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Comparative Human Toxicity Impact of Electricity Produced from Shale Gas and Coal

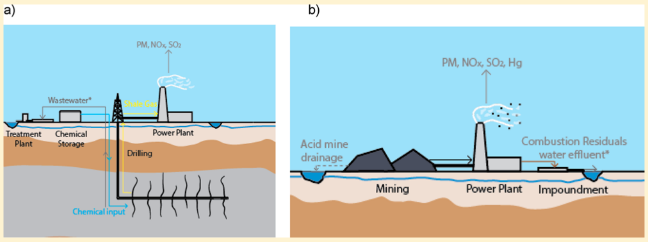
Lu Chen,[†,‡] Shelie A. Miller,*[,†,‡] and Brian R. Ellis[‡]

[†]Center for Sustainable Systems, School for Environment and Sustainability, University of Michigan, 440 Church Street, Ann Arbor Michigan 48109, United States

[‡]Department of Civil and Environmental Engineering, University of Michigan, 2350 Hayward Street, Ann Arbor Michigan 48109, United States

S *Supporting Information*

ABSTRACT: The human toxicity impact (HTI) of electricity produced from shale gas is lower than the HTI of electricity produced from coal, with 90% confidence using a Monte Carlo Analysis. Two different impact assessment methods estimate the HTI of shale gas electricity to be 1−2 orders of magnitude less than the HTI of coal electricity (0.016−0.024 DALY/GWh versus 0.69−1.7 DALY/GWh). Further, an implausible shale gas scenario where all fracturing fluid and untreated produced water is discharged directly to surface water throughout the lifetime of a well also has a lower HTI than coal electricity. Particulate matter dominates the HTI for both systems, representing a much larger contribution to the overall toxicity burden than VOCs or any aquatic emission. Aquatic emissions can become larger contributors to the HTI when waste products are inadequately disposed or there are significant infrastructure or equipment failures. Large uncertainty and lack of exposure data prevent a full risk assessment; however, the results of this analysis provide a comparison of relative toxicity, which can be used to identify target areas for improvement and assess potential trade-offs with other environmental impacts.



■ INTRODUCTION

Potential contamination of drinking water is a major concern surrounding hydraulic fracturing operations. Recent studies quantify water quality issues associated with shale gas,[1−3] although the magnitude and frequency of contamination events are still not well documented. While concerns of water contamination and VOC emissions associated with shale gas production may be warranted, it is also important to contextualize the potential human toxicity impacts of shale gas relative to other sources of electricity.

A growing number of studies investigate the potential environmental impacts related to hydraulic fracturing. A recent analysis suggests that spills of fracturing fluid could impact soil health due to the ionic strength of flowback water, although the study found that human health concerns were minimal.[4] A variety of studies have highlighted concerns regarding major water withdrawals and contamination events due to stray gas leakage, accidental spills, and wastewater disposal.[3,5−7] Data are scarce regarding changes to air quality associated with hydraulic fracturing, although some localized analyses are being conducted.[8] VOC signatures from winter haze events have been linked to the Bakken,[9] although it is difficult to differentiate between activities associated with hydraulic fracturing and other oil and gas extraction activities. Other analyses suggest the contribution of VOCs to changes in air quality is minimal,[8,10] indicating the need for further investigations into VOC emissions associated with hydraulic fracturing. Additional studies have begun to assess the toxicity of components of fracturing fluids and flowback water.[11] Life cycle assessments of electricity produced from shale gas have focused on a variety of environmental concerns, including greenhouse gas emissions, water use, and criteria air emissions;[1,12−16] however, there does not appear to be a comprehensive toxicity analysis comparing electricity produced from shale gas and coal.

The relative human toxicity impact (HTI) of electricity produced from shale gas and coal are compared to better understand how increased penetration of shale gas will affect overall toxic releases associated with the power sector. HTI is commonly used within life cycle assessment to quantify the inherent toxicity burden associated with emissions from a product or process.[17] The HTI is measured as disability-adjusted life years per unit of electricity generation (DALY/GWh) and is calculated by means of generic fate and exposure assumptions applied consistently across all systems. The HTI serves as a useful screening metric and an initial step toward a full risk assessment. Characterization factors (CFs), sometimes referred to as human toxicity potentials (HTP), estimate human health damage for a wide range of chemicals and are

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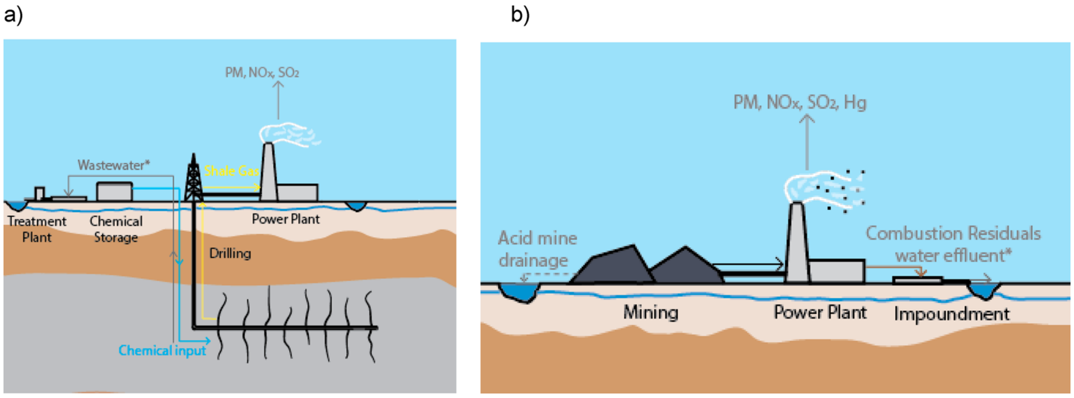

Figure 1. (a) Sketch of shale gas electricity system, including hydraulic fracturing operations and electricity generation; (b) Sketch of coal electricity system, including coal mining and electricity generation.

expressed as DALY/kg emission. The results of life cycle impact assessment are subject to a great deal of variability and uncertainty. Therefore, this analysis uses a variety of approaches to test the robustness of the results. Two scenarios, representing a baseline case and an accidental release scenario, are used to examine different sets of potential assumptions. Both the USEtox 2.0 and ReCiPe 2016 impact assessment methods are used to demonstrate how different sets of CFs may influence the results.[18,19] Local sensitivity analysis in the form of one-at-a-time perturbation is used to assess the degree of influence associated with individual parameters. Finally, Monte Carlo Analysis is used as a global sensitivity method to simulate how both variability and uncertainty across parameter ranges affect the outcome of the analysis.

This study conducts a comparative analysis of the toxicological human health effects of electricity produced from shale gas and coal. Human toxicity is a single life cycle impact assessment indicator and is not a comprehensive analysis of environmental factors. A complete life cycle assessment (LCA) should include a broader suite of impacts, including climate change, ecosystem quality, resource depletion, land use, and water use. The evolving nature of research on the environmental impacts of hydraulic fracturing is characterized by data limitations and lack of consensus on individual indicators such as climate change.[20] Focused, in-depth analyses on discrete indicators are still needed to be able to compile the necessary information to complete a comprehensive LCA. Therefore, the results of this analysis on human toxicity must be used alongside compatible studies focusing on other indicators in order to understand the overall environmental context.

■ MATERIALS AND METHODS

The scope of this analysis is a human toxicity impact assessment of direct chemical emissions associated with shale gas and coal electricity. The inventory includes chemical emissions that occur during resource extraction and electricity generation, the stages with the greatest associated toxicity. Baseline case and accidental release scenarios are constructed for both energy systems. The baseline scenario approximates normal operating conditions, while the accidental release scenario simulates major unintended releases of emissions for each system to calculate an upper toxicity limit. For the shale gas system, the study includes aquatic emissions of fracturing fluid chemicals and produced water associated with shale gas extraction, air emissions of NO_x, PM, and VOCs during shale gas extraction, and emissions of PM, VOCs, NO_x and SO_x during electricity generation. To compensate for data scarcity associated with shale gas production and improve confidence in the robustness of the results, the toxicity of the shale gas system is overestimated whenever there is insufficient or missing data. For the coal system, the study estimates the toxicity associated with effluent loadings from coal mine outfalls, air emissions of PM, Hg, VOCs, NO_x, and SO_x during electricity generation, and aqueous emissions from coal ash impoundments. For the coal accidental release scenario, emissions of metals associated with a coal ash spill are also included. Chemical emissions associated with transportation, infrastructure, and cooling water are not included in the analysis, due to minor contributions to overall toxicity.[1,21] Figure 1 depicts the shale gas and coal systems.

Pennsylvania is used as the point of origin for both shale gas and coal, given the abundance of both sources of energy within the region. When Pennsylvania specific data are not available, alternate data sets are used that can be reasonably assumed to be consistent with coal and shale gas resources originating from the region. National averages are used for efficiency and air emissions associated with natural gas and coal electricity generation, as the resources produced within the region are distributed nationally.[22]

Human Toxicity Calculations. Chemical emissions that have a direct toxicological effect on human health are included in the analysis. Factors that are nontoxicity related (e.g., noise, light, stress), have an indirect effect on human health (e.g., ozone depletion, climate change), or cause environmental damage not related to human health (e.g., salinity, pH, turbidity) are outside the scope of the analysis. Toxic releases are measured using disability-adjusted life years (DALY) per GWh of electricity produced. A DALY is a common metric to measure impact on human health and corresponds to the number of years lost due to poor health, disability, or premature death.[18,19]

To calculate HTI of electricity produced from coal and shale gas, the USEtox 2.0 (released in 2015) and ReCiPe 2016 characterization factors are used.[18,19] USEtox and ReCiPe are impact assessment methods that estimate the toxicity of a given quantity of emissions by providing CFs that estimate human health damage for a wide range of chemicals. Human toxicity CFs are averages for North America. Both USEtox and ReCiPe are long-term exposure assessment methods and consider the impact on the general population. The time horizon is 100 years for USEtox and infinite for ReCiPe's Egalitarian

Table 1. Assumptions Used for Baseline and Accidental Release Shale Gas Scenarios and Parameter Ranges Found within the Literature for Comparison[a]

parameter	unit	baseline case	accidental release	literature range	reference
average fracturing fluid volume	m^3/well	13 000	13 000	3500–26000	27
flowback water percentage share of injected fluid	%	15	15	10–15	1,26
produced water percentage share of injected fluid	%	5	5	1–7	1
shale gas electricity generation efficiency	Mcf/kWh	0.0101	0.0101	0.00964–0.0134	28–30
shale gas well lifetime span	year	15	15	5–30	31
shale gas production volume	MMcf	4300	4300	1600–9000	32,33
barium removal efficiency	%	90	0	0–99	34
fracturing fluid release rate	% released	1	100	see details in text	3

[a] Calculations and further justifications are found within the SI.

assumptions. Human health impacts due to acute workplace exposures are not taken into account. USEtox 2.0 is a scientific consensus model recommended as the preferred database for calculating HTI by the United Nations Environment Programme and the Society of Environmental Toxicology and Chemistry's Life Cycle Initiative. ReCiPe was developed by a consortium of life cycle assessment practitioners. The two methods were developed independently and both provide end point level CFs for carcinogenic and noncarcinogenic toxicity. Life cycle impact assessment is an accepted method to quantify the relative hazard and importance of pollutants when data for a full-scale risk assessment are not available.[23]

CFs from both models are reported due to the large degree of uncertainty in developing appropriate CFs and differences in CFs for key constituents of this analysis. The sensitivity analysis addresses concerns regarding CF uncertainty in greater detail. eq 1 is the generic form for calculating HTI for an individual emission.

$$\mathrm{HTI}_i = \mathrm{CF}_i \times E_i \tag{1}$$

Where i represents each unique chemical, HTI_i is the human toxicity impact for each emission (DALY/GWh), CF_i is the characterization factor for each chemical (DALY/kg emission), E_i is the emissions of each chemical associated with a given amount of electricity generation (kg/GWh). Total HTI (HTI_t) for life cycle electricity generation is the sum of the individual chemical impacts (eq 2).

$$\mathrm{HTI}_t = \sum \mathrm{HTI}_t \tag{2}$$

The HTI of PM reported in this analysis is the aggregate human health damages resulting from both $PM_{2.5}$ and PM_{10} formed from both primary (directly emitted to the atmosphere) and secondary (generated by chemical reactions with NH_3, NO_x and SO_x) sources.

PM has a different mechanism for impacting human health than other chemicals in the inventory and the CF for PM is derived via a different approach. The ReCiPe model contains a CF for $PM_{2.5}$, as well as for NH_3, NO_x, and SO_2, which contribute to secondary PM aerosol formation. USEtox does not include a CF for PM, so the USEtox method is supplemented with a method described by Gronlund et al.[24] Emission data collected from twenty-three natural gas and 13 coal-fired power plants within Pennsylvania are used to calculate the HTI of PM, using production-weighted averages of electricity generation for each plant.[25] The Supporting Information (SI) contains relevant assumptions and calculations for the HTI of PM.[24,25]

Shale Gas. In an effort to be conservative due to uncertainty and lack of data, assumptions are made to overestimate the potential toxic emissions for the shale gas baseline whenever limited data inhibit the analysis. A number of pathways exist for chemicals within fracturing fluid and produced water to enter the environment. Potential emission pathways include storage failures of fracturing fluids and produced water at the surface, produced water migration through the subsurface into groundwater, contamination from faulty casings, and as effluent from wastewater treatment plants.[26]

To estimate potential releases of fracturing fluid chemicals, baseline and accidental release scenarios are constructed. Data from 2990 wells from FracFocus within Pennsylvania are used, containing a total of 368 chemicals. FracFocus 3.0 is a machine-readable database that includes self-reported chemicals used in fracturing fluid formulations from over 62 000 wells throughout the United States.[27] The ID number, total fracturing fluids volume, purpose of each chemical (corrosion inhibitor, proppant, etc.), and the concentration of each component were recorded for each well. To account for differences in chemical compositions due to geographic and operational variations, average well information for each Pennsylvania County represented in FracFocus as of October 2015 was used. Not all wells use the same chemicals, so the average concentration was used for the 100 most frequently used chemicals from 2010 to 2015 (SI Table S1). Most of the fracturing formulations surveyed within FracFocus include less than 40 chemicals, so using the average concentration of the 100 most frequently used chemicals overestimates the HTI for shale gas.

Currently, no database exists that systematically records spills and other releases.[35] The baseline case is conservative to compensate for data gaps in toxicity and chemical composition. The baseline case assumes that 1% of fracturing fluids are emitted to surface water without any treatment, although the discharge could occur via any of the potential emissions release mechanisms. A recent study estimated the total known spill volume of fracturing fluid from 6000 wells to be 174 m^3 from 2008 to 2013,[35] averaging 0.03 m^3 per well during that period. The assessment report from the EPA lists the median spill volume as 1.6 m^3 per well that reports a spill (range 0.19–72 m^3), with 0.4–12.2 spills per 100 wells, which is similar to data reported elsewhere,[36,37] although it does not necessarily account for spills that go unreported. Of the spills that are reported, the EPA estimates that only 9% of spills reach surface water. Assuming 13 000 m^3 of fracturing fluid for each well (Table 1),[27] a more reasonable spill rate is 0.001% of injected fluids, as opposed to the 1% assumed in the baseline scenario.

In addition to fracturing fluids injected into the wells, produced water has the potential to contaminate drinking water sources. The chemical composition of flowback and produced water are different, with larger concentrations of naturally dissolved materials in produced water than flowback water. Some of the chemicals in produced water, especially organic compounds, originate from the injected fracturing fluids. It is difficult to differentiate naturally occurring organic compounds from those that derive from fracturing fluid formulations. Because of the difficulty distinguishing between the sources of chemicals, all chemicals within produced water are treated as a separate source of emissions, effectively double-counting the fracturing fluid chemicals contained in produced water. The volume of flowback and produced water is assumed to be 20% of the injected water volume and have the average chemical composition of a compilation of 35,000 data entries for the Marcellus shale gas region reported by Abualfaraj et al.[38] (SI Table S2).

Produced water from hydraulic fracturing operations is assumed to be sent to a dedicated water treatment plant, which is the most common method of disposal in Pennsylvania.[39] Alternative methods of disposal include injection into Class II disposal wells and recycling for additional shale gas extraction operations.

Chemical constituents of produced water can enter surface water via the same mechanisms as fracturing fluids, and also as effluent from wastewater treatment due to incomplete removal. For the baseline shale gas case, 10% of the chemicals within produced water are assumed to be discharged to surface water. For context, a recent study estimated the total known spill volume of produced water from 6000 wells to be 473 m^3 from 2008 to 2013,[35] averaging 0.079 m^3 per well. The assessment report from the EPA lists the median spill volume of produced water as 37.5 m^3 per well that reports a spill, with 0.4−12.2 spills per 100 wells. Assuming 2700 m^3 of produced water per well (Table 1), then the spill rate of produced water is likely less than 1%. In addition, some fraction of the constituents of produced water are discharged as effluent from the wastewater treatment facility, so the overall estimate of 10% discharge is assumed to be reasonable and likely an overestimate for the baseline case.

The accidental release scenario for shale gas assumes 100% of injected fracturing fluid and 100% of produced water is directly discharged to surface water over the entire lifetime of the well. While this scenario is highly improbable, it provides a maximum threshold for possible toxicity loads associated with shale gas.

For air emissions from shale gas fields, values are obtained from a comprehensive study by Roy et. al, which quantifies total load of NO_x, $PM_{2.5}$, and VOC emissions per well associated with hydraulic fracturing operations.[40] Their study estimates total load, but does not provide compositional analysis of VOCs. In the absence of compositional data from this study, the CF for benzene was used for all VOCs in this analysis. VOC composition from oil and gas sites tend to have significant proportions of alkanes and alkenes that have lower toxicity than aromatics. Therefore, the use of the benzene CF is considered an overestimation of VOC contribution to HTI.

Coal. An EPA study on coal mine discharge[41] was used in conjunction with annual Pennsylvania coal production estimates[42] to estimate toxicity associated with coal mining.[41] The study calculated loading of aluminum, iron, manganese, and total suspended solids in Pennsylvania streams impacted by acid mine drainage.

To estimate human toxicity of mercury air emissions from coal-fired power plants, data was collected from the 100 largest power producers in the United Sates in 2013.[43] Combustion residuals, also known as coal ash or fly ash, are assumed to be stored in surface impoundments. Some of the chemicals within the coal ash leach out of the disposal site and are released to the environment (SI Table S3).[44] The coal ash effluent values are assumed to be representative of coal originating in Pennsylvania, given the locations of the power plants where data were obtained. An EPA study on power plant effluent, which sampled coal-fired power plants in the Appalachian region,[45] reported similar values. It is assumed that, in addition to ash, the coal ash impoundment also receives discharges of flue gas desulfurization wastewater and transport water for fly and bottom ash. These waste streams appear to be included in reported outflow values and are therefore not calculated as separate emissions.

Electricity generation was calculated using nameplate capacities of each power plant, assuming continual operation of all plants, 365 days/year, which overestimates the generation capacity of each plant, underestimating the HTI associated with electricity production. Wet disposal of coal ash is not the only possible method of disposal. Coal ash can also be reused in engineering applications or landfilled in a dry state. This choice is further addressed in the sensitivity analysis and discussion.

The accidental release scenario for coal uses the same assumptions for all parameters of the baseline case, and also includes unintentional release of coal ash into surface water, similar to the impoundment failures that occurred in Kingston, TN in 2008 and Eden, NC in 2014. Although major releases of coal ash spills are infrequent, they have historical precedent and their inclusion provides an analogous system to compare with the accidental release scenario for shale gas.

For the coal accidental release scenario, postremediation data from the Kingston coal ash spill were used.[46] Remediation efforts took place in 2009 and 2010 leaving 0.18 million m^3 of ash in the Emory River. The density of ash is assumed to be 1500 kg/m^3 and the HTI is calculated based on the volume of the ash left in the Emory River postremediation. Babbit et al. calculated the average coal ash generation from four power plants in Florida as 0.0568 kg/kWh.[21] The National Renewable Energy Laboratory estimates the average coal ash generation rate as 0.1 kg/kWh.[47] The smaller coal ash generation value is used in this analysis in order to provide a more conservative HTI estimate.

Data Limitations. The scope of this study is intended to address the full life cycles of shale gas and coal electricity; however, data limitations necessitated a number of choices to be made regarding the life cycle inventory of both shale gas and coal. Because of these limitations, systematic choices were made to overestimate the coal inventory while underestimating the shale gas inventory.

Most studies that report VOCs or nonmethane hydrocarbons report total load but do not provide compositional analysis. Benzene is used as the CF for all VOCs in this analysis, which likely overestimates VOC contributions to total HTI for all scenarios.

Data relevant to calculating the HTI of coal mining is rarely reported in a format that is conducive to translation into inventory data. Mining emissions tend to be reported as concentrations, without sufficient data to relate concentrations to mass emission/mass coal extracted needed for the inventory (e.g., streamflow, time since coal extraction, overall amount of coal extracted from site). Although other studies have found that coal mining may have significant human health impacts on

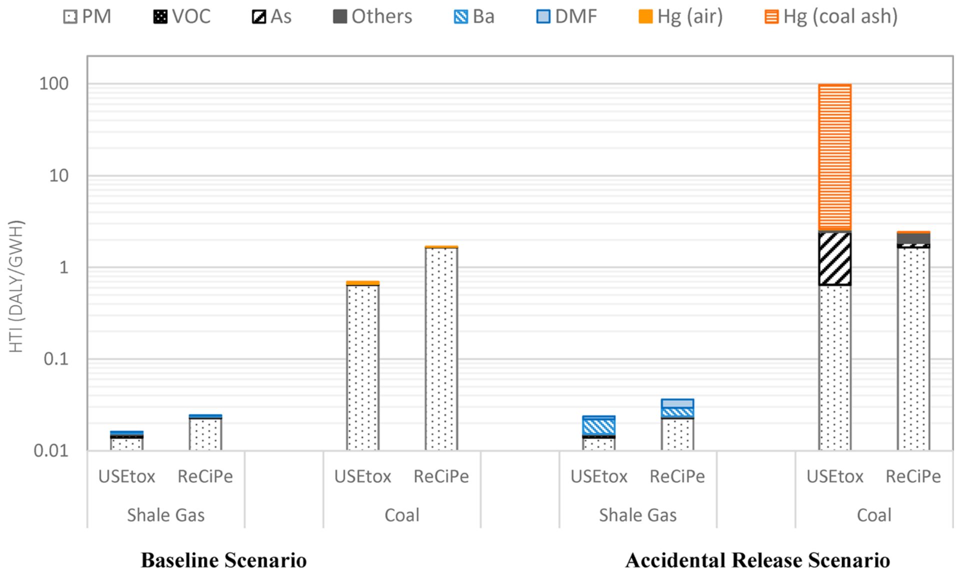

Figure 2. Baseline and accidental release scenarios for electricity produced from shale gas and coal using the USEtox and ReCiPe2016 methods to quantify HTI. Data is depicted on a log scale. Major components of HTI include particulate matter (PM), volatile organic compounds (VOC), arsenic (As), barium (Ba), N, N′-dimethylformamide (DMF), and mercury (Hg). Constituents that appear in both systems are depicted in gray scale, whereas those only found in coal system are in orange and those only found in shale gas system are in blue.

nearby communities,[48−50] lack of appropriate data inhibit inclusion of several aspects of the coal system into this analysis. Potential toxicity impacts are associated with cadmium and selenium in mine drainage; however, limited data prevented quantification of loading data from either element.[41] Similarly, calculation of HTI associated with coal mining does not include particulate matter or other air emissions associated with mining that are analogous for the data obtained for the shale gas baseline case. Fugitive coal dust emissions along transportation routes has been linked to chronic community-level exposures of particulate matter and metals,[51] but the format of data available inhibits translation into inventory metrics useful for this analysis.

In addition to limitations associated with life cycle inventory collection, ReCiPe and USEtox do not contain a uniform list of chemicals, so a given chemical may be associated with a CF in one database and not in another. Characterization factors are also highly uncertain and a standard operating assumption is that a CF may vary by 3 orders of magnitude in either direction.[18,19]

Different oxidation states of metals have different toxicities; however, oxidation states are not available from the data sets used in this analysis. To be consistent with the systematic overestimation of the shale gas HTI, the more toxic oxidation state is assigned for shale gas emissions whenever available. Similarly, mercury in the coal system is assumed to be present in its inorganic form, even though it has the potential to form organic mercury, such as methylmercury and dimethylmercury, which are more toxic than inorganic mercury.[52] CFs for radionuclides are included in ReCiPe but not in USEtox. The effects on results of these issues are further discussed in the Results and Discussion.

Uncertainty and Sensitivity Analysis. A Monte Carlo Analysis (MCA) is used to determine the range of possible results and the extent of overlap between the shale gas and coal cases. Triangular distributions are assigned to each of the parameters within the model, using the most likely value and upper and lower range boundaries (see Table 1 and SI Tables S10−S12). For the CFs, which span 6 orders of magnitude, triangular distributions are assigned to the exponent of each parameter to reduce positive bias in sampling. The results of 10 000 trials are reported.

A one-at-a-time perturbation method is used to assess the sensitivity of the results to each input parameter. The effect of each parameter on the HTI is determined by changing its value to the extreme ends of its range while keeping all other parameters at their initial model value (see Table 1 and SI Tables S10−S12). CFs from USEtox are used as the default for both the sensitivity analysis and the MCA, unless a CF was available only in ReCiPe.

In addition to the sensitivity analysis, two alternate operating scenarios are explored for coal. The scenarios are disposal of coal ash via a dry storage method and implementation of the Mercury Air Toxics Standards (MATS) regulation.[53]

■ RESULTS AND DISCUSSION

Figure 2 shows that the baseline scenario for coal-fired electricity has a greater HTI than both the baseline and accidental release scenarios for shale gas, by at least an order of magnitude. The HTI baseline values are 0.016 DALY/GWh (USEtox) and 0.024 DALY/GWh (ReCiPe) for shale gas and 0.69 DALY/GWh (USEtox) and 1.7 DALY/GWh (ReCiPe) for coal. A prior study on coal toxicity using CML2001, another popular impact assessment method, estimated an HTI of coal electricity to be between 0.24 DALY/GWh to 2.2 DALY/GWh,[54] which aligns with the results of this analysis.

Particulate matter is the dominant toxicity contributor for both shale gas (86% USEtox, 93% ReCiPe) and coal (92% USEtox, 98% ReCiPe), and includes calculation of primary emissions of particulate as well as secondary aerosol formation (see SI Tables S6−S9). Other contributors to the HTI of the coal baseline are air emissions of mercury (5.6% USEtox, < 1% for ReCiPe) and arsenic (1.1% USEtox, 1.3% ReCiPe). Other contributors to the shale gas baseline are VOCs (5.4% USEtox, < 1% ReCiPe) and barium (4.4% USEtox and 2.2% ReCiPe) due to the high concentration of barium in the produced brine

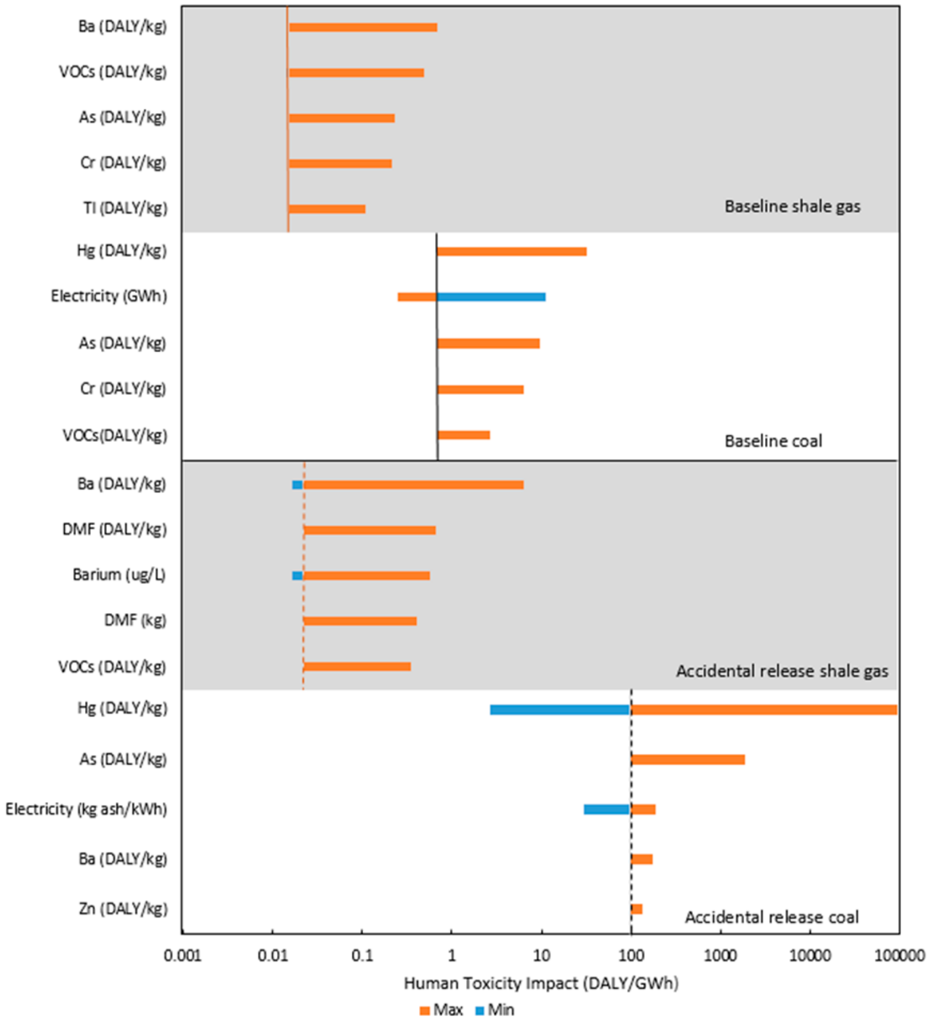

Figure 3. One-at-a-time sensitivity analysis of USEtox results shown on a log scale. Each of the 137 parameters used within the analysis is varied across its range (SI Tables S6−S8), leaving all other parameter values constant. The five most influential components for HTI estimation are shown for maximum (orange) and minimum (blue) values of each parameter's distribution. Each of the vertical lines corresponds to the initial scenario results. The solid and dashed lines are the results of the baseline and accidental release for shale gas (orange) and coal (black). Parameters representing concentrations of a chemical are reported as μg/L; those representing CF of a chemical are reported as DALY/kg; those representing mass of each chemical are reported as kg or kg/kWh.

and the large volume of produced water (SI Table S2). A variety of chemicals in produced water make up the remainder of the contributions to the HTI of shale gas, including arsenic (1.3% USEtox, 2.2% ReCiPe) and N, N′-dimethylformamide (DMF) (<1% both USEtox, ReCiPe), a compound commonly found within fracturing fluid as a corrosion inhibitor.

Other contributors to shale gas toxicity include fracturing fluid chemicals that are used for their properties as biocides, friction reducers, and iron control. Biocides and corrosion inhibitors are reported elsewhere as the main toxic chemical additives of concern within fracturing fluids.[55] Although some concerns have been raised regarding emissions of radioactive elements such as radium,[1,34,56] radionuclides are not major factors in this analysis due to lower CF estimates (Table S2); however, it should be highlighted that the toxicity pathways of radionuclides are considerably different than other chemical compounds in this analysis and may warrant further analysis. Even though some individual components of fracturing fluids have relatively large CF values, fracturing fluids are only used during completion of a shale gas well. As the volume of produced gas increases over the lifetime of the well, the impact of fracturing chemicals on a per GWh basis decreases (SI Figure S1). Meanwhile, the emissions generated during electricity production, such as particulate matter, have a first order producing relationship with electricity generation, so the emission intensity is constant on a per GWh basis, resulting in a larger contribution to HTI.

Figure 2 also provides estimates of accidental release conditions, resulting from major unintended releases within each system. The coal accidental release scenario uses data from historic coal ash spills (SI Table S5), whereas the accidental release scenario for shale gas represents an implausible upper bound assumption of complete release of all fracturing fluid and produced water directly to surface water throughout the entire well lifetime (Table 1). Under the accidental release scenario, a major spill of coal ash results in an HTI that is 2−3 orders of magnitude greater than the accidental release scenario for shale gas, with calculated HTI values of 0.024 DALY/GWh (USEtox) and 0.036 DALY/GWh (ReCiPe) for shale gas and 98 DALY/GWh (USEtox) and 2.4 DALY/GWh (ReCiPe) for coal. Under the accidental release scenario for coal, mercury from coal ash is the largest contributor to HTI for USEtox

(97%) but has a negligible contribution to HTI for ReCiPe (<1%) due to a much lower CF for mercury in ReCiPe. Particulate emissions remain the dominant contributor for the HTI of ReCiPe (68%). Other major components of the HTI for the coal accidental release scenario include arsenic in coal ash (1.8% USEtox, 8.2% ReCiPe) and a variety of other coal ash constituents (<1% USEtox, 23% ReCiPe). While particulate matter from electricity generation continues to be the dominant factor for the accidental release scenario of shale gas (58% USEtox, 62% ReCiPe), the chemicals in produced water are a larger proportion of the HTI, including barium (29% USEtox, 15% ReCiPe), and DMF (7.1% USEtox, 19% ReCiPe).

The accidental release scenarios for shale gas are still one to two orders of magnitude lower than the baseline case for coal (Figure 2 and SI Tables S1−S2), indicating that the toxicity of electricity from shale gas is lower than the toxicity of electricity produced from coal even under the most extreme set of assumptions. Figure 2 highlights the well-known variability and uncertainty associated with impact assessment calculations. The ReCiPe and USEtox methods provide results for the accidental release scenarios for both shale gas and coal that differ by an order of magnitude, deriving from discrepancies in CFs within the databases (SI Table S1, S2, S4). For example, the CF associated with mercury in ReCiPe is significantly lower than the mercury CF in USEtox. Despite the variations and discrepancies between the two models, the overall trends of the analysis are similar. The rationale for using both USEtox and ReCiPe in this analysis was to determine whether different toxicity models might yield fundamentally different results; however, the results of the analysis appear to indicate that while the calculated contributions of individual chemicals to overall toxicity may differ, the overall conclusions are similar irrespective of the impact assessment method used.

To more directly address the large uncertainty and variability in HTI estimation, a one-at-a-time sensitivity analysis is employed to test the robustness of the results. The sensitivity analysis varies operational parameters (e.g., chemical concentrations, water volumes, well lifetime) across their range of minimum and maximum reported values and CFs by 3 orders of magnitude in each direction (SI Tables S10−S12).

Figure 3 can be used to gauge the robustness of model results by indicating which parameters have the greatest influence on the scenarios. Figure 3 demonstrates how the results would change if any of the parameters existed at the extreme ends of their range. Increasing any individual parameter to its maximum value within either of the shale gas scenarios will not surpass the toxicity of the corresponding coal scenarios. Similarly, no parameters exist in the coal scenarios that have minimum values that would cause the HTI of coal to fall below the HTI of the corresponding shale gas scenarios. A CF for barium that is 3 orders of magnitude greater than its recommended value could make the accidental release scenario for shale gas more toxic than the baseline case for coal. Although CFs are not available for some of the chemical constituents of fracturing fluids, the available CFs of fracturing fluid chemicals are, on average, 8 orders of magnitude lower than the CF of mercury air emissions (SI Tables S1 and S4). It is assumed that any gaps in data are unlikely to significantly affect overall results.

Figure 3 allows examination of individual parameters contributions to the sensitivity of the results. A Monte Carlo Analysis (MCA) allows a greater understanding of the overall robustness of the results, as reported in Figure 4.

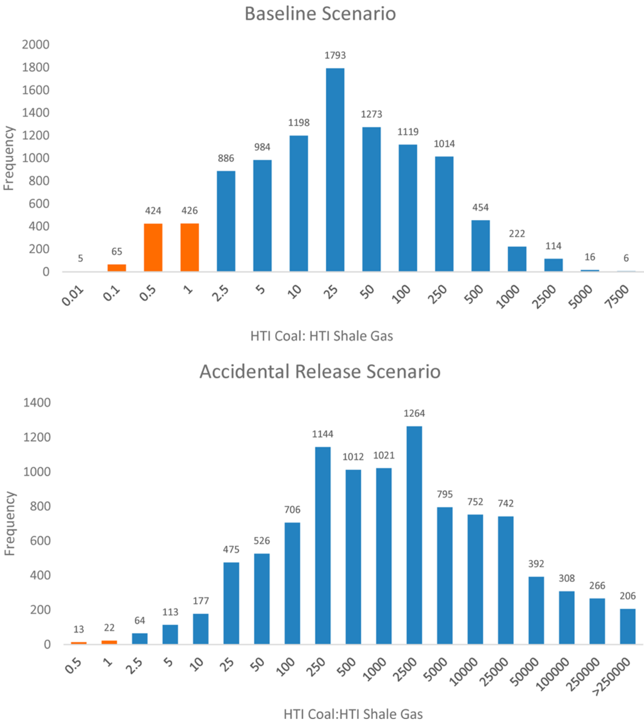

Figure 4. Histogram of MCA results for the baseline and accidental release scenarios, reported as the ratio of the HTI of coal to the HTI of shale gas for 10,000 trials. Each bar represents the number of values in each bin, with the value on the *x*-axis indicating the highest value of the bin range. Ratios greater than 1, depicted in blue, indicate an HTI of coal greater than the HTI of shale gas. Ratios less than 1, depicted in orange, indicate an HTI of coal less than the HTI of shale gas.

Results are reported as the ratio of individual trials to ensure that any shared parameters, such as chemical characterization factors, are the same for both shale gas and coal for each trial run. The results of the Monte Carlo Analysis indicate that the HTI for the shale gas baseline is lower than the coal baseline in 90.8% of the trials, suggesting 90% confidence in the overall finding that the HTI of shale gas is lower than the HTI of coal. The median value of 10 000 MCA trials indicates the HTI of coal is 17 times greater than the shale gas baseline case. For the accidental release scenario, 99.6% of MCA trials result in a coal HTI that is greater than the shale gas HTI, with the median value of the HTI of coal 830 times greater than shale gas. Therefore, best available information indicates that the coal system releases more toxic emissions than the shale gas system.

If new regulations are enacted with respect to the coal industry, the results of the analysis have the potential to change. Restrictions regarding the use of surface impoundments to store coal ash slurry will reduce leachate and the potential for future unintentional releases of coal ash.[57] Leachate emissions are a minor contributor to the HTI of the coal baseline case (SI Table S4); nevertheless, a shift to an alternate coal ash disposal method would reduce the HTI of the coal baseline (1.5% reduction in USEtox, 1.3% reduction in ReCiPe), but not enough to have an effect on the comparative results with shale gas. Similarly, full compliance with the Mercury and Air Toxics Standards[53] would reduce the HTI associated with air emissions of Hg by about 23%, which corresponds to a 2% reduction in overall HTI. These scenarios highlight the dominance of particulate matter's contribution to the HTI of the coal baseline. Given the large amount of particulate matter associated with coal electricity, major efforts to reduce toxicity from other sources will be insufficient to reduce the HTI of coal to be similar to that of shale gas. Even when particulate matter is not taken into account, the HTI of coal is greater than the HTI of shale gas. While particulate matter is the greatest contributor to human toxicity for both coal and shale gas, the accidental release cases highlight the importance of appropriate treatment and containment of aquatic releases. Aquatic emissions can become major contributors to the overall HTI when waste products are inadequately disposed or there are infrastructure or equipment failures.

The results of this analysis indicate that electricity produced from shale gas has a lower HTI than electricity produced from coal. As the electricity portfolio continues to shift from coal to natural gas, the overall toxicity burden of the electricity sector can be expected to decrease. New regulations on the coal industry, such as those regarding coal ash disposal and mercury air emissions, will have some impact on the HTI of electricity production. Technology improvements in hydraulic fracturing, such as replacing some chemicals with less toxic ones and reducing water usage, is likely to reduce the HTI of electricity produced by shale gas as well. Although translation of HTI into actual human health effects can only be obtained through a more detailed risk assessment, the results of this analysis indicate that the overall toxicity burden of the power sector will be reduced by substituting natural gas for coal. The results of the analysis are robust under both baseline and accidental release scenarios.

This analysis does not imply that concerns regarding emissions associated with shale gas production are unfounded, only that the toxicity burden is lower than that from coal. Additional information is needed to quantify the relative impacts of factors that are nontoxicity related (e.g., noise, light, stress) or cause environmental damage not related to human health (e.g., salinity, pH, turbidity). The analysis also does not take into account the complexity of the electricity sector, as it does not quantify the extent to which shale gas actually displaces coal or its potential to impede penetration of renewable energy alternatives.

Given the complexity of the shale gas system and its increasing importance to the energy sector, individual impact metrics for shale gas should not be evaluated in isolation. This analysis provides a robust and detailed analysis of human health impacts that can be used in conjunction with studies that are emerging on other critical factors of interest to inform a more complete depiction of the aggregate environmental impact of shale gas.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b03546.

> Parameters used for HTI calculations of shale gas and coal scenarios. Sensitivity analysis input data and results. Influence of well lifetime on shale gas results (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*Phone: 734-763-8645; e-mail: sheliem@umich.edu.

ORCID

Shelie A. Miller: 0000-0003-0379-3993

Brian R. Ellis: 0000-0002-7253-4285

Notes

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