

HOLISTIC RISK ASSESSMENT OF SURFACE WATER CONTAMINATION BY NATURALLY
OCCURRING RADIOACTIVE MATERIAL IN OIL PRODUCED WATER FROM THE BAKKEN SHALE

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MASTER OF SCIENCE

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ABSTRACT

The risks to the environment and human health due to hydraulic fracturing (HF) in onshore unconventional oil and gas (O&G) development have been studied in the past but results are inconclusive. A common shortcoming in previous studies is the absence of social risk perception and awareness analysis. This thesis research proposes the combination of statistical methods to analyze risks to human health due to improper management of produced water, the major by-product of HF. This study focuses on the Bakken Shale located in North Dakota. A risk assessment of radium-226 was performed from a technical perspective only. A second assessment, focused on lead-210, combined technical analysis with risk perception and awareness of ND residents. Results indicate that the latter offers more holistic information that could greatly contribute to the mitigation of risks in O&G development by creation and implementation of standards and regulations that consider technical and social aspects.

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DEDICATION

To my beloved parents Rodolfo and Edna. Without you none of my successes would have been possible.

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LIST OF ABBREVIATIONS

AWWA	American Water Works Association
Ba	Barium
BORA.....	Barrier and operational risk analysis
Bq	Becquerel
Ca	Calcium
DoD.....	Department of Defense
EIA.....	U.S. Energy Information Administration
EPA	U.S. Environmental Protection Agency
ERA	Environmental (ecological) risk assessment
ETA	Event tree analysis
ETA/DTA	Event/Decision tree analysis
FEP	Features, events or processes
FMEA	Failure modes, effects, and analysis
FTA	Fault tree analysis
FTA/LTA.....	Fault/Logic tree analysis
GHG	Greenhouse gases
GIS	Geographical information system
HAP	Hazardous air pollutants
HAZID	Hazard identification
HAZOP.....	Hazard and operability studies
HF.....	Hydraulic fracturing
HOF.....	Human and organizational factors
IAM	Integrated assessment model
MCL.....	Maximum contaminate level
mSv	Millisieverts
ND	North Dakota

TENORM.....Technologically enhanced naturally occurring
radioactive material

Th-232Thorium-232

TWATotal weight average

U.K.....United Kingdom

U-238Uranium-238

United States.....U.S.

USDOE.....U.S. Department of Energy

USGS.....United States Geological Survey

VFAVolatile fatty acids

WHOWorld Health Organization

O&GOil and gas

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1. INTRODUCTION

1.1. Background

Energy development in the United States (U.S.) has changed over the last decade. The country used to depend on foreign oil and gas (O&G) deposits but now, using unconventional methods, it is one of the top producers in the world. Deposits of O&G in very impermeable rocks such as shale and tight sands are considered unconventional as compared to conventional deposits such as sand and carbonates. The production of O&G from unconventional reservoirs requires a combination of two techniques, horizontal drilling and hydraulic fracturing (HF). With these techniques combined, producers are able to cover larger areas and stimulate more production of O&G using large amounts of water (up to 49,200 m³) mixed with chemicals (~1% of the total volume) and proppant (~5% of total volume).

The purpose of applying large amounts of water and additives at high pressure inside the O&G wellbore is to break the formation rock, allowing the release of O&G which flow up to the surface where they are collected. Along with the O&G, water returns to the surface as produced water which is a combination of injected fluid and indigenous formation water. The latter is water naturally present in the reservoir with geological characteristics specific to the location. Produced water requires special management due to high total dissolved solids (TDS), naturally occurring radioactive material (NORM) (mostly radium), other inorganic elements, and organic elements.

The improper management of water throughout the process of O&G production could affect the environment and consequently human health. Some of these consequences are water shortage, spills of chemicals, migration of produced water into groundwater reservoirs, and produced water spills and leaks. The impacts of these events have been studied around the globe where O&G production, both onshore and offshore, take place.

Most of the studies on the impacts of O&G development have focused on the technical aspects only (e.g. mechanical and software failures). This technical or engineering approach

focuses on defining risks based on probabilities or expected values and measuring the uncertainties involved. The results from this type of risk assessment only show one side of the issue leaving aside social aspects including human errors and impacts to society. In addition, despite being a well-known and established technology, HF and its impacts on the environment and human health are still a controversial subject and information is still scarce. Many stakeholders, from laypeople to politicians and environmental groups, are involved in the debate. This has resulted in a divided population between proponents and opponents of unconventional O&G development. Nevertheless, it is imperative to assess from an objective and holistic approach the risks involved in the O&G industry.

1.2. Research problem statement

The expansion of unconventional O&G development using HF has resulted in multiple studies. Although these studies have become more available, the information on risks related to the water and produced water management in unconventional O&G development and the impacts in the U.S. is still insufficient. There are evident gaps in data on NORM in produced water and the impacts on the environment and human health. Data on NORM, especially radium 226 (Ra-226) the most common radionuclide present in unconventional O&G produced water, has been collected in different States such as Pennsylvania and used to create standards and regulations. However, in North Dakota (ND) which is the second largest producer of unconventional oil in the country, research on Ra-226 in produced water from the Bakken Shale is extremely scarce. The lack of data is even more evident for other radionuclides including Ra-226's decay product, lead 210 (Pb-210). Studies on Pb-210 in produced water have not been conducted in ND despite its known harmful health effects, especially on children, mobility and relatively long life in the environment, and the large volumes of produced water generated annually in the State.

Moreover, the studies available to the public are typically focused on other locations and references to ND are limited. These studies were conducted mostly from an engineering

approach, the preferred method to assess risks. Applying only this approach results in an incomplete analysis of the risks under review. Thus, scarce data and narrow view of the risks to the environment and human health merits the improvement of current techniques used to study and measure the real risks involved in unconventional O&G development. This improvement includes conducting risk assessment from a social perspective. To date, no research has been conducted to understand public risk perception of hazards associated with produced water from HF in ND.

1.3. Research objectives

The objectives of this thesis research are:

1. To review different risk assessment techniques and select the most applicable ones to onshore unconventional O&G development.
2. To conduct a risk assessment from an engineering perspective on human exposure to Ra-226 in produced water from the Bakken Shale.
3. To develop a holistic risk assessment method by incorporating social awareness and perception with risk characterization of surface water contamination due to Pb-210 in produced water from the Bakken Shale.
4. To understand the factors that shape social risk perception and awareness of produced water from different stakeholders in ND.

1.4. Thesis organization

The introduction of this thesis (Chapter 1) includes the sections: background, research problem statement, objectives, and thesis organization. In Chapter 2, different risk assessment techniques applicable to study onshore unconventional O&G production and the risks to water quantity and quality associated with HF and produced water management are reviewed and summarized. Constraints in performing risk assessment are identified including gaps in databases. Each risk associated with water and produced water management and mitigation strategies are discussed.

Chapter 3 presents a study on the risk assessment of human exposure to Ra-226 in produced water spills via food and drinking water consumption. The assessment only considers Ra-226 because it is the most predominant radionuclide in produced water. A multivariate regression model was developed to predict Ra-226 in produced water which then was further analyzed by assessing the human exposure to the radionuclide in the event a produced water spill reaches a surface water body. Using food transfer factors found in the literature, the annual effective dose rate of Ra-226 for an adult in ND was estimated for three different scenarios. With this, the total annual effective dose was calculated and used to determine the risk of human exposure to Ra-226 in produced water.

Chapter 4 presents a holistic risk assessment method developed to characterize the risks of surface water contamination due to Pb-210 in oil produced water from the Bakken Shale. Because of limited data on Pb-210, a simulation model was developed to determine its concentration based on its parent Ra-226 and historical TDS levels in produced water. Scenarios where produced water spills could reach surface water were analyzed by applying the typical four steps of the risk assessment process. The scenarios evaluated are: (1) storage tank overflow, (2) leakage in equipment, and (3) spills related to trucks used to transport produced water. Risk perception and awareness of produced water from different stakeholders, which are based on a survey described in Chapter 5, were incorporated into the assessment.

Chapter 5 presents a survey that collected data on risk perceptions and awareness of produced water from ND residents. This chapter focuses on presenting and analyzing the results of the survey to understand the risk perception of select ND stakeholder groups regarding produced water management and NORM. The software Qualtrics was used to create an online survey, collect data, and perform statistical analysis. Finally, Chapter 6 presents conclusions and recommends future work.

2. LITERATURE REVIEW

2.1. Introduction

O&G resources can be classified as conventional or unconventional depending on the geological formation. Conventional deposits, sand and carbonates such as limestone, have high permeability that allows the fluids (O&G) to flow into the wellbores (Freyman, 2014; Scanlon et al., 2014; USDOE, 2013). Unconventional O&G deposits are trapped inside rocks such as shale and tight sands which have high porosity and limited permeability (Freyman, 2014; Scanlon et al., 2014). These characteristics make production difficult, requiring stimulation to allow O&G to flow to the wellbore at an acceptable rate (Scanlon et al., 2014). The technologies of horizontal drilling and high-volume HF have been combined to achieve the flow of hydrocarbons resulting in recent growth in onshore unconventional O&G development. In the U.S. from 2011 to 2013, 95% of oil production growth and 100% of natural gas production growth came from the Bakken, Niobrara, Marcellus, Utica, Permian, Haynesville and Eagle Ford (Figure 2.1) (EIA, 2014).

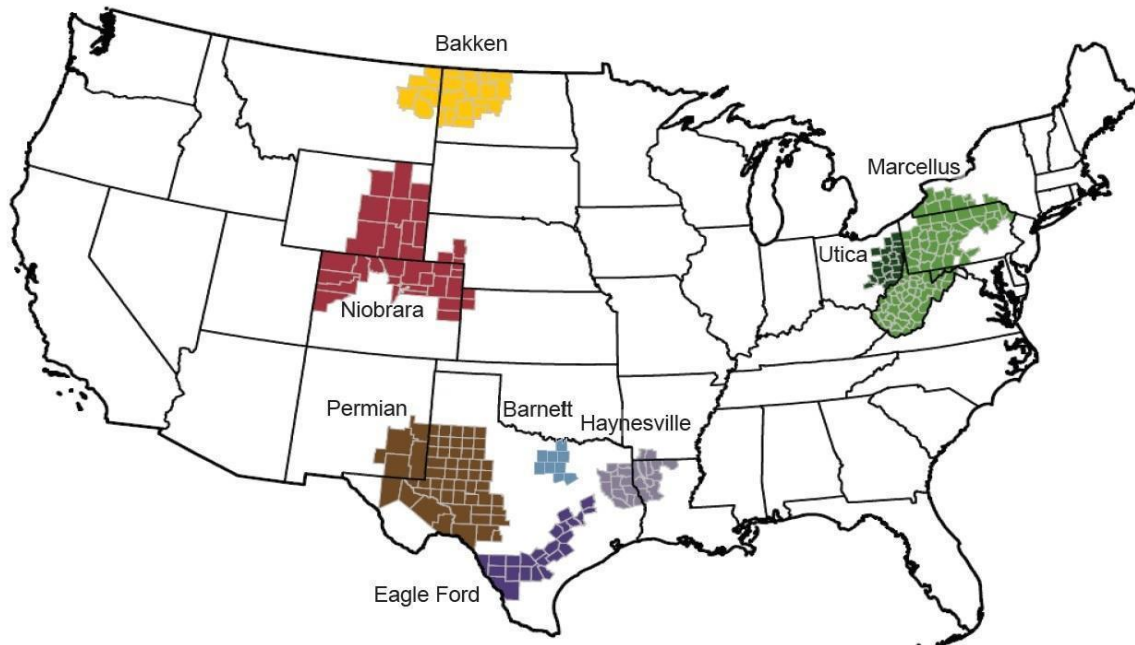


Figure 2.1. Most important unconventional O&G regions in the U.S. Adapted from EIA (2014).

This chapter focuses on the following four shale formations due to their contribution to the total unconventional O&G produced in the U.S.: Bakken (ND), Barnett (Texas), Eagle Ford (Texas), and Marcellus (Pennsylvania). The Eagle Ford ranked first in unconventional oil production in 2013 while the Bakken ranked second; together representing 67% of total unconventional oil production in the U.S. in 2013 (Scanlon et al., 2014). Shale gas development was first assessed in the Barnett, which was also the first shale in the world to be fully developed using HF (Nicot et al., 2014; USDOE, 2014). In 2013, Pennsylvania produced more than 3 thousand cubic feet of natural gas, mostly from the Marcellus Shale, making it the largest gas play in the U.S. (Pennsylvania Department of Environmental Protection, 2013).

Onshore unconventional O&G development and production have boosted the U.S. economy, but along with this benefit environmental risks have emerged, similar to any other large volume extraction of underground resources. Some of the arguments by shale gas proponents are clean energy, future energy independence, economic benefits and jobs creation, reduction in greenhouse gases (GHG) emissions, and modest environmental risks (Stern et al., 2014). On the other hand, the opponents argue that safeguards and monitoring are not adequate, operations present significant risks, there are impacts to the environment, human health, and society, GHG emissions could increase due to methane escape, and dependence on shale O&G is a step back from progress towards renewable energy (Stern et al., 2014). In addition, regulations in different States show inconsistencies and there are not enough staff and expertise to track, coordinate and prevent risks (Stern et al., 2014; Wiseman, 2014).

Unconventional O&G development using HF requires millions of cubic meters of water and chemicals, some of which are known to affect human health, and contaminate air and water (EPA, 2012). This could lead to impacts on water availability, human health, agriculture, livestock, and wildlife. This literature review focuses only on risks to water quantity and

quality. Although studies on this subject have become more available, the information on risks related to the water and produced water management in unconventional O&G development and the impacts is still insufficient. Consequently, risk assessment in unconventional O&G merits further investigation.

The results from further research could contribute to practices in other countries that have unconventional O&G resources. The top 5 countries with technically recoverable shale oil resources are Russia, U.S., China, Argentina, and Libya while the top 5 countries with shale gas resources are China, Argentina, Algeria, U.S., and Canada (EIA, 2013). Some of these countries are still assessing the feasibility of unconventional O&G production but application of HF is likely to occur in the future.

The review begins with explaining the basics of unconventional O&G development process. The different stages in the water life cycle throughout the development process are described, including the possible risks to water quantity and quality. Risk assessment techniques applicable to unconventional oil and gas are discussed. Finally, results from these assessments are reviewed to determine what is missing.

2.2. Unconventional O&G development process

The unconventional O&G development process (Figure 2.2) begins with planning for the water sources, the amount of water needed, and proper produced water management (American Petroleum Institute, 2010). Water is obtained from the source and transported to the well site, by pipeline or truck, where it is stored before chemical mixing. The next step is drilling which requires drilling mud; typically, a mixture of water, mud, and drilling additives (Lutz et al., 2013). During drilling, the well casing made of steel pipes is installed using cement to isolate all formations that contain water, oil, gas, coal or a combination (American Petroleum Institute, 2010; ND Century Code, 2012). Once the well is constructed, the HF process begins with mixing the water with additives. Using pumping trucks, the HF fluid is injected into the well at high enough pressure to fracture the formation rock to enable the

release of O&G. Prior to production, the flowback process begins which is designed to capture the initial production that contains a high percentage of produced water, a mixture of injected and formation water stored in the shale (EPA, 2012). Once the flowback decreases gradually, the transition to formation water and hydrocarbons occurs (Water Environment Federation, 2013). During the production phase the O&G enters the wellbore and then is collected at the surface while the formation water is released during the lifespan of the well (Nicot et al., 2014; Water Environment Federation, 2013). Both the injected and formation water (produced water) are recovered and then subjected to one of the following three options: 1) disposal, 2) treatment and reuse; or 3) treatment and disposal. The HF process is then repeated if needed to continue stimulating the O&G production until the well is no longer productive (American Petroleum Institute, 2010). Once this happens, the well is plugged or isolated with cement barriers before abandonment (American Petroleum Institute, 2010).

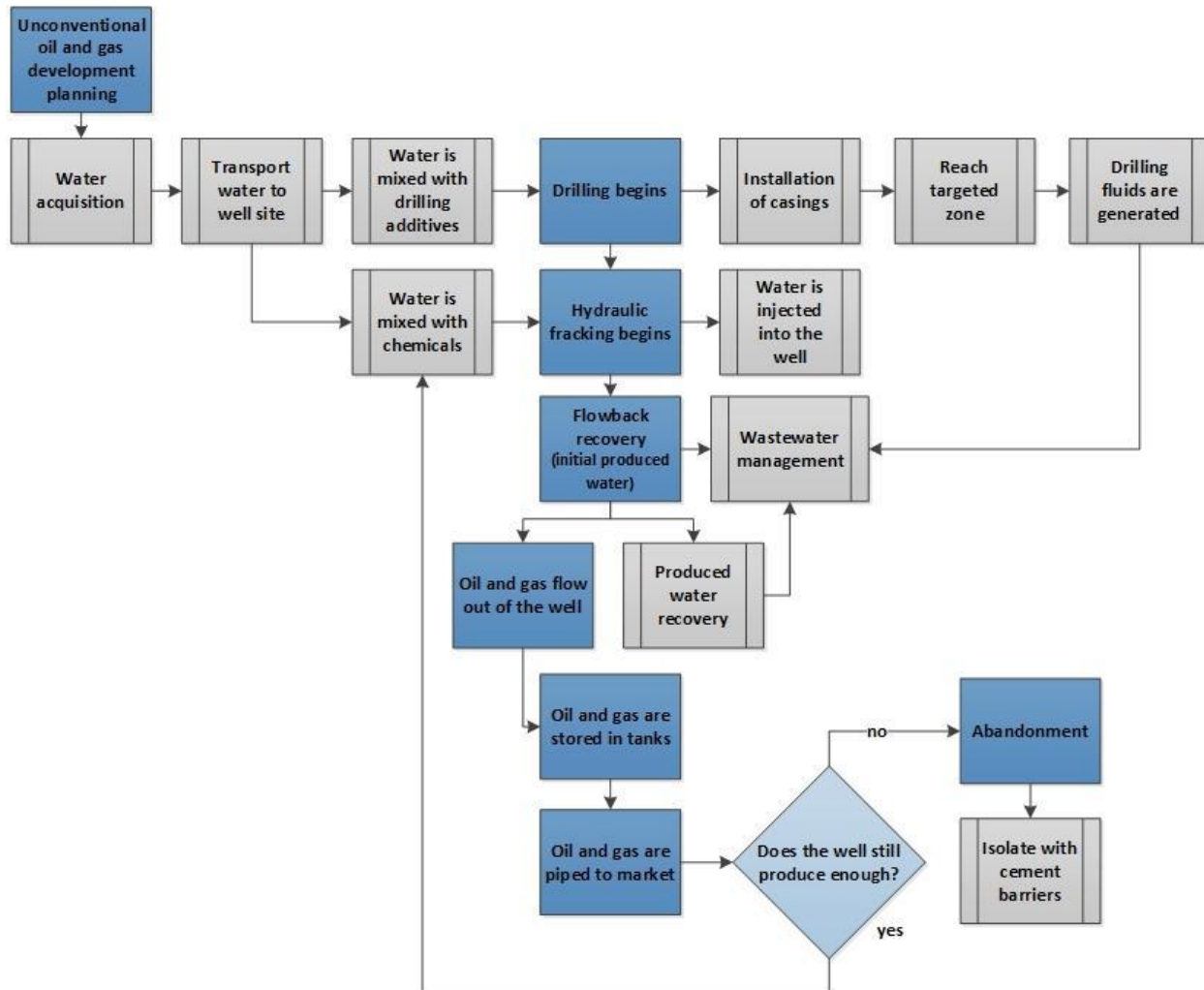


Figure 2.2. General process of unconventional oil and gas production.

2.3. Water lifecycle in unconventional O&G production

There are five major stages for the water life cycle associated with unconventional O&G development (EPA, 2012). These stages are: 1) water acquisition, 2) chemical mixing, 3) well injection (HF), 4) flowback process and produced water generation, and 5) treatment and disposal (EPA, 2012). Possible risks in each stage of the water life cycle are shown in Table 2.1.

Table 2.1. Possible risks on the different water life cycle stages.

Stage	Possible risks
Water acquisition	Water shortage or limited access for other users' needs causing stress on water resources particularly during drought season.
Chemical mixing	Spills of chemicals could cause surface water and/or groundwater contamination. Health problems due to chemical exposure.
Well injection	Casing failure or induced fractures in the rocks could serve as pathway for HF fluid migration into water resources.
Flowback process and produced water	Surface spills, infiltration in the ground from the reserve pits or tanks, leaks from pipes, and effects on human health due to exposure to the chemicals, brine and other natural radioactive material.
Produced water treatment and waste disposal	Spills and leakage during on-site treatment, storage and transportation to off-site treatment facilities or disposal. Limitation of the treatment plants to completely eliminate contaminants which reach streams and impair drinking water sources. Deep-well injection could induce earthquakes and cause well casing failure.

2.3.1. Water acquisition

The amount of water used per well varies from 7,600 to 15,200 m³ and up to 49,200 m³, depending on the geological characteristics, well construction (depth and length), and fracturing operations (chemicals used and fracture stimulation design) (American Petroleum Institute, 2010; EPA, 2012; Freyman, 2014). Of the total water, 10% is used during drilling, 89% for HF, and the rest is consumed by infrastructure (USDOE, 2014). In ND's Bakken Shale, it is estimated that each well requires ~8,700 m³ of water for drilling and HF (ND State Water Commission, 2014). In Texas, for the Barnett Shale the estimation is 18,900 m³/well while in the Eagle Ford is 18,200 m³/well (Nicot et al., 2014; Scanlon et al., 2014). In

Pennsylvania, the Marcellus Shale requires ~11,400 to 18,900 m³/well (Lutz et al., 2013). From January 2011 to May 2013, there were a total of 39,294 shale oil and gas wells across the U.S. which equals to 5.95×10^8 m³ of water (assuming 15,150 m³/well) or the water consumed by 3 million Texans in a year (Environment Texas Research and Policy Center, 2013; Freyman, 2014).

Sources of water for onshore unconventional O&G production vary by region. The main source of water in the Bakken shale is the Missouri River although groundwater is sometimes used where access to the river is restricted (ND State Water Commission, 2014). In 2012, from the 4.35×10^8 m³ of water consumed in ND, 1.74×10^7 m³ (or 4%) were used for fracturing purposes (ND State Water Commission, 2014). In the Barnett shale, operators depend on both groundwater and surface water, relying on different aquifers such as Ogallala and Carrizo-Wilcox and the Brazos River basin (Freyman, 2014; Nicot et al., 2014). In the Eagle Ford, 90% of the new oil wells use groundwater (RRC of Texas, 2013). The amount of water used in the Barnett for HF purposes was 3.18×10^7 m³ in 2011 while the Eagle Ford used 6.74×10^7 m³ in 2013 (Nicot et al., 2014; Scanlon et al., 2014). In Pennsylvania, the Marcellus shale wells use surface water from the Susquehanna and Delaware River basins but in recent years the operators have switched to reusing and recycling, reaching almost 90% of the wastewater in 2012 (Pennsylvania Department of Environmental Protection, 2014). Mining, where HF is included, accounts for 2% of the total water withdrawals in the State which withdraws 3.67×10^7 m³ of water every day or ~13.4 billion cubic meter per year (Pennsylvania Department of Environmental Protection, 2009).

2.3.2. Chemical mixing

The fracturing fluid is composed of water (~94%), proppant (~5%) and other chemical additives (~1%) (American Petroleum Institute, 2010; EPA, 2012; Halliburton, 2015). The mixture varies according to the well location and operator but a typical combination requires 3 to 12 chemical additives (FracFocus, 2014a). Sand is commonly used as the proppant to

help keep the fractures open and release the hydrocarbons (O&G). Other options for proppant are resin coated sand, intermediate strength proppant ceramics, and high strength proppants such as sintered bauxite and zirconium oxide (Arthur et al., 2008). In addition, a generic formula for the additives is acid, acid/corrosion inhibitor, biocide, breaker, clay and shale stabilization/control, crosslinker, friction reducer, gel, iron control, non-emulsifier, pH adjusting agent, scale inhibitor and surfactant (FracFocus, 2014b). The concentrations of some of these additives are detailed in Table 2.2 (Chesapeake Energy Corporation, 2011a). According to the Environmental Protection Agency - EPA (2012), there are seven chemicals often used in the mixture appearing in over 2,500 products reported by 14 operators between 2005 and 2009. These chemicals are methanol, isopropanol, crystalline silica, 2-butoxyethanol, ethylene glycol, hydrotreated light petroleum distillates, and sodium hydroxide (EPA, 2012). The concentrated additives are mixed with water and proppant using blender trucks (Figure 2.3). Then, the fracturing fluid is transferred to pumping trucks which inject the fluid into the well (American Petroleum Institute, 2010; EPA, 2012).

2.3.3. Well injection (HF)

The fracturing fluid is injected inside the well using pumping trucks at high pressures to break the formation rock, allowing the release of O&G which flow through the wellbore up to the surface where they are collected (EPA, 2012). Depending on the well depth it may require several injections or stages (EPA, 2011a). The depth where HF takes place depends on the geological formation usually being thousands of feet away from groundwater resources (AWWA, 2013). In ND, potable water is located at 610 m deep while the oil-bearing formations are generally at 3,050 m under the surface (ND State Water Commission, 2014). In Texas, the Eagle Ford wells have an average depth of 2,750 m while the Barnett Shale is located at 2,300 m from the surface and groundwater is found at 370 m (Chesapeake Energy Corporation, 2011a; Scanlon et al., 2014; USDOE, 2014). The well depth on the Marcellus

Shale range from 1,520 to 2,440 m while groundwater is found at 260 m from the surface (EPA, 2012; USDOE, 2014).

Table 2.2. Common groups of chemical additives in the HF fluid and associated chemical compounds and concentrations.

Additive	Chemical	Concentration
Friction reducer	Polyacrylamides	500 - 1,000 ppm (parts per million) (0.05-0.1% of total fluid)
Biocide	Glutaraldehyde, glutaraldehyde/ quaternary amine blends, tetrakis hydroxymethyl phosphonium sulfate, 2,2- dibromo, 3-nitrilopropionamide, and sodium hypochlorite	75 - 500 ppm (0.075 - 0.05% of total fluid)
Scale inhibitor	Polymers (carboxylic acid and acrylic acid)	75 - 120 ppm (0.075% - 0.12% of total fluid)
Substitute	Potassium chloride	500 - 2,000 ppm (0.05% - 0.2% of total fluid)
Surfactant	Laurel sulfates, and fluoro and nano-surfactants	500 - 1,000 ppm (0.05% - 0.1% of total fluid)
Dissolvent	Hydrochloric acid	0.08% - 2.1% of total (as acid volume). 0.012% - 0.31% of total (as active acid)

Table 2.2. Common groups of chemical additives in the HF fluid and associated chemical compounds and concentrations (continued).

Additive	Chemical	Concentration
Acid corrosion inhibitor	Formic acid, amines, amides, and amido-amines	2,000 - 5,000 ppm of acid volume 0.0004% - 0.0043% of total (temperature and time dependent)
Iron control	Citric acid, acetic acid, thioglycolic acid, and ethylenediamine tetraacetic acid	5,000 ppm of acid volume 0.0004% - 0.011% of total fluid

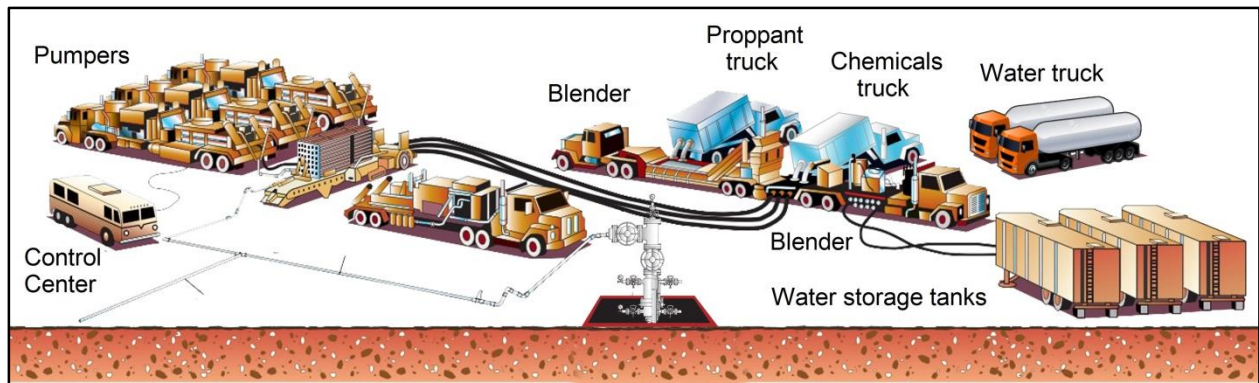


Figure 2.3. HF units used to stimulate O&G production. Adapted from American Petroleum Institute (2010).

2.3.4. Flowback process and produced water

After the fracturing fluid is injected, the pressure is reduced allowing the fluid to come back to the surface. This fluid is called produced water and is composed of the injected fluid and the formation water. Flowback process water is a subset of produced water and is defined by the time period in which it returns. The contents in produced water of major concern are salt, oil and grease, natural organic and inorganic compounds, chemical additives, and natural radioactive materials from the shale formation (National Petroleum Council, 2011). The EPA

has identified over 1,000 chemicals that are reported to be used in fracturing fluids or found in produced water (EPA, 2012). The content of produced water, as well as the amount, varies for every well depending on the formation that is being stimulated.

The fraction of the water volume injected that returns to the surface during the first 10 days is between 10% to 25% for the Barnett and Marcellus Shale (Chesapeake Energy Corporation, 2011b; Wilson and VanBriesen, 2012). The Bakken Shale returns 15 to 40% of the water volume injected during the initial flowback, which is considered relatively high (Boschee, 2015; Energy and Environmental Research Center, 2010). On the other hand, in the Eagle Ford less than 15% of the volume injected returns immediately to the surface (Boschee, 2015; Maguire-Boyle and Barron, 2014). Some of the causes for which the remaining water, called residual treatment water, does not return to the surface include the fluid being trapped inside the shale matrix due to capillary and osmotic forces, formation pressure decrease, and fracturing fluid traveling beyond the capture zone (Engelder et al., 2014; EPA, 2004).

The content of produced water is relatively high in total dissolved solids (TDS), which are tied to salinity. The TDS ranges are up to 632,689 mg/L in the Bakken, up to 476,500 mg/L in the Marcellus, 21,581–300,155 mg/L in the Barnett, and 1,033–317,876 mg/L in the Eagle Ford (Blondes et al., 2014). Common elements in produced water are sodium, calcium, magnesium, chloride, potassium, iron, strontium, barium, lithium, and silicon (Maguire-Boyle and Barron, 2014; Wilson and VanBriesen, 2012). In addition, NORM, mostly radium isotopes, have been detected on soil near road spreading (Skalak et al., 2014), spill sites related to HF activities (Warner et al., 2013b), soil and sludge from reserve pits (Rich and Crosby, 2013), and soil and pipe-scale at oil production sites (Zielinski et al., 2001).

It is important to note that some of the elements found in produced water have different charges (ions) and depending on the electrical charge some can be more harmful to humans than others (American Chemical Society, 2006). For example, heavy metals such as

lead with double positive charge (Pb^{2+}) are of great concern if they reach drinking water because of their toxic effects in the human body (American Chemical Society, 2006). Similarly, radionuclides that are part of the NORM in produced water undergo radioactive decay which releases radioactive particles and depending on the particle, this radiation can be harmful to humans. The three types of radiation emitted are alpha, beta, and gamma from which alpha is the most dangerous if it is inside the body because it can be absorbed by cells (King, 2016). On the other hand, beta and gamma radiation are the most dangerous if the source is outside the body because they can penetrate the skin (King, 2016). This is relevant for produced water management because radium-226, the most predominant radionuclide in this type of waste, emits mostly alpha particles and lead-210 (a decay product of radium-226) emits beta particles (American Chemical Society, 2006).

The EPA requires the operators to apply reduced emissions completions, also known as reduced flaring completions or green completions, to separate gas from solids and liquids during the initial high-rate flowback process and production (EPA, 2011a). This is done to reduce gas emissions to the atmosphere and the need for flaring (EPA, 2011a). According to the EPA (2014), after HF there are three stages of fluid handling namely initial flowback stage, separation flowback stage, and production stage. During the initial stage the flowback is stored in vessels, any gas can be vented or flared, and the hydrocarbons present are filtered out and sold (EPA, 2011a). The initial stage then shifts to separation stage when sufficient gas is present for a separator to operate. During the separation stage, the flowback is routed to the equipment that separates solids, gas, liquid hydrocarbons and water (EPA, 2014). These separations can be done using two, three or four phase separation hydrocyclones (Ditria and Hoyack, 1994; Manning and Thompson, 1995). Once the flowback process is completed the production stage begins where phase separation is also applied and liquids, including produced water, are stored and the gas or oil recovered is routed to flow line or collection system (EPA, 2014).

2.3.5. Produced water treatment and waste disposal

The separated produced water is managed using one of the following techniques: evaporation ponds, deep-well injection, on-site treatment, off-site treatment, and centralized treatment plants. In ND, evaporation ponds are not used for wastewater disposal; instead, the brine is stored in tanks before underground injection (ND Century Code, 2012). In 2012, the Industrial Commission of ND modified the oil industry regulations with the purpose of reducing the use of evaporation ponds including the prohibition of open pits to store liquids left over from the drilling process for oil wells drilled below 1,520 m and reclamation of the pit within one year after completing the well (Industrial Commission of ND, 2012; ND Department of Mineral Resources, 2011). In Texas, collecting pits for produced water storage are allowed before disposal and evaporation ponds; however, the practice of the latter varies throughout the State (RRC of Texas, 2015a; Texas Administrative Code, 1977). In the west, evaporation before closing the reserve pit is allowed because the rates of evaporation are favorable compared to the east where pits are generally dewatered due to low evaporation rates (RRC of Texas, 2015a). The on-site treatment, which is a mobile solution to the wastewater at the wellhead without the need of storage units, is applied in less frequency in the Barnett (Easton, 2013; Nicot et al., 2014).

In Pennsylvania, the use of impoundments has been banned (Easton, 2013). Another method is deep-well injection of wastewater in class II wells, which are exclusive for fluids associated with the O&G industry (EPA, 2012). This method is used the most in ND and Texas while in Pennsylvania it is used in less frequency due to lack of sufficient class II wells (Nicot et al., 2014). Because of this, Pennsylvania has to send the wastewater to Ohio where more injection wells are available (Detrow, 2012). In 2009-2010, of the total produced water reported in the Marcellus Shale, 77.5% was treated in industrial wastewater treatment plants, 16% was reused in other wells, 5% was treated in municipal wastewater facilities, 1% was classified as unknown disposal, 0.5% was disposed in deep-wells, and 0.007% was spread on

roads (Rozell and Reaven, 2012). In 2011, the Pennsylvania Department of Environmental Protection requested companies to reduce disposal through wastewater treatment plants as an effort to protect surface waters (Ferrar et al., 2013). As a result, between 2012 and 2014 the amount of the produced water disposed through municipal wastewater facilities was almost 0%, more than 69% was reused, and the rest was disposed mostly through industrial treatment facilities and deep-well injection (Pennsylvania Department of Environmental Protection, 2014).

2.4. Risk assessment

2.4.1. Risk assessment methods used in industries

Risk analysis is used for characterizing, managing, and informing others about an identified risk. The risk, or potential loss, is related to the probability of exposure to a hazardous event and the consequences of it (Modarres, 2006). In addition, risk is the combination of possible consequences and associated uncertainties (Aven et al., 2007). According to the Food and Drug Administration, risk analysis has three elements: risk assessment, risk management and risk communication which interact with each other (Fjeld et al., 2007). The process of characterizing the potential risks and its magnitude, through quantitative estimates and qualitative expressions, is called risk assessment (Fjeld et al., 2007; Modarres, 2006; National Research Council, 1983). Risk assessment uses deterministic or stochastic processes to characterize risks. Deterministic processes can be quantitative, qualitative or hybrid while stochastic processes are based on classic statistical approach and the accident or abnormal action forecasting methods (Marhavilas and Koulouriotis, 2012a, 2012b). The classification of major risk assessment processes for both deterministic and stochastic approaches are shown in Table 2.3.

Table 2.3. Risk assessment processes and techniques.

Process		Technique	
Deterministic	Qualitative	<ul style="list-style-type: none"> • Check lists • What if analysis • Safety audits • Task analysis 	<ul style="list-style-type: none"> • STEP technique • Hazard and operability studies (HAZOP)
	Quantitative	<ul style="list-style-type: none"> • Proportional risk assessment technique • Decision matrix risk assessment • Risk measures of societal risk • Quantitative risk assessment (QRA) • Quantitative assessment of domino scenarios • Weighted risk analysis 	
	Hybrid	<ul style="list-style-type: none"> • Human error analysis techniques • Fault tree analysis (FTA) • Event tree analysis (ETA) • Risk-based maintenance 	
Stochastic	Classic statistical approach	<ul style="list-style-type: none"> • Epistemic models (predictive epistemic approach) • Probability distributions (e.g. Exponential and normal) • Event data-models (e.g. Rate model, time and risk model, and poisson model) 	
	Accident forecasting modeling	<ul style="list-style-type: none"> • Time-series • Markov chain analysis • Grey model • Scenario analysis 	<ul style="list-style-type: none"> • Regression method • Neural networks • Bayesian networks

There are many methods and techniques used in risk assessment which can be combined or customized to fit the specific needs of the industry. Nuclear and aerospace are two major industries that have contributed significantly in developing very robust risk assessment methods and techniques over the years. Table 2.4, although not a comprehensive list, provides examples of risk assessment methods and techniques used in different industries.

Table 2.4. Risk assessment methods and techniques used in different industries.

Method	Industries	Techniques
Major hazard risk assessment (MHRA) (Iannacchione et al., 2008)	Minerals	<ul style="list-style-type: none"> • Workplace risk assessment and control • The preliminary hazard analysis • Failure modes, effects and analysis (FMEA) • Fault/Logic tree analysis (FTA/LTA), event/decision tree analysis (ETA/DTA) and/or logic gate • Bow tie analysis – qualitative • Work process flow chart • Exposure and risk
HAZOP (Dunjó et al., 2010; Iannacchione et al., 2008)	Chemical, pharmaceutical, petroleum, and petrochemical	<ul style="list-style-type: none"> • Systematic brainstorming (guide words) • IEC Standard 61882 • Process flow diagrams (PFDs) • Piping and instrumentation diagrams (P&IDs)

Table 2.4. Risk assessment methods and techniques used in different industries (continued).

Method	Industries	Techniques
Standard Practice for System Safety (DoD, 2000; U.S. Army Environmental Center, 2001)	Defense	<ul style="list-style-type: none"> • Systematic hazard analysis process • Iterative process ends when residual risk is at an acceptable level • Programmatic ESOH evaluation • Department of Defense (DoD) Standards (5000.1, 5000.2, 5000.2-R) • Hazardous material management program • Risk matrix • Monte Carlo simulation • Decision analysis • Sensitivity analysis
Probabilistic risk assessment (PRA) (National Aeronautics and Space Administration, 2011)	Nuclear and aerospace	<ul style="list-style-type: none"> • Master logic diagrams • Block diagram and event sequence diagram • FTA and ETA • Exponential distribution based on constant failure rate • Lognormal distribution • Sampling process (e.g., Monte Carlo) • Bayes' Theorem • Human reliability analysis

The MHRA was developed in Australia specifically to evaluate mining operations by combining informal, basic-formal, and advanced-formal techniques (Iannacchione et al., 2008). In addition, MHRA uses several techniques shown in Table 2.4. The use of different

techniques at every stage of the process allows evaluating more data and obtaining more complete results. Another method is the HAZOP which was developed in the United Kingdom (U.K.) in the 1960's for risk assessment in the chemical industry. It is a team-based systematic approach that uses a brainstorming process to identify the potential ways a process can deviate from its safe operating conditions (Kolluru et al., 1996). The disadvantages of HAZOP are high resource requirements and incomplete analysis for large events (Kolluru et al., 1996).

The Standard Practice for System Safety is used by all departments and agencies from the DoD to address environmental, safety, and health risks that could result in the development, test, production, use, and disposal of DoD systems, subsystems, equipment, and facilities (DoD, 2000). PRA plays an important role in decision-making related to designs, manufacture, operations, policy, and regulations (Modarres, 2006). It is used to evaluate complex systems, such as those in nuclear and aerospace industries, and its objectives are usually design improvement, risk acceptability, decision support, regulatory and oversight support, and operations and life management. To achieve these objectives, the PRA is combined with qualitative and quantitative results from deterministic processes to analyze risks and make decisions (Modarres, 2006).

The methods and techniques are not exclusive to one industry and can be selected and combined depending on the objectives of the analysis, type and availability of data, and preference of the analyst. Tables 2.5 and 2.6 summarize some of the most common deterministic and stochastic techniques, their type, advantages, and disadvantages.

Table 2.5. Deterministic risk assessment techniques used in different industries.

Technique	Application	Advantages	Disadvantages
Safety audits (Cacciabue, 2004; Marhavidas and Koulouriotis, 2012b)	Operational procedures are inspected according to safety programs (norms and standards). It is used to study human factors.	The evaluations are recurrent which could ensure safety levels and detect risk early.	Limited to the identification of safety critical factors.
Fault trees and event trees (Aven and Kristensen, 2005; Iannacchione et al., 2008; NASA, 2011)	Fault trees: Failure relationship of more complex events with more basic events. Event trees: Practical quantification of accident scenarios. Probabilities and expected values from hard data and expert opinions. The uncertainty can be expressed by confidence intervals.	Well-suited to quantitative analysis when probabilities can be assigned.	High level of details needed for each event. Mostly used for timed events.
Risk matrix (Iannacchione et al., 2008)	Qualitative categories are defined (low-to-high or unlikely-to-likely).	Used in many qualitative risk analysis techniques.	Ranking of risk is subjective.

Table 2.6. Stochastic risk assessment techniques used in different industries.

Technique	Application	Advantages	Disadvantages
Monte Carlo simulation (Safety and Reliability Society, 2011)	The probability of a variable is determined by random numbers. By repeating this process, the distribution of the output random variable may be built up, from which estimates of the parameters of interest may be calculated.	Good for complex systems that may be subject to change later. Very flexible.	Large calculations. Solutions are not exact and depend on the number of runs.
Bayesian analysis (Aven and Kristensen, 2005)	Probability is a measure of uncertainty which is divided in two parts: variation in the population and uncertainty about what value is the true value of this chance.	Can be used with fault trees and event trees.	Uses subjective probability distribution.
Probabilistic distributions (Aven and Kristensen, 2005; Pidgeon, 1998; Safety and Reliability Society, 2011)	Quantifications of risk are based on statistics using historical data resulting in numbers that are not facts. Assumptions are necessary to obtain sufficient volume of data.	Understanding the distribution of random events allows users to apply practical solutions to operational problems.	Risk expressed by probabilities is subjective. This narrow view of risk alone cannot establish safety levels. Statistics may result in low risk numbers.

Table 2.6. Stochastic risk assessment techniques used in different industries (continued).

Technique	Application	Advantages	Disadvantages
Numerical models (Aven and Kristensen, 2005)	Theories and laws used to simplify representations of the world. Needs a balance between simplicity and accuracy.	Different choices depending on the context. Uncertainties are assessed. Not limited to the engineering community.	Not useful if not considered sufficient accurate.

2.4.2. Risk assessment in the O&G industry

Risk assessment in the O&G industry has evolved since the 1960's when risk was controlled only by applying proper safety management. In addition, risk estimates were very uncertain and data was very limited. It was until the 1970's to the 1990's when risk analysis was established as a technique to support regulatory decisions and safety management systems (Aven and Kristensen, 2005). Risk assessment in the O&G industry is widely used in offshore operations and in some countries such as U.K. and Norway companies are required to perform risk analysis prior to operations (Cai et al., 2013; Skogdalen and Vinnem, 2012). Table 2.7, which is by no means exhaustive, shows some examples of the risk assessment methods and techniques that have been used in the O&G industry.

The environmental (ecological) risk assessment (ERA) was developed by the EPA to evaluate the likelihood of adverse ecological effects caused by exposure to physical, chemical or biological stressors (EPA, 1998). The assessment is performed using data from field or laboratory studies or from models which produce two types of outputs: quantitative risk estimates and qualitative conclusions (EPA, 1998). In addition, it intends to transform scientific data into information about the effects of human activities on the environment (EPA, 1998). The EPA, U.S. National Imagery and Mapping, U.S. Department of Energy (USDOE), Russian Federal Center of Geological Systems, and the Ministry of Defense of the Russian Federation conducted an ERA of O&G activities in the Priobskoye oil field in western Siberia (EPA, 1998). Different techniques were applied including geographical information system (GIS) database, national security systems-derived products, environmental impact assessment, and algorithms (EPA, 1998).

Table 2.7. Risk assessment methods and techniques used in the O&G industry.

Method	Technique	
ERA (EPA, 1998)	<ul style="list-style-type: none"> • GIS • Historical Imagery Data • National Security Systems Imagery Data 	<ul style="list-style-type: none"> • Boolean Logic • Environmental Impact Assessment • Hazard Assessment
Barrier and operational risk analysis (BORA) (Aven et al., 2006)	<ul style="list-style-type: none"> • Risk Influencing Factors (RIFs) • Barrier Block Diagrams and Influence Diagrams 	<ul style="list-style-type: none"> • Frequencies/Probabilities • Event Trees and Fault Trees • Checklists and Manual Inspection
Hazard identification (HAZID) (McCoy et al., 1999; Silvanita et al., 2011)	<ul style="list-style-type: none"> • FMEA • FTA/LTA 	<ul style="list-style-type: none"> • ETA • HAZOP
Layers of protection analysis (LOPA) (Habibi et al., 2013; Summers, 2003)	<ul style="list-style-type: none"> • PFDs • P&IDs 	<ul style="list-style-type: none"> • Hazard Scenarios • Risk Tolerance Criteria • HAZOP
QRA (Standards Norway, 2010)	<ul style="list-style-type: none"> • RIFs • Frequencies/ Probabilities • Sub-Models • FTA and ETA 	<ul style="list-style-type: none"> • PFDs and P&IDs • FMEA • Task Analysis

The BORA uses a detailed and quantitative model of barrier performance. The barriers are used to prevent initiating events from happening and reduce consequences. The BORA

method includes development of the risk model, assignment of probabilities of events, identification and assessment of risk influencing factors, and calculation of specific probabilities (Aven et al., 2006). The HAZID method is based on fault propagation and event tree analysis to evaluate failure sequence and consequences (McCoy et al., 1999). The basic process consists of decomposing the plant or system into equipment or units and then creates a model for each unit. The connection between the units is analyzed as well as a model of the fluids in the system. The LOPA is a semi-quantitative method that estimates hazards based on the HAZOP output and the adequacy of protection layers used to mitigate risk (Habibi et al., 2013; Summers, 2003). The LOPA compares a scenario or impact event with a benchmark or the target factor to measure the gap between the existing situation and the tolerable level of risk (Habibi et al., 2013).

The QRA was exclusively used in the offshore industry in Norway during the 1980's but it was implemented in the U.K. afterwards (Skogdalen and Vinnem, 2012). This technique combines sub-models to analyze individual and societal risks and defines individual risk as the probability of an unprotected person to get hurt in a hazardous location (Marhavilas and Koulouriotis, 2012b). The disadvantage of QRA is that it does not include human and organizational factors (HOFs) such as working practice, communication, and procedures. Despite this, Norway and U.K. regulations require to include HOFs in offshore QRAs so there have been efforts to develop methods to formally include these factors (Skogdalen and Vinnem, 2011). Cai et al. (2013) applied Bayesian networks in QRA of subsea blowout preventer operations. The approach in this study consists of five steps: 1) Translate the process flow chart into a Bayesian network, 2) classify the influencing factors of the nodes into human, hardware, software, mechanical, or hydraulic, 3) establish single Bayesian networks for each factor, 4) integrate the single networks into the main Bayesian network, and 5) analyze the Bayesian network model. The analysis shows that the factors that affect safety the most are mechanical and hydraulic, the least important are software and hardware,

and human factors are in the middle (Cai et al., 2013). Although this study is about offshore O&G industry, it could be modified to apply on onshore operations.

2.4.3. Recent risk assessment in unconventional O&G development

2.4.3.1. Engineering approach

For many years, the engineering approach to assess risk has been the preferable method in many areas, including the O&G industry (Aven and Kristensen, 2005; Jacquet, 2014). This approach defines risks based on probabilities or expected values which are complemented with the estimation of uncertainties using different techniques such as Bayesian networks (Aven and Kristensen, 2005).

In 2010, the EPA started working on a study titled "Potential Impacts of Hydraulic Fracturing on Drinking Water Resources" at the request of the U.S. Congress. This study intends to assess and determine the risks HF has on drinking water by "identifying the driving factors that may affect the severity and frequency of such impacts" (EPA, 2012). The progress report published in 2012 collects information about existing data, scenario evaluations, laboratory studies, toxicity assessment and case studies. In June 2015, the EPA released a second draft for review titled "Assessment of the Potential Impacts of Hydraulic Fracturing for Oil and Gas on Drinking Water" which will be used to develop the final report.

The USDOE published a report "Environmental Impacts of Unconventional Natural Gas Development and Production" in May 2014 which summarizes, from different publications, the potential environmental impacts of operations within the lower 48 States shale plays (USDOE, 2014). The type of environmental impacts documented are greenhouse emissions and climate change, air quality, water use and quality, induced seismicity, and land use and habitat fragmentation.

Intensive water usage during unconventional O&G production could decrease water availability, especially in arid regions or during drought season (USDOE, 2013b). Drought risk assessment is necessary to enhance energy security by forecasting and quantifying risk.

Several studies have been performed using different methods such as the Standardized Precipitation Indices and the Palmer Drought Severity Index (Strzepek et al., 2010). Markovian models have been applied for hydrological processes but they are not adequate to describe drought events due to longer time dependence (Chung and Salas, 2000). Chung and Salas (2000) proposed the use of low-order discrete autoregressive moving average models combined with probability distribution of drought events, expected values, variances, and Monte Carlo simulation to describe the associated risks. Other widely used methods are remote sensing and GIS which use satellite derived indices and exact spatial information to analyze drought-risk sensitive areas and quantify the risk (Lin and Chen, 2011; Vicente-Serrano, 2006; Wu and Wilhite, 2004).

Rozell and Reaven (2012) studied the likelihood of water contamination during the production of natural gas from the Marcellus Shale. The risks were calculated using probability bounds analysis based on different sources including databases from State environmental agencies. The study presents the best and worst case scenarios with the aim of providing a technique for decision-making instead of exact results. In addition, the study only focuses on the Marcellus Shale and makes several assumptions due to the lack of information.

The environmental and public risk of different pathways of contamination focusing on fluid containment and transport systems was analyzed by Ziemkiewicz et al. (2014). The pathways analyzed are integrity of the lining in pits and impoundments and pipelines used to transport fluids to and from the sites studied. Data were collected in the field, including water samples, and with it a probabilistic analysis was performed using event trees and categorization by severity ranking. Likelihood was calculated by taking the ratio of the number of times the problem was observed to the total number of sites evaluated. A binomial distribution was developed based on a population of 70 pits and impoundments with a sample size of 14 sites.

Soeder et al. (2014) performed an engineering risk assessment using an integrated assessment model (IAM), which has been used to assess carbon dioxide storage in geologic systems. The approach to evaluate environmental risk elements is similar for shale formations. Hence, the IAM can be modified for HF by including risk elements that are different from CO₂ storage sites. The process intends to identify short and long term risks known as features, events or processes (FEP). The FEP analysis uses high-fidelity models to evaluate the risks. The output from these numerical models is simplified to reduced-order models which are then used for the IAM (Soeder et al., 2014). The IAM uses laboratory analysis (e.g. microbiological analysis and cement/wellbore analysis), data collected in the field, and numerical modeling (e.g. Monte Carlo simulations of field-scale performance). The disadvantage of the IAM process is that health and ecosystem impacts are beyond its scope.

Casing and cement impairment in conventional and unconventional O&G wells in Pennsylvania was studied by Ingraffea et al. (2014) using statewide data and the Cox proportional hazards model. The Cox regression, or proportional hazards regression, is a semi-parametric and multivariate analysis that uses the hazard function to study the survival of an individual or object based on a rate instead of a proportion (Ingraffea et al., 2014b). With this model, it was possible to capture temporal and geographic dimensions and hazards ratios of the count of impairment events that were inspected (Ingraffea et al., 2014a). Siegel et al. (2015) collected samples from water wells that were located near 661 O&G wells to study the relationship of methane migration and proximity to existing O&G wells. Four statistical tests were used which are: 1) test of proportions to compare samples with a threshold concentration, 2) logistic regression to find a trend, 3) survival analysis to compare statistical distributions between two groups, and 4) correlation analysis between methane concentration and distance (Siegel et al., 2015).

Risk assessment from an engineering approach has its challenges including the difficulty of assessing uncertainties and assigning probabilities and appropriate values for

estimations, ability to distinguish between objective knowledge and subjective judgments, difficulty of working with intangibles and uncertainties, and failure to include temporal data (Aven et al., 2007; Ingraffea et al., 2014a). Some risks are easier to manage than others and the manageability and uncertainties depend on the stage of development of the system (Aven et al., 2007). In the early stages of the system development, the uncertainties and manageability are larger. In addition, non-disclosure agreements that allow companies to hold back contamination reports limit the data availability for risk assessments. Some results from risk assessments regarding contamination cannot always be attributed to HF due to insufficient relevant databases, and lack of pre- and post- information on the presence of methane and petroleum byproducts in the basins (Adgate et al., 2014).

2.4.3.2. Holistic approach

It is common to include a cost/benefit or cost/effectiveness analysis to the risk assessment but this is not the most adequate approach for unconventional O&G because there will always be an economic justification (Aven et al., 2007). Similarly, using the engineering approach solely will result in an incomplete analysis or even bias since there has been cases of overconfidence in judgments by experts (Aven and Kristensen, 2005; Pidgeon, 1998). A better approach would be a balance between scientific judgments and social beliefs (Pidgeon, 1998; Renn et al., 1992). This unification is based on the idea that hazards are related to psychological, social, institutional and cultural processes in ways that can affect perceptions of risks and dictate risk behaviors (Renn et al., 1992).

Recent expansion of O&G activities has caught the attention of different stakeholders including the general public. This has resulted in sociological studies that consider the public engagement in risk characterization and decision making (North et al., 2014). A review of the different parameters affecting how people perceive risk revealed that the two most important factors are familiarity with the process and trust (Wachinger et al., 2013). Theodori et al. (2014) conducted a survey in the Marcellus Shale to study social perception of HF and found

that almost half of the respondents are unfamiliar with the practice and that the natural gas industry is considered the least trustworthy source of information. The public mistrust and perception of lack of transparency produce higher levels of stress and with it other health problems (Adgate et al., 2014).

Natural resources dependent communities are often benefited with employment opportunities and business; however, these benefits are short-term (Jacquet, 2014). Previous studies show that massive industrialization and worker immigration in a short period of time result in overwhelmed housing supplies, stressed municipal services (e.g. potable water) and government programs, and disruption in social and economic patterns (Jacquet, 2014). Also, national and regional surveys indicate that perceptions of the impacts are polarized, for example, between financial gain and environmental impacts (Jacquet, 2014). Negative public perceptions of unconventional O&G production can be improved by developing non-toxic fracturing chemicals, community adaptation, use of alternative water sources, full communication between and among the stakeholders, and sharing more information and educating the public about wastewater treatment technologies (Theodori et al., 2014).

Perry (2012) discusses the first draft report by the EPA, *Potential Impacts of Hydraulic Fracturing on Drinking Water Resources*, and mentions the flaws it may have regarding the lack of social factors. Thus, the author proposes the use of an iterative analytic-deliberative process where deliberation is made by all the parties involved resulting in better long-term decision making. The uncertainties and risks regarding social, community, and human health factors (societal cost) are evaluated not only by quantitative measurements but by using local, community and temporal scale and other qualitative criteria (Perry, 2012). The analytic-deliberative process is a promising alternative for a holistic approach to risk governance; however, it has not yet been completely adopted in shale risk assessment (North et al., 2014). Likewise, Aven (2012), Aven and Kristensen (2005), and Pidgeon (1998) proposed a

combination of engineering and social science research to open up for new ways of measuring risk and its uncertainties.

Just like risk assessment from an engineering approach, social science research also faces limitations. Some of these constraints are self-selected populations, small sample sizes, short follow-up times and unclear loss to follow-up rates, limited exposure measurements, unavailable exposure data, and if available it is inconsistent, particularly for non-cancer health effects (Adgate et al., 2014). One way to perform risk assessment in the O&G industry effectively would be combining the available information, both engineering and social sciences databases, and use the different techniques mentioned in the previous section to estimate the gaps. This holistic approach would be able to move narrow risk concept based on probabilities to a broader view (Aven and Kristensen, 2005; Pidgeon, 1998). Furthermore, this broader view can be used to make decisions based on scientific judgment and perception of the public (Pidgeon, 1998). An option to perform holistic risk assessment in unconventional O&G is the approach proposed by Aven and Kristensen (2005) shown in Figure 2.4.

1. Data (facts)

- **Quantitative:** Engineering risk assessments (e.g. probabilities, expected values, uncertainties).
- **Qualitative:** Human and organizational factors (e.g. working environment conditions, safety management, attitudes and culture).
- **Observational:** Historical data (e.g. field data, state and federal spills reports, fatalities, and other major accidents).
- **Simulated:** Results from computational tool (e.g. studies on shale behavior stimulated with HF).

2. Risk analysis descriptions

- Perform risk assessment: hazard identification, dose-response assessment, exposure assessment, and risk characterization which is both quantitative and qualitative (National Research Council, 1983).

3. Perceived risk information

- Include social science research about risk perception: National and private surveys (e.g. interviews and questioners to understand risk-related behavior, general public perception, and other stakeholders).

4. Judgements by special competence people

- Take into account evaluations by the people in the risk and safety field.
- Involve ethical, strategic, and political concerns (Aven et al., 2007).
- Safety management reviews can be included.

5. Expert groups opinion

- Technical community: Multidisciplinary conclusions about system performance, safety levels, and uncertainties. Subjective opinions from experts in the field.

6. Representatives from various interested parties

- Discussion on several judgements and views from different parties to build trust and consensus (e.g. decision makers, stakeholders, individual or organization, internal or external).

Figure 2.4. Holistic approach components to perform risk assessment.

2.5. Results from previous risk assessments related to unconventional O&G

Data have shown that most environmental or safety incidents related to shale gas wells result from operations not being performed according to the recommended engineering practices or procedures (Soeder et al., 2014). The potential risks in every stage of the water life cycle that could result from improper procedures and management are discussed in details below.

2.5.1. Water acquisition

ND is less susceptible to water stress compared to Texas but is still prone to droughts and floods. The ND State Water Commission issues groundwater and surface water withdrawal permits and requires annual reports from users. Furthermore, water from the Missouri River is readily available but only ten miles of it are accessible to O&G operators due to a restriction imposed by the U.S. Army Corps of Engineers (ND State Water Commission, 2014). Despite this, in 2012 HF used 2.08×10^7 m³ of water, more than the amount used by Fargo, the largest city in the State with a population of 110,000 people (Freyman, 2014).

HF is intensively used in Texas where more than half of the wells are located in regions that have medium to extreme high water stress (Freyman, 2014). This means that water is limited since it is already used for other purposes. The climate in Texas ranges from semiarid west to sub-humid east, and in the past years drought has been exceptional or extreme. In 2011, 88% of the State faced maximum drought, with record temperatures above 100°F (Scanlon et al., 2013). Although water used per well has increased in recent years in the Barnett Shale, so has the length of the lateral or horizontal portion of the well, indicating that the water used per length has remained constant (Nicot et al., 2014; Scanlon et al., 2014). Despite this, it is expected that by 2020 the water used for HF in Texas will reach 1.51×10^8 m³ per year or the equivalence of 19,700 Olympic-sized swimming pools (Freyman, 2014).

Pennsylvania is considered to be at low risk of droughts and groundwater challenges (Freyman, 2014). Despite this, the majority of wells (62%) in the Marcellus Shale are located

in regions considered to have medium water stress, particularly during summer months. This makes the risk of water shortage to be associated mostly with the time of withdrawal rather than the quantity of water available (Freyman, 2014). From 2005 to mid-2013, Pennsylvania used $\sim 1.14 \times 10^8 \text{ m}^3$ of water for HF purposes which equal the residential water use of almost 156,000 people in the same time period (Environment America, 2013; PA Public Utility Commission, 2014). The Commissions of the Susquehanna River Basin and the Delaware River Basin have different roles in the natural gas development, from issuing permits for water withdrawals to monitoring wastewater storage, treatment, and disposal (Delaware River Basin Commission, 2010; Susquehanna River Basin Commission, 2013).

The impacts of drought can be environmental or socioeconomic including land degradation, desertification, water scarcity, agriculture and food security, services (e.g. water and energy supply), and conflict over resources (Yan, 2010). A study in which the Standardized Precipitation Indices and the Palmer Drought Severity Index were combined showed that meteorological drought (based on precipitation alone) is expected to increase in some regions of the U.S. while the hydrological drought (based on precipitation and temperature) will affect most of the country by 2050 (Strzepek et al., 2010). Since the beginning of the 20th century, temperature across the U.S. has been increasing to the point that 60% of the country experienced some level of drought during summer of 2012 (USDOE, 2013b). In the last decade, there have been several events that reflect the vulnerability of the energy sector due to decrease water availability including the prohibition by the city of Grand Prairie (Texas) to use city water for HF because of extreme drought in the Fall of 2011 and high prices for water or water access denied to operators for several weeks in Kansas, Texas, Pennsylvania, and ND in 2012 (USDOE, 2013b).

Mitigation of water scarcity can be achieved by prioritizing the application of integrated, cross-enterprise water management which includes best practices, investment in new technology and application of strategies designed locally because of regional regulations

and specific environmental attributes of the shale play (Gay and Slaughter, 2014; Mauter et al., 2014). Some of the critical attributes of a shale play are geographic distribution, diversity of hydrospheres, land surface, and biospheres which lead to regional stressors (Mauter et al., 2014).

2.5.2. Chemical mixing

In 2012, the EPA identified more than 1,000 chemicals used in HF from which 27 chemicals (Table 2.8) are known or suspected carcinogens, or listed as hazardous air pollutants that may impact drinking water. In addition, 82 chemicals are considered confidential business information and therefore undisclosed to the public (EPA, 2012). A major concern in this stage is the possible risk of fracturing fluid spills and contamination of drinking water sources but data to quantify the risk are not available. Several databases were analyzed by the EPA (2012) and the information regarding incidents is unclear.

It is difficult to quantify the risk of contamination directly related with chemicals and produced water when the reports do not specify the content of the fluids spilled. In addition, most of these spills are reported only by the media (EPA, 2012). The ND Department of Health (ND DoH) database shows almost 8,000 spills of oil, brine, and other chemicals between 2000 and 2013 (Cwiak et al., 2015). Figure 2.5 shows the location of the spills reported by the ND DoH.

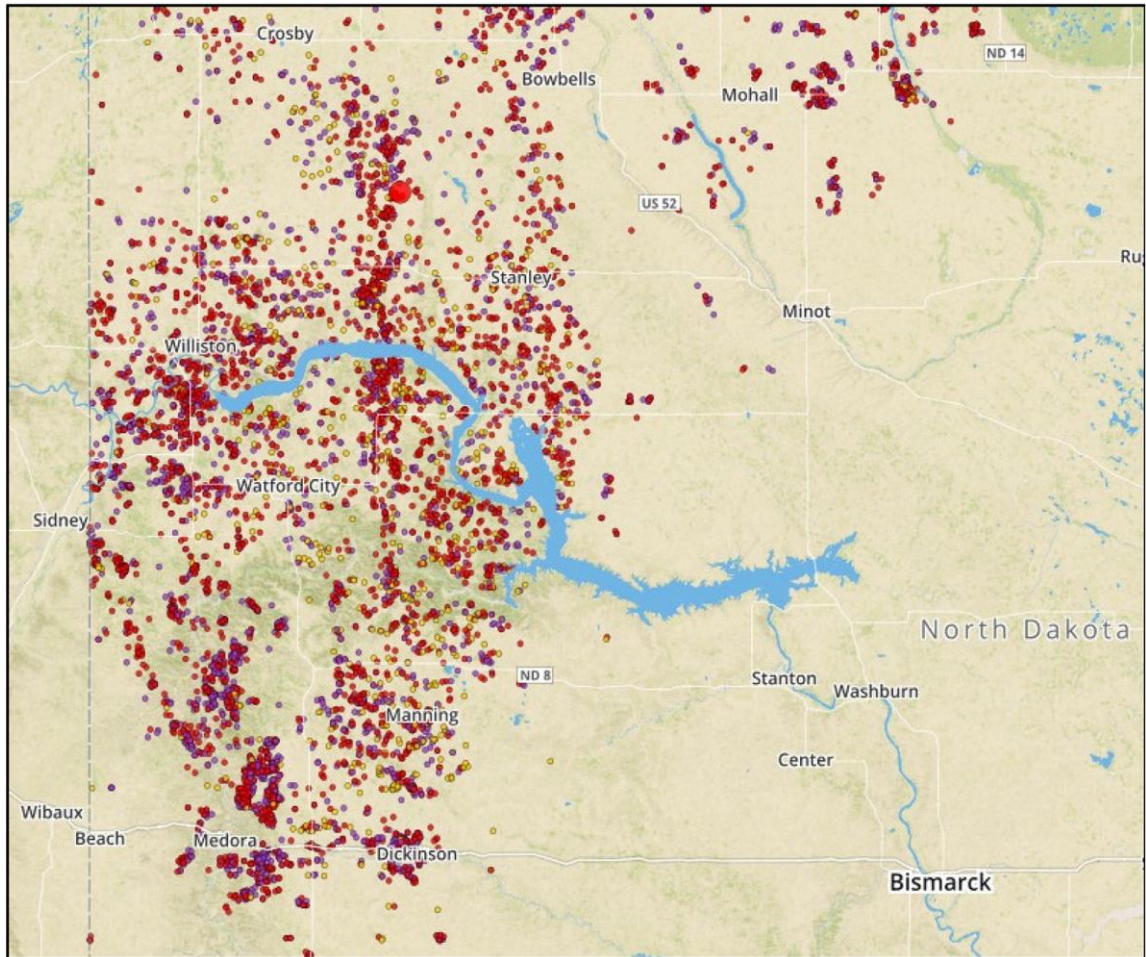


Figure 2.5. Spills in ND from 2000 to 2013. Red: oil, purple: brine, and yellow: other. Adapted from Gage Cartographics (2014).

Table 2.8. Chemicals suspected to be carcinogens, hazardous air pollutants (HAP) or regulated under the Safe Drinking Water Act (SDWA) and number of products used in HF that contain these chemicals (EPA, 2012).

Chemicals	Category	No. of products	Chemicals	Category	No. of products
Methanol	HAP	342	Phenol	HAP	5
Ethylene glycol	HAP	119	Benzene	Carcinogen, SDWA, HAP	3
Naphthalene	Carcinogen, HAP	44	Di (2-ethylhexyl) phthalate	Carcinogen, SDWA, HAP	3
Xylene	SDWA, HAP	44	Acrylamide	Carcinogen, SDWA, HAP	2
Hydrochloric acid	HAP	42	Hydrofluoric acid	HAP	2
Toluene	SDWA, HAP	29	Phthalic anhydride	HAP	2
Ethylbenzene	SDWA, HAP	28	Acetaldehyde	Carcinogen, HAP	1
Diethanolamine	HAP	14	Acetophenone	HAP	1
Formaldehyde	Carcinogen, HAP	12	Copper	SDWA	1
Thiourea	Carcinogen	9	Ethylene oxide	Carcinogen, HAP	1

Table 2.8. Chemicals suspected to be carcinogens, hazardous air pollutants (HAP) or regulated under the Safe Drinking Water Act (SDWA) and number of products used in HF that contain these chemicals (EPA, 2012) (continued).

Chemicals	Category	No. of products	Chemicals	Category	No. of products
Benzyl chloride	Carcinogen, HAP	8	Pb	Carcinogen, SDWA, HAP	1
Cumene	HAP	6	Propylene oxide	Carcinogen, HAP	1
Nitrilotri-acetic acid	Carcinogen	6	p-Xylene	HAP	1
Dimethyl formamide	HAP	5			

The entities in charge of keeping track of O&G spills in Texas are the Railroad Commission (RRC) and the Commission on Environmental Quality. The Texas RRC keeps reports of spills which are categorized as crude, combined liquids, gas well liquid, or products. In 2013-2014, there were 2,316 reported spills from which 312 are classified as gas well liquid or products (RRC of Texas, 2015b). In Pennsylvania, according to the EPA, in the period of 2006 to 2012, there were 4,319 inspections with violations in the Marcellus Shale region. Once again, the nature of the incidents is not clear.

During chemical mixing there is also a concern on exposure to some of the additives in the fracturing fluid that are known to be toxic; however, the maximum exposure levels without any adverse effects are not clear. Due to lack of data about pre- and post- drilling activities, extensive and long-term studies on chemicals exposures and health effects are not available (EPA, 2015). Also, the data required for this type of studies is usually extensive and difficult to collect (Shonkoff et al., 2014; Stern et al., 2014). To date, there are no population-

based studies that explain the health impacts of unconventional gas production related to water contamination (Adgate et al., 2014). The seven chemicals most used in the fracturing fluid, mentioned in section 3, and their effects on health are shown in Table 2.9.

Colborn et al. (2011) found that from the total chemicals reported to be used in HF to extract natural gas, more than 75% could affect the skin, eyes, and other sensory organs, and the respiratory and gastrointestinal systems. Also, around 40-50% could affect the nervous system, immune and cardiovascular system and kidneys. In addition, of the total chemicals used 37% affect the endocrine system and 25% could cause cancer (Colborn et al., 2011).

2.5.3. Well injection (HF)

Groundwater contamination could be caused by fluids migration through natural or induced fractures. Previous studies suggest that pathways for gas can also serve as pathways for HF fluid migration (Osborn et al., 2011; Warner et al., 2012). Osborn et al. (2011) analyzed 68 private groundwater wells to determine the concentrations of dissolved salts, water isotopes, and isotopes of dissolved carbon, boron, and radium. From these wells, 60 were also analyzed for methane and higher-chain hydrocarbon content. The study found that 85% of the wells contained methane within the defined action level for hazard migration. However, gas found in water wells and shallow aquifers is believed to be a natural and common phenomenon (Osborn et al., 2011; Siegel et al., 2015).

Table 2.9. Most common chemicals used in HF fluid and their limits and health effects.

Chemical	Level	Health effects
Methanol	0.5 mg/kg-day reference dose, intake level at or below which no health effects are likely to occur even with long-term daily exposures (Saba et al., 2012).	Narcosis, metabolic acidosis. Severe abdominal, leg, and back pain occur and visual degeneration can lead to blindness. 80 to 150 ml of methanol is usually fatal to humans.
Isopropanol	400 ppm (980 mg/m ³) total weight average (TWA) - OSHA* permissible exposure limit (PEL)**	Narcosis, mild eye, nose, and throat irritation.
Crystalline silica	50 µg/m ³ proposed PEL by the OSHA.	Silicosis, lung cancer, chronic obstructive pulmonary disease, and kidney disease.
2-Butoxyethanol	240 mg/m ³ OSHA PEL, 1.6 mg/m ³ EPA inhalation reference concentration (RfC).	Mild irritation. Not likely to be carcinogenic to humans at or below the RfC.
Ethylene glycol	100 mg/m ³ threshold limit value. NIOSH*** recommended exposure limit (REL) has not been established.	Irritation-eye, nose, throat, skin.

Table 2.9. Most common chemicals used in HF fluid and their limits and health effects (continued).

Chemical	Level	Health effects
Hydrotreated light petroleum distillates	100 mg/m ³ NIOSH recommended TWA 10 hours.	Irritation, nausea, headache, drowsiness, symptoms of drunkenness, lung congestion, convulsions, coma.
Sodium hydroxide	2 mg/m ³ TWA OSHA PEL.	Ulceration of nasal passages. Eye, skin, and respiratory irritation.

* OSHA: Occupational Safety and Health Administration.

** PEL's are based on 8-hour time weight average exposure limit.

*** NIOSH: National Institute for Occupational Safety and Health.

Results from Engelder et al. (2014) indicate that the flowback and produced water that remain inside the well do not pose a threat to shallow aquifers by migrating upward along natural pathways because the capillary and osmotic forces keep the fluids permanently inside the shale matrix. Vengosh et al. (2014) reported that fractures after the hydraulic stimulation are less than 600 m above well perforation which is insufficient to reach groundwater resources. In addition, Reilly et al. (2015) found through chemical analysis that the most common source of groundwater contamination is septic effluent. Similar to the observations made by Osborn et al. (2011), Jackson et al. (2013) found that 82% of the drinking water samples contained methane and from the different factors analyzed, distance to gas wells was the dominant one. In the same study both biogenic and thermogenic sources were considered and overall the results show that methane found is of thermogenic origin which suggests that it reaches shallow water through casing failures or imperfections in cement annulus of the gas wells (Jackson et al., 2013). However, a more recent study has found no relationship between dissolved methane concentrations and proximity to existing O&G wells (Siegel et al., 2015). Siegel et al. (2015) analyzed groundwater samples from locations near

gas wells and found no evidence of systematic increased methane concentration closer to these wells.

Groundwater contamination could be also caused by HF fluid migration due to casing failure. According to the study conducted by Rozell and Reaven (2012), the probability of a well failing is in the range of 2.0×10^{-8} and 2.0×10^{-2} and the chance of a well leaking per year is from 1×10^{-6} to 0.1. Ingraffea et al. (2014a) indicate that Pennsylvania records show that between 0.7% and 9.1% of the O&G wells developed since 2000 show a loss of well integrity and the higher risks are observed in unconventional wells. However, the hazard modeling conducted in the same study indicates that the loss of structural integrity is actually 12% for unconventional wells drilled since 2009. Furthermore, this and other studies indicate that the most common methane migration mechanism, if not coming from a natural source, is this loss of integrity of the cement and casing of the wells (Ingraffea et al., 2014a; Vengosh et al., 2014).

2.5.4. Flowback and produced water production

The risk of spills and/or leaks could result in surface and groundwater contamination similar to the stages of chemical mixing and well injection (EPA, 2012). A constraint to assess risks associated with this stage is that specific spill data related to produced water is not completely available to the public. During 2012, in ND 25.5 million barrels of brine were generated and there were 141 reports of pipeline leaks from which approximately 8,000 barrels of brine were spilled (Al Jazeera America, 2014). The Texas RRC and the Commission on Environmental Quality track spills mostly from oil, gas and liquid condensate but there are no reports related to HF fluids (EPA, 2012).

Flowback and produced water contain high concentrations of different contaminants which complicate the treatment to reach acceptable levels for discharge and reuse. Some of the organic and inorganic contaminants found in produced water are listed in Tables 2.10 and 2.11, respectively (Agency for Toxic Substances and Disease Registry, 1999; Maguire-Boyle

and Barron, 2014; Orem et al., 2014; Wilson and VanBriesen, 2012). Organic constituents can be originated from the produced water, the shale itself, the oil in the shale, or the fracturing fluid (added chemicals) (Orem et al., 2014). One additive used intensively in the fracturing fluid is gel, generally guar gum and its derivatives, to increase the viscosity of water and improve the transport of sand into the fractures (Lester et al., 2014). The gel does not pose a threat to health but it may have effects on membrane separation treatment processes affecting the efficiency of contaminants removal (Lester et al., 2014).

Some studies indicate that the radioactivity in most produced waters is directly proportional to the content of salts (Brown, 2014; Fisher, 1998; Vengosh et al., 2014). The content of Ra-226 in the Marcellus Shale produced water can be higher than 10,000 pCi/L (picocuries per liter) while the standard for drinking water (Ra-226 and Ra-228) is just 5 pCi/L (Brown, 2014; Osborn et al., 2011). In ND some radioactive material has been found in different waste streams from O&G activities, mostly scale in equipment, in concentrations above natural backgrounds (Argonne National Laboratory, 2014). Also, one study conducted in the Barnett Shale found that the total beta radiation in one reserve pit was eight times higher than the regulatory limit (Brown, 2014).

Table 2.10. Organic content of produced water from typical shale gas wells and effects on health.

Compound type	Level/source	Health effect of different compounds
Dissolved organic carbon	Hydrocarbons found in the produced water at levels as high as 5,500 mg/L	<ul style="list-style-type: none"> • Cyclic octaatomic sulfur: Microbiological activity indicator • Straight chain alkanes/alkenes: Mucosal irritation in nasal turbinates and larynx in rat, cystic uterine endometrial hyperplasia in mice, and carcinogenic potential • Aromatics and aliphatics: Hepatic and renal effects, hemolytic anemia, and respiratory irritant effects in animals • Carboxylic acids: Low genotoxic potential
Added organic chemicals	Found in the flowback water at levels > 1,000 µg/L per individual compound	<ul style="list-style-type: none"> • Aliphatic hydrocarbons (solvents): Respiratory irritant effects in animals, asphyxia and chemical pneumonitis • Brominated nitrilopropionamides and hexahydro-1, 3, 5-trimethyl-1, 3, 5-triazine-2-thione (biocide): Developmental, reproductive, mutagenic, carcinogenic, or neurological effects • Ethylene glycol and derivatives (cross linker and scale inhibitors): Central nervous system depression, cardiopulmonary effects, and renal damage • Guar gum and diesel fuel (gelling agent): Guar gum does not pose a threat but diesel fuel contains known carcinogens

Table 2.10. Organic content of produced water from typical shale gas wells and effects on health (continued).

Compound type	Level/source	Health effect of different compounds
Added organic chemicals	Found in the flowback water at levels > 1,000 µg/L per individual compound	<ul style="list-style-type: none"> • Ethanol (foaming agent): Malnutrition, effects on hepatic metabolism and immunological functions • Methanol (corrosion inhibitor): Visual disturbances, neurological damage, dermatitis • Fatty acid phthalate esters (breaker): Liver effects
Benzene, toluene, ethylbenzene, and xylene	Contained in diesel used as a gelling agent	<ul style="list-style-type: none"> • Cancer risk, neurological effects, primarily central nervous system depression, ototoxicity, hemato-logical, immunological, and lymphoreticular effects.
Polycyclic aromatic hydrocarbons	Lower than off-shore produced waters	<ul style="list-style-type: none"> • Carcinogenic, reproductive problems in mice, and respiratory effects
Volatile fatty acids (VFAs)	Produced by bacteria. Maxi-mum level of 53.7 mg/L.	<ul style="list-style-type: none"> • Aliphatic acid anion (primarily acetate): Induces headache in sensitized rats, corrosive for the skin, eye damage, and mucous membranes irritation. VFAs are responsible for unpleasant odor in wastewater

Table 2.11. Inorganic contents of produced water from typical shale gas wells and the effects on health.

Contents	Health effects	Regulated levels
Sodium	Unlikely to have adverse health effects	20-60 mg/L for esthetic effects (recommended)
Calcium and magnesium	Causes hardness in water but don not represent a threat to health	500 mg/L TDS
Potassium	High doses can affect health in people with kidney disease, heart disease, coronary artery disease, hypertension, diabetes, adrenal insufficiency, and people with limited renal reserve are more vulnerable.	4.7 g/day adequate intake for adults
Iron	Not a threat to health	0.3 mg/L recommended
Strontium	Strontium accumulates in bones. Children are more vulnerable to excess strontium.	4.0 mg/L lifetime health advisory level
Barium	Causes increase in blood pressure	2 mg/L
Chloride	Unlikely to have adverse health effects	250 mg/L
Bromide	In high and chronic doses, vomiting or stupor, depression, loss of muscle coordination and psychoses. Increases formation of disinfection by-products that are carcinogenic and potentially teratogenic	1.0 ppm

2.5.5. Produced water treatment and waste disposal

Surface spills can be caused by leaking reserve pits and pipes, transportation accidents, and improper treatment followed by stream discharge. All of these scenarios

present a threat of drinking water contamination (EPA, 2012). The purpose of wastewater treatment is to eliminate the TDS, which are mostly derived from the subsurface or lower the concentration to acceptable level before discharge (Lutz et al., 2013). However, not all treatment plants have the capacity or the technology to successfully remove naturally occurring salts (Wilson and VanBriesen, 2012). Another limitation of wastewater treatment plants is that the content of the produced water is difficult to predict because it varies with time, location, and composition of the fracturing fluid (Barbot et al., 2013).

Ferrar et al. (2013) found that in Pennsylvania prior to a voluntary cessation of off-site treatment (requested by the Pennsylvania Department of Environmental Protection) in 2011, the concentrations of contaminants in the wastewater treatment plants effluents were above quality criteria. After the cessation, the contaminant concentrations in the receiving waters decreased suggesting that on-site treatment is more effective than off-site plants (Ferrar et al., 2013). Brine treatment can reduce concentrations of NORM to more than 90% (Warner et al., 2013a). However, there are still high levels of NORM in receiving stream sediments which pose the risk of bioaccumulation in the food chain (Brown, 2014; Warner et al., 2013a). Health effects of radium consumption in drinking water include tooth fracture, anemia, cataracts, and cancer if exposure is chronic (Rich and Crosby, 2013). In addition, trihalomethanes and other disinfection by-products are produced during the water treatment process due to the elevated bromide and chloride concentration and their reaction with organic compounds which present health risks (Brown, 2014; EPA, 2012; Vengosh et al., 2014; Warner et al., 2013).

Wilson and VanBriesen (2012) found that operators in Pennsylvania have been shifting to recycling and reusing methods and have reduced discharges to surface water bodies. The study shows an increasing rate of water reuse within operations and treatment at publicly owned treatment works and centralized waste treatment plants with effluent limitations established by the EPA (EPA, 2003; Wilson and VanBriesen, 2012). These non-discharging

methods have reduced the levels of bromide that were being released to the environment, but acceptable levels for water treatment plants have not been determined (Wilson and VanBriesen, 2012).

Transportation of wastewater for treatment or disposal requires a considerable number of trucks which increases the probability of traffic accidents that could result in spills. In the Bakken Shale region, there was an increase of 68% of crashes involving trucks from 2006 to 2010 (Environment America, 2013). In the Eagle Ford region, the Texas Department of Transportation reported a 40% increase in fatal motor vehicle accidents from 2008 to 2011 (Adgate et al., 2014). Likewise, the Crash Reporting System from the Pennsylvania Department of Transportation reported an increase in accidents involving heavy trucks between 1997 to 2011 (Adgate et al., 2014).

According to the EPA, $7.6 \times 10^6 \text{ m}^3$ of brine is disposed per day in the 144,000 class II injection wells all over the country. Deep-well injection is one of the most common methods used but additional research is required to determine the long term impacts, especially on groundwater and seismic activity, and to accommodate the demand of produced water volumes (Arthur et al., 2008). There is one study by the EPA (1998b) that determined that the probability of wastewater migration to groundwater resources is very unlikely and depends on the thickness of the low permeability strata overlying the formation.

3. RISK ASSESSMENT OF HUMAN EXPOSURE TO Ra-226 IN OIL PRODUCED WATER FROM THE BAKKEN SHALE

3.1. Introduction

Oil produced water from the Bakken Shale in ND contains relatively high levels of TDS which, based on previous studies, indicate possible elevated levels of NORM (Godoy and Petinatti da Cruz, 2003; Hamlat et al., 2001; Rich and Crosby, 2013; Rowan et al., 2011). This NORM can contain different radioactive elements including Ra-226 which is a major source of radioactivity in produced water (Chriss and Bursh, 2002; Vandenhove et al., 2005; White, 1992; Zhang et al., 2014). In order to develop successful handling and treatment planning of produced water, it is important to characterize its content especially for major contaminants such as Ra-226.

Similar to the effects of TDS on radioactivity levels, there is a strong correlation between Ra-226 and high levels of barium (Ba) and strontium (Sr) which occur simultaneously in produced water from unconventional oil and gas extraction (Barbot et al., 2013; Jerez Vegueria et al., 2002; Zhang et al., 2014). Radium is classified in the alkaline-earth group because its properties are similar to calcium (Ca), Sr, and Ba (Zhang et al., 2014). One of these properties is high electropositive (+2) character of Ra which means that it loses electrons very easily and becomes a cation. Because of this, Ra reacts with many insoluble products such as sulfates (Vandenhove et al., 2005). Pure radium sulfate (RaSO_4) in produced water is not common; instead Ra co-precipitates with other carrier solids such as Ba and Sr (Ceccarello et al., 2004; Doerner and Hoskins, 1925; Zhang et al., 2014). These reactions occur when the injected HF water mixes with the formation water which has different chemical characteristics. Consequently, the chemical equilibrium is disturbed resulting in precipitation of sulfates and carbonates (E&P Forum, 1994).

High levels of sulfate in produced water react with Ba and Sr producing barium sulfate (BaSO_4) and strontium sulfate (SrSO_4), respectively, which promote scale formation in oil

processing equipment (Al-Masri and Aba, 2005; Ceccarello et al., 2004; Lee and Neff, 2011). Due to co-precipitation of Ra-226 with BaSO₄ and SrSO₄, scale can contain relatively high levels of radioactivity (Al-Masri and Aba, 2005; U.S. Geological Survey, 1999; Vandenhove et al., 2005). When the co-precipitation occurs, part of the Ra content stays in the scale while the rest remains in the produced water (Godoy, 1996; Heaton and Lambley, 1995; Logan, 2015). In ND, the average Ra-226 level in equipment scale is 548 pCi/g (Argonne National Laboratory, 2014). Lauer et al. (2016), the only study on produced water from ND, found the average Ra-226 level to be 851 pCi/L (n = 3).

High levels of Ra-226 in produced water poses risks to humans and the environment. The generation of produced water in ND has increased over the years and is expected to climb another 328% between 2014 and 2035 (Kurz et al., 2016). Also, the number of incidents related to produced water spills in the Bakken has increased since 2007 (Kurz et al., 2016; Lauer et al., 2016). Incorrect management of large volumes of produced water containing NORM could affect soil used for agricultural purposes and contaminate surface water bodies that serve as irrigation and drinking water supplies to nearby towns in the State.

The media that could transport Ra-226 into the environment and thus, reach humans are water, air, and soil (Pacific Northwest National Laboratory, 2008; Staven et al., 2003). Ra-226 entering surface water bodies is particularly possible since it is, under certain conditions, very soluble and mobile in aquatic environments (Rajaretnam and Spitz, 2000). Ra may be incorporated into the food chain by plant uptake and animal tissue bioaccumulation. When the contaminated water, crops, and animals are consumed by humans, the risk of Ra entering the body and accumulating in bones arises (Fisher, 1996). The results of long-term exposure to radium include anemia, cataracts, fractured teeth, bone cancer, and even death (Agency for Toxic Substances and Disease Registry, 1990).

In ND, 60% of the surface water available is used for crop irrigation (ND DoH, 1999a). Also, the average consumption of fish and shellfish in the State is 0.33 g/kg-day which is

above the national average consumption of 0.22 g/kg-day (EPA, 2013). The increase of produced water spills along with the high reliability on surface water for drinking, crop irrigation, and fishing purposes could lead to enhanced exposure of the ND residents to Ra-226.

Previous studies have developed multivariate regression models based on the correlation of Ra-226 with other inorganic elements found in equipment scale (Al-Masri and Aba, 2005; Jerez Vegueria et al., 2002; Vidic, 2015). Using the same principle, this study proposes a method to predict Ra-226 content in produced water based on Sr, Ba, and Ca levels. The objective of this study is to investigate the human exposure to Ra-226 through ingestion of contaminated surface water, potatoes and fishes. Due to limited data on Ra-226 levels in produced water from the Bakken, the simulated Ra-226 concentration based on cations and food transfer factors were used to perform the risk assessment.

3.2. Methodology

3.2.1. Method overview

Three different scenarios for human exposure to Ra-226 were selected: 1) ingesting treated water from a lake contaminated with produced water, 2) consuming potatoes which have been irrigated with surface water contaminated with produced water, and 3) consuming fish caught in a lake contaminated with produced water. Only exposure to Ra-226 in water via oral ingestion was considered in scenario 1. For scenario 2, soil to plant transfer factors found in the literature were used to investigate the accumulation of Ra-226 in potatoes, a major agricultural product in ND (Northern Plains Potato Growers Association, n.d.; Carvalho et al., 2009; Tagami and Uchida, 2009; Staven et al., 2003; Watson et al., 1983; Pietrzak-Flis et al., 1995; Schuttelkopf and Kiefer, 1982). Likewise, exposure to Ra-226 accumulated in fishes in scenario 3 was estimated using transfer factors reported in previous research (Clulow et al., 1998; Hosseini et al., 2008; IAEA, 2001; Meinhold et al., 1996).

First, a simulation was used to determine the average concentration of Ra-226 in produced water, as well as, the concentration that remains in surface water after radionuclide decay and transportation processes. The software used to perform the simulations were Palisade's decision tools @Risk® and @StatTools⁷ and the Canadian Centre for Environmental Modelling and Chemistry's Quantitative Water Air Sediment Interaction model (QWASI).

The average Ra-226 concentration in produced water was calculated based on the Ba, Ca, and Sr contents in produced water from the Bakken formation. A multivariate regression model (hereinafter the Model) between Ra-226 and major minerals in produced water from different locations in the U.S. was developed. These locations were Louisiana, Mississippi, Pennsylvania, Texas, and others. The Model was used to predict Ra-226 concentration in produced water using data on major cations in produced water from oil wells in ND, which was obtained from the United States Geological Survey (USGS) Produced Water Database.

The next step was to determine the exposure to Ra-226 for each scenario. The result from the Model was used as the input in QWASI to simulate the fate of Ra-226 in a lake. The QWASI output (remaining concentration of Ra-226) was used in all the scenarios. In the first scenario, the amount of Ra-226 that remains in drinking water was calculated using the average removal efficiency of different water treatment methods. In the second scenario, food transfer factors (partition coefficients) specific to potato were used to calculate the amount of Ra-226 transferred from irrigation water to soil and then, from soil to the edible tuber product. In the third scenario, the concentration of Ra-226 in lake water that transfers to the fish muscle was estimated using food transfer factors specific to fishes. Using the estimated transference of Ra-226 concentrations from contaminated surface water to drinking water, potatoes, and fishes, the effective annual dose rate per capita was estimated. Finally, a sensitivity analysis was conducted to re-evaluate the results taking into account variations of $\pm 25\%$ and $\pm 50\%$ on the Ra-226 concentration in produced water simulated with the Model. The complete methodology is summarized in Figure 3.1.

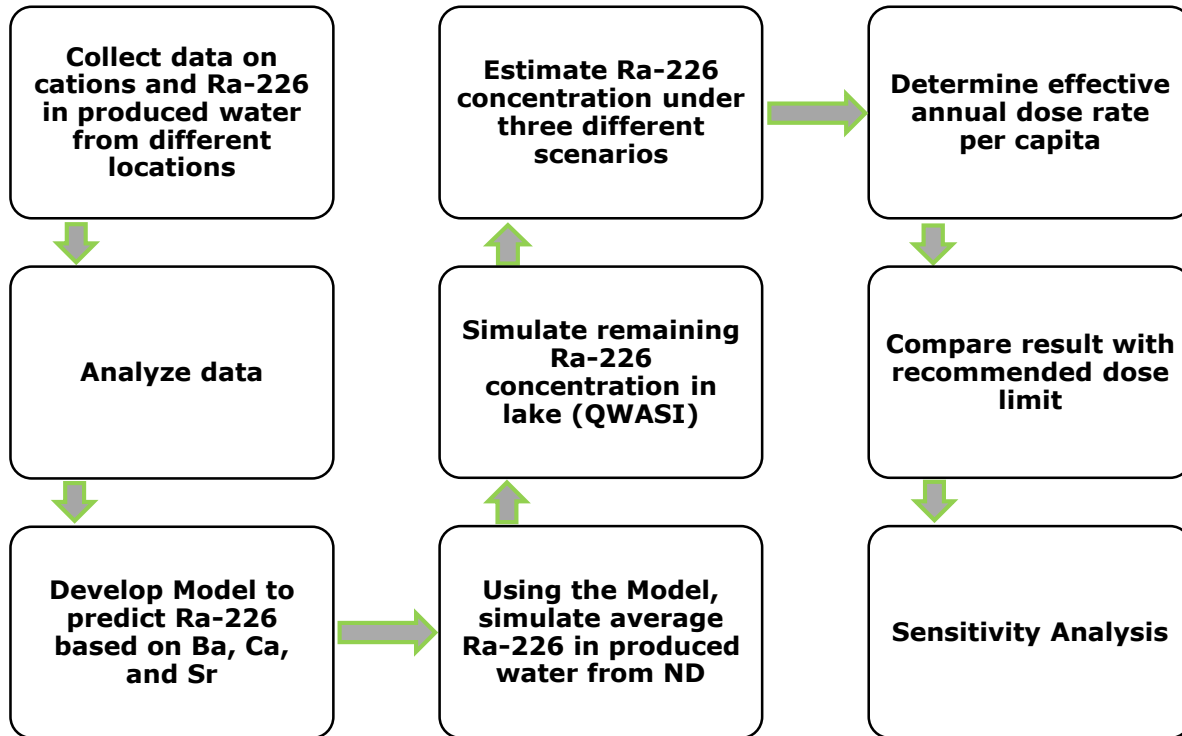


Figure 3.1. Process used to conduct risk assessment of Ra-226 human exposure.

3.2.2. Sources of data

The main data used for this research was the chemical contents of produced water from oil wells in ND collected from the latest version (v2.2) of the USGS Produced Water Database available online and Lauer et al. (2016). The produced water data from Mississippi and Pennsylvania was also obtained from the same USGS Database. The data from Louisiana, Texas, and others was from peer-reviewed journal articles because the USGS Produced Water Database only shows incomplete data on these States (Alley et al., 2011; Landa and Reid, 1983; Silva et al., 2012; Warner et al., 2013). The transfer factor for potatoes and fishes were obtained from multiple sources (Carvalho et al., 2009; Clulow et al., 1998; Hosseini et al., 2008; IAEA, 2001; IAEA, 1981; Meinhold et al., 1996; Pietrzak-Flis et al., 1995; Staven et al., 2003; Tagami and Uchida, 2009; Watson et al., 1983).

3.2.3. Determination of Ra-226 concentration in produced water from the Bakken

3.2.3.1. Regression model

Available data on Ra-226 content in produced water from the Bakken formation is very scarce. Because of this, a simulation Model was developed based on the correlation between radioactivity levels and major cations in produced water. Levels of Ra-226, Ba, Ca, and Sr from at least four different locations were used to develop a multivariate regression model. This Model was then used with the ND data to simulate the local average Ra-226 concentration. Table 3.1 summarizes the amount of data and sources used to develop the Model and to perform the simulation for ND.

Table 3.1. Produced water data for multivariate regression model and simulation.

Location	Datasets	Average values				Source
		Ba (mg/L)	Ca (mg/L)	Sr (mg/L)	Ra-226 (pCi/L)	
Louisiana	1	44	1,590	56	176	(Landa and Reid, 1983)
Mississippi	17	64	26,018	1,314	563	(Blondes et al., 2016)
Pennsylvania	40	1,151	13,437	1,925	1,244	(Blondes et al., 2016; Warner et al., 2013)
Texas	1	147	10,880	1,750	2,300	(Silva et al., 2012)
Various (max. value)	Not specified	7	52,920	2	262	(Alley et al., 2011)
ND	21	19	17,302	1,082	---	(Blondes et al., 2016; Lauer et al., 2016)

3.2.3.2. Ra-226 simulation for ND

The total number of data points (n=60 for each cation and Ra-226) were used to analyze the relations between Ba, Ca, and Sr (independent variables) and Ra-226 (dependent

variable) to determine how the independent variables influence the concentration of Ra-226. A multiple regression model (equation 3.1) was developed using logarithmic transformations of the average values of each cation (n=5) shown in Table 3.1, excluding ND. This was done with the purpose of reducing error, skewedness, and possible negative values for Ra-226 (Baker, 2006; Hopkins, 2003). The Model developed with Palisade's @StatTools7 is:

$$\mathbf{Log(^{226}Ra) = 1.625 + 0.168Log(Ba) + 0.308Log(Ca) + 0.208Log(Sr)} \quad (3.1)$$

For details on the development of the Model, please see Tables A1 and A2 in Appendix A. The constant value or percentage weight for each independent variable indicate the percentage change in Log(Ra-226) when the variable changes by 1% (Baker, 2006). In this case, there is a 16.80% increment in Log(Ra-226) for every 1% variation in Log(Ba). Likewise, for Log(Ca) and Log(Sr), the changes in Log(Ra-226) are 30.80% and 20.80%, respectively. This is in alignment with other research that found higher rates of radium co-precipitation with Ba than with Sr which means that Sr presence increases radium concentration in solution (Al-Masri and Aba, 2005; Wilson, 2012; Zhang et al., 2014). Also, TDS has a strong, positive correlation with Ra-226 and because most of the TDS content comprises inorganic salts including Ca, the percentage weight of Ca in the Model is the highest of the three cations (Fisher, 1996; Rowan et al., 2011; Tinto and Solomon, 2012).

The Ba, Ca, and Sr datasets (each containing 21 data points) from ND were analyzed with Palisade's @Risk to determine the type of distribution each dataset follows. By using the probability distribution instead of the average values in the Model, the variability in the data was captured and more realistic values were obtained. The results indicate that Ba follows triangular distribution and Ca and Sr Weibull distribution. For details on the type of distribution for each dataset, please see Appendix A, Tables A3 and A4.

The @Risk software was used to simulate 100,000 times the level of Ra-226 taking into account the distribution of each independent variable in Equation 3.1. To determine the central tendency of the simulated Ra-226 values, the median was used instead of the mean.

The median is preferred over the mean when the data is skewed, which was the case in this study. The median of the Ra-226 simulation was 535 pCi/L (see Figure A1 in Appendix A). This concentration falls in the range of 527-1,210 pCi/L (n = 3) found by Lauer et al. (2016), the only study to date that reports Ra-226 levels in produced water samples from the Bakken Shale.

3.2.4. Exposure assessment

The next step in the analysis was to assess the human exposure to Ra-226 via food chain. The assessment focused on calculating the amount of Ra-226 transferred from surface water contaminated with produced water to drinking water, potato skin, and fish. The simulated Ra-226 concentration in produced water of 535 pCi/L (obtained in subsection 3.2.3.2) was used. In addition, scientific peer-reviewed literature values of transfer factors were applied (Carvalho et al., 2009; Clulow et al., 1998; Hosseini et al., 2008; IAEA, 2001; IAEA, 1981; Meinhold et al., 1996; Pietrzak-Flis et al., 1995; Staven et al., 2003; Tagami and Uchida, 2009; Watson et al., 1983).

When a contaminant enters an aquatic environment, many physical and chemical processes occur, and attenuate and/or transform the contaminant. This is the case for Ra-226. The QWASI model was used to determine the concentration that remains in a lake after a spill or produced water enters a surface water body. For this simulation, the characteristics of Lake Sakakawea located in the Missouri River basin in central ND were used. This lake was considered in the simulation analysis because it is a major source of surface water for several towns in the State, is located near oil well production sites, and is one of the top fishing spots in ND (City of Williston, 2014; ND DoH, 1999b; ND Tourism Division, n.d.).

The Ra-226 concentration in air was not included in the QWASI analysis since the data is not collected in ND (Otto, 2016). Contribution through atmospheric deposition is assumed to be negligible based on secular equilibrium between the parent radionuclide Ra-226 and the daughter radionuclide radon-222 (Rn-222) (Argonne National Laboratory, 2005). This means

that the activity of Ra-226 is slightly different than Rn-222, thus, assumed equal and because previous research found low levels of Rn-222 over surface water, the contribution of atmospheric Ra-226 was not included in the analysis (Jasaitis et al., 2016).

Transfer factors are used to estimate the amount of contaminant bioaccumulated in organisms such as plants and animals (Staven et al., 2003). Ra-226 may transfer to agricultural products through two routes, irrigation water deposition and root uptake from soil (Pacific Northwest National Laboratory, 2008). Therefore, the total concentration of Ra-226 in plants can be calculated by adding Ra-226 concentrations calculated by Equations 3.2 and 3.3 based on water-to-plant transfer factor and soil-to-plant transfer factor, respectively. (Carvalho et al., 2009; Pacific Northwest National Laboratory, 2008):

$$TF_{water-plant} = \left(\frac{Ra-226 \text{ concentration in plant in } Bq \text{ kg}^{-1} \text{ dry weight}}{Ra-226 \text{ concentration in water in } Bq \text{ L}^{-1}} \right) \quad (3.2)$$

$$TF_{soil-plant} = \left(\frac{Ra-226 \text{ concentration in plant in } Bq \text{ kg}^{-1} \text{ dry weight}}{Ra-226 \text{ concentration in soil in } Bq \text{ kg}^{-1} \text{ dry weight}} \right) \quad (3.3)$$

Similarly, the amount of Ra-226 bioaccumulated in the fish tissue can be determined using the transfer factor measured in fresh weight animal product as follows (Staven et al., 2003):

$$TF_{water-fish} = \left(\frac{Ra-226 \text{ concentration in fish in } Bq \text{ kg}^{-1} \text{ fresh weight}}{Ra-226 \text{ concentration in the water in } Bq \text{ L}^{-1}} \right) \quad (3.4)$$

3.3. Results and discussion

3.3.1. Exposure assessment

3.3.1.1. Scenario 1: Drinking water

The first scenario covers previous actual cases in ND in which produced water spills have reached a surface water body. For example, there was a produced water spill in William County in January 2015 where 3×10^6 gallons were discharged from which a fraction (unknown) affected the Blacktail Creek and Little Muddy River. This event poses human health risks because both water bodies discharge into the Missouri River, a drinking water source

(Lauer et al., 2016). To determine the concentration of Ra-226 that remains in an impacted surface water body, the QWASI model and the characteristics of Lake Sakakawea were used. QWASI calculates the concentrations in the air, water, and sediment compartments of a lake based on the fugacity concept. For details on QWASI, please see Mackay et al. (2014); Webster et al. (2005); Woodfine et al. (2000); and Mackay and Diamond (1989). Several modifications for Ra-226 were required because fugacity uses measurable vapor pressure (please see Figures A2-A8, Appendix A).

Based on the QWASI analysis, an initial Ra-226 concentration of 535 pCi/L (previously calculated in section 3.2.3.2) would result in a final concentration of 125.64 pCi/L (1.27×10^{-1} ng/L). If a lake containing 125.64 pCi/L is used as a drinking water source, the ultimate concentration after water treatment process for radionuclides removal (88% average removal efficiency of lime softening, ion exchange, and activated carbon) will be 15.08 pCi/L or 0.56 Bq/L (Becquerel per liter) (EPA, 2015). This amount is more than three times the maximum contaminate level (MCL) of 5 pCi/L (combined Ra-226 and Ra-228) allowed in drinking water established by the EPA (Agency for Toxic Substances and Disease Registry, 1990).

3.3.1.2. Scenario 2: Potato skin bioaccumulation

In this scenario, the amount of Ra-226 transferred from irrigation water and soil to potatoes was calculated. Potato was selected because of its high consumption in the State compared to other products. Transfer factors are site specific but for this study, several ratios for water-to-plant and soil-to-plant found in the literature were considered (Tables A5 and A6, Appendix A). The transfer factors from previous research used for this analysis were calculated using soils with similar characteristics to the ones found in ND. Table 3.2 shows the transfer factors, Ra-226 concentration in irrigation water, and Ra-226 concentration in soil used to determine the total amount of Ra-226 bioaccumulated on the potato skin.

Table 3.2. Data used to calculate Ra-226 bioaccumulation in potatoes.

Parameter	Values		Source	Distribution
Water-to-Plant Transfer Factor (Bq/kg per Bq/L)	Mean	4.70	(Carvalho et al., 2009)	Normal (assumed)
	Std. Dev.	0.14		
Soil-to-Plant Transfer Factor (Bq/kg per Bq/kg soil)	Mean	5.8×10^{-3}	(Carvalho et al., 2009; IAEA, 1981; Pietrzak-Flis et al., 1995; Staven et al., 2003; Tagami and Uchida, 2009; Watson et al., 1983)	Normal (assumed)
	Std. Dev.	1.8×10^{-3}		
Ra-226 in contaminated irrigation water	125.64 pCi/L		This study (Scenario 1)	N/A
	4.65 (Bq/L)*			
Ra-226 in soil (Bq/kg)	Mean	90.70	(Lauer et al., 2016)	Inverse Gaussian (@Risk best fit)
	Shape factor	16.70		

* 1 pCi = 0.037 Bq.

Some considerations for this scenario are:

- Water-to-plant transfer factor: Due to limited information, only two data points from Carvalho et al. (2009) were used.
- Soil-to-plant transfer factor: Assumed normal distribution based on mean and standard deviation from seven data points found in the literature (Carvalho et al., 2009; IAEA, 1981; Pietrzak-Flis et al., 1995; Staven et al., 2003; Tagami and Uchida, 2009; Watson et al., 1983).
- Ra-226 concentration in soil: 21 data points from Lauer et al. (2016) fitted using @Risk best fit tool.

The data in Table 3.2 and the considerations listed above were used to calculate the amount of Ra-226 that could accumulate on the potato skin. @Risk was setup to run 100,000 iterations which resulted in a final Ra-226 concentration of 22.38 Bq/kg dry weight of potatoes. For comparison, the natural radioactivity levels in white potatoes is 0.05-0.10 Bq/kg (1.0-2.5 pCi/kg) (Idaho State University, 2011)

3.3.1.3. Scenario 3: Fish tissue bioaccumulation

A similar process used in scenario 2 was applied in scenario 3. Using a Ra-226 concentration of 125.64 pCi/L (previously calculated in section 3.3.1.1) in a lake used for recreational fishing such as Lake Sakakawea, the amount bioaccumulated in fish was calculated. For this case, six different transfer factors found in the literature (Clulow et al., 1998; Hosseini et al., 2008; IAEA, 2001; Meinhold et al., 1996) (Table A7, Appendix A) were used to estimate the mean value for the transfer factor for fishes (6.60 Bq/kg per Bq/L) and standard deviation (3.30 Bq/kg per Bq/L) which were used in the simulation. Assuming the transfer factor follows a normal distribution, @Risk was used to perform 100,000 iterations of Equation 3.4 to calculate the transfer of Ra-226 to fishes. The simulation results indicate that the total amount bioaccumulated in the fish tissue is 30.68 Bq/kg fresh weight. As a comparison, the global concentration of radioactivity in fishes is 2.4 Bq/kg fresh weight (Aarkrog et al., 1997).

3.3.2. Risk assessment

The amount of Ra-226 transferred from oil produced water to drinking water and food calculated in the previous section can be further analyzed to determine the radiation exposure of the local population. There is plenty of reports and guidelines from national and international organizations (e.g. the EPA, the International Atomic Energy Agency, and the World Health Organization (WHO)) dedicated to analyze human health risks associated with radioactive material. For the purposes of this study which only focuses on three foodstuffs, a simplified process was used to calculate the amount of Ra-226, measured in millisievert

(mSv), entering the human body via food chain. Table 3.3 summarizes the data used to calculate the annual dose received by adults in ND.

Table 3.3. Data used to calculate annual Ra-226 dose rate per individual in ND.

Parameter	Scenario 1	Scenario 2	Scenario 3
	Drinking Water	Potatoes with Skin	Fish
(1) Consumption rate per year	730 L ^a	48.40 kg (US Department of Agriculture Economic Research Service, 2015)	8.43 kg ^b (EPA, 2013)
(2) Effective ^c dose/unit intake via ingestion	2.8×10^{-7} (Sv/Bq) (IAEA, 2014)		
(3) Ra-226 concentration simulated in this study	0.56 Bq/L	22.38 Bq/kg	30.68 Bq/kg
(4) Annual Ra-226 dose rate ^d (mSv)	0.11	0.30	0.08

^a Assumed 2 liters per day.

^b Average adult weight in the US = 82 kg (Centers for Disease Control and Prevention, 2016).

^c Committed or received dose.

^d (1) × (2) × (3) = (4).

The total annual Ra-226 dose rate that an adult in ND would receive if he/she consumes water, potatoes and fish that have been in direct contact with surface water contaminated with produced water is 0.49 mSv. The global average annual human exposure to radiation (different radionuclides besides Ra-226) from natural sources is 2.40 mSv/year from which 0.30 mSv is due to ingestion of food and drinking water (United Nations Scientific Committee on the Effects of Atomic Radiation, 2000). If the scenarios were considered individually, none of the estimated annual dose rate exceeds the global average. However, by considering a diet including the three foodstuffs evaluated, the total contribution of Ra-226 via food chain is higher than the global average. The interpretation of these results

indicate that the level of risk of human exposure to Ra-226 could be high if produced water in ND is not handled correctly.

3.3.3. Sensitivity analysis

To analyze the effects of possible underestimation (25% and 50%) or overestimation (-25% and -50%) of the actual Ra-226 concentration in produced water, a sensitivity analysis with four different scenarios was conducted. Table 3.4 shows a comparison between the results obtained with the base concentration of 19.80 Bq/L (535 pCi/L) and the four different variations in the sensitivity analysis.

Table 3.4. Sensitivity analysis results. $\pm 25\%$ and $\pm 50\%$ scenarios evaluated compared to based concentration.

Levels of Ra-226	Base	-25%	+25%	-50%	+50%
Concentration in produced water (initial) (Bq/L)	19.80	14.85	24.75	9.90	29.70
Concentration in lake (final) (Bq/L)	4.65	3.70	6.19	2.50	7.39
Concentration in drinking water (Bq/L)	0.56	0.44	0.74	0.30	0.89
Concentration in potato (skin) (Bq/kg)	22.38	17.92	29.62	12.28	35.26
Concentration in fish (Bq/kg)	30.68	24.42	40.85	16.50	48.77
Annual dose rate (mSv)	0.49	0.40	0.67	0.27	0.79

The sensitivity analysis revealed that variation in the simulated Ra-226 concentration in produced water (535 pCi/L) by $\pm 25\%$ and $\pm 50\%$ still presents risks to human health. The concentration of Ra-226 that would remain in drinking water exceeds EPA's MCL (5 pCi/L or 0.19 Bq/L) in all the sensitivity analysis scenarios. In addition, the annual dose rate exceeds the average annual human exposure to radiation through food and drinking water ingestion (0.30 mSv) in all scenarios except in the scenario where the Model overestimated the actual

level of Ra-226 in produced water by 50% (9.90 Bq/L or 268 pCi/L) which is considerably below the low range reported by Lauer et al. (2016) (527-1,210 pCi/L, n = 3).

3.4. Summary

The concentration of Ra-226 in produced water varies based on several factors including geological characteristics of the Bakken Shale. A Model was developed to predict Ra-226 using the known levels of the most common cations present in produced water and the result is agreeable with a range reported by the only experimental study available with extremely limited data. This study goes beyond the calculation of Ra-226 in produced water by estimating the risks to human health in the event that produced water reaches a surface water body that is used as a source for drinking water, irrigation water, and recreational fishing. A proper risk assessment considers the complete diet of the population as well as other sources of radionuclides exposure such as inhalation. However, the results which only take into account the contribution of Ra-226 via ingestion of water, potatoes, and fish indicate that the levels are above the average annual radiation dose from natural sources. In addition, the sensitivity analysis shows that there could be risks to human health even when the Ra-226 levels in produced water vary significantly compared to the simulated concentration.

With the Model, the final concentrations of Ra-226 can be calculated for any sample of produced water from the Bakken Shale if major cation levels are known. As more data becomes available, the Model can be refined to provide more accurate results. In addition, the analysis methods applied in the study could be used for any other cases in the U.S. where unconventional oil and gas production is being practiced with a few modifications including the specific characteristics of produced water, the impacted surface water body, and biota consumption rates/types. Overall, the results presented in this study can be treated as a warning and a reference to conduct further investigations. Studies such as the one presented here can greatly contribute to understand the health risks associated with unconventional oil

and gas production and develop risk management strategies to mitigate risks of introducing contaminants from produced water into the human food chain.

4. HOLISTIC RISK ASSESSMENT OF SURFACE WATER CONTAMINATION DUE TO Pb-210 IN OIL PRODUCED WATER FROM THE BAKKEN SHALE

4.1. Introduction

Oil produced water generated during the flowback process and production stage contains different constituents including chemical additives, salt, oil and grease, natural organic and inorganic compounds, and natural radioactive materials. Leakage, spills, and discharges of produced water present a threat to human health and the environment because of the relatively high concentrations of these contaminants (Guerra et al., 2011; Vengosh et al., 2014; Vidic et al., 2013). Research on water contamination due to oil and gas production has been conducted on specific constituents, such as salinity and organic compounds, but studies about radioactive materials are still scarce (Barbot et al., 2013; Paschoa, 1998). This is especially true for ND, the second largest oil producing State in the country (Argonne National Laboratory, 2014; Scanlon et al., 2014).

NORM is found in geologic formations and distributed naturally while the technologically enhanced NORM (TENORM) is the result of human activities, such as O&G production, which transport the radionuclides and increase the concentrations and the probability of exposure (Rich and Crosby, 2013). Some underground geological reservoirs exploited to produce O&G in the U.S. contain uranium-238 (U-238) and thorium-232 (Th-232) which are sources for TENORM (Argonne National Laboratory, 2014).

U-238 is naturally found in the environment and its decay series includes Pb-210 along with Rn-222 and polonium-210. Stable isotopes of Pb are found in both terrestrial and freshwater ecosystems but their concentrations vary based on the location (Tamponnet, 2009). Once the radioactive material reaches the environment, there are different pathways to human exposure such as surface and shallow groundwater contamination. The fate and transport of Pb-210 in the environment have not been studied as widely as other radionuclides (Argonne National Laboratory, 2014; Tamponnet, 2009).

Previous TENORM studies have focused mostly on Ra-226, Ra-228, and Rn-222, which are the predominant radioisotopes present in natural gas production liquid and solid wastes (Godoy and Petinatti da Cruz, 2003; Rich and Crosby, 2013; Rowan et al., 2011). The radioactivity levels in produced water vary from background values to thousands of pCi/L and usually Ra-226 is found in high concentrations (Chriss and Bursh, 2002). Because of this, State regulations typically are based upon total Ra (Ra-226 plus Ra-228) (Argonne National Laboratory, 2014). However, Pb-210 and other radionuclides have been found in O&G production wastes but there is limited information about potential radiation exposure to workers and general public (Rich and Crosby, 2013).

Produced water quality varies according to several factors including geological formation, location, depth, stimulation method, and the chemicals applied. According to previous research, there is a strong correlation between TDS concentration and Ra activity in produced water from reservoirs with similar lithological characteristics (Fisher, 1998; Godoy and Petinatti da Cruz, 2003; Rowan et al., 2011). Fisher (1998) found that TDS levels higher than ~35,000 mg/L result in high radium activity of more than ~100 pCi/L. Produced water from the Williston Basin, where the Bakken Shale is located, is known to have some of the highest TDS concentrations in the U.S. ranging from a few hundreds to up to 632,700 mg/L (Gleason and Tangen, 2014). Based on Fisher (1998), radioactivity in produced water from the Bakken Shale should be high due to elevated salinity.

In 2014, the Argonne National Laboratory published a report entitled "Radiological Dose and Risk Assessment of Landfill Disposal of Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) in North Dakota" in response to a request by the ND DoH to support possible changes in the regulations regarding TENORM (Argonne National Laboratory, 2014). Based on this report, regulations on solid waste could be safely modified as long as the maximum exposure to landfill workers does not exceed 100 millirem per year (Argonne National Laboratory, 2014). The report covers the human health risks of Ra-226,

Ra-228, Pb-210, and Th-232 in solid wastes but excludes produced water. Very little research related to Pb-210 in produced water has been conducted despite that Pb is known for its harmful effects, especially on children, such as damage to the brain and nervous system, slowed growth and development, learning and behavior problems, and hearing and speech problems (Centers for Disease Control and Prevention, 2013).

This research focuses on assessing the risks of surface water contamination caused by spills of produced water from the Bakken formation containing Pb-210. To achieve this, historical data on produced water from ND was analyzed to simulate the radiation levels coming from Pb-210 because this information is not publicly available. With this data, a holistic risk assessment of surface water contamination due to Pb-210 was performed considering three different scenarios: (1) storage tank overflow, (2) leakage in equipment, and (3) spills related to trucks used to transport produced water. Simulation was applied in this study by using Palisade's decision tool @Risk® and the Canadian Centre for Environmental Modelling and Chemistry's QWASI model. In addition, social risk perception and awareness of produced water risks of different stakeholders in ND were measured. The objectives of this study are:

- To simulate average Pb-210 concentration in produced water from the Bakken Shale.
- To quantify the risks of surface water contamination due to Pb-210 under three different scenarios.
- To perform a holistic risk assessment by incorporating the results of a survey that measures risk awareness and perception of the issue.

4.2. Methodology

4.2.1. Method overview

A risk assessment focused on three scenarios that could result in surface water contamination was conducted (Figure 4.1). The first scenario is storage tank overflow that reaches surface water that is nearby the oil well pads. The second scenario is leakage in pipes, storage units, and other equipment, that reaches surface water. The third scenario is

accidents related to trucks transporting produced water which cause spills that reach surface water. These scenarios were selected because they were found to be common in reported spills and media coverage in ND. Other scenarios that could contribute to surface water contamination such as runoff and melting snow were not considered because of supporting data unavailability and high variability of environmental conditions.

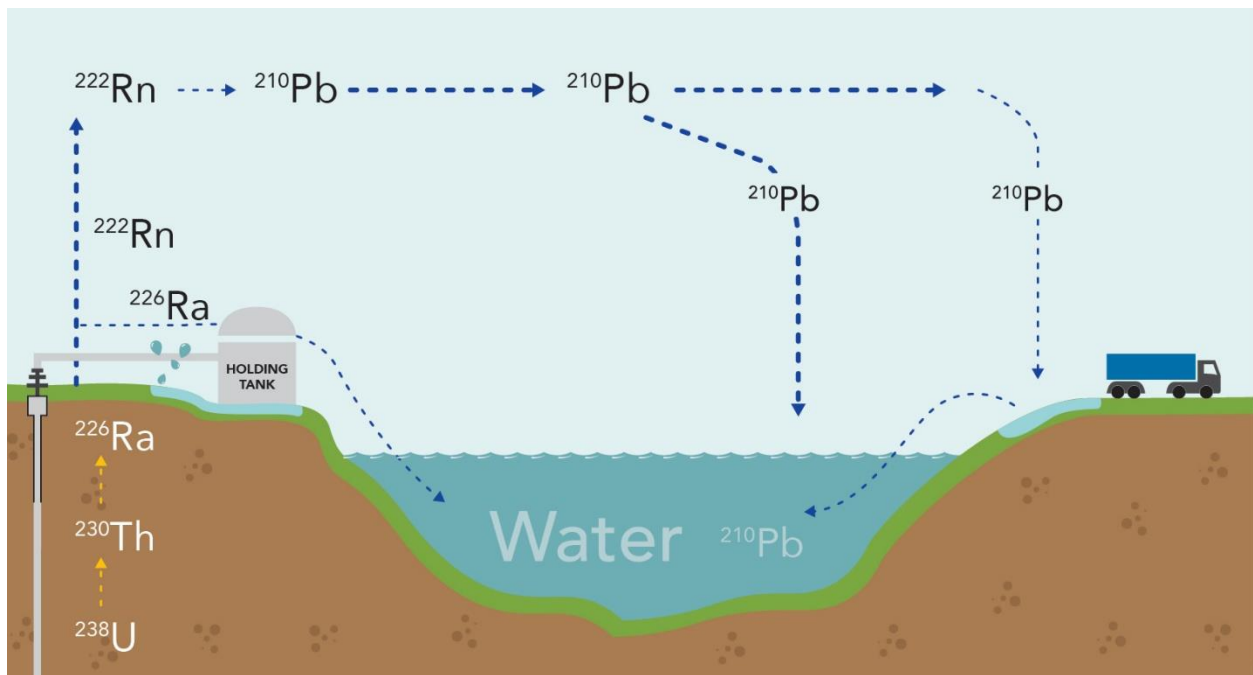


Figure 4.1. Scenarios that could result in surface water contamination due to Pb-210.

This research uses a combination of stochastic and deterministic methods to analyze the risks of Pb-210 in each scenario. These methods were combined because of the variability in the type of data used. First, the historical TDS data, reported spills, and truck crashes were analyzed using Palisade's @Risk® tool. Second, the Ra-226 level in produced water from the Bakken Shale was calculated based on a generalized linear regression model between observed TDS and Ra-226 levels in produced waters from different locations including Pennsylvania, Texas, New York, and Wyoming.

Third, the Rutherford-Soddy's decay law was used to simulate the decay of Ra-226 into Pb-210. The probability distribution parameters of Ra-226 were used to capture the variability in the data due to different factors which could not have been considered if point estimates were used. The predicted Pb-210 concentration was used to assess the exposure by oral ingestion. The exposure concentration was estimated using the QWASI model that simulates chemical fate in lakes. Furthermore, a survey was distributed in several counties in ND to collect data from different stakeholders including general public, oil field and truck operators, and emergency management personnel and/or individuals in charge of produced water management (hereinafter experts). The data was analyzed to quantify awareness and perception of the risks of each scenario where produced water could reach surface water. The results from the survey were included in the risk characterization of each scenario.

Finally, a semi-quantitative risk assessment matrix was used to translate the results obtained in the previous steps of the risk assessment into quantitative measurement based on low, medium-low, medium-high, and high risk. Also, a sensitivity analysis was carried out to evaluate the variation in the Pb-210 levels if the simulated Ra-226 concentrations were underestimated or overestimated by 25% to 50% compared to actual levels. The entire methodology is summarized in Figure 4.2.

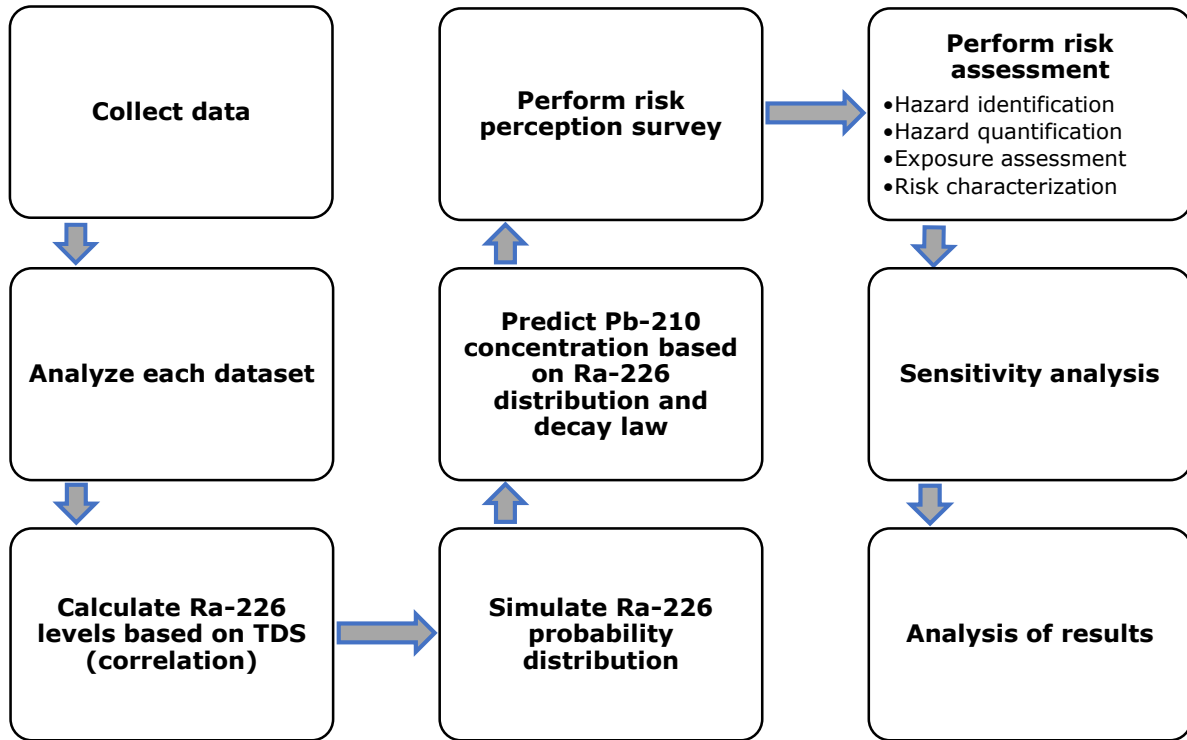


Figure 4.2. Process followed to perform a holistic risk assessment of Pb-210 surface water contamination.

4.2.2. Sources of data

The chemical quality, particularly TDS, of produced water in the Bakken was obtained from the USGS Produced Water Database (Blondes et al., 2014). Please see Table B1 in Appendix B for sample TDS data. Also, the environmental incident reports available at the ND DoH website were used (ND DoH, 2015). This dataset is a collection of reported oilfield environmental incidents that caused any discharge of liquids or solids that might cause pollution of water bodies. The information used in this study is associated with saltwater spills from October 7, 2014 to October 5, 2015 (Table B2, Appendix B). In addition, the ND Department of Transportation (ND DoT) report called “2014 North Dakota Crash Summary” was analyzed to collect information regarding truck crashes in the State (Figures B1 and B2, Appendix B) (ND DoT, 2015).

4.2.3. Determination of Pb-210 concentration

There were two steps in determining Pb-210 concentration. Ra-226 concentration was estimated and then Pb-210 was calculated based on the decay of Ra-226. In ND, data on radium in produced water has not been reported in the USGS Produced Water database and only one study by Lauer et al. (2016) has measured Ra-226 in produced water from the Bakken but the data was based on only three samples. Because of this, it is necessary to use a simulation approach to determine the radium concentration. Since there is no available data on Ra-226 from the Bakken Shale, three different datasets from previous studies were analyzed (Table B3, Appendix B). The first dataset, which comes from Pennsylvania and New York, was obtained from Rowan et al., 2011. The second set, which is from Texas, was found in Fisher (1995). Lastly, the third set, which is from Pennsylvania and Wyoming, was retrieved from the USGS produced water database for the wells that were monitored for TDS and Ra-226 levels. Using a total of 331 data points from the three datasets a linear regression model with an R-squared value of 0.80 (Tables B4 and B5, Appendix B) was developed as follows:

$$\mathbf{Log(^{226}Ra) = 1.12 \times Log(TDS) - 3.33} \quad (4.1)$$

This generalized model (hereinafter the Model) was used to simulate the Ra-226 levels for the Bakken. The Model is more representative because it captures variability introduced by several factors from different locations and hence provides a more generalized pattern. It is important to note that the results obtained with the Model are estimates since its parameters were calculated from historical data and for this reason, treated as random variables. The observed Ra-226 data in ND by Lauer et al. (2016), which is very limited (three produced water samples), will be used to validate the Model results.

According to the Rutherford-Soddy's decay law, the atoms of a radioactive substance change randomly from a parent element to another daughter element at a specific rate (Braun, 1983). The decay constant λ indicates how fast the substance decays or disintegrates which can be measured by the half-life ($t_{1/2}$) of the isotope, or the time it takes for half of the

original quantity of isotope to decay (Braun, 1983). The fraction of atoms that disintegrate after time t can be expressed as:

$$N(t) = N_0 e^{-\lambda t} \quad (4.2)$$

Where N_0 is the amount of atoms at $t = 0$ and $\lambda = \frac{\ln 2}{t_{1/2}}$

Depending on the activity relationship between the parent compound and the daughter product, there can be no equilibrium, transient equilibrium, or secular equilibrium (Prince, 1979). When Ra-226 decays, it does not disappear but changes into new nuclear species including Pb-210. Because the half-life of Ra-226 (1,600 years) is much longer than the half-life of Pb-210 (22 year), they are in secular equilibrium (Prince, 1979). During the lifespan of an oil well in the Bakken, which is approximately 30 years, there is no significant change in Ra-226 and it is constantly feeding Pb-210 (Braun, 1983; ND Petroleum Council, 2012). However, Ra-226 is unsupported because its parent, U-238, remains in the reservoir (Smith, 2011). Because of the significant difference between the half-lives of Ra-226 and Pb-210, the activity of both are the same over time meaning that the Pb-210 concentration at time t (N_t) can be determined using the concentration of Ra-226 at $t = 0$ (N_0).

4.2.4. Social perception

Risk assessment from an engineering or technical point of view alone might deliver biased results (Perry, 2012). This is because risk measurement is usually narrowed down into probabilities and consequences, leaving aside the public awareness of risk (Leiserowitz, 2005; Slovic, 2000). Produced water spills are mostly caused by human errors. As a result, the perception of risk can be a critical factor for quantifying risk level. A holistic approach to analyze risk incorporates risk perceptions which are created based on different dimensions including technical knowledge, psychological and social factors, and personal experiences (Leiserowitz, 2005; Slovic, 2000). It is believed that risk perception influences risk behaviors, meaning that the more aware a person is about a specific risk the more protective actions the person is willing to take and vice versa (Slovic and Peters, 2006a; Xiaohao et al., 2010).

A survey was developed to collect risk perception of different stakeholders in ND including the general public, operators, and experts. The survey was used to obtain data regarding the knowledge, opinions, and concerns of different stakeholders affected directly and indirectly by the oil production in the State. The survey questions were focused on determining the risk awareness and perception, and influence of produced water management on human behavior or action. The survey was conducted completely online where the participants were asked to answer 21 to 25 questions, depending on their responses, in a time period of 5 to 10 minutes. The regions of interest were Cass county (where Fargo, ND is located) and the 17 oil-producing counties in the State. These counties are: Billings, Bottineau, Bowman, Burke, Divide, Dunn, Emmons, Golden Valley, McHenry, McKenzie, McLean, Mountrail, Renville, Slope, Stark, Ward, and Williams (ND Petroleum Council, 2015).

4.2.5. Holistic risk assessment

The steps followed for the assessment include hazard identification, hazard quantification, exposure assessment, and finally, risk characterization. Hazard identification set the relation between scenarios where produced water could be discharged and the initiating events or hazard that could cause those scenarios. Hazard quantification established the probability of each scenario occurring. The exposure routes to Pb-210 can be through oral ingestion, dermal contact or inhalation (Emmanuel et al., 2007). In general, about 80% of the Pb-210 internal exposure in humans is through ingestion and 20% through inhalation but this can vary depending on the location (Jaworowski, 1969; Tamponnet, 2009). In the risk characterization, the data obtained in the first three steps was linked and the social risk perceptions and awareness about produced water from different stakeholders were included.

4.3. Results and discussion

4.3.1. Analysis of data

The historical data of TDS (n=7,907) in the Bakken was analyzed using Palisade's @Risk® which showed that the data follows a distribution with a mean equal to 264,512 mg/L

and a standard deviation of 76,754 mg/L. From all the spills that occurred from October 7, 2014 to October 5, 2015, there were 609 contained and 112 uncontained spills and the volume ranged from 42 gallons to 2,940,000 gallons (1 - 70,000 barrels) (ND DoH, 2015). The average volume for uncontained spills is 2,757 gallons without taking into account a major incident which released almost 3 million gallons of produced water in Williams County in January of 2015. This incident was treated as an extremely large rare event and thus, not included to avoid the influence of extreme rare events or outliers.

From 2009 to 2014, there were 3,283 crashes involving trucks and 6,029 crashes involving truck tractors with average annual increments of 17.4% and 16.6%, respectively (Figure B1 in Appendix B) (ND DoT, 2015). These numbers include other accidents besides those associated with produced water transportation. The ND DoT was contacted to obtain data regarding the distribution of these accidents in the different Counties in the State (R. Hair, personal communication, August 21, 2016). From the total amount of crashes occurred in the State in 2014, 5.5% involved truck units and more than 60% of these accidents took place in the major oil-producing counties in the State (Figure B2 in Appendix B).

4.3.2. Pb-210 simulation

The TDS data points obtained (n=7,907) from the Bakken were plugged in the Model to calculate the Ra-226 concentrations (n=7,907) and then, this dataset was fitted using @Risk® best fit tool (Table B4, Appendix B). The results from the analysis indicate that the simulated Ra-226 data follows a distribution with a mean equal to 557 pCi/L and a standard deviation of 179 pCi/L (Figure B3, Appendix B). The average Ra-226 value falls within the range (527-1,210 pCi/L) found by Lauer et al. (2016) based on three produced water samples in ND. Because the mean and the standard deviation capture the Ra-226 variability in the data, it was important to include these parameters in the Rutherford-Soddy's decay law to calculate the Pb-210 concentration. For this analysis, the initial Pb-210 concentration was assumed to be equal to the Ra-226 concentration ($N_0 = {}^{226}\mathbf{Ra}$) due to secular equilibrium and

after one year of produced water generation ($t = 1$), half-life of 22 years for Pb-210, and $\lambda = \frac{\ln 2}{22}$ then, equation (4.2) is now:

$$^{210}\text{Pb} = ^{226}\text{Ra} \times e^{-\lambda(1)} \quad (4.3)$$

Equation (4.3) was used to simulate Pb-210 in @Risk® (Figures B4 and B5, Appendix B) which estimated an average concentration of 540 pCi/L. The concentration is the same for the three scenarios under evaluation assuming all produced water spills had the same chemical composition. In addition, the @Risk® tool was used to create a probability distribution (Figure B6, Appendix B) of Pb-210 using the results from 100,000 Monte Carlo iterations and 1,000 bootstrap samples. The Monte Carlo method and bootstrapping were used to obtain better estimations.

4.3.3. Risk assessment

The first step is hazard identification. Hazard events identified for each scenario under evaluation are:

- Storage tank overflow: Storage capacity limitation, human error, and heavy rain or storm.
- Equipment leakage: Equipment failure and human error.
- Truck accident during transportation of produced water: Insufficient storage capacity, human error, road conditions, and weather conditions.

Incident reports available on the ND DoH website were used for hazard quantification. From all the incidents related to produced water spills, contained and uncontained, reported on the website from October 7, 2014 to October 5, 2015, 70% was due to leakage, 15% was due to storage unit overflow, 9% was unspecified incident, and 7% was related to truck incidents (ND DoH, 2015). The leakage incidents were mostly caused due to equipment failure including pipeline integrity, valve/piping connections, stuffing box, and treater leaks. Only one of the spill reports indicated that the produced water spill was caused by a truck rolling

into a ditch while the rest of the spills classified as truck incident were caused by truck overflow and operator error.

Based on the ND DoH reports, the total number of spills that reached surface water was very low. Less than 3% indicated that the spilled water was in contact with surface water. From this fraction of spills, less than 6% was caused by storage tank overflow, almost 89% by leakage, and 6% by truck incident. These numbers are based on historical data and should not be treated as predictions. The average volume of produced water that reached a water body in each scenario was 420 gallons due to storage unit overflow, 23,562 gallons due to leakage (without taking into account the spill in Williams County in Jan. 2015), and 1,680 gallons by truck incident. However, it is important to clarify that among the water bodies that were in contact with the produced water were small pools, riffle streams, stock ponds, and slough water which are not considered sources for drinking water. It should be noted that the reports do not indicate whether these small water bodies are directly connected to drinking water sources. Only the incident in Williams County in January 2015, where almost 3 million gallons were spilled, impacted a drinking water source. The spill affected the Blacktail Creek and Little Muddy River which flow into the Missouri River, a drinking water source (Lauer et al., 2016).

In the second step of the risk assessment, the exposure to Pb-210 by oral ingestion was calculated based on the average initial concentration of 540 pCi/L determined in section 4.3.2. The exposure assessment was based on the scenario where produced water is accidentally spilled and reaches a surface water body that serves as human drinking water source. The actual exposure to Pb-210 is the concentration that remains after the contaminant undergoes different chemical and physical processes, which happen naturally, and after the surface water is treated by different methods.

When Pb-210 reaches a lake, several interactions, which change the initial amount of contaminant, occur. A lake system consists of three different compartments which are water,

sediments, and air (Mackay and Diamond, 1989). Environmental fate models have been developed and used for the past decades to understand the fate and transport potential of chemicals in each of the compartments (Woodfine et al., 2000). For this analysis, the QWASI spreadsheet model version 1.00 was used due to its simplicity to illustrate the variation of Pb-210 concentration once it reaches the environment.

The QWASI model uses the fugacity concept, which is suitable for chemicals with measurable vapor pressure, but since metals, such as Pb, have zero or negligible vapor pressure some modifications are required (Mackay et al., 2014). As a basic explanation, fugacity is the tendency of a chemical of escaping from the phase it is in (i.e. solid, liquid, or gas) (Valsaraj and Melvin, 2000). The processes simulated with QWASI include advective flow, sediment deposition, resuspension, burial, and sediment-water diffusion (Woodfine et al., 2000). Details on the fugacity model and the equations used in QWASI are available in Mackay et al. (2014), Webster et al. (2005), Woodfine et al. (2000), and Mackay and Diamond (1989).

For the exposure scenario where produced water reaches a surface water body, the characteristics of Lake Sakakawea were used. This lake was chosen because it is a major source of surface water for several cities in ND including Dickinson, Williston, Garrison, and Parshall (City of Williston, 2014; ND DoH, 1999), and because it is located close to the regions with high density of oil wells. Table B5 in Appendix B summarizes the inputs and the sources used in the simulation. Some required inputs were not found in the literature and in these cases the default values given by QWASI were applied (Figures B7 and B8, Appendix B). In addition, processes in the air compartment were ignored because Pb atmospheric deposition in ND is not significant (Figure B9, Appendix B) (EPA, 2011b).

The results from QWASI indicated that the Pb-210 concentration that remains in the water compartment is 1.81×10^{-3} ng/L or approximately 140 pCi/L. For detailed results, please see Figures B10 and B11 in Appendix B. Among methods to remove radioactive

materials from drinking water are lime softening, ion exchange, and activated carbon (EPA, 2015). The average removal efficiency of these three methods is approximately 88% (EPA, 2015) which means that the final concentration after treating the water is 17 pCi/L.

Other unit used to measure radioactivity is Bq/L and since 1 pCi/L is equal to 0.037 Bq/L (Keith et al., 2012) then, 17 pCi/L is equivalent to 0.63 Bq/L. For comparison, some of the maximum allowable levels of Pb in drinking water are 0 mg/L established by the EPA, 1 pCi/L or less based on the National Interim Primary Drinking Water Regulations, 0.1 Bq/L according to the WHO, and 0.2 Bq/L in Canada (Shammas and Wang, 2016; EPA, 2012; Velten and Jacobs, 1982; WHO, n.d.). The concentration of Pb-210 to which a person could be exposed to in the simulated case exceeds these standards.

To characterize the risk of each scenario, a 4×4 risk matrix (Food and Agriculture Organization of the United Nations, n.d.) shown in Figure 4.3 was used. The risk matrix, or C × L matrix, combines consequences (C) and the probability or likelihood (L) of those consequences happening. As seen in Table 4.1, C and L are divided into four different levels. The L levels are remote, unlikely, possible, and likely which are defined based on the probability of the event occurring (Table 4.1) (Food and Agriculture Organization of the United Nations, n.d.; Mosleh and Bari, 1998; Stouffer et al., 2015).

Table 4.1. Measurements of risk likelihood and risk consequence or impact (Pb-10 concentration in contaminated water) used for the risk matrix.

Level	Likelihood		Consequence or Impact	
	Description	Characteristics	Description	Characteristics (Contaminant Level)
1	Remote	It has never been heard of but it is not impossible. Probability < 2%.	Minor	< 0.1 Bq/L
2	Unlikely	Not expected to occur but it has been known to occur else- where. Probability of 2 – 10%.	Moderate	0.1 - 10 Bq/L
3	Possible	Evidence suggests that it may occur in some circumstances. Probability of 10 – 35%.	Major	10 - 100 Bq/L
4	Likely	It will probably occur. Regular and strong evidence. Probability of 40 – 100%.	Extreme	> 100 Bq/L

The four levels of C are minor, moderate, major, and extreme. They are based on the maximum allowable concentration of the chemical under study (Brereton and Alenbach, 1998; Stouffer et al., 2015). For this risk assessment, the limit value used for Pb-210 in drinking water was 0.1 Bq/L established by the WHO (n.d.). This recommended value was used

because a limit in terms of radioactivity has not been established in the U.S. (Velten and Jacobs, 1982). Table 4.1 describes the different consequence levels.

Each of the three scenarios that could result in surface water contamination due to Pb-210 was analyzed using the risk matrix. The C is the same for the three scenarios in which people could be in direct contact with Pb-210 by drinking water containing 0.63 Bq/L. This concentration is 6 times higher than the guidance level established by the WHO. This means that the C is level 2 or moderate impact. The L of each scenario happening (Table B6, Appendix B) is about 6% for storage unit spill, 89% for equipment leakage, and 6% due to truck incident. By looking at Table 4.1, the L of scenario 1 is considered unlikely (level 2), for scenario 2 is likely (level 4), and for scenario 3 is unlikely (level 2).

After multiplying the C and L in each case, scenario 1 and 3 have a score of 4 while scenario 2 is 8. Based on Figure 4.3, the risk can be classified low if the score is 1-2 (green), medium-low if it is 3-4 (yellow), medium-high if it is 6, 8 or 9 (orange), and high if the score is between 12 or 16 (red) (Food and Agriculture Organization of the United Nations, n.d.). In accordance with the numerical score and color code, the risks of scenarios 1 and 3 are considered medium-low while scenario 2 is classified as medium-high risk (Figure 4.3a).

4.3.4. Social risk perception survey results

The survey was developed to capture the general risk perception that oil produced water from ND causes to different stakeholders including general public, operators, and experts. The total number of respondents that completed the survey was 191 with a 19.4% dropout rate which is within the average range of 15% to 30% for online surveys (Galesic, 2006). From all the respondents, 35% did not indicate their place of residence within the State, 46% lived in Cass County while the remaining 19% were in other counties including 11 oil-producing counties. The majority of participants was 18 to 24 years old and had a 4-year college degree (38% and 39%, respectively). For the purpose of this study, only 7 questions out of 25 were used because they were focused on measuring the level of risk perception and

awareness of produced water. The remaining questions are presented and analyzed in Chapter 5.

The participants were asked to rank their level of concern of each scenario where produced water could reach surface water. For the storage tank overflow scenario, 41% responded to be not at all to slightly concerned. For the equipment failure and truck accident scenarios, the majority indicated to be moderately to extremely concerned (57% and 49%, respectively). Some questions in the survey were focused on obtaining the opinion of experts in ND which represented 8% of the total number of participants. The experts were asked to rank the awareness of general public, oil field operators, and truck operators regarding produced water. The respondents indicated that the general public is slightly aware (38%) while oil field operators (56%) and truck operators (44%) are extremely aware. The experts were also asked to rank the level of risk perception of each stakeholder in each scenario. According to the experts, the general public seems to have the highest risk perception on each of the scenarios. On the other hand, the oil field operators and truck operators have lower risk perceptions except on the scenarios where they are in direct contact with produced water, namely equipment and trucks, respectively.

The answers from all the participants were quantified in order to integrate them with the results from the risk matrix in the previous section. There are different methods to interpret raw data from surveys including percent agree, rating and ranking average, top box, and Z-score to percentile rank (MeasuringU, 2011; SurveyMonkey, 2016). For the interpretation of the survey results, a combination of percent agree and rating and ranking was applied along with a modified version of the methodology proposed by Plattner et al. (2006).

The model developed by Plattner et al. (2006) combines technical risk measurement and a weighted mean of different perception affecting factors (PAF). Equation (4.4) defines

risk as the sum of hazard (objective or technical risk) and outrage (perceived risk) (Plattner et al., 2006; Sandman, 1999):

$$\mathbf{Risk} = \mathbf{hazard} + \mathbf{outrage} \quad (4.4)$$

Where hazard (R_{tech}) is equal to $C \times L$ and outrage (R_{perc}) can be calculated with the following equation (Plattner et al., 2006):

$$\mathbf{R}_{perc} = \mathbf{R}_{tech} \times \frac{\sum_{i=1}^n (\mathbf{paf}_i \cdot \mathbf{a}_i)}{\sum_{i=1}^n \mathbf{a}_i} \quad (4.5)$$

For this study, four PAF (\mathbf{paf}_i) were considered, each one with an assigned weight (\mathbf{a}_i). The values the PAF can take vary from 0.5 to 2 so that the R_{tech} can be reduced by half, increased by 2 folds, or in between. The weight assigned to each PAF ranges from 0 to 1 so that the total sum of the weights equals 1. The weights were assigned based on the importance of the survey question or PAF to capture the risk perception of each stakeholder. The four PAF and their respective weights are: 1) level of concern with produced water – 15%; 2) level of familiarity with produced water management – 20%; 3) level of awareness with produced water content – 35%; and 4) opinion of experts on risk perceptions – 30%.

Likert scales were used in the survey to measure the level of concern, awareness, and familiarity. Each scale has 5 levels and each one has a score (i.e. 5, 4, 3, 2, 1). For the quantification process, it was assumed that the less concern/aware/familiar a person was, the higher the score. These survey scores were transformed into PAF-values with a range of 0.5-2. For details, please see Tables B7-B9 in Appendix B. For example, most of the participants indicated to be not familiar at all (survey score = 5) with produced water management, then the PAF-value assigned was 2. Likewise, most of the people indicated to be slightly aware (survey score = 4) with the produced water content so the PAF-value in this case is 1.625. The opinion of experts on risk perception of each stakeholder on each scenario was also transformed into PAF-value and then the average was determined (Table B10, Appendix B). The transformed values for each risk level are 2, 1.25, and 0.5 for low, medium, and high,

respectively. In scenario 1 the results were: general public – medium (1.25), oil field operator – low (2), and truck operator – low (2). In this case the average is 1.75. For scenarios 2 and 3, the average is 1.

Table 4.2. Parameters used to calculate the holistic risk score of each scenario evaluated. The technical risk scores were obtained from the 4x4 risk matrix. The perceived risk scores were obtained from the survey and converted to PAF values.

Parameter	Scenario 1	Scenario 2	Scenario 3
Technical risk	4	8	4
Perceived risk (PAF):			
Level of concern with produced water	1.625	0.5	0.5
Level of familiarity with produced water-MGMT	2	2	2
Level of awareness with produced water-content	1.625	1.625	1.625
Experts' opinion-risk perception	1.75	1	1

The next step was to calculate the risk of each scenario using the information from Table 4.2 and equation 4.5. For scenario 1, the overall risk was calculated as follows:

$$\text{Risk}_{\text{perc, sc-1}} = 4 + \left[4 \times \frac{(1.625 \times 0.15) + (2 \times 0.20) + (1.625 \times 0.35) + (1.75 \times 0.30)}{1} \right]$$

$$\approx 11$$

Using the same method, the calculated risks for scenarios 2 and 3 are approximately 19 and 9, respectively (see Appendix B for further detail). The results indicate that by adding the quantified social risk perception the risk of each scenario incremented. Based on the color code from a 5x5 matrix the risk is low if the score is 1-3 (green), medium-low if it is 4-6 (yellow), medium-high if it is 8-12 (orange), and high if the score is between 15 and 25

(Figure 4.3b). Then, scenarios 1 and 3 are medium-high risk and scenario 2 is considered high risk.

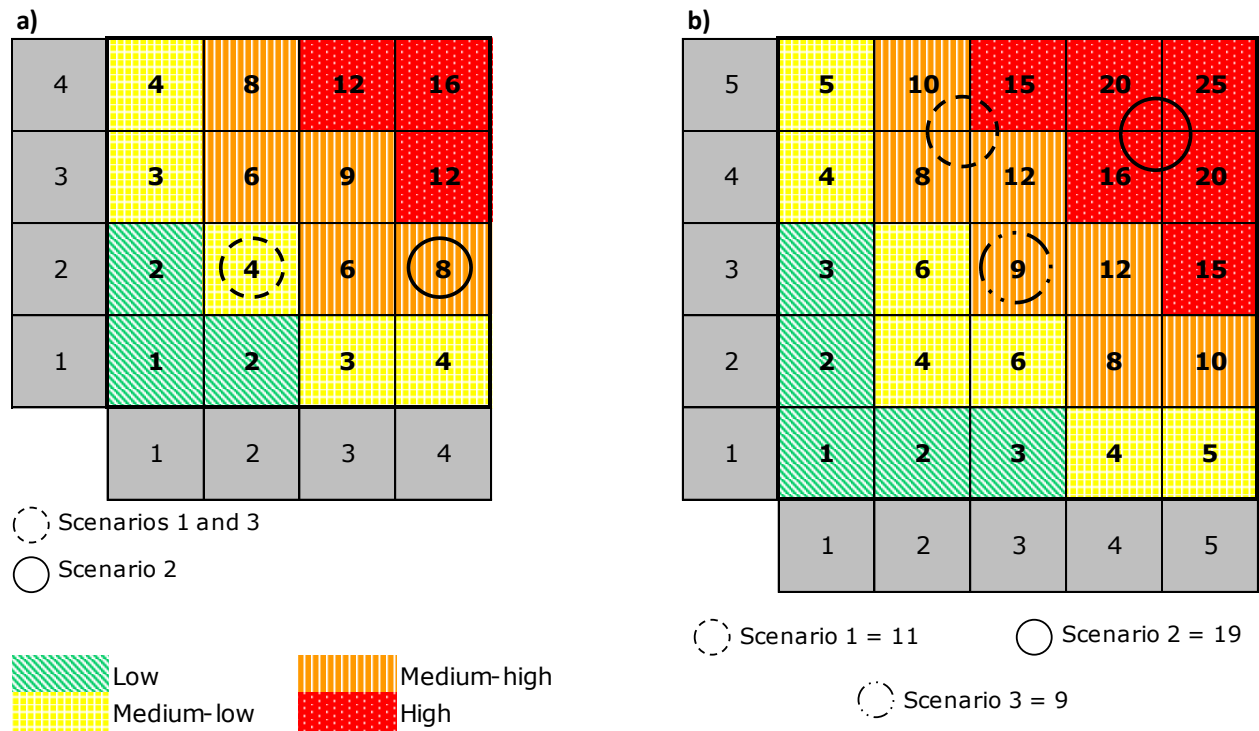


Figure 4.3. Scores for each scenario using a: a) 4x4 risk matrix for technical risk and b) 5x5 risk matrix for combined technical and social risks.

The data collected with the survey indicate that the public is not getting enough objective information about produced water coming from the oil wells in ND. This has caused overestimated and biased risk perceptions. According to the results, people overestimated the risk of each scenario by 25% - 75%. Poor social perception or lack of awareness about risk involved poses more risk on people that do not take preventive measures.

4.3.5. Sensitivity analysis

In this study, the main input to simulate Pb-210 concentration was the Ra-226 initial concentration. If the real average level of radium in produced water was underestimated or overestimated, the average concentration of Pb-210 would be different. To know how much

the output would change if actual Ra-226 level varied between 25%-50%, a sensitivity analysis of four scenarios (Table 4.3) was performed.

Table 4.3. Four different sensitivity analysis scenarios in which the initial concentration varied $\pm 25\%$ and $\pm 50\%$ were evaluated. The final Pb-210 remaining in treated water is shown (in Bq/L) for each sensitivity analysis scenario.

Scenario	Initial Pb-210 (pCi/L)	Pb-210 predicted by QWASI (pCi/L)	Pb-210 in treated water (Bq/L)
Ra-226 25% underestimation	674.54	173.41	0.77
Ra-226 25% overestimation	404.81	104.35	0.46
Ra-226 50% underestimation	809.52	208.70	0.93
Ra-226 50% overestimation	269.85	69.36	0.31

Table 4.3 shows that the mean Pb-210 concentration varied significantly compared to the mean base (540 pCi/L) when the Ra-226 input changed. To understand the effects of these variations, each sensitivity scenario was evaluated in the QWASI model. In addition, the Pb-210 level (in Bq/L) that would remain in the water after treatment was calculated and compared with the 0.63 Bq/L that would remain if the base concentration was considered.

The concentration level of Pb-210 after treatment in each scenario was almost 3 to 9 times higher than the recommended limit of 0.1 Bq/L established by the WHO. This range falls in the level 2 category (or moderate) of the measurement of risk consequence or impact shown in Table 4.1, the same category obtained using the base concentration. This indicates that even if the simulated Pb-210 concentration varies 25% to 50% from the actual value, the risk of water contamination in scenarios 1 and 3 would still be considered medium-high while scenario 2 would still be high risk.

4.4. Summary

This study analyzed from an exploratory perspective the impacts of Pb-210 in produced water from unconventional oil production in ND. A method to predict Pb-210 in the water environment based on Ra-226 in produced water was developed. Also, a holistic process that incorporates technical and social risk was created. With this study, although with some limitations, it was found that the risks of surface water contamination with Pb-210 are not negligible and call for attention.

Undoubtedly more research on this subject is necessary to refine and complement the findings of this study. However, the results have several significant implications for assessing the real impacts of the incorrect management of produced water. First, the lack of data indicate that it is imperative to request oil operators to report radioactivity levels in produced water. Second, improving the spill reporting system could reduce the social risk perception since more and better information would be available. Finally, this study offers the opportunity to utilize the holistic process in future work that could be expanded to other scenarios such as runoff and melting snow which were not covered in this research.

5. PERCEIVED RISKS OF PRODUCED WATER MANAGEMENT AND NATURALLY OCCURRING RADIOACTIVE MATERIAL CONTENT IN NORTH DAKOTA

5.1. Introduction

ND developed its first successful oil well in 1951 and since then the State has become the second largest oil producer in the country and ranks 19th worldwide (ND Petroleum Council, 2015). Sixty-five years have elapsed which makes oil production an old and well established practice in ND. However, the most recent oil boom and new drilling and completion techniques have resulted in controversy dividing the population between proponents and opponents for oil development (Brown and Yücel, 2013; Holeywell, 2011; Siegler, 2014).

O&G production in the U.S. has changed over the years, especially in ND. When oil was first discovered in the 1950's, it was welcomed by the State since there was a nationwide race to discover O&G during that time (Bluemle, 2001). However, oil production in the 1950's was limited because there was no supply shortage in the country compared to other decades, such as the 1970's crisis (Bluemle, 2001; History, 2010). Nowadays, O&G production using the unconventional methods of HF and horizontal drilling have changed the way people see energy development in the country. Information availability has increased and people are more informed about the events occurring in their communities (Energy In Depth, n.d.; Western States Petroleum Association, 2013). This increase in awareness has resulted in relatively new and stricter environmental regulations regarding O&G compared to earlier development in the country (Kao and Gao, 1998; Kusnetz, 2012).

Unconventional O&G development requires the injection of millions of gallons of water with chemicals additives under high pressure. The injected water returns to the surface (flowback water) through the wellbore mixed with water that is naturally present in the reservoir (formation water). This mixture is called produced water which requires special management techniques for recycling or disposal due to its unique characteristics. Produced water contains constituents such as salinity, organics and inorganics, and NORM and some of

the constituents are known to affect human health and the environment if improperly managed (EPA, 2012). Over the years, research has been conducted regarding contaminants in produced water and this information has become more publicly available (Energy In Depth, n.d.; Western States Petroleum Association, 2013). In addition, media coverage on accidents and spills related to oil development and production has increased (Weber et al., 2014). These two factors, among others, have called the attention in ND and nationwide regarding the risks associated with produced water.

Risk perception cannot be directly measured in a quantitative way because it is based on different factors such as emotion, trust, values, knowledge, and experiences (Jones, 2012; Leiserowitz, 2005; Slovic, 2000). Different methods, including physical scaling and multivariate analysis, have been used to translate risk attitudes and perceptions into numerical measurements (Slovic et al., 1982). Surveys have been widely used in the past to collect data regarding risk attitude in different areas such as economics and psychology (Xiaohao et al., 2010). Surveys are effective tools to measure valid predictors of risk attitudes in real situations if designed appropriately and specific enough to collect more than general measures of concern (Leiserowitz, 2005; Xiaohao et al., 2010).

Previous research has attempted to understand the perception public and stakeholder groups have regarding energy development, including HF. Surveys have been conducted at the state level, for example, in Pennsylvania, Ohio, and New York, and nationwide to collect public opinion data (Boudet et al., 2014). Data collected in these studies could be further analyzed by combining social and technical perspectives. In the past, risk analysis of new technologies has focused mostly on technical aspects leaving aside social aspects resulting in a narrow view of the real risks (Skogdalen and Vinnem, 2011). The results from social risk perception studies can be incorporated in risk assessments of O&G development to deliver holistic results that can be used to create appropriate safety measurements inside and outside the production fields.

The holistic study of risks in oil development and production is particularly incomplete in ND where very little data on social aspects have been collected despite it is one of the largest oil producing States in the country. Among the few studies conducted in ND are Weber et al. (2014), focused on social services, and Cwiak et al. (2015) which reported the thoughts, observations, and opinions of emergency management personnel about the direct and indirect impacts of oil production in the State. No study on social risk perception in ND has focused on produced water, the largest by-product of oil production.

The number of produced water spills in ND has increased over the years with approximately 3,900 reports since 2007 (Lauer et al., 2016). This increase is directly correlated with the amount of oil extracted (Lauer et al., 2016). In addition, most of the accidents in the exploration and production industry are caused by human error (Boschee, 2014). Here lies the importance of studying the social perception of produced water in ND because understanding how risk perception influences judgment and safety culture can lead to improved regulations and standards (Boschee, 2014).

The objective of this research is to determine the risk perception and awareness of produced water management in different stakeholder groups in ND as well as to identify the most influencing variables that shape those perceptions. An online survey was developed using the web-based software Qualtrics (2016). The same software was used to collect and analyze the raw data obtained with the survey in order to understand the reasoning behind the perceptions that produced water management and contents have on people. The questions were aimed at four different stakeholder groups: general public, oil field operators, produced water hauling truck operators, and people whose jobs involve direct produced water management, decision making regarding health and safety, and/or emergency management personnel (hereinafter experts). The survey was distributed in numerous counties in ND.

5.2. Methodology

5.2.1. Survey questionnaire development

The survey consisted of a questionnaire with 25 questions administered completely online. The survey was built using the Qualtrics online tool which in addition was used to collect the data, conduct the statistical analysis, and create reports. The NDSU Institutional Review Board approved this study including the survey (Certification of Exempt Human Subjects Research #EN 16229, please see Appendix D). Most of the questions used a Likert scale from which the participants chose their answer. The Likert scale is a popular method used to measure attitudes and behaviors from one extreme to another (e.g. Not at all familiar to extremely familiar) (Losby and Wetmore, 2012).

The complete survey is available in Appendix C. The first set of questions in the survey was used to collect demographic characteristics and the main source of information among the participants. Also, the general attitudes the participants had towards the economy of the State and if they work or used to work in the oil field industry in ND was collected. In addition, the participants were asked to indicate if they know someone that works or used to work in the oil industry. A section of the survey also focused on collecting affective imagery data by using the method of word associations (Leiserowitz, 2005). The participants were asked to rank (negative or positive) the initial thought or image that comes to their mind when they read "fracking wastewater" and "natural radioactive material." This was done with the objective of analyzing the relationship between image perception and risk perception (Leiserowitz, 2005).

Later, the respondents indicated their level of awareness and familiarity with the contents and management of produced water, as well as, their level of concern with the associated risks. Moreover, the questionnaire included a section where participants indicated their level of trust in different organizations involved directly or indirectly with produced water management. Towards the end of the survey, participants were asked if their jobs involve

responsibilities directly related to oil production and produced water management or if they were part of the emergency management personnel or a key response partner in ND. For the ones that responded negative, that was the end of the survey. Those who responded affirmative were categorized as experts and were redirected to further questions. Experts were asked to rank the level of awareness of the general public, the oil field operators, and the hauling truck operators about the risks of produced water. In addition, the experts were requested to gauge the risk perception of each stakeholder group on three different scenarios where produced water could reach a surface water body.

5.2.2. Survey testing and validation

The survey testing and validation process consisted of a pilot test. Four potential respondents, including colleagues of the authors and a layperson, were selected and asked to complete the survey. The most important aspects evaluated on the pilot test were completion time, wording, and questions accessibility (e.g. type of question such as multiple choice). Each respondent provided feedback on the content which was used to modify the questions and language to make the survey clearer.

5.2.3. Sample selection and size

Four different stakeholder groups were selected in order to obtain a more generalized risk perception in ND. They were general public, oil field operators, produced water hauling truck operators, and experts. The latter group was of particular importance because their job involves decision making regarding produced water management and/or health and safety measurements, and collaboration with people that work in the oil field. It was assumed they have a broader view of the situation because of their daily job experiences. For this survey, the voluntary sample method was used which means that the sample was made up of people who self-selected into the survey because they had some level of interest in produced water management and content.

The Qualtrics sample-size calculator was used to determine the necessary number of participants. The calculator uses the Cochran equation (Israel, 2013):

$$n = \frac{z^2 \times \hat{p}(1-\hat{p})}{ME^2} \quad (5.1)$$

Where z is the z -score based on the confidence interval, \hat{p} is the population standard deviation, and ME is the desired margin of error. Since \hat{p} was unknown in this case, the recommended conservative assumption of $\hat{p} = 0.5$, or the maximum variability, was used (Israel, 2013). To calculate the sample size, a total population of 379,000 habitant in Cass County and the 17 oil-producing counties (Cubit Planning Inc., 2016) was used, a 90% confidence interval, and a margin of error equal to 5%. A targeted sample size of 271 was obtained using the Qualtrics sample-size calculator (Qualtrics, 2016). It should be noted that since the survey was also distributed among NDSU colleagues, Cass County was included in the targeted population.

5.2.4. Survey distribution

The survey was distributed in different locations in ND including Cass County and 17-oil producing counties, which are: Billings, Bottineau, Bowman, Burke, Divide, Dunn, Emmons, Golden Valley, McHenry, McKenzie, McLean, Mountrail, Renville, Slope, Stark, Ward, and Williams (ND Petroleum Council, 2015). The link to the online questionnaire was distributed using different means including NDSU's listserv, direct e-mail invitations to experts in different Counties, and two Facebook community pages focused on the Bakken oil field. Every participant that followed the link received an invitation and a consent form (for details please see Appendix C) which included an explanation of the research study, contact information, the objective of the study, and information about the survey (e.g. length, risks, and benefits). The survey was active from April 5, 2016 to April 19, 2016.

5.2.5. Data collection and statistical analysis

The objective of the statistical analysis was to determine the most influential variables on risk perception as well as the interrelation between the variables. The data from each completed survey was automatically collected and stored online by Qualtrics (2016). This software was used in combination with Microsoft Excel for the statistical analysis section usually composed of three phases: data preparation, descriptive statistics, and inferential statistics (Trochim, 2006). The data preparation consisted of downloading the initial raw data report from Qualtrics. The descriptive statistics were used to present graphically the results based on the most frequently selected answers on each questions and on some, the mean and standard deviation. Finally, inferential statistics using cross-tabulation along with Chi-Square analysis were obtained to support the conclusions regarding what the population thinks about produced water in ND.

5.3. Data analysis

A total of 237 surveys were started during the active period of two weeks, less than the targeted sample size ($n=271$), calculated in section 5.2.3, possibly due to the limitations of the distribution means used. From the total number of surveys started, 191 participants completed the survey, that is a 19.4% dropout rate which is within the average range of 15% to 30% for online surveys (Galesic, 2006). The response rate (80.6%) was automatically calculated by Qualtrics. The average time the participants took to respond all the questions was 10 minutes. The following subsections describe the results on each part of the survey.

5.3.1. Demographics

From all the respondents, 35% did not indicate their place of residence within the State of ND, 46% indicated to live in Cass County while the remaining 19% were in other counties including 11 oil-producing counties. More than 60% of the participants were between 18 to 34 years old. The highest level of education completed by almost 40% of the respondents

was a 4-year college degree. In addition, 89% of the people have never worked in the oil industry but 71% indicated to know someone that does or used to do so.

5.3.2. Source of information

Media coverage can affect risk perception and the extent of the influence depends on the medium, the message, and the viewer (Boudet et al., 2014; Plattner et al., 2006). The majority of the people (68%) use the internet as their main source of news while the second most used medium is television (19%). The effects of internet on risk perception has not been studied as widely as other media such as television and newspapers (Boudet et al., 2014). However, studies such as Park and Sohn, (2013), Cacciatore et al. (2012), and Gerhards and Schafer (2010) found that the internet is a good source of news which are more focused on the environment but at the same time is more biased and thus less credible compared to television and newspaper.

5.3.3. State and local economics

Two questions were included to understand how satisfied the current residents are with their communities. The first question asked to rank the satisfaction with living/investing in the State of ND and 70% of the people responded to be somewhat to extremely satisfied. The second question asked the participants to agree or disagree with the following statement: "Areas where produced water is stored or transported are likely to be unattractive to new residents, business development, and tourist." Almost half of the people (46%) responded that they somewhat agree or strongly agree with the statement. Although the survey did not measure the proximity between the residence of the participant and the closest oil well, the results are in line with previous research which has reported the Not In My Backyard (NIMBY) effect (Huijts et al., 2007; Krause et al., 2014). The NIMBY sentiment is based on the idea that participants will perceive positively a new technology that generates local economic benefits as long as the individual is far from an undesirable facility (Krause et al., 2014).

5.3.4. Risk perception images

When analyzing a situation, people tend to balance risks and benefits. Each side of the balance affects people's perception which creates negative or positive reactions (Boudet et al., 2014). According to Leiserowitz (2005), affect refers to the "positive or negative quality of a stimulus." All the affective images associated with "fracking wastewater" given by the participants were classified in 25 different categories from which the top 7, shown in Figure 5.1, was considered since it represents 55% of the respondents.

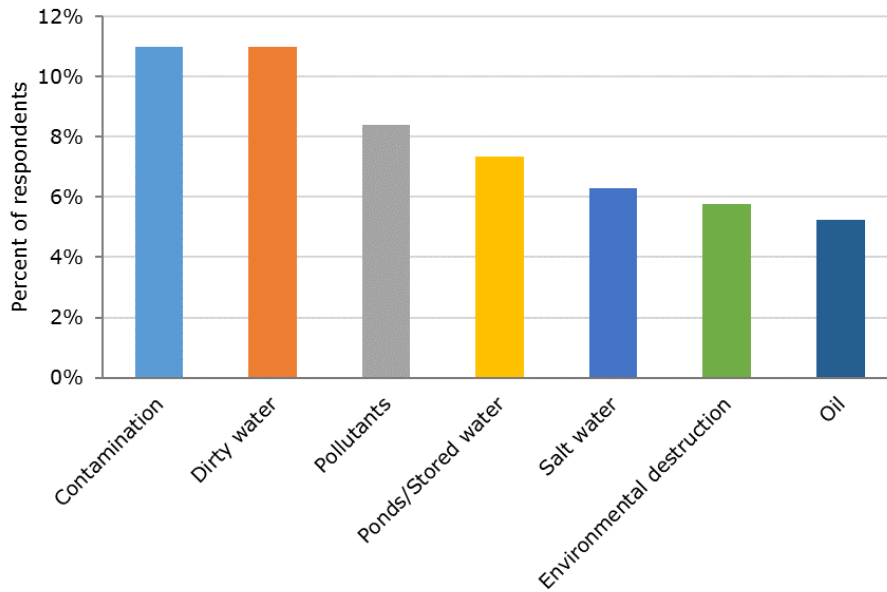


Figure 5.1. Top categories of the affective images participants associated with "fracking wastewater."

The majority of people associated produced water or fracking wastewater with "contamination" and "dirty water." In addition, the participants were asked to rank their image using a scale ranging from -5 (very negative) to +5 (very positive). Most of the participants (59%) rated between -3 to -5 which is considered to be between moderately to very negative (Figure 5.2).

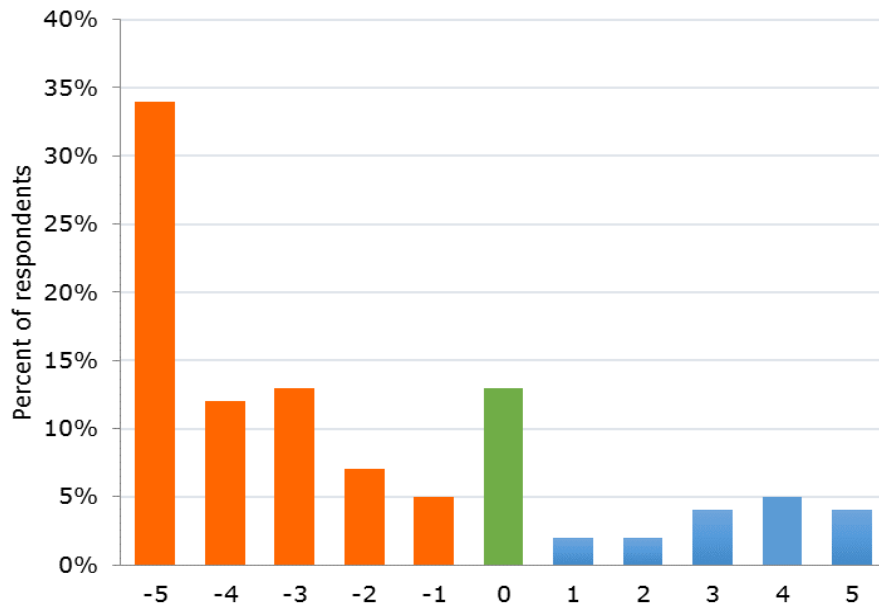


Figure 5.2. Rating of affective images for produced water given by the participants (-5 = very negative and +5 = very positive).

Figure 5.3 shows the top 6 out of 25 categories used to classify the thoughts and images that participants associated with “natural radioactive material.” These categories represent 54% of the total answers. Almost 20% of the participants think that “natural radioactive material” is “dangerous” while the second most selected category was “natural and/or safe.” The most frequently selected option to rank the thought or image associated with “natural radioactive material” was neutral (rate = 0) by 27% of the participants. The opposite affective imagery given to produced water and natural radioactive material could be explained by the fact that people usually have a lower risk perception for natural situations than man-made situations (McCrary and Baumgarten, 2004).

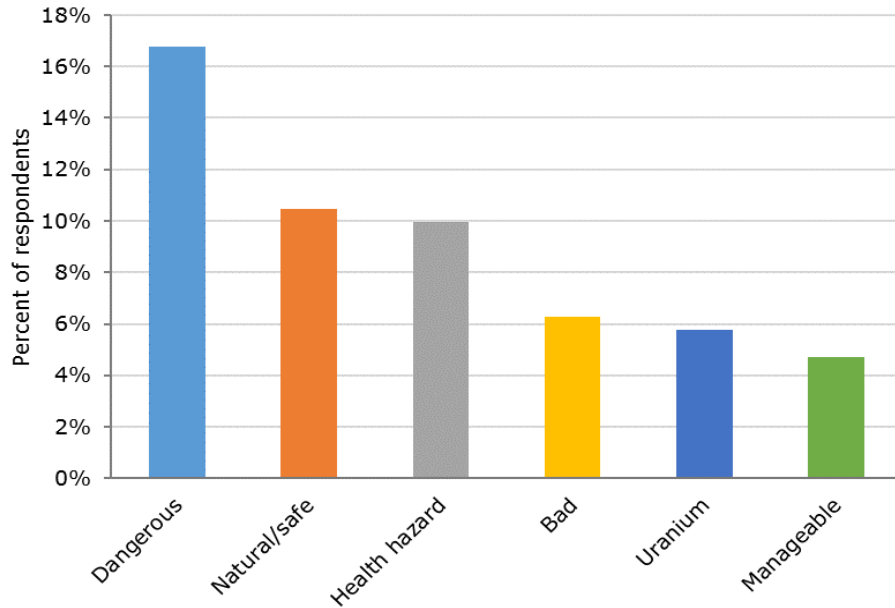


Figure 5.3. Top categories of the affective images participants associated with “natural radioactive material.”

5.3.5. Produced water handling and contents

The survey asked the participants to rank their level of familiarity with produced water handling. More than half of the people (58%) responded they are slightly familiar to not familiar at all with the processes of storage and transportation of produced water. Similarly, the participants indicated their level of awareness with the contents of produced water such as contaminants and chemical additives. More than half (52%) said they are slightly aware to not at all aware with the produced water content. On the other hand, the majority (56%) of the participants indicated they are slightly to moderately familiar with natural radioactive material and its effects on human health. However, there is still a considerable amount of participants (44%) that did not know that produced water might contain NORM.

5.3.6. Health and safety risks

In order to measure the perception of harmful effects caused by produced water on public health and safety, the participants were asked to rank their level of concern of three different scenarios (Table 5.1). The level of concern was measured using a Likert scale with

five points coded as follows: 1 – Not at all concerned, 2 – Slightly concerned, 3 – Somewhat concerned, 4 – Moderately concerned, and 5 – Extremely concerned. Table 5.1 shows the scenarios presented to the participants, the mean and standard deviation of the responses, and the most frequently selected answer for each one. Despite expressing concern in the three scenarios, it seems that people have not taken preparedness actions since most of the participants (72%) said that they do not know whom to contact to report a produced water spill.

Table 5.1. Level of concern on three different scenarios where produced water could affect human health and the environment. From 1 – Not at all concerned to 5 – Extremely concerned.

Question (scenario)	Mean	Std. Dev.	Most selected (%)
How concerned are you that storing produced water in tanks might have harmful effects on public health and safety in your area?	3.09	1.43	Slightly concerned (24.1%)
How concerned are you that failure of equipment used to handle produced water (e.g. pipelines) might have harmful effects on public health and safety in your area?	3.52	1.37	Extremely concerned (32.5%)
How concerned are you that transporting produced water by truck might have harmful effects on public health and safety in your area?	3.20	1.45	Extremely concerned (24.6%)

5.3.7. Level of trust

All participants were requested to indicate the level of trust in different organizations involved, directly or indirectly, with produced water management. The groups evaluated were oil operators, truck companies, State/local government, Federal government, and the EPA.

Once again, a Likert scale with four points was used. The scale options were: 1 – No trust at all, 2 – Little trust, 3 – Quite bit of trust, and 4 – A lot of trust. Table 5.2 shows the results on the level of trust of the group evaluated by the participants.

Table 5.2. Level of trust in different organizations. From 1 – No trust at all to 4 – A lot of trust.

Organization	Mean	Std. Dev.	Most selected (%)
Oil operators	2.07	0.86	Little trust (43%)
Truck companies	2.18	0.79	Little trust (49%)
State/local government	2.43	0.86	Quite a bit of trust (39%)
Federal government	2.35	0.87	Little trust (43%)
EPA	2.69	0.97	Quite a bit of trust (36%)

Based on the mean, the EPA’s score was the highest but not by far which indicates that overall, the trust level in all the organizations evaluated is low. On the other hand, over 40% of the participants indicated some level of confidence that State agencies will provide honest and accurate information about the safety of produced water handling and disposal.

As mentioned before, risk perception of people is created based on different factors and one of them is the social influence of different organizations and individuals involved, in this instance, in oil production in ND. Not everybody processes information the same way, especially if technical knowledge is required to understand a new technology. In this case, a person will rely on others, including professionals and other organizations such as State and Federal government, to form an opinion (Huijts et al., 2007; Boschee, 2014). The groups shown in Table 5.2 were selected because they tend to have more influence in public acceptance due to their role in policy making (Huijts et al., 2007). The results seem to be in agreement with previous research that has found that trust in government agencies and

industries is relatively low while trust in organizations that appear to share values with the respondent is higher (MacGregor et al., 1994; Siegrist et al., 2000; Upreti, 2004).

5.3.8. Questions to experts

From all the participants that completed the survey, 16 (8%) indicated to work directly with oil production and produced water management or as part of the emergency management personnel in ND. These participants, considered experts, were asked to rank the produced water risk awareness of the general public, the oil field operators, and the produced water hauling truck operators. The majority of expert respondents (57%) said that the public is not at all aware to slightly aware. The oil operators are considered to be extremely aware by 56% of the experts. Moreover, most of the experts (63%) think that the produced water hauling truck operators are moderately to extremely aware.

Finally, the last question in the survey directed to the experts asked to rank the level of risk perception of each stakeholder group in three different scenarios where a produced water release from containment (i.e. storage, equipment, and truck) could reach a surface water body. The results are summarized in Table 5.3. The level of risk perception for each case shown in Table 5.3 was the most frequently selected option (based on percentage) by the experts.

Table 5.3. Opinion of experts about the risk perception of each stakeholder in three different scenarios. The risk perception level is based on the most selected option (%).

Scenario	General public	Oil field operators	Truck operators
Storage tank overflow	Medium (56%)	Low (56%)	Low (56%)
Equipment failure	High (50%)	Medium (38%)	Medium (38%)
Truck accident	High (50%)	Low (44%)	High (38%)

The experts reported that the general public has the highest risk perception for all of the scenarios and the oil field operators have the lowest risk perception. Experts believe that

hauling truck operators have a high risk perception of accidents associated with truck transportation. This could be because drivers' relative exposure to risks during transportation is higher than other stakeholder groups (Flin et al., 1996). Based on these results, experts think that there is a big gap in the level of awareness and risk perception between the general public and the oil and truck operators.

5.4. Cross-tabulation results

To further understand the results, the relationships between variables using the cross-tabulation tool available in Qualtrics (2016) was analyzed. Each cross-tabulation result given by Qualtrics is accompanied by a Chi-Square analysis which determines whether or not two qualitative variables are independent (Qualtrics, 2016). This is done using the hypothesis testing where the null hypothesis is that there is no relationship between the variables while the alternative hypothesis is that dependency exists between the variables. The most important statistical indicator to look at is the p-value which suggests that the variables are dependent if it is less than or equal to the significance level (α), which in this case was 0.05 (Martz, 2012). To know more about the cross-tabulation and Chi Square analysis, Michael (2013) is recommended.

A key objective of this research was to understand the variables that has the most influence on the risk perception of ND residences. Unfortunately, the variables of location, age, level of education, and main source of news were not cross-tabulated because some demographic categories were found to be under represented in the results. In order to understand how the other variables influence risk perception, cross-tabulations were performed for the following categories:

- Work or have worked in the oil field
- Know someone that works or have worked in the oil field
- Familiarity with produced water management
- Familiarity with NORM

- Affective imagery of produced water
- Affective imagery of NORM
- Having a job directly related to produced water management and/or health and safety measurements (i.e. experts)
- Knowledge of produced water spills reporting system

Based on the Chi-Square analysis, there was no significant dependency between “working or have worked in the oil field” and the “level of concern with produced water harming human health and the environment.” In addition, the participants that “know someone that works or have worked in the oil field” indicated to be more concerned with harmful effects of produced water in the scenario of truck accidents (p-value = 0.02) but the same tendency was not observed for storage tank (p-value = 0.16) and equipment failure (p-value = 0.07).

The influence of level of knowledge was analyzed by cross-tabulating familiarity with produced water management and NORM with the level of concern in different scenarios. People that indicated to be more concerned tended to be less familiar with the processes of produced water storage and transportation. However, no association was obtained for the level of familiarity with natural radioactive material. As a general observation, people tend to fear more the unknown, or uncertain situations, and the level of perceived risk affects the willingness to adopt self-protective behaviors (Coppola, 2015; Weinstein et al., 1991).

A risk can be perceived based on how people feel about it and one way to measure that feeling is through images (Slovic and Peters, 2006b). The score given by the participant to the image or thought associated with produced water and NORM was cross-tabulated with the level of concern of three scenarios (i.e. storage, equipment, and truck). For the produced water, in all the scenarios there was an association (p-value < 0.0001) between negative score and high risk perception. Similarly, the level of concern and the NORM affective imagery

were found to be dependent in each scenario: storage (p-value = 0.00001), equipment (p-value = 0.003), and truck (p-value = 0.0006).

The dependence between being an expert and the risk perception could not be supported by the Chi-Square analysis since the p-values for each scenario was larger than 0.05. However, based on the most selected option, the experts tended to be more concerned about the scenario where equipment fails than the scenarios of storage tank overflow and truck accidents. Finally, not knowing how to report a produced water spill seemed to be directly related to a higher level of risk perception in the scenario of equipment failure (p-value = 0.0009) but not for the other two scenarios.

5.5. Discussion

The cross-tabulation analysis conducted in this study gives a broad picture of the risk perception among people in ND. The main focus was to measure the impacts of the level of knowledge and experiences have on risk perception of produced water from unconventional oil production. To further understand demographics and their effects on social risk perception, it is recommended to perform a statewide survey with a larger sample of the population and using other means of distribution besides internet although it is expected to obtain similar results as reported by other research which found different levels of risk perception based on media use, gender, age, and formal education. For example, Boudet et al. (2014, 2016) reported higher risk perception of new energy technologies among newspaper readers, women, older people, and individuals with more formal education.

Overall, based on the statistical analysis of this survey, people have little knowledge regarding the management and content of produced water. One would think that because most of the participants (71%) indicated to know someone that works in the oil field they would have a more reliable source of information. However, in this case it could be that the quantity of the information is not the problem but the quality (Gower, 2006).

Not knowing where to report a produced water spill could affect the safety of people and impact the environment. Also, not knowing about the monitoring and reporting mechanisms in place and oil field employee training could also contribute to higher risk perceptions. This has caused different levels of concern with the scenarios where produced water could reach surface water being equipment failure the one with the highest risk perception among the respondents.

Studying social risk perception could contribute significantly in the development of prevention plans and risk management. By measuring risk perception, subjective information can be combined with objective data obtained through technical analyses. With this, better and more responsible decisions regarding produced water management can be made. All the stakeholders affected directly and indirectly should make collective efforts to control risk without hindering economic development in their communities.

5.6. Summary

Very little research has been conducted to understand the social impacts created by unconventional O&G production especially the effects of produced water. Through the development and implementation of an online survey, this study collected data on social risk perceptions of produced water in ND. The cross-tabulation analysis revealed that the most important variables that influence risk perception are the images and thoughts associated with produced water, level of knowledge about produced water handling and content, and knowing how to proceed in case of a spill of produced water. By understanding the risk perception of different stakeholder groups and controlling the most influencing factors, risk mitigation plans can be improved and in turn a more transparent risk communication between the parties involved can be achieved. It is important to note that the survey used in this study has its limitation and results should be used with caution. The findings represent current perception of a small sample of the population in ND. Due to the lack of information on the subject in the State, this study can contribute to future investigation that improves the

understanding of the variables that affect risk perception of produced water in ND and other locations where unconventional O&G production is practiced.

6. CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE WORK

The work presented in this thesis can be grouped in three parts: 1) a comprehensive review of the risks associated with onshore unconventional O&G development (Chapter 2), 2) a technical assessment of risks of human exposure to Ra-226 in produced water from the Bakken Shale (Chapter 3), and 3) a holistic assessment of risks of contamination due to Pb-210 in produced water (Chapter 4) and the social perceptions and awareness associated with those risks in the State of ND (Chapter 5).

In the first part of the thesis, the weakest areas where further study is necessary were identified. Many gaps in data were encountered, especially on the characteristics of fracturing fluid and produced water. The little data found was difficult to access for different reasons including confidentiality, paid sources, and produced water spills reporting systems that are not user-friendly. The literature review led to the selection of the least studied type of contaminant in ND and the most suitable risk assessment techniques. The contaminants selected are Ra-226 and Pb-210 due to their ability to mobilize in the environment, their effects on human health, and the scarcity of data on NORM. The risk assessment techniques selected are a combination of stochastic and deterministic methods due to data unavailability and variability.

In the second part of the thesis a method was developed to perform risk assessment from a technical or engineering approach focused on the human exposure to Ra-226 via food and water consumption. Due to the little data found specific to Ra-226 in ND, simulation techniques were used. No study of this kind has been conducted in ND and only one study has reported levels of Ra-226 in produced water from the Bakken Shale. Because of this, a regression model was developed to predict Ra-226 concentration in produced water based on the levels of the most common cations barium, strontium, and calcium. This model could be used to estimate the Ra-226 for any sample of produced water from the Bakken Shale. The results from the regression model were used in a simplified risk assessment to estimate the

human exposure to Ra-226 via consumption of fish, potatoes with skin, and drinking water. The assessment estimated the total annual effective dose rate of Ra-226 for an adult in ND to be 0.49 mSv. This result is above the global average annual effective dose rate via food and drinking water which is 0.30 mSv. Although the method used in this chapter is a simplification of a proper human health risk assessment, the results indicate that there is potential risk to residents in ND which merits further investigation. The methodology and data presented in this chapter could be used as a baseline for future work. A proper human health risk assessment should measure the Ra-226 levels in foodstuff from a typical diet for a ND resident and levels from other exposure routes such as inhalation.

The third and last part of the thesis presents a novel method to analyze the risks of surface water contamination due to Pb-210 in produced water from the Bakken Shale from a holistic perspective. The results from Chapter 5 were combined with Chapter 4 to characterize the risks. Although findings from the study include a low probability of a produced water spill reaching surface water, the consequence of this event could have a great impact since the simulated concentration of Pb-210 in drinking water was found to be higher than the recommended value established by the WHO. After including the results from the risk perception survey (Chapter 5), the assessment indicates that the risks of contamination of the three scenarios evaluated in Chapter 4 are between medium-high to high. The issues that emerge from high-impact, low-probability events, such as insufficient preparedness of governments and industry, have not been widely studied. In addition, research on the impacts these events have in society is still in the early stage. Thus, results from the study presented in Chapter 4 is valuable for future research which should collect more data on actual Pb-210 level, refine the simulation model developed, and incorporate survey results from a more varied demographics.

Results from the survey indicate that the most important variables that seem to positively or negatively influence the risk perception and awareness are the images and

thoughts associated with produced water, level of knowledge about produced water handling and content, and knowing how to proceed in case of a spill of produced water. It is recommended to conduct a statewide survey that reaches out to the populations that were underrepresented in this study. The demographics that need to be further analyzed are different media use, gender, age, and formal education.

Overall, social risk perception could be in alignment with actual technical risk if availability of objective information is improved. The results obtained with a holistic approach are more comprehensive than the results from a technical risk assessment. The identification and understanding of outside factors, such as risk perception and awareness of risk, could contribute to the creation of plans, standards, and regulations to mitigate risks if combined with technical analyses. Risk communication is important but it has never been more important in the management of high-impact, low-probability events like the ones associated with unconventional O&G. By reducing the risks of environmental contamination and human health exposure to contaminants, the perceptions in different stakeholders can be improved and, thus, decrease the opposition to onshore production of O&G using unconventional methods.

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**APPENDIX A. SUPPORTING MATERIAL FOR CHAPTER 3: RISK ASSESSMENT OF
HUMAN EXPOSURE TO Ra-226 IN OIL PRODUCED WATER FROM THE BAKKEN
SHALE**

A.1. Multivariate Regression Model

Table A1 shows the average values of the five different datasets used for the development of the multivariate regression model. The average values transformed to log normal values were used in the analysis. Table A2 shows the results of the regression analysis performed in Microsoft Excel.

Table A1. Average values and Log values used for the multivariate regression model.

Data Location	Ba in mg/L (Log)	Ca in mg/L (Log)	Sr in mg/L (Log)	Ra-226 in pCi/L (Log)	Source
Louisiana	44 (3.78)	1,590 (7.37)	56 (4.03)	176 (5.17)	(Landa and Reid, 1983)
Mississippi	64 (4.16)	26,018 (10.17)	1,314 (7.18)	563 (6.33)	(Blondes et al., 2016)
Pennsylvania	1,151 (7.05)	13,437 (9.51)	1,925 (7.56)	1,244 (7.13)	(Blondes et al., 2016; Warner et al., 2013)
Texas	147 (4.99)	10,880 (9.29)	1,750 (7.47)	2,300 (7.74)	(Silva et al., 2012)
Various (max. value)	7 (1.95)	52,920 (10.88)	2 (0.69)	262 (5.57)	(Alley et al., 2011)

Table A2. Multivariate regression model results.

	Coeff.	Standard Error	t Stat	p-value	Lower 95%	Upper 95%	Lower 95.0%	Upper 95.0%
Intercept	1.62523	4.62568	0.35135	0.78490	-57.1496	60.4001	-57.1496	60.4001
Log Ba (mg/L)	0.16803	0.56304	0.29843	0.81536	-6.98613	7.32220	-6.98613	7.32220
Log Ca (mg/L)	0.30794	0.43172	0.71329	0.60555	-5.1776	5.79349	-5.1776	5.79349
Log Sr (mg/L)	0.20752	0.34331	0.60447	0.65386	-4.15469	4.56973	-4.15469	4.56973

A.2. ND Produced Water Data

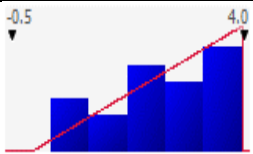
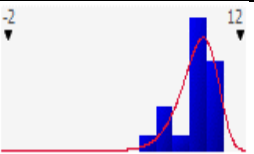
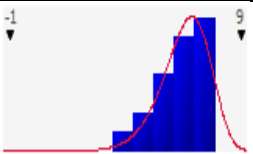
Produced water data from the Bakken Shale was obtained from two different sources: USGS Produced Water Database v2.2 and Lauer et al. (2016) Table A3 shows the data (in Log) from each source.

Table A3. Produced water data from the Bakken Shale.

Source	Log Ba	Log Ca	Log Sr	Source	Log Ba	Log Ca	Log Sr
USGS	3.81	10.04	7.71	USGS	0.54	8.95	6.05
USGS	3.81	10.04	7.72	USGS	1.61	7.50	5.30
USGS	3.14	9.68	7.31	USGS	2.30	10.33	5.70
USGS	1.70	8.22	5.86	USGS	2.71	10.06	6.77
USGS	2.09	9.47	6.83	USGS	3.40	10.83	7.75
USGS	2.09	9.39	6.83	USGS	3.87	10.79	7.61
USGS	0.34	7.60	4.98	Lauer et al.	2.22	9.40	6.65
USGS	0.36	7.65	4.95	Lauer et al.	2.52	9.06	6.31
USGS	3.03	9.77	7.13	Lauer et al.	3.27	5.92	3.50
USGS	3.31	9.82	7.22	Lauer et al.	1.85	9.64	6.88

Each dataset was analyzed using Palisade's @Risk to determine the type of distribution followed by each cation. Table A4 shows the results of the best fit analysis.

Table A4. @Risk best fit results.

Parameter	Log Ba	Log Ca	Log Sr
Best Fit	RiskTriang (0,3.8712,3.8712)	RiskWeibull (10.246,9.6994)	RiskWeibull (7.7423,6.8882)
Minimum	0	0	0
Maximum	3.8712	+Infinity	+Infinity
Mean	2.5808	9.2369	6.4765
Mode	3.8712	9.6026	6.7662
Median	2.7374	9.3586	6.5697
Std. Deviation	0.9125	1.0861	0.9907
Graph			

A.3. Ra-226 Simulation Results

Equation 3.1 in Chapter 3 and the three different distributions for the cations shown in the previous sections, A.2, were used in @Risk to simulate 100,000 times the concentration of Ra-226 in produced water. Figure A1 shows the results in Log scale. The median is 6.2823 or $e^{6.2823} = 535$ pCi/L.

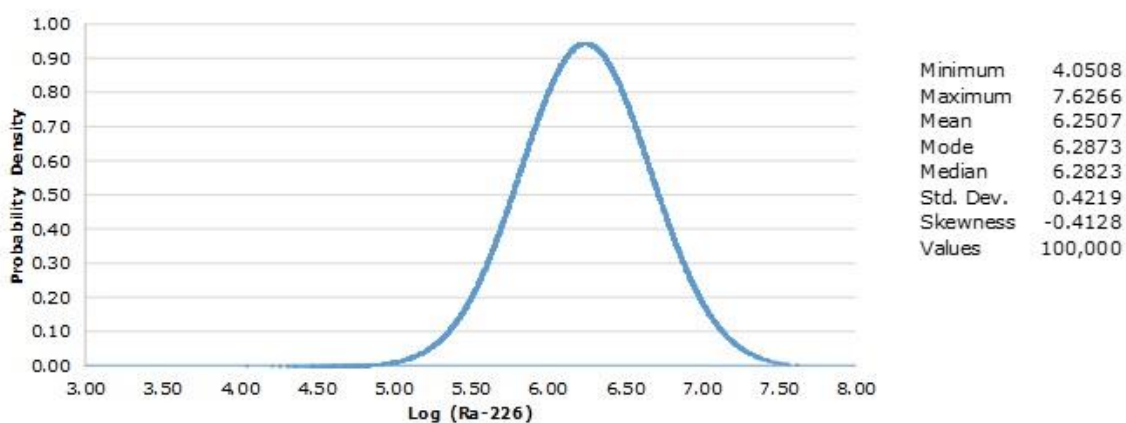


Figure A1. Ra-226 probability distribution.

A.4. QWASI Simulation Inputs and Outputs

Chemical Name	Radium-226	
CAS	7440-14-4	
Molar Mass (g/mol)		226
Data Temperature (°C)		20
Melting Point (°C)		700
Vapor Pressure (Pa)		1.00E-22
Solubility in Water (g/m ³)		1.96E+05
Henry's Law Constant (Pa·m ³ /mol)		1.00E-10
Reaction Half-Lives (hours)		
In Water		1.40E+07
In Sediment		1.40E+07
Partition Coefficients		
	unitless	
logK _{OW}		0
Air-Water (K _{AW})		0.00E+00
Aerosol-Air (K _{QA})	←	0.00E+00
	OR	
Aerosol-Water (K _{QW})	←	0.00E+00
	L/kg	
K _{OC}		1.00E+02
Suspended Particles-Water		38000
Suspended Particles-Water (Inflow)		38000
Sediment-Water		21000
Resuspended Sediment-Water		21000

Figure A2. QWASI input – Chemical properties.

Environment Name	Lake Sakakawea		<input type="button" value="Clear Form"/>	
Dimensions				
Water Surface Area (m ²)		1.23E+09		
Water Volume (m ³)		27999996000		
Sediment Active Layer Depth (m)		0.005		
Concentration of Solids				
Aerosols in Air (µg/m ³)		24		
in Water Column (mg/L)		5		
in Inflow Water (mg/L)		30		
in Sediment (vol/vol)		0.15		
Density (kg/m³)				
Aerosols		1000		
Particles in Water		1400		
Sediment Solids		1400		
Organic Carbon (OC) Content (mass/mass)				
Particles in Water		0.4		
Particles in Water (inflow)		0.4		
Sediment Solids		0.03		
Resuspended Sediment		0.025		
Rates				
Water Inflow (m ³ /h)		1536000		
Water Outflow (m ³ /h)		5880000		
Sedimentation (g/m ² -day)		1.2		
Sediment Burial (g/m ² -day)		0.4		
Sediment Resuspension (g/m ² -day)		0.6		
Aerosol Deposition (m/h)		7.2		
Scavenging Ratio (vol air/ vol rain)		133000		
Rain Rate (m/year)		0.4		
Mass Transfer Coefficients (m/h)				
Volatilization - Air Side		1		
Volatilization - Water Side		0.01		
Sediment-Water Diffusion		0.0004		
Additional Comments				
Click Add to DB to add this environment to the Environment				
<input type="button" value="Add to DB"/>				

Figure A3. QWASI input – Environmental properties.

EMISSION RATES

Directly Discharged into Water	0	kg/year	0.000E+00	mol/h
Concentration in Inflow Water	0.54	ng/L	2.389E-09	mol/m ³
Concentration in Air	0.00E+00	ng/m ³	0.000E+00	mol/m ³

Figure A4. QWASI input – Emission rate.

QWASI RESULTS

Chemical Name Radium-226
Environment Name Lake Sakakawea

Fugacity Ratio 1.44455E-07

Subcooled Liq. Vap. Press. 6.92258E-16 Pa

Partition Coefficients	dimensionless	L/kg
Suspended Particles-Water	5.32E+04	3.80E+04
Suspended Particles-Water (Inflow)	5.32E+04	3.80E+04
Sediment-Water	2.94E+04	2.10E+04
Resuspended Sediment-Water	2.94E+04	2.10E+04

	Volume Fraction	Volume	Z Value	VZ
		m ³	mol/m ³ .Pa	mol/Pa
Air: Bulk	-	-	4.10E-04	-
Gas Phase	1.00E+00	-	4.10E-04	-
Aerosols	2.40E-11	-	0.00E+00	-
Water: Bulk	-	2.80E+10	1.19E+10	3.33E+20
Liquid Phase	1.00E+00	2.80E+10	1.00E+10	2.80E+20
Suspended Particles	3.57E-06	1.00E+05	5.32E+14	5.32E+19
Inflow Water: Bulk	-	-	2.14E+10	-
Liquid Phase	1.00E+00	-	1.00E+10	-
Suspended Particles	2.14E-05	-	5.32E+14	-
Sediment: Bulk	-	6.15E+06	4.41E+13	2.71E+20
Pore Water	8.50E-01	5.23E+06	1.00E+10	5.23E+16
Solids	1.50E-01	9.23E+05	2.94E+14	2.71E+20
Resuspended Sediment	-	-	2.94E+14	-
Rain	-	-	1.00E+10	-

Figure A5. QWASI output – Results part 1.

Advection, Reaction and Intercompartmental Transfers	Flow Rate (G) m ³ /h	D Value mol/Pa·h
Water Inflow	1.54E+06	1.54E+16
Suspended Particle Inflow	3.29E+01	1.75E+16
Water Outflow	5.88E+06	5.88E+16
Suspended Particle Outflow	2.10E+01	1.12E+16
Rain Dissolution and Deposition	5.62E+04	5.62E+14
Dry Aerosol Deposition	2.13E-01	0.00E+00
Wet Aerosol Deposition	1.79E-01	0.00E+00
Volatilization	0.00E+00	0.00E+00
Sedimentation	4.39E+01	2.34E+16
Water-Sediment Diffusion	4.92E+05	4.92E+15
Sediment Resuspension	2.20E+01	6.46E+15
Sediment Burial	1.46E+01	4.31E+15
Reaction in Water	-	1.65E+13
Reaction in Sediment	-	1.34E+13

Rate Constants	h⁻¹
Reaction in Water	4.95E-08
Reaction in Sediment	4.95E-08

Total D values	mol/Pa·h
From Air	5.62E+14
From Water	9.83E+16
Inflow	3.29E+16
From Sediment	1.57E+16

Intercompartmental D Values		mol/Pa·h
air to water		5.62E+14
water to air		0.00E+00
water to sediment		2.83E+16
sediment to water		1.14E+16

	Fugacity Pa	Activity
Air: Bulk	0.00E+00	0.00E+00
Water: Bulk	4.72E-20	6.82E-05
Inflow Water: Bulk	1.12E-19	1.61E-04
Sediment: Bulk	8.51E-20	1.23E-04

Total Chemical Mass in Water-Sediment System							
	3.88E+01	mol					
	8.77E+00	kg					

	Concentrations			Amounts			
	mol/m ³	g/m ³	µg/g dry wgt	mol	kg	%	
Air: Bulk	0.00E+00	0.00E+00	-	-	-	-	
Gas Phase	0.00E+00	0.00E+00	-	-	-	-	
Aerosols	0.00E+00	0.00E+00	0.00E+00	-	-	-	
Water: Bulk	5.62E-10	1.27E-07	-	1.57E+01	3.55E+00	4.05E+01	
Liquid Phase	4.72E-10	1.07E-07	-	1.32E+01	2.99E+00	3.41E+01	
Suspended Particles	2.51E-05	5.67E-03	4.05E-03	2.51E+00	5.67E-01	6.47E+00	
Inflow Water: Bulk	2.39E-09	5.40E-07	-	-	-	-	
Liquid Phase	1.12E-09	2.52E-07	-	-	-	-	
Suspended Particles	5.94E-05	1.34E-02	9.59E-03	-	-	-	
Sediment: Bulk	3.75E-06	8.48E-04	-	2.31E+01	5.21E+00	5.95E+01	
Pore Water	8.51E-10	1.92E-07	-	4.45E-03	1.00E-03	1.15E-02	
Solids	2.50E-05	5.65E-03	4.04E-03	2.31E+01	5.21E+00	5.95E+01	
Rain	0.00E+00	0.00E+00	-	-	-	-	

Figure A6. QWASI output – Results part 2.

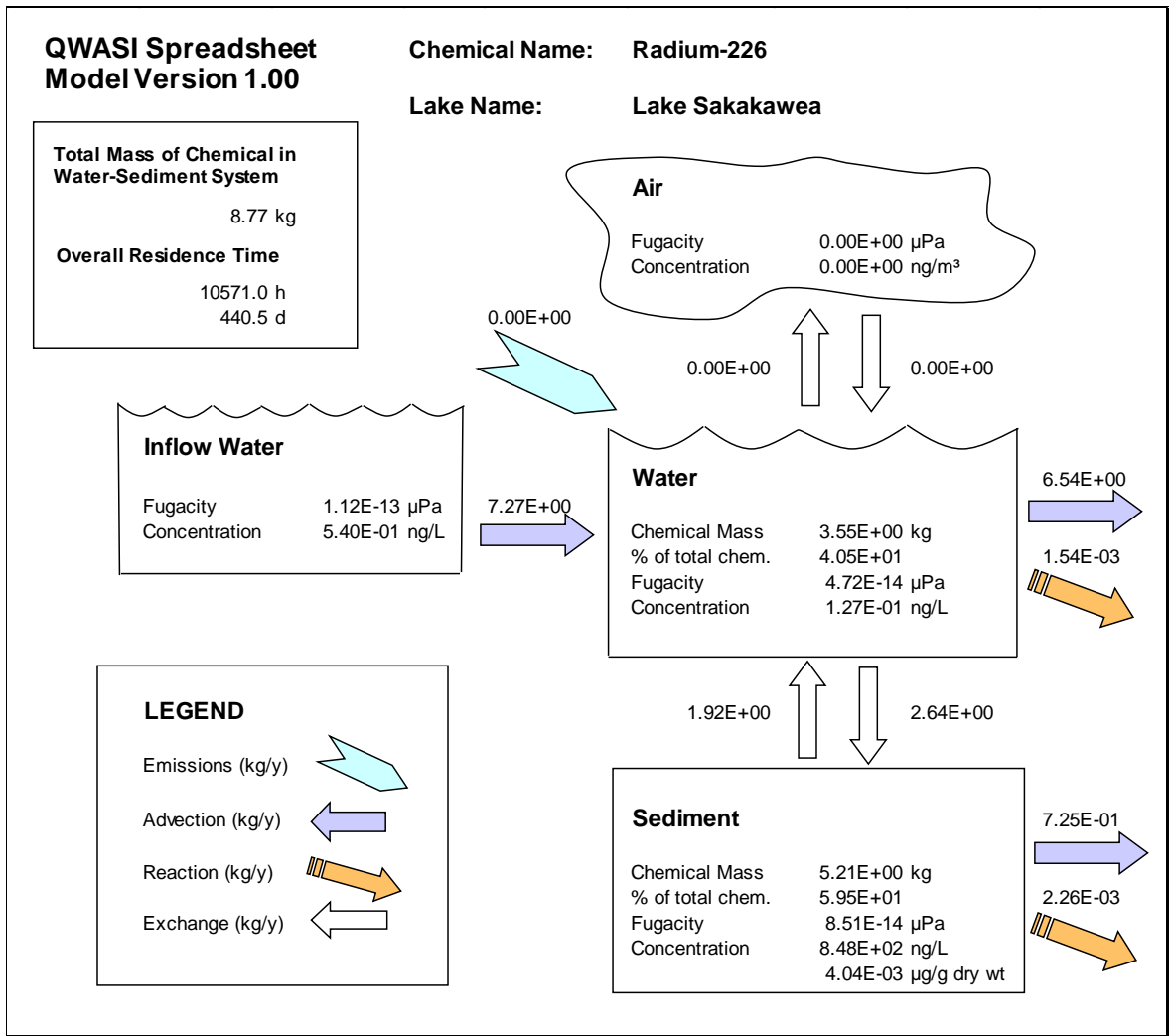


Figure A8. QWASI output – Diagram.

A.5. Scenario 2: Ra-226 Transferred to Potatoes

Table A5 shows the results from the statistical analysis of the data on Ra-226 concentration in soil obtained from Lauer et al. (2016). One data point ($x=2,802$ Bq/kg) was not used since it was treated as an extreme event.

Table A5. Summary of statistical analysis of Ra-226 in soil from ND.

One Variable Summary	Ra-226 in soil (Bq/kg)
Mean	100.29
Variance	43496.21
Std. Dev.	208.56
Skewness	3.89
Kurtosis	19.01
Median	26.00
Mean Abs. Dev.	106.27
Mode	18.00
Minimum	12.00
Maximum	959.00
Range	947.00
Count	21.00
Sum	2106.00
1st Quartile	21.00
3rd Quartile	70.00
Interquartile Range	49.00

Results of @Risk best fit analysis of data on Ra-226 in soil from ND (Table A5):

- Distribution Type: Inverse Gaussian
- Mean: 100.29 Bq/kg
- Shift Factor: 9.6284
- Distribution Shape Parameter: 16.70 Bq/kg

Table A6 summarizes the sources used to calculate the mean and standard deviation of the soil-to-plant transfer factor and the water-to-plant transfer factor.

Table A6. Soil-to-Plant and Water-to-Plant transfer factors for Ra-226.

Transfer Factor	Source
Soil-to-plant	
1.00E-02	(Carvalho et al., 2009)
1.80E-03	(Tagami and Uchida, 2009)
1.60E-03	
2.00E-03	(Staven et al., 2003)
3.00E-03	(Watson et al., 1983)
1.12E-02	(Pietrzak-Flis et al., 1995)
1.10E-02	(Schuttelkopf and Kiefer, 1982)
Water-to-plant	
4.80E+00	(Carvalho et al., 2009)
4.60E+00	

Statistical analysis results using the data in Table A6:

- Soil-to-Plant Transfer Factor (Bq/kg per Bq/kg soil)
 - Distribution Type: Normal (assumed)
 - Mean: 5.8×10^{-3}
 - Standard deviation: 1.8×10^{-3}

- Water-to-Plant Transfer Factor (Bq/kg per Bq/L)
 - Distribution Type: Normal (assumed)
 - Mean: 4.70
 - Standard deviation: 0.14

Data from Tables A5 and A6 and Equation A1 were used in @Risk to simulate 100,000 times the concentration of Ra-226 transferred to potatoes.

$${}^{226}\text{Ra}_{potato} = (TF_{soil-plant} \times {}^{226}\text{Ra}_{soil}) + (TF_{water-plant} \times {}^{226}\text{Ra}_{water}) \quad (\text{A1})$$

A.6. Scenario 3: Ra-226 Bioaccumulated in Fish

Table A7 shows the six different transfer factors found in the literatures that were used in the simulation of scenario 3.

Table A7. Water-to-Fish transfer factors for Ra-226.

Water-to-Fish Transfer Factor	Source
3.59E+00	(Meinhold et al., 1996)
5.00E+00	(IAEA, 2001)
4.38E+00	(Hosseini et al., 2008)
1.10E+01	(Clulow et al., 1998)
1.05E+01	
5.00E+00	

Statistical analysis results using the data in Table A7:

- Distribution Type: Normal (assumed)
- Mean: 6.60
- Standard deviation: 3.30

Data from Table A7 and Equation A2 were used in @Risk to simulate 100,000 times the concentration of Ra-226 bioaccumulated in the fish tissue.

$${}^{226}\text{Ra}_{\text{fish}} = TF_{\text{water-fish}} \times {}^{226}\text{Ra}_{\text{water}} \quad (\text{A2})$$

**APPENDIX B. SUPPORTING MATERIAL FOR CHAPTER 4:
HOLISTIC RISK ASSESSMENT OF SURFACE WATER CONTAMINATION DUE TO Pb-
210 IN OIL PRODUCED WATER FROM THE BAKKEN SHALE**

B.1. Sources of Data

B.1.1. TDS

The chemical quality of the produced water in the Bakken was obtained from the USGS Produced Water Database (Blondes et al., 2016). The database was filtered to just ND to obtain the 7,907 TDS data points used in the analysis. An example of 12 data points used is shown in Table B1.

Table B1. Example of data on TDS levels in produced water from ND used in the analysis.

IDUSGS	IDORIG	IDDB	SOURCE	BASIN	STATE	TDS
27682	33000001	USGSMAN	PAN AMERICAN PETROLEUM CORPORATION	WILLISTON BASIN	ND	42,299
27683	33000002	USGSMAN	PAN AMERICAN PETROLEUM CORPORATION	WILLISTON BASIN	ND	230,908
27684	33000003	USGSMAN	PAN AMERICAN PETROLEUM CORPORATION	WILLISTON BASIN	ND	98,856
27685	33000004	USGSMAN	PAN AMERICAN PETROLEUM CORPORATION	WILLISTON BASIN	ND	80,934
27686	33000005	USGSMAN	STANOLIND OIL AND GAS COMPANY	WILLISTON BASIN	ND	383,980
27687	33000006	USGSMAN	STANOLIND OIL AND GAS COMPANY	WILLISTON BASIN	ND	387,060
27688	33000007	USGSMAN	STANOLIND OIL AND GAS COMPANY	WILLISTON BASIN	ND	213,135
27689	33000008	USGSMAN	STANOLIND OIL AND GAS COMPANY	WILLISTON BASIN	ND	331,130
27690	33000009	USGSMAN	STANOLIND OIL AND GAS COMPANY	WILLISTON BASIN	ND	345,460
27691	33000010	USGSMAN	STANOLIND OIL AND GAS COMPANY	WILLISTON BASIN	ND	335,296
27692	33000017	USGSMAN	STANOLIND OIL AND GAS COMPANY	WILLISTON BASIN	ND	307,735
27693	33000018	USGSMAN	STANOLIND OIL AND GAS COMPANY	WILLISTON BASIN	ND	319,626

B.1.2. Produced water spills

The environmental incident reports available at the ND DoH website (ND DoH, 2015) were used to determine the most common causes of produced water spills and the frequency of these scenarios. A sample of the spills obtained from the ND DoH database is shown in Table B2.

Table B2. Example of produced water spill reports in ND used in the analysis. SW = Saltwater, Vol. = Volume.

Incident ID	Date Reported	Date Incident	County	Oil Vol.	Oil Units	SW Vol.	SW Units	Contained
20151005100644	10/5/2015	10/4/2015	Williams			100	Barrels	Yes
20151005085231	10/5/2015	10/4/2015	Billings	2	Barrels	2	Barrels	Yes
20151005164310	10/5/2015	10/4/2015	Divide	30	Barrels			Yes
20151005094755	10/5/2015	10/4/2015	Divide	150	Barrels			Yes
20151005164545	10/5/2015	10/4/2015	McKenzie					Yes
20151003172900	10/3/2015	10/3/2015	McKenzie	10	Barrels			Yes
20151003180124	10/3/2015	10/2/2015	Williams					Yes
20151003100622	10/3/2015	10/1/2015	Williams	1	Barrels			Yes
20151002080758	10/2/2015	10/1/2015	Bowman	5	Barrels			Yes
20151002150106	10/2/2015	9/30/2015	Divide					Yes
20151001124756	10/1/2015	9/29/2015	Bowman	4	Barrels	9	Barrels	Yes
20151001143042	10/1/2015	9/24/2015	Williams					Yes
20150930155417	9/30/2015	9/30/2015	Burke	2	Gallons	5	Barrels	Yes
20150930140819	9/30/2015	9/29/2015	Mountrail			3	Barrels	Yes
20150930145326	9/30/2015	9/29/2015	McKenzie			1	Barrels	Yes
20150930152830	9/30/2015	9/30/2015	McKenzie	1	Barrels	0	Barrels	Yes
20150930110503	9/30/2015	9/29/2015	Dunn					Yes

Table B2. Example of produced water spill reports in ND used in the analysis (continued). SW = Saltwater, Vol. = Volume.

Incident ID	Date Reported	Date Incident	County	Oil Vol.	Oil Units	SW Vol.	SW Units	Contained
20150930155800	9/30/2015	9/29/2015	Dunn					Yes
20150929110011	9/29/2015	9/29/2015	McKenzie	15	Barrels			Yes
20150706142628	7/6/2015	7/6/2015	Dunn	6	Barrels	2	Barrels	No
20150520164421	5/20/2015	5/20/2015	Burke	3	Barrels	2	Barrels	No
20150514174342	5/14/2015	5/14/2015	Williams	1	Gallons	1	Gallons	No
20150506143342	5/6/2015	5/4/2015	Burke	5	Barrels	1	Barrels	No
20150430144112	4/30/2015	4/29/2015	Williams	1	Barrels	1	Barrels	No
20150415135347	4/15/2015	4/14/2015	Williams	2	Gallons	2	Gallons	No
20150319104347	3/19/2015	3/19/2015	Williams			1	Barrels	No
20150225131537	2/25/2015	2/24/2015	Burke			1	Barrels	No
20150225121205	2/25/2015	2/25/2015	Williams	4	Barrels	1	Barrels	No
20150123160451	1/23/2015	1/23/2015	Burke	6	Barrels	2	Barrels	No
20141105160942	11/5/2014	11/5/2014	Williams	1	Barrels	1	Barrels	No

B.1.3. Truck accidents in ND

The ND DoT report entitled “2014 North Dakota Crash Summary” was analyzed to determine the number of crashes involving trucks and truck tractors (ND DoT, 2015). Unfortunately, information of accidents involving vehicles transporting produced water specifically was not available. The ND DoT was contacted to obtain data regarding the distribution of these accidents in the different Counties in the State (R. Hair, personal communication, August 21, 2016). Figure B1 shows the accidents involving trucks and truck tractors that occurred in ND between 2009 and 2014. Figure B2 shows the number of accidents that occurred in each of the 17 oil-producing Counties in the State.

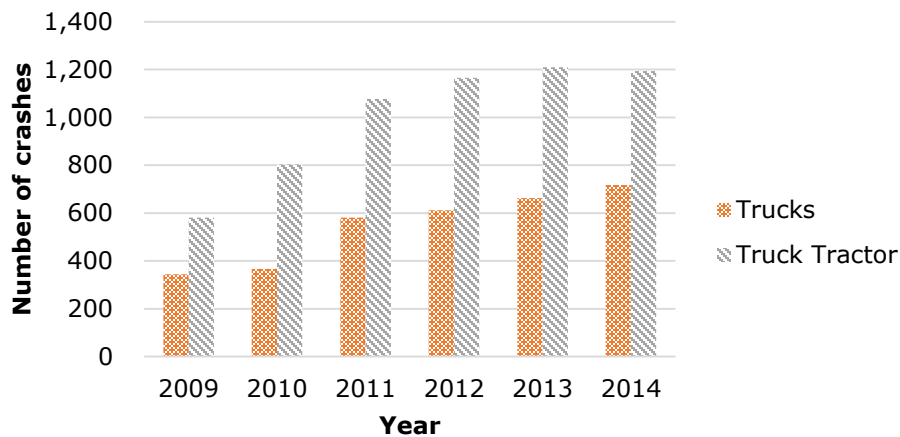


Figure B1. Number of crashes involving trucks and truck tractors from 2009-2014 in ND.

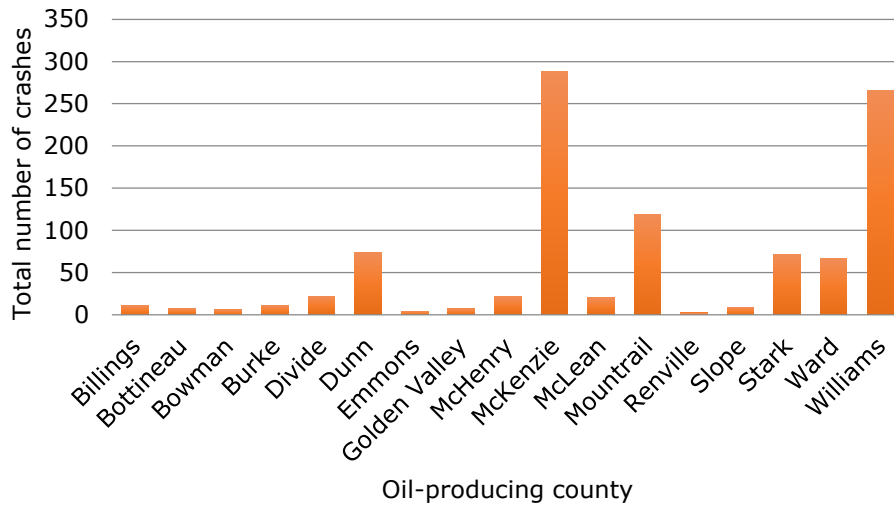


Figure B2. Number of truck and truck tractor crashes in oil-producing counties in 2014.

B.2. Determination of Pb-210 concentrations

B.2.1. Estimation of Ra-226

A generalized regression model for Ra-226 and TDS was developed using three different datasets. Table B3 summarizes the information about each dataset. The results from the generalized regression model using the total 331 data points are shown in Table B4. The results from ANOVA used to determine the generalized regression model, or the Model, are shown in Table B5.

Table B3. Datasets used to develop the Model.

Dataset	Number of Data Points	Source
Pennsylvania and New York	65	(Rowan et al., 2011)
Texas	142	(Fisher, 1995)
Pennsylvania and Wyoming	124	(Blondes et al., 2016)
Total data points	331	

Table B4. Summary of the regression statistics.

Summary Output	
Multiple R	0.896920328
R Square	0.804466074
Adjusted R Square	0.803871746
Standard Error	0.610527047
Observations	331

Table B5. Summary of ANOVA results.

ANOVA	Degrees of freedom	SS	MS	F	Significance F
Regression	1	504.535035	504.5350355	1353.57246	1.2E-118
Residual	329	122.632537	0.372743276		
Total	330	627.167573			

	Coeff.	Std. Error	t-Stat	P-value	Lower 95%	Upper 95%	Lower 95.0%	Upper 95.0%
Intercept	-3.3337	0.1341	-24.854	1.667E-77	-3.597	-3.069	-3.597	-3.069
Log (TDS)	1.11828	0.0303	36.790	1.24E-118	1.0584	1.1780	1.0584	1.1780

Using the results from the ANOVA analysis, the Model was determined as follows:

$$\mathbf{Log(Ra226) = 1.12 \times Log(TDS) - 3.33} \quad \text{(B1)}$$

The Model was applied to the TDS levels from ND (n=7,907) to calculate the Ra-226 levels. An example of some simulated Ra-226 values obtained from the Model is shown in Table B6.

Table B6. Selected simulated Ra-226 concentrations.

TDS	$\text{Log}(\text{Ra226}) = 1.12 \times \text{Log}(\text{TDS}) - 3.33$	Ra-226
35,047	1.76	57.55
35,074	1.76	57.59
35,247	1.76	57.91
35,368	1.76	58.14
35,544	1.77	58.46
35,669	1.77	58.69
35,897	1.77	59.11
35,940	1.77	59.19
35,940	1.77	59.19
36,014	1.77	59.33
36,067	1.77	59.42
36,070	1.77	59.43
36,280	1.78	59.82
36,300	1.78	59.85
36,335	1.78	59.92
36,374	1.78	59.99
36,649	1.78	60.50
37,000	1.79	61.15
37,300	1.79	61.70

The software @Risk® was used to analyze the simulated Ra-226 and to find the best distribution that represented the 7,907 data points. Figure B3 shows the probability distribution obtained with @Risk®.

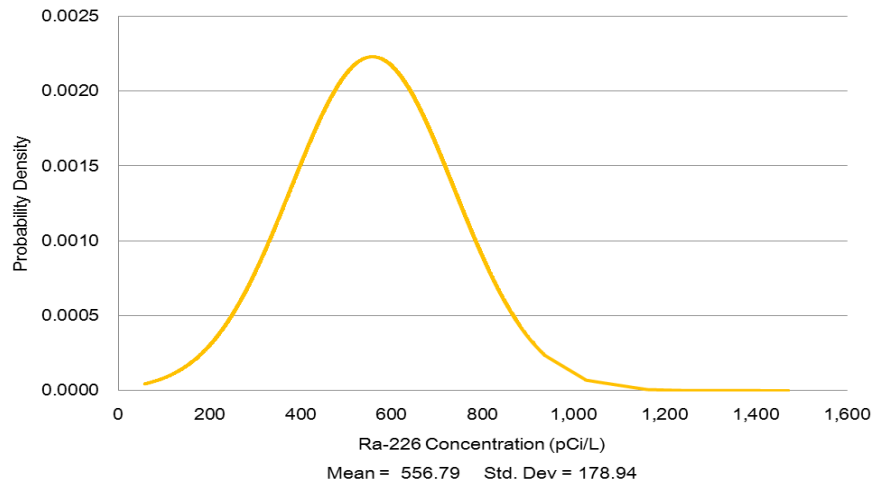


Figure B3. 7,907 data points fitted to obtain Ra-226 probability distribution.

B.2.2. Estimation of Pb-210

The Pb-210 levels were obtained using the Ra-226 distribution and the Rutherford-Soddy's decay law equation. Once again, @Risk® was used to calculate the Pb-210 levels by applying 100,000 Monte Carlo iterations and bootstrapping (1,000). Figure B4 shows a screenshot of the model setup used with @Risk®. Cell B6, Ra-226 concentration, varied according to the mean and the standard deviation shown in Figure B3. Cell E6, final Pb-210 concentration, was calculated using the decay equation and the initial Ra-226 level (cell B6). Figure B5 shows the first 10 simulated results out of the 100,000 data points obtained.

	A	B	C	D	E	F
1	Half-life	22.2				
2	Lambda (λ)	0.031223				
3						
4	$N(t) = N_0 e^{-\lambda t}$					
5				<i>t in years</i>	1	
6	Ra-226	556.7916		Pb-210	539.6756	
7						

Figure B4. Model setup to simulate Pb-210 based on lambda, $t = 1$ year, and initial Ra-226 concentration.

@RISK Data	
Performed By: Luisa	
Date: Sunday, April 10 2016 03:26:41 p.m.	
Name	Pb-210 1 year
Description	Output
Iteration / Cell	E7
1	389.344532
2	575.1920423
3	626.1452702
4	619.9275651
5	612.6542206
6	486.5044013
7	765.6020135
8	440.7518143
9	503.0432519
10	507.5022253

Figure B5. Example of @Risk® output for Pb-210 levels.

Figure B6 shows the probability distribution of the 100,000 simulated data points for Pb-210.

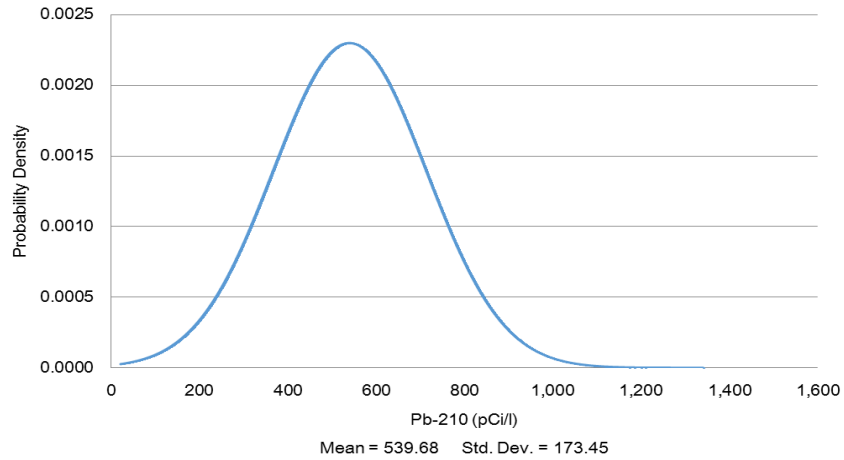


Figure B6. 100,000 data points fitted to obtain Pb-210 probability distribution.

Figures B3 and B6 are very similar because Ra-226 was used to generate Pb-210 and since the decay period of one year is not significant there is not a considerable change between the initial (parent radionuclide) and final (daughter radionuclide) concentrations.

B.3. Risk assessment

B.3.1. Exposure assessment

The exposure to Pb-210 by oral ingestion was calculated based on the average initial concentration of 540 pCi/L after one year of produced water generation. The actual exposure to Pb-210 is the concentration that remains after the contaminant undergoes different chemical and physical processes, which happen naturally, and after the surface water is treated by different methods. The QWASI model was used to calculate the final concentration of Pb-210 in Lake Sakakawea. Table B7 summarizes the inputs and the sources used in the QWASI simulation and Figures B7-B9 show how the model was set up using the spreadsheet model version 1.00. Figures B10 and B11 show the complete results from the QWASI simulation.

Table B7. Summary of inputs used in the QWASI model to simulate the final Pb-210 concentration.

Environmental properties of Lake Sakakawea			Environmental properties of Lake Sakakawea (Continued)		
Lake Dimensions	Value	Source	Rates	Value	Source
Water surface area (m ²)	1.23×10 ⁹	(Bolgrien et al., 2009)	Aerosol deposition (m/h)	7.2	(Mackay and Diamond, 1989)
Water volume (m ³)	27.99×10 ⁹	(ND DoH, 2001)	Scavenging ratio (air _{vol} /rain _{vol})	133,000	
Sediment active layer depth (m)	0.005	Default	Rain rate (m/year)	0.4	(Natural Resources Conservation Service, 2008)
Concentration of Solids	Value	Source	Mass transfer coefficients (MTC) (m/h)	Value	Source
In water column (mg/L)	5	(Bolgrien et al., 2009)	Air side coefficient	1	(Mackay and Diamond, 1989)
In inflow water (mg/L)	30		Water side coefficient	0.01	
Of aerosol in air (µg/m ³)	24	(Johnson, 2006)	Sediment-water diffusion	0.0004	
In sediment (m ³ /m ³)	0.15	Default	Chemical properties of Pb-210		
Density of solids (kg/m ³)	Value	Source	Parameter	Value	Source
In water	1,400	(U.S. Army Corps of Engineers, 2009)	Molar mass (g/mol)	210	(GSI Environmental, 2014)
In sediment	1,400		Data temperature (°C)	20	
In aerosols	1,000	(Hinds, 1999)	Melting point (°C)	327	
OC fraction of solids	Value	Source	Vapor pressure (Pa)	1×10 ⁻⁷	(Bradley et al., 2014)
In water column	0.4	Default	Solubility in water (g/m ³)	1×10 ⁻⁶	
In sediment	0.03	Default	Henry's Law constant	1×10 ^{-10 a}	Assumed
In inflow water	0.4	Default	Reaction half-life in water and sediment (h)	192,720	(Prince, 1979)
In resuspended sediment	0.025	Default	Log KOW	0.729	(GSI Environmental, 2014)
Rates	Value	Source	Air-water (KAW)	0	Assumed
Water inflow (m ³ /h)	1.54×10 ⁶	(U.S. Army Corps of Engineers, 2013)	Aerosol-air (KQA)	0	Assumed
Water outflow (m ³ /h)	5.88×10 ⁶		KOC	1	(GSI Environmental, 2014)
Sedimentation (g/m ² · day)	1.2	Default	Emissions of Pb-210		
Sedimentation burial (g/m ² · day)	0.4	Default	Parameter	Value	Source
Sedimentation resuspension (g/m ² · day)	0.6	Default	Concentration in inflow water (ng/L)	7.04×10 ^{-3 b}	Based on 540 pCi/L

Footnotes for Table B5.

^a For metals, the Henry's Law constant is equal to zero (Illinois EPA, 2015) but QWASI requires the user to introduce a non-zero value.

^b Conversion from pCi/L to ng/L:

Initial concentration = 540 pCi/L

Molar mass of Pb-210 = 210 g/mol

Pb-210 half life in years = 22.2

Pb-210 half life in seconds = 7.001×10^8

Decay constant = 9.90×10^{-10}

Avogadro's number = 6.02×10^{23}

1. Convert pCi/L to disintegrations/s

$$dps = 540 \text{ pCi/L} * \frac{0.037 \text{ Bq/L}}{1 \text{ pCi/L}} = 19.98 \text{ dps}$$

2. Calculate the number of atoms

$$\# \text{ atoms} = \frac{19.98 \text{ dps}}{\text{decay constant}} = 2.02 \times 10^{10} \text{ atoms}$$

3. Calculate the amount of moles

$$\text{Moles} = \frac{\# \text{ atoms}}{\text{Avogadro's number}} = 3.35 \times 10^{-14} \text{ moles}$$

4. Convert moles to mass

$$3.35 \times 10^{-14} \text{ moles} * \frac{210 \text{ g}}{\text{mol}} = 7.04 \times 10^{-12} \text{ g} = 0.00704 \text{ ng}$$

ENVIRONMENTAL PROPERTIES

Please complete all fields or select an environment from the database:

Environment Name:

Dimensions	
Water Surface Area (m ²)	1.23E+09
Water Volume (m ³)	27999996000
Sediment Active Layer Depth (m)	0.005

Concentration of Solids	
Aerosols in Air (µg/m ³)	24
in Water Column (mg/L)	5
in Inflow Water (mg/L)	30
in Sediment (vol/vol)	0.15

Density (kg/m ³)	
Aerosols	1000
Particles in Water	1400
Sediment Solids	1400

Organic Carbon (OC) Content (mass/mass)	
Particles in Water	0.4
Particles in Water (inflow)	0.4
Sediment Solids	0.03
Resuspended Sediment	0.025

Rates	
Water Inflow (m ³ /h)	1536000
Water Outflow (m ³ /h)	5880000
Sedimentation (g/m ² -day)	1.2
Sediment Burial (g/m ² -day)	0.4
Sediment Resuspension (g/m ² -day)	0.6
Aerosol Deposition (m/h)	7.2
Scavenging Ratio (vol air/ vol rain)	133000
Rain Rate (m/year)	0.4

Mass Transfer Coefficients (m/h)	
Volatilization - Air Side	1
Volatilization - Water Side	0.01
Sediment-Water Diffusion	0.0004

Additional Comments:

Click **Add to DB** to add this environment to the Environment

Figure B7. QWASI model setup - Environmental properties.

CHEMICAL PROPERTIES

← hover cursor over red triangle for info
 Please complete all required fields below
 or
 Select a chemical from the database:

* indicates optional input

Chemical Name		References and Notes
CAS	Lead-210	
Molar Mass (g/mol)	210	
Data Temperature (°C)	20	
* Melting Point (°C)	327	
* Vapor Pressure (Pa)	1.00E-07	
* Solubility in Water (g/m ³)	1.00E-06	
Henry's Law Constant (Pa·m ³ /mol)	1.00E-10	
Reaction Half-Lives (hours)		
In Water	1.93E+05	
In Sediment	1.93E+05	
Partition Coefficients		
	unitless	
* logK _{OW}	0.729	
Air-Water (K _{AW})	0.00E+00	calculated
Aerosol-Air (K _{QA})	0.00E+00	
Aerosol-Water (K _{QW})		

Figure B8. QWASI model setup - Chemical properties.

EMISSION RATES				
Directly Discharged into Water	0	kg/year	0.000E+00	mol/h
Concentration in Inflow Water	0.00704	ng/L	3.352E-11	mol/m ³
Concentration in Air	0	ng/m ³	0.000E+00	mol/m ³
Additional Comments				

Figure B9. QWASI model setup – Emission rates.

QWASI RESULTS				
Chemical Name	Lead-210			
Environment Name	Lake Sakakawea			
Fugacity Ratio	0.000816245			
Subcooled Liq. Vap. Press.	0.000122512 Pa			
Partition Coefficients	dimensionless	L/kg		
Suspended Particles-Water	5.60E-01	4.00E-01	(estimated using Koc and OC content of suspended particles)	
Suspended Particles-Water (Inflow)	5.60E-01	4.00E-01	(estimated using Koc and OC content of inflow particles)	
Sediment-Water	4.20E-02	3.00E-02	(estimated using Koc and OC content of sediment solids)	
Resuspended Sediment-Water	3.50E-02	2.50E-02	(estimated using Koc and OC content of resuspended sediment)	
	Volume Fraction	Volume	Z Value	
		m ³	mol/m ³ .Pa	
			VZ	
			mol/Pa	
Air: Bulk	-	-	4.10E-04	-
Gas Phase	1.00E+00	-	4.10E-04	-
Aerosols	2.40E-11	-	0.00E+00	-
Water: Bulk	-	2.80E+10	1.00E+10	2.80E+20
Liquid Phase	1.00E+00	2.80E+10	1.00E+10	2.80E+20
Suspended Particles	3.57E-06	1.00E+05	5.60E+09	5.60E+14
Inflow Water: Bulk	-	-	1.00E+10	-
Liquid Phase	1.00E+00	-	1.00E+10	-
Suspended Particles	2.14E-05	-	5.60E+09	-
Sediment: Bulk	-	6.15E+06	8.56E+09	5.27E+16
Pore Water	8.50E-01	5.23E+06	1.00E+10	5.23E+16
Solids	1.50E-01	9.23E+05	4.20E+08	3.87E+14
Resuspended Sediment	-	-	3.50E+08	-
Rain	-	-	1.00E+10	-
Advection, Reaction and Intercompartmental Transfers	Flow Rate (G)	D Value		
	m ³ /h	mol/Pa-h		
Water Inflow	1.54E+06	1.54E+16		
Suspended Particle Inflow	3.29E+01	1.84E+11		
Water Outflow	5.88E+06	5.88E+16		
Suspended Particle Outflow	2.10E+01	1.18E+11		
Rain Dissolution and Deposition	5.62E+04	5.62E+14		
Dry Aerosol Deposition	2.13E-01	0.00E+00		
Wet Aerosol Deposition	1.79E-01	0.00E+00		
Volatilization	0.00E+00	0.00E+00		
Sedimentation	4.39E+01	2.46E+11		
Water-Sediment Diffusion	4.92E+05	4.92E+15		
Sediment Resuspension	2.20E+01	7.69E+09		
Sediment Burial	1.46E+01	6.15E+09		
Reaction in Water	-	1.01E+15		
Reaction in Sediment	-	1.89E+11		
	Rate Constants	h ⁻¹		
		mol/Pa-h		
	Reaction in Water	3.60E-06		
	Reaction in Sediment	3.60E-06		
	Total D values	mol/Pa-h		
	From Air	5.62E+14		
	From Water	6.47E+16		
	Inflow	1.54E+16		
	From Sediment	4.92E+15		
	Intercompartmental D Values	mol/Pa-h		
	air to water	5.62E+14		
	water to air	0.00E+00		
	water to sediment	4.92E+15		
	sediment to water	4.92E+15		
	Fugacity	Activity		
	Pa			
Air: Bulk	0.00E+00	0.00E+00		
Water: Bulk	8.61E-22	7.03E-18		
Inflow Water: Bulk	3.35E-21	2.74E-17		
Sediment: Bulk	8.61E-22	7.03E-18		
	Total Chemical Mass in Water-Sediment System			
		2.41E-01	mol	
		5.06E-02	kg	

Figure B10. Complete results from the QWASI model – Part 1.

	Concentrations			Amounts		
	mol/m ³	g/m ³	µg/g dry wtg	mol	kg	%
Air: Bulk	0.00E+00	0.00E+00	-	-	-	-
Gas Phase	0.00E+00	0.00E+00	-	-	-	-
Aerosols	0.00E+00	0.00E+00	0.00E+00	-	-	-
Water: Bulk	8.61E-12	1.81E-09	-	2.41E-01	5.06E-02	1.00E+02
Liquid Phase	8.61E-12	1.81E-09	-	2.41E-01	5.06E-02	1.00E+02
Suspended Particles	4.82E-12	1.01E-09	7.23E-10	4.82E-07	1.01E-07	2.00E-04
Inflow Water: Bulk	3.35E-11	7.04E-09	-	-	-	-
Liquid Phase	3.35E-11	7.04E-09	-	-	-	-
Suspended Particles	1.88E-11	3.94E-09	2.82E-09	-	-	-
Sediment: Bulk	7.37E-12	1.55E-09	-	4.53E-05	9.52E-06	1.88E-02
Pore Water	8.61E-12	1.81E-09	-	4.50E-05	9.45E-06	1.87E-02
Solids	3.62E-13	7.59E-11	5.42E-11	3.34E-07	7.01E-08	1.38E-04
Rain	0.00E+00	0.00E+00	-	-	-	-

MASS BALANCES		
System	kg/year	mol/h
Total Chemical Inputs	9.47E-02	5.15E-05
Emission	0.00E+00	0.00E+00
Inflow	9.47E-02	5.15E-05
Air to water transfer	0.00E+00	0.00E+00
Total Chemical Losses	9.47E-02	5.15E-05
Outflow	9.31E-02	5.06E-05
Water to air transfer	0.00E+00	0.00E+00
Total Transformation	1.60E-03	8.67E-07
Sediment Burial	9.74E-09	5.30E-12

Residence Time	h	d
Water	4.68E+03	1.95E+02
Sediment	2.69E+05	1.12E+04
System	4.68E+03	1.95E+02

Water	kg/year	mol/h
Total Chemical Inputs	1.03E-01	5.57E-05
Emission	0.00E+00	0.00E+00
Inflow	9.47E-02	5.15E-05
Air to water transfer	0.00E+00	0.00E+00
Sediment to water transfer	7.79E-03	4.24E-06
Total Chemical Losses	1.03E-01	5.57E-05
Outflow	9.31E-02	5.06E-05
Water to air transfer	0.00E+00	0.00E+00
Water to sediment transfer	7.79E-03	4.24E-06
Transformation in water	1.60E-03	8.67E-07

Response Time	h	d
	4.33E+03	1.80E+02

Sediment	kg/year	mol/h
Total Chemical Inputs	7.79E-03	4.24E-06
Water to sediment transfer	7.79E-03	4.24E-06
Total Chemical Losses	7.79E-03	4.24E-06
Sediment to water transfer	7.79E-03	4.24E-06
Transformation in sediment	3.00E-07	1.63E-10
Sediment Burial	9.74E-09	5.30E-12

Response Time	h	d
	1.07E+01	4.46E-01

Rate Details	kg/year	mol/h
Emission to Water	0.00E+00	0.00E+00
Water Inflow	9.47E-02	5.15E-05
Particle Inflow	1.14E-06	6.18E-10
Water Outflow	9.31E-02	5.06E-05
Particle Outflow	1.86E-07	1.01E-10
Rain Dissolution	0.00E+00	0.00E+00
Dry Aerosol Deposition	0.00E+00	0.00E+00
Wet Aerosol Deposition	0.00E+00	0.00E+00
Absorption	0.00E+00	0.00E+00
Volatilization	0.00E+00	0.00E+00
Sedimentation	3.90E-07	2.12E-10
Water-Sediment Diffusion	7.79E-03	4.24E-06
Sediment-Water Diffusion	7.79E-03	4.24E-06
Sediment Resuspension	1.22E-08	6.62E-12
Sediment Burial	9.74E-09	5.30E-12
Water Transformation	1.60E-03	8.67E-07
Sediment Transformation	3.00E-07	1.63E-10

Water Response Times		
	h	d
Water Inflow	1.82E+04	7.60E+02
Particle Inflow	1.52E+09	6.33E+07
Water Outflow	4.76E+03	1.98E+02
Particle Outflow	2.38E+09	9.92E+07
Rain Dissolution	4.99E+05	2.08E+04
Dry Aerosol Deposition	N/A	N/A
Wet Aerosol Deposition	N/A	N/A
Volatilization/Absorption	N/A	N/A
Sedimentation	1.14E+09	4.74E+07
Water-Sediment Diffusion	5.69E+04	2.37E+03
Sediment Resuspension	3.64E+10	1.52E+09
Transformation	2.78E+05	1.16E+04

Sediment Response Times		
	h	d
Sedimentation	2.14E+05	8.92E+03
Water-Sediment Diffusion	1.07E+01	4.46E-01
Sediment Resuspension	6.85E+06	2.85E+05
Sediment Burial	8.56E+06	3.57E+05
Transformation	2.78E+05	1.16E+04

Figure B11. Complete results from the QWASI model – Part 2.

B.3.2. Holistic risk characterization

B.3.2.1. Risk characterization results

The Pb-210 exposure concentration was considered to be 0.63 Bq/L in all scenarios. The likelihood level was determined based on the probability of each scenario. Then, the consequence level and the likelihood level are:

Consequence = 6 times the WHO guidance level, moderate impact (level 2)

Likelihood = See Table B8

Table B8. Level of risk likelihood for each scenario.

Scenario	Probability	Level
1	6%	2
2	89%	4
3	6%	2

Based on a 4 × 4 risk matrix, the risks of scenarios 1 and 3 are considered medium-low (score=4) while scenario 2 is classified as medium-high risk (score=8).

B.3.2.2. Holistic risk characterization results

Tables B9-B12 show the conversion of qualitative results from the survey to quantitative measurements.

Table B9. Transformation of levels of concern into PAF-values.

Concern scale	Value in survey	PAF-value
Not at all concerned	5	2
Slightly concerned	4	1.625
Somewhat concerned	3	1.25
Moderately concerned	2	0.875
Extremely concerned	1	0.5

Table B10. Transformation of levels of familiarity into PAF-values.

Familiarity scale	Value in survey	PAF-value
Not familiar at all	5	2
Slightly familiar	4	1.625
Moderately familiar	3	1.25
Very familiar	2	0.875
Extremely familiar	1	0.5

Table B11. Transformation of levels of awareness into PAF-values.

Familiarity scale	Value in survey	PAF-value
Not at all aware	5	2
Slightly aware	4	1.625
Somewhat aware	3	1.25
Moderately aware	2	0.875
Extremely aware	1	0.5

The risk perception of each stakeholder from the experts' perspective was measured based on the most selected option (Table B12). The transformed PAF-values are: low (2), medium (1.25), and high (0.5).

Table B12. Experts' opinion – Risk perception of each stakeholder.

Scenario	General public	Oil field operators	Truck operators	Average
Storage tank overflow	Medium (1.25)	Low (2)	Low (2)	1.75
Equipment failure	High (0.5)	Medium (1.25)	Medium (1.25)	1
Truck accident	High (0.5)	Low (2)	High (0.5)	1

The holistic risk characterization of each scenario was obtained using the following equation:

$$Risk = R_{tech} + \left[R_{tech} \times \frac{\sum_{i=1}^n (pa_{fi} \cdot a_i)}{\sum_{i=1}^n a_i} \right] \quad (B2)$$

Scenario 1

$$Risk_{perc, sc-1} = 4 + \left[4 \times \frac{(1.625 \times 0.15) + (2 \times 0.20) + (1.625 \times 0.35) + (.75 \times 0.30)}{1} \right] \approx 11$$

Scenario 2

$$Risk_{perc, sc-2} = 8 + \left[8 \times \frac{(0.5 \times 0.15) + (2 \times 0.20) + (1.625 \times 0.35) + (1 \times 0.30)}{1} \right] \approx 19$$

Scenario 3

$$Risk_{perc, sc-3} = 4 + \left[4 \times \frac{(0.5 \times 0.15) + (2 \times 0.20) + (1.625 \times 0.35) + (1 \times 0.30)}{1} \right] \approx 9$$

Since the awareness has 5 levels, a 5 × 5 matrix color code was used to translate the risk levels of each scenario. Scenario 1 and 3 are considered medium-high while scenario 2 is considered high.

**APPENDIX C. SUPPORTING MATERIAL FOR CHAPTER 5: PERCEIVED RISKS OF
PRODUCED WATER MANAGEMENT AND NATURALLY OCCURRING RADIOACTIVE
MATERIAL CONTENT IN NORTH DAKOTA**

C.1. Survey Informed Consent

Dear participant:

My name is Luisa Torres and I am working on a research study with Dr. Eakalak Khan and Dr. Om Yadav through the Department of Civil and Environmental Engineering and the Department of Industrial Engineering, North Dakota State University, respectively.

You are being invited to participate in this research study on perceptions of risk of water contamination due to oil produced water (or fracking wastewater) from the Bakken. We are recruiting adults who have little or in depth knowledge about oil production in North Dakota, work or have worked in the oil fields or know someone that has, and currently is a resident in North Dakota.

The objective of this research is to understand the perception of risk of different interested parties regarding oil production and produced water. Also, to measure awareness among people regarding management of produced water storage and transportation. Participation involves only the completion of an online survey and is entirely voluntary. Based on the extent of your responses, the survey could take between 10 and 15 minutes to complete.

There are no risks associated with this study. While you will not experience any direct benefits from participation, information collected in this study may benefit others in the future by helping to understand the risk perception of oil production in North Dakota.

All of your responses to this survey will remain anonymous and cannot be linked to you in any way. No identifiable information about you will be collected at any point during the study. You may change your mind and cease participation at any point during the completion

of the survey instrument. If you decide to complete the survey, you may choose to retain this consent form for your records. **Your completion of the survey indicates your consent to participate in the survey.**

If you have any questions regarding the survey or this research project in general, please contact Dr. Eakalak Khan at (701) 231-7717 or eakalak.khan@ndsu.edu. If you have questions about the rights of human participants in research, or to report a problem, you may contact the NDSU Human Research Protection Program, at (701) 231-8995, toll-free at 1-855-800-6717 or via email at ndsu.irb@ndsu.edu.

If you would like to complete the survey, please click on "Start survey" below.

Thank you for your participation in this study.

Luisa F. Torres

Graduate student

C.2. Survey Questions

- **Demographic Characteristics**

1. Location: Please select the county in which you reside currently.
2. Age: What is your age?
3. Education: What is the highest level of education you have completed?
 - Less than High School
 - High School/GED
 - 2-year College Degree
 - 4-year College Degree
 - Master's Degree
 - Doctoral Degree
 - Professional Degree (JD, MD)
4. Do you work or have you worked in the oil industry in North Dakota?
 - Yes or No
5. Do you know someone that works or has worked in the oil industry in North Dakota?
 - Yes or No

- **Sources of information**

6. What is the main source you use to get the latest news?
 - Television
 - Internet
 - Print
 - Radio

- **General attitudes and feelings about state and local economics**

7. How satisfied or dissatisfied would you be living or investing in North Dakota?

- 1 – Extremely dissatisfied
- 2 – Somewhat dissatisfied
- 3 – Neither satisfied nor dissatisfied
- 4 – Somewhat satisfied
- 5 – Extremely satisfied

8. Areas where produced water is stored or transported are likely to be unattractive to new residents, business development, and tourist:

- 1 – Strongly disagree
- 2 – Somewhat disagree
- 3 – Neither agree or disagree
- 4 – Somewhat agree
- 5 – Strongly agree

- **Risk perception images**

9. Give the first thought or image that comes to mind when you heard or read “fracking wastewater.”

10. Please rank this thought or image based on the scale ranging from -5 (very negative) to +5 (very positive).

11. Give the first thought or image that comes to mind when you heard or read “natural radioactive material.”

12. Please rank this thought or image based on the scale ranging from -5 (very negative) to +5 (very positive).

- **Awareness of produced water handling and content**

13. How familiar are you with the processes of storage and transportation of produced water?

- 1 – Not at all familiar
- 2 – Slightly familiar
- 3 – Moderately familiar
- 4 – Very familiar
- 5 – Extremely familiar

14. How aware are you with the content of produced water? (e.g. chemicals additives and contaminants)

- 1 – Not at all aware
- 2 – Slightly aware
- 3 – Somewhat aware
- 4 – Moderately aware
- 5 – Extremely aware

15. How familiar are you with natural radioactive material and its effects on human health?

- 1 – Not at all familiar
- 2 – Slightly familiar
- 3 – Moderately familiar
- 4 – Very familiar
- 5 – Extremely familiar

16. Did you know that produced water might contain levels of natural radioactive material? (For more information: <http://www.epa.gov/radiation/tenorm-oil-and-gas-production-wastes>)

- Yes or No

- **Perceptions of health and safety risks**

17. Please indicate your level of concern for each question:

Table C1. Level of concern on different scenarios.

Question (scenario)	Not at all concerned	Slightly concerned	Somewhat concerned	Moderately concerned	Extremely concerned
How concerned are you that storing produced water in tanks might have harmful effects on public health and safety in your area?					
How concerned are you that failure of equipment used to handle produced water (e.g. pipelines) might have harmful effects on public health and safety in your area?					
How concerned are you that transporting produced water by truck might have harmful effects on public health and safety in your area?					

- **Trust in State officials and oil operators**

18. Indicate the degree of trust in the organizations either directly or indirectly involved in produced water management.

Table C2. Level of trust in different organizations.

Organization	No trust at all	Little trust	Quite a bit of trust	A lot of trust
Oil operators				
Truck companies				
State/local government				
Federal government				
Environmental Protection Agency				

19. How confident are you that the State agencies (e.g. Department of Health and Department of Mineral Resources) will provide honest and accurate information about the safety of produced water handling and disposal?

- 1 – Not at all confident
- 2 – Not too confident
- 3 – Somewhat confident
- 4 – Very confident

20. Do you know who to contact to report a produced water spill in your area?

- Yes or No

21. As part of your daily job, do you have responsibilities directly related to oil production and produced water management or do you work in one of the areas listed below?

- Emergency management in ND
- Key response partner (i.e. public health, law enforcement, fire, emergency medical services)
 - Yes or No

If yes, please answer the following questions.

- **Further questions**

22. Based on your experience, how aware do you think the general public is about produced water risks in North Dakota?

- 1 – Not at all aware
- 2 – Slightly aware
- 3 – Somewhat aware
- 4 – Moderately aware
- 5 – Extremely aware

23. Based on your experience, how aware do you think the **operators in the oil field** are about produced water risks in North Dakota?

- 1 – Not at all aware
- 2 – Slightly aware
- 3 – Somewhat aware
- 4 – Moderately aware
- 5 – Extremely aware

24. Based on your experience, how aware do you think the **hauling truck operators** are about produced water risks in North Dakota?

- 1 – Not at all aware
- 2 – Slightly aware
- 3 – Somewhat aware
- 4 – Moderately aware
- 5 – Extremely aware

25. Based on your experience, how do you think each group ranks the risks of each scenario listed below? (low, medium, high)

Table C3. Experts’ opinion on rank of risk of different stakeholders.

Scenario	General Public	Operators in the oil field	Hauling truck operators
Produced water storage tank overflows and reaches a surface water body			
Equipment leakage (e.g. pipelines) reaches a surface water body			
Truck accident spills produced water and reaches a surface water body			

APPENDIX D. IRB CERTIFICATION OF EXEMPT HUMAN SUBJECTS RESEARCH

LETTER



March 29, 2016

Dr. Eakalak Khan
Civil and Environmental Engineering

Re: IRB Certification of Exempt Human Subjects Research:
Protocol #EN16229, "Holistic Risk Assessment of Surface Water Contamination Due to Oil Produced Water from the Bakken, North Dakota"

Co-investigator(s) and research team: Dr. Om Yadav, Luisa F. Torres

Certification Date: 3/29/2016 Expiration Date: 3/28/2019
Study site(s): varied
Sponsor: n/a

The above referenced human subjects research project has been certified as exempt (category # 2b) in accordance with federal regulations (Code of Federal Regulations, Title 45, Part 46, Protection of Human Subjects). This determination is based on the original protocol submission with updated training documentation (received 3/25/2016).

Please also note the following:

- If you wish to continue the research after the expiration, submit a request for recertification several weeks prior to the expiration.
- The study must be conducted as described in the approved protocol. Changes to this protocol must be approved prior to initiating, unless the changes are necessary to eliminate an immediate hazard to subjects.
- Notify the IRB promptly of any adverse events, complaints, or unanticipated problems involving risks to subjects or others related to this project.
- Report any significant new findings that may affect the risks and benefits to the participants and the IRB.

Research records may be subject to a random or directed audit at any time to verify compliance with IRB standard operating procedures.

Thank you for your cooperation with NDSU IRB procedures. Best wishes for a successful study.

Sincerely,

A handwritten signature in black ink that reads "Kristy Shirley".

Digitally signed by Kristy Shirley
DN: cn=Kristy Shirley, o=NDSU,
ou=Institutional Review Board,
email=kristy.shirley@ndsu.edu,
c=US
Date: 2016.03.29 10:47:16 -0500

Kristy Shirley, CIP, Research Compliance Administrator

For more information regarding IRB Office submissions and guidelines, please consult http://www.ndsu.edu/research/integrity_compliance/irb/. This Institution has an approved FederalWide Assurance with the Department of Health and Human Services: FWA00002439.

INSTITUTIONAL REVIEW BOARD

NDSU Dept 4000 | PO Box 6050 | Fargo ND 58108-6050 | 701.231.8995 | Fax 701.231.8098 | ndsu.edu/irb

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