspect of hormone action is a clear indicator of potential resultant health outcomes (Zoeller et al., 2012). Mammalian reporter gene assays are often used due to high sensitivity and the translational potential of results (Naylor, 1999; Soto et al., 2006). Yeast receptor screens tend to be less sensitive, though are less susceptible to toxicity (Leusch et al., 2010). Due to these factors, we opted to couple mammalian and yeast bioassays to assess differences between the systems and to ensure that toxicity concerns would not prevent characterization of EDC activities at these sites. We further used authentic standards to convert receptor activities to equivalent concentrations of well-described control chemicals, facilitating the translation of in vitro results, as exposure to EDCs has been linked to a number of negative health outcomes in laboratory animals at environmentally relevant concentrations, wildlife and humans (Akingbemi and Hardy, 2001; Christiansen et al., 2008; Kelce and Wilson, 1997; Kidd et al., 2007; Mendiola et al., 2011; Sumpter and Jobling, 1995; Tyler et al., 1998).

The site examined herein was a West Virginia wastewater injection disposal facility that included an injection disposal well, several lined holding ponds and brine storage tanks, and a small stream that flows through the site (Fig. 1). This stream flows into the Wolf Creek downstream, and eventually into the New River, a drinking water source for local communities and important recreational area. A second tributary of Wolf Creek was identified as a background, non-impacted site, and samples were collected from both streams and assessed for agonist and antagonist activities for the estrogen (ER), androgen (AR), progesterone (PR), glucocorticoid (GR), and thyroid (TR) receptors. From our prior work with individual UOG chemicals and mixtures, we hypothesized that the disposal facility