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Measurement of diesel combustion-related air pollution downwind of an experimental unconventional natural gas operations site

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GRAPHICAL ABSTRACT

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ABSTRACT

Background & aim: Unconventional natural gas (UNG) extraction activities have considerable potential to affect air quality. However, there are few published quantitative observations of the magnitude of such impacts. To provide context, we compared measured exposures to diesel engine exhaust close to industrial fracking equipment at an UNG training simulation site in Łowicz, Poland to pedestrian exposures to traffic-related air pollution in the city centre of Glasgow, UK.

Methods: We made mobile and static measurements at varying distances from sources in both of the above locations with a portable aethalometer (Aethlabs AE51) for black carbon (BC) and portable monitors (Aeroqual Series-500) for nitrogen dioxide (NO_2) and ozone (O_3). Duplicate BC measurements were compared with NO_2 observations, after correction of the NO_2 sensor response for O_3 interference effects.

Results: Duplicate BC instruments provided similar real-time measurements (r = 0.92), which in turn were relatively highly correlated with NO_2 observations at 5-min temporal resolution at the UNG experimental site (r = 0.75) and on the walking route in Glasgow city centre (r = 0.64) suggesting common diesel sources for NO_2 and BC in both locations. Average BC and NO_2 concentrations measured approximately 10 m downwind of diesel fracking pumps were 11 and 113 $\mu g/m^3$ respectively. These concentrations were approximately 37 times and 4 times higher than upwind background BC and NO_2 concentrations at the site; and approximately 3 times higher than average BC and NO_2 concentrations measured in traffic influenced areas in Glasgow.

Conclusions: Marked elevations of BC and NO_2 concentrations were observed in downwind proximity to industrial fracking equipment and traffic sources. This suggests that exposure to diesel engine exhaust emissions from fracking equipment may present a significant risk to people working on UNG sites over extended time

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periods. The short time resolution of the portable instruments used enabled identification of likely sources of occupational and environmental exposure to combustion-related air pollutants.

1. Introduction

Unconventional natural gas (UNG), e.g. tight gas and shale gas, has been defined as gas trapped within relatively impermeable rock formations (Wang et al., 2014). Extraction of UNG as a substitute for conventional hydrocarbon sources has transformed natural gas production worldwide, especially in the USA. However, the development of UNG in countries outside the USA, including the UK, has been constrained as a result of concerns over possible impacts on human health and ecosystems. Current evidence regarding possible public health risks associated with UNG development is limited in detail and quality, resulting in intense debate around potential public health issues (Adgate et al., 2014; Hays et al., 2015; Kovats et al.; Werner et al., 2015). Controversy of this nature is often a direct consequence of uncertainties in scientific assessment (NPL, 2015), which in turn hampers policy decisions, and can result in high levels of anxiety within local communities (Larock and Baxter, 2013); and may produce adverse health effects not directly related to the extent of exposure to environmental pollution (Chapman et al., 2013). For example, in Scotland limitations in scientific evidence about possible health effects culminated in a moratorium on shale gas and coal bed methane extraction to allow a health impact assessment to be completed (Scottish-Government, 2015). In 2017 the Scottish Government announced an indefinite continuation of this moratorium with particular concerns being noted about the insufficiency of epidemiological evidence on the health risks associated with UNG industrial activities (Scottish-Government, 2017; SEPA, 2017b).

Air quality is one of several health concerns related to UNG developments (Field et al., 2014). There are analogies between the challenges of assessing potential public health risks from air pollution from UNG development and existing areas of environmental health research including traffic-related (HEI, 2010) and waste management related environmental pollution (Mohan et al., 2009; UK-Committee-on-Toxicity, 2010; Vrijheid, 2009). UNG operations, including well construction/operation and transportation of gas/other materials from the site, release a range of air pollutants (including nitrogen dioxide (NO_2), ozone (O_3), particulate matter (PM), carbon monoxide (CO), sulphur dioxide (SO_2) and benzo(a)pyrene (BaP) (Field et al., 2014)). These pollutants have guideline values specified by the World Health Organization (WHO) and Health, Safety and Executive (HSE) for ambient (Krzyzanowski and Cohen, 2008) and occupational (HSE, 2013; SCOEL, 2014) exposure respectively (Table 1). Another pollutant of concern associated with diesel combustion is black carbon (BC), which is mostly found within fine particle fractions (Viidanoja et al., 2002). Currently, there are no legislative air quality standards for BC (EEA, 2013) despite its increasingly clear association with negative health outcomes (Englert, 2004; Heal and Quincey, 2012; WHO, 2012) and its recognition as a 'marker' of local combustion emissions. Exposure to diesel exhaust particles is often characterised by exposure to elemental carbon, and in some cases black carbon has been used as a proxy exposure metric in environmental (Grahame et al., 2014; Janssen et al., 2011) and occupational (Forder, 2014) situations.

A small number of studies highlight risks of adverse health effects in communities exposed to atmospheric emissions from nearby UNG facilities (Elliott et al., 2017; Elsner and Hoelzer, 2016; Islam et al., 2016; Paulik et al., 2016). For example, McKenzie et al. (2012) collected air samples of volatile organic compounds (VOCs) and conducted health risk assessments in two groups living near UNG developments in Colorado; and observed that residents living within 0.8 km of UNG sites had a higher risk of developing cancer and non-cancer diseases. McKenzie et al. (2014) observed that maternal residence within 16 km from a gas well was associated with a raised prevalence of congenital heart defects and neural tube defects in offspring. McKenzie et al. (2016); Rabinowitz et al. (2015); Rasmussen et al. (2016) observed that frequent respiratory symptoms were experienced by people living less than 1 km close to the natural gas wells. However, in common with other areas of environmental health, in these studies of UNG sites there are crucial limitations in characterisation of exposure to potential hazards (Lioy, 2015), which prevent reliable confirmation or refutation of causal linkage between potential environmental influences on human health through misclassification bias (Armstrong, 1998).

In addition to uncertainties about environmental exposures affecting local communities, there is relatively limited published information on assessment of occupational exposures of people working in UNG operations. The most common sources of occupational exposure are likely to include: (1) direct and fugitive emissions of hydrocarbons from the well and associated infrastructure; (2) deliberate venting and flaring of gas and related petroleum products; (3) diesel exhaust emissions from hydraulic fracturing equipment, trucks and generators (Adgate et al., 2014; Bogacki and Macuda, 2014).

The implementation of adequate air pollution monitoring is vital for effective management of risks to human health from UNG operations. This includes the collection of information on spatial and temporal patterns of air pollution from measurements and/or modelling to estimate human exposure at specific locations (Adgate et al., 2014; McKenzie et al., 2014; Werner et al., 2015). For example, Zielinska et al. (2014) observed variations in benzene, NO_2 and VOCs concentrations from preliminary surveys and continuous measurements at several distances from the operation of UNG and within residential areas.

Knowledge about the potential health implications of air pollution from UNG operations can be substantially improved through the deployment autonomy of portable measurement systems. In this pilot study, we deployed miniature low power [hence portable and readily deployable (Lewis and Edwards, 2016; Snyder et al., 2013)] sensor-based systems to measure exposures upwind, within and downwind of operation of hydraulic fracturing equipment. Our study investigated short-term simultaneous exposure to BC, NO_2 and O_3 in different locations at a simulated UNG site in Poland. To the best of our knowledge few if any simultaneous measurements of these pollutants close to UNG sites have been published in primary scientific literature providing a challenge for comparison of our measurements with existing knowledge. Therefore we also made measurements of BC, NO_2 and O_3 within road-traffic influenced locations in Glasgow city-centre to provide context to the concentrations we measured at the UNG site. The results of our study have implications for best practice in protecting workers in close proximity to hydraulic fracturing equipment; and illustrate and

Table 1
Guidelines for environmental (Krzyzanowski and Cohen, 2008) and occupational exposure (HSE, 2013; SCOEL, 2014) to air pollutants measured in this study.

Air pollutant:	Guideline values (Environmental exposure)		Guideline values (Occupational exposure)	
	Averaging time:	Limit value:	Averaging time:	Limit value:
NO_2	1 h	200 μg/m^3	8 h	0.955 mg/m^3
	1 year	40 μg/m^3	15 min	1.91 mg/m^3
O_3	1 h	120 μg/m^3	15 min	0.4 mg/m^3

begin to address some of the exposure assessment challenges associated with UNG development.

2. Methods

2.1. Monitoring site

Field measurements were made on 22 June 2015 during a field visit to an UNG service company's logistics and storage site in Łowicz county (52.087°N, 19.916°E), 76 km west of Warsaw, Poland (Fig. 1). This service company provided drilling, hydraulic fracturing, well testing services and seismic data acquisition services to the onshore oil and gas industry. The site was of similar scale (area = 8862 m^2) to an active UNG site and consisted of: an administrative and laboratory building; open storage area for heavy equipment; indoor storage warehouses; and a paved yard.

The company set up a simulated hydraulic fracturing site in the yard consisting of a control cabin, three diesel-powered truck-mounted fracking pumps, a fluid blender and manifold truck (Fig. 2). The fracking equipment was using a Diesel Engine MTU 12V4000, Rated power: 2250 BHP, driving a Stewart & Stevenson High Pressure Pump Model FT-2251T. The fuel used was Diesel #2. We are not aware of the use of any fuel additives. These simulated hydraulic fracturing procedures are done as part of training for the company's operators. The fracking equipment was not connected to a wellhead, but was run under equivalent pressures and loads to an active site (Josifovic et al., 2016). This experimental design allowed us to assess the effects of the diesel engines used to power the hydraulic fracturing equipment on nearby air quality in absence of pollution from natural gas, flowback water (potential for 'degas' emissions), and gas infrastructure including compressors and flare-stacks. Table 2 summarises information on the large fracking equipment plant operated during our measurements.

2.2. Sampling strategy

We monitored air quality at selected locations to provide a brief sample estimate of exposure of the staff on the site, ranging from staff indoors in the administration building and control cabin to mechanics working on and around the pumps, informed by site survey (Fig. 2). Wind direction was estimated using a compass and visual observation from the movement of nearby vegetation, and the direction in which visible exhaust smoke from machinery travelled. The wind was generally from south to south-westerly directions for the duration of the mobile and static measurements. Mobile measurements were made while walking with the air monitors in background, upwind and downwind areas across the site (Fig. 2b). Static measurements were made at single upwind/downwind locations when the researcher stood with the monitors for 2 min to characterise emissions of air pollutants near the fracking pumps. The walking paths were approximately 960 m in length, and walking speed was approximately 0.8 m s^{-1} (typical pedestrian walking speed (Shi and Ng, 2017)). The downwind walking paths and static monitoring locations were selected in areas where peak downwind concentrations were expected.

We monitored concentrations of pollutants alternately in different microenvironments at the experimental UNG facilities (Table 3). The majority of monitoring locations were located within 100 m of diesel fracking pumps (Fig. 2b) to characterise exposure to combustion-related pollutants of the personnel who work in close proximity to this machinery. We also made indoor measurements in an office in the administrative building situated to the north-west of the UNG simulation site. We measured pollutant concentrations during four time periods between 11:00 and 16:00. This included: mobile background measurements (between 11:00 and 12:00); indoor measurements in the office (between 13:00 to 14:30); and mobile and static measurements in the vicinity of operating hydraulic fracturing equipment (12:00 to

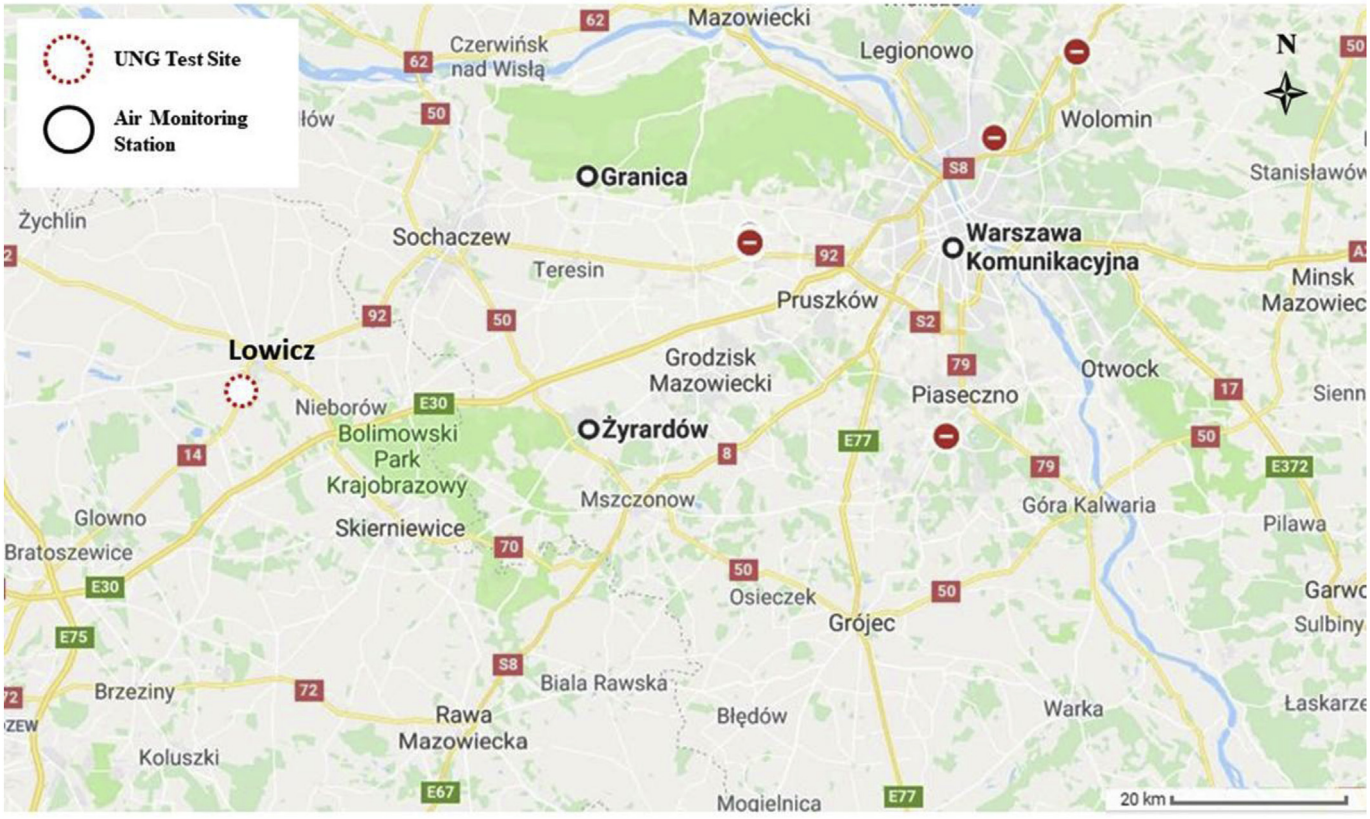

Fig. 1. Location of UNG test site in Łowicz, Poland in relation to local air monitoring stations located in Granica (rural background), Zyrardow-Roosevelta (urban background), and Warszawa-Komunikacyjna (traffic). Source: https://www.google.co.uk/maps/.

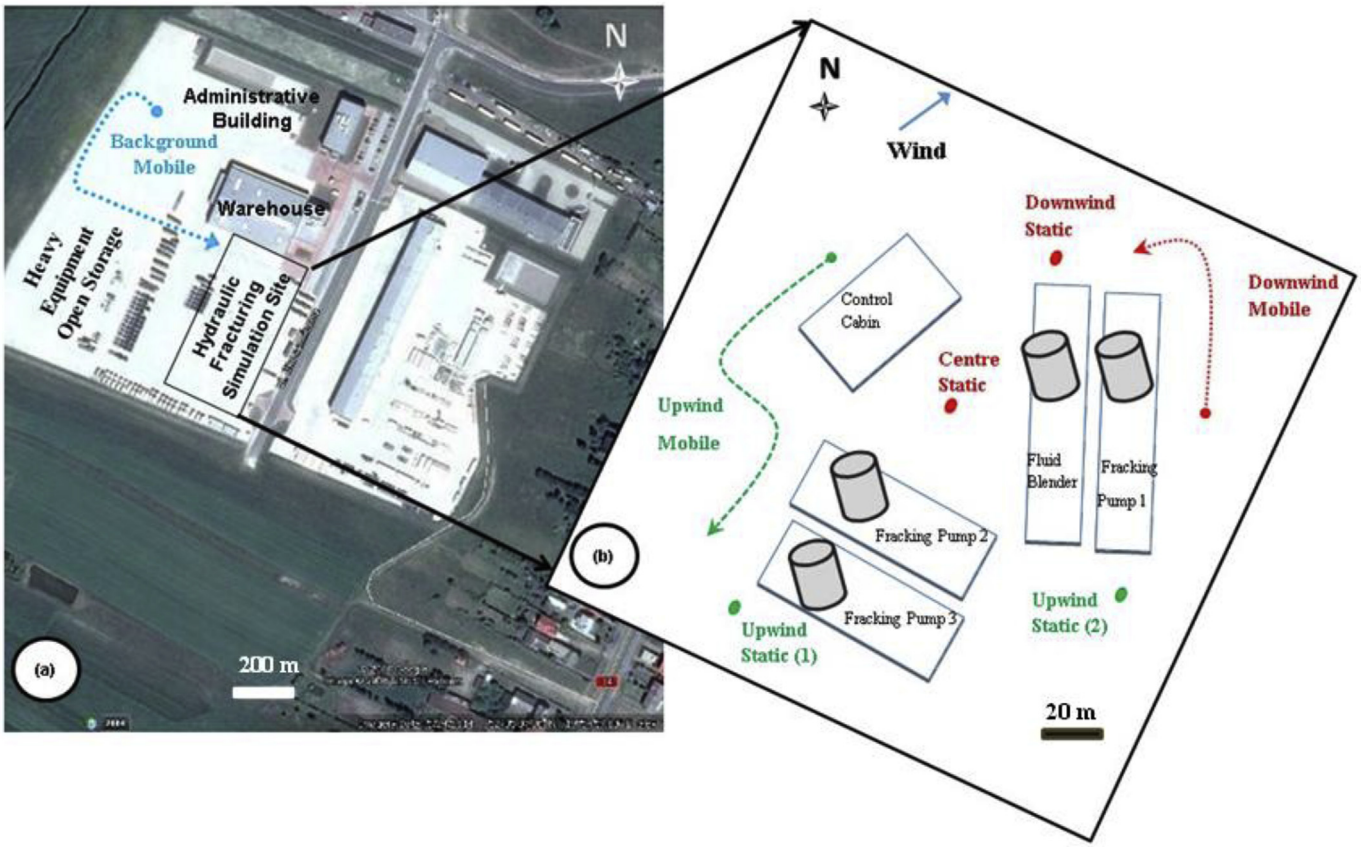

Fig. 2. (a). Aerial photograph (source: https://earth.google.com/web/) of experimental UNG site in Łowicz county, Poland. The blue line represents the path of the mobile background measurements; (b) Schematic diagram of site, indicating locations of mobile and static measurements: centre of site; downwind; and upwind. The cylinder symbols represent locations of diesel exhaust stacks. The mobile measurements started from '●' and finished at the arrow tip. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

12:45 and 15:00 to 16:00). We monitored BC, NO_2 and O_3 between 12.00 and 12.45; however we could only monitor BC between 15:00 and 16:00 because of rain (the handheld NO_2 and O_3 instruments were not weather-proof). We made upwind and downwind BC measurements during the second period, but static measurements could not be made in the centre of the site due to safety procedures. We focused our analyses on the first sampling period as it was more complete in terms of pollutant metrics and locations monitored. However the data recorded for the second period are presented as context to the analyses of the first period. A summary of measurement locations and durations are presented in Table 4. All times indicated were local times.

Both static and mobile measurements of upwind background concentrations were made (Fig. 2b) to allow the cross-site gradient of air pollution concentrations to be compared to the nearest local authority air monitoring station (Fig. 1). Additional mobile background measurements were made while walking from the administrative building towards to the east side of the heavy equipment open storage area (Fig. 2a).

2.3. Black carbon (BC) concentrations

We measured BC with two portable aethalometers (AE51; AethLabs, USA). This instrument has been widely used to measure personal exposure to BC in different microenvironments (Dons et al., 2012, 2013, 2014). The AE51 aethalometer quantifies BC by attenuation of laser light through accumulated sampled particles on a PTFE coated glass fibre filter strip (T60). Particle laden air is sampled through a 1 m conductive tube with a conductive plastic rain hood to prevent water

Table 2
Diesel powered hydraulic fracturing equipment used during test measurements.

Fracking equipment	Task	Diesel engine:
Stewart & Stevenson High Pressure Fracking Pump - Model FT-2251T	Trailer-mounted fracturing unit capable of pumping inhibited acid and other proppant laden stimulation fluids	MTU 12V4000, Rated power: 2250 BHP http://www.mtuonsiteenergy.com/products/diesel-generator-sets/mtu-4000-ds/mtu-12v4000-ds1750-50hz/
Stewart & Stevenson MT-132HP Blender	Trailer-mounted hydraulic fracturing blender designed to mix slurries for fracturing treatments and provide boost pressure. Maximum proppant load = 21 250 pounds per minute and maximum concentration = 400 kg/m^3.	Front deck engine: Caterpillar Model C18; Rated power = 755 BHP. Supplies power for hydraulic system. Rear deck engine: Caterpillar Model C18. Rated power = 800 BHP. Supplies power for the centrifugal discharge pump.
ET-26ACD Stewart & Stevenson Data Acquisition Centre	A controlled area to control and monitor well treatments	20 KW generator

Table 3
Mobile and static monitoring locations at the experimental UNG site.

Sample location name (& code)	Details of sample location (Fig. 2b)
Upwind Static (1) - US(1)	12 m from fracking pumps 2 and 3
Upwind Static (2) - US(2)	15 m from fracking pump 1
Downwind Static	10 m from fluid blender and fracking pump 1
Centre Static (CS)	Between control cabin, fracking pump 2 & fluid blender
Background Mobile	While walking from administrative building to east side of heavy equipment open storage area (Fig. 3a)
Upwind Mobile (UM)	While walking from control cabin control to fracking pump 3
Downwind Mobile (DM)	While walking from east side to north side of fracking pump 1
Indoor	Static sample collected from an office in administrative building

Table 4
Summary of black carbon (BC), nitrogen dioxide (NO_2) and ozone (O_3) measurements at different monitoring locations.

Monitoring location:	Measurement duration (min):	Air pollutants measured:
Outdoor background	60	BC, NO_2 and O_3
Indoor office room in administrative building	90	BC, NO_2 and O_3
First monitoring period	45	BC, NO_2 and O_3
Second monitoring period	45	BC only

ingress to the instrument.

In this paper, we refer to the aethalometers according to their serial number (AE51-1204 and AE51-1303). The aethalometers were calibrated by the manufacturer and new filters were fitted prior to conducting the field measurements described here. The instruments were set to record data every second, with a flow rate of 100 ml/min. BC data was downloaded using MicroAethCOM software v2.2.4.0, and processed using an optimised noise adjustment (ONA) method ($\Delta ATN = 0.01$) to retain the highest possible temporal resolution (Hagler et al., 2011). We averaged 1-s BC data to 1-min intervals, to allow comparison with measurements recorded at 1-min intervals from the NO_2 and O_3 monitors. Average data from the 2 instruments is presented. The MicroAeth BC sensor has a response time of 1 s, and measurement resolution of 0.001 μg/m^3.

2.4. Nitrogen dioxide (NO_2) and ozone (O_3) concentrations

We measured NO_2 and O_3 with Aeroqual Series 500 monitors (Aeroqual Ltd, New Zealand). In these monitors NO_2 was quantified using an electrochemical sensor (GSE 0–1 ppm) and O_3 was quantified with a gas sensitive metal oxide semiconductor sensor (OZU 0–0.15 ppm). The Aeroqual NO_2 and O_3 monitors were set to record data at 1-min intervals. The Aeroqual NO_2 and O_3 sensors have response times of 30 s and 60 s respectively. The resolution of both Aeroqual sensors specified by the manufacturer was 1 ppb (*i.e.* ~ 2 μg/m^3).

The Aeroqual NO_2 sensor has previously been shown to be affected by cross sensitivity to O_3, and consequently we corrected measured concentrations following the method described by Lin et al. (2015). We deployed the Aeroqual NO_2 and Aeroqual O_3 monitors at a UK government Automatic Urban and Rural Network urban background monitoring station at Townhead, Glasgow (55.866 ºN, 4.244 ºW). Reference analysers for NO_2 and O_3 at this site are a Teledyne API200A chemiluminescence analyser and a Thermo 49i UV absorbance analyser respectively. The concentrations measured by the reference analysers were subject to national QA and QC procedures (DEFRA, 2010) We co-located the Aeroqual monitors within 2 m horizontal distance, and 1.5–2 m vertical distance of the reference analyser inlets at Townhead between 17:00 on 12 May 2015 to 08:00 on 18 May 2015, 35-days prior to the measurements at the UNG site. A linear regression equation was calculated to describe the relationship between the difference in NO_2 concentration measured by the Aeroqual NO_2 monitor and the reference NO_2 analyser, and concurrent O_3 concentrations measured by the Aeroqual O_3 monitor (*Aeroqual_O3* (μg/m^3)) (Lin et al., 2015):

$$NO_2 difference = 0.97\ Aeroqual O_3 - 32.46 \qquad R_2 = 0.71 \tag{1}$$

where:

NO_2_difference = observed difference (μg/m^3) between NO_2 concentration measured by the Aeroqual NO_2 monitor and the reference NO_2 analyser.

Equation (1) allows the concentrations measured by the Aeroqual NO_2 monitor to be corrected to account for the Aeroqual NO_2 instrument cross-sensitivity to O_3:

$$NO_2\_corrected = NO_2\_Aeroqual - NO_2\_difference \tag{2}$$

Where:

NO_2_corrected = calibrated Aeroqual NO_2 concentrations (μg/m^3)
NO_2_Aeroqual = unadjusted Aeroqual NO_2 concentrations (μg/m^3)

The Aeroqual O_3 monitor measurements were also adjusted using a calibration relationship derived from the co-deployment of Aeroqual O_3 sensor alongside the reference O_3 analyser at Townhead:

$$O_3 corrected = 1.14\ Aeroqual O_3 + 13.1 \qquad R_2 = 0.78 \tag{3}$$

Where:

O_3_corrected = calibrated Aeroqual O_3 concentrations (μg/m^3)
Aeroqual_O_3 = unadjusted O_3 concentrations (μg/m^3)

Similar to the findings of our earlier studies (Lin et al., 2015) we observed limited and inconsistent effects of temperature and relative humidity during the calibration of the monitors and therefore these meteorological variables were not included in our analyses. The weather conditions during calibration and subsequent instrument deployment were not dissimilar.

2.5. Field deployment procedure

The duplicate BC instruments were carried separately in two backpacks carried by 2 researchers on the same sampling route and times. We fitted the NO_2 and O_3 monitors securely in the external mesh pockets of one of the backpacks. The internal clocks of all instruments were synchronised with internet time servers. All monitors were switched on at the same time and the sampling duration was recorded. We recorded a time-activity diary by hand during measurement periods.

2.6. Fixed air monitoring station

Local air pollution and meteorological data was obtained from monitoring stations on the Warsaw Regional Inspectorate of Environmental Protection internet site (http://sojp.wios.warszawa.pl/?

par = 90). These air quality measuring stations are operated by local government in Mazowieckie province. The nearest fixed monitoring sites were a rural background site (Granica-KPN; 50 km North East), an urban background site (Zyrardow-Roosevelta; 45 km East), and a traffic site (Warszawa-Komunikacyjna; 85 km East) (Fig. 1).

2.7. Additional BC and NO_2 measurements in Glasgow city centre

To provide context to the BC and NO_2 exposure measurements at the UNG site we made additional measurements of the same pollutants on a pre-defined walking route in Glasgow city centre using the portable monitors (AE51 Aethalometer & Aeroqual) during 8 repeat walks over a four week period (30 June 2015–24 July 2015). The measurements were made in the middle of the day, avoiding rush-hours but at times when the roads were relatively busy with traffic. Emissions estimates derived from traffic counts used in air quality models for Glasgow City centre suggest that in some of the busy streets on the walking route diesel buses may contribute approximately 70–80% of NO_x emissions (SEPA, 2017a). Correspondingly the pollution composition measured in these streets in Glasgow City centre was anticipated to be broadly comparable with the diesel-dominated conditions close to the experimental UNG site.

To better understand the influence of nearby traffic on the BC and NO_2 concentrations measured by the instruments in Glasgow, we examined detailed spatiotemporal patterns for an example study period on 24 July 2015 (between 11:50 and 13:00 British Summer Time) using ArcGIS v.10.2.2 (Esri, USA). We also examined bivariate relationships between observed BC and NO_2 concentrations for the different study periods in Poland and Glasgow using reduced major axis regression (Ayers, 2001).

3. Results and discussion

3.1. Calibration of aerqual sensor data

We used the procedure developed by Lin et al. (2015) to calibrate the Aeroqual NO_2 and Aeroqual O_3 sensors by comparison to reference NO_2 and O_3 instruments at the DEFRA Automatic Urban Network (AUN) monitoring site at Townhead in Glasgow, 35 days prior to the field visit to the experimental unconventional natural gas site in Poland. The measurements available for the Townhead AUN site were at hourly resolution. For O_3 the range of reference concentrations during calibration mostly encompassed the range of calibrated measurements (range of hourly averages of reference analyser O_3 concentrations during calibration of 8–75 μg/m^3 *cf.* range for calibrated 1-min O_3 measurements of 13–86 μg/m^3 at the UNG site in Poland; 4 out of 210 calibrated measurements slightly exceeded the above range of reference analyser O_3 concentrations during calibration). For NO_2 the range of reference concentrations during calibration was more limited than we observed in the 1-min resolution measurements made in Poland (range of hourly average reference analyser NO_2 concentrations during calibration of 5–56 μg/m^3 *cf.* range of calibrated 1-min NO_2 measurements at the UNG site in Poland of 0–287 μg/m^3). This means that NO_2 concentration estimates above the concentration ranges observed during calibration (30 out of 210 calibrated measurements) were extrapolated rather than interpolated from the calibration equations. This practical limitation is mitigated to some extent by wider manufacturer calibration ranges (range of standards in calibration certificates for O_3 sensors: 4–264 μg/m^3; range of standards in calibration certificates for NO_2 sensors: 13–1300 μg/m^3); and by recently published laboratory evaluations conducted by another research group where a relatively linear overall response of the Aeroqual sensors was noted over a calibration range of ~10–~350 μg/m^3 (Miskell et al., 2018).

Four negative calibrated observations of 1-min NO_2 concentrations were estimated for field measurements at locations upwind of the UNG site in Poland (*i.e.* negative calibrated observations of: −5, −7, −8, −10 μg/m^3). These negative calibrated observations represented ~2% of the total number of calibrated observations (*i.e.* 4 out of a total of 214 calibrated 1-min NO_2 concentration estimates). We censored the 4 negative calibrated NO_2 estimates to half of the lowest reference analyser concentration in the calibration dataset where we observed linear responses in the calibration relationships (*i.e.* half of the minimum reference analyser concentration of 5 μg/m^3 described in section 2.4). The censorship of the 4 negative estimates to 2.5 μg/m^3 is consistent with the average NO_2 concentration (2.7 μg/m^3) at the Granica-KPN rural background site over the 1100–1300 time period of our field NO_2 measurements. The censorship of the calibrated NO_2 estimates in this way had minimal effect on the computed mean upwind concentrations (Table 5 and Table S1) and no effect on our overall conclusions. As context, in the previous research that developed the above method of calibration (Lin et al., 2015), 25 negative calibrated NO_2 values were generated out of a test dataset of 849 hourly values (*i.e* ~3%) on occasions when the Aeroqual O_3 signal was relatively high compared to the Aeroqual NO_2 signal.

Table 5
Summary statistics of black carbon (BC), nitrogen dioxide (NO_2) and ozone (O_3) concentrations at different monitoring locations during operation of hydraulic fracturing pumps.

Site	Start Time	End Time	Black Carbon (μg/m^3)			Nitrogen dioxide (μg/m^3)			Ozone (μg/m^3)		
			Mean	Max	Min	Mean	Max	Min[b]	Mean	Max	Min
Background Mobile	11:00	12:00	0.0	0.0	0.0	19	34	< 5	58	78	25
UNG (1) - First monitoring period:											
Upwind Mobile	12:01	12:13	0.1	0.3	0.0	36	53	< 5	22	69	13
Centre Static	12:14	12:16	16.9	27.5	9.7	193	257	125	13	13	13
Downwind Mobile	12:17	12:32	8.1	22.9	0.7	110	288	32	22	52	13
Downwind Static	12:33	12:35	46.2	51.2	42.1	129	173	88	41	55	13
Upwind Static (1)	12:37	12:39	0.1	0.1	0.1	49	70	35	48	65	13
Upwind Mobile	12:40	12:45	0.2	0.3	0.1	13	45	< 5	73	86	64
Administrative Building	13:00	14:30	0.8	1.9	0.4	41	60	16	18	59	13
UNG (2) - Second monitoring period[a]:											
Upwind Static (2)	15:00	15:26	0.4	0.9	0.3	–	–	–	–	–	–
Downwind Mobile	15:27	15:31	2.4	8.1	0.5	–	–	–	–	–	–
Downwind Static	15:32	15:34	7.8	10.9	5.1	–	–	–	–	–	–
Upwind Static (1)	15:36	15:38	0.3	0.4	0.3	–	–	–	–	–	–
Upwind Mobile	15:39	15:45	0.5	0.6	0.3	–	–	–	–	–	–

[a] NO_2 and O_3 concentrations were not measured during the second monitoring period because of rainfall.
[b] Minimum NO_2 concentrations were estimated as 2.5 μg/m^3. See section 3.1 for details.

3.2. BC, NO_2 and O_3 concentrations measured at UNG test site

The concentrations of all the measured pollutants at the UNG test site increased markedly during fracking pump operation and gradually declined after the fracking pumps were switched off, during both the first and second monitoring periods (Fig. 3). Short-term peaks in BC (approx. 50 μg/m^3) and NO_2 (approx. 290 μg/m^3) were probably the result of local exhaust plumes from the fracking pump engines, indicating how site operators working close to operational pumps may experience high short-duration peak exposures.

We are not aware of other published measurements of this nature at UNG sites. As context, in an occupational study, 8-h average BC exposures have been observed in municipal household waste collectors (10.1 μg/m^3) working close to refuse trucks and refuse truck drivers (7.8 μg/m^3) in Korea (Lee et al., 2015). Personal exposures to BC in aircraft passengers have been measured during transit (average = 3.78 μg/m^3) and during flights (average = 0.20 μg/m^3) with maximum 30 s-average BC concentrations of 24 μg/m^3 observed in a terminal transfer bus at a Brazilian airport (Targino et al., 2017).

We divided the measurements into two discrete time-series to interpret measured pollution concentrations during the first (Fig. 3b) and second (Fig. 3c) 45-min duration monitoring periods when the fracking pumps were operated. Concentrations of BC, NO_2 and O_3 varied substantially between the upwind and downwind sampling locations. Average mobile background measurements of BC, NO_2, O_3 concentrations were 0.008 μg/m^3, 19 μg/m^3 and 40 μg/m^3 respectively. During the first monitoring period, maximum BC (51 μg/m^3) and NO_2 (287 μg/m^3) concentrations were recorded in downwind static and downwind

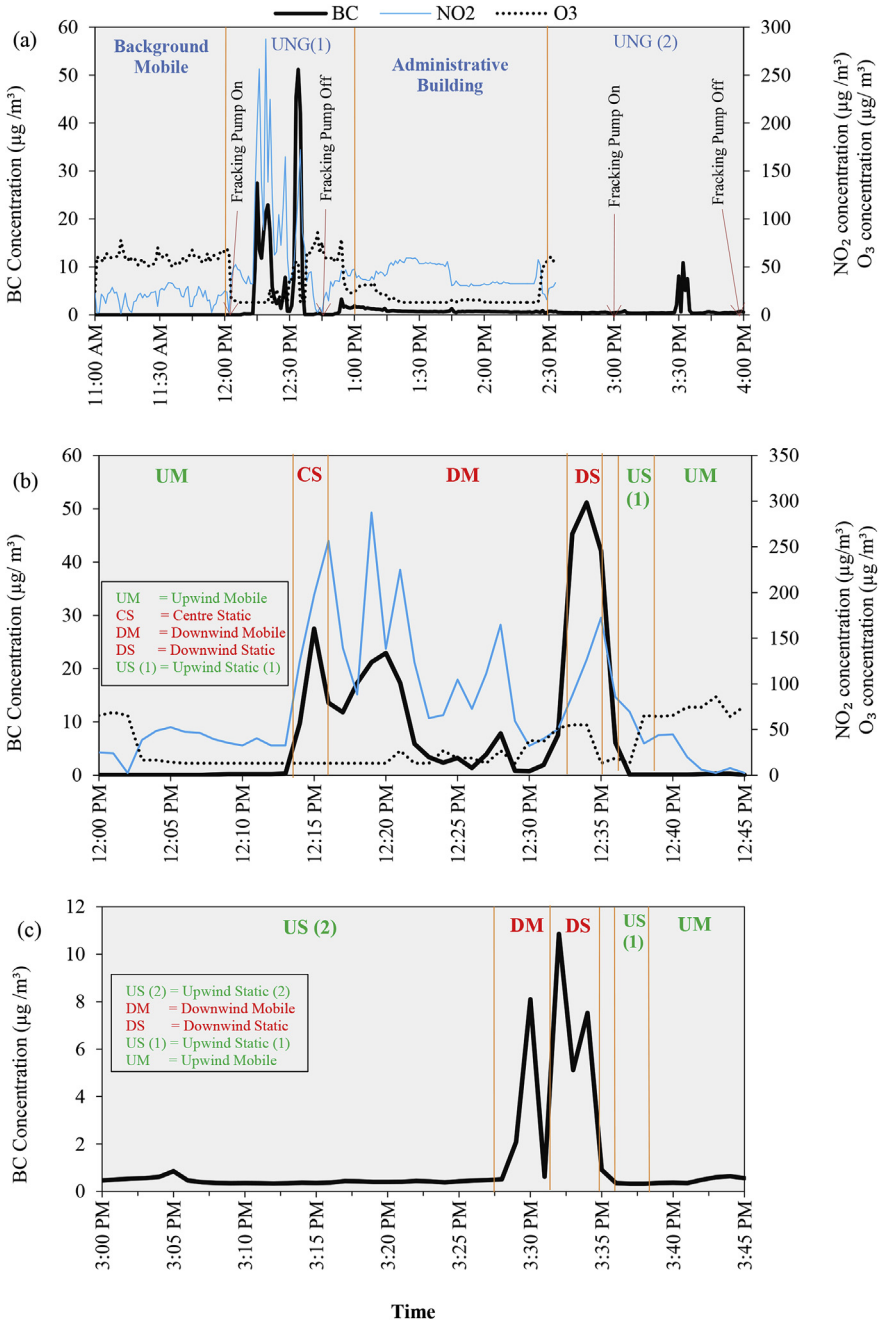

Fig. 3. (a) Air pollutant concentrations measured at different locations within UNG site during both monitoring periods. (b) BC, NO_2 and O_3 concentrations at UNG test site during the first monitoring period, from 12:00 to 12:45. (c) BC concentrations during second monitoring period at UNG test site, between 15:00 to 15:45. All data are presented at 1-min average time resolution.

Table 6
Meteorological and air pollution data obtained from monitoring sites near the UNG site (Fig. 1). Data were obtained from the Warsaw Regional Inspectorate of Environmental Protection's website (http://sojp.wios.warszawa.pl/?par=90). 1-hour resolution data was selected to match the monitoring period on 22 June 2015 between 11:00 to 16:00.

Time	Granica-KPN (Rural Background)				Zyrardow-Roosevelta (Urban Background)					Warszawa-Komunikacyjna (Traffic)				
	Temp (°C)	RH (%)	Wind speed (m/s)	NO_2 (μg/m^3)	Temp (°C)	RH (%)	Wind speed (m/s)	$PM_{2.5}$ (μg/m^3)	PM_{10} (μg/m^3)	Temp (°C)	RH (%)	$PM_{2.5}$ (μg/m^3)	PM_{10} (μg/m^3)	NO_2 (μg/m^3)
11:00	[a]	–	–	–	18.1	47	0.89	4.7	8.5	18.5	46	11.2	30.3	79.2
12:00	19.3	44	2.12	1.9	18.9	44	1.02	4.1	5.6	19.7	40	8.3	19.8	61.4
13:00	19.7	42	2.02	3.5	19.8	42	1.06	2.8	4.2	20.2	38	9.4	24.7	81.3
14:00	19.9	42	1.97	2.3	20.0	41	0.97	6.4	18.3	20.2	38	9.1	22.8	69.7
15:00	19.2	44	1.78	2.4	19.2	42	0.99	9.1	22.6	20.5	37	10.0	28.1	93.9
16:00	19.2	46	2.19	2.8	18.9	49	0.80	11.5	24.0	20.7	37	10.6	24.4	101.4

[a] Data not available.

mobile monitoring locations respectively. In contrast, the highest O_3 concentration (86 μg/m^3) was recorded during mobile upwind measurements. In the first monitoring period, peaks of BC and NO_2 coincided at centre static, downwind mobile and downwind static monitoring locations. BC concentrations were lower during the second monitoring period with several peaks observed at mobile and static downwind monitoring locations. As context, Zhu et al. (2002) compared differences in the concentration of combustion-related pollutants upwind and downwind of a major highway with heavy-duty diesel traffic in Los Angeles, and observed average BC concentration 17 m downwind (21.7 μg/m^3) approximately 5 times higher than average upwind concentration (4.6 μg/m^3).

We examined air pollutant and meteorological data from the nearest rural, urban background and traffic monitoring sites (Fig. 1, Table 6). On the day of measurements at the experimental UNG site average air temperature and relative humidity were 19 °C and 42%, which were expected for the time of year. Average NO_2 concentrations at the rural monitoring site (3 μg/m^3) were lower than 1-h average background monitoring at the UNG experimental site (19 μg/m^3). Average NO_2 downwind of UNG operations at the UNG site (113 μg/m^3; Table S1) was of similar magnitude to maximum hourly NO_2 concentration (101 μg/m^3) at the traffic site in Warsaw (Table 6).

3.3. Contrasting personal exposure to BC and NO_2 in urban traffic and industrial UNG environments

To provide context to the measurements at the UNG site we made measurements of personal exposure of pedestrians to BC and NO_2 in Glasgow city centre. Measurements were made within a few metres of roadways. Marked pollution gradients in both BC and NO_2 were observed on the walking routes depending on proximity to local traffic (Fig. 4 illustrates the spatial pattern of pollutants measured on the final walk selected from 8 separate walks in Glasgow City centre (Table 7)). The spatial pattern was relatively consistent between pollutant metrics.

BC and NO_2 at the UNG site and Glasgow city centre are compared in Fig. 5. Average concentrations of BC and NO_2 measured downwind of the UNG site (11 and 111 μg/m^3 respectively) were approximately 37 times and 4 times higher than the upwind background BC and NO_2 concentrations at the UNG site (0.3 and 21 μg/m^3 respectively) and approximately 3 times higher than in Glasgow city centre (3.7 and 42 μg/m^3). BC measurements made at the UNG site (downwind) were also higher than observations in New York city (1.5–6.2 μg/m^3) reported by Kinney et al. (2000). These comparisons are intended to be indicative only as they are subject to substantial uncertainty associated with varying emission and meteorological conditions.

BC and NO_2 were positively correlated at the UNG site (r = 0.75 for 5-min averaging periods) and Glasgow city centre (r = 0.65 for 5-min averaging periods) (Fig. S3). This is consistent with observed correlation of BC and NO_2 at roadway intersections in Baltimore, USA (r = 0.83 for 2-week averaged data (Riley et al. (2016)); an urban site in Kuopio, Finland (r = 0.84 for 24-h averaged periods (Penttinen et al., 2000)); and at motorway sites in Toronto, Canada (r = 0.89 for 10-min averaged data (Beckerman et al., 2008)). Lower correlations were observed for BC and NO_2 measured at 1-min averaging periods compared to 5-min and 15-min averaging periods (Fig. S3).

We characterised the bivariate relationship between BC and NO_2 by calculating reduced major axis (RMA) regression statistics (Ayers, 2001) and observed similar-magnitude RMA slopes and intercepts at the UNG site and in Glasgow (Table 7). The regression slopes that we calculated in the present study were an order of magnitude less than equivalent statistics calculated in a previous study that compared BC (averages of repeated 5-min measurements using AE51 instrument) and NO_2 concentrations (1-week average measured using passive diffusion tubes) in Glasgow (Gillespie et al., 2017). The higher correlations and different regression slopes observed between BC and NO_2 concentrations in the present study compared to equivalent statistics reported by

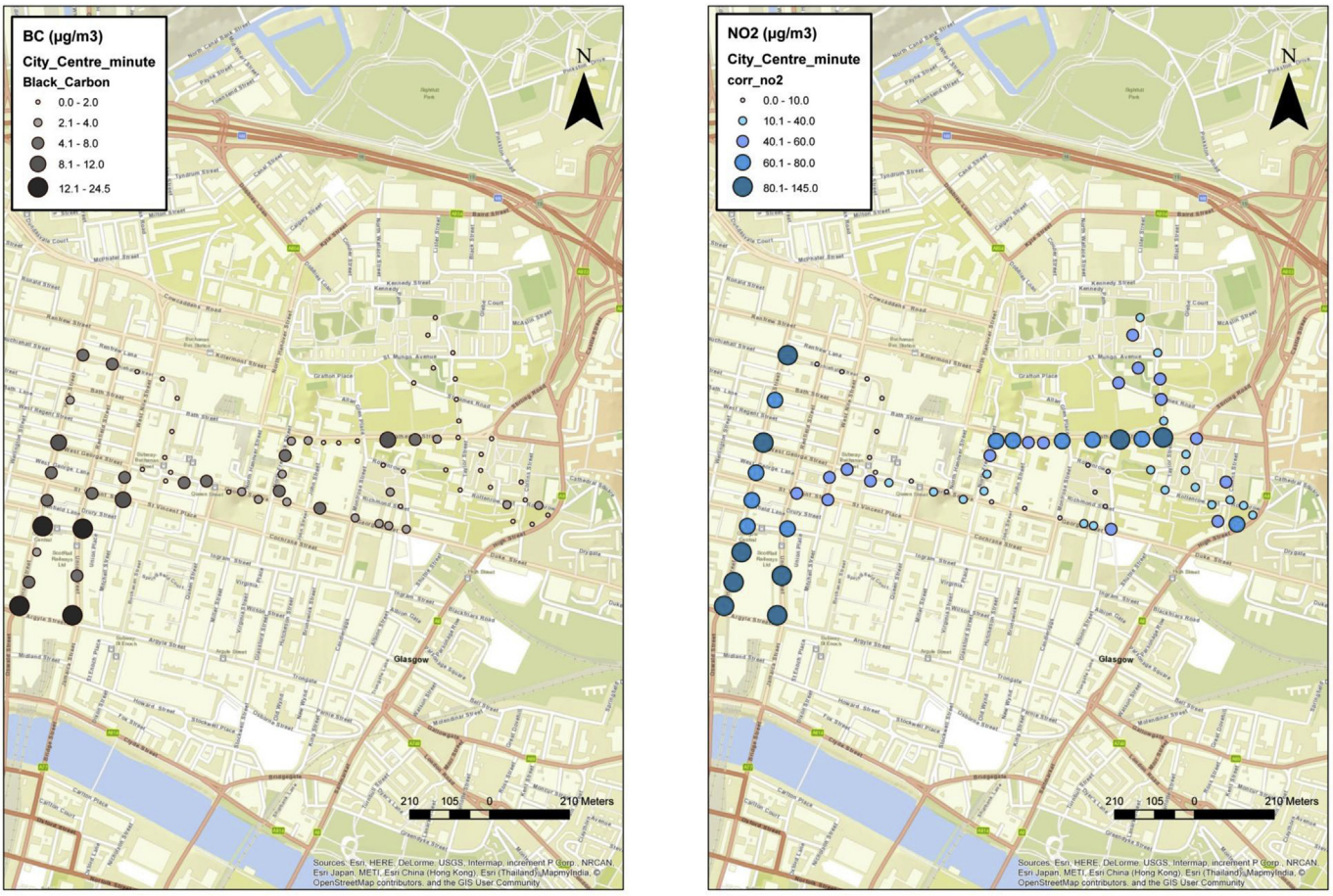

Fig. 4. Examples of mobile measurements of BC (left hand map) and NO_2 (right hand map) made in Glasgow city centre on 24 July 2015.

Table 7
Summary of Reduced Major Axis (RMA) regression statistics for NO_2 *vs*. BC for 5-min averaging periods for mobile measurements in Poland and Glasgow city centre. Equivalent RMA statistics for static 'spot' measurements reported by Gillespie et al. (2017) are given for comparison.

Study name	n	R^2	slope	intercept
UNG test site Poland	42	**0.57**	0.14	−3.81
Glasgow city centre (exc. 6/7/2015 & 15/7/2015)	81	**0.38**	0.12	−2.2
Gillespie et al. (2017)	18	**0.75**	9.12	24.6
Individual study days for Glasgow city centre:				
30/6/2015	12	**0.43**	0.25	−9.54
6/7/2015	15	0.12	0.17	−9.7
9/7/2015	15	**0.64**	0.12	0.10
14/7/2015	15	**0.59**	0.12	−2.96
15/7/2015	14	0.02	0.25	−10.12
22/7/2015	10	**0.43**	0.15	−5.22
23/7/2015	15	**0.65**	0.08	−1.99
24/7/2015	14	**0.42**	0.07	0.87

Significant correlations (P = 0.05) are presented in **bold** text.

Gillespie et al. (2017) may be the result of simultaneous measurement of both metrics over a larger concentration range over shorter time periods.

4. Conclusions and recommendations

We deployed portable BC, NO_2 and O_3 monitors at an experimental UNG test site in Poland and observed that locations downwind of fracking pumps, where site operators frequently work, had average concentrations of BC and NO_2 approximately 3 times higher than in a city centre area in Glasgow, UK. We observed approximately 37-fold and 4-fold increases in BC and NO_2 concentrations between upwind and downwind locations in close proximity to diesel-powered fracking pumps. The concentrations of BC and NO_2 downwind of the fracking pumps at this UNG site were approximately 3 times higher than the average concentrations of these pollutants that we observed in Glasgow city centre.

These observations have important implications for best practice in UNG site design in that control cabins should always, where practical, be situated in the predominantly upwind direction. The site that we worked at was running two to three pumps under simulated ideal operating conditions. In practice sites often run up 15–18 fracking pumps and typical operation time during fracking operations is 100–150 min (Josifovic et al., 2016). Under real operating conditions, the pumps may often run under non-optimal conditions e.g. through partial pump failure, and or increased load through complex geotechnical capacity. Hence further assessment through monitoring the effect of emissions from arrays of fracking pumps under 'typical' operating conditions on nearby air quality is an important recommendation from our study. Extension of our preliminary approach to more detailed monitoring campaigns would inform environmental management decisions about engineering controls e.g. examination of the cost-benefit considerations relevant to potential reduction of diesel emissions from UNG sites through alternative sources of power (e.g. from gas turbines, electric pumps) and measures to reduce power consumption by pumps (Josifovic et al., 2016).

In addition to measuring on-site exposure, the monitoring techniques and equipment that we have used could be adapted to quantify personal exposures to emissions to air pollutants generated during UNG operations, and to quantify exposures in nearby communities. Personal

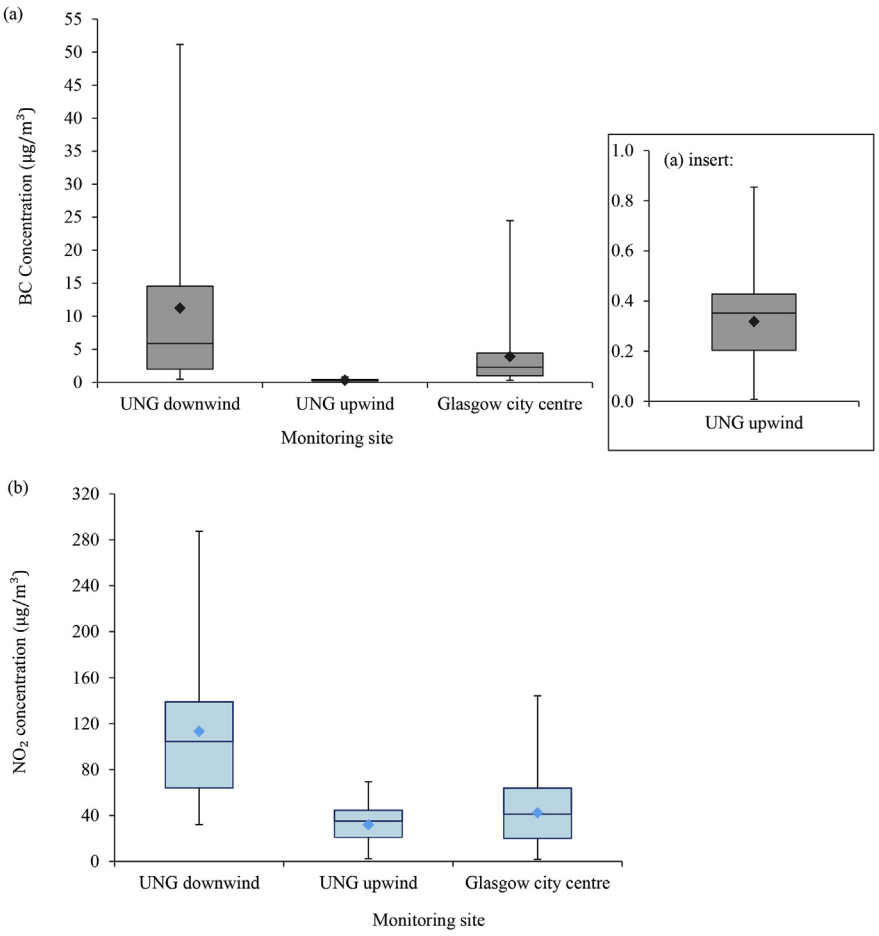

Fig. 5. Descriptive statistics for BC (a) and NO_2 (b) concentrations measured downwind of fracking pumps (all measurements combined from 2 monitoring periods, n = 23), upwind of fracking pumps (all measurements combined from 2 monitoring periods, n = 53) and on walking route in Glasgow city centre (all measurements made on 24 July 2015, n = 72). Underlying data represent pollutant concentrations averaged over 1-min intervals. The plots represent 3rd and 1st quartiles by upper and lower end of box respectively, and 95th and 5th percentiles by ends of upper and lower whiskers respectively. The diamond points represent the arithmetic mean BC and NO_2 concentrations. Numerical values are given in Table S1 in Supplementary Information. Approximately 2% (4 out of 214) of estimated NO_2 concentrations were small negative numbers (censored to 2.5 µg/m^3 as described in section 3.1) as a result of the calibration procedure to remove the effect of O_3 interference on the NO_2 sensor (Lin et al., 2015). Fig. 5 (a insert): expanded detail of BC measurements upwind of fracking pumps.

exposure characterisation would be advantageous to UNG extraction operators by enabling quantitative assessment of the benefits and costs of implementing personal protective equipment (PPE) to reduce employee exposure to diesel engine emissions (HSE, 2012) including assessment of possible use of respirators (Penconek et al., 2013). Characterisation of exposure in nearby communities will benefit from combination of recent developments in portable environmental monitoring instruments with open-access data analytical methods to interpret wind direction effects on observations downwind of emission sources (Azarmi et al., 2016; Grange et al., 2016).

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.06.032.

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