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Environmental Health Sciences

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Seth B. Shonkoff,[1,2] Jake Hays,[3,4] and Madelon L. Finkel[4]

[1]Physicians Scientists and Engineers for Healthy Energy, Oakland, California, USA;

[2]Department of Environmental Science, Policy, and Management, University of California, Berkeley, Berkeley, California, USA; [3]Physicians Scientists and Engineers for Healthy Energy, New York, New York, USA; [4]Department of Public Health, Weill Cornell Medical College, New York, New York, USA

Address correspondence to Seth B. Shonkoff, 436 14th Street, Suite 808, Oakland, CA, 94612 USA. Telephone: 510.899.9706. E-mail: sshonkoff@psehealthyenergy.org

Short running title: Public Health and Shale Gas

Abstract

Background: The United States has experienced a boom in natural gas production due to recent technological innovations that have enabled this resource to be produced from shale formations.

Objectives: This review discusses the body of evidence that focuses on exposure pathways to evaluate the potential environmental public health impacts of shale gas development. It highlights what is currently known and identifies data gaps and research limitations by addressing matters of toxicity, exposure pathways, air quality, and water quality.

Discussion: There is evidence of potential environmental public health risks associated with shale gas development. A number of studies suggest that shale gas development contributes to levels of ambient air concentrations known to be associated with increased risk of morbidity and mortality. Similarly, an increasing body of studies suggest water contamination risks exist through a variety of environmental pathways, most notably during wastewater transport and disposal and via poor zonal isolation of gases and fluids due to structural integrity impairment of cement in gas wells.

Conclusion: Despite a growing body of evidence, a number of data gaps persist. Most importantly, there is a need for more epidemiological studies to assess associations between risk factors, such as air and water pollution and health outcomes among populations living in close proximity to shale gas operations.

Introduction

Technological innovations in drilling and well stimulation techniques have led to the production of natural gas from previously inaccessible geological formations, such as shale. Proponents of modern gas development argue that it has created a unique economic and political opportunity. Some in the public health community, however, have concerns about the potential for the extraction process to negatively impact the environment and human health (Finkel et al. 2013; Goldstein et al. 2012; Saberi 2013; Witter et al. 2013).

Producing natural gas from shale and tight gas formations in an economically feasible manner frequently requires a new constellation of existing technologies: high volume, slickwater, hydraulic fracturing from clustered, multi-well pads using long, directionally-drilled laterals. This method can involve drilling a well vertically thousands of feet below the surface and then directionally (horizontally) for up to two miles. An average of two to five million gallons of fluid consisting of water, proppant (often crystalline silica), and chemicals (some of which are known carcinogens or otherwise toxic) are injected into the well at a pressure high enough to fracture the shale rock (EPA 2010a). Often referred to as slickwater, chemicals are added to the fracturing fluid in order to decrease its friction. The fracturing fluid creates and expands cracks in the shale. When the pressure is released, the cracks are held open by the sand, allowing the tightly held gases to flow into the cracks and up the production casing. The gas is then collected, processed, and sent through transmission pipelines to market. In 2012, shale gas constituted nearly 40% of US gas production, up from 2% in 2000 (Hughes 2013).

Natural gas has a variety of attractive attributes. In the current market, it is a relatively inexpensive and abundant fuel. When combusted for electricity generation it emits fewer health

damaging contaminants and approximately 50% less carbon dioxide emissions when compared to burning coal (US EIA 2013). Yet, emerging scientific evidence suggests that there may be health risks associated with the development of shale gas.

In this review we discuss the body of scientific literature relevant to the environmental public health impacts of shale gas production. We highlight what is currently known and identify data gaps and research limitations.

Methods

Scope of review

This review primarily draws upon literature directly pertinent to the human health dimensions of shale and tight gas development. Tight gas refers to natural gas produced from reservoir rocks of low permeability, such as shale or sandstone. Shale gas and other forms of tight gas are referred to as "unconventional" due to their atypical reservoirs, which require new production techniques. However, the review references some studies that do not directly evaluate unconventional natural gas operations, but that are nonetheless relevant to various aspects of the overall process (e.g., particulate matter pollution, ozone, etc.). In the case of ozone, for instance, we analyzed top down studies that measure tropospheric concentrations rather than studies that supply bottom up measurements (e.g., leakage rates). Materials included in this review are predominantly sourced from the peer-reviewed scientific literature but include, where appropriate, government reports and other grey literature. Although the production chain of gas development is far-reaching, this review focuses on the processes that begin with trucking the water, sand, chemicals, and other materials to the well pad and ends with the disposal of wastewater. Evidence suggests that these processes present the greatest risks to environmental public health and therefore have received

the most attention in the scientific literature (Korfmacher et al. 2013; McKenzie et al. 2012; Rozell and Reaven 2012; Witter 2013).

Terminology

Terminology is important when discussing modern forms of natural gas development. In part due to a lack of well-defined, uniform terminology, there has been confusion regarding which processes constitutes this type of development. The terms, "hydraulic fracturing" or "fracking" are regularly used in the popular media as umbrella terms that are colloquially used to describe the entire process of shale gas and other forms of unconventional natural gas development, from land clearing and well spudding to transmission of natural gas to market. However, taken literally "hydraulic fracturing" only refers to the well stimulation processes and excludes other potentially more health and environmentally impactful processes, including but not limited to well drilling, fracturing fluid production, wastewater disposal, transportation of materials, and the processing, compression, and transmission of gas and liquids.

Many of the studies cited in this review may also apply to shale (tight) oil development and other forms of oil and gas development using well stimulation techniques that include matrix acid stimulation, acid fracturing, and steam injection. However, these other techniques are beyond the focus of this review. Additionally, the term "unconventional oil and gas development" can also refer to bitumen/tar sands extraction and processing, and other types of fossil fuel development that employ novel engineering and production techniques to obtain resources from unconventional resources (e.g., coal bed methane), that are beyond the scope of this review. The majority of the environmental public health-relevant scientific literature on modern oil and gas production has focused on the development of natural gas from shale formations and so this review uses the term, *shale gas development*. However, this review discusses, where appropriate,

scientific literature on other forms of unconventional or tight gas development that include the most prominent and relevant features of shale gas development, such as high volume, horizontal, hydraulic fracturing.

Identification of relevant studies

The literature directly relevant to the environmental public health dimensions of shale gas development is still limited. For this reason, we adopted a broad search strategy comprised of the following:

- Systematic searches in three peer-reviewed science databases across multiple disciplines: *PubMed* (http://www.ncbi.nlm.nih.gov/pubmed/), *Web of Science* (http://www.webofknowledge.com), and *ScienceDirect* (http://www.sciencedirect.com)

- Searches in existing collections of scientific literature on this subject, such as The Marcellus Shale Initiative Publication Database at Bucknell University (http://www.bucknell.edu/script/environmentalcenter/marcellus), complemented by Google (http://www.google.com) and Google Scholar (http://scholar.google.com)

- Manual searches (hand-searches) of references included in all peer-reviewed studies that pertained directly to shale gas development

For bibliographic databases, this review used a combination of Medical Subject Headings (MeSH)-based and keyword strategies, which included the following terms as well as relevant combinations thereof: shale gas, shale, hydraulic fracturing, fracking, drilling, natural gas production, Marcellus, Barnett, Denver-Julesberg Basin, air pollution, methane, water pollution, public health, water contamination, fugitive emissions, air quality, epidemiology, unconventional gas development, and environmental pathways.

At the time of writing, this search identified a total of 211 peer-reviewed publications that pertain directly to shale gas development. This database can now be accessed online and will continue to be updated with relevant literature (http://psehealthyenergy.org/site/view/1180). Of these 211 publications, only 33 presented original data that met our inclusion criteria and which were considered relevant as primary literature.

Inclusion/exclusion criteria

From the studies identified through February 1, 2014, we excluded non-relevant technical papers and studies related to economics, climate change, sociology, regulation, seismicity, water usage, social stress and quality of life considerations. While we excluded commentaries from the results of this review, we cite a few to provide documentation of particular considerations among the public health community. We included studies with direct pertinence to the environmental public health and environmental exposure pathways (i.e., air and water) associated with shale and tight gas development. In this regard, we supplemented the shale gas literature with studies that evaluated particular environmental pathways and health outcomes. For instance, we included studies directly related to the health impacts of tropospheric ozone, fine particulate air pollution, and endocrine disrupting chemicals. While this review excludes the vast majority of non-peer-reviewed scientific literature, it references environmental impact statements and other government reports where appropriate.

Results

The environmental public health framework and possible exposure pathways

The environmental exposure pathway framework is often used to describe associations between pollutant sources and health effects via emissions, environmental concentrations of pollutants,

pollutant exposure pathways (through mouth, nose, ears, eyes, skin, etc.), and dose (i.e., micrograms of pollutant ingested per day) (Figure 1) (ATSDR 2005).

Sources of health-relevant environmental pollution are located in a number of places and through multiple processes in the lifecycle of shale gas development. These sources include the shale gas production and processing activities (i.e., drilling, hydraulic fracturing, hydrocarbon processing and production, wastewater disposal phases of development); the transmission and distribution of the gas to market (i.e., in transmission lines and distribution pipes); and the transportation of water, sand, chemicals, and wastewater before, during, and after hydraulic fracturing.

We begin with a brief introduction of what is known regarding the toxicity and possible exposure pathways of the hydraulic fracturing fluids used in the well stimulation process. We then discuss the current scientific understanding of air quality concerns associated with shale gas development. Lastly, we discuss the current scientific understanding of water pollution risks and exposure pathways associated with the processes.

Hydraulic fracturing fluids: chemical toxicology and exposure pathways

Shale gas development uses organic and inorganic chemicals known to be health damaging in fracturing fluids (Aminto and Olson 2012; US HOR 2011). These fluids can move through the environment and come into contact with humans in a number of ways, including surface leaks, spills, releases from holding tanks, poor well construction, leaks and accidents during transportation of fluids, flowback and produced water to and from the well pad, and in the form of run-off during blowouts, storms, and flooding events (Rozell and Reaven 2012). Further, the mixing of these compounds under conditions of high pressure, and often, high heat, may synergistically create additional, potentially toxic compounds (Kortenkamp et al. 2007;

Teuschler and Hertzberg 1995; Wilkinson 2000). Compounds found in these mixtures may pose risks to the environment and to public health through numerous environmental pathways, including water, air, and soil (Leenheer et al. 1982).

Chemicals are used in the drilling and fracturing processes as corrosion inhibitors, biocides, surfactants, friction reducers, gels, and scale inhibitors, among other uses (Aminto and Olson 2012; NYS DEC 2011; Southwest Energy 2012). Examples include methanol, ethylene glycol, naphthalene, xylene, toluene, ethylbenzene, formaldehyde, and sulfuric acid, some of which are known to be toxic, carcinogenic, and associated with reproductive harm (Colborn et al. 2011; NYS DEC 2011). Many of these compounds are also regulated in other industries under the Safe Drinking Water Act (SDWA) and the Clean Water Act (CWA) as hazardous water pollutants (Safe Drinking Water Act of 1974; Clean Water Act of 1972; US HOR 2011).

Many of the chemical compounds used in the process lack scientifically based maximum contaminant levels (MCLs), which render a quantification of their public health risks more difficult (Colborn et al. 2011). Moreover, uncertainty about the chemical make-up of fracturing fluids persists due to the limitations on required chemical disclosure, driven by the Energy Policy Act of 2005 (Energy Policy Act of 2005). For instance, in many states, companies are not mandated to disclose information about the quantities, concentrations, or identities of chemicals used in the process on the principle that trade secrets might be revealed (Centner and O'Connell 2014; Centner 2013; Maule et al. 2013).

Some companies make efforts to be more transparent in the disclosure of chemicals used in the process. FracFocus (www.fracfocus.org) was developed as an online, voluntary chemical disclosure registry and some agencies (e.g., Bureau of Land Management) have suggested that it

be used as a regulatory compliance tool (FracFocus 2014; Konschnik et al. 2013). However, the registry has been criticized due to uncertainty surrounding the timing, substance, and omissions of the disclosed data on the website (Konschnik et al. 2013).

Because of limited information that is available, researchers have sought to acquire more information on the chemical make-up of fracturing fluids through other means. For example, using Material Safety Data Sheets (MSDSs), Colborn et al. (2011) identified chemical information for 353 of 632 chemicals contained in 944 products used for natural gas operations in Colorado (Colborn et al. 2011). This study represents one of the first attempts to conduct a chemical hazard assessment by identifying some of the compounds in fracturing fluids.

It should be noted that the scope of Colborn et al. (2011) is limited in that it does not measure exposure, dose, or health outcomes across populations. The researchers identified Chemical Abstract Services (CAS) numbers for the chemicals and used these in systematic searches of databases such as TOXNET (http://toxnet.nlm.nih.gov). Based upon the results of these searches, the researchers classified the compounds into twelve different health effects categories. At certain concentrations or doses, more than 75% of the chemicals identified are known to negatively impact the skin, eyes, and other sensory organs, the respiratory system, the gastrointestinal system, and the liver; 52% have the potential to negatively affect the nervous system; and 37% of the chemicals are candidate endocrine disrupting chemicals (Colborn et al. 2011).

Endocrine disrupting chemicals (EDCs) present unique hazards, particularly during fetal and early childhood growth and development (Diamanti-Kandarakis et al. 2009). They can affect the reproductive system and epigenetic mechanisms leading to pathology decades after exposure

(Zoeller et al. 2012). EDCs have challenged traditional concepts in toxicology because effects at higher doses do not always predict effects at low doses (Vandenberg et al. 2012). In other words, the dose does not always make the poison.

Kassotis et al. (2013) measured surface and ground water samples in Colorado for estrogen and androgen receptor activities using reporter gene assays in human cell lines. Samples collected from the more intensive areas of natural gas development exhibited statistically significantly more estrogenic, anti-estrogenic, or anti-androgenic activities than references sites with either no operations or fewer operations (Kassotis et al. 2013). The concentrations of chemicals detected were in high enough concentrations to interfere with the response of human cells to male sex hormones and to estrogen. This study demonstrates that EDCs are a potential health concern in natural gas operations and suggests that chemicals used in the process should be screened for EDC activity.

Air quality

Air pollutant emission sources from shale gas development can be grouped into two main categories: 1) emissions from drilling, processing, well completions, servicing, and other gas production activities; and 2) emissions from transportation of water, sand, chemicals, and equipment to and from the well pad.

Air pollution: drilling, well stimulation, gas production, processing, and servicing

The literature suggests that shale gas development processes emit hazardous air pollutants including, but not limited to benzene, toluene, ethylbenzene, and xylene (BTEX compounds), formaldehyde, hydrogen sulfide, acrylonitrile, methylene chloride, sulfuric oxide, nitrogen oxides, volatile organic compounds (VOCs), trimethylbenzenes, aliphatic hydrocarbons, diesel

particulate matter, and radon gas (McKenzie et al. 2012; Pétron et al. 2012; Roy et al. 2013). These emissions can result in elevated air pollution concentrations that exceed US EPA guidelines for both carcinogenic and non-carcinogenic health risks (McKenzie et al. 2012; MSI 2011).

A hazard assessment by McKenzie et al. (2012) used EPA guidance to estimate chronic and sub-chronic non-cancer hazard indices and cancer risks from exposure to hydrocarbons for residents living > ½ mile from wells and for those living ≤ ½ mile from wells in Colorado (McKenzie et al. 2012). The study found that residents living ≤ ½ mile from wells are at a greater risk for health effects from exposure to natural gas development than those living > ½ mile from wells. Notably, the study found a sub-chronic non-cancer hazard index (HI) of 5 for those living ≤ ½ mile compared to an HI of 0.2 for those living > ½ mile from wells driven primarily from exposure to trimethylbenzenes, xylenes, and aliphatic hydrocarbons (McKenzie et al. 2012). Unfortunately, baseline air quality data prior to this study were not available. However, the statistically significant spatial associations between air quality and shale gas development are an indicator that air quality may be negatively impacted and health risks may increase during various stages of shale gas development.

A study by Bunch et al. (2013), however, found that shale gas production activities did not result in community-wide exposures to concentrations of volatile organic compounds (VOCs) at levels that would pose a health concern. Bunch et al. (2013) examined VOC concentration data from seven air monitors at six locations in the Barnett Shale region in Texas. These measurements were then compared to federal and state health-based air comparison values (HBACVs) in order to determine possible acute and chronic health effects; none of the concentrations exceeded acute HBACVs (Bunch et al. 2013). Air quality data included in this study were generated from

monitors focused on regional atmospheric concentrations of pollutants. Conversely the McKenzie et al. (2012) study included samples at the community level in close proximity to gas development. Finer geographically scaled samples often capture local atmospheric concentrations that are more relevant to human exposure. This may be a primary reason why health hazard estimates differed between the two studies.

Roy et al. (2013) estimated emissions of nitrogen oxides (NO_x), VOCs, and particulate matter (PM) to present an air emissions inventory for the development of natural gas in the Marcellus Shale region for 2009 and 2020. In 2020, shale gas development activities are predicted to contribute 6-20% [12%] of the NO_x emissions and between 6-31% [12%] of anthropogenic VOC emissions in Pennsylvania (Roy et al. 2013). However, these estimates are based on assumptions of improvements in gas production, completion, and processing infrastructure. If source-level emissions remain the same as in 2009, Marcellus VOC emissions are predicted to constitute approximately 34% (19%-62%) of the regional anthropogenic VOC emissions in 2020 (Roy et al. 2013). Increases in emissions of VOCs and NOx, which are precursors of tropospheric ozone formation could complicate ozone management in the region and may offset ozone precursor emission reductions in other sectors at a time when several regions in Pennsylvania struggle to be within ozone attainment (Roy et al. 2013).

In another study focused on hydrocarbon emissions, Colborn et al. (2012) assessed air quality in western Colorado using weekly air samples taken before, during, and after drilling and hydraulic fracturing on a new natural gas well pad (Colborn et al. 2012). The data showed numerous chemicals in the air associated with natural gas development operations, most notably methane, ethane, propane, and other alkanes. Many non-methane hydrocarbons (NMHCs), which were observed during the initial drilling phase, are associated with multiple health effects. Notably,

thirty of the NMHCs observed in the field were EDCs. In addition to the direct air pollution associated with natural gas drilling and processing (NMHCs, VOCs, etc.) outlined above, there are also indirect pollution concerns such as the secondary atmospheric formation of tropospheric (ground-level ozone) (Colborn et al. 2012).

Studies indicate that shale gas development is associated with the production of secondary pollutants such as tropospheric (ground-level) ozone, which is formed through the interaction of methane (CH_4), VOCs and nitrogen oxides in the presence of sunlight (Jerrett et al. 2009; US EPA 2013). Tropospheric ozone is a strong respiratory irritant associated with increased respiratory and cardiovascular morbidity and mortality (Jerrett et al. 2009; UNEP 2011). While toxicological data suggests that pure methane is not by itself health damaging minus its role as an asphyxiant and an explosive, methane is a precursor to global tropospheric ozone.

Pétron et al. (2012) analyzed data collected at the NOAA Boulder Atmospheric Observatory (http://www.esrl.noaa.gov/psd/technology/bao) and filtered by wind sector that indicated a high alkane and benzene signature from the direction of the Denver-Julesburg Basin, an area of considerable oil and gas development (Pétron et al. 2012). The study found that an estimated 4% (range: 2.3 to 7.7%) of all natural gas (comprised mostly of CH_4) that was produced was being accidentally leaked or purposefully vented to the atmosphere (Pétron et al. 2012). Karion et al. (2013) observed significant methane leaks in the Uintah Basin shale gas field. A range of 6.2% to 11.7% of total gas production was estimated to be leaking to the atmosphere (Karion et al. 2013).

A national methane emissions study by Miller et al. 2013, which combined ground and aerial sampling of methane with computer modeling, found that atmospheric levels of methane due to

oil and gas extraction could be 4.9 ± 2.6 times greater than current estimates from the Emissions Database for Global Atmospheric Research (EDGAR) (http://edgar.jrc.ec.europa.eu/index.php) and the United States Environmental Protection Agency (US EPA) (Miller et al. 2013). Although it is difficult to distinguish the sources of methane between shale and non-shale development and between oil and gas production, Peischl et al. (2013) estimated that 17% of gross methane production from oil and gas activities in the Los Angeles Basin are leaked or vented to the atmosphere (Peischl et al. 2013).

Some studies have modeled ozone impacts associated with shale gas operations. Kemball-Cook et al. (2010) modeled ozone precursor emissions (VOCs and NO_x) in the Haynesville shale play that lies beneath the Northeast Texas/Northwest Louisiana border. Photochemical modeling showed increases in 2012 8-hour ozone design values of up to 5 parts per billion (ppb) which, along with the amount of projected emissions, give cause for concern about future atmospheric concentrations of ozone in Texas and Louisiana (Kemball-Cook et al. 2010). Olaguer (2012) used The Houston Advanced Research Center (HARC) neighborhood air quality model to simulate ozone formation near a hypothetical natural gas processing facility, using estimates based on both regular and non-routine (e.g. flaring) emissions (Olaguer 2012). This model predicted that under average conditions using regular emissions associated with compressor engines may significantly increase ambient ozone in the Barnett Shale formation (> 3ppb 2 km downwind from facility) (Olaguer 2012).

Substantial air quality impacts from oil and natural gas operations in Wyoming, Colorado, Utah, and Texas have also been directly measured (Carter et al. 2012; Edwards et al. 2013; US DOE 2011). In February 2009, Schnell and colleagues studied air quality in the rural Upper Green River Basin (UGRB) of Wyoming near the Jonah-Pinedale Anticline natural gas field. The study

observed high photochemical ozone concentrations in the UGRB in the winter, reporting readings of up to 140 ppb, just less than double the EPA ozone concentration limit 75 ppb (http://www.epa.gov/air/criteria.html). Prior to 2005, typical wintertime ozone concentrations in this area were 30 ppb to 40 ppb (Pinto 2009). This suggests an association between the increase in NO_x and VOC emissions from oil and gas development activities in the area (Schnell et al. 2009). In 2011, a study conducted for the Wyoming Department of Environmental Quality, found that the eight-hour ozone concentrations in the UGRB exceeded the US EPA ozone eight-hour standard thirteen days (MSI 2011) and exceeded the US EPA science-recommended limit of 65 ppb twenty-five days (Weinhold 2008).

Additionally, in Utah in the winter of 2010 there were 68 days when ozone levels exceeded the EPA ozone standard of 75 ppb and in 2011 there were readings more than double the EPA standard (UT DEQ 2013). Results of a study conducted by the US EPA and the National Oceanic and Atmospheric Administration (NOAA) (http://www.noaa.gov) indicate that ozone precursor emissions (VOCs and NO_x, primarily) from oil and gas development in the Uintah Basin in Utah are a primary factor (UT DEQ 2013).

As mentioned, crystalline silica sand is used as a proppant (to "prop" open cracks in the target formation to allow gas to flow up the well) and is delivered by trucks to the drilling site. The transportation of this sand in trucks and trains and mixing it into fracturing fluids with sand movers, conveyer belts, and blender hoppers at the well site is known to release silica dust into the air where well pad workers can be exposed (Esswein et al. 2013). Workers experience the most direct exposure, however, silica dust may also be an air contaminant of concern to nearby residents. The etiological association between respiratory exposure to silica dust and the development of silicosis is well known (CDC 1992; CDC 2002). Silicosis is a progressive lung

disease where tissue in the lungs reacts to silica particles causing inflammation and scarring, decreasing the ability of the lungs to take in oxygen (CDC 1992; CDC 2002). Respiratory exposure to silica is also associated with other diseases such as chronic obstructive pulmonary disease (COPD), tuberculosis, kidney disease, autoimmune conditions, and lung cancer (CDC 2002).

Esswein et al. (2013) collected full shift air samples at eleven sites in five states in cooperation with industry partners to determine levels of worker exposure. Of the 111 air samples, 51.4% showed silica exposures greater than the calculated Occupational Safety & Health Administration (OSHA) permissible exposure level (PEL) and 68.5% showed exposures greater than the NIOSH recommended exposure limit (REL) of 0.05 milligrams per cubic meter (Esswein et al. 2013). Further, Esswein and colleagues noted that the type of respirators worn by workers were not sufficiently protective in some cases given the magnitude of silica concentrations (Esswein et al. 2013).

Air pollution: transportation

Each well requires on average between two to five million gallons of water per hydraulic fracturing event (EPA 2010a). Water is generally not pumped directly to wells and is instead transported by diesel trucks, each of which has an approximate capacity of 3,000 gallons (EPA 2011b). It is estimated that approximately 2,300 heavy-duty truck trips are required for each horizontal well during early stages of shale gas development (NYS DEC 2011). With thousands of such wells concentrated in high development regions, increased levels of truck traffic and elevated levels of diesel-associated air pollution will be brought to these areas.

The pollutant of primary health concern emitted from the transportation component of shale gas development is fine diesel particulate matter (dPM). Diesel particulate matter is a well-understood health damaging pollutant that contributes to cardiovascular illnesses, respiratory diseases (e.g., lung cancer) (Garshick et al. 2008), atherosclerosis, and premature death (Pope et al. 2004; Pope 2002). For instance, the California Air Resources Board (2008) indicates that there is an expected 10% (CI: 3% to 20%) increase in the number of premature deaths per 10 $\mu g/m^3$ increase in $PM_{2.5}$ exposure (Tran et al. 2008). Particulates can also concentrate associated products of incomplete combustion and act as a delivery system to the alveoli of the human lung when their diameter is less than 2.5 microns (Smith et al. 2009). In addition to diesel PM, as previously mentioned, nitrogen oxides (NO_x) and volatile organic compounds (VOCs) – other prevalent pollutants in diesel emissions – react in the presence of sunlight and high temperatures to produce tropospheric (ground-level) ozone.

Water quality

Rozell and Reaven (2012) conducted a risk assessment that identified five main pathways of water contamination in the shale gas production process: 1) the transportation spills of fracturing fluid or produced water; 2) well casing leaks; 3) leaks through fractured rock; 4) drilling site discharge; and 5) wastewater disposal (Rozell and Reaven 2012). The assessment found that wastewater disposal carries a potential risk of water contamination several orders of magnitude larger than the other pathways (Rozell and Reaven 2012).

Other studies suggest that structural impairment of cement that is used to prevent trans-zonal gas migration in the wellbore are the most common mechanism through which groundwater can become contaminated (Vidic et al. 2013). Indeed, state environmental regulators at the Pennsylvania Department of Environmental Protection (DEP) found that oil and gas

development polluted water supplies for at least 161 residences in PA between 2008 and 2012, primarily due to cement structural integrity in wells and wellbores (Legere 2013). While we touch upon the five aforementioned pathways, for the purpose of this review we focus most of our discussion on well casing leaks, drilling site discharge, and wastewater disposal, as these are generally regarded as the most viable means of water contamination (Rozell and Reaven 2012; Vidic 2013).

Flowback and produced water

Estimates of the proportion of fracturing fluid that returns to the surface as flowback and produced waters range from 9% to 80% with most estimates around 35% (US EPA 2010a; Horn 2009; NYS DEC 2011). These wastewaters contain the chemicals used in the fracturing fluid as well as compounds found deep in geological strata, such as salts, chlorides, heavy metals (e.g., cadmium, lead, arsenic, etc.), organic chemicals (e.g., benzene, toluene, ethylbenzene, xylene), bromide, and, depending upon the geology, naturally occurring radioactive materials (e.g., radium-226). Many of these naturally occurring compounds are known to be associated with human health effects when exposure is sufficiently elevated (Balaba and Smart 2013; Colborn et al. 2011; Haluszczak 2013). A proportion of flowback and produced waters are treated and released as effluent or for other beneficial uses such as irrigation for agriculture. However, many of the chemicals persist in high quantities because treatment facilities are unable to screen for and eliminate the complex array of compounds and products of synergistic interactions between them (Ferrar et al. 2013; Hladik et al. 2014; Lutz et al. 2013).

Flowback and produced water is sometimes treated at facilities and then discharged into surface waters (Ferrar et al. 2013). A study by Warner et al. (2013a) examined water quality and isotopic compositions of discharged effluents, surface waters, and stream sediments associated with a

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Marcellus wastewater treatment facility site. The findings suggest that insufficiently treated flowback and produced water with elevated concentrations of contaminants associated with shale gas development is entering local water supplies, even after treatment. The study found elevated levels of chloride and bromide downstream along with radium-226 levels in stream sediments at the point of discharge that were ~ 200 times greater than upstream and background sediments and well above regulatory standards (Warner et al. 2013a). These types of water emissions may increase the health risks of those that rely on these surface and hydrologically contiguous groundwater sources for drinking (Wilson and VanBriesen 2012) and sources of food (i.e., fish protein) (Papoulias and Velasco 2013).

A meta-analysis of chemical and physical characterizations of produced waters from shale gas found that most of the produced waters generated by shale gas development were classified as saline (>30,000 mg/l) or hyper-saline (>40,000 mg/l) (Alley et al. 2011). The treatment of this produced water for beneficial use often involves reverse osmosis, a practice that may generate a waste stream too large to justify the activity (Alley et al. 2011). Alley and colleagues (2011) also found that prior to treatment, produced waters can exceed toxicity thresholds of contaminants of concern (COCs) including, but not limited to phosphates, cadmium, aluminum, barium, chloride, strontium, radium-226, bromine, lithium and magnesium. Toxicity thresholds used in this meta-analysis were LC_{50} values of *Ceriodaphnia dubia* Richard, *Daphnia magna* Straus, and *Pimephales promelas* Rafinesque and water use criteria under the Food and Agricultural Organization of the United Nations (FAO) Guidelines for agricultural uses and the United States Environmental Protection Agency (US EPA) Water Quality Criteria (WQC) for surface discharge (Alley et al. 2011).

The results of Alley et al. (2011) agree with other reporting that samples of fracturing fluids, drilling muds, and flowback and produced waters in wastewater surface containment ponds contain chemicals that at elevated doses or certain concentrations have been associated with health effects ranging from skin and eye irritation to neurological and nervous system damage, cancer, and endocrine disruption (Colborn et al. 2011). Moreover, between July 2009 and June 2010, 192.5 million gallons of produced water (PW) was reported in Pennsylvania alone, with uncertainties as to the location and type of disposal to be employed (PA DEP 2010).

It should also be noted that the handling and disposal of flowback and produced water hold implications for air quality due to volatile compounds, such as benzene, toluene, ethylbenzene, and xylene (BTEX) that are often mixed with the fluids. This may be particularly relevant when wastewater is stored in surface containment ponds and misted into the air to promote evaporation (Colborn et al. 2011).

Gas and fluid migration

Sub-surface gas and fluid migration is most commonly associated with impaired structural integrity of well cement and, to a lesser extent, well casings. Failures in well barriers may allow intrusion of gases and fluids from producing formations below the casing shoe or from shallower gas and fluid-bearing formations intersected by the wellbore to lower-pressure annuli. This may result in annular gas flow or sustained casing pressure (SCP) and become a pathway for gas migration to the surface, which is a known mechanism of emissions of gases to the air and migration of gases and fluids to groundwater (Bruffato et al., 2003; Watson and Bachu 2009). Methane and other hydrocarbons can also migrate along improperly plugged wells, through an inadequately sealed annulus, or between geological zones due to cement failures in the wellbore (Vidic et al. 2013).

Leaking oil and gas wells are recognized as a potential mechanism of subsurface migration of methane, as well as heavier n-alkanes and other non-methane volatile organic compounds (NMVOCs) to groundwater and the atmosphere, contributing risks to drinking water and air quality, respectively (Bourgoyne et al. 2000; Brufatto et al. 2003; Chilingar and Endres 2005; Watson and Bachu 2009). Cement failures in onshore and offshore wells are reported to occur in between 2% and 50% of all wells, providing pathways for gas migration to occur in the wellbore (Bourgoyne et al. 2000; Brufatto et al. 2003; Watson and Bachu 2009).

Methane has a low solubility (26 mg/L at 1 atm, 20°C) (Vidic et al. 2013) and is relatively unreactive compared to longer-chain and unsaturated hydrocarbons (Jackson et al. 2011). As such, it is typically regarded as non-toxic and is not regulated in the United States as a solute in water wells. However, there are no peer-reviewed studies on the health effects of chronic exposure to lower concentrations of methane in drinking water or indoor or outdoor air (Jackson et al. 2011). Further, if there is a pathway for methane migration, there could be a pathway for associated health-damaging gases co-produced with methane.

Some attention has been paid to the flammability of methane, the risk of explosions, and the risk of asphyxiation (in high indoor concentrations, primarily). For example, in 2007, methane contaminated a water well and a home exploded in Geauga County near Cleveland, Ohio and the Ohio Department of Natural Resources blamed a faulty concrete casing in a nearby gas well (OH DNR 2008). Similarly in Pavillion, WY high concentrations of methane were found in drinking water wells that was attributed to gas production activities (DiGiulio et al. 2011). The EPA also concluded that methane from geological layers not targeted for gas production migrated up the wellbore and to an aquifer due to well cement failures in Parker County, Texas (EPA 2010a).

In certain regions, methane can naturally occur in aquifers and there are conflicting scientific opinions about whether its presence is caused or exacerbated by shale gas development (Davies 2011; Saba and Orzechowski 2011; Schon 2011). However, there are convincing findings that shed light on the likelihood that shale gas development is associated with high methane levels in drinking water wells. Osborn et al. (2011) found that communities in Pennsylvania with active shale gas development (one or more gas wells within 1 km) were found to have statistically significantly higher concentrations of methane in their water wells than in non-extraction sites (no shale gas wells within 1 km) (Osborn et al. 2011). The chemical signature of the methane found in the active area drinking water wells indicated that it came from a high-pressure, deep earth source (thermogenic methane). Alternatively, the methane from non-active sites had signatures of shallow earth origins (biogenic methane). This suggests that shale gas production processes were the source of the methane contamination.

Building upon previous work by Osborn et al. (2011), Jackson et al. (2013) analyzed 141 drinking water wells across northeastern Pennsylvania. The researchers found methane in 82% of the samples (115 of 141 of the wells), with average concentrations six times higher for homes that were less than one kilometer from natural gas wells (59 out of 141). These data, based on isotopic signatures and gas ratios, suggest that a subset of homeowners living less than one kilometer from shale gas wells had drinking water that was contaminated with stray gases associated with gas development activities (Jackson et al. 2013).

There is also evidence of existing pathways in some locations between deep underlying formations and shallow drinking water aquifers (Vengosh et al. 2013). Myers (2012) demonstrated this in a modeling study that suggested pathways that would allow for the transport of contaminants from the fractured shale to aquifers (Myers 2012). Warner et al. (2012) found

evidence of possible migration of Marcellus brine through naturally occurring pathways based on strong geochemical fingerprints in salinized groundwater samples (Warner et al. 2012).

Both of these studies suggest that migration through fractured rock can serve as a sub-surface contamination pathway to underground sources of drinking water. They also highlight the significance of the specific geographic regime, as some shallow drinking water resources are at more risk for contamination than others. Another study in areas of the Fayetteville Shale suggests that methane contamination of shallow groundwater may not be a problem in certain shale formations (Warner et al. 2013b). This difference may be attributed to geological variations across geographic space, including the presence of intermediate gas bearing formations that are found overlying parts of some shale plays (e.g., Marcellus), but not others (e.g., Fayetteville).

Additionally, a study that evaluated water quality in private drinking water wells near natural gas operations in the Barnett Shale formation in Texas found higher levels of arsenic, selenium, strontium and total dissolved solids (TDS) in wells located within 3 km of active gas wells (Fontenot et al. 2013). The study used historical data from the region as a baseline to determine the contamination rates before the expansion of natural gas operations. While heavy metals were known to occur at low levels in aquifers in the region, concentrations were significantly higher in areas of active development (Fontenot et al. 2013). The authors were able to link contamination to natural gas activities, however, the specific factor responsible for contamination (e.g., well casing failures, mobilization of natural constituents, hydrogeochemical changes from lowering the water table, etc.) remains undetermined (Fontenot et al. 2013).

Researchers have been challenged in their ability to link associations between water contamination and unconventional natural gas development to any particular part of the process.

After complaints about the taste and odor of well water from local residents, the EPA initiated a ground water investigation in the town of Pavillion, WY (DiGiulio et al. 2011). The observed water wells were located in an area known as the Pavillion gas field, which contained 169 gas production wells and 33 containment ponds used for storage/disposal of drilling wastes and produced and flowback waters from unconventional natural gas development of a sandstone formation.

From 2009 to 2011 the EPA conducted four sampling events meant to determine the presence (not extent) of ground water contamination in the formation. Elevated concentrations of benzene, toluene, ethylbenzene, and xylenes (BTEX) were detected in sampling wells at concentrations of 246, 617, 67, and 750 micrograms per liter, respectively (DiGiulio et al. 2011). Trimethylbenzenes and diesel range organics were detected at concentrations up to 105 and 4,050 micrograms per liter, respectively and total purgeable hydrocarbons were detected in the ground water samples near the containment ponds (DiGiulio et al. 2011). While these initial data indicate ground water impacts that seem likely to be associated with unconventional gas production practices (EPA 2011a), the results of this study have been contested and it is still unclear which part of the gas development process (if any) is responsible for the contamination. Further, there are geological differences between sandstone and shale, and fracturing is often conducted closer to the surface in sandstone formations. However, the findings suggest an association between water contamination and production activities that are also used in shale gas development.

Site discharge and improper waste disposal

Fracturing fluids and produced waters can also contaminate underground sources of drinking water during waste management and disposal. Flowback and produced waters are often

contained in evaporation ponds, pits, and tanks, in some cases in very close proximity to residences (Bamberger and Oswald 2012; Rozell and Reaven 2012). These containment ponds are often, but not always lined to protect against leakage, although case studies have documented reported ruptures to these liners that may have led to water and soil contamination and contributed to fish and livestock deaths (Bamberger and Oswald 2012). An analysis of waste obtained from reserve pits has also shown the potential for exposure to technologically enhanced naturally occurring radioactive material (TENORM) and potential health effects from individual radionuclides (Rich and Crosby 2013).

Groundwater contamination can also result from surface spills at active well sites. Gross et al. (2013) analyzed data from the Colorado Oil and Gas Conservation Commission (COGCC) (http://cogcc.state.co.us) and noted 77 reported surface spills (associated with less than 0.5% of active wells) impacting groundwater in Weld County, Colorado. The groundwater samples were analyzed for BTEX components (benzene, toluene, ethylbenzene, and xylene). Most notably, benzene measurements exceeded US EPA National Drinking Water maximum contaminant levels (MCL of benzene is 5 ppb) in 90% of the samples (Gross et al. 2013). Baseline-sampling measurements were not available and therefore the background BTEX concentrations remain unclear. However, natural groundwater concentrations are typically low near deposits of crude oil, coal, and natural gas (Gross et al. 2013).

Discussion

Future research needs

There is a growing body of scientific literature on the environmental public health dimensions of shale gas development, however our review indicates that a number of important data gaps persist. Emissions and atmospheric concentration measurements should be conducted among

diverse geographies and indoors as well as outdoors to help to estimate the types and magnitude of exposures of populations to pollutants associated with shale gas development. Additionally, studies that take into account personal exposures and time-activity patterns of individuals would be helpful to assess epidemiologically meaningful exposures. This could include studies of individuals in populations that use personal monitors and conduct household sampling of drinking water in conjunction with health records that look at disease outcomes.

Perhaps the most important information gap is the lack of epidemiologic studies. There is a need to assess the strength of the association between risk factors, such as air pollution, water contamination and health outcomes among populations living in close proximity to shale gas development activities compared to populations living in areas without active shale gas development activities. While lacking in definitive proof of cause and effect, self-reporting health surveys and environmental testing have suggested possible adverse health outcomes from shale gas development in Pennsylvania (Steinzor et al. 2013). Of particular interest are the epidemiological studies on vulnerable populations including pregnant women, young children, the elderly, and those with compromised immune systems that live, work, and play in close proximity to shale gas development. Further occupational health studies are also needed, as workers are likely to be the first and the most exposed demographic from shale gas development.

There have been some efforts in epidemiology and risk assessment, including a recent retrospective cohort study by McKenzie et al. (2014) that examined associations between maternal residential proximity to natural gas development. This study estimated associations between maternal exposure to natural gas development and a number of birth outcomes. The evidence found no positive association between density and proximity of wells within a 10-mile radius of maternal residence and prevalence of oral clefts, preterm birth, or term low birth

weight. However, the researchers did observe a positive association between density and proximity of pregnant mothers to shale gas development and the prevalence of congenital heart defects and possibly neural tube defects in their newborns (McKenzie et al. 2014).

There have been some other epidemiologic efforts as well, including a study funded by The American Natural Gas Alliance (ANGA) (http://anga.us) that evaluated associations between childhood cancer incidence in Pennsylvania and hydraulic fracturing sites (Fryzek et al. 2013). The authors included 29,000 hydraulically fractured wells drilled between 1990 and 2009 in their analysis and obtained childhood cancers from the Pennsylvania cancer registry for this time period. However, shale gas development did not begin in Pennsylvania until 2006 when four wells of this type were drilled. In fact, only 726, or 2.5% of the 29,000 wells in their database are the relevant to directionally drilled shale gas wells. Unfortunately this exposure misclassification and the disregard for the extended latency periods of many childhood cancers render this study inconclusive as to the effect of shale gas development on childhood cancer rates. This study demonstrates the need for more epidemiological assessments that pay attention to the latency periods of environmentally mediated diseases.

Epidemiological investigations are challenged by the difficult task of identifying specific risk factors and the uncertainty in exposure classification due to the fact that compounds used in shale gas development are often not disclosed. In these cases of uncertainty a comprehensive water monitoring and, under certain circumstances, a biomonitoring program that uses both targeted and non-targeted strategies would be useful. Targeted testing for specific compounds known to be associated with shale gas development in the drinking water supplies as well as the blood and urine of a representative sample of those living in close proximity to shale gas development could generate useful data. Non-targeted techniques including time-of-flight mass

spectrophotometers (TOF-MS) may also be helpful. Rather than monitoring for individual chemicals, TOF-MS has been important for the progress of biomonitoring in recent years by allowing researchers to monitor for tens of thousands of organic compounds at a time. This enables researchers to circumvent policy issues that do not require companies to disclose the compounds they employ in their activities, such as is the case in many regions throughout the United States.

Even with full disclosure of the chemicals added to fracturing fluid, the ability to link chemicals to specific health outcomes remains difficult. Fracturing fluids and flowback and produced wastewaters are complex mixtures of chemicals with individual and possibly cumulative and synergistic properties. Many health outcomes are not specific to chemicals associated with shale gas development (e.g., headaches can be caused by a number of factors, rashes can be non-specific, and asthma can be induced through a number of pathways), complicating the task of assessing associations between exposures and health outcomes. In turn, more exposure assessment and water and air monitoring should be undertaken to investigate the full suite of compounds emitted to the environment from these activities.

The chemicals contained in fracturing fluids are often not publically disclosed due to trade secret laws and exemptions under the 2005 Energy Policy Act that further confound environmental public health research (Energy Policy Act of 2005). Moreover, the US EPA does not regulate the injection of fracturing fluids under the Underground Injection Control (UIC) program of the Safe Drinking Water Act (SDWA) (http://water.epa.gov/type/groundwater/uic/regulations.cfm). The non-disclosure of these chemicals creates research barriers due to the fact that it is difficult to monitor for unknown compounds.

Limitations

This review represents a near exhaustive review of the peer-reviewed scientific literature on the environmental public health dimensions of shale gas development. The literature cited here is limited by the publication date of this paper. Data available on this subject in the future may change the scientific understanding of the environmental public health concerns of shale gas development. Thus, this review of the literature should only be viewed as a substantive summary of what is known to date.

Conclusion

This paper has reviewed the growing body of evidence of potential environmental public health dimensions of shale gas development. Scientific modeling and field investigations have helped to illuminate the emerging environmental issues with which shale gas production may be associated. A number of studies suggest that shale gas development contributes to levels of ambient air concentrations known to be associated with increased risk of morbidity and mortality (Colborn et al. 2012; Kemball-Cook et al. 2010; McKenzie et al. 2012; McKenzie et al. 2014). Similarly, some evidence supports theories of water contamination risks through a variety of pathways, most notably during wastewater transport and disposal and through failed cement in wells with poor structural integrity (Vengosh 2013; Warner et al. 2013a; Vidic 2013). The existing peer-reviewed scientific data suggest that there are potential risks, which could possibly influence public health. It is clear that more research is needed to more fully understand the magnitude of these concerns. As shale gas development activities have accelerated dramatically over the past decade, the need for well-designed empirical studies becomes increasingly apparent.

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Figure legend

Figure 1. Environmental exposure pathway. The environmental exposure pathway provides an analytical framework to describe, in broad terms, the connections between pollutant sources and human health outcomes. This framework begins with the emission source, in this case a well pad and associated infrastructure, which emit a variety of contaminants into the air, water, and soil. The concentrations of pollutants in the air, water, and soil that result from these emissions influence the magnitude of human exposures through organs such as the nose, mouth, and skin. Once the level of exposure is identified, it is then possible to estimate the dose, or how much of the pollutant is ingested in a given period of time. The dose, in turn, determines the health outcome.

Figure 1.

SOURCE ▶▶ EMISSIONS ▶▶ CONCENTRATION ▶▶ EXPOSURE ▶▶ DOSE ▶ HEALTH EFFECTS