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EDITORIAL
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More than a decade of hydraulic fracturing and horizontal drilling research

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Unconventional natural gas and oil withdrawals in the United States (US) increased dramatically starting in the mid-2000s (Fig. 1), further enabled by the onset of multi-well pads with horizontal drilling in 2007. This expansion was met with great scientific interest in the technological progress and its interplay with people and the environment. Broadly speaking, research on the topic of "hydraulic fracturing" grew from 30 to 50 publications per year at the onset to just under 1000 publications per year today. However, studies of the geochemistry and biology of horizontal drilling with hydraulic fracturing (HDHF) have occurred at much lower rates (less than 100 total publications per year; fewer than a few dozen on geochemistry alone). Nevertheless, great progress has been made to identify the principal effects associated with US domestic, unconventional energy extraction on natural systems (see Vengosh *et al.*, *Environmental Science & Technology*, 2014).[1] We convened this themed issue as co-guest editors in order to showcase some of those most recent advances on the environmental geochemistry and biology of hydraulic fracturing.

One of the drivers of recent discoveries in this area are novel analytical, genomic, and informatic tools to evaluate chemical and biological impacts associated with HDHF. These tools span from developments in chemical instrumentation and improved software for compound identification, to improved laboratory accounting of complex chemical mixtures and their interferences, to high-throughput nucleic acid sequencing, assembly, and availability of (meta)genomic datasets, and evaluation of large historic or geospatial datasets for impact or risk assessment. Ultimately, these approaches have been deployed to assess (a) the naturally-occurring and induced changes in the biology and chemistry of hydraulically fracturing systems, (b) the source and fate

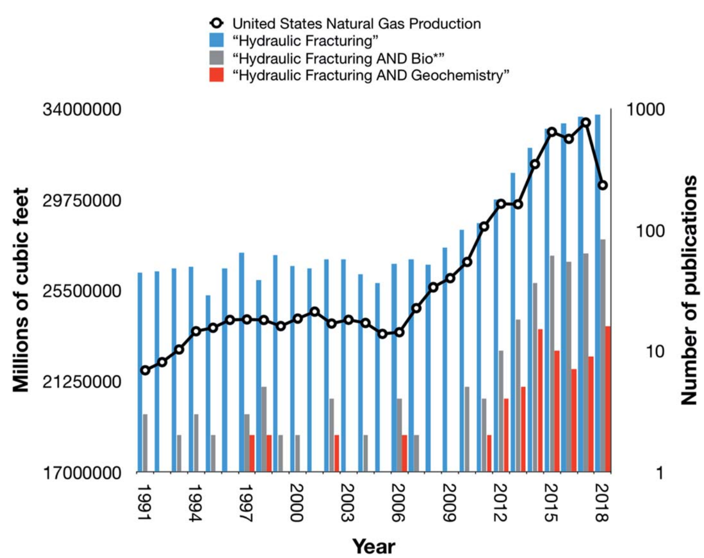

Fig. 1 The expansion of natural gas extraction and research in the United States and globally, respectively. Production data were obtained from the Energy Information Administration (eia.gov), where 2018 extraction data represent a partial year (January through October, 2018). Publication data were found using a Web of Science™ search and the search terms with Boolean operators are shown in the legend [all data accessed on December 31, 2018].

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of organic and inorganic constituents in flowback and produced waters (and treatments thereof), and (c) interactions between and among those components.

We encourage you to explore the details of the collection, and note that sweeping summaries regarding HDHF's influence on environmental processes and impacts should be avoided, in part, because the science in this field is still unfolding through longer-term studies and improved sample collection access. Defining the chemical class of interest (*e.g.*, hydrophobic organic, hydrophilic organic, inorganic, inorganic and radioactive, light gases or natural gas products, or biological constituents) and geophysical and biogeochemical parameters of the relevant environments will be critically important for framing such discussions. For example, in this issue, Rogers *et al.* (DOI: 10.1039/c8em00291f) demonstrated biodegradation of the commonly disclosed HDHF chemicals polyethylene glycol and polypropylene glycol and identified important co-occurring biological drivers (*i.e.*, an increase in primary alcohol dehydrogenase genes) in the lab, and Luek *et al.* (DOI: 10.1039/c8em00331a) see persistent influences of HDHF activities on organic sulfur composition 10 months after fracturing in the field. Interpretation of field results requires robust analytical methods (see comparison of methods used to study formaldehyde leachate from proppants by Schenk *et al.*, DOI: 10.1039/c8em00342d), as well as knowledge of analytical artifacts such as matrix effects, as nicely shown by Nell and Helbling (DOI: 10.1039/c8em00135a). Relatedly, Tasker *et al.* (DOI: 10.1039/c8em00359a) provide results of an ambitious laboratory intercomparison to evaluate analytical uncertainty among different labs with respect to inorganic chemicals and naturally occurring radioactive materials (NORMs). NORMs, in particular, have been a central focus in HDHF geochemical research, due to their human health implications, distribution in the environment as the result of management practices (McDevitt *et al.*, DOI: 10.1039/c8em00336j), and challenges and opportunities in waste management (Ouyang *et al.*, DOI: 10.1039/c8em00311d; Ajemigbitse *et al.*, DOI: 10.1039/c8em00248g). Interrogation of solids in flowback and produced water, especially as those return to the surface and become oxygenated (*e.g.*, on the way to and during treatment), was described by Flynn *et al.* (DOI: 10.1039/c8em00404h), identifying silica-enriched iron(III) oxyhydroxides and barite–celestine as dominant mineral phases.

Waste management implications for organic chemical constituents, such as dissolved organic carbon, were studied by Akyon *et al.* (DOI: 10.1039/c8em00354h), whose results for Utica and Bakken Shale residual fluids waters were largely consistent with previous work for simulated fluids (Kekacs *et al.*, *Biodegradation*, 2015),[2] demonstrating that the majority of, but not all, DOC is readily biodegraded in aerobic systems. Exploring the native microbial community and associated biomarker composition of the Marcellus Shale and overlying Mahantango Formation, Akondi *et al.* (DOI: 10.1039/c8em00444g) discovered a transition from ester-linked phospholipid fatty acids (PLFAs) to diglyceride fatty acids (DGFAs) going from the lower to higher permeability overlying formation, where the DGFA profile was consistent with physiological or nutrient-limitation stress exposure. The impact of chemical stressors (*e.g.*, known toxicants in HDHF additives) on microbiological make-up was reported by Santos *et al.* (DOI: 10.1039/c8em00338f), who observed an increase in the fatty acids that would act to reduce membrane permeability (*e.g.*, saturated/unsaturated ratio and higher branched and cyclopropane fatty acids) in the presence of toxic compounds.

A strong motivator for uncovering fundamental geochemical and biological processes is to enable prediction of risk or impacts associated with HDHF. To that end, Stringfellow and Camarillo (DOI: 10.1039/c8em00351c) explore HDHF chemical disclosure databases and Wen *et al.* (DOI: 10.1039/c8em00385h) evaluate groundwater quality data over 100 years in an area with rich conventional and unconventional gas development to determine what, if any, change in groundwater chemistry has occurred as a result of HDHF activities. This work showed that there were almost no statistically significant differences in groundwater quality metrics, except for small variations in road salt indicators (*e.g.*, Cl^-) attributed to surface activities. In absence of any substantial effects observed after 10 years of unconventional development, Wilson and co-workers (DOI: 10.1039/c8em00300a) present an intriguing approach, wherein one can use hydrological knowledge to predict regions of groundwater vulnerability (*i.e.*, likely regions of contamination in the event of spills or well failures) to improve risk assessment, especially for mobile, persistent chemicals (Rogers *et al.*, *Environmental Science & Technology*, 2015)[3] that may be released near the surface.

More than a decade of research, while notable on human or microbiological timescales, is relatively short from a hydrological perspective. Thus, while some critical questions have been addressed, many remain and may continue to emerge. Further, we note that unconventional shale gas and tight oil regions are heterogeneous, with variable geochemistries, microbial communities, hydraulic connectivities, and differential magnitudes of exploration. As HDHF practices are transferred more broadly to unconventional formations around the world, the analytical advances and fundamental, transferrable understanding described in this themed issue should aid and inform that development.

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