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Assessment of the Acute and Chronic Health Hazards of Hydraulic Fracturing Fluids

Elizabeth V. Wattenberg,[1] Jeffrey M. Bielicki,[2,3] Ashley E. Suchomel,[1] Jessica T. Sweet,[1] Elizabeth M. Vold,[1] and Gurumurthy Ramachandran[1]

[1]Division of Environmental Health Sciences, University of Minnesota, Minneapolis, Minnesota
[2]Department of Civil, Environmental, and Geodetic Engineering, The Ohio State University, Columbus, Ohio
[3]The John Glenn School of Public Affairs, The Ohio State University, Columbus, Ohio

There is growing concern about how hydraulic fracturing affects public health because this activity involves handling large volumes of fluids that contain toxic and carcinogenic constituents, which are injected under high pressure through wells into the subsurface to release oil and gas from tight shale formations. The constituents of hydraulic fracturing fluids (HFFs) present occupational health risks because workers may be directly exposed to them, and general public health risks because of potential air and water contamination. Hazard identification, which focuses on the types of toxicity that substances may cause, is an important step in the complex health risk assessment of hydraulic fracturing. This article presents a practical and adaptable tool for the hazard identification of HFF constituents, and its use in the analysis of HFF constituents reported to be used in 2,850 wells in North Dakota between December 2009 and November 2013. Of the 569 reported constituents, 347 could be identified by a Chemical Abstract Service Registration Number (CASRN) and matching constituent name. The remainder could not be identified either because of trade secret labeling (210) or because of an invalid CASRN (12). Eleven public databases were searched for health hazard information on thirteen health hazard endpoints for 168 identifiable constituents that had at least 25 reports of use. Health hazard counts were generated for chronic and acute endpoints, including those associated with oral, inhalation, ocular, and dermal exposure. Eleven of the constituents listed in the top 30 by total health hazard count were also listed in the top 30 by reports of use. This includes naphthalene, which along with benzyl chloride, has the highest health hazard count. The top 25 constituents reportedly used in North Dakota largely overlap with those reported for Texas and Pennsylvania, despite different geologic formations, target resources (oil vs. gas), and disclosure requirements. Altogether, this database provides a public health tool to help inform stakeholders about potential health hazards, and to aid in the reformulation of less hazardous HFFs.

Keywords Bakken Shale Play, health hazards, hydraulic fracturing, hydraulic fracturing fluid, toxicity

Address correspondence to Elizabeth V. Wattenberg, Division of Environmental Health Sciences, University of Minnesota, MMC #807, 420 Delaware Street SE, Minneapolis, MN 55455; e-mail: watte004@umn.edu

INTRODUCTION

Since 2005, there has been a major increase in the use of hydraulic fracturing to produce natural gas and oil in the United States.[1] Hydraulic fracturing involves injecting large volumes of highly pressurized fluid, which contains a variety of physical and chemical constituents, through wells that extend thousands of feet below the surface of the earth, resulting in the release of gas and oil from newly created or reopened fractures in tight shale formations.[2–4] The production of natural gas from hydraulic fracturing in the United States increased 13-fold between 2005 and 2012, and by 2013 accounted for over 30% of gas production in the United States.[5] As a result of a similar increase in the use of hydraulic fracturing to produce oil, during 2013 the United States domestic oil production levels exceeded imports for the first time since 1995.[6,7] Major sites of hydraulic fracturing occur in various regions of the United States, including the Barnett shale play in Texas and the Marcellus shale play in the Northeast, where the primary resource is natural gas, and the Bakken shale play in North Dakota and Montana, where the primary resource is oil.[1] Hydraulic fracturing can use millions of gallons of fluid that contains toxic and carcinogenic constituents; furthermore, the constituents are often transported through areas where existing roads and other infrastructure are strained by the large increase in heavy truck traffic and vehicle emissions.[3,8,9,10–12] Altogether, this raises concerns about how hydraulic fracturing activity affects public health.

Hydraulic fracturing fluids (HFFs) contain several constituents, each of which serves a specific purpose. The exact composition of the HFF varies from well to well, depending upon the geologic conditions, whether gas or oil is produced, and the company operating the site.[1,2] Proppants such as crystalline silica are used to keep the fractures in the shale formations open in order to maintain the flow of gas and oil.[2,4] Other constituents are added to the fluid to suspend proppants during transport, increase fluid viscosity, reduce surface tension to allow the fluid and gas to flow more freely, and to maximize the fluid travel distance to extend the fracture length of the well.[1] Constituents are also added to maintain the wells by preventing corrosion, the buildup of minerals, and the growth of bacteria.[1,3,10] Altogether, the constituents typically comprise up to 2% of the HFF volume.[4]

HFFs pose both immediate (acute) and longer-term (chronic) health risks to workers who may be directly exposed to them, and present longer-term health risks to the general public who may be exposed to toxic constituents through air and water contamination, spills, and improper disposal of waste.[1,2,4,13] Hazard Identification, which focuses on the types of toxicity substances may cause, is an important component of the complex health risk assessment of HFFs, which ultimately requires analysis of exposures, fate, and transport of constituents in the environment, and determining the risks of toxicity associated with the levels of constituents detected in air and water.[11] While the health hazards of some of the HFF constituents are well characterized, such as crystalline silica, a commonly used proppant that causes silicosis and is a known human carcinogen,[14,15] the health hazards of many other constituents, in particular those that are used to produce oil, are not yet well characterized.

Data on HFFs used in North Dakota were used to develop a practical, transparent, and adaptable database tool for the hazard identification of a wide range of HFF constituents, which can be used to inform workers, community residents, and other stakeholders. North Dakota data were used because this is one of the few states that require disclosure of hydraulic fracturing activities and HFF constituents.[16] Furthermore, while other studies have focused on health hazards from hydraulic fracturing to produce natural gas,[9,11,13,17] studies investigating activities that primarily produce oil, as is the case in North Dakota, are lacking. This database tool can be used to help identify the constituents of highest concern, and thus to help set priorities for assessing and managing the health risks associated with HFFs.

METHODS

Identification of HFF Constituents

Information was collected from the FracFocus Chemical Disclosure Registry on hydraulic fracturing activity in North Dakota between December 2009 and November 2013.[18] FracFocus is a national hydraulic fracturing chemical registry managed by the Interstate Oil and Gas Compact Commission and the Groundwater Protection Council.[18] The FracFocus database contains one report for each hydraulic fracturing job. A complete report includes: hydraulic fracturing date, county of operation, operator name, well name and number, latitude and longitude, true vertical depth, total water volume used, and HFF constituents; the constituent data include the product trade name, supplier, purpose of the product, ingredients, Chemical Abstract Service Registration Number (CASRN), maximum ingredient concentration in the additive, and the maximum ingredient concentration in the HFF. Data on individual hydraulic fracturing jobs were imported from SkyTruth, an online database that converts the FracFocus files into a standardized format that is usable by Microsoft Excel.[19]

According to the FracFocus reports, 43 companies (listed as "operators") conducted 3,448 distinct hydraulic fracturing jobs in 2,850 distinct wells in North Dakota during this period. Hydraulic fracturing was reported for one well in 2009, 6 wells in 2010, 643 wells in 2011, 1705 wells in 2012, and 512 wells by November 2013.

Data were compiled on the purpose and reports of use for each identified constituent (see the Health Hazard Database in Online Supplemental Material). If more than one purpose was listed, the most commonly reported purpose(s) was used (typically at least 5% of the reports). If no purpose was listed, the CASRN was used to search for a purpose associated with the constituent as listed by FracFocus or the United States Environmental Protection Agency.[3,18] If a purpose associated with the CASRN was found, it was entered into the database. Not available (represented as "NA") was entered when no purpose could be identified.

Review of HFF Constituent Health Hazards

The match between the reported CASRN and the reported constituent name was checked by searching CHEMIDplus, a database of chemical compounds maintained by the National Library of Medicine. If the constituent name did not match the CASRN, the CASRN was used to identify the matching name through CHEMIDplus. Of the 569 reported constituents, 359 listed a CASRN; 347 of these could be identified by a valid CASRN and matching constituent name. Out of these 347, 168 constituents had at least 25 reports of use, nine had 20–23 reports of use, 23 had 15–19 reports of use, 16 had 10–13 reports of use, and 32 had 5–9 reports of use. The remainder had four or less reports of use.

The human health hazards of constituents that had at least 25 reports of use were evaluated based on 13 endpoints that reflect likely routes of exposure for workers (inhalation, ocular, dermal) and the general public (inhalation and oral), likely durations of exposure of concern to workers (acute and chronic) and the general public (chronic), and major toxic effects of concern (see Health Hazard Database in Online Supplemental Material). The 13 health hazard endpoints are presented in Table I.

The health hazards for each constituent were identified by searching eleven publicly available databases and registries maintained by federal and state agencies and international organizations: Agency for Toxic Substances and Disease Registry, European Chemicals Agency, European chemical Substances Information System, Hazardous Substance

TABLE I. Thirteen Health Hazard Endpoints used for Evaluating Constituents of HFF Based on Route of Exposure and Acute or Chronic Effects

Route of exposure	Acute	Chronic
Oral	–	carcinogenicity
Inhalation	–	carcinogenicity
Oral	–	neurotoxicity
Inhalation	neurotoxicity	neurotoxicity
Oral	–	reproductive/developmental toxicity
Inhalation	–	reproductive/developmental toxicity
Oral	–	other
Inhalation	other	other
Inhalation	respiratory tract irritation	–
Ocular	eye irritation or damage	–
Dermal	skin irritation or damage	–

Notes: "Oral chronic other" and "inhalation chronic other" include health hazard data that did not fall into the other oral and inhalation chronic categories listed, respectively. "Inhalation acute other" includes health hazard data that did not fall into one of the other inhalation acute categories listed. The criteria used to determine that a constituent is a potential carcinogen or that there was no evidence of carcinogenicity after study completion are explained in the Key to the Health Hazard Database in the online supplemental material.

Database, Integrated Risk Information System (IRIS), International Agency for Research on Cancer Monographs on the Evaluation of Carcinogenic Risks to Humans, International Programme on Chemical Safety, International Uniform Chemical Information Database, National Toxicology Program, Proposition 65 list, and Toxicology Data Network.

All 11 of the databases were searched for information on each constituent. Data were used if the amount of detail reported was sufficient to evaluate the quality of the study (e.g., animal studies reported species, route of exposure, doses, number of study subjects, appropriate controls) and the data came from a reputable source (e.g., a report from a reputable work group such as the IRIS, peer-reviewed literature). If studies of equally high-quality reported conflicting results regarding the potential to cause toxicity, the assumption was made that the positive results indicated a potential hazard. This is a conservative assumption that protects health. Health hazard information from human studies came mainly from epidemiology studies. Case studies were only used if several case studies reported the same health effects and the health effects noted were consistent with what is known about the health hazards of the chemical from other studies, such as animal studies.

The database (included in Online Supplemental Material) also lists the following regulatory or guidance information when available: threshold limit value-time weighted average (TLV-TWA), TLV-short-term exposure limit (TLV-STEL), TLV-Ceiling, vapor hazard ratio (VHR) (TLV-TWA), VHR (TLV-STEL), VHR (TLV-Ceiling), recommended exposure limit (REL) TWA, REL STEL, REL Ceiling, permissible exposure limit (PEL) TWA, PEL STEL, PEL Ceiling, maximum contaminant level goal (MCLG), maximum contaminant level (MCL), oral slope factor, reference dose (RfD), reference concentration (RfC), health-based drinking water guidance (HBDG), and HDBG based on a cancer endpoint (cHBDG) (Health Hazard Database in Online Supplemental Material). The HBDG and cHBDG were calculated based on a method used by the Minnesota Department of Health using RfDs published on IRIS (see key to Health Hazard Database in Online Supplemental Material).

Health Hazard Counts

To calculate the health hazard counts, one point was assigned for each of the following conditions indicated in the database: (+) positive in a toxicity test based on animal studies; (+H) positive based on data from human studies. No points were assigned under the following condition: (−) no evidence of toxicity after study completion. An entry of (SDS) indicates that a health hazard was reported in a Safety Data Sheet, but could not be confirmed through searching the 11 databases. A blank entry indicates that no information was found on the endpoint. These data gaps indicate uncertainty in the potential hazards and thus a count for uncertainty was calculated by assigning one point to each (SDS) and blank entry. Counts for the following categories were generated (the maximum possible count in each category is shown in parenthesis): total health hazard count (13); chronic oral endpoints (4); chronic inhalation endpoints (4); total chronic endpoints (8); total acute endpoints (5). Counts were also generated for data gaps to give an indication of uncertainty: total unknown chronic (8); total unknown acute (5). As an example, Table II shows how the health hazard count for naphthalene was determined.

RESULTS

HFF Constituents

Health hazard information was located for 113 (67%) of the 168 constituents that had at least 25 reports of use (see Health Hazard Database in Online Supplemental Material). The most common health hazard endpoints found were those associated with acute toxicity, which is particularly relevant for occupational exposures (Figure 1). The database search indicated that 110 (65%) of the constituents could potentially cause some type of acute toxicity (Figure 1A, white pie slice), although data were not found for any of the acute endpoints for 58 (35%) of the constituents (Figure 1A, hatched pie slice). Approximately 50% of the constituents could potentially cause respiratory tract irritation (80/168), eye irritation or damage (92/168), or skin irritation or damage (90/168) (Figure 1B,

TABLE II. Health Hazard Count for Naphthalene

Acute Endpoint	Count	
Inhalation neurotoxicity	+	
Inhalation other	data gap	
Respiratory tract irritation	+	
Eye irritation or damage	+	
Skin irritation or damage	+	
Total Acute	**4**	
Chronic Endpoint	Oral Count	Inhalation Count
Carcinogenicity	data gap	+
Neurotoxicity	data gap	+
Reproductive/ Developmental	+	-
Other	+	+
Total Chronic	**2**	**3**
Total Unknown Count (Acute + Chronic)		3
Total Health Hazard Count (Acute + Chronic)		**9**

white pie slices). For each of these endpoints data were not available for over 40% of the constituents (Figure 1B, hatched pie slices). Data were available to indicate that acute inhalation exposure to 15 (9%) of the constituents could potentially cause neurotoxicity (Figure 1B, white pie slice), while 4 (2%) of the constituents tested negative for acute inhalation neurotoxicity (Figure 1B, black pie slice). Data on neurotoxic effects from acute inhalation exposure were not available for 149 (89%) of the constituents (Figure 1B, hatched pie slice).

Far less information was found for chronic toxicity than acute toxicity. Although available data indicate that 49 (29%) of the constituents can potentially cause some type of chronic toxicity (Figure 2A, white pie slice), data were not available for any of the chronic endpoints for 99 (59%) of the constituents (Figure 2A, hatched pie slice).

Chronic oral toxicity is relevant for community members who may be concerned about long-term exposure to contaminated drinking water. The database search identified 6 (4%) constituents that tested positive for carcinogenicity, and 5 (3%) constituents that tested negative, but data on carcinogenicity were not available for 157 (93%) constituents (Figure 2B, white, black, and hatched pie slices, respectively). More information that was available for reproductive and developmental toxicity; 24 (14%) constituents tested positive, 25 (15%) tested negative, and no data were available for 119 (71%) constituents

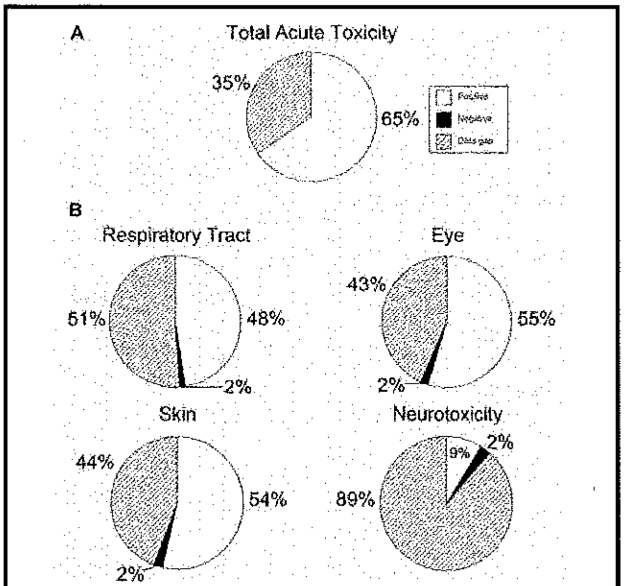

FIGURE 1. Percentages of HFF constituents associated with (A) any of the acute toxicity endpoints and (B) the indicated type of acute toxicity. Data are available to indicate the constituents can potentially cause toxicity (white slice, Positive); no evidence of acute toxicity was found after study completion (black slice, Negative); no data on acute toxicity was found (hatched slice, Data gap).

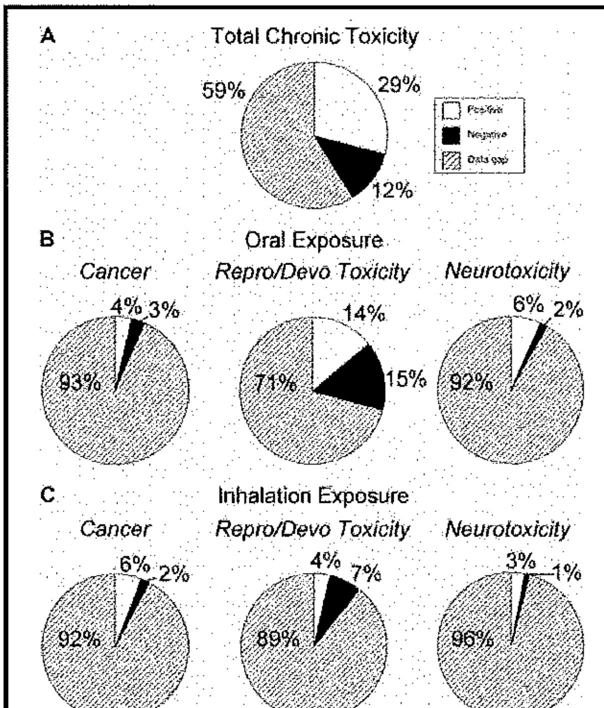

FIGURE 2. Percentages of HFF constituents associated with (A) any of the chronic toxicity endpoints, (B) the indicated type of chronic oral toxicity, and (C) the indicated type chronic inhalation toxicity. Data are available to indicate the constituents can potentially cause toxicity (white slice, Positive); no evidence of chronic toxicity was found after study completion (black slice, Negative); no data on chronic toxicity was found (hatched slice, Data gap).

(Figure 2B, white, black, and hatched pie slices, respectively). The amount of information that was available for neurotoxicity was similar to that of carcinogenicity. Whereas 11 (6%) constituents tested positive and 3 (2%) tested negative, no data were found for 154 (92%) constituents (Figure 2B, white, black, and hatched pie slices, respectively).

Chronic inhalation exposure is relevant for workers, who may be exposed over several years of working in the industry, and for the general public who are concerned about multiple pathways of exposure. Health effects due to chronic inhalation exposure presented a major data gap. For example, 10 (6%) constituents tested positive for carcinogenicity, 3 (2%) tested negative, and data were not available for 155 (92%) constituents (Figure 2C, white, black, and hatched pie slices, respectively). Likewise, data on reproductive and developmental toxicity were not available for 150 (89%) constituents, although 6 (4%) constituents tested positive, and 12 (7%) tested negative (Figure 2C, hatched, white, and black pie slices, respectively). Data on neurotoxicity were not available for 161 (96%) constituents (Figure 2C, hatched pie slice). Five (3%) constituents tested positive, however, and 2 (1%) tested negative for neurotoxicity (Figure 2C, white and black pie slices, respectively).

Health Hazard Ranking

The constituents that are used most frequently and have the highest health hazard counts (the more types of toxicity a constituent can potentially cause, the higher the health hazard count) are likely to be of most concern for public health. Table III shows the top 30 constituents ranked by the number of times they were reportedly used. A purpose was identified for all of these constituents except for sorbitan, mono-9-octadecenoate, (Z)-. Eleven of the constituents ranked in the top 30 by reports of use are also listed among the top 30 constituents ranked by total health hazard count (see Table IV, bold, italic rows): naphthalene, sodium chlorite, methanol, isopropyl alcohol, 1,2,4 trimethylbenzene, ethylene glycol, acetic acid, sodium hydroxide, ammonium chloride, crystalline silica (quartz), and crystalline silica (cristobalite).

Health hazard data were located for 27 of the constituents ranked in the top 30 by reports of use (Table III). These data indicate that all of 27 of these constituents can cause some type of acute toxicity, while almost half of them (13) can potentially cause some type of chronic toxicity. Crystalline silica, a commonly used proppant, had the highest reports of use (3,223). Importantly, the total health hazard count for crystalline silica (4) is based in part on human data, which indicate that inhalation exposure can cause cancer and silicosis. Workers are most likely to be exposed to crystalline silica dust while it is being offloaded from trucks at the well pad.[15] Crystalline silica and other proppants remain within the rock formations after injection of the HFF, and are therefore unlikely to be present in the produced or flowback water.[18] Naphthalene had the highest total health hazard count (9) among the 30 constituents (Table III) and is ranked seventh by reports of use. It is used as a surfactant and may be a component of produced and flowback water.[18] The available data indicate that naphthalene can potentially cause both acute and chronic toxicity (carcinogenicity, neurotoxicity, reproductive/developmental toxicity, and other effects through inhalation or oral exposure). Information for the 13 health hazard endpoints was not located for potassium metaborate, potassium formate, or alcohols, C12-16, ethoxylated, ranked 16, 19, and 30 by reports of use, respectively. Potassium metaborate and potassium formate are crosslinkers, which combine with other constituents to form salts that are components of produced water.[18] Alcohols, C12-16, ethoxylated is a friction reducer, which is typically degraded in the rock formation by micro-organisms, such that only a small amount is present in the produced water.[18] The total health hazard counts each of these three constituents is zero, and the total unknown chronic and total unknown acute counts are the maximum possible (eight and five, respectively), indicating a high level of uncertainty about the health hazards of potassium metaborate, potassium formate, and alcohols, C12-16, ethoxylated.

Some type of health hazard guidance or regulatory value was available for 15 of the constituents ranked in the top 30 by reports of use, including all 11 of the constituents that are also ranked in the top 30 by health hazard count (Tables III and IV). In addition, data were available to calculate the vapor hazard ratio (ratio of the vapor pressure in mm Hg to the occupational exposure limit in ppm) for five of the constituents. The vapor hazard ratios were all below 1. A rough rule of thumb in industrial hygiene practice is that if this ratio is less than 1, then good general ventilation, which is available at outdoor locations, is sufficient for adequate control of exposure. The exception is acetic acid, which has a vapor hazard ratio of 1.6 based on the TLV-TWA (adequate control requires good general ventilation with capture at emissions points).

Table IV lists the constituents that are listed in the top 30 by total health hazard count. In cases where constituents had the same total health hazard count, the order was determined based on the amount of available human data, and then on the uncertainty count. For example, benzyl chloride and naphthalene each have total health hazard counts of nine. Benzyl chloride is listed first because the uncertainty count is four, whereas the uncertainty count for naphthalene is three. Acrylamide, formaldehyde, and sodium chlorite each have hazard counts of eight. Acrylamide has human data to support two health hazard endpoints and is thus listed third. Formaldehyde has human data to support one health hazard endpoint and is thus listed fourth. The health hazard score for sodium chlorite is all based on data from animal studies and is thus listed fifth. The available data indicated that all 30 constituents can potentially cause some type of both chronic and acute toxicity. Twenty-six constituents were positive for at least two chronic endpoints, 9 had data to indicate they are potentially carcinogenic, 13 had data to indicate they are neurotoxic, and 17 had data to indicate they can potentially cause reproductive/developmental toxicity. Furthermore, approximately 80% were positive for at least three acute health hazard endpoints.

TABLE III. Top 30 Constituents of HFFs Ranked by Reports of Use

Rank	Name (CASRN)	Purpose	Reports of use	Total health hazard count	Guidance value
1	Crystalline silica (quartz) (14808-60-7)	Proppant	3223	4	TLV-TWA, REL TWA, PEL TWA
2	Potassium hydroxide (1310-58-3)	Crosslinker/pH Buffer	2341	3	TLV-Ceiling, REL Ceiling
3	Guar gum/ (polysaccharide blend) (9000-30-0)	Gelling agent	2212	1	NA
4	Methanol (67-56-1)	Corrosion inhibitor/Scale inhibitor/Surfactant/Non-emulsifier	2153	7	TLV-TWA, TLV-STEL, VHR (TLV-TWA, TLV-STEL), REL TWA, REL STEL, PEL TWA, RfD, RfC, HBDG
5	Solvent naphtha, (petroleum), heavy aromatic (64742-94-5)	Non-emulsifier	2081	3	NA
6	Hydrotreated light petroleum distillate (64742-47-8)	Friction reducer/Gelling agent/Crosslinker	2076	2	NA
7	Naphthalene (91-20-3)	Surfactant	2008	9	TLV-TWA, TLV-STEL, VHR (TLV-TWA, TLV-STEL), REL TWA, REL STEL, PEL TWA, RfD, RfC, HBDG
8	Sodium hydroxide (1310-73-2)	pH Buffer/Biocide	2001	5	TLV-Ceiling, REL Ceiling, PEL TWA
9	Sodium chloride (7647-14-5)	Breaker	1816	3	NA
10	Ammonium peroxydisulfate (7727-54-0)	Breaker	1694	3	NA
11	Ethylene glycol (107-21-1)	Crosslinker/Scale inhibitor/Friction reducer	1572	6	RfD, HBDG
12	Ethanol (64-17-5)	Biocide/surfactant	1540	4	TLV-STEL, VHR (TLV-STEL), REL TWA, PEL TWA
13	1,2,4 Trimethyl-benzene (95-63-6)	Surfactant	1423	6	REL TWA, RfC
14	Poly(oxy-1,2-ethanediyl),alpha-(4-nonylphenyl)-omega-hydroxy-, branched (127087-87-0)	Surfactant	1403	2	NA
15	Isopropyl alcohol (67-63-0)	Non-emulsifier/Corrosion inhibitor/Surfactant	1394	6	TLV-TWA, TLV-STEL, VHR (TLV-TWA, TLV-STEL), REL TWA, REL STEL, PEL TWA

(Continued on next page)

TABLE III. Top 30 Constituents of HFFs Ranked by Reports of Use *(Continued)*

Rank	Name (CASRN)	Purpose	Reports of use	Total health hazard count	Guidance value
16	Potassium metaborate (13709-94-9)	Crosslinker	1343	0	NA
17	Sodium chlorite (7758-19-2)	Breaker	1330	8	MCLG, MCL, RfD, HBDG
18	Amorphous silica (7631-86-9)	Proppant	1286	2	REL TWA, PEL TWA
19	Potassium formate (590-29-4)	Crosslinker	956	0	NA
20	Glutaraldehyde (111-30-8)	Biocide	937	4	TLV-Ceiling
21	Sodium perborate tetrahydrate (10486-00-7)	Surfactant/Breaker	918	2	NA
22	2-Ethylhexanol (104-76-7)	Surfactant/Non-emulsifier	867	4	NA
23	Mullite (1302-93-8)	Proppant	865	3	NA
24	Sorbitan, mono-9-octadecenoate, (Z)-(1338-43-8)	NA	857	1	NA
25	Ammonium chloride (12125-02-9)	Crosslinker/Scale inhibitor	844	5	TLV-TWA, TLV-STEL, REL TWA, REL STEL
26	Potassium carbonate (584-08-7)	Buffer	833	3	NA
27	Polyethylene glycol (25322-68-3)	Biocide	814	2	NA
28	Crystalline silica (cristobalite) (14464-46-1)	Proppant	766	4	TLV-TWA
29	Acetic acid (64-19-7)	pH Buffer	756	5	TLV-TWA, TLV-STEL, VHR (TLV-TWA, TLV-STEL), REL TWA, REL STEL, PEL TWA
30	Alcohols, C12-16, ethoxylated (68551-12-2)	Friction reducer	746	0	NA

Notes: The highest rank is 1 and the lowest rank is 30. NA indicates not available.

Table V compares the top 25 constituents by reports of use for North Dakota, Texas, and Pennsylvania as reported to FracFocus between December 2009 and November 2013. According to the reports from this period, 22,678 distinct hydraulic fracturing jobs were conducted in 20,913 distinct wells in Texas, and 5,101 distinct hydraulic fracturing jobs were conducted in 2,986 distinct wells in Pennsylvania. Although natural gas is the primary resource recovered by hydraulic fracturing in Texas and Pennsylvania, several of the frequently reported constituents are the same as those frequently reported for North Dakota. North Dakota and Texas have 16 out of 25 constituents in common, while North Dakota and Pennsylvania have eleven out of 25 in common (bold, italic entries). This indicates that many constituents are commonly used in hydraulic fracturing for both the recovery of natural gas and oil. Furthermore, 10 of the constituents ranked in the top 25 by reports of use in Texas and eight of those ranked in the top 25 by reports of use for Pennsylvania are also among the constituents listed in the top 30 by total health hazard count for North Dakota. This consistency highlights

TABLE IV. Top 30 Constituents of HFFs Ranked by Total Health Hazard Count

Rank	Name (CASRN)	Purpose	Rank by reports of use	Health hazard count	Chronic	Acute	Unknown chronic	Unknown acute
1	Benzyl chloride (100-44-7)	NA	157	9	4	5	4	0
2	***Naphthalene (91-20-3)***	***Surfactant***	***7***	***9***	***5***	***4***	***2***	***1***
3	Acrylamide (79-06-1)	Friction reducer	164	8	5	3	3	1
4	Formaldehyde (50-00-0)	Biocide	81	8	3	5	4	0
5	***Sodium chlorite (7758-19-2)***	***Breaker***	***17***	***8***	***3***	***5***	***5***	***0***
6	Dimethylformamide (68-12-2)	Corrosion inhibitor	166	7	3	4	4	1
7	***Methanol (67-56-1)***	***Corrosion inhibitor/Scale inhibitor/Surfactant/Non-emulsifier***	***4***	***7***	***4***	***3***	***4***	***2***
8	Butyl alcohol (71-36-3)	Surfactant	79	7	3	4	4	1
9	Acetone (67-64-1)	Anti-bacterial	116	7	3	4	4	0
10	***Isopropyl alcohol (67-63-0)***	***Non-emulsifier/Corrosion inhibitor/Surfactant***	***15***	***6***	***2***	***4***	***6***	***1***
10	Sodium bromate (7789-38-0)	Breaker	47	6	3	3	5	2
11	***1,2,4 Trimethylbenzene (95-63-6)***	***Surfactant***	***13***	***6***	***2***	***4***	***5***	***1***
11	Propargyl alcohol (107-19-7)	Corrosion inhibitor	147	6	2	4	5	1
11	Dichloroethyl ether (DCEE) (111-44-4)	Surfactant	168	6	2	4	6	0
12	***Ethylene glycol (107-21-1)***	***Crosslinker scale inhibitor/Friction reducer***	***11***	***6***	***4***	***2***	***4***	***1***
13	Magnesium silicate hydrate (talc) (14807-96-6)	NA	74	5	3	2	5	3
14	***Acetic acid (64-19-7)***	***pH Buffer***	***29***	***5***	***1***	***4***	***6***	***1***

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TABLE IV. Top 30 Constituents of HFFs Ranked by Total Health Hazard Count *(Continued)*

Rank	Name (CASRN)	Purpose	Rank by reports of use	Health hazard count	Chronic	Acute	Unknown chronic	Unknown acute
15	***Sodium hydroxide (1310-73-2)***	***pH Buffer/ Biocide***	***8***	***5***	***2***	***3***	***6***	***2***
15	***Ammonium chloride (12125-02-9)***	***Crosslinker/Scale inhibitor***	***25***	***5***	***2***	***3***	***6***	***2***
15	n-Propanol (71-23-8)	Crosslinker	54	5	1	4	7	1
15	Sodium bromide (7647-15-6)	Biocide	131	5	3	2	5	3
15	2,2',2''-Nitrilotriethanol (102-71-6)	Crosslinker/Breaker	136	5	2	3	6	2
15	Zirconium dichloride oxide (7699-43-6)	NA	154	5	2	3	6	2
16	Tetrakis (hydroxymethyl) phosphonium sulfate (55566-30-8)	Biocide	39	5	1	4	6	1
17	Boric acid (10043-35-3)	Crosslinker	49	5	2	3	5	1
17	Quaternary ammonium compounds, benzyl-C12-16-alkyldimethyl, chlorides (68424-85-1)	Biocide	142	5	2	3	4	2
18	Ethylene glycol mono-n-butyl ether (111-76-2)	Surfactant/Non-emulsifier	50	5	1	4	4	1
19	Isobutyl alcohol (78-83-1)	Non-emulsifier	125	5	2	3	3	0
20	***Crystalline silica (quartz) (14808-60-7)***	***Proppant***	***1***	***4***	***2***	***2***	***6***	***3***
20	***Crystalline silica (cristobalite) (14464-46-1)***	***Proppant***	***28***	***4***	***2***	***2***	***6***	***3***

Notes: The highest rank is 1 and the lowest rank is 19. If constituents had the same total health hazard count, constituents with the higher number of endpoints based on human data were given a higher rank. In cases were the rank remained the same, regardless of human data, constituents with higher uncertainty counts were given the higher rank. *Chronic* indicates total chronic endpoint count, *Acute* indicates total acute endpoint count; *Unknown chronic* indicates total unknown chronic endpoint count, *Unknown acute* indicates total unknown acute endpoint count. "NA" indicates that the information for indicated purpose was not identified. Bold, italics indicates that the constituents are also ranked in the top 30 by reports of use (see Table III).

TABLE V. Top 25 Constituents Ranked by Reports of Use for North Dakota, Texas, and Pennsylvania

	North Dakota		Texas		Pennsylvania	
Rank	Name (CASRN)	Total Health Hazard Count	Name (CASRN)	Total Health Hazard Count	Name (CASRN)	Total Health Hazard Count
1	Crystalline silica (quartz) (14808-60-7)	4	***Crystalline silica (quartz) (14808-60-7)***	***4***	Hydrochloric acid (7647-01-0)	4
2	Potassium hydroxide (1310-58-3)	3	***Methanol (67-56-1)***	***7***	***Methanol (67-56-1)***	***7***
3	Guar Gum/(polysaccharide blend) (9000-30-0)	1	Hydrochloric acid (7647-01-0)	4	***Hydrotreated light petroleum distillate (64742-47-8)***	***2***
4	Methanol (67-56-1)	7	***Hydrotreated light petroleum distillate (64742-47-8)***	***2***	***Crystalline silica (quartz) (14808-60-7)***	***4***
5	Solvent naphtha, (petroleum), heavy aromatic (64742-94-5)	3	***Ethylene glycol (107-21-1)***	***6***	Propargyl alcohol (107-19-7)	6
6	Hydrotreated light petroleum distillate (64742-47-8)	2	***Ammonium peroxydisulfate (7727-54-0)***	***3***	***Ethylene glycol (107-21-1)***	***6***
7	Naphthalene (91-20-3)	9	***Guar gum/(polysaccharide blend) (9000-30-0) Isopropyl alcohol (67-63-0)***	***1***	***Glutaraldehyde (111-30-8)***	***4***
8	Sodium hydroxide (1310-73-2)	5		***6***	2-Dibromo-3-nitrilopropionamide (10222-01-2)	4
9	Sodium chloride (7647-14-5)	3	***Sodium hydroxide (1310-73-2)***	***5***	***Ammonium chloride (12125-02-9)***	***5***
10	Ammonium peroxydisulfate (7727-54-0)	3	***Potassium hydroxide (1310-58-3)***	***3***	Polyethylene glycol (25322-68-3)	2
11	Ethylene glycol (107-21-1)	6	***Sodium chloride (7647-14-5)***	***3***	***Isopropyl alcohol (67-63-0)***	***6***
12	Ethanol (64-17-5)	4	Propargyl alcohol (107-19-7)	6	Citric acid (77-92-9)	3
13	1,2,4 Trimethyl-benzene (95-63-6)	6	Acetic acid (64-19-7)	5	***Guar gum/(polysaccharide blend) (9000-30-0)***	***1***
14	Poly(oxy-1,2-ethanediyl),alpha-(4-nonylphenyl)-omega-hydroxy-, branched (127087-87-0)	2	***Glutaraldehyde (111-30-8)***	***4***	***Ethanol (64-17-5)***	***4***
15	Isopropyl alcohol (67-63-0)	6	***Ethanol (64-17-5)***	***4***	Ethylene glycol mono-n-butyl ether (111-76-2)	5
16	Potassium metaborate (13709-94-9)	0	Ethylene glycol mono-n-butyl ether (111-76-2)	5	Quaternary ammonium compounds, benzyl-C12-16-alkyldimethyl, chlorides (68424-85-1)	5

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TABLE V. Top 25 Constituents Ranked by Reports of Use for North Dakota, Texas, and Pennsylvania *(Continued)*

	North Dakota		Texas		Pennsylvania	
Rank	Name (CASRN)	Total Health Hazard Count	Name (CASRN)	Total Health Hazard Count	Name (CASRN)	Total Health Hazard Count
17	Sodium chlorite (7758-19-2)	8	Citric acid (77-92-9)	3	Sodium persulfate (7775-27-1)	3
18	Amorphous silica (7631-86-9)	2	***Solvent naphtha, (petroleum), heavy aromatic (64742-94-5)***	***3***	Hemicellulase enzyme (9012-54-8)	0
19	Potassium formate (590-29-4)	0	***Naphthalene (91-20-3)***	***9***	***Sodium hydroxide (1310-73-2)***	***5***
20	Glutaraldehyde (111-30-8)	4	Phenol-formaldehyde novolak resin (9003-35-4)	0	Tributyl tetradecyl phophonium chloride (81741-28-8)	NA
21	Sodium perborate tetrahydrate (10486-00-7)	2	Hexamethylenetetramine (100-97-0)	2	***Sodium chloride (7647-14-5)***	***3***
22	2-Ethylhexanol (104-76-7)	4	***Poly(oxy-1,2-ethanediyl),alpha-(4-nonylphenyl)-omega-hydroxy-, branched (127087-87-0)***	***2***	1-Decanaminium, N-decyl-N,N-dimethyl-, chloride (7173-51-5)	NA
23	Mullite (1302-93-8)	3	***1,2,4 Trimethylbenzene (95-63-6)***	***6***	Alcohols, C12-16, ethoxylated (68551-12-2)	0
24	Sorbitan, mono-9-octadecenoate, (Z)-(1338-43-8)	1	Alcohols, C12-14, ethoxylated propoxylated (68439-51-0)	0	4,4-Dimethyloxazolidine (51200-87-4)	NA
25	Ammonium chloride (12125-02-9)	5	Polyethylene glycol nonylphenyl ether (9016-45-9)	2	3,4,4-Trimethyl-oxazolodine (75673-43-7)	NA

Notes: Constituents listed under Texas or Pennsylvania that are shown in bold italics are also listed in the top 25 by reports of use for North Dakota (Table III). Constituents that have a total health hazard count of 5 or greater, plus crystalline silica (quartz), are also listed in the top 30 constituents ranked by hazard count for North Dakota (Table IV). "NA" indicates that the constituent was not among the 168 constituents for which health hazard counts were generated.

the common use of hazardous chemicals in hydraulic fracturing in various regions of the country. Only four constituents that were reported to be used in Pennsylvania did not have health hazard counts from the Health Hazard Database in Online Supplemental Material (tributyl tetradecyl phophonium chloride, 1-decanaminium, N-decyl-N,N- dimethyl-, chloride, 4,4-dimethyloxazolidine, 3,4,4-trimethyloxazolodine), which indicates that this database may be useful for locations of hydraulic fracturing beyond North Dakota.

HFFs contain several constituents, which raises the concern that the mixture of constituents may pose a greater health hazard than the individual constituents. In North Dakota an average of 29 constituents were reported to be used in each hydraulic fracturing job between December 2009 and November 2013. The maximum number of constituents reportedly used in a single hydraulic fracturing job was 130. To roughly indicate the overall health hazard of HFFs, health hazard counts of each constituent in the HFF mixture were summed. Figure 3 provides a histogram of the overall health hazard counts for the mixtures. The distribution is approximately normal, with an average health hazard count of 65 (standard deviation of 25). The highest overall health hazard count was 132 for a mixture that contained 61 identifiable components (12 components were reported as trade secrets). The acute health hazards accounted for 71% of the count. For chronic oral exposure, the highest counts for carcinogenicity, neurotoxicity, and reproductive/developmental toxicity in a HFF were three, six, and nine, respectively. For chronic inhalation exposure, the highest counts for carcinogenicity, neurotoxicity, and reproductive/developmental toxicity in a HFF

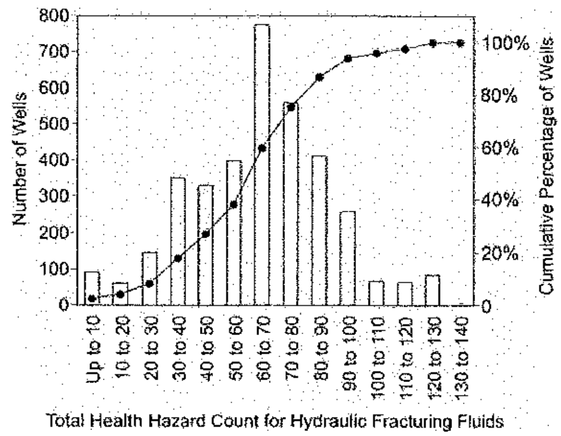

FIGURE 3. Total health hazard counts for the mixtures of constituents found in HFFs. Total health hazard counts were determined for 3,068 distinct HFFs reportedly used in North Dakota. The primary Y-axis is the frequency of the health hazard count. The X-axis is the health hazard count, binned by increments of 10. The secondary Y-axis is the cumulative percentage of wells equal to or less than the Health Hazard Count.

were five, three, and four, respectively. The low chronic counts, relative to the high acute counts, are likely due to a lack of data, as approximately 59% (99/168) of the constituents lack data on any chronic endpoint in contrast to only 35% (58/168) of constituents lacking data on any acute endpoint. Health hazard counts ranged from 34–96 for those HFFs that had the average number of constituents reported (29). This suggests that the health hazards of HFFs may vary between jobs, as the composition of the HFF is formulated for specific conditions.

DISCUSSION

An important first step in the complex health risk assessment of hydraulic fracturing is the identification of known and potential health hazards associated with the constituents of HFFs. The health hazard information is used together with information on exposure, fate and transport of the constituents, and levels of constituents that cause concern about toxicity, to assess the potential overall health risks under specific conditions that occur in the workplace and broader community. To assess the potential health hazards posed by HFFs, a health hazard database was developed and health hazard counts were generated for chronic and acute endpoints, which are applicable for both occupational and general public exposures. The database represents hazard identification, which focuses on identifying potential types of toxicity. A variety of stakeholders can use the health hazard database to identify constituents of concern, and to guide decisions regarding further research, regulation, and management of the fluids used for hydraulic fracturing. For example, health hazard counts were integrated with data on the reports of use to help identify constituents of highest concern due to both high potential for toxicity and frequent use. This type of analysis can help set priorities for pursuing further research such as toxicity testing or exposure assessment that can reduce uncertainty about health hazards and refine risk assessments. It can also help set priorities for risk management, such as remediation and reformulation of fracturing fluids. For example, because constituents serve specific purposes within the HFFs, the health hazard database could be used to help formulate an HFF with the least hazardous constituents feasible. Although the HFF constituents listed in the database are those used in North Dakota, many of the most frequently reported constituents are also frequently reported in Texas and Pennsylvania, regions of the country that are major areas of hydraulic fracturing for the recovery of natural gas. Therefore, this database can have applications for broader geographical areas and hydraulic fracturing practices.

Importantly, a regulatory or guidance value was located for all eleven of the constituents that were listed both in the top 30 by health hazard count and the top 30 by reports of use. This indicates that these substances have already raised sufficient concern for various organizations, such as the United States Environmental Protection Agency (USEPA) and the American Conference of Governmental Industrial Hygienists (ACGIH), to issue limits for exposure. Similarly, these constituents may have relatively high health hazard counts partly because they have undergone more extensive toxicity testing than substances that are not as commonly used. For example, naphthalene has a high total health hazard count (9 out of a maximum possible count of 13) together with a low uncertainty count (2 for chronic toxicity data gaps out of a maximum possible count of 8; 1 for acute toxicity data gaps out of a maximum possible count of 5). The low uncertainty count, in particular, indicates that naphthalene has been studied relatively thoroughly. Having results from rigorously conducted health hazard studies helps improve the accuracy of risk assessments, and consequently contributes to well-informed risk management practices.

This analysis was based on thorough searches of a wide range of national and international public databases, but gaps in health hazard data posed a major source of uncertainty in the analysis presented here. Data were not complete for all constituents. For example, the health hazard count for potassium formate, ranked 19th by reports of use, is zero. This count reflects a high level of uncertainty, as opposed to low toxicity, because information on the 13 health hazard endpoints was not found for this constituent. There are several possible reasons for a lack of health hazard data. For example, hazard data may exist but it may not be available through the public databases that were searched. Another explanation could be that the necessary studies have not been conducted. The constituent may have been a low priority for toxicity testing because it has not been commonly used in the past, it is relatively new, has had very specific conditions for use, or because exposure or toxicity through a specific route of exposure, such as inhalation, is not likely. Hazard uncertainty was addressed by calculating an uncertainty count for each constituent. The uncertainty count was applied to help rank constituents by health hazard; if constituents had the same total health hazard count, the constituent with the greater uncertainty count was considered to pose the greater health hazard. This is based on the conservative, health protective assumption that uncertainty could mean greater toxicity. The uncertainty count, used together with other information, could also be used to set priorities for toxicity testing or help inform decisions about HFF formulation, as the management of known hazards is better informed than the management of unknown hazards.

The analysis presented here highlights the need for a mechanism for disclosing the potential toxicity of proprietary substances that are used in HFFs. The disclosure laws in North Dakota provide trade secret exemptions for chemicals considered by the operator to be confidential business information.[16] Consequently, over 36% (210/569) of the constituents reported in North Dakota between December 2009 and November 2013 were listed as "business proprietary," "trade secret," or "confidential business information." Furthermore, approximately 77% of the hydraulic fracturing jobs used to compile these data reported at least one constituent as confidential business information. Importantly, these labels prevent independent health hazard assessment of these constituents, and thus pose a barrier to a thorough risk assessment and informed risk management of hydraulic fracturing activity. One approach to reduce this source of uncertainty is to require the disclosure of all HFF constituents. Alternatively, the results of toxicity testing could be reported to an independent organization that could make the health hazard data public, while maintaining the trade secret.

The analysis also revealed the need for quality control of the data reported to fracfocus.org. No agency or organization is responsible for overseeing the quality, completeness, and consistency of the reports.[20] Accordingly, different types of errors were found in the FracFocus database, some of which limited the analysis. For example, entries were found in which the maximum concentration of the constituent in the HFF was listed as over 27,000%. Other errors included mismatches between constituent name and CASRN (57/359, 16%), reported CASRNs that were not listed in CHEMIDplus (12/359, 3%), and constituents that were listed with a CASRN, but for which the purpose was never identified (182/359, 51%). It is likely that these issues are input errors or honest mistakes, but developing a mechanism for quality control of databases that disclose the constituents used in HFFs would be a great benefit to all stakeholders with an interest in hydraulic fracturing.

CONCLUSIONS

As the use of hydraulic fracturing methods by the oil and gas industry expands, attention needs to be paid to the human health risks to those employed in the supply chain, and to the general public. This study investigated the known and potential hazards from the constituents used in HFFs, and established the differences between those that are acute (primarily occupational) and those that are chronic (occupational, general public). This study serves as a point of departure for future investigations into the risks and management of hydraulic fracturing, ranging from life-cycle assessments to risk assessments that incorporate environmental and occupational exposure, and environmental fate and transport modeling.

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SUPPLEMNTAL MATERIAL

Supplemental data for this article can be accessed at tandfonline.com/uoeh. AIHA and ACGIH members may also access supplementary material at http://oeh.tandfonline.com/.

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