Environmental Science
Water Research & Technology

PAPER

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Cite this: *Environ. Sci.: Water Res. Technol.*, 2018, **4**, 325

Wastewater from hydraulic fracturing in the UK: assessing the viability and cost of management†

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The safe and effective management of wastewaters from unconventional hydrocarbon production using the hydraulic fracturing (fracking) process poses a major challenge. Exploitation of unconventional hydrocarbons, such as shale gas, remains controversial in the UK primarily due to concerns surrounding the hydraulic fracturing process required to extract the resource. The key issue of how waste fluids produced by hydraulic fracturing in the UK will be safely managed has yet to be adequately addressed, and the capacity for the specialist treatment required is currently uncertain. To address this critical knowledge gap we review, for the first time, the available management options for these waste fluids in the UK. We find that these are limited in comparison to the options available in the U.S., due to uncertainty surrounding whether wastewater injection wells will be permitted in the UK. Consequently, it is highly probable that these fluids will need to be treated and safely disposed of at the surface. In order to constrain the composition of wastewater which will require treatment in the UK, we analyse the only existing data set of returned waters from hydraulic fracturing (n = 31). We supplement this with measurements of wastewater from UK conventional onshore hydrocarbon (n = 3), and offshore hydrocarbon (n = 14), operations which produce water from similar formations as those currently targeted for shale gas exploration. Comparison of this limited UK data to the more extensive unconventional production dataset from the United States (n = 3092) provides confidence in our projected UK wastewater compositions. We find that the high level of salinity and concentration of naturally occurring radioactive material (NORM) in UK wastewaters will be problematic to treat for disposal into a freshwater environment. We use our data compilation to estimate costs of treating such wastewaters in a number of relevant scenarios. We find that the projected salinity in FP waters from UK hydraulic fracturing operations can be treated at a cost of between $2701 (~£2000) and $1 376 093 (~£1 047 000) per well, requiring between 2 and 26% of expected revenue. Additional costs, specific to the UK of up to £163 450 per well, will be incurred due to the legislative requirement for disposal of NORM concentrated sludge in permitted landfill sites. We find that existing capacity to receive NORM waste at currently permitted UK treatment facilities is limited, and that this will pose management problems if wastewaters are generated from multiple unconventional wells simultaneously.

Received 7th November 2017,
Accepted 27th November 2017

DOI: 10.1039/c7ew00474e

rsc.li/es-water

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Water impact

If poorly managed, wastewater generated during hydraulic fracturing for shale gas can pose a significant environmental threat. This research assesses the viability and cost of wastewater management options for the UK shale gas industry. Using data from the UK and the US, volumes and chemistries of wastewater to be managed are predicted, and assessed against current UK capacity.

Introduction

Hydrocarbon production from shale formations has become an increasingly prominent source of energy over the last decade, yet exploitation of the resource remains controversial. The majority of this controversy is due to concerns surrounding the hydraulic fracturing (fracking) process which is required to extract the hydrocarbons, and the management of resulting wastewater.[1–5] During this process, injected fluids consisting of 99.5% fresh water and proppant (to maintain fracture connectivity) and 0.5% chemical additives such as biocides, surfactants, viscosity adjusters, cross-linkers, breakers, corrosion inhibitors, bactericide, and friction reducers[6] react with the freshly fractured and exposed minerals, and mix with the formation fluids within the shale rocks being targeted. On de-pressurisation of the well

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† Electronic supplementary information (ESI) available. See DOI: 10.1039/c7ew00474e

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following the fracturing process, these fluids are returned to the surface having inherited heavy metals, naturally occurring radioactive material (NORM), salts and hydrocarbons from interaction with the rocks and fluids at depth. The chemistry of fluids returned is determined by the chemistry of fracturing fluid, geochemistry of the rock, and the fluid chemistry of any formation waters residing in the pore space,[7] with potential influence from the chemistry of residual drilling fluid. Volumes of fluid returned vary depending on (1) the volume of fluid injected, (2) local geological characteristics that control water retention in the subsurface such as imbibition and extent of fracture network[8] and (3) well length.[9] We collectively define and refer to these fluids as flowback and produced water (FP water), after Nicot *et al.* 2014.[10]

FP waters generated by unconventional hydrocarbon operations – such as hydraulic fracturing or coal bed methane extraction – require appropriate management to ensure that they are stored, transported, treated and disposed of in a way that minimises the operational need for fresh water, maximises the efficiency and cost of the processes and reduces risks posed to the environment. The expected volumes and chemistries of FP water, and the capacity for their treatment[11] or disposal in the UK is poorly understood, as outlined in a recent joint report from the Natural Environment Research Council (NERC), National Science Foundation (NSF), and Royal Society of Chemistry (RSC).[12] This report is the only literature published in the public domain that covers the management of FP waters from shale gas extraction in the UK to date, and concludes that the potential cost of treatment of FP waters could render extraction of shale gas reserves in the UK uneconomic.[12] However, this conclusion was based on a limited review of available data that made no comprehensive assessment of UK FP water management options. To address this critical knowledge gap we review the available options for management of these wastes in the UK.

Options for FP water management in the UK

FP water can be disposed of through injection into geological formations, such as depleted oil and gas reservoirs, or saline aquifer bodies.[6,13,14] This relies on the geological suitability and capacity for subsurface storage and cannot be uniformly depended upon.[15,16] Currently, concerns exist over the suitability of underground disposal for the UK, due to observed increases in induced and triggered seismicity across the U.S. (where injected fluids lubricate critically stressed fault systems or high injection rates are used[17–19]) and evidence of surface water degradation within the vicinity of injection sites.[20] Through examination of publically available data, we have established that 99.9% of the produced water generated by conventional onshore oil and gas operations in the UK is reinjected back into the formation from which it was recovered.[21] This practice requires no interim treatment as the FP water composition is similar to that of the formation water. Hence, there is currently no precedent for the treatment of large volumes of FP water that could result from extensive shale gas exploitation in the UK.

Reinjection of wastewater is frequent practice for enhancing hydrocarbon recovery and providing pressure support for depleted conventional reservoirs.[13] Between 20 and 25% of water produced from conventional operations in the UK sector of the North Sea is reinjected for this purpose.[22] However, in the case of unconventional operations, the storage space required does not exist within the low porosity, and relatively impermeable, shale rocks to allow the reinjection of FP waters. Disposal of these waters by injection into the subsurface would require the existence of additional geological storage space with specific characteristics, such as sealing cap rocks and lithological heterogeneity. No comprehensive assessment of such sites in the UK has yet been undertaken, and combined with public concerns around such disposal, the viability of injection of FP water into the subsurface for disposal is uncertain.

Without prior treatment or separation, FP water disposal through underground injection also confines large volume of fresh water used in the fracturing process to waste, which will significantly increase the amount required for UK shale gas operations. Further, the disposal of liquid wastes into landfill sites is currently prohibited in England and Wales by The Landfill (England and Wales) Regulations 2002, as result of the EU Directive 99/31/EC on the landfill of waste (Landfill Directive) which also applies to Scotland and Northern Ireland. Whilst the Environment Agency (England and Wales) and Scottish Environmental Protection Agency (SEPA) will classify the FP water produced by the hydraulic fracturing process as mining waste,[11] the legality of permitting this to be disposed of through deep injection *via* disposal wells has yet to be established.[23,24]

Reuse of FP waters in subsequent hydraulic fracturing operations (internal reuse) is frequent practice in the in the Marcellus Shale region of the U.S., where up to 90% of produced waters were reused in 2012. However reuse values vary significantly by region with only 5–10% reuse in the Barnett Shale region.[25–27] Reuse can reduce fresh water consumption, but can lead to concentration of contaminants by repetitive exposure of the waters to fresh rock surfaces and multiple stages of mixing with fracturing additives.[28] Operators opting for internal reuse in the U.S. have observed reduced well clean out times as untreated FP waters can reach chemical equilibrium with the rock formation considerably faster than fresh waters.[29] Although internal reuse can eradicate the need for treatment to discharge standards in the interim, most operators opt for some degree of water treatment to reduce risk of reduced hydrocarbon production[9] and scaling of infrastructure.[30] These factors, combined with the financial incentive proven in the U.S.[31] strongly imply that even with internal reuse in place, a degree of FP treatment will be required in the UK.

FP waters can be thoroughly treated to discharge standards and be directly discharged to a surface water body such

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as river, estuary or canal. Discharge of FP waters to surface water requires the waters to be treated to an environmentally acceptable level, guided by the Water Framework Directive (2000/60/EC) before they can be released to the environment, (S.I. 1†). This will require specialist treatment focusing on removal of the most concentrated, and highest risk contaminants. Treatment can be performed on-site, using mobile treatment technology, or off-site *via* transport to a specialist treatment works.

Treatment of salinity

FP waters typically contain levels of total dissolved solids (TDS) ranging from 50 000–250 000 mg L^{-1},[32] meaning that 5–25% of the mass of every litre of FP water is comprised of contaminants in solution. These dissolved solids are predominantly sodium and chloride salts and hence *TDS* is commonly referred to as *salinity*.[33] Current UK regulations for disposal to freshwater water courses require the salinity of FP waters to be reduced to a maximum concentration of 200 mg L^{-1} Na, and 250 mg L^{-1} Cl (S.I. 1†), equating to a 99.5–99.9% reduction in concentration.

Treatments to reduce or remove TDS from FP waters include distillation, and reverse osmosis.[13] Distillation, evaporation, or compression use heat to evaporate the contaminated water in an enclosed environment whereby the potable water re-condenses, leaving the contaminants concentrated within the residue. Reverse osmosis utilises fluid pressure differences to pass FP water through a membrane filter removing some contaminants. Reverse osmosis is more cost effective and less energy intensive than distillation, however its application is limited to waters with TDS below 50 000 mg L^{-1}, restricting its suitability to treat most FP waters, including the ones considered in this study. Additionally, osmotic membranes can require frequent maintenance and replacement due to fouling and permeability reduction.[3]

The unrecoverable waste, or sludge, becomes concentrated with contaminants not targeted by this treatment such as heavy metals or naturally occurring radioactive materials and will require additional focused treatment or permanent disposal in permitted landfill.

Treatment of naturally occurring radioactive materials (NORM)

Rocks rich in organic matter, such as gas-bearing shales or other hydrocarbon source rocks, contain naturally occurring radionuclides, including uranium (^{238}U) and thorium (^{232}Th). These radionuclides decay to form a number of daughter particles, notably radium (Ra^{226}), which is particularly soluble[34] and therefore partial to leaching by formation or injected waters. During hydraulic fracturing the injected fluids mix with the radium enriched formation waters within the rock, and subsequently contaminate the FP water with naturally occurring radioactive material (NORM). The decay of radionuclides causes radiation to be emitted prompting negative health impacts at critical doses,[28] so monitoring and appropriate control of NORM in waste streams is extremely important.

NORM contaminated waste streams generated by the conventional hydrocarbon industry can be re-injected into the source formation onshore or offshore to provide pressure support to enhance reservoir recovery, or can be diluted and dispersed offshore into the marine environment within set limits.[35,36] As previously mentioned, the capacity and legality of re-injection of FP waters in the UK is uncertain. Additionally, the dilution and dispersion of pre-treated FP waters high in NORMs directly to the offshore environment will not be feasible due to limitations on marine contamination enforced by The OSPAR Treaty (2007) as outlined in Section B.44, UK NORM Waste Management Strategy.[37]

Concentrations of radionuclides in unconventional waste streams are expected to be 1.5 times higher than in conventional hydrocarbon waste streams as a result of the direct contact of the waters with the radionuclide-rich hydrocarbon bearing shale rocks.[38] NORM levels will become concentrated within precipitates that form during evaporation treatment and in the sludge collected by filtration, sedimentation, or separation. The disposal of this precipitate is subject to the Radioactive Substances Regulations (2011) conditions of low-level radioactive waste only within an appropriately permitted landfill. The UK Department for Energy and Climate Change (DECC) highlighted a lack of capacity to treat NORM-contaminated FP water in their Strategic Environmental Assessment for Further Onshore Oil and Gas Licensing (2013).

Treatment of heavy metals

Human exposure to heavy metals in water can cause serious negative health effects, and therefore regulations exist to minimise exposure from treated and discharged wastewaters.[39] A review of research on wastewater treatment for heavy metals by Fu & Wang (2011) identifies seven key treatment methods; (1) chemical precipitation, (2) ion-exchange, (3) adsorption, (4) filtration, (5) coagulation & flocculation, (6) flotation, and (7) electrochemical treatment.

Chemical precipitation (1) utilises chemicals to cause metal ions to precipitate out of solution which can then be removed by filtration or sedimentation.[40] Ion-exchange (2), specifically targets the metals required for removal with exchangeable cations.[41] Adsorption (3) removes contaminants of concern by collecting them on an adsorbent surface creating adsorbate film.[40] Filtration (4) utilises a barrier screen designed to trap or block any particulate material from passing through with the treated effluent. Coagulation and flocculation (5) occurs by a flaking agent causing particles to coagulate that can then be sifted or filtered out. Flotation (6) brings the contaminants to the surface of the fluid by attachment to air bubbles. Electrochemical treatment (EC) (7), alters the surface charge on contaminant particles held in the water causing them to separate and accumulate in an easily removable mass.

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A comprehensive study of the performance of UK wastewater treatment plants[42] highlighted a wide variation in removal rates of between 25–79% for heavy metals. This variation was attributed to variations in composition of wastewater being treated and the operation parameters at individual treatment works.[42] Removal of trace metals dissolved in solution currently pose the most significant challenge to the industry in comparison to other trace contaminants, so it will be pertinent to consider the contribution shale gas operations may make to the waste load requiring treatment in future work. However, in this work, due the limited dataset available from UK operations at present, we focus on assessing options for treatment of the two major problem contaminants in UK FP waters; salinity and NORM.

Methods

Our review of the options for FP water management in the UK highlights that some degree of water treatment will be required following hydraulic fracturing. Hence, we consider the costs associated with this treatment, and estimate the capacity within existing UK facilities to receive and treat such wastes.

Constraining expected volumes of UK FP water

The volume of fluid injected to undertake the hydraulic fracturing process depends on; the depth and length of well to be fractured, the number and length of stages to be perforated, the properties of the fracturing fluid, and the geological characteristics of the formation.[43,44] Concurrently, volumes of FP water produced vary according to the volume of fluid injected, local geological characteristics that control water retention in the subsurface such as imbibition and extent of fracture network,[8] and the well length.[9] To date, only one well, targeting the Carboniferous Bowland Shale Formation, located at Preese Hall in Lancashire, England, has been subjected to high volume hydraulic fracturing in the UK. In 2011, Cuadrilla Resources injected 8399 m^3 of fluids, and perforated six fracturing stages in this well, before operations ceased and approximately 8000 m^3 of FP water was produced at the surface.

As this operation provides the only FP water volume datum for the UK at present, additional estimates of the likely volume of FP water returned to the surface can be made using injection volume estimates from the British Geological Survey and published ranges of percentages of injected fluid returned by the in the following eqn (1) (S.I.4 & 5†).

$$V_r = V_i \times P_r \quad (1)$$

where V_r is volume of wastewater returned (m^3), V_i is the volume of fluid injected (m^3), and P_r is the percentage of fluid typically returned.

Calculating costs of FP water treatment

The cost of FP water treatment can be determined by the following equation (adapted from Webb & Woodfield, 1981):

$$C = R + VP + \left(\frac{St}{Ss} \times S\right) \quad (2)$$

where; C is the total cost to the operator, R is the cost of receiving the waste at the facility in question including storage and transport costs, V is the volumetric cost of waste treatment imposed by the plant, P is the cost per volume of the primary treatment applied to the waste, St is the Total Suspended Solid (TSS) concentration of the waste, Ss is the Total Dissolved Solid (TDS) concentration of the waste, and S is the cost of the treatment of solids.

To forecast the cost of treatment of wastewater (C), the volume of waste to be treated (V) and the chemistry, or solid salinity, (St, Ss) must be determined and combined with the cost of auxiliary treatment (R, P, S).

We apply the results of the volume calculation (S.I.2†) to a simplified cost projection eqn (3) to identify the expected range in cost of treatment for our simulated range of FP water volumes. To provide a contextual and holistic estimate of the likely cost of treatment, published costs (C_{tr}) for reverse osmosis (RO) and distillation by mechanical vapour compression (MVC), were used[45] (S.I.3†). These costs were supplemented with a cost range provided by a specialist treatment plant in Pennsylvania, U.S. which uses a combination of chemical adjustment, filtration, settling, and distillation to produce dischargeable and re-usable effluent.[46]

A Monte Carlo simulation (n = 10 000) was performed to estimate the range in total cost likely to arise from changes in expected injected and returned volumes of fluid, and to estimate the sensitivity of each parameter within the calculation to the total cost of treatment. The input parameters for Monte Carlo simulations are summarised in Fig. 1, and provided in full in S.I.2.†

$$C_{to} = V_i \times P_r \times C_{tr} \quad (3)$$

where C_{to} is the total cost of wastewater treatment, V_i is the volume of fluid injected (m^3), P_r is the percentage of fluid typically returned, and C_{tr} is the cost of treatment per m^3.

Assessing the capacity for FP water treatment in the UK

Non-specialist public treatment works in the UK are unable to treat highly saline and NORM contaminated waters.[16,47] Hence, the removal of salinity (TDS) and NORM can only be undertaken at specially permitted treatment facilities.[37] There are currently four treatment facilities in the UK that are appropriately permitted to handle liquid waste containing

		Minimum	Maximum	Source
$V_{injected}$		7,000m^3	18,000m^3	See S.I.4
$P_{returned}$		10%	70%	See S.I.5
$C_{treatment}$	RO	\$0.50/m^3	\$12.33/m^3	See S.I.3.1
	MVC	\$0.47/m^3	\$25.16/m^3	See S.I.3.1
	CBD	\$50.30/m^3	\$164.50/m^3	Eureka Resources [46]

Fig. 1 Table of end-member variables used for Monte Carlo simulations, see ESI† for more detail.

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NORM.[48–51] Publically available permitting documents for these four sites provide information on the environmentally agreed limits for accumulation and disposal of aqueous radioactive waste on a daily, monthly and annual basis. Amalgamating these provides the total daily accumulation and disposal limits for the UK (correct as of November 2017). Using the measured radioactivity in FP waters from the 2011 hydraulic fracturing at Preese Hall and the projected volumes of FP water, we estimate the range in radioactivity expected to be received and disposed of by the sites listed (eqn (4)).

$$A_p = A_m \times V_p \quad (4)$$

where A_p is the projected total radioactivity (Bq), A_m is the measured radioactivity of FP waters (Bq L^{-1}), and V_p is the projected volume of FP waters (m^3).

Results

Volumes of FP water

The British Geological Survey estimate between 7000 m^3 and 18 000 m^3 of injection fluid will be required for each UK hydraulic fracturing operation[52] (S.I.4†). The American Petroleum Institute cite that between 10% and 70% of this fluid typically returns to the surface[53] (S.I.5†). Using eqn (1) with the bounding values listed in Fig. 1, allows calculation of expected return water volumes of between 700 m^3 and 4900 m^3, if 7000 m^3 is injected. If the volume injected is nearer to the upper bound of 18 000 m^3, then the range of returned waters increases to 1800–12 600 m^3. These do not represent exhaustive volume limits, as corroborated by the wider range of volumes recorded in the U.S.,[43] but provide reasonable estimates within the confines of existing data for the UK.

To improve upon this volume estimate, a Monte Carlo analysis (n = 10 000) was performed on this volume projection to determine the 95% confidence intervals, given a uniform probability distribution assumed between the BGS end-member values. These simulations determine that in 95% of cases the FP water volume is likely to be between 1253 m^3 and 10 544 m^3, and only in 1% of cases will the FP water volume exceed 12 224 m^3. The volume of water retained by the formation could vary from 2227 m^3 to 12 754 m^3 under these injection volume scenarios.

In comparison, the return of 8000 m^3 fluids from the 8399 m^3 injected by Cuadrilla Resources in the only hydraulic fracturing of a shale formation in the UK to date, highlights that 95% of the injected fluids returned to the surface. However, this well was not completed and produced in the planned fashion, with no shut-in period due to the triggered seismic events, and hence these ratios are not expected to be representative and are not used in the modelled volumes of FP water. The fluid volume injected at Preese Hall is well within the 5th and 95th percentile volumes of fluids injected for hydraulic fracturing in 38 530 wells in the U.S. between 1st January 2011 and 28th February 2013 which range from 135 m^3 to 32 700 m^3 respectively, with a median volume of 6800 m^3.[43] However, these data include volumes for fractured vertical wells and coal bed methane operations, which typically use significantly less fluids. A higher and more concise volume range would be expected should the hydraulic fracturing of only high volume, horizontally drilled wells be considered, but unfortunately, the U.S. data sampled is not reported by well type.

Cost of FP water treatment

The expense and energy intensity of FP water treatment is dependent on the composition of the water to be treated as defined by eqn (2). The minimum, mean, and maximum, levels of dissolved solids measured in the FP waters collected by Cuadrilla for treatment, at Preese Hall were; 94 000 mg L^{-1}, 128 750 mg L^{-1} and; 210 000 mg L^{-1},[54] respectively. Whilst these data are limited by being from a single well, this provides the best estimates of future compositions of UK FP waters currently available. Confidence in this salinity range is provided from examination of data from offshore operations that produce waters from wells drilled to the Bowland Shale or underlying Visean limestone formations. The salinities of these waters range from 164 460 mg L^{-1} to 398 240 mg L^{-1},[55] indicating that higher salinities may be possible from fracturing of other areas of the Bowland Shale. These offshore produced waters are typically discharged directly to the sea, and are only treated to separate co-produced hydrocarbons to the limit of 30 mg L^{-1}.[56]

Additionally, produced waters from conventional onshore production at Kirby Misperton in Yorkshire, England from the Permian Kirkham Abbey and Permo-Triassic Sherwood Sandstone Formations overlying the Carboniferous Bowland Shale exhibit a range of salinities from 180 000 mg L^{-1} to 349 000 mg L^{-1}.[57] These comparisons provide confidence that the salinity levels measured at Preese Hall are comparable, on average lower, than salinities measured from fluid produced during offshore Carboniferous production and onshore Permo-Triassic production as outlined in Fig. 2.

The cost of treatment is dependent on feedwater chemistry (eqn (2)) meaning estimates of overall cost are limited by the assumption that FP water chemistry will not change throughout the life of the well. However, salinity is known to vary significantly within the first weeks of production and then increase with time as fluids are produced.[58] Measured TDS values in FP waters from the U.S. range from 35 to 358 000 mg L^{-1},[59] so scenarios beyond those considered in this analysis are possible. However, this is the best data available at present, until further UK hydraulic fracturing operations are undertaken and can easily be adapted when additional FP water chemistry data becomes available.

Using the data outlined, a Monte Carlo simulation was performed to predict the range of costs that will be incurred by operators for treatment of FP waters from a single UK well using eqn (3) (S.I.2.4†). The calculated costs range from $553 (low volume reverse osmosis for <50 000 mg L^{-1} TDS waters) up to $2 023 797 (high volume combined treatment techniques of high TDS waters) per well, with a 95% confidence interval of $2701–1 376 093. Only in 1% of cases where combined treatment techniques are used will the cost exceed $1 819 846 per well. The method of treatment and volume of

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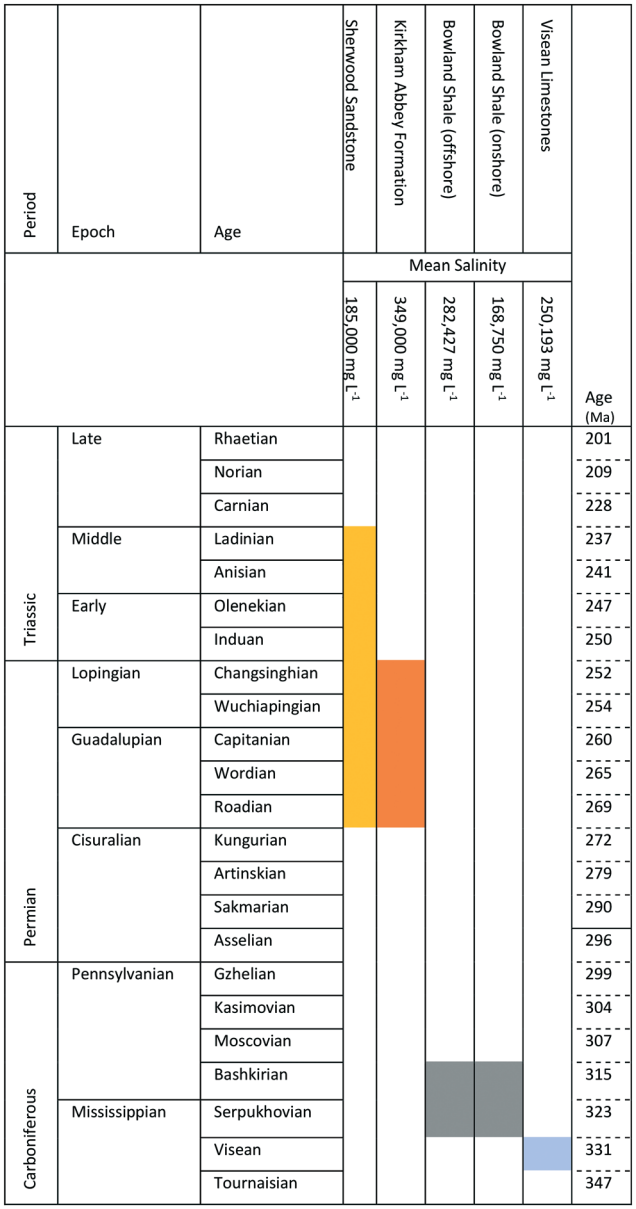

Fig. 2 Stratigraphic chart annotated with mean salinities (mg L^{-1}) of FP and produced water data by source formation for the UK.

water to be treated play a significant role in determining the overall cost of treatment. The variability in cost of treatment projected is greatest when the volumes of water produced are the highest. The range in cost of the treatment techniques considered is inherently linked to the energy requirements of those techniques. FP water containing a higher concentration of dissolved solids requires more energy to treat (more pressure for osmosis and greater heat for distillation) and therefore is more costly per unit. The range of costs applied in this analysis show the variability in treatment cost due to range in chemistry of feedwater and processes utilised for treatment (S.I.3.1†).

NORM are removed from FP waters during treatment by mechanical vapour compression (MVC) when radioactive nuclides are precipitated within the sludge generated as a by-product. Consequently, any sludge produced during MVC of FP waters is contaminated with NORM and must be disposed of to a Radioactive Substances Regulations (RSR) permitted landfill site or to the LLW (Low Level Waste) Repository, in Cumbria. The cost of landfill disposal in the UK (gate fee plus landfill tax) per tonne varied from £89–135, with a median of £100 per tonne, in 2014/15.[60] Due to the cost incurred by obtaining a RSR permit to receive LLW[61] there is likely to be an additional charge for NORM contaminated waste corresponding to the radioactivity level of the waste. The LLW Repository pricing structure declares that the price for receipt of waste is based on "weight, volume, material type, radioactivity levels, hazardous content, packaging requirements, transportation mode and location". They estimate a charge to the operator of £500 per m^3 for very low level radioactive waste (VLLW, <4 MBq per tonne), and £3038 per m^3 for low level waste (LLW, <4 GBq per tonne alpha & <12 GBq per tonne beta or gamma), with an additional charge per mega-Becquerel of £9 for U^{238}, and £55 for Th^{232}, including Ra^{226}.[62] Additional costs will be incurred due to the need to transport the NORM contaminated sludge for disposal offsite.

Capacity for NORM treatment in the UK

Collation of publically available data[48–51] allows calculation of a total daily accumulation limit for NORM in the UK to be 1.92×10^8 Bq for the ^{238}U group, and 3.82×10^7 Bq for the ^{232}Th group (Fig. 3). The total daily disposal limits are 1.15×10^7 Bq for the ^{238}U group, and 4.09×10^7 Bq for the ^{232}Th group. These limits apply to the parent radionuclides and groups of daughter radionuclides within the same decay chain. Although there are no limits placed upon the volume of treated waste discharged from these plants, there are volumetric limits imposed upon the quantity of waste containing NORM that can be received per day, equating to 826 m^3 or (826 000 L) per day. The maximum radioactivity per volume of waste that can therefore be received is 232 Bq L^{-1} for the ^{238}U group and 46 Bq L^{-1} for the ^{232}Th group.

The maximum activity of ^{226}Ra (^{238}U group) recorded in FP waters from hydraulic fracturing in the UK is 90 Bq L^{-1} (day 158), with activities of 14, 6, and 17 Bq L^{-1} measured 0, 50, and 70 days respectively from initial flowback. As these values are only from one fracturing event they should not be considered as exhaustive or necessarily representative figures for the UK.[37] However, the recorded ranges in activity, and their increase in time as FP water is generated at the surface, are consistent with other studies of NORM in FP waters.[34] ^{226}Ra levels of up to 626 Bq L^{-1} have been recorded in U.S. unconventional FP waters,[59] with a median value of 39 Bq L^{-1}. U.S. conventional hydrocarbon produced waters have been reported to have activities of up to 196 Bq L^{-1}, with a median of 12 Bq L^{-1}.[59] Comparatively, the activity of from offshore disposal of produced water from conventional oil and gas operations to the UK sector of the North Sea from 2005–2012 was 5.9×10^8 and 13.3×10^8 Bq per day, for the ^{238}U and ^{232}Th group, respectively.

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Facility Name	Operator	Location	Permit No	Radionuclide or Group of Radionuclides	Accumulation Limit (Bq/day)	Disposal Limit (Bq/day)
FCC Knostrop	FCC Environment (FCC Recycling UK Ltd)	Leeds	BB3538DH	^{238}U	1.54E+07	5.33E+06
				^{232}Th	1.79E+07	4.73E+06
Bran Sands	Northumbrian Water Ltd.	Middlesbrough	PB3438DJ	^{238}U	1.50E+08	3.33E+06
				^{232}Th	1.50E+07	3.33E+07
Castle Environmental	Castle Oils Ltd.	Stoke-on-Trent	QB3339DQ	^{226}Ra	2.44E+07	8.22E+04
				^{228}Ra	3.33E+06	6.03E+05
FCC Ecclesfield	FCC Environment (FCC Recycling UK Ltd)	Sheffield	ZB3395DX	^{238}U	2.57E+06	2.74E+06
				^{232}Th	2.00E+06	2.19E+06
TOTAL				**^{238}U**	**1.92E+08**	**1.15E+07**
				^{232}Th	**3.82E+07**	**4.09E+07**

Fig. 3 Waste treatment facilities in the UK, and their permitted radioactivity accumulation and disposal limits as outlined in their environmental permits by the Environment Agency.

Using the activity data from the Preese Hall well, a Monte Carlo simulation was used to predict the likely range of activities of FP waters using eqn (4). The results indicate that the total ^{226}Ra activity per hydraulically fractured well in the UK will range from 9.5×10^4 Bq to 1.3×10^6 Bq in 95% of cases, and will only exceed 1.5×10^6 Bq in 1% of cases. These values lie within the current calculated treatment capacity for the ^{238}U group, though only when considering the projected activity for the parent radionuclide. Constraining the implications of multiple daughter radionuclide groups on the accumulation and disposal capacity is not possible at present due to the limited data available. These results show that it is unlikely that FP waters produced from a single well will cause significant stress to the existing treatment facilities, provided they are divided between all available treatment plants. However, the uncertainty surrounding the volume of waste generated during fracturing could pose a threat to the capacity to treat NORM in the medium to longer term.

The Environment Agency enforce volumetric as well as activity limits on the volume of aqueous waste containing NORM that can be received at available treatment sites, currently amounting to 825 m^3 per day across the UK. If the volume of FP water produced during fracturing exceeds this the capacity of the available treatment facilities could become critically stressed. Hence, without alternate storage options or emergency treatment capacity, operations would be forced to cease until the fluids can be appropriately handled.

Implications for FP treatment in the UK

Cost of salinity treatment

The values of salinity measured in FP waters from the hydraulic fracturing of the Bowland Shale in the UK range from 98 000 to 210 000 mg L^{-1}. Salinity of FP water can vary geographically within a single shale basin due to lithological heterogeneity and distribution of relatively saline horizons.[34,63] However, these values lie well within the reported salinity ranges observed in FP waters from unconventional production in the Haynesville, Marcellus and Barnett shale plays of the U.S. (35–358 000 mg L^{-1}). Additionally, they are lower than measured salinities of produced water from conventional hydrocarbon extraction onshore (180 000–349 000 mg L^{-1}) and offshore (164 000–398 240 mg L^{-1}) in similar formations in the UK (Fig. 4).

It is clear that FP water with higher levels of contaminants, including TDS, will be more costly and energy intensive to treat. Waters with TDS levels above 50 000 mg L^{-1} can only be treated by distillation (MVC), as filtration and osmosis has been proven to be ineffective at high salinities.[64]

The 95% confidence range of estimated costs of desalination, per well, for FP waters from fracturing of the Bowland Shale in the UK by MVC range from $3952 to $196 484. Disposal of desalination by-products such as sludge contaminated with NORM will significantly increase the cost projections. The U.S. Energy Information Administration calculate that the mean estimated ultimate recovery (EUR) from shale gas wells is 1.8×10^6 MMBtu.[65] With the current price for natural gas at $2.94 per MMBtu (correct as of November 2017), each well can be estimated to generate on average $5 292 000 in revenue. Therefore, up to 3.7% of the total revenue from a single shale gas well could be absorbed by desalinising treatment costs alone. Variations in the price of natural gas can cause the percentage of expected revenue required for FP water treatment by MVC to vary from 2% up to 5%.

Additionally, if multiple treatment processes are required to target multiple contaminants within the waste, the cost can increase significantly. Eureka Resources, a specialised FP water treatment company based in Williamsport (PA), typically charge $50.30–164.50 per m^3 to treat FP water at their designated facility.[46] If these prices are to reflect the total

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	UK			USA	
Total Dissolved Solids (mg L^{-1})	**Offshore Conventional**	**Onshore Conventional**	**Onshore Unconventional**	**Onshore Conventional**	**Onshore Unconventional**
Sample size	*n=14*	*n=3*	*n=24*	*n=88 261*	*n=174*
Minimum	164 460	180 000	**94 000**	0.1	**35**
Maximum	398 240	349 000	**210 000**	528 700	**358 000**
Mean	273 217	239 666	**168 750**	83 490	**65 120**
Median	280 430	190 000	**180 000**	51 220	**24 700**
		99% confidence interval in min-max to correct for outliers		156 – 392 963	**37 – 355 751**

Fig. 4 Table of summary statistics for comparable data sets of salinity measured in onshore and offshore operations in the UK and USA.

combined treatment cost in the future for the UK, between \$107 683 to \$1 376 093 (95% CL), or up to 26% of the estimated revenue per well would be required. The U.S. has also benefitted from the use of portable treatment facilities that reduce salinity and remediate against some contaminants on-site before re-use. These reduce the transport requirements associated with off-site waste treatment, but increase the likelihood of onsite spills as a result of increased on site waste handling.

Cost of naturally occurring radioactive material treatment

Aqueous NORM waste in the UK is treated by following the principal of 'dilute and disperse' within the sewerage and water treatment system.[37] Given that underground disposal of FP waters has been discounted as an option for the UK to date, treatment for re-use or effluent discharge will be required.

It is also pertinent to consider the volume of solid or sludge NORM waste generated as a by-product. Radium, the most common NORM in FP water, is chemically similar to other alkaline earth elements such as magnesium (Mg), barium (Ba), strontium (Sr), and calcium (Ca) and so readily co-precipitates generating NORM concentrations in scale and sludge produced during MVC treatment. This sludge by-product will require disposal to landfill with the appropriate radioactivity permits unless exempt due to low activity by the Radioactive Substances Regulations (2011).

Within the context of eqn (2), the S value for FP water containing NORM will be inflated by the cost of obtaining and maintaining a RSR permit, with application fees for 2017 ranging from £980–2640, and subsistence costs ranging from £154 to £3940 per year depending on the conditions and nature of the mining waste activity proposed.[61] Additionally, disposal of these sludge wastes at landfill will incur a 'gate fee' and landfill tax, between £89–135 per tonne in 2014/15.[60] Utilising the known salinity of waters returned at Preese Hall, the total mass of solids available for removal from waters returned during fracturing of a single well can be projected to be between 10 and 3269 kg (53–2053 kg, 95% CL). Between 1 and 99% of the volume of sludge is comprised of residual wastewater[66] and therefore the total volume to be disposed of varies in proportion to the percentage water content. Dewatering processes significantly reduce the volume of waste to be disposed,[66] but if 100% recovery of solids is assumed and no dewatering performed the volume of sludge to be disposed varies from 10–329 600 kg (0.01–326.9 t). This equates to an additional cost of £1.00–326.90 in regular landfill gate fees per well, or up to £163 450 (£500 per m^3 of sludge) at the LLW Repository exclusive of permitting levies for the cost incurred in obtaining an RSR permit, transportation costs, and radioactivity charges such as those imposed at the LLW Repository.[62] The maximum projected disposal cost of NORM waste of £163 450 equates to an additional 3% of the estimated overall revenue generated from a single well. Therefore under these scenarios is unlikely that the additional costs of disposing this low level radioactive sludge will render unconventional extraction uneconomic, however the issue of available capacity remains pertinent.

Lessons for the UK on FP water management from the U.S.

A number of the FP water management options practised in the U.S. have caused surface contamination, which has impacted the ecology and environment. It is crucial that the UK learns from the mistakes and subsequent regulatory improvements made in the U.S. to prevent similar problems occurring in the UK.

Treating waters returned by hydraulic fracturing operations at centralised or municipal treatment works in the U.S. has led to increased concentrations of contaminants such as TDS, bromide, and chloride in the receiving waters due to the incompatibility of the treatment facilities with the waters concerned.[4,67,68] Inadequate treatment of these waters has also been shown to reduce water quality downstream from treatment works.[16,68–71] Consequently, treatment of water in public or municipal works has since been banned by the U.S. Environmental Protection Agency, and treatment may now only be performed by specialist or designated treatment works.[72]

Leaks and spills associated with transport and storage of these waters can also cause detriment to water quality, as well as due to casing failure or poor well integrity at deep injection sites.[73] Containment of waters within unlined surface

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ponds or impounds can allow leaching into the water table and contamination of nearby ground and surface waters.[74] However, this practice will not be permitted in the UK,[75] and regulatory changes in some states in the U.S. mean that full lining of any hydraulic fracturing site is now required, which combined with a means of secondary containment of FP water has significantly reduced contamination risks.[76]

Conclusions & recommendations

We find that disposal options in the UK are limited by current restrictions on the underground injection of waste. Hence it is certain that some form of treatment will be required before waters will be re-usable, either for hydraulic fracturing operations or externally, or for safe to discharge to the environment. We find that whilst the salinity and NORM levels in future FP waters from UK hydraulic fracturing operations can be treated, this will cost between $2701 and $1 376 093 per well for salinity. This will require up to 26% of the revenue generated by a typical shale gas well, and up to £163 450 (3% of revenue) for NORM management.

We have found that there is currently limited treatment capability for receiving returned waters from unconventional gas operations in the UK that are contaminated with NORM, and no uniquely dedicated treatment plants currently exist. As the UK shale gas industry is in its infancy, the limited treatment capacity for returned waters should not pose a problem in the short term, unless especially high volumes of FP water are experienced that cannot be received at the currently permitted facilities.

However, significant expansion of the shale gas industry resulting in simultaneous FP water production from multiple wells would critically stress the current capacity to receive, treat and dispose of NORM contaminated, highly-saline wastewaters. We strongly recommend that this area receives further attention from the emergent unconventional gas industry, the established waste water management industry and regulatory bodies in the UK, in order to produce a coherent strategy for the for the future management of FP waters. Our work has found that no such co-ordinated strategy currently exists, and limited FP management capacity in the UK will present a significant hurdle to future expansion of the industry unless it is urgently addressed.

We advise that future shale gas activities in the UK make their returned water composition data publically available so as to improve upon the estimates presented in this study. We further recommend that future treatment options are reviewed in light of new data once it becomes available.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This work was completed as part of the FracRisk Project funded by the European Union's Horizon 2020 Research and Innovation programme under Grant Agreement No. 636811. The authors would like to thank Dr. Mark Naylor for his help with conducting Monte Carlo analyses presented in this paper.

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