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Effects of 'pre-fracking' operations on ambient air quality at a shale gas exploration site in rural North Yorkshire, England



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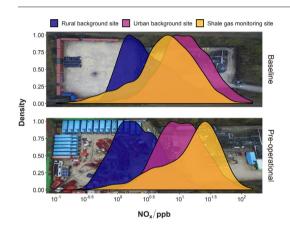
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HIGHLIGHTS

First observational assessment of incremental air quality impacts of operations at a shale gas site in the UK.

- Observations show due to baseline variability one annual cycle of measurements is needed to establish a local climatology.
- During "pre-operational phase" NO increased 3-fold from combination of increased vehicle activity and site operations.
- Although NO₂ also increased, air quality limit values for NO₂ were not exceeded

GRAPHICAL ABSTRACT



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ABSTRACT

Rural observations of air quality and meteorological parameters ($N_{\rm X}$, $N_{\rm S}$, $N_{\rm S}$, $N_{\rm S}$) were made over a 2.5-year period (2016–2018) before, during and after preparations for hydraulic fracturing (fracking) at a shale gas exploration site near Kirby Misperton, North Yorkshire, England. As one of the first sites to apply for permits to carry out hydraulic fracturing, it has been subject to extensive regulatory and public scrutiny, as well as the focus for a major programme of long-term environmental monitoring. A baseline period of air quality monitoring (starting 2016) established the annual climatology of atmospheric composition against which a 20-week period of intensive activity on the site in preparation for hydraulic fracturing could be compared. During this 'preoperational phase' of work in late 2017, the most significant effect was an increase in ambient NO (3-fold) and $N_{\rm O_X}$ (2-fold), arising from a combination of increased vehicle activity and operation of equipment on site. Although ambient $N_{\rm O_X}$ increased, air quality limit values for $N_{\rm O_2}$ were not exceeded, even close to the well-site. Local ozone concentrations during the pre-operational period were slightly lower than the baseline phase due to titration with primary emitted NO. The activity on site did not lead to significant changes in airborne particulate matter or non-methane hydrocarbons. Hydraulic fracturing of the well did not subsequently take place and the on-site equipment was decommissioned and removed. Air quality parameters then returned to the original (baseline) climatological conditions. This work highlights the need to characterise the full annual climatology of

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air quality parameters against which short-term local activity changes can be compared. Based on this study, changes to ambient $\mathrm{NO_x}$ appear to be the most significant air quality ahead of hydraulic fracturing. However, in rural locations, concentrations at individual sites are expected to be below ambient air quality limit thresholds. © 2019 Published by Elsevier B.V.

1. Introduction

The UK relies heavily on natural gas as an energy resource (BEIS, 2017) and exploratory studies indicate that there are significant onshore gas reserves that could be accessed by hydraulic fracturing or "fracking" as it is more commonly known (Andrews, 2013).

Shale gas extraction in the UK is at an exploratory stage at present. At the time of preparing this paper, shale gas exploration wells have been drilled at only a few sites in England. One of these (KM) is near Kirby Misperton in the Vale of Pickering, North Yorkshire. Following on from the hydraulic fracturing in 2011 which resulted in seismic activities of 2.3 $\rm M_{\rm L}$, there was a period of investigation that resulted in a number of studies and reviews, including the Royal Society and Royal Academy of Engineers (Mair et al., 2012). Most of the reviews revealed uncertainties due to lack of baseline data and it was required as evidence to support risk based assessments.

The impacts of shale gas-related air pollution in the UK remain uncertain, but include possible atmospheric emissions of both reactive air pollutants and greenhouse gases. Therefore, understanding the emissions at the first shale gas sites in the UK is important since it has the potential to help inform future decision making and constrain cumulative impact assessments should the sector expand.

The development and operation of a shale gas site consists of several different stages, each of which has the potential for emissions to the atmosphere; these are listed in Table 1. In England, and subject to permissions being granted, shale gas wells can be fracked relatively close to residential areas (<1 km in the case of Kirby Misperton), and concerns have been raised about the impact of operations on local air quality. Atmospheric emissions are expected from mobile machinery, compressors and pumps, venting and flaring, fugitive emissions of produced gas, and road transport to and from site (Public Health England, 2014). The most significant potential air quality pollutants are listed in Table 1.

There have been numerous studies of the effect of unconventional oil and gas extraction on air quality in some areas of the United States. For regional air quality the emission of non-methane hydrocarbons (NMHC) and nitrogen oxides (NO_x) are of most interest as some NMHC can be toxic (such as benzene) and can also contribute to photochemical ozone production (Edwards et al., 2014). The Eagle Ford area

 Table 1

 List of potential atmospheric pollutants from the shale gas operation life cycle.

Stage	Source of emission	Potential pollutants	
Well drilling and cementing	Dust	PM	
	Diesel generators	PM, NMHC, NO _x	
	Traffic	PM, NMHC, NO _x	
	Chemical processing	0 ₃	
	Fugitive	NMHC, H ₂ S	
Pre-operational phase	Dust	PM	
	Diesel generators	PM, NMHC, NO _x	
	Traffic	PM, NMHC, NO _x	
	Chemical processing	03	
Hydraulic fracturing	Dust	PM	
	Diesel generators	PM, NMHC, NO _x	
	Traffic	PM, NMHC, NO _x	
	Chemical processing	03	
	Fugitive	NMHC, H ₂ S	
Well production	Traffic	PM, NMHC, NO _x	
	Chemical processing	0_{3}	
	Fugitive	NMHC, H ₂ S	
Site restoration and well abandonment	Fugitive	NMHC, H ₂ S	

has been extensively studied with increases in methane, NMHC and ozone reported in oil and gas producing regions (Pacsi et al., 2015; Roest and Schade, 2017; Schade and Roest, 2016; Schade and Roest, 2018). Edwards et al., 2014, Schade and Roest, 2018, observed high levels of saturated hydrocarbons whose dominant sources were fugitive and evaporative emissions from oil and gas production. A major source of NO_x was the diesel powered compressor engines.

The Eagle Ford region was dominated by oil production which is not expected at any of the UK sites so it may be wiser to contrast results from the Marchellus Shale region in the US which is dominated by gas production. Fugitive emissions are expected to be dominated by methane and ethane, which are less reactive hydrocarbons than those observed at Eagle Ford, which may have implications for different atmospheric chemistry in this region. Studies have again found the air quality to be impacted in this region (Williams et al., 2018; Ren et al., 2019).

More recent studies have attempted source apportionment for nitrogen oxides, another major air pollutant that has been observed to increase in shale gas areas in the US but a limited number of studies on this are available (Prenni et al., 2016; Majid et al., 2017).

Emissions are often mitigated to some extent by control measures in place under the prevailing national regulatory system (and the degree of enforcement) and the type of operation, and so the experiences and impacts on air quality are not necessarily directly transferable between countries (or site operators). Superimposed are the modulating effects of different atmospheric conditions (e.g. wind speed, solar insolation, boundary layer depth, background atmospheric composition, etc.) which will also affect how fracking activities impact on atmospheric composition in different locations.

To determine the impacts of shale gas operations on air quality requires before, during and after observations; something that has not always been available in the context of the USA. Recognising the importance of this, an environmental monitoring programme was initiated around the KM site in 2016. The monitoring is independent of industry and the UK regulator but is supported by grant funding from the UK Government's Department for Business, Energy and Industrial Strategy (BEIS) (Ward et al., 2016; Ward et al., 2017; Ward et al., 2018). The project involves the monitoring of groundwater, surface water, seismicity and ground motion, soil gas, radon and air quality. This paper discusses elements of the air quality monitoring and some of the impacts observed at the KM site.

A recommendation in a UK Government report on greenhouse gases (MacKay and Stone, 2013) was that UK shale gas exploration should be accompanied by careful monitoring of all aspects of exploration (pre, during and post) and that this should include air quality pollutants. Managing and improving air quality in the UK is driven by European (EU) legislation on ambient air quality standards and also commitments to transboundary emissions, through the National Emissions Ceiling Directive and the UNECE Gothenburg Protocol. The 2008 Ambient Air Quality Directive (2008/50/EC) sets legally binding limits for outdoor air pollutants that impact on human health, of relevance here are the limits for NO₂, O₃, benzene, 1,3 butadiene, PM₁₀ and PM_{2.5}.

There is considerable existing infrastructure in the UK for measuring air quality, most significantly the AURN (Automatic Urban and Rural Network, https://uk-air.defra.gov.uk/networks/network-info?view=aurn) air-quality network which is used for compliance reporting against European air-quality directives. There are currently 127 operational sites measuring a range of parameters including most commonly nitrogen oxides, particulate matter, ozone, and less frequently carbon monoxide, sulfur dioxide and NMHCs. UK monitoring is however focused

predominately on locations with the poorest air quality and there can be large distances between AURN sites, especially in rural areas (which generally have better air quality, and are therefore not a priority for measurement). A report from the Defra Air Quality Expert Group (AQEG, 2018) on the potential air quality impacts of shale gas stated that "given none of the ambient monitoring stations in the current national network are well placed for baseline monitoring of shale gas activities, additional monitoring will be required" (Air Quality Expert Group to the Department for Environment and (AQEG), 2018). As a result, new observations have been made to provide a representative climatology of rural UK air quality conditions near the KM shale gas site, with this location being subsequently used to determine any increment in environmental pollution during shale gas operations including fracking.

We refer to observations made before any industrial activity at the KM site as the 'baseline' period of measurement. Our definition of baseline is data at a specific fixed location that is statistically representative of the local atmospheric composition, and which reflects the influences of preexisting local, regional and global pollution sources. To be representative, the baseline dataset must include inputs to air sampled over a period of time that is sufficient to capture typical ranges in emissions and variability in meteorological conditions. An atmospheric baseline can then provide values against which any incremental impacts of new emissions, new pollution sources, or policy interventions can potentially be assessed at a later date. The baseline for air pollution can be expected to incorporate effects of wind direction, time of day, and season for example. In an atmospheric context, we consider the minimum baseline period to be at least one-year in duration given the strong seasonal effects on air quality of meteorology, emissions and atmospheric chemistry. Arguably atmospheric baseline periods could be even longer, if there is a need to capture longer-term year to year variability, for example in circulation patterns and the balance of cyclonic to anticyclonic flows.

This paper discusses air quality parameters measured at the KM site. A baseline of greenhouse gas sources and climatology will be presented in future work.

2. Methods

2.1. Site selection

The siting of a baseline monitoring station should enable the sampling of locally representative atmospheric composition over a wide

range of meteorological conditions. A further consideration should also be that measurement sites are located appropriately to future-proof any role in potential operational monitoring requirements. The position of the KM monitoring site was selected so as to be predominantly downwind of any shale gas-related operations to support the assessment of any incremental impacts of exploratory activity (Smedley et al., 2015). The KM monitoring station is situated at the edge of the shale gas well-pad approximately 1 km to the west of the village of Kirby Misperton, North Yorkshire (Fig. 1).

The site consists of a mains-powered outdoor weather proof enclosure containing the air quality measurement instrumentation and a meteorological station. Table 2 lists the parameters and measurement techniques used the KM monitoring station.

Gas phase air inlets were initially positioned on 3 m high pylons. These were later extended to a height of 9 m, on top of a noise reduction barrier installed around the perimeter of the site. The Particulate Matter (PM) inlet was fixed at 3 m through the measurement period since this was a fixed, calibrated and heated inlet line. The installation of the barrier near the sampling site has meant that, between September 2017 and end of March 2018, PM observations were affected as a result of some wind recirculation and pooling at very low windspeeds.

2.2. Calibration and quality assurance

A quality assurance (QA) and quality control (QC) system for air quality and greenhouse gas concentration data was established covering all aspects of monitoring, including equipment evaluation, site operation, site maintenance and calibration, data review and ratification. All gas instrument calibrations were traceable through a chain to international reference standards to ensure comparability with similarly calibrated instrumentation, and included quantified uncertainties in the dataset.

The calibration methods for all measurements are detailed below. The ozone instrument provides an absolute measurement but was verified annually using a Model 49i-PS Primary Standard over the calibration range 0–500 ppb. The primary standard is itself checked annually against a certified source by the National Physical Laboratory (NPL, http://www.npl.co.uk). The $\rm NO_x$ instrument was calibrated and zeroed monthly using a 100 ppb NO standard in nitrogen linked to an NPL binary standard, the Global Atmospheric Watch (GAW) accredited Central Calibration Laboratory for $\rm NO_x$ and is also then referenced to the

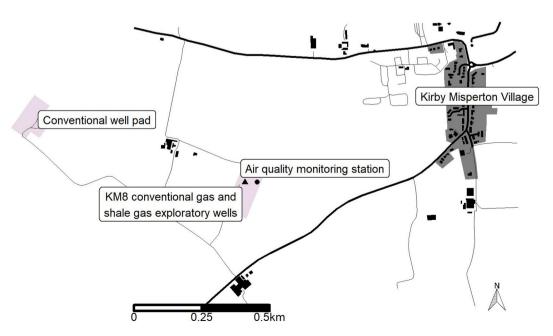


Fig. 1. Locations of the KM monitoring station (circle) and the KM8 well site (triangle).

 Table 2

 Instrumentation details at the KM measurement station. Note: measurements of methane and carbon dioxide (not listed) were also recorded and will be reported in future work.

Species	Instrument details	Frequency	Precision/uncertainty
Meteorological data (temperature, wind speed wind direction, relative humidity)	Luft WS-500UMB compact weather station	1 min	
NO, NO ₂	Teledyne T200UP Chemiluminescence/photolytic converter $NO/NO_2/NO_x$	1 min	NO_2 precision = 0.75 ppb NO precision = 0.65 ppb
O_3	ThermoFisher Model 49i Ozone Analyser	1 min	Precision = 0.74 ppb
PM ₁ , PM _{2.5} , PM ₄ , PM ₁₀	Fidas 200	1 min	$PM_{2.5}$ uncertainty = 0.44 $\mu g/m^3$ PM_{10} uncertainty = 0.64 $\mu g/m^3$
H ₂ S	ThermoFisher Model 250 H ₂ S/SO ₂ model	1 min	Precision = 1.0 ppb
SO_2	ThermoFisher Model 250 H ₂ S/SO ₂ model	1 min	Precision = 1.4 ppb
NMHC (C ₂ –C ₆)	SilcoCan Air sampling canister (3 L) GC-FID analysis for C_2 - C_6 NMHC	Weekly	<10% uncertainty for all NMHC

GAW scale. Monthly calibrations were only possible due to logistics and location of the measurement enclosure on the industrial site. All calibration slopes, offsets and drifts were collated and all data corrected for any instrument drift. The latest set of calibrations show an instrument drift of 0.3% over a period of 3 months. Any major drifts observed over the measurement period were corrected for. Internal gas phase titration with ozone is carried out yearly to calculate the NO $_2$ conversion efficiency and 5 point calibration carried out for NO. Precision is shown in Table 2.

NMHC observations are also referenced to the GAW scale using a thirty component mix of hydrocarbons at mixing ratios of ~4 parts per billion, provided by NPL. Measurements of a target gas mixture (cylinder of ambient air) were also routinely made in order to check instrument sensitivity drift and calculate the measurement uncertainty over the measurement period (in accordance with the quality assurance procedures defined by the ACTRIS (European Research Infrastructure for the observation of Aerosol, Clouds and Trace gases) VOC measurement guidelines, (ACTRIS, 2014). Metadata including precision and guidance on use of the data was prepared for each instrument and made available to view publicly on the Centre for Environmental Data Analysis (CEDA, http://www.ceda.ac.uk/) after final QC approval.

3. Results and discussion

3.1. Summary of air pollutants annual means

Fig. 2 shows a summary of the annual mean concentrations (for PM) and mixing ratios (for gases) of selected air pollutants at KM for the period Feb 2016–December 2017. As might be expected for a rural UK location none of the monitored air pollutants exceeded annual mean limit values (blue bar, Fig. 2), or approached the 75% threshold of the Ambient Air Quality Directive where air quality issues would typically need to be taken into account in the UK planning and development decision making process.

The most significant difference in annual mean values was the change in annual mean NO (and then included in the subsequent summation as NO_x) between 2016 and 2017, with NO increasing from 1.3 ppb to 5.3 ppb and the total NO_x from 5.9 ppb to 10.9 ppb, both as an annual mean. There was a slight decrease in annual mean O_3 between 2016 and 2017, related to direct titration via the increase in NO in 2017. These differences between years are associated with the preparation of the site for fracking and are discussed further later.

The distribution of $13\,C_2-C_6$ non-methane hydrocarbons is shown in Fig. 3. At present, there are only four automated hydrocarbon monitoring instruments in the UK air pollution network and none of these are in rural Northern England, so these measurements provide new insight into current background conditions. The relative profile of NMHCs was very similar between 2016 and 2017, as were the absolute amounts. Of the NMHCs measured, only benzene has a specific limit value in the EC Directive on air quality with an annual mean limit value of 5

 $\mu g\ m^{-3}$ (approximately 1.88 ppb). Annual mixing ratios of benzene in 2016 and 2017 were 0.10 and 0.14 ppb respectively, far below the limit value.

3.2. Effect of seasonality on air quality

Fig. 4 shows monthly mean mixing ratios for selected pollutants at the KM site. O₃ has a broad seasonal cycle, peaking in the spring and early summer months. This is typical for the UK, NO_x (Jan 2016 is missing as the instrument was not installed until Feb) also shows a seasonal cycle, with an increase in the winter months, due to a combination of an increase in emissions and meteorological impacts, as the boundary layer depth is shallower. The NMHCs show a summer minimum due to increased hydroxyl radical removal. The hydroxyl radical (OH) is the major chemical scavenger in the troposphere and it controls the lifetime of NMHCs in the atmosphere. The concentrations of OH depend on the photolysis of O_3 and therefore are higher in the summer months. Mean mixing ratios for both NO_x and NMHCs differ in the winter months between 2016 and 2017 with ethane, propane, acetylene and benzene a factor of 2-3 higher in 2016 and NO_x a factor of 2-3 higher in 2017. For the NMHC this is again due to higher emissions in the winter (e.g. heating) and the combinations of the shallow boundary layer. In 2017 there were also additional factors to consider which are discussed later.

3.3. Influence of wind direction

To enable a full baseline assessment it is important to examine the influence of wind direction on the observed mixing ratios of each

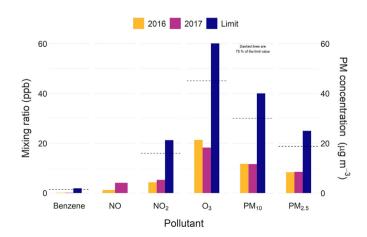


Fig. 2. Mean annual mixing ratios and annual mean concentrations of key pollutants at the KM measurement site compared with the EU Directive limit values for 2016 and 2017. The dotted line is the 75% thresholds for air quality limits.

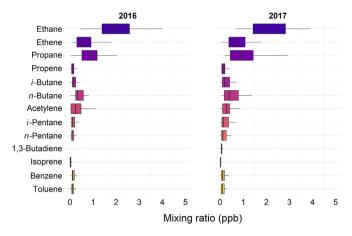


Fig. 3. Selected hydrocarbon boxplot of annual hydrocarbon mixing ratios measured at the KM measurement site in 2016 and 2017. Vertical bars are median values. The left and right edges of the box correspond to the 25th and 75th percentiles respectively. The horizontal whiskers show the largest or smallest values no further than 1.5 times the interquartile range respectively. Data beyond the end of the whiskers are not included here.

pollutant. Bivariate polar plots using wind speed and direction coloured by pollutant mixing ratio (Fig. 5) reveal an additional source for NO in 2017 which was not present in 2016. This additional source drives the increase in the annual concentration seen in Fig. 2. There are two clear sources of NO, one to the west of the monitoring station at low wind speeds therefore close to the measurement site and the other to the north east in higher wind speeds therefore further away. The $\rm O_3$ shows modest decreases in 2017 in the areas of highest NO $_x$. O $_3$ in 2018 looks lower on average but this is due to the majority of the summer data being missing due to instrument failure.

In September 2017, the site operator started to bring equipment required for hydraulic fracturing on to the shale gas well pad. Fig. 6 illustrates the amount of additional equipment brought on to the site for this stage. The image on the left shows the well pad when all the additional infrastructure was removed in April 2018. All that can be seen is the

existing conventional well pad (right hand side), the bags of sand (white and blue bags in the centre of the picture) and the container wall around the left hand side of the site (red containers). In contrast the picture taken in December 2017, shows the existing conventional well and sand bags, the container wall has been extended to include scaffolding and noise reducing material. The blue containers contain the fracking fluids and water and the majority of the red equipment is the drilling rigs and external generators.

This included an increase in traffic movements as well as pumps and generators being operated at the site. We define this phase of preparative work as the pre-operational period. It ran from the 19th September 2017 to the 1st February 2018. Research estimating emissions from shale gas operations (Robinson, 2012) found the main sources of NO_x emissions were from freight vehicles and drilling infrastructure. The majority of the hydraulic fracturing infrastructure was in place by December 2017. Seasonal polar plots for 2016, 2017 and 2018 are shown in Fig. 7, indicating that the additional source of NO was first present in Autumn 2017 and was visible until Spring 2018. This is further evidence that the statistical increase NO_x is a result of activities during the pre-operational phase at KM. A time series for the NO_x data for the whole measurement period is shown in Fig. 8 and clearly shows the increase during the "pre-operational" period.

 NO_x is generated by many different combustion sources and whilst NO has a short lifetime, measured in minutes before conversion to NO_2 , it means the footprint of sources extends many hundreds of metres away from the monitoring station (which was located at the well-site itself). If the pre-operational activities at KM were the only temporary incremental source of NO_x then the increment seen in atmospheric concentrations could be assigned solely to that source. Measurements made here would provide some guide to how concentrations may change at other similar locations. We do note however that the operator's well-site and supply chain activities were not the only new temporary source of NO_x added during the pre-operational phase. Further NO_x sources were introduced as a result of campaigning and protest activity including blockading and slow walking of vehicles, along with additional idling vehicles near the site from protest activities and the related policing and media. It is not possible to disentangle the

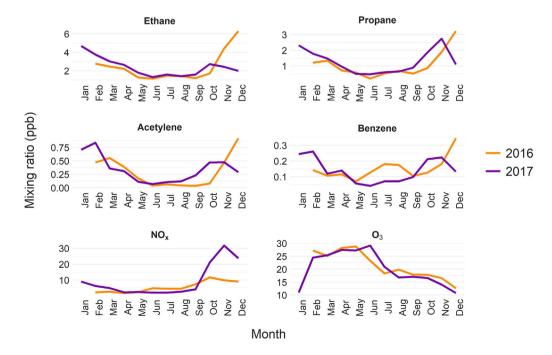


Fig. 4. Monthly averaged observed mixing ratios for NO_x , O_3 and selected hydrocarbons at the KM measurement site in 2016 and 2017. There were no NO_x measurements in January 2017 due to the instrument not being available.

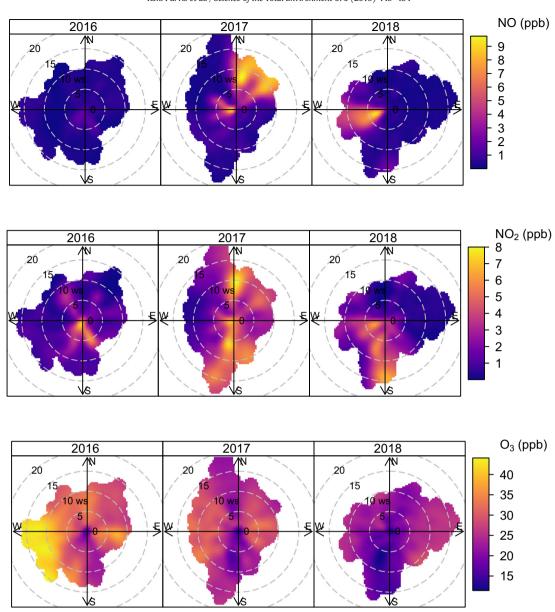


Fig. 5. Polar plots of mean mixing ratios of NO (a), NO₂ (b) and O₃ (c) at the KM measurement site (N.B. O₃ is missing data from May–July 2018 due to instrument failure).



Fig. 6. Images of the Third Energy Kirby Misperton well site in December 2017 (left hand side) and April 2018 (right hand side), illustrating the extra equipment brought onto the site in preparation for shale gas extraction. (Credit: Eddie Thornton).

relative proportions of these two sources, but it is important to identify that site preparation itself leads to incremental NO_x emissions from the operator, but also from any other new vehicle or combustion-related sources introduced near to well-sites. A study investigating the environmental impacts of traffic for the UK found that the impact of a single well pad can create substantial increases in local air quality pollutants during key periods. Modelling results showed that the total daily NO_x emissions would increase by 18-20% for a scenario with 71 vehicle movements from the site per day (Goodman et al., 2016), the vehicle movement at KM was much lower than this yet still saw an 50% increase in NO_x emission. Another modelling study showed that in the UK, local NO₂ concentrations could increase by up to 30 ppb (Archibald et al., 2018). Analysis of NO_x over oil and gas active areas in the US also found that the increase in NO_x emissions (Majid et al., 2017) and found strong correlations between NO₂ measurements from satellites and annual oil production. Other studies from the US also showed an increase in NO_x emission in areas of oil and gas production (Prenni et al., 2016) and similar trends seen in Canada from satellite measurements (McLinden et al., 2016). However, it is difficult to contrast the situation in KM with those published as hydraulic fracturing never took place and equipment was removed in in spring 2018.

3.4. Pre-operational period – effect on NO_x distribution

The probability density functions (PDFs) for NO_x measured at KM for the baseline and pre-operational periods are shown in Fig. 9. The periods used in this analysis are baseline: 19/9/16-31/12/16 and pre-operational: 19/9/17-31/12/17. Choosing the same time periods for the two years helps minimize (although does not eliminate) differences due to seasonal factors such as meteorology.

Data from two further Defra AURN sites are included in this analysis for comparison. High Muffles (HM) is a remote background site on the North Yorkshire Moors approximately 30 km from KM. York Bootham is in the City of York, the nearest major urban location to KM and is typical of a UK urban background site. During the baseline period data from the KM site falls somewhere between the remote rural background at

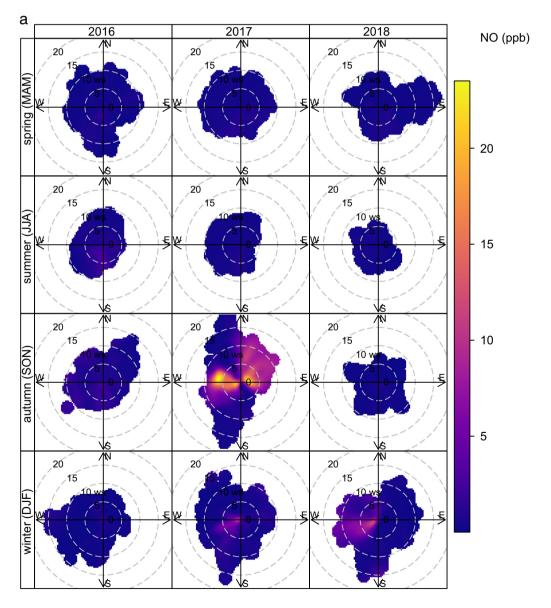


Fig. 7. Polar plots of mean seasonal mixing ratios of NO (a) and NO_2 (b) at the KM measurement site throughout 2016 and 2017. The pre-operation phase is most clearly visible as an increment in NO during autumn (SON) 2017. Data shown are one-minute averages of calibrated measured concentration.

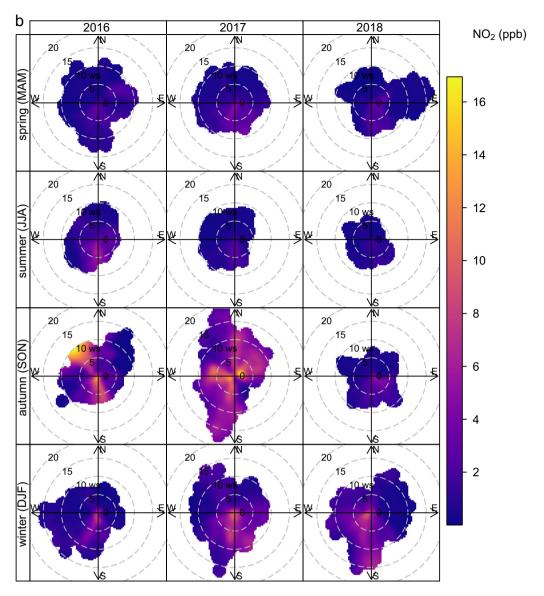


Fig. 7 (continued).

High Muffles and the York urban background in terms of both NO and NO₂ mixing ratio distributions (Fig. 7). Whilst KM is a countryside location, there is habitation nearby along with roads and agricultural activity. This means that KM during the baseline phase cannot be considered to be a remote/pristine environment from an air pollution perspective. During the pre-operational phase a significant shift occurs however as a result of increases in NO emissions at KM. This changes the distribution to give abundances that are higher than those measured in urban York. There is also a shift in NO₂, albeit smaller. The NO₂ distribution in the pre-operational phase was more similar to an urban background site than a rural background site. The NO₂ mixing ratios at the KM site in this pre-operational period were higher than those at York Bootham but did not exceed air quality standards. Edwards et al. (2014), observed high O₃ concentrations in the US in winter, a period not usually associated with high ozone events. It was concluded that high VOC concentrations coupled to a snow-covered surface and shallow boundary layer optimized the ozone production efficiency. There was no significant increase in NMHCs observed at KM well site during the pre-operational period suggesting that pre-operational activities did not lead to substantial increases in local NMHCs emissions.

4. Conclusions

Observations of air quality made at a shale gas well pad in North Yorkshire 2016–2018 have provided a baseline dataset against which the local effects of fracking and other operational shale gas activities can be later evaluated statistically and comparatively. The observations show considerable baseline variability in air pollution throughout the year and indicate that at least one full annual cycle of measurements is needed to establish a local climatology against which operational increments can be evaluated. The preparation work for hydraulic fracturing involving the delivery of equipment to the site and its operation led to significant increases in NO_x emissions, changing the site characteristics to be more similar to the UK urban background during this period. Whilst NO_x concentrations were seen to increase over the preoperational phase, no other pollutants showed a statistically significant change. Despite the increase in NO_x, there was continued attainment of all relevant air quality standards, i.e. no EU Air Quality Directive limits were exceeded. Operations were suspended at the site and hydraulic fracturing did not proceed. As a result, the equipment was removed for the site in January 2018. Air quality then returned to previous

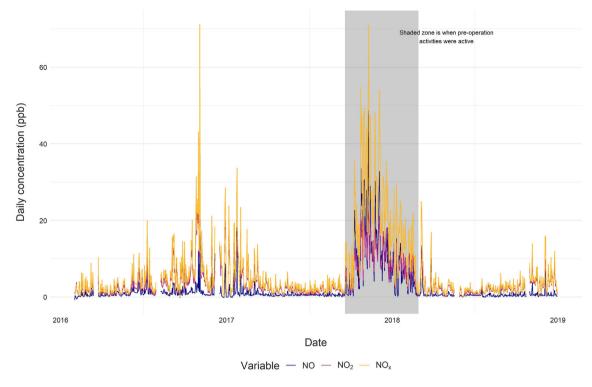


Fig. 8. Daily average of NO_x mixing ratio (ppb) at the Kirby Misperton measurement site from 2016 to 2018. The shaded area shows the pre-operational period. Data shown are one-minute averages of calibrated measured concentration.

baseline conditions. Although direct comparisons with elsewhere is not possible since hydraulic fracturing did not ultimately take place, the increase in NO_x emissions for the 3 month period of the operational stage is comparable with the increases in NO_x seen in other areas of active oil and gas extraction. This study provides a close examination of the impacts of a single pre drilled well, whereas other studies suc has Prenni et al., 2016 have looked at the cumulative influence of >10,000 wells. The emissions of air pollutants observed are consistent with the only

modelling study published to date for air pollutants from fracking which estimated that NO_x emission from traffic could increase emissions by 20%–30% (Goodman et al., 2016).

This study is the first observational assessment of the incremental air quality impacts of operations at a shale gas site in the UK. It highlights that pre-operational activities, through use of onsite diesel power generation as well as increased road transport, have the potential to increase $\rm NO_x$ emissions, and lead to significant localized increases in NO

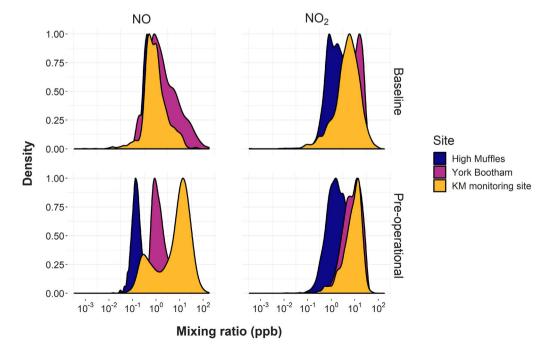


Fig. 9. Normalised probability density functions of NO and NO_2 during the baseline and pre-operational periods at KM well-site compared to a rural background site (High Muffles) and an urban background site (York Bootham). The baseline period is defined as 19/09/2016-31/12/2016 and the pre-operational period is defined as 19/09/2017-31/12/2017.

and NO_2 concentrations. Air pollution emissions are potentially impactful in the UK, and particularly in England, since exploration may well by necessity occur near populated areas, given the much higher UK population density. Other air pollutants were less affected during the preoperational phase, suggesting that a regulatory focus on ensuring the use of best available technology for NO_x abatement (both road transport and non-mobile machinery) would be most beneficial during this phase of site operations. Since the well-site did not proceed to hydraulic testing it has not yet been possible to determine the scale of air quality impacts during fracking operations, but all monitoring discussed in this paper remains in place should that occur in the future.

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References

- ACTRIS, 2014. WP4-NA4: Trace Gases Networking: Volatile Organic Carbon and Nitrogen Oxides, Deliverable D4.9: Final SOPs for VOCs Measurements.
- Air Quality Expert Group to the Department for Environment & Affairs (AQEG), 2018. Potential Air Quality Impacts of Shale Gas Extraction in the UK.
- Andrews, I.J., 2013. The Carboniferous Bowland Shale Gas Study: Geology and Resource Estimation. British Geological Survey for Department of Energy and Climate Change, London UK.
- Archibald, A.T., Ordonez, C., Brent, E., Williams, M.L., 2018. Potential impacts of emissions associated with unconventional hydrocarbon extraction on UK air quality and human health. Air Qual. Atmos. Health 11, 627–637.
- BEIS, 2017. UK Energy in Brief.
- Edwards, P.M., Brown, S.S., Roberts, J.M., Ahmadov, R., Banta, R.M., deGouw, J.A., Dube, W.P., Field, R.A., Flynn, J.H., Gilman, J.B., Graus, M., Helmig, D., Koss, A., Langford, A.O., Lefer, B.L., Lerner, B.M., Li, R., Li, S.M., McKeen, S.A., Murphy, S.M., Parrish, D.D., Senff, C.J., Soltis, J., Stutz, J., Sweeney, C., Thompson, C.R., Trainer, M.K., Tsai, C., Veres, P.R., Washenfelder, R.A., Warneke, C., Wild, R.J., Young, C.J., Yuan, B., Zamora, R., 2014. High winter ozone pollution from carbonyl photolysis in an oil and gas basin. Nature 514. 351.
- Goodman, P.S., Galatioto, F., Thorpe, N., Namdeo, A.K., Davies, R.J., Bird, R.N., 2016. Investigating the traffic-related environmental impacts of hydraulic-fracturing (fracking) operations. Environ. Int. 89–90, 248–260.
- MacKay, D., Stone, T., 2013. Potential Greenhouse Gas Emissions Associated With Shale Gas Extraction and Use.

- Mair, R., Bickle, M., Goodman, D., Koppleman, B., Roberts, J., Selley, R., Shipton, Z., Thomas, H., Walker, A., Woods, E., Younger, P., 2012. Shale Gas Extraction in the UK: A Review of Hydraulic Fracturing.
- Majid, A., Martin, M.V., Lamsal, L.N., Duncan, B.N., 2017. A decade of changes in nitrogen oxides over regions of oil and natural gas activity in the United States. Elementa (Wash. D.C.) 5.
- McLinden, C.A., Fioletov, V., Krotkov, N.A., Li, C., Boersma, K.F., Adams, C., 2016. A decade of change in NO2 and SO2 over the Canadian oil sands as seen from space. Environ. Sci. Technol. 50, 331–337.
- Pacsi, A.P., Kimura, Y., McGaughey, G., McDonald-Buller, E.C., Allen, D.T., 2015. Regional ozone impacts of increased natural gas use in the Texas power sector and development in the Eagle Ford shale. Environ. Sci. Technol. 49, 3966–3973.
- Prenni, A.J., Day, D.E., Evanoski-Cole, A.R., Sive, B.C., Hecobian, A., Zhou, Y., Gebhart, K.A., Hand, J.L., Sullivan, A.P., Li, Y., Schurman, M.I., Desyaterik, Y., Malm, W.C., Collett, J.L., Schichtel, B.A., 2016. Oil and gas impacts on air quality in federal lands in the Bakken region: an overview of the Bakken Air Quality Study and first results. Atmos. Chem. Phys. 16. 1401–1416.
- Public Health England, 2014. Review of the Potential Public Health Impacts of Exposures to Chemical and Radioactive Pollutants as a Result of the Shale Gas Extraction Process.
- Ren, X.R., Hall, D.L., Vinciguerra, T., Benish, S.E., Stratton, P.R., Ahn, D., Hansford, J.R., Cohen, M.D., Sahu, S., He, H., Grimes, C., Fuentes, J.D., Shepson, P.B., Salawitch, R.J., Ehrman, S.H., Dickerson, R.R., 2019. Methane emissions from the Marcellus Shale in southwestern Pennsylvania and northern West Virginia based on airborne measurements. J. Geophys. Res.-Atmos. 124, 1862–1878.
- Robinson, A., 2012. Air Pollution Emissions from Shale Gas Development and Production. Roest, G., Schade, G., 2017. Quantifying alkane emissions in the Eagle Ford Shale using boundary layer enhancement. Atmos. Chem. Phys. 17, 11163–11176.
- Schade, G.W., Roest, G., 2016. Analysis of non-methane hydrocarbon data from a monitoring station affected by oil and gas development in the Eagle Ford shale, Texas. Elementa (Wash. D.C.) 4, 1–17.
- Schade, G.W., Roest, G., 2018. Source apportionment of non-methane hydrocarbons, NOx and H2S data from a central monitoring station in the Eagle Ford shale, Texas. Elementa (Wash. D.C.) 6.
- Smedley, P.L.W., Allen, Rob S., Baptie, Grant, Daraktchieva, Brian, Jones, Zornitza, Jordan, David G., Purvis, Colm J., Cigna, Ruth M., Francesca, 2015. Site selection strategy for environmental monitoring in connection with shale-gas exploration: Vale of Pickering, Yorkshire and Fylde, Lancashire. British Geological Survey Open Report.
- Ward, R.S., Allen, G., Baptie, B.J., Daraktchievea, Z., Jones, D.G., Jordan, C.J., Purvis, R.M., Smedley, P.L., 2016. Environmental baseline monitoring - Vale of Pickering: phase I - final report (2015/16). British Geological Survey (96 pp. (OR/16/002).
- Ward, R.S., Smedley, P.L., Allen, G., Baptie, B.J., Daraktchieva, Z., Horleston, A., Jones, D.G., Jordan, C.J., Lewis, A., Lowry, D., Purvis, R.M., Rivett, M.O., 2017. Environmental baseline monitoring project. Phase II, final report. British Geological Survey 163 pp. (OR/ 17/049).
- Ward, R.S., Smedley, P.L., Allen, G., Baptie, B.J., Cave, M.R., Daraktchieva, Z., Fisher, R., Hawthorn, D., Jones, D.G., Lewis, A.C., Lowry, D., Luckett, R., Marchant, B.P., Purvis, R.M., Wilde, S., 2018. Environmental baseline monitoring: phase III final report (2017–2018). British Geological Survey Open Report.
- Williams, P.J., Reeder, M., Pekney, N.J., Risk, D., Osborne, J., McCawley, M., 2018. Atmospheric impacts of a natural gas development within the urban context of Morgantown, West Virginia. Sci. Total Environ. 639, 406–416.