



Arsenic release to the environment from hydrocarbon production, storage, transportation, use and waste management[☆]

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ABSTRACT

Arsenic (As) is a toxic trace element with many sources, including hydrocarbons such as oil, natural gas, oil sands, and oil- and gas-bearing shales. Arsenic from these hydrocarbon sources can be released to the environment through human activities of hydrocarbon production, storage, transportation and use. In addition, accidental release of hydrocarbons to aquifers with naturally occurring (geogenic) As can induce mobilization of As to groundwater through biogeochemical reactions triggered by hydrocarbon biodegradation. In this paper, we review the occurrence of As in different hydrocarbons and the release of As from these sources into the environment. We also examine the occurrence of As in wastes from hydrocarbon production, including produced water and sludge. Last, we discuss the potential for As release related to waste management, including accidental or intentional releases, and recycling and reuse of these wastes.

1. Arsenic in hydrocarbons: origins, analytical methods and concentrations

Arsenic (As) is a known toxin and carcinogen (NRC, 1999, 2001), causing a wide range of adverse human health impacts (Kapaj et al., 2006). Research on As in the environment has been extensive over the past two decades. From that work, much has been learned about As mobility in the environment (e.g., Smedley and Kinniburgh, 2002; Ravenscroft et al., 2009). Two challenging aspects related to As include identifying the As source(s) and the release mechanism(s). Arsenic is present in both geogenic (e.g., rocks, soils, sediments) and human (e.g., industrial, munitions, agricultural) sources. From these sources, As can be released via both naturally occurring processes and human activities, such as mining, agriculture, waste disposal, and accidental releases. Although disentangling the source and release mechanism is difficult, it is critical for understanding how As is released to the environment, so that further release can be managed or prevented.

Although significant work has been conducted on geogenic sources of As, one source that has received less attention is hydrocarbons. Because most hydrocarbons formed over geologic time from burial of marine organisms, As that was contained within organisms becomes

incorporated within the hydrocarbon. Thus, to examine As in hydrocarbons, we first need to outline how As occurs in marine environments and how it is taken up by organisms.

1.1. Origins of As in hydrocarbons

Arsenate (As(V)) and arsenite (As(III)) are the dominant oxidation states of inorganic As in marine ecosystems (Neff, 1997). In addition to these inorganic forms, there are also organic forms of As, including methylarsonic acid (MMA) and dimethylarsinic acid (DMA), found in seawater (Andreae, 1986; Cullen and Reimer, 1989; Neff, 1997). The average concentration of As in modern seawater ranges from 1 to 4 µg/L (Andreae, 1979; Andreae and Andreae, 1989; Neff, 1997; Reimer et al., 2010), but that can vary seasonally (Byrd, 1988; Neff, 1997). The lowest concentrations of As in seawater are measured in the spring when As is taken up by phytoplankton and suspended matter; concentrations increase in the fall from release of As from sediments into the water column (Neff, 1997), similar to the seasonal patterns observed for phosphate (Maher, 1984; Riedel, 1993; Sanders and Riedel, 1993). Relative concentrations of inorganic and organic forms of As are also influenced by concentrations of oxygen and phytoplankton and the

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species composition of phytoplankton (Waslenchuk, 1978; Sanders, 1985; Phillips, 1990).

Marine biota, including zooplankton, phytoplankton, and macroalgae, can take up As from seawater and process it through several metabolic pathways. Inorganic As(V) initially taken up by organisms is reduced to As(III), and then oxidized and methylated to produce primarily MMA, DMA, and other organoarsenic compounds, which are then excreted back into the ocean (Andreae and Klumpp, 1979; Sanders and Windom, 1980; Andreae, 1983; Phillips, 1990; Neff, 1997). Methylation of inorganic As has long been considered as a detoxification process (i.e., the Challenger process (Challenger, 1945)), but more recent work has shown that the products of methylation, including MMA and DMA, have toxic properties (Reimer et al., 2010). The substantial research on As uptake and transformation within marine biota reveals that the pathways are complicated and are confounded by analytical limitations and the occurrence of multiple processes; for example, in animals, it can be difficult to disentangle the processes within the animal vs. those mediated by bacteria within the organism (Francesconi and Edmonds, 1996).

In addition to excreting As, marine organisms can also accumulate As. Arsenic concentrations are higher in marine biota, including zooplankton, phytoplankton, and macroalgae, than they are in terrestrial organisms; some marine organisms are known to accumulate As to concentrations several thousand-fold above the As concentration in seawater (Lunde, 1977; Puri and Irgolic, 1989; Phillips, 1990). For example, some species of macroalgae, including the brown algae, can accumulate As concentrations in excess of 100 mg/kg (Lunde, 1977; Phillips, 1990; Rahman et al., 2012). The reasons for the high accumulation of As in macroalgae “remains one of the unsolved mysteries of arsenic chemistry” (Reimer et al., 2010).

Analysis of As species in marine organisms, including algae and animals, shows the presence of a host of water-soluble and lipid-soluble fractions, including arsenobetaine, arsenosugars and arsenolipids (Lunde, 1977; Andreae, 1983; Cullen and Reimer, 1989). Work conducted in the 1970s and 1980s on organoarsenic chemistry focused on pathways for metabolism of As compounds in marine biota (Andreae, 1983) and bioaccumulation of organoarsenic compounds such as arsenobetaine in higher order marine organisms, including fish, crustaceans, bivalves, snails, cephalopods and marine mammals (Edmonds and Francesconi, 1988; Hanaoka et al., 1992; Neff, 1997). Because seafood is an important food source for many populations, knowledge of the metabolism and toxicity of organoarsenic compounds in humans has been the subject of recent work (Taylor et al., 2017).

As discussed by Phillips (1990), there are wide differences in As concentrations and forms in marine biota. The reasons for these variations are not fully understood; however, differences in metabolism, phosphate concentrations, and uptake rates, in addition to sampling of different parts of biota, likely influence the concentrations and forms (Andreae and Klumpp, 1979; Sanders and Windom, 1980; Phillips, 1990; Sanders and Riedel, 1993; Cutter et al., 2001). Readers who are interested in As cycling in seawater and uptake into marine organisms topics can refer to key reviews of these topics (Cullen and Reimer, 1989; Francesconi and Edmonds, 1996; Reimer et al., 2010; Rahman et al., 2012).

In this chapter, we focus on As within a select group of hydrocarbons and source rocks, including crude oil, natural gas, oil shale, gas shale, and oil sands. We provide short descriptions of these sources in Table 1, but for a more in-depth discussion of hydrocarbon formation, we refer the reader to textbooks (e.g., Tissot and Welte, 1984; Jahn et al., 1998). Arsenic can be contained in hydrocarbons because, as discussed above, it can accumulate in the organisms that are the source of the hydrocarbons or it can be contained in the geologic medium in which the hydrocarbons accumulate (Fig. 1). There is an extensive literature on trace elements in hydrocarbons because many trace elements, most notably nickel (Ni) and vanadium (V), form metallo-porphyrin complexes in oil and have been used to provide information on the origin, migration and maturation of crude oils (Jacobs and Filby, 1983; Hitchon and Filby,

Table 1

Descriptions of hydrocarbon sources discussed in this chapter. Information modified from Tissot and Welte (1984) and Jahn et al. (1998).

Hydrocarbon type	Description and formation	Organic composition of hydrocarbon
Crude oil	Complex mixture of mostly organic compounds derived from marine organisms that die and undergo diagenesis, under which sedimentary organic material is converted into hydrocarbons.	Composed of alkanes (paraffins), cycloalkanes (naphthenes or cycloparaffins), aromatic hydrocarbons and nonhydrocarbons (sulfur, oxygen and/or metal containing compounds).
Natural gas	Gases that form from decomposition of organic material, including plants, animals and microorganisms under increased pressure and temperature of diagenesis. Can also be produced in lower temperature environments through anaerobic decomposition.	Primarily consists of methane, but also contains ethane, propane, butanes and other hydrocarbons, as well as gases such as carbon dioxide and hydrogen sulfide.
Oil shale	Organic rich fine-grained sedimentary rock that contains kerogen, an organic compound mixture. Formed under lacustrine, marine or terrestrial depositional environments.	Kerogen has a variety of compositions that depend on origin. Humic kerogen formed from land plants; planktonic kerogen formed from plankton. Forms at lower temperature than oil and gas.
Gas-bearing shale	Also called tight shale or black shale, contains high concentrations of organic matter (TOC 3–15%). Gas can be trapped in pore spaces.	Natural gas (see above).
Oil sands	Also called tar sands. Mixture of sand, clay, water, and bitumen, which is a thick (viscous to solid) substance that forms in different ways, often from oil degradation by bacteria.	Bitumen contains high molecular weight hydrocarbon molecules that have high viscosity and high density.

1984; Filby, 1994). In addition, trace elements have also been used to characterize depositional environments of shales, including gas shales (see Tribouvillard et al., 2006; Rimstidt et al., 2017).

Despite the abundance of papers on trace elements in crude oils and gas shales, and a plethora of papers on As in coal, there are no recent papers, to our knowledge, that focus on the behavior of As through the formation, production, and use of hydrocarbons, which is the central goal of this review. Before we address the larger goal, however, we must first consider the sampling and analytical methods used to measure As in hydrocarbons and associated products, as accurate measurements of As and its organic and inorganic species are critical for understanding how As behaves within the hydrocarbon life cycle.

1.2. Sampling and analytical methods for As in hydrocarbons

Arsenic compounds, including volatile, organic forms, are difficult to sample and measure in hydrocarbons. First, there is potential for contamination from external sources; thus ensuring a representative sample is challenging (Stigter et al., 2000). Second, it is important to store and preserve samples to stabilize the As species (Brandão et al., 2006; Harrington et al., 2010; Reimer et al., 2010) as biological activity, exposure to air and other processes can cause changes in species over time (Reimer et al., 2010). Third, extracting As compounds, especially organic compounds, from hydrocarbon matrices involves several intricate steps (Harrington et al., 2010; Mello et al., 2012). Because concentrations of these As compounds can be in the part per billion or even part per trillion range, each step must be conducted carefully to prevent changes in As speciation and to avoid contamination. Last, there are many analytical approaches to choose from; each has advantages and

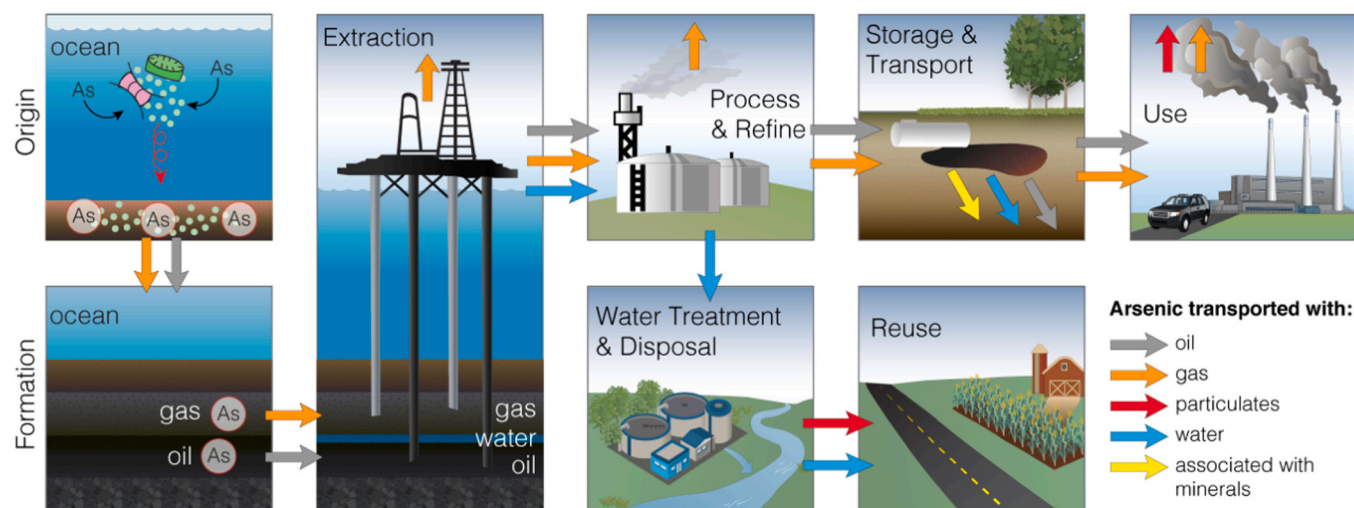


Fig. 1. Schematic of As in hydrocarbon production, starting with incorporation of As from marine organisms into hydrocarbons, through the processes of hydrocarbon production, process and refining, storage, and use, and the parallel process of wastewater treatment, disposal and reuse. The colored arrows show the main transport modes of As, including with oil, natural gas, particulates, and water. “Associated with minerals” refers to the adsorption of As to minerals such as iron oxides and clays in aquifers.

disadvantages with respect to detection limit, interferences, efficiency, and cost. Mello et al. (2012) reviewed sample preparation and analytical methods for metals in crude oil and serves as an excellent reference for this information.

Prior to analysis, As species must first be extracted from the hydrocarbon and/or geologic medium. Arsenic compounds can be extracted from crude oil in several ways, but the choice must take into account a variety of factors. Oxidation, for example, will impact the ability to analyze speciated As forms (Puri and Irgolic, 1989). Other methods include boiling water and acid digestion methods, but these can also impact As speciation (Puri and Irgolic, 1989). The matrices of oil sands and oil shales have additional challenges in that the bitumen (oil sands) or kerogen (oil shale) must be first separated from the mineral fraction. There are several approaches for separation, which often include use of solvents, hot water extractions (Jacobs and Filby, 1983) or acid digestions (Bicalho et al., 2017). Extracting volatile As species from natural gas is particularly challenging (Puri and Irgolic, 1989; Delgado-Morales et al., 1994b; Krupp et al., 2007; Xu et al., 2015; Cachia et al., 2017). Extraction methods include use of adsorption tubes or solutions (silver nitrate or nitric acid) to take up the volatile species such as trimethylarsine (TMA) from natural gas prior to analysis.

Methods to analyze trace concentrations of As, especially organic As compounds, have evolved significantly. For decades, methods such as neutron activation (INAA or NAA), X-ray fluorescence (XRF), atomic absorption spectroscopy (AAS) and inductively coupled plasma mass spectrometry atomic emission spectrometry (ICPAES), have been successfully used to measure As. Each of these methods has advantages and disadvantages in terms of need for sample pretreatment and digestion, sample pre-concentration, matrix interference, issues of sample stabilization, sensitivity and detection limits (Puri and Irgolic, 1989). The advent of hydride generation helped to lower detection limits and to identify methylated As compounds.

More recently, the application of inductively coupled plasma mass spectrometry (ICPMS) to As analysis has allowed for higher sensitivity with lower detection limits. Coupling of ICPMS with methods to separate As compounds via high performance liquid chromatography (HPLC) or other liquid chromatography methods prior to analysis allows for high sensitivity detection of multiple As species, including inorganic and organic forms (e.g., Garbarino et al., 2002; Harrington et al., 2010) in aqueous samples. Gas chromatography (GC) coupled with ICPMS has also been used to directly measure As species in natural gas without needing to extract the species first (Krupp et al., 2007).

Most papers on methods focus on analyzing As in specific hydrocarbon sources, such as crude oil (Puri and Irgolic, 1989; Hardaway et al., 2004; Sánchez et al., 2013), oil sands (Bicalho et al., 2017), gasoline, diesel and naphtha (Brandão et al., 2006), and natural gas (Irgolic et al., 1991; Delgado-Morales et al., 1994b; Krupp et al., 2007; Xu et al., 2015; Cachia et al., 2017; Freije-Carrelo et al., 2017). As there are dozens of papers on analysis of As species in different hydrocarbon products, the reader is directed to these papers in addition to reviews (e.g., Nadkarni, 1991; Cassella et al., 2002; Sánchez et al., 2013) for more information.

1.3. Arsenic concentrations vary by hydrocarbon type

1.3.1. Crude oil

Arsenic within marine organisms is likely the dominant source of As contained in crude oil (Irgolic et al., 1991). Arsenobetaine, the primary organic As compound in marine organisms, is likely decarboxylated at elevated temperatures and pressures of diagenesis (Puri and Irgolic, 1989; Irgolic et al., 1991) to form a variety of organoarsenic compounds within the oil (Filby, 1994) that are often associated with asphaltenes (Filby, 1994). Concentrations and forms of As in crude oil are controlled by the depositional environment, the organic material, the degree of maturation, the chemical composition of the oil, and the diagenetic history (Filby, 1994). However, there are still many questions about the genesis of As compounds in hydrocarbons that require additional work (Puri and Irgolic, 1989). Concentrations of As measured in crude oils compiled from the literature are shown in Table 2 (Becker et al., 2012; Chifang et al., 2005; Jungers et al., 1975; Kitto, 1993; Olsen et al., 1995; Puri and Irgolic, 1989; Reboucas et al., 2003; Shah et al., 1970; Stigter et al., 2000; Trindade et al., 2006; Hitchon and Filby, 1982). The concentrations are also shown in a boxplot in Fig. 2 to compare with As concentrations in other hydrocarbons.

1.3.2. Natural gas

Conventional natural gas, similar to crude oil, is derived from marine organisms that decompose under elevated temperature and pressures. Also similar to crude oil, As that is contained within organisms is the likely source of volatile As in natural gas due to decarboxylation reactions at high temperatures (Puri and Irgolic, 1989). The main form of As identified in natural gas is TMA (Irgolic et al., 1991; Delgado-Morales et al., 1994b), accounting for 55–80% of the total As (Irgolic et al., 1991), with lower concentrations of dimethylethylarsine (DMEA),

Table 2

Arsenic concentrations in crude oil and petroleum products compiled from the literature. Data reported were collected from primary sources; averages reported in secondary sources are not included. Concentrations reported as mass per volume were converted to mass per mass assuming densities of 873 kg/m³ (crude oil), 740 kg/m³ (naphtha) and 749 kg/m³ (gasoline). Mass concentrations were converted to mg/kg to allow for comparison. HG = hydride generation, AES = atomic emission spectroscopy, AAS = atomic absorption spectroscopy, INAA = instrumental neutron activation analysis, ICPMS = inductively coupled plasma mass spectrometry. - = not reported.

Type	Mean As mg/kg	Median As mg/kg	Min As mg/kg	Max As mg/kg	#	Location or Crude Oil Types	Extraction or dissolution method	Analytical method	Reference
Crude oil	19.47	0.92	0.04	578	50	—	Extraction with boiling water or nitric acid, mineralization with nitric & sulfuric acid	HG-AES	Puri and Irgolic (1989)
Crude oil	0.103	0.008	0.02	1.988	69	Alberta, Canada	NA	INAA	Hitchon and Filby, 1982
Crude oil	0.262	0.111	0.046	1.11	10	Libya, U.S. (California, Louisiana, Wyoming)	Benzene	INAA	Shah et al. (1970)
Crude oil	0.011	0.0129	< 0.01	0.037	23	Arabian, Iranian, Ural, Kuwait	Sulfuric acid/hydrogen peroxide digestion	HG- AAS	Stigter et al. (2000)
Crude Oil	3.49	0.24	0.04	26.2	8	China	Benzene-methanol rinse	INAA	Chifang et al. (2005)
Crude oil	0.131	0.108	< 0.06	0.280	4	North Sea	Xylene	ICPMS	Olsen et al. (1995)
Naphtha	0.0022	0.0021	0.0008	0.0039	11	Brazil, Argentina, Morocco, Algeria	None	Electrothermal AAS	Reboucas et al. (2003)
Gasoline	0.0263	—	—	—	3	—	Cold finger system	HG-AAS	Becker et al. (2012)
Gasoline	0.966	—	—	—	3	—	Nitric acid / hydrogen peroxide	HG-AAS	Trindade et al. (2006)
Gasoline	—	—	< 0.001	0.013	44	Cities across U.S.	Oxidation	Spark source MS	Jungers et al. (1975)
Fuel oil	0.020	0.018	0.0079	0.048	19	Maryland, U.S.	Ashing	INAA	Kitto (1993)

methyldiethylarsine (MDEA), and triethylarsine (Irgolic et al., 1991; Krupp et al., 2007). Although there are microbially mediated processes that generate TMA, Puri and Irgolic (1989) suggest that the majority of TMA in natural gas is a product of diagenetic reactions and not from modern microbial activity. Although As measurements in natural gas have been reported in a few studies (Irgolic et al., 1991; Krupp et al.,

2007; Cachia et al., 2017), the sampling strategies, phases sampled and measurement units are significantly different, making concentrations difficult to summarize and compare.

1.3.3. Oil shale

Arsenic within oil shale deposits is present in both the kerogen and in

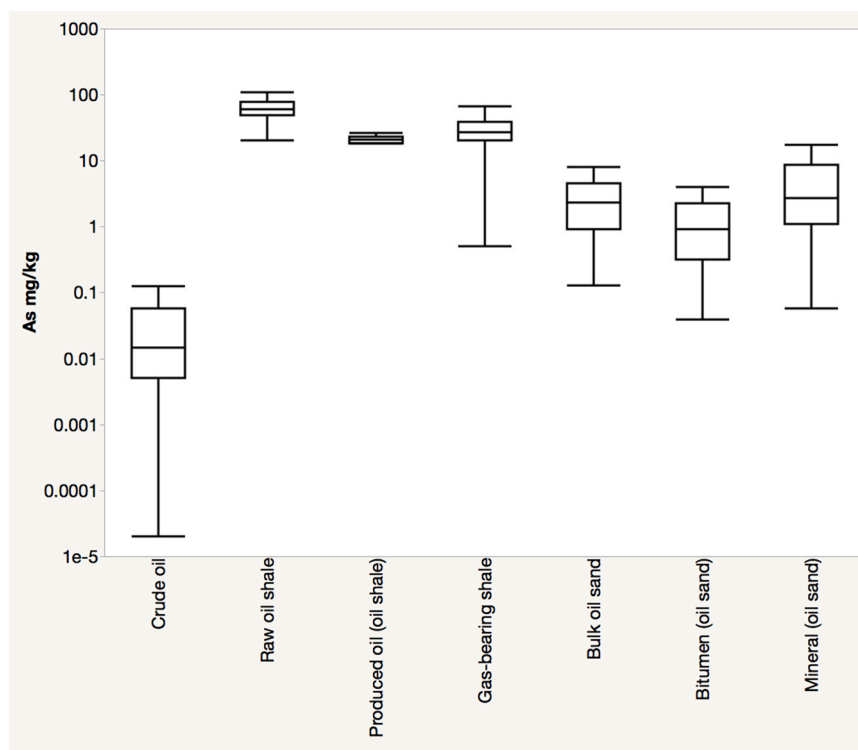


Fig. 2. Boxplots of arsenic concentrations (log scale) in select hydrocarbons discussed in the text. Concentration data were gathered from the references shown in Tables 2–5. The middle line in the boxplots show the median value, the ends of the box represent the 1st and 3rd quartiles, and the whiskers show the 1st quartile - 1.5*(interquartile range) and the 3rd quartile + 1.5*(interquartile range).

shale rock. Kerogen can contain elevated As concentrations from the As contained in the algae and higher plant material, a precursor to humic kerogen (Fish, 1983). Arsenic can also occur in shale within minerals such as pyrite or adsorbed to clays (e.g., Chermak and Schreiber, 2014). Concentrations of As in raw shale (shale + kerogen) and the oil produced through retorting (produced oil) compiled from several studies are shown in Table 3 (Fruchter et al., 1978; Shendrikar and Faudel, 1978; Wildeman and Meglen, 1978; Fish, 1983; Fish et al., 1987; Patterson et al., 1987; Patterson, 1988). The concentrations are also shown in a boxplot in Fig. 2 to compare with As concentrations in other hydrocarbons.

A variety of As species have been measured in raw shale, including organic and inorganic forms (Fish, 1983; Schraufnagel, 1983; Cramer et al., 1988; Puri and Irgolic, 1989). Cramer et al. (1988) found that oxidized As was associated with oxygen and/or carbon whereas reduced As species was associated with sulfur (S) or metals; the form of As species differed between kerogen and shale. Organic forms of As that have been identified in raw shale or in extracts of raw shale include MMA and phenylarsonic acid (Fish, 1983; Schraufnagel, 1983; Cramer et al., 1988). Weiss et al. (1983) speculated that organoarsenic in the produced oil is associated with iron (Fe)-humic complexes. Although more recent papers have not addressed this specific question, studies have found that As can bind with Fe-complexed natural organic matter in aquatic systems (Ritter et al., 2006; Sharma et al., 2010; Liu et al., 2011; Mikutta and Kretzschmar, 2011), suggesting that this association is a possibility.

1.3.4. Gas-bearing shales

Similar to oil shale, As in gas-bearing shales is contained within both the organic material and the shale itself. Shale often contains pyrite, which can contain trace to percent levels of As, and clay minerals to which As can adsorb (Leventhal, 1998; Balaba and Smart, 2012; Chermak and Schreiber, 2014; Phan et al., 2015; Stuckman et al., 2019). Concentrations of As measured in gas-bearing shales compiled from the literature are shown in Table 4 (Leventhal, 1980; Frost et al., 1985; Bracht, 2010; Balaba and Smart, 2012; Phan et al., 2015; Wang et al., 2015, 2016; Stuckman et al., 2019) The concentrations are also shown in a boxplot in Fig. 2 to compare with As concentrations in other hydrocarbons.

1.3.5. Oil sands

Arsenic concentrations in oil sands, including the bulk rock, the bitumen and the mineral fraction (also called sediment residue or fines), have only been reported in a few studies (Table 5) (Bicalho et al., 2017; Jacobs and Filby, 1983; Hitchon and Filby, 1982; Hosterman et al., 1990). Similar to the sources above, As is contained in both the organic and inorganic fractions. The concentrations are also shown in a boxplot in Fig. 2 to compare with As concentrations in other hydrocarbons.

Table 3

Arsenic concentrations in oil shale compiled from the literature. Raw oil shale = shale + kerogen. Produced oil = oil produced through retorting of the kerogen in the shale. Fm = formation. HPLC = high performance liquid chromatography, GFAA = graphic furnace absorption spectrometry, AAS = atomic absorption spectrometry, INAA = instrumental neutron activation, ICPAES = inductively coupled plasma atomic emission spectroscopy, - not reported, * additional methods used to analyze gaseous forms.

Mean As in raw shale mg/kg (n)	Mean As in produced oil mg/kg (n)	Location	Extraction, dissolution ore preparation method	Analytical method for As	Reference
20 (1)	–	Green River Fm, U.S.	Organic solvents	HPLC-GFAA	Fish (1983)
–	23 (1)	Green River Fm, U.S.	Organic solvents	HPLC-GFAA	Fish et al. (1987)
62 (3)	21.2 (3)	Green River Fm, U.S.	Nitric acid/sulfuric acid digestion	AAS	Shendrikar and Faudel (1978)
81 (3)	12 (2)	Green River Fm, U.S.	Fusion, acid digestion	INAA*	Fruchter et al. (1978)
75 (1)	26 (1)	Green River Fm, U.S.	Shale: fusion, acid digestion. Oil: acid digestion	INAA	Wildeman and Meglen (1978)
8.5 (4)	–	Rundle deposit, Australia	–	INAA	Patterson (1988)
50 (1)	23 (1)	Julia Creek oil shale, Australia	Organic solvents	INAA or ICPAES	Patterson et al. (1987)

2. Arsenic released from hydrocarbon production

Arsenic can be released to the environment through hydrocarbon production, in which hydrocarbons are recovered or extracted from the source, followed by processing and refining. After extraction from the geologic source, hydrocarbons are processed using different physical, chemical and thermal methods to extract the specific product and to remove non-hydrocarbon components, such as water, gases, and contaminants such as S, nitrogen (N) and metals/metalloids. Hydrocarbons are then further refined to generate specific commodities or to meet specific qualifications. For example, in oil refineries, crude oil is converted into gasoline, fuel oil and jet fuel, in addition to other products such as naphtha, lubricants, petroleum coke asphalt, gases and petrochemicals, among others. To refine crude oil, processes include separation, distillation, and various types of cracking, blending, and extractions. Some of these processes can release As (and other trace elements) to the environment, described in more detail below.

2.1. Arsenic release from conventional crude oil production

Primary recovery of conventional crude oil relies on natural pressure to bring oil to the surface, followed by use of pumps. Secondary recovery methods include water flooding or natural gas flooding, for which two wells are needed to inject and produce the hydrocarbon. Tertiary, or enhanced oil recovery, methods include thermal recovery, gas injection (CO₂) to lower viscosity and increase oil flow, and chemical injection, done by injecting chemicals such as polymers to help improve recovery (Hsu and Robinson, 2019).

Oil extracted using these methods can contain water, gases, and other compounds that need to be separated and managed. Produced water, which includes formation and injected water, creates a large volume waste stream. For example, in 2003, an estimated 667 million metric tons (about 800 million m³) of produced water were discharged to the ocean from offshore facilities throughout the world (Neff et al., 2011). Because produced water is in contact with geologic formations, it can contain elevated inorganic solutes, including As. Another waste product from oil and gas production is the cuttings from drilling, which may also release As depending on their content. More information on produced water and cuttings will be discussed in Section 4.

After extraction, crude oil undergoes processing to separate oil from gas and water and remove salt, S and other compounds (Jahn et al., 1998), followed by processing including separation, distillation, and various types of cracking, blending, and extractions. Several of these processes require use of catalysts, which often contain Ni. Because As can react with Ni to form nickel arsenide, which causes catalyst poisoning and deactivation, many refineries use guard catalysts with high surface area to remove As from feedstocks (Bhan, 2004). Once concentrated on the guard catalysts, the As can be properly disposed.

Table 4

Arsenic concentrations in gas-bearing shale compiled from the literature. Fm = formation. ICP = inductively coupled plasma, MS = mass spectrometry, OES = optical emission spectroscopy, INAA = instrumental neutron activation analysis. GFAAS = graphite furnace atomic absorption spectroscopy, XRF = x-ray fluorescence, - = not reported.

Mean As mg/kg	Median As mg/kg	Min As mg/kg	Max As mg/kg	n	Location	Extraction or dissolution method	Analytical method	Reference
22.1	16	4.9	90	14	Marcellus Fm, U.S.	Digestion with HNO ₃ , HF, HCl, HClO ₄	ICPMS	Phan et al. (2015)
30.5	20	12	67	5	Marcellus Fm, U.S.	Fusion with LiBO ₂	ICPOES and ICPMS	Stuckman et al. (2019)
32.5	26	4.9	165	658	Marcellus Fm, U.S.	Fusion with Na peroxide	ICPOES and ICPMS	Bracht (2010)
149	153	139	154	3	Marcellus Fm, U.S.	Digestion with HNO ₃ and HF	GFAAS	Balaba and Smart (2012)
25.0	23	13	50	7	Antrim Shale, U.S.	-	XRF	Leventhal (1980)
27.6	24	0.50	190	293	New Albany Shale, U.S.	-	INAA	Frost et al. (1985)
60.5	-	-	-	2	Bakken Fm, U.S.	Digestion with HNO ₃	ICPMS	Wang et al. (2016)
25.2	-	-	-	2	Eagle Ford Fm, U.S.	Digestion with HNO ₃	ICPMS	Wang et al. (2015)

Table 5

Arsenic concentrations in oil sands compiled from the literature. Concentrations include those analyzed from bulk material, bitumen, and mineral (sediment residue, fines) fractions. INAA = neutron activation, ICP-SFMS = inductively coupled plasma sector field mass spectrometry, - = not reported.

Mean As in bulk material mg/kg (n)	Mean As in bitumen mg/kg (n)	Mean As in mineral mg/kg (n)	Location	Extraction method	Analytical method	Reference
-	0.56 (6)	6.3 (2)	Athabasca I, Canada	Soxhlet extraction with toluene	INAA	Jacobs and Filby (1983)
-	0.146 (6)	15.4 (2)	Athabasca II, Canada	Soxhlet extraction with toluene	INAA	Jacobs and Filby (1983)
-	0.361 (2)	-	Athabasca, Canada	Soxhlet extraction with benzene	INAA	Hitchon and Filby, (1982)
1.2 (7)	0.3 (7)	1.1 (7)	Athabasca, Canada	Microwave digestion with HNO ₃ -HBF ₄	ICP-SFMS	Bicalho et al. (2017)
4.39 (18)	2.73 (21)	4.90 (2)	U.S., Canada, Trinidad/Tobago, Venezuela	Benzene (for bitumen)	INAA	Hosterman et al. (1990)

2.2. Arsenic release from conventional production of natural gas

As discussed in Section 1, natural gas can contain volatile As compounds such as TMA (Irgolic et al., 1991). Because TMA is volatile, it can be released to the atmosphere during natural gas extraction and production. Through analysis of volatile As in natural gas and condensate from an off-shore platform, Krupp et al. (2007) calculated As release from the wells assuming different gas to condensate ratios. The calculated As release from these wells ranged from about 8000 g/m³ to ~16,000 g/m³. Due to lack of data on As release from natural gas production at the global scale, it is currently not possible to assess how As release from natural gas compares with other sources of volatile As, such as volcanoes, mining, smelting, and coal combustion. Research on persistence of TMA in the atmosphere (Pantsar-Kallio and Korpela, 2000) suggests that it decomposes quickly, but estimates on global As release from natural gas (or other hydrocarbon) production have not been made.

During processing, natural gas undergoes physical and chemical treatment to purify the gas and to remove unwanted gases and condensates, including As (Krupp et al., 2007). Methods for As removal include use of chemicals such as potassium peroxydisulfate (Delgado-Morales et al., 1994a) and separation methods such as pulsed sieved plates (Chaturabul et al., 2012). Once the As has been removed from the gas, it can be disposed of properly.

2.3. Arsenic release from gas-bearing shales

In cases where natural gas cannot be produced commercially from gas-bearing shales without altering the rock permeability or fluid viscosity (Hsu and Robinson, 2019), the gas can often be extracted using hydraulic fracturing (discussed in more detail in Section 4). In this process, recovery of the gas requires injection of a fracturing fluid and a

proppant under high pressure in order to create fractures and increase rock porosity and permeability. The introduction of fluids to gas-bearing shales at high pressure during the fracturing process can cause geochemical reactions at depth within the shale (Vidic et al., 2013), the *in situ* fluids, and the fracturing fluids (Soeder and Kappel, 2009; Marcon et al., 2017).

Interactions of fluids with shale can stimulate a variety of biogeochemical processes. Shales primarily consist of mixtures of fine-grained quartz and clay minerals and can contain other minerals including feldspars, carbonate minerals, sulfide minerals, and oxide minerals in addition to organic carbon (Yaalon, 1962; Vine and Tourtelot, 1970; O'Brien and Slatt, 1990; Slatt and Rodriguez, 2012; Chermak and Schreiber, 2014). These minerals host a variety of trace elements, including As (Leventhal, 1998; Chermak and Schreiber, 2014). Fluid interaction with these minerals can trigger geochemical reactions such as pyrite oxidation, carbonate dissolution, clay transformation, desorption of trace elements, and complexation of trace elements. In addition to these reactions, high salinity total dissolved solids (TDS) fluids can be flushed from pore spaces (Wang et al., 2015, 2016; Wilke et al., 2015; Marcon et al., 2017). These downhole reactions affect mobilization of major and minor elements, including As. Introduction of oxidized fluids into shales can promote pyrite oxidation, which, if the pyrite is As-bearing, can release As, sulfate and protons (Nordstrom, 1982; Rimstidt and Vaughan, 2003). However, if the shale contains carbonate, dissolution of carbonate minerals can buffer the acid production. The impact of these acid producing and buffering reactions affect pH, Fe, S and As cycling, since As adsorbs to Fe oxides produced from pyrite oxidation, but desorption of As from metal oxides and clays increases with increasing pH (Smedley and Kinniburgh, 2002). Experimental studies of the Eagle Ford, Bakken and Marcellus shales show that interaction of the shales with simulated fracturing fluids of different chemistries results in mineral dissolution, precipitation, and changes in

pH, all of which can influence As mobility (Wang et al., 2015, 2016; Marcon et al., 2017).

In addition to geochemical reactions, microbial processes also strongly influence redox reactions involving As (Cullen and Reimer, 1989; Ahmann et al., 1994; Newman et al., 1998; Stolz and Oremland, 1999; Inskeep et al., 2002; Oremland and Stolz, 2003; Santini et al., 2002; Oremland and Stolz, 2005; Santini and Ward, 2018). Bacteria can reduce As(V), oxidize As(III), and methylate As for a variety of reasons, including detoxification and generation of energy. These organisms are phylogenetically diverse and can be found in a wide range of environments (Oremland and Stolz, 2003; Santini and Ward, 2018). Because As (III) and As(V) exhibit different adsorption characteristics (Dixit and Hering, 2003), reduction or oxidation of As can impact the ability of As species to attach to mineral surfaces (Langner and Inskeep, 2000; Zobrist et al., 2000). Organisms that reduce Fe and S can also affect As mobility. For example, reductive dissolution of hydrous ferric oxides (HFOs) can change the surface area and properties of the mineral, resulting in changes in sorption/desorption mechanisms (e.g., Tadanier et al., 2005). Rates of reductive dissolution are affected by Fe mineralogy and the surface adsorption of reaction products, including Fe(II) and As species (Ford, 2002; Roden and Urrutia, 2002; Hansel et al., 2004). In addition, bacterial sulfate reduction generates sulfide, which can bind with both Fe and As to remove As from water (Moore et al., 1988; Rittle et al., 1995; Kirk et al., 2004; Keimowitz et al., 2007; Sun et al., 2016).

Fluids that are produced during hydraulic fracturing, including flowback water, formation water, injected fluids, water condensing from the gas phase, and mixtures of these, are called produced water (Engle et al., 2014). Disposal of produced water and sludge/drilling cuttings after they are brought to the surface and the potential reuse or recycling of these materials are discussed in more detail in Section 4.

2.4. Arsenic release from production of oil shale

Oil shale can be produced *ex situ* through mining or *in situ* through introduction of heated water under pressure. Kerogen can be extracted from shale using pyrolysis (or retorting), which uses heat in the absence of oxygen to release the hydrocarbon from the shale. More information on retorting is provided in other sources (Piver, 1983). During this process, kerogen decomposes into vapors, which are cooled and condensed into oil.

Several studies conducted in the late 1970s examined As partitioning during the retort process and showed that the majority of As mass during the retort process remains with the shale. Shendrikar and Faudel (1978) simulated the retorting process and analyzed trace elements in raw and processed shale, the produced oil, and water. Results show that 95% of As ended up in the processed (also called spent) shale, 5% ended up in the produced oil and negligible mass partitioned into retort water. Fruchter et al. (1978) also conducted a mass balance on trace elements during retorting and included analysis of As in gas. Results showed almost all the mass (reported as 100%) remained in the processed shale, 5% partitioned into the produced oil, and less than 0.1% of the initial As mass was measured in the retort water and gas. Fruchter et al. (1978) report that most of As in gas occurred as arsine (AsH₃); measurements before and after the gas went through an emissions control device showed that As in off gas after treatment was reduced to about 10% of the initial concentration, with the remaining mass trapped as particulates in a filter.

The inclusion of As in oil produced from oil shale creates several problems during refining (Sikonia, 1985). First, the arsine gas can oxidize to form solid As trioxide, which can clog filters and over time, pipes, as shown in the Fruchter et al. (1978) study. Second, as discussed previously for crude oil, As will poison catalysts that are used in cracking and other processes involved to refine the hydrocarbon. Thus, pre-treatment of the produced oil is needed to remove As. In addition to the issues of elevated As concentrations in produced oil, there are several environmental issues related to As in oil shale production.

Because most of the As mass remains with the shale during retorting, the processed shale reflects a waste product that must be managed, similar to the cuttings waste produced during the hydraulic fracturing process for production of unconventional oil and gas (see discussion of solids management in Section 4). In addition, although the percent mass of As is low in retort water compared with the mass that remains in the shale, even low concentrations of As in water are toxic for humans.

2.5. Arsenic release from production of oil sands

The hydrocarbon extracted from oil sands consists of mostly bitumen, which is denser and more viscous than crude oil. Oil sands are produced using open pit mining or steam extraction (Hsu and Robinson, 2019; AGI, 2020). In open pit mining, the oil sands, a mix of sand, water and bitumen, are mined and then separated into sand, water, and bitumen (AGI, 2020). In steam extraction, steam is injected through wells into deeper deposits to heat sand and make bitumen flow more easily. The bitumen is recovered in a separate well to bring to the surface. Once the bitumen is extracted, it is often diluted before being processed (Hsu and Robinson, 2019).

After extraction, bitumen is transported to a processing (often called upgrading) facility, where it is converted into synthetic crude oil (AGI, 2020). This is done by chemical treatment, including mixing the bitumen with lighter oils or chemicals, and fractionation/cracking to break down the larger bitumen molecules into smaller ones. From here, the synthetic crude oil is sent via pipelines to a refinery, where it is converted into gasoline, diesel and other products, similar to the refining processes described above for conventional oil refining.

Arsenic release to the environment from oil sands production has been evaluated in several studies, but to date, there is no consensus in the literature on whether significant As release occurs from oil sands development. Kelly et al. (2010) reports that As can be released via airborne emissions from the mining operations and from upgrading facilities processing the ABS (Athabasca Bituminous Sands) deposit, which can introduce particulate As into snow, which then, during snowmelt, releases particulate As to tributaries (Kelly et al., 2010). However, particulate As is also associated with land clearing, mining and road dust associated with oil sands development (Kelly et al., 2010). Bari et al. (2014) found elevated As in atmospheric deposition at two sites located within 20 km of ABS development compared to locations further away. Timoney and Lee (2009) examined environmental impacts of oil sands production in Alberta; results suggest that oil sands development resulted in a variety of impacts, including increasing As concentrations in sediment of Lake Athabasca to levels that exceed freshwater guidelines for fish protection. In contrast, several other studies have not observed a significant connection between As and oil sands production. Donner et al. (2019) examined As in moss from bogs in northern Alberta near the ABS deposits. Examination of a peat core showed that As increased from atmospheric deposition in the early 1900s but declined in the mid-1900s, suggesting that the elevated As was associated with worldwide coal combustion that decreased due to implementation of air pollution controls (Donner et al., 2019). Several other studies (Shotyk et al., 2016; Donner et al., 2017; Owca et al., 2020) suggest that oil sands production has not significantly increased metals and trace element pollution and that observed changes are likely due to other human activities (i.e., coal combustion) or natural processes. Additional research could address questions about release of As, and other trace elements, from oil sands production.

3. Arsenic release from hydrocarbon storage, transportation and use

Arsenic can be introduced into the environment through both direct and indirect mechanisms associated with hydrocarbon storage, transportation, and use (Fig. 1). Leaks of hydrocarbons from above and below ground storage tanks and releases from pipelines, and truck, rail and

water carriers (tankers, barges) have had widespread impact on both human and environmental health. Because As is contained within hydrocarbons (see Section 1), direct release of As can occur from these leaks and spills into the atmosphere, soil, surface water, groundwater and the ocean; however, the quantification of these loads is challenging due to the lack of As analyses for site-specific hydrocarbon sources. Arsenic can also be directly released to the atmosphere through volatile and particulate emissions resulting from hydrocarbon combustion. In addition to direct release, biodegradation of hydrocarbons coupled to Fe-reduction can trigger indirect release of geogenic As to groundwater, a process that has been recently documented in detail at the U.S. Geological Survey (USGS) Bemidji research site, discussed in more detail below.

3.1. Arsenic release from hydrocarbon storage and transportation

Due to the vast volumes of hydrocarbons that are stored and transported across land, inland waterways and the ocean, leaks and spills are common and often have widespread and catastrophic impact. Hundreds of books, papers and news articles have been written on the topic of hydrocarbon spills in terrestrial and marine environments and cannot be repeated here. Below, we highlight some key points that pertain to As.

Hydrocarbons are stored in both above ground storage tanks (ASTs) and underground storage tanks (USTs). Historically, leaking USTs have been a major source of soil and groundwater pollution; in the United States, there are currently ~542,000 USTs that store petroleum and other hazardous products (USEPA, 2020). The U.S. Environmental Protection Agency (USEPA, 2020) reports over 557,000 confirmed releases from leaking USTs since 1984, with close to 494,000 cleanups completed. Published estimates of the volume of hydrocarbons that have been released from storage tanks could not be found for this review.

Hydrocarbons can also be released to the environment during transport from oil wells to refineries (upstream) and from refineries to markets (downstream). Transportation modes include pipeline, tanker/ barge, rail and truck. The U.S. Department of Transportation Bureau of Transportation Statistics (USDOT, 2020a) provides data on transportation of crude oil and refined petroleum products in the United States. In 2018, these data show that almost 75% of combined crude oil and petroleum products were transported in the United States via pipeline, with 15% by water carrier and 10% by rail. Refined petroleum products, such as gasoline, are also transported regionally via trucks. Leaks and spills from these transportation modes release hydrocarbons to soil, surface water, groundwater and the ocean. For example, from 2010 through April 2020, ~4100 petroleum pipeline spills were reported in the United States, totaling 42 million gallons of released hydrocarbons (PHMSA, 2020). Rail transport of crude oil and petroleum products has increased significantly within the United States since 2010 (USDOT, 2020a), thus increasing the likelihood of spills from rail accidents.

Oil spills in the marine environment can release massive volumes of hydrocarbons through tanker accidents and blowouts and also from operational discharges and pipeline leaks (Jernelöv, 2010). For example, the Deepwater Horizon accident of 2010 released an estimated 134 million gallons of crude oil into the Gulf of Mexico (NOAA, 2020). Oil releases during the Persian Gulf War in the early 1990s were even more extensive, releasing an estimated 462 million gallons into the Persian Gulf (Michel, 2011). In addition to these catastrophic releases, spills occur frequently from vessels and pipelines (several thousand incidents per year; hundreds of thousands of gallons released per year (USDOT, 2020b)) in navigable inland waters. For example, in 2010, a pipeline leak released between 800,000 and one million gallons of dilute bitumen produced from oil sands into the Kalamazoo River, Michigan (USEPA, 2010), which was the largest release of oil into an inland waterway in the United States. (Zhu et al., 2018). In addition to these accidents, operational discharges (tank washing, flushing of ballast water) also release hydrocarbons to aquatic systems. Combined, these

releases have had widespread and long-term ecological effects on marine fisheries and wildlife, which have had substantial impacts on the economy of the regions.

As presented in Section 1, hydrocarbons contain metals and trace elements, the concentrations of which are highly variable. Although there are few reports of metals and trace elements analyzed from marine spills, Santos-Echeandía et al. (2008) sampled the water column for copper (Cu), Ni and V above the wreckage of the Prestige, a tanker carrying 77,000 tons of heavy fuel oil, which sank in 2002 in the Atlantic Ocean near the coast of NW Spain. Using the data, the authors were able to estimate mass releases of these metals into the water column. Most of the contamination occurred in the upper water column (top 50 m) and was associated with seawater mixing with the fuel oil. Unfortunately, there are no similar studies that have been conducted focusing on As release from marine oil spills. Because there is no average As concentration that can be applied to all hydrocarbon sources, and the estimates of spilled volumes of oil have wide ranges and high uncertainties, we did not attempt in this paper to quantify the load of As directly released from hydrocarbon spills. Future work could focus on making these calculations to allow for an estimate of direct As release from hydrocarbon spills.

3.2. Indirect release of As to groundwater at hydrocarbon-impacted sites

Indirect release of As into groundwater can occur through the process of biodegradation of hydrocarbons. Geogenic As in aquifers is often associated with HFOs because As strongly adsorbs to these minerals. In oxic, circum-neutral pH conditions, As has a stable association with these minerals and is not released to groundwater. However, in the presence of a source of biodegradable organic carbon, such as hydrocarbons, bacteria can couple the oxidation of the organic carbon with the reduction of the As-bearing HFOs, which can result in As release to groundwater. Thus, although As may not be introduced to the aquifer directly from hydrocarbons, it can be mobilized through introduction of hydrocarbons.

This mechanism of indirect As release at petroleum-impacted sites was introduced in the literature in the 2000s (Brown et al., 2010; Burgess and Pinto, 2005; Ghosh et al., 2003; Hering et al., 2009). In 2011, the American Petroleum Institute published a Groundwater Arsenic Manual (API, 2011) which presents several case studies of As mobilization in groundwater at petroleum-impacted sites, including two refineries, a former reserve pit, and a former fuel storage facility. The manual focuses on practical aspects, including identifying petroleum-impacted sites where indirect As release may occur, and providing tools for assessing and managing As at sites where As release has occurred. Recent work at the USGS Bemidji Site has shed light on the cycling of geogenic As resulting from a hydrocarbon spill, described in the case study below.

3.2.1. Case study: arsenic cycling in a hydrocarbon plume

As a result of a crude oil pipeline burst in 1979, 1.7×10^6 L (about 10,700 barrels) of crude oil were released onto a glacial outwash deposit in northern Minnesota, near Bemidji (Baedecker et al., 1993; Essaid et al., 2011). After pooling at the surface, oil percolated through the vadose zone to the water table. Over the past 40 years, extensive biodegradation of the hydrocarbons has resulted in establishment of anoxic conditions in the plume. The dominant electron-accepting processes coupled with biodegradation are Fe-reduction and methanogenesis (Cozzarelli et al., 2010).

Due to the extensive Fe-reduction associated with biodegradation of hydrocarbons, geogenic As associated with HFOs in the glacial aquifer has been released into groundwater, forming its own plume (Cozzarelli et al., 2016). Because the site has been instrumented and sampled for decades as a USGS research site, there is an extensive spatial and temporal dataset on organic solutes, inorganic solutes, and field parameters (Essaid et al., 2011) that allow for an in-depth analysis of As cycling in

the aquifer. Fig. 3 shows the conceptual model of As cycling in the hydrocarbon plume, based on geochemical characteristics including benzene, dissolved oxygen, dissolved Fe and dissolved As concentrations.

Ziegler et al. (2017a) conducted chemical extractions of aquifer sediment in different redox zones of the aquifer and found an As-rich Fe “curtain” at the anoxic-oxic transition (leading edge of the plume); this transition zone has migrated over time as the plume has evolved. Results suggest that the aquifer has the capacity to naturally attenuate the As plume via sorption of As to HFOs.

Using both groundwater and sediment data, Ziegler et al. (2017b) conducted a mass balance of Fe and As in the Bemidji aquifer. Mass balance results showed that almost all of the Fe and As mass (99.9%, 99.5%, respectively) in the aquifer is associated with sediment. Results also suggested that over the lifetime of the plume, close to 80% of As originally associated with sediment has been mobilized to groundwater, but only a small amount (<1%) remains in groundwater due to the process of re-sorption onto HFOs. At the broader scale, this study documented that sediments can act as both sources and sinks for As, and that the source-sink relationship changes over time due to redox conditions.

Reactive transport modeling of As cycling at the Bemidji site (Ziegler, 2018) has allowed for long-term simulation of As release from aquifer sediments, subsequent mobilization in groundwater, and eventual attenuation of As due to resorption processes. In the reactive transport model, which is constrained by almost 30 years of data, As behavior is controlled by release from HFOs due to Fe reduction coupled with hydrocarbon biodegradation, and resorption to HFOs at the leading edge. Over time, however, as the hydrocarbon plume expands, resorbed As is re-released into groundwater due to an expanded zone of Fe reduction. This cycling suggests that although As adsorption is an important process for retaining As in aquifers, sorption can be transient and should not be assumed to provide a permanent sink for As in aquifer systems.

3.3. Arsenic release from hydrocarbon combustion

Combustion of hydrocarbons can release trace elements such as As into the atmosphere, both directly through stack emissions from refineries and indirectly from hydrocarbon combustion (Linak and Wendt, 1993; Stigter et al., 2000). Although most of the research on metal and metalloid release from combustion has centered on emissions from coal-fired power plants, combustion of hydrocarbons can also release toxic elements, including As.

Toxic elements can be released from combustion in both vapor phase, which can nucleate, condense and coagulate into aerosols, and as fine particulates (Linak and Wendt, 1993). The dynamics of these

releases are complex and are discussed elsewhere (see Linak and Wendt, 1993). Much of this material is removed from emissions prior to release. Removal efficiency is controlled by the particle size and type of technology used.

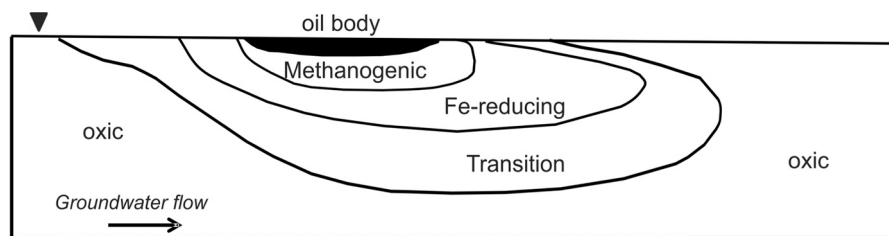
Because As concentrations vary in different fossil fuels (see Section 1), release of As through combustion also varies based on the fuel type. Vouk and Piver (1983) studied metal and trace element release from combustion of fossil fuels, including coal, fuel oil, and gasoline. Using concentration data of each source and combustion data from power plants from the late 1970s, the authors estimated mobilization through combustion. Results show that coal combustion resulted in the highest release of As, followed by fuel oil and gasoline. Natural gas was not examined because it was not widely used in the 1970s when these data were collected. The USEPA also examined released of hazardous air pollutants (HAPs), including As, from combustion of fossil fuels as a directive of the amendments to the Clean Air Act of 1990 (French et al., 1998). They collected data from 52 utilities, including coal, oil and natural gas plants, and used the data to estimate HAP emissions across the U.S. Data show that emissions from coal-fired plants have the highest As emissions (70 tons per year in 2010), followed by oil-fired plants (3 tons per year in 2010), and natural gas-fired plants (0.25 tons per year in 2010) (French et al., 1998).

Buerki et al. (1989) analyzed trace elements in aerosols released from traffic, industrial plants burning heavy oil, residential heating, and incineration plants in Switzerland. Results showed that although heavy oil concentrations of As were below detection of 1 mg/kg, particulates measured from heavy oil combustion residue had an average As concentration of 230 mg/kg.

More recently, there has been increasing interest in fine airborne particulate matter (PM) released from emissions. Huffman et al. (2000) conducted combustion experiments on residual fuel oils, and collected and separated PM from emissions into different size fractions, focusing on particles less than 2.5 μm in diameter (PM_{2.5}). Using scanning electron microscopy to examine particulates, Huffman et al. (2000) showed the highest As concentrations were in the PM_{2.5} fraction, ranging from 18.7 to 49.9 mg/kg; As concentrations in the size fraction greater than 2.5 ranged from 1.7 to 11.0 mg/kg. Future work to examine fine particulate matter could provide additional insight, as inhalation of these particles can have adverse health impacts, including lung cancer (see Hu et al., 2012; Mao et al., 2020).

4. Arsenic release from hydrocarbon production wastes

During development of hydrocarbon resources, liquid and solid wastes are produced that can contain As, due to the presence of As in the



Zone	DO mg/L	Benzene $\mu\text{g/L}$	Diss. Fe mg/L	Diss. As $\mu\text{g/L}$
Methanogenic	<0.1	800-4000	>30	>50
Fe-reducing	<0.1	5-800	1-30	5-50
Transition	0.1-1	1-5	0.5-1.0	1-5
Oxic	>1	<1	<0.5	<0.5

Fig. 3. Conceptual model of As cycling in the Bemidji plume based on the geochemical data in the table. Zones refer to terminal electron-accepting processes. Arsenic is released to groundwater in the methanogenic and Fe-reducing zones but retained in the transition zone. Modified from Cozzarelli et al. (2016)

source rocks and hydrocarbon phases, as discussed in Section 1. When these wastes are intentionally or unintentionally released to the environment, As can enter the air, water, soil and plant systems. Furthermore, release of wastes containing biodegradable carbon into soils or sediments containing geogenic As can result in the indirect release of As into surface and groundwater due to biogeochemical reactions as discussed in Section 3.2. In this section, we discuss As concentrations in these waste materials, focusing on the recent literature, due to the large increase in waste production during the last decade of expanding unconventional oil and gas (UOG) resource development. We review some of the water quality and ecological concerns related to development of oil and gas resources that include potential waste releases. We also review emerging issues related to beneficial reuse of waste materials as it relates to potential concerns regarding As mobilization.

4.1. Arsenic in oil field brine/produced water, wastewater

In conventional oil and gas (OG) wells, produced water is composed largely of a mixture of the brine present in the reservoir and any water injected to facilitate recovery of the hydrocarbons (Engle et al., 2014; Orem et al., 2014; Akob et al., 2015; Rowan et al., 2015). For continuous reservoirs, such as shale, recovery of the resource requires injection of a fracturing fluid and a proppant under high pressure (hydraulic fracturing) in order to create fractures and increase rock porosity and permeability. Produced water from unconventional resources (referred to here as UOG wastewater), therefore, contains inorganic and organic constituents present in the formation, as well as additives. The composition of UOG wastewater is variable, depending on the producing basin, and can range in salinity from like that of drinking water to several times more saline than sea water. UOG wastewater from Marcellus Shale gas wells, for example, typically contains high concentrations of TDS (approximately 200,000 mg/L), chloride (Cl), bromide (Br), sodium (Na), calcium (Ca), strontium (Sr), barium (Ba), radium (Ra) and organic compounds (Haluszczak et al., 2013; Orem et al., 2014; Akob et al., 2015; Rowan et al., 2015). Some produced waters from the Marcellus

Table 6

Arsenic concentrations in produced waters from the USGS Produced Waters Geochemical Database (Blondes et al., 2018), n = 284 samples with reported As concentrations. CBM = coal bed methane. SD = standard deviation, - = not reported.

Basin	Type	n	Range (mg/L)	Average (mg/L)	SD (mg/L)
Appalachian	Shale gas	89	0.009–0.151	0.074	0.036
Gulf Coast	Conventional hydrocarbon	31	0.0001–0.056	0.008	0.013
Raton	CBM	29	0.01–0.06	0.019	0.017
Black Warrior	CBM	26	0.001–0.475	0.024	0.090
Powder River	CBM	43	0.00007–0.4	0.019	0.066
Powder River	Conventional hydrocarbon	9	0.0019–5.1	0.763	1.571
Green River	Conventional hydrocarbon	12	0.0003–0.7	0.064	0.192
Green River	Tight gas	7	0.004–0.03	0.012	0.008
Green River	CBM	4	0.0003–0.02	0.006	0.008
Williston	Conventional hydrocarbon	12	0.01–0.316	0.170	0.098
Williston	Tight oil	11	0.063–0.566	0.338	0.175
Arkla	Conventional hydrocarbon	4	3–7	5.5	1.5
Big Horn	Conventional hydrocarbon	2	0.005–0.051	0.028	0.023
Hanna	Tight gas	2	0.001–0.003	0.002	0.001
Wind River	Tight gas	1	-	0.011	-
Wind River	CBM	1	-	0.2	-
Wind River	Conventional hydrocarbon	1	-	1.12	-

reach 360,000 mg/L (Shaffer et al., 2013). Marcellus Shale UOG wastewaters are enriched in As and selenium (Se) (Balaba and Smart, 2012), due to the substantial amount of these elements in the shale itself. Balaba and Smart (2012) underscore the difficulties of accurately measuring As concentrations in highly saline water samples, where analytical interferences cause challenges with both accuracy and precision. Those interferences can require a high level of sample dilution, resulting in many non-detects in UOG wastewater samples. Jubb et al. (2020) found commercial laboratories typically reported As values in UOG wastewater samples as below the detection limit due to the over-dilution of samples required by ICP-MS analyses. This could explain the limited data available for As in UOG wastewaters that have high salinity. In their study, Jubb et al. (2020) analyzed samples from five petroleum reservoirs by ICP-OES utilizing an argon humidifier and a high-salinity nebulizer and were able to detect and quantify trace elements, including As, with minimal, or no, dilution.

The USGS Produced Waters Geochemical Database (PWGD) reports on the chemistry of 114,943 samples of produced waters from hydrocarbon and geothermal energy development (Blondes et al., 2018). Of these samples, 284 report As concentrations in produced waters from hydrocarbon development (See Table 6). The highest concentrations of As (up to 7 mg/L) appear to be found in produced water from conventional hydrocarbon wells.

In addition to the samples reported in the USGS PWGD, chemical analyses of shale gas wastewater were collected from the Pennsylvania (PA) Department of Environmental Protection (PADEP) by Shih et al. (2015) for Marcellus Formation wells from 2009 to 2011. Arsenic concentrations were reported in a range from 23 to 99 µg/L in the sample reports. Maguire-Boyle and Barron (2014) sampled produced water from wells in the Marcellus (PA), Eagle Ford (Texas), and Barnett (New Mexico) formations and reported values of As ranging from below detection in the Eagle Ford to 26.1 mg/L in the Marcellus. In a review of produced water chemistry (Alley et al., 2011), gas field waters had substantially greater As concentrations, up to 11 mg/L, than either coal bed methane waters (up to 0.06 mg/L), or oil field waters (up to 0.857 mg/L); such differences are likely due to the very different depositional histories of the source rock.

4.2. Arsenic in sludges, drill cuttings, and other oil-field wastes

Shale gas development can generate substantial quantities of solid drilling waste that can be rich in As; this waste has the potential to be released into the environment during reuse, storage or disposal. In 2011, most of the 725,000 tons of drill cuttings produced in PA was disposed of in landfills; by 2013 this had increased to 1.2 million tons (Maloney and Yoxtheimer, 2012; Phan et al., 2015). Understanding the concentration and distribution of As in these waste materials is critical to predicting the potential for release of As during their use or disposal. Phan et al. (2015) carried out sequential extractions on drill cuttings and core material from the Marcellus Shale and surrounding units to identify the As reservoirs and concentrations. The As was primarily associated with sulfide minerals (16–90 mg/kg), and suggests the potential for As mobilization if drill cuttings are exposed to oxidizing conditions. Stuckman et al. (2015) investigated the leaching potential of As from Marcellus drill cutting wastes and found that As was equally present in oxidizable and reducible phases. They found that oil-based drill cuttings were especially susceptible to leaching As, along with Fe, uranium (U), manganese (Mn), and antimony (Sb), raising concerns for exposure of such cuttings to rainwater when stored in open pits. Stuckman et al. (2019) also studied leaching of metals from drill cuttings and cores collected at the Marcellus Shale Energy and Environmental Laboratory in West Virginia (WV). They found that As, Ni, V, cadmium (Cd), and U were mainly correlated with the organic carbon and S content in drill cuttings. Arsenic was leachable from both oxidizable and reducible phases, such as Fe oxides, and was influenced by the composition of the shale as well as the drilling muds containing minerals such as barite,

clays and organic additives. In contrast, Eagle Ford core leaching studies by Wang et al. (2015), which contained calcite in great excess compared to pyrite, resulted in As concentrations remaining low at all oxidant levels (only up to 3%) and below the EPA maximum contaminant levels of 10 µg/L for drinking water, an observation attributed to the relatively low amounts of As in the original solid sample and desorption processes.

Little is known about the potential for As release from the subsurface disposal of solid wastes produced during hydrocarbon production. Disposal in landfills exposes these waste materials to an anoxic environment, which could lead to the release of trace elements into groundwater. The presence of biodegradable organic carbon in landfills can develop an anoxic groundwater environment, similar to the anoxic conditions that develop from introducing hydrocarbons to aquifers (discussed in Section 3), which can result in the release and mobilization of As contained in the wastes as well as indirect release of geogenic As. For example, in a study at the Norman Landfill, in Norman, Oklahoma, Cozzarelli et al. (2011) found that As, Ni, Ba, Sr, Cd, chromium (Cr), and cobalt (Co) had substantially higher concentrations in anoxic wells downgradient from the municipal landfill than in oxygenated background wells.

4.3. Arsenic in drinking water/stream water from oil and gas production

There have been numerous recent investigations into potential impacts on water quality in regions of extensive oil and gas development. Fontenot et al. (2013) examined TDS, As, Ba, Se, and Sr in the Barnett Shale region and found that some private water well samples within a 3 km range of shale gas operations exceeded EPA's Drinking Water Maximum Contaminant Limit (MCL). Arsenic, Se, Sr, Ba, and TDS reached their highest concentrations in areas of active extraction in close proximity to natural gas wells, with 29 of 90 groundwater wells exceeding the As MCL of 10 µg/L. Concentrations were significantly higher in active extraction areas compared to reference samples and historical samples. Arsenic concentrations in active extraction areas ranged from 2.2 to 161.2 µg/L, with an average of 12.6 µg/L. The maximum concentration of As detected in a sample from an active extraction area was almost 18 times higher than both the maximum concentration among the non-active/reference area samples and historical levels from this region. In the active extraction region, 29 of 90 water wells exceeded EPA's As MCL. The mechanism responsible for the As in groundwater was not identified, and it was noted that the mobilization of geogenic constituents into private wells could have been caused by mechanical disturbances from intense drilling activity, reduction of the water table by drought or groundwater withdrawals, and faulty gas-well drilling equipment or well casings (Fontenot et al., 2013).

Knee and Masker (2019) evaluated whether stream water quality constituents, including As, were related to the intensity of UOG development in catchments in the Marcellus Shale region. They found significant positive relationships between As concentration in streams and UOG well density but the mechanism of mobilization was not identified. Mumford et al. (2020), however, found no statistically significant relationship between the intensity, presence, or absence of shale gas development and any signal in a comprehensive set of chemical constituents, including As, in their study of 25 small PA state forest watersheds over the course of 2 years. In another large-scale study of water quality data from PA watersheds, Olmstead et al. (2013) also found that the presence of shale gas wells upstream from a particular watershed did not result in increased Cl concentrations downstream; however, the release of treated wastewater from shale gas wells by permitted wastewater treatment plants did appear to influence stream Cl concentrations.

Arsenic concentrations in groundwater and base-flow surface waters in the Monongahela River Basin (WV) were examined in another area of rapidly expanding development of the Marcellus Formation by Chambers et al. (2015). Arsenic concentrations in groundwater samples ranged from less than 0.02 µg/L to 6.3 µg/L, whereas concentrations in

the base-flow samples ranged from 0.06 µg/L to 1.4 µg/L. Although Chambers et al. (2015) note that comparisons of data sets were complicated by the wide range of reporting levels in the historical data, they concluded that no significant difference was observed among the gas-producing regions for groundwater or base-flow samples and no sample from either the groundwater survey or the base-flow survey exceeded the 10 µg/L MCL for As. These studies provide useful baseline surface-water and groundwater As concentration data that can be used for comparisons in the event of releases of UOG products or wastes in these watersheds.

Oil and gas wastewater management practices, such as deep well injection (e.g., Akob et al., 2016; Kassotis et al., 2016; Orem et al., 2017), surface water discharge (e.g., Ferrar et al., 2013; Warner et al., 2013; Skalak et al., 2014), and accidental wastewater releases (Laurer et al., 2016; Cozzarelli et al., 2017) have been studied for their potential water-quality implications. In one of the earlier studies, Warner et al. (2013) analyzed the effluents discharged from a brine treatment facility receiving Marcellus wastewater, upstream and downstream waters, and sediment in order to assess environmental impacts of shale gas wastewater disposal on surface-water quality and stream sediments. Although they did not report As concentrations, the authors observed the discharge of wastewater effluent had a measurable impact on the water quality of the stream. In a study of a large wastewater spill from a ruptured pipeline in the Williston Basin, Cozzarelli et al. (2017) reported geochemical signatures and biological effects from the wastewater spill were present in the stream at least 6 months after the spill; however, As concentrations were not detected at concentrations above the background stream concentrations.

4.4. Impacts of reuse and recycling of oil and gas wastewater on As release and mobilization

Oil and gas (OG) wastewater, as well as some of the constituents in the wastewaters, can be put to beneficial use. Engle et al. (2014) outlined some of the potential beneficial uses of OG wastewater, including agricultural uses in crop production and livestock management. Reuse of OG wastewater for dust suppression and deicing was identified in 13 states by Tasker et al. (2018). In PA, Michigan (MI), Ohio (OH), and New York (NY), spreading high TDS produced waters on roads has occurred for decades to provide dust suppression on dirt roads during warmer months and deicing of paved roads during the winter (Skalak et al., 2014).

Reuse and recycling issues center on two broad categories in terms of potential human health effects relating to As. The first is the potential human exposure by ingesting drinking water contaminated with As and the second is from consumption of food in which As may have bioaccumulated. Potential health effects of As ingestion have been well-documented and include endocrine disruption (Balise et al., 2019) and developmental effects and risks of miscarriage as summarized in Webb et al. (2014). Developmental exposure to As has also been associated with altered metabolism (Davey et al., 2008; Rodriguez et al., 2016). In addition, As can impact the immune system and increases vulnerability to disease (Taheri et al., 2016). Elliott et al. (2017) named As a priority pollutant of concern in UOG wastewater, including it on a list of 67 chemicals based on known or suspected reproductive and developmental toxicity. However, studies on the specific role of As from oil and gas wastewaters reuse practices or releases are lacking.

Skalak et al. (2014) found accumulation of Ra, Sr, Ca, and Na in sediments adjacent to roads where the beneficial use of road-spreading of OG wastewaters was conducted for deicing purposes. They hypothesized that these sediments represent a potential non-point source of these elements into the environment. Little is known about the As content of wastewaters reused for road spreading, as most states require limited chemical analyses or pretreatment (McDevitt et al., 2020), often only oil-water separation. Among the states surveyed by McDevitt et al. (2020), only NY and North Dakota (ND) monitored lead (Pb) and As

concentrations.

In western states of the U.S., where water scarcity is prevalent, more focus has been on the potential use of OG wastewater on agricultural lands. Such uses require a thorough understanding of the complex interactions between wastewater constituents and soil composition. Irrigation with OG wastewater can have negative effects on soil health, crop yields, and microbial diversity (Miller et al., 2020). Kondash et al. (2020) conducted experiments to assess the potential environmental effects of using oilfield produced water (OPW) blended with surface water for irrigation in California. The authors noted that salt and boron (B) accumulated in the irrigated soils and have the potential to pose long-term risks to soil sodification, groundwater salinization, and plant health. McLaughlin et al. (2016) underscored the importance of considering mixture interactions between organic components often added during hydraulic fracturing and inorganic OG wastewater constituents. They studied the chemical and biological transformation mechanisms for 3 common hydraulic fracturing products and showed that co-contaminant fate and toxicity involved complex interactions that need to be considered when predicting potential environmental impacts.

The leaching of As and metals from the solids into the fluid phase can be enhanced at the high ionic strengths of UOG wastewaters, which has implications for the reuse of OG wastewaters for irrigation (see review by Lazar et al. (2020)). Scanlon et al. (2009) found that an influx of saline water caused mobilization of As from sediments due to a counterion effect, where the competition for sorption sites may enhance As mobilization. This process could also mobilize As from sediments after the accidental release of saline wastewaters. In another study, Chen et al. (2017) conducted experimental studies that included evaluation of As (V) mobility in soils subjected to artificial OG wastewater and found that the composition of the wastewater (largely the salinity), rather than the soils composition, controlled the mobility and bioavailability of As, underscoring the need to fully understand chemical constituents in waters intended for reuse.

Societal interest in OG wastewater reuse to solve water scarcity and disposal limitations is driving a lot of interest in treatment technologies in order to address concerns about potential environmental and human health effects. Simões et al. (2020) conducted a bibliometric analysis using the Scopus database and found 2434 documents on “produced water” treatment, of which 1305 were scientific articles, between 1969 and 2017; approximately 60% of the papers were published since 2010. The treatment technologies discussed included physical, chemical, membrane, biological, and combined treatments. Gregory et al. (2011) review some of the specific treatment approaches and other water management challenges when dealing with these high salinity waters.

5. Conclusions and opportunities for future research

The release of As, a known human toxin and carcinogen, to the environment is of critical concern for human health. Through an extensive literature review, we outline the occurrence of As in hydrocarbons and the key processes driving As release from hydrocarbon production, storage, transportation, use and waste management. Due to uptake of As in marine biota from seawater, As is incorporated into hydrocarbons in both organic and inorganic forms. Some hydrocarbon sources, such as oil shales, have much higher As concentrations than others. When hydrocarbons are extracted and processed, stored and transported, and utilized via combustion, As can be released to air, land and water. In addition, the extensive volumes of water produced during oil and gas production can contain As. Recycling and reuse of this produced water are of both scientific and societal interest, but treatment is challenged by the high saline content of these waters.

Although there has been substantial progress in building scientific knowledge of As release from hydrocarbons and subsequent mobility in the environment, there is much yet to be learned. First, there remain challenges for analysis of As in hydrocarbons and their waste products, resulting in wide ranges of concentrations reported, with little

knowledge of controls on those concentrations. To better understand the role of hydrocarbon-related As in the global As cycle and in potential impacts on water and soil quality and ecosystem and human health, continued improvements in digestion/extraction and analytical methods are needed for accurate detection of low As concentrations in organic phases such as oil and gas and in high-saline waters. A recent study by Tasker et al. (2019) underscores this analytical challenge in OG wastewaters. Tasker et al. (2019) conducted an inter-laboratory comparison of results obtained by 15 different laboratories analyzing the same 3 OG wastewater samples. Less than 7 of the 15 labs were able to report detectable concentrations of As, Ni, Cu, Zn, Cr and Pb with accuracies of approximately $\pm 40\%$. Although the labs used analytical instrumentations with low instrument detection capabilities for trace metal analyses, the large dilution factors needed to accommodate the high salt content combined with low trace metal concentrations in the wastewaters limited the analytical accuracy. In addition, the analysis of volatile As phases requires sophisticated instrumentation that are likely not available at typical environmental monitoring laboratories.

Second, outside of research studies, measurements of As in oil, gas and energy-associated waste are not routinely made. In the U.S., although there are regulatory standards for allowable As concentrations in water, food, soils and air (for occupational exposure), there are not similar regulatory limits for As in hydrocarbons. However, due to the poisoning of catalysts by As in the refining process, refineries have installed treatment technologies to remove As before reacting with these catalysts; thus, there is incentive for refineries to remove As. Without these measurements of As concentrations, calculations of As release from hydrocarbon extraction, processing, use and waste disposal can only be estimated.

Third, research on the overall toxicology and health effects of hydrocarbon production could aid in identifying both the broad effects of hydrocarbon production on humans and the effects of specific activities and contaminants. Recently, Cushing et al. (2020) examined the impact of natural gas flaring on birth outcomes; results suggested that exposure to flare events was associated with 50% higher odds of preterm birth. In a review of the potential health effects of UOG development, the Health Effects Institute (HEI, 2020) emphasized that more studies are needed with the objective of quantifying potential human exposure to water contamination from UOG processes. They noted that the inability to definitively attribute measured contaminant concentrations in groundwater and surface water to UOG sources, as compared to other potential sources, has led to controversy about impact assessment. This issue applies to assessing whether any potential effects from As in these waters can be attributed to hydrocarbon sources or wastes.

Fourth, the relative importance of the As content of hydrocarbon products and wastes to potential toxicological effects on exposed organisms is largely unknown. There have been some studies showing some correlations of effects, but not causation. For example, Debenest et al. (2012) studied the ecotoxicological effects of wastewater effluents generated during oil sands bitumen extraction on a microalgae, *Pseudokirchneriella subcapitata*. They found the high toxicity of oil sand processing waters could be related to metals and metalloids detected in the wastewater as indicated by high Pearson correlation values between cell growth rate and the concentrations of As, Ni, Cr, Cu, Pb, As, aluminum and beryllium. It is likely that the toxicological effect is due to a mixture of contaminants, however, the relative effect of individual chemical components of the wastewater on microalgae growth could not be teased out.

Last, research contributing to treatment methods that prevent release of As from hydrocarbon extraction, production, use and waste and those that remove, immobilize or attenuate As in air, water, soil and sediment, could minimize exposure of humans and ecosystems to the deleterious effects of As.

CRediT authorship contribution statement

Madeline Schreiber: Conceptualization, Visualization, Writing - original draft (Sections 1–3), Writing - review & editing. **Isabelle Cozzarelli:** Conceptualization, Visualization, Writing - original draft (Sections 4 and 5), Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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