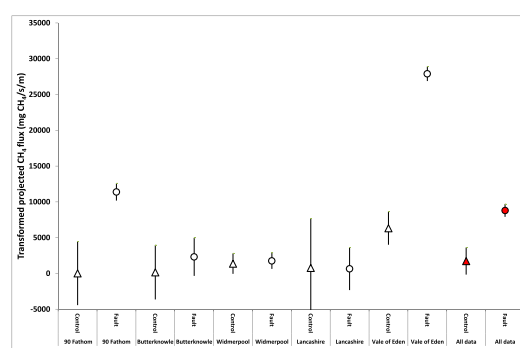


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GRAPHICAL ABSTRACT

- Fugitive emissions of CH₄ from basin-bounding faults in the UK
- Fault surveys had a significantly higher CH₄ flux than control surveys.
- No apparent link in CH₄ flux to presence or absence of hydrocarbons.
- Estimated flux from faults 11.5 ± 6.3 t CH₄/km²/yr



ABSTRACT

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This study considered whether faults bounding hydrocarbon-bearing basins could be conduits for methane release to the atmosphere. Five basin bounding faults in the UK were considered: two which bounded potential shale gas basins; two faults that bounded coal basins; and one that bounded a basin with no known hydrocarbon deposits. In each basin, two mobile methane surveys were conducted, one along the surface expression of the basin bounding fault and one along a line of similar length but not intersecting the fault. All survey data was corrected for wind direction, the ambient CH₄ concentration and the distance to the possible source. The survey design allowed for Analysis of Variance and this showed that there was a significant difference between the fault and control survey lines though a significant flux from the fault was not found in all basins and there was no apparent link to the presence, or absence, of hydrocarbons. As such, shale basins did not have a significantly different CH₄ flux to non-shale hydrocarbon basins and non-hydrocarbon basins. These results could have implications for CH₄ emissions from faults both in the UK and globally. Including all the corrected fault data, we estimate faults have an emissions factor of 11.5 ± 6.3 t CH₄/km/yr, while the most conservative estimate of the flux from faults is 0.7 ± 0.3 t CH₄/km/yr. The use of isotopes meant that at least one site of thermogenic flux from a fault could be identified. However, the total length of faults that penetrate through-basins and go from the surface to hydrocarbon reservoirs at depth in the UK is not known; as such, the emissions factor could not be multiplied by an activity level to estimate a total UK CH₄ flux.

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1. Introduction

With the introduction of high-volume hydraulic fracturing drilling techniques to extract unconventional hydrocarbons from shale formations, there has been increasing concern over the potential contamination of groundwater aquifers and the possible migration of gas and fluids. Some studies have suggested hydraulic fracturing fluids could have migrated to groundwater aquifers along natural fractures (Llewellyn et al., 2015), or that well integrity issues (Davies et al., 2014) have the potential to cause fluid migration to groundwater aquifers from active wells (Inggraffea et al., 2014), possibly as a consequence of poor cementing (Darrah et al., 2014). Fugitive emissions of methane (CH₄) at the ground surface can occur from abandoned wells, whether they have undergone decommissioning (Boothroyd et al., 2016) or remain unplugged (Kang et al., 2014; Townsend-Small et al., 2016). Natural migration of hydrocarbons has been identified along permeable pathways (Grasby et al., 2016; Warner et al., 2012) and Moritz et al. (2015) found deep thermogenic CH₄ could have migrated naturally along faults to aquifers, where it mixed with and was transformed into biogenic CH₄. Lavoie et al. (2016) suggested this process may have also occurred from shallower formations along small scale fracture networks, leading to microbial degradation of thermogenic volatiles in groundwater aquifers. It is important to establish what the cause of groundwater contamination or surface emissions of hydrocarbons is, whether from well integrity issues (Darrah et al., 2014); stimulated fractures connecting to natural faults and fractures (Reagan et al., 2015); or natural migration of fluids (Molofsky et al., 2011). An understanding of baseline conditions is thus required prior to any hydraulic fracturing taking place to determine whether natural seepage occurs.

It is important to understand the extent to which fault zones act as conduits for fluid flow, including hydrocarbons (such as CH₄), and CO₂, when considering the potential impact of hydraulic fracturing processes from shale gas basins. Modelling work has suggested that while fault zones may act as pathways for fluid flow – including frack fluid and brines – (Kissinger et al., 2013; Lange et al., 2013) such a scenario is only likely under certain geological conditions, such as high pressures induced by hydraulic fracturing if a highly permeable ($9.0 \times 10^{-14} \text{ m}^2$) fault zone is present (Kissinger et al., 2013). A study of natural and stimulated hydraulic fractures found the vertical extent of most natural fractures was between 200 and 400 m with a maximum recorded height of 1106 m (Davies et al., 2012). For stimulated fractures in the Barnett, Woodford, Marcellus, Niobrara and Eagle Ford shale gas formations, fracture propagation was typically <100 m and the maximum was 588 m (Davies et al., 2012).

Davies et al. (2013) indicated the maximum height of stimulated hydraulic fractures connecting to pre-existing fractures and hydraulic fractures was 1000 m. It was expected that in the case of stimulated hydraulic fractures in shale basins, overpressure in oil and gas operations would reduce when pumping stops, meaning fractures would be likely to close due to confining stresses. Nonetheless, transmission of fluids through pre-existing fracture systems could not be discounted and consideration of local geology was cited as an important stage prior to allowing fracking operations in a given area (Davies et al., 2013). It is therefore important to consider whether fractures could propagate and connect with larger scale faults, potentially providing pathways for fluid migration if permeability is high, such as with proppant used to keep fractures open under hydraulic fracturing. The importance of vertical separation between stimulated hydraulic fractures and overlying aquifers and the possibility of connections between stimulated and natural fractures allowing fluid flow to overlying aquifers was also highlighted by Jackson et al. (2015), who noted that for ~44,000 wells studied in the USA, the average fracturing depth was 2500 m but 16% were fractured <1600 m and 6% <900 m from the surface. It is consequently important to understand the behaviour of shale basins prior to any hydraulic fracturing processes taking place, so as to understand whether natural leakage of CH₄ and CO₂ from geological

sources already takes place, but also the propensity for fault zones to enhance fugitive emissions following hydraulic fracturing.

To identify whether elevated concentrations of CH₄ in the atmosphere have been transported through fault networks, it is necessary to determine what the source of any elevated concentration is. Indeed, this is a limitation of studies where faults are inferred to transport thermogenic CH₄ but where this is not verified (Vottatori et al., 2014). Isotopic measurements of $\delta^{13}\text{C-CH}_4$ can be used to distinguish between thermogenic and microbial sources of CH₄, typically ranging from $\delta^{13}\text{C-CH}_4$ -50 to -20‰ and -110 to -50‰ respectively (Whiticar, 1999). Mobile devices have been effectively used to monitor CO₂ and CH₄ concentrations along with isotopic compositions, enabling the identification of fugitive emissions from urban pipeline leaks (Jackson et al., 2014; Phillips et al., 2013), oil and gas production pads (Brantley et al., 2014) and coal seam gas fields (Maher et al., 2014). Numerous studies have calculated greenhouse gas budgets for shale operations (Burnham et al., 2012; Howarth et al., 2011; O'Donoghue et al., 2014; O'Sullivan and Paltsev, 2012) yet no consideration has been given to the release of thermogenic CH₄ naturally via fault zones and the potential consequence were stimulated hydraulic fractures to connect with natural fractures and provide a permeable pathway for fluid migration to the surface.

There are numerous examples of gaseous migration along fault zones to the surface, both of CH₄ and other gases. In Italy, endogenous migration of CO₂ was found to be greater in grassland with surface expression of faults compared to unfaulted grassland, with an extra soil CO₂ component 0.3–4.0 times background biological soil production of CO₂ (Etiope, 1999). Vottatori et al. (2014) found peaks in CO₂ and ²²²Rn in Greece were concentrated around fault zones and suggested gas micro-seepage from deep sources through the fault zone was the cause. Similarly, deep-seated faults in Poland have been identified to act as conduits for gas migration – including noble gases, CO₂ and CH₄ (Kotarba et al., 2014). In the Paradox Basin, central Utah, USA, a naturally leaking CO₂ rich system caused enrichment of CO₂ in groundwater, leading to precipitation of travertine mounds from springs and geysers, with vertical migration along faults also transporting hydrocarbons (Dockrill and Shipton, 2010). Migration of CO₂ through fault zones in the Paradox Basin has taken place over hundreds of thousands of years, with enhanced fluid flow concentrating in localised areas rather than across the entire fault system (Burnside et al., 2013). Methane seepage was highly localised along the most permeable sections of faults in Bacau, Romania (Baciu et al., 2008). Geothermal spring temperature measurements have been used as an analogue of convective heat transport along fault zones and evidence of high-permeability flow paths (Fairley and Hinds, 2004). Etiope and Klusman (2002) assessed a range of routes via which CH₄ emissions were possible from fault zones, including: Fischer-Tropsch reactions in geothermal systems; microseepage via buoyant flux of CH₄ or otherwise faults increasing the flow rate of microbubbles; and gas vents. Microseeps have been shown to occur across both onshore and offshore Europe, with estimated fluxes of CH₄ in Europe at 0.8 Tg yr^{-1} and total seepage estimated at 3 Tg yr^{-1} (Etiope, 2009). Faults and fracture networks were suggested to act as preferential pathways of degassing (Etiope, 2009) and Tang et al. (2013) reported hydrocarbon microseepage, including CH₄, through faults in the Yakela condensed gas field in the Tarim Basin, Xinjiang, China. Faults were speculated to act as conduits for coalbed methane migration (Boardman and Rippon, 1997; Creedy, 1988) and have been identified as one of three likely causes of CH₄ migration from coal seams in Ukraine (Alsaab et al., 2009). Furthermore, in the Ruhr Basin, Germany, emissions of thermogenic CH₄ were found where coal bed methane accumulated at the top of Carboniferous sediments (Thielemann et al., 2000). This meant that thermogenic CH₄ emissions were restricted to a few natural faults only, rather than being widespread (Thielemann et al., 2000).

Overpressure can be an important factor affecting fluid migration, with increased pressure driving fluid flow and keeping fractures open

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