



Emissions of organic compounds from produced water ponds I: Characteristics and speciation

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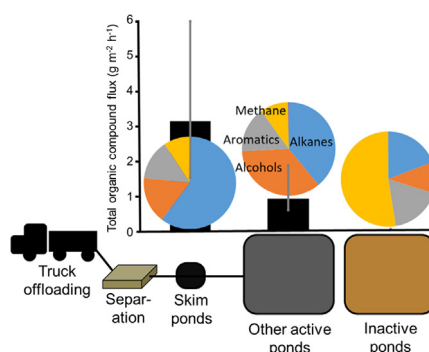
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HIGHLIGHTS

- Produced water ponds are a significant source of organics to the atmosphere.
- Prior to this work, emissions from produced water had not been adequately characterized.
- Produced water ponds are a significant source of hydrocarbons and alcohols.
- Methanol and C6–C9 alkanes and aromatics dominate fluxes from produced water ponds.

GRAPHICAL ABSTRACT



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ABSTRACT

We measured fluxes of methane, a suite of non-methane hydrocarbons (C₂–C₁₁), light alcohols, and carbon dioxide from oil and gas produced water storage and disposal ponds in Utah (Uinta Basin) and Wyoming (Upper Green River Basin) United States during 2013–2016. In this paper, we discuss the characteristics of produced water composition and air–water fluxes, with a focus on flux chamber measurements. In companion papers, we will (1) report on inverse modeling methods used to estimate emissions from produced water ponds, including comparisons with flux chamber measurements, and (2) discuss the development of mass transfer coefficients to estimate emissions and place emissions from produced water ponds in the context of all regional oil and gas-related emissions.

Alcohols (made up mostly of methanol) were the most abundant organic compound group in produced water (91% of total volatile organic concentration, with upper and lower 95% confidence levels of 89 and 93%) but accounted for only 34% (28 to 41%) of total organic compound fluxes from produced water ponds. Non-methane hydrocarbons, which are much less water-soluble than methanol and less abundant in produced water, accounted for the majority of emitted organics. C₆–C₉ alkanes and aromatics dominated hydrocarbon fluxes, perhaps because lighter hydrocarbons had already volatilized from produced water prior to its arrival in storage or disposal ponds, while heavier hydrocarbons are less water soluble and less volatile. Fluxes of formaldehyde and other carbonyls were low (1% (1 to 2%) of total organic compound flux). The speciation and

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magnitude of fluxes varied strongly across the facilities measured and with the amount of time water had been exposed to the atmosphere. The presence or absence of ice also impacted fluxes.

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1. Introduction

Emissions to the atmosphere from the oil and gas industry include the greenhouse gases methane and carbon dioxide (Brandt et al., 2014; Howarth et al., 2011; Karion et al., 2013), as well as a suite of volatile organic compounds (VOC) and nitrogen oxides (NO_x) (Edwards et al., 2014; Lyman and Tran, 2015; McDuffie et al., 2016; Prenni et al., 2016; Rappenglück et al., 2014), which can react in the atmosphere to form ozone and particulate pollution. Emissions from the oil and gas sector and their impacts on air quality and climate have been studied extensively (Allen et al., 2013; Brandt et al., 2014; Colborn et al., 2014; Hendler et al., 2009; Howarth et al., 2011; Karion et al., 2013; Warneke et al., 2014), and regional (Ahmadov et al., 2015; Bar-Ilan et al., 2006) and national (Maasackers et al., 2016) inventories exist. In many cases, however, emissions inventories underestimate emissions measured using top-down approaches that capture emissions from entire fields or basins (Ahmadov et al., 2015; Brandt et al., 2014; Karion et al., 2013). This discrepancy could be due to underestimates in the emission factors or activity data used to generate emissions inventories. It could also be due to emission sources that are excluded from current emissions inventories.

Produced water storage and disposal ponds are not included in the Western Regional Air Partnership (WRAP) oil and gas emissions inventories for the western United States (Friesen et al., 2009), which have been used in the U.S. Environmental Protection Agency National Emissions Inventory (NEI) and a number of other official and unofficial inventory efforts. Authors of oil and gas emissions inventories have cited a lack of survey responses about produced water ponds, or expected low emissions, as reasons for excluding them (Bar-Ilan et al., 2006; Friesen et al., 2009).

Almost no studies of emissions from produced water ponds have been conducted. Field et al. (2015) measured the composition of an emission plume that was influenced by a produced water disposal facility. They found high concentrations of toluene and xylenes, though the measurements they collected were off-site and could have been influenced by other sources. Thoma (2009) measured organic compound emissions from two produced water disposal facilities in Colorado over a few days. More complete emissions data from produced water ponds are needed so this source category can be included in emissions inventories and adequately accounted for in air emissions regulations.

Produced water represents a large portion of the material brought to the surface during the oil and gas extraction process. More than 5 barrels of water are produced per barrel of oil in the United States, and 182 barrels of water are produced per Mmcf of natural gas (Clark and Veil, 2009). Most produced water in the United States is injected back into the subsurface, but about 2% is disposed of in surface ponds, which allow it to evaporate into the atmosphere (Clark and Veil, 2009). In the arid western United States, the percentage of produced water disposed of by evaporation is higher. In the Uinta Basin (in north-eastern Utah; location of the majority of the measurements collected for this study), 11% (about 11 million barrels annually) is disposed of this way (Chidsey, 2015). Water is also often stored in open ponds prior to subsurface injection.

Produced water is a complex solution containing hydrocarbons and other organic and inorganic compounds (Benko and Drewes, 2008; Clark and Veil, 2009; Dórea et al., 2007; Lu et al., 2006; Neff et al., 2011; Tibbetts et al., 1992; Utvik, 1999). Because constituents of produced water vary by region, well location, treatment methods and well depth, no absolute compositional definition exists for produced water (Veil et al., 2004). Total dissolved solids (TDS) in produced

water range in concentrations from near zero to more than 75,000 ppm (average of 13,200 for the Uinta Basin), and inorganic ions tend to be dominated by Na-Cl (Zhang et al., 2009). Produced water tends to be rich in hydrocarbons, especially aromatics (Dórea et al., 2007; Field et al., 2015) and can contain high concentrations of water-soluble organics like methanol (Veil et al., 2004). Water from gas production tends to contain more light aromatics, including benzene, toluene, ethylbenzene, and xylene, than water from oil production (Benko and Drewes, 2008). Water disposed of or stored in open ponds often includes both produced water and fluid that flows back from the well to the surface after hydraulic fracturing (i.e., flow-back water).

Here we report on fluxes of a suite of organic compounds and carbon dioxide from produced water ponds at eight produced water disposal facilities in the Uinta Basin of Utah and the Upper Green River Basin of Wyoming during 2013–2016. This paper focuses on produced water composition and on the speciation, magnitude, and variability of fluxes from produced water to the atmosphere, using flux chamber measurements. Companion papers will discuss (1) methods and results from inverse modeling estimates of emissions from produced water ponds, including comparisons with flux chamber results (Tran et al., 2017), and (2) analysis of mass-transfer coefficients for compounds in produced water and development of facility-level and basin-level emission estimates (Mansfield et al., 2017). This study included measurements from ponds only and did not consider emissions from tank storage, transport, or processing of produced water.

2. Experimental

2.1. Study locations

The produced water disposal facilities sampled for this study were located in the Uinta Basin, Utah, and the Upper Green River Basin, Wyoming, in the United States. While the majority of produced water is injected into the subsurface without spending time in open ponds, the facilities sampled in this study all employed ponds, either for storage of water prior to injection or for evaporative disposal. Access was granted to some of the facilities on condition of anonymity, so facility names and other identifying information are not disclosed. Table S1 provides information about the number and types of measurements that occurred at each facility.

The produced water disposal facilities sampled in this study functioned as follows: (1) Produced water was separated from hydrocarbon liquids and natural gas by gravity and collected in a storage tank at an oil or gas well site. In some cases, water was piped directly to the disposal facility, rather than being stored in a tank. (2) Produced water was trucked or piped to a disposal facility, where it was released into a closed tank or vessel for additional gravity-based separation of water from oil. Some facilities employed more sophisticated techniques to separate water from oil (e.g., centrifugal separation), while some did not use any closed-tank separation. Water in well-site storage tanks was usually heated during cooler seasons, so water was usually warm when it arrived at disposal facilities (sometimes exceeding 40 °C). (3) Water was transferred from separation tanks into open ponds. Ponds varied from less than 0.1 to several hectares in size. (4) Once in a pond, water was often transferred to additional ponds. Facilities we studied had from two to ten individual ponds. Most facilities utilized a small netted pond (often called a skim pond) downstream from the separation tank to catch additional residual oil before transfer to larger evaporation or storage ponds. (5) For some facilities, water was injected into the subsurface after storage in open ponds. For others, water

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