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Point source attribution of ambient contamination events near unconventional oil and gas development

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HIGHLIGHTS

- Air quality characterized *in situ* with mobile mass spectrometry
- Episodic BTEX contaminated events attributed to specific anthropogenic processes
- Emissions from unconventional oil and gas development influence regional air quality

GRAPHICAL ABSTRACT

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ABSTRACT

We present an analysis of ambient benzene, toluene, and xylene isomers in the Eagle Ford shale region of southern Texas. *In situ* air quality measurements using membrane inlet mobile mass spectrometry revealed ambient benzene and toluene concentrations as high as 1000 and 5000 parts-per-billion, respectively, originating from specific sub-processes on unconventional oil and gas well pad sites. The detection of highly variant contamination events attributable to natural gas flaring units, condensate tanks, compressor units, and hydrogen sulfide scavengers indicates that mechanical inefficiencies, and not necessarily the inherent nature of the extraction process as a whole, result in the release of these compounds into the environment. This awareness of ongoing

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contamination events contributes to an enhanced knowledge of ambient volatile organic compounds on a regional scale. While these reconnaissance measurements on their own do not fully characterize the fluctuations of ambient BTEX concentrations that likely exist in the atmosphere of the Eagle Ford Shale region, they do suggest that contamination events from unconventional oil and gas development can be monitored, controlled, and reduced.

1. Introduction

There is a growing societal concern regarding the relationship between unconventional oil and gas development (UD) and environmental quality in petroliferous shale energy basins. Recent investigations have reported the migration of methane gas in shallow groundwater (Jackson et al., 2013; Osborn et al., 2011), the leaching of harmful chemical compounds into the soil and water (Drollette et al., 2015; Hildenbrand et al., 2015; Hildenbrand et al., 2016; Lauer et al., 2016; Llewellyn et al., 2015; Thacker et al., 2015), and the mishandling of produced fluids (Lauer et al., 2016) each of which can have a deleterious effect on the quality of surface and sub-surface water resources.

The majority of the air quality studies performed near UD have focused primarily on the detection and quantification of methane emissions (Allen et al., 2013; Harriss et al., 2015; Yacovitch et al., 2015). A recent review of the literature by Moore et al. attributed rogue hydrocarbon emissions to unconventional drilling and hydrocarbon processing (Moore et al., 2014). Air quality measurements in both the Bakken and Marcellus shale regions have also detected methane emissions from non-sputtering flares, which exhibited a 98% destruction removal efficiency (Caulton et al., 2014a). In these regions the greatest methane emissions were attributed to unidentified venting practices (Caulton et al., 2014a). These particular findings corroborate data collected in the Barnett, Denver-Julesburg, Pinedale, and Western Gulf basins where a weak correlation between emission and production rates was observed, indicating that maintenance-related stochastic processes and the design of production/control equipment are factors determining emission levels (Brantley et al., 2014).

Methane emissions have also been quantified in parts of southwestern Pennsylvania *via* an instrumented aircraft platform. These measurements revealed an average of 34 g of CH_4/s per well from seven well pad sites that were determined to be in the drilling phase, which accounted for up to 30% of the observed regional flux (Caulton et al., 2014b). While a potent greenhouse gas, rogue methane emissions of these proportions likely do not have an immediate effect on human health, as these concentrations are orders of magnitude below what would be required for human asphyxiation. In the Barnett Shale region of northern Texas, several point sources have been identified as potential contributors to the emission of NO_x and volatile organic compounds (VOCs), chemicals that can negatively impact human health. These include compression units (upstream), engine exhausts and condensate and oil tanks, in addition to production, well drilling, hydraulic fracturing, well completions, natural gas processing and transmission lines (midstream), each of which can contribute fugitive emissions intermittently (Armendariz, 2009). Two recent investigations in the Barnett shale region reported the emission of various chemical species from gas production sources, particularly lower molecular weight alkanes (<C6) (Zielinska et al., 2014) and aromatics (Bunch et al., 2014). However, these detections were not detectable beyond a distance of ~100 m in the downwind direction of the source in question, and all ambient

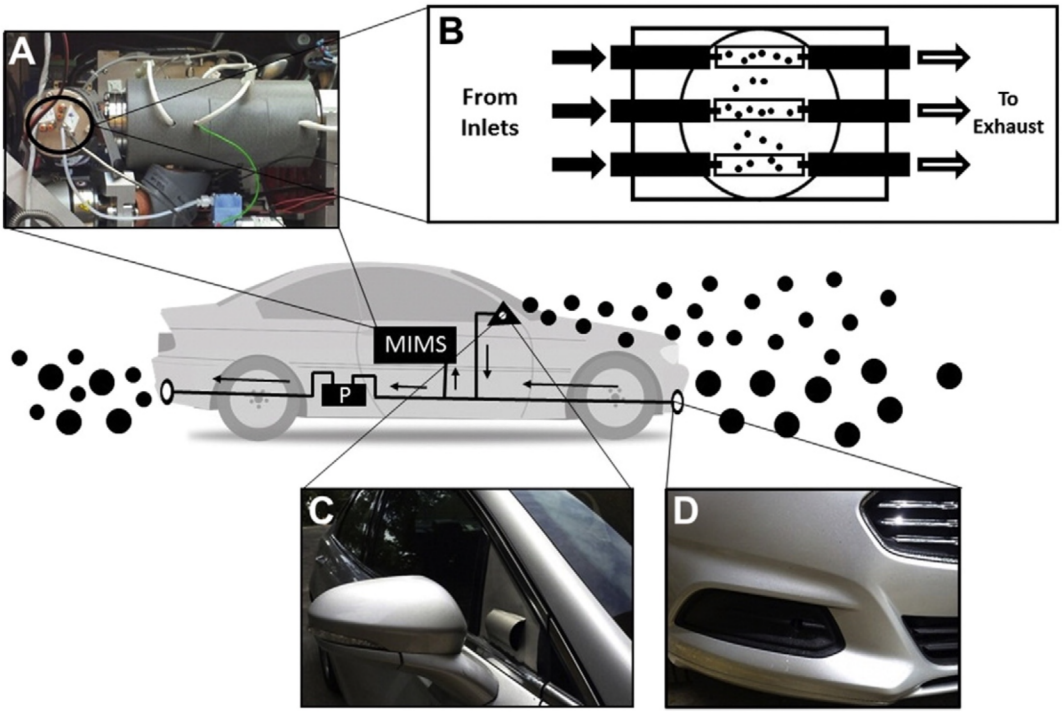

Fig. 1. Anatomy of the MIMS system. (A) Membrane inlet and linear quadrupole housing, (B) membrane inlet schematic, (C) quarter glass inlet, and (D) fog lamp inlet. Sampling of target atmosphere is provided by one of two inlets, either in a custom 'quarter glass' inlet or through a modified foglamp inlet. Atmosphere is subsequently metered for constant flow. Air is then directed into a membrane inlet of the installed mass spectrometer. All gases then exit *via* an exhaust port. Plots are created by associating mass spectra obtained with the GPS location at point of sample.

VOC concentrations were below the health-based safe exposure levels (Zielinska et al., 2014), except for 1,2-dibromoethane, which is not known to be associated with UD (Bunch et al., 2014). On the contrary, in the Marcellus shale region, Litovitz et al. estimated that an individual compressor station (midstream) operating at full capacity could emit 11–45 Mg (10^6 g) of VOC and 46–90 Mg of NO_x per year (Litovitz et al., 2013). These findings were corroborated by mobile measurements revealing individual compressor stations emitting up to 0.162 tons of NO_x, 371 tons of CO_2, and 4.94 tons of CH_4 per day, respectively (Goetz et al., 2015), with similar detected emissions believed to potentially affect compliance with federal ozone standards (Swarthout et al., 2015). With our current understanding it is difficult to quantify the long-term effects of these emissions within the context of human health; however, they do present a significant burden to the surrounding environment.

Previous measurements in the Eagle Ford Shale region have revealed saturated hydrocarbons attributable to UD, which dominate hydroxyl radical reactivity at levels similar to other shale basins (Schade and Roest, 2016), and the observation that total nonmethane hydrocarbon (TNHMC) concentrations were higher in 10/12 mobile monitoring efforts collecting data downwind (relative to upwind) of UD activities (Sullivan, 2014). We wished to build upon these finding by utilizing a novel mobile mass spectrometry platform for the detection, quantification, and point sourcing of ambient VOCs in the Eagle Ford Shale region. Previously, this technology has been used to covertly detect clandestine methamphetamine operations as a result of its ability to identify a multitude of unique chemical species *in situ* (Mach et al., 2015). Measurements were collected throughout the Western Gulf Basin to initially characterize regional air quality within the context of noxious aromatic chemicals and subsequently, to quantify the extent to which UD activities have contributed to the detected abnormalities. These data identify individual point sources of atmospheric contamination in addition to exposing significant variability between individual oil and gas operations employing similar sub-processes.

2. Materials and methods

2.1. Study area

All mobile mass spectrometry measurements were collected throughout the Eagle Ford region in southern Texas. Both sampling excursions in June and November of 2015 were guided by third-party local landowners with extensive knowledge of the local roadways and all data were collected in a blind fashion, without knowledge of the detected masses and/or their concentrations during data collection. Individual UD extraction sites (pads) with producing UD wells were examined in accordance with the rights afforded to the participating mineral rights lessors. Anecdotal cases of contamination where air quality was perceived to have been degraded were also analyzed with full consent of the participating landowners. Collectively, 12,821 full mass scans from 0 to 200 *m/z* were performed *in situ* during the regional and UD pad specific analyses across 13 counties in the Western Gulf Basin.

2.2. Instrumentation

The mass spectrometer (MS) and residual gas analyzer were configured into the passenger seat of an electric hybrid vehicle as had been described previously (Mach et al., 2015). Briefly, atmosphere is introduced into the system through a glass aperture that provides an inlet in which a diaphragm pump continuously pumps through air. Utilizing an atmospheric sampling tube, a smaller diaphragm pump samples a small quantity of air to pass through the tubular membrane inlet to the MS (MIMS) (Fig. 1). The system repeatedly scans masses and tags each scan with associated latitude and longitude coordinates using a Python script and an Arduino-based microcontroller with global positioning system (GPS) capabilities. The post-processing of data utilizes Google Earth to plot the acquired intensities for a given mass, which are represented by concentration-based colored circles on a map. Maps are of one target mass, plotting detected concentration for a given region. Concentration determination is performed in conjunction with a Flexstream (KIN-TEK, Texas, USA) gas standards generator (Supplementary Fig. 1). Instrument calibration is performed by exposing test air to the inlet, with multiple standardized analyte concentrations, while monitoring signal intensity fluctuations given in the MS. This provides a correlatable intensity for a certain concentration. Once maximum diffusion through the membrane has occurred for a given concentration of test air, the signal is stable. The National Oceanic and Atmospheric Agency (NOAA) and the Environmental Protection Agency's (USEPA) Areal Locations of Hazardous Atmospheres (ALOHA) software was used in

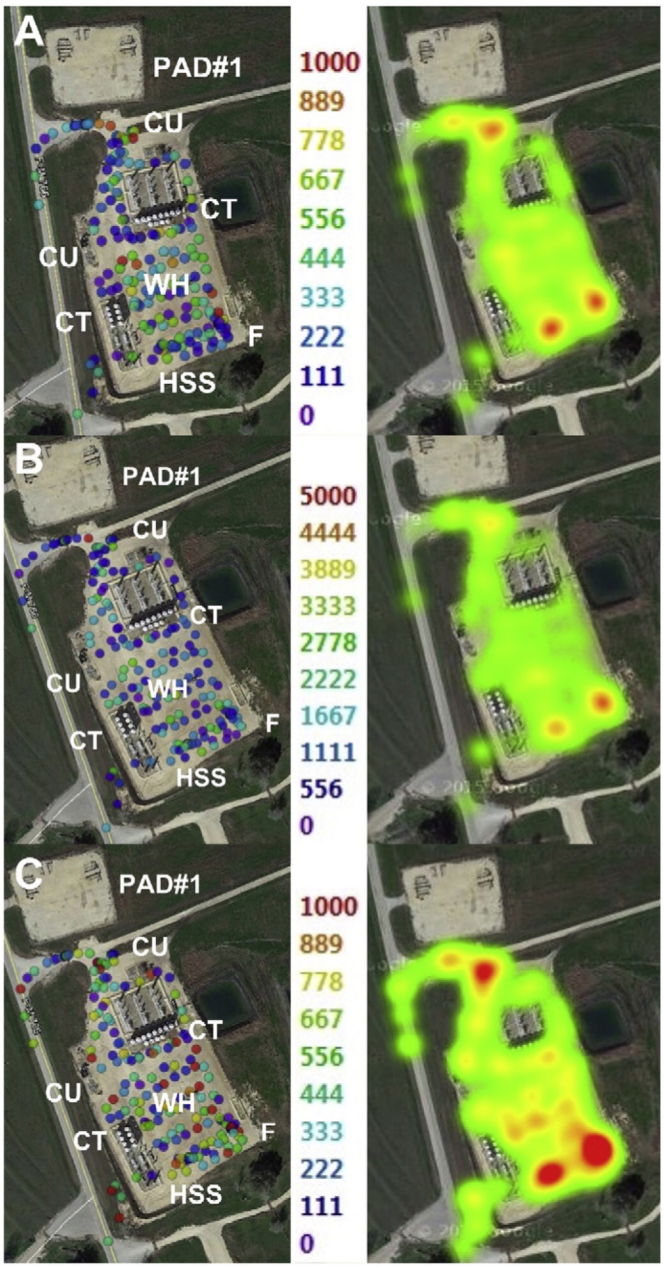

Fig. 2. (A) Benzene, (B) toluene, and (C) total xylene isomer concentrations on and around pad site #1 in the Eagle Ford shale region. The components under investigation include wellheads (WH), condensate tanks (CT), compressor units (CU), H_2S scavenger (HSS), and the gas flaring station (F). Individual concentrations are illustrated in parts-per-billion.

conjunction with local weather data to provide a theoretical interpretation of effluent plume diffusion, which enabled the rendering of heat map concentration gradients.

3. Results and discussion

3.1. Unconventional oil extraction site analysis

With the consent of participating landowners, mobile mass spectrometric measurements were acquired on and around pad sites containing UD extraction activities (Supplementary Table 1). The basic anatomy of an UD pad site in the Eagle Ford region is fairly standard with some variation from pad to pad. This includes, but is not limited to, gas flaring stations, condensate tanks, compressor units, wellheads, heater-treaters, hydrogen sulfide (H_2S) scavengers, and additional auxiliary machinery. By quantifying ambient benzene, toluene, ethylbenzene, and total xylenes (BTEX), it was discovered that the emission of these compounds was highly variable among individual sites. Heat map renderings were generated to illustrate an average of the atmospheric concentrations on each site to identify individual emission sources (Figs. 2 and 3). These data take into account distance between points and the number of points per unit area, and are not artificially represented as a 'hotter' area due to more data points (*i.e.*, extrapolations correct for spatial autocorrelation). Hot regions, represented by red, are higher in concentration compared to green areas (Figs. 2 and 3, right-hand panels). On the first pad site, elevated benzene, toluene, and xylenes concentrations could be attributed to the gas flaring station and a H_2S scavenger on the south side of the site, and condensate tanks and a compressor unit located on the north side of the site (Fig. 2). Similarly, ambient BTEX detected on three other pad sites could be sourced to two flaring stations (pads 2 and 4), condensate tanks (pad 2), and a compressor unit (pad 3) (Supplementary Figs. 2 and 3). However, an analysis of pad 5 revealed very limited BTEX attributable to any of these processes, and a H_2S scavenger was identified as the primary contributing source on pad 6 (Fig. 3). It was also determined that the individual wellheads on each of the six test sites were not contributing to BTEX contamination events. Weather conditions were optimal for the identification of individual point sources with no precipitation and winds <5 mph throughout the collection of the pad site measurements.

The discovery that individual processes are not emitting BTEX into the atmosphere in a systematic and uniform fashion is certainly comforting; however, the operational inefficiencies identified from these data are significant within the context of air quality standards. The Occupational Safety and Health Administration (OSHA) has identified 1000 parts-per-billion (ppb) as the permissible exposure limit

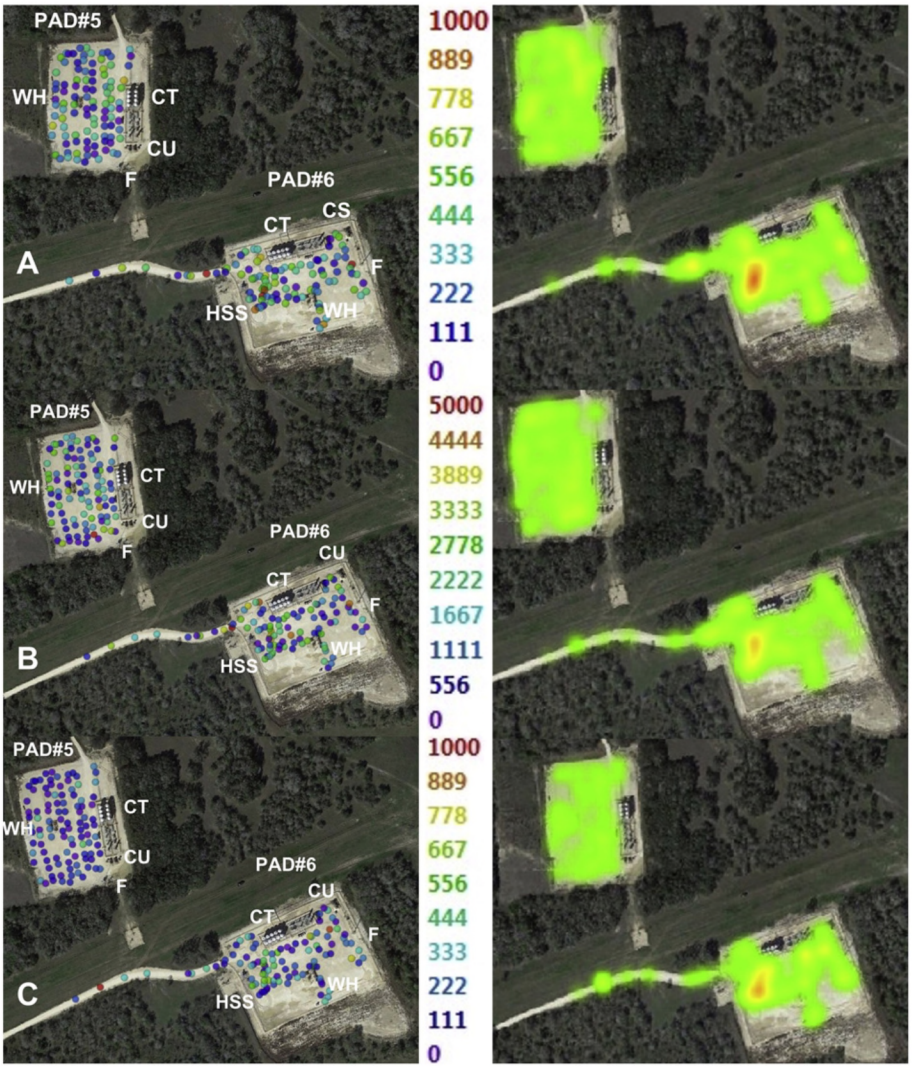

Fig. 3. (A) Benzene, (B) toluene, and (C) total xylene isomer concentrations on and around pad sites #5 and 6 in the Eagle Ford shale region. The components under investigation include wellheads (WH), condensate tanks (CT), compressor units (CU), H_2S scavenger (HSS), and gas flaring stations (F). Individual concentrations are illustrated in parts-per-billion.

(PEL) for benzene in the air as an 8-hour time-weighted average (TWA). However, the OSHA action level is 500 ppb as an 8-hour TWA and the US National Institute for Occupational Safety and Health (NIOSH) maintains a lower recommended exposure limit (REL) of 100 ppb over 8 h. Benzene concentrations did not exceed the 1000 ppb OSHA standard level on any of the six pad sites; however, there were a number of individual measurements above the 500 ppb threshold, particularly on pads 1 and 6 (Figs. 2 and 3). Ambient benzene concentrations of this magnitude would be of concern to human health if they persisted for periods of time >8 h. Toluene was found to be the most prevalent air contaminant, though ambient toluene concentrations did not exceed 5000 ppb, which is well below the OSHA PEL and NIOSH REL of 200,000 and 100,000 ppb, respectively. Similarly, total ambient xylene isomer concentrations did not exceed the NIOSH air quality standard of 100,000 ppb on any of the six pad sites; they were measured to reach a maximum concentration of 1000 ppb. Collectively, these data suggest that individuals working on UD extraction sites for an extended period of time (8 h) could be subjected to potentially harmful levels of ambient benzene if the detected concentrations persisted at or above the observed levels and if the proper safety precautions are not implemented. As it relates to the 6 UD pad sites that were examined in this study, ambient toluene and total xylene isomer concentrations were of far less concern as these light aromatics were found orders of magnitude below their respective occupational health standards.

3.2. Regional air quality analysis

With the detection of ambient BTEX attributable to specific components of the unconventional oil and gas extraction process and the identification of varying degrees of mechanical inefficiencies, we performed a subsequent analysis throughout multiple contiguous counties to assess regional air quality within the context of all Eagle Ford shale activities as a whole (Supplementary Dataset). Air quality measurements collected proximally to producing UD oil wells resulted in detectable BTEX, albeit at lower concentrations than was observed on individual pad sites. While traveling along state highways and local county roads surrounded by active producing UD oil wells, ambient benzene and toluene concentrations did not exceed 500 and 2000 ppb, respectively (1000 ppb benzene and 5000 toluene ppb observed on individual pad sites- Figs. 2 and 3). Environmental factors such as volatilization, degradation, and diffusion likely explain the observed concentration gradient with respect to distance to individual point sources of BTEX emissions (Field et al., 1992). Interestingly, ambient benzene and toluene concentrations were found to be higher in the areas surrounded by UD than the local interstate where heavy traffic and vehicular exhaust, but not neighboring UD activity, were contributing to BTEX and VOC emissions (Fig. 4) (Ho et al., 2009). In areas engaged with producing UD oil wells, including portions of Wilson, Karnes, Dewitt, Gonzales, and Lavaca counties, the ambient concentrations of benzene and toluene ranged between 100 and 400 and 200–2000 ppb, respectively. In particular, a 32 km stretch of county highway surrounding the township of Helena in Karnes County, exhibited the highest concentrations of ambient BTEX (Fig. 4- right panels). Karnes is the most active county in the Western Gulf Basin with 3751 Eagle Ford UD wells as of February of 2015. Furthermore, several measurements collected within 3.2 km of Helena exhibited toluene concentrations orders of magnitude greater (>1600 ppb) than those observed in the rest of the regional dataset (Fig. 5), indicating a single significant or a cluster of multiple contributing point sources.

4. Conclusions

Collectively, these data represent a snapshot of regional air quality in southern Texas with ambient BTEX concentrations within the acceptable limits set forth by OSHA and NIOSH for short-term exposures. However, these measurements on their own do not fully characterize the range of ambient BTEX concentrations that likely exist due to fluctuations in anthropogenic activities and weather. To this point, Brown et al. state that the current monitoring protocols and air quality standards are inadequate given that various pathophysiologies can be associated with an acute exposure to toxic compounds near unconventional oil and gas drilling sites (Brown et al., 2014). This includes respiratory, neurological, and dermal responses as well as vascular bleeding, abdominal pain, nausea, and vomiting (Brown et al., 2014). Subsequent air quality monitoring efforts around UD operations will provide additional characterization contamination events, further elucidating the frequency and duration of emissions from individual point sources. Such efforts will ultimately provide greater context to the measurement of UD-related emissions, allowing us to make better inferences regarding the risks to human health.

Ambient benzene detections near UD gas flares, condensate tanks, and compressor units were found to be as high at 1000 ppb, similar to the ambient levels detected near UD wells in the Barnett shale in northern Texas (Armendariz, 2009). In contrast, elevated levels of any light aromatics compounds (benzene, toluene, *etc.*) were not detected in a recent analysis of Marcellus Shale natural gas development sites (Goetz et al., 2015). Collectively, the highly variable nature of the findings presented here, in relation to other investigations of air quality in

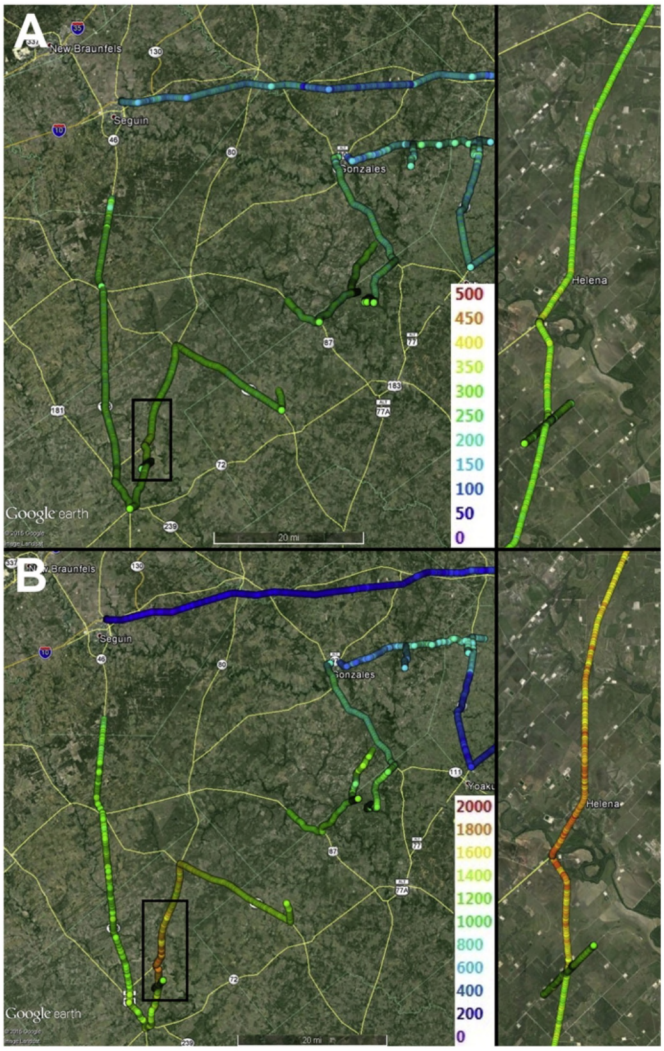

Fig. 4. (A) Benzene and (B) toluene concentrations throughout the counties in the Western Gulf Basin most actively engaged in unconventional oil and gas development. The area illustrated in the two right insets is an area in Karnes County with ambient benzene and toluene concentrations above the background concentrations observed in the surrounding areas. Individual concentrations are illustrated in parts-per-billion.

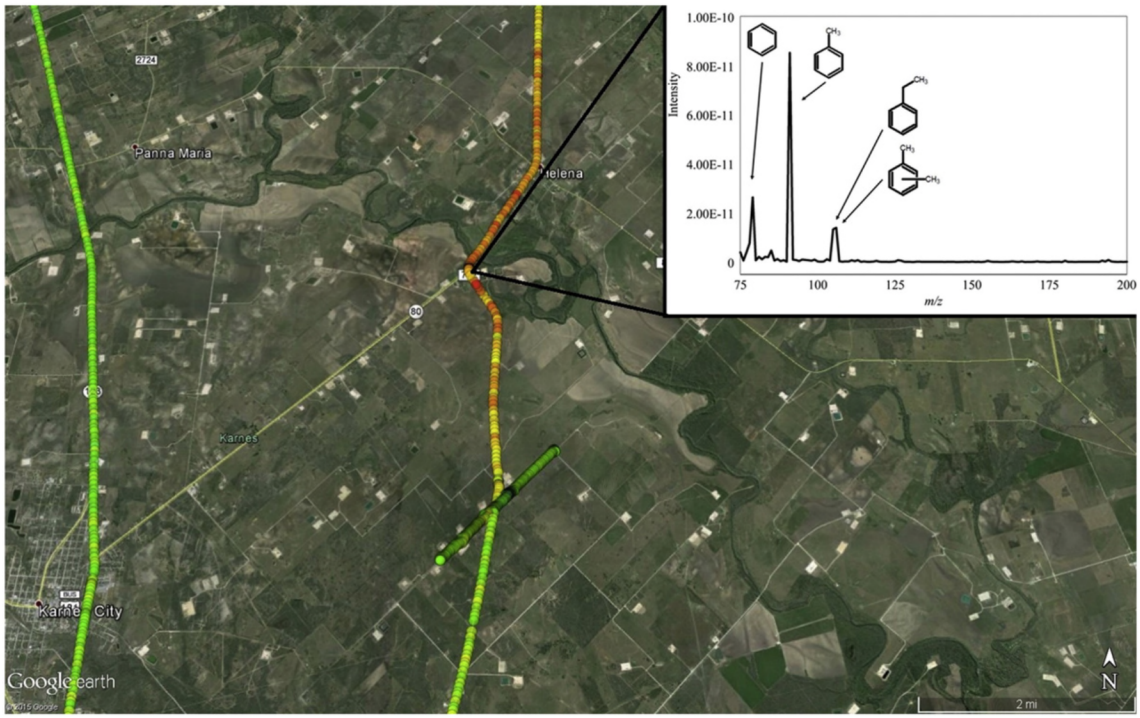

Fig. 5. Elevated toluene concentration in Karnes County, Texas. The upper right-hand inset illustrates chromatographic peaks corresponding to the detection of benzene (78 *m*/*z*), toluene (92 *m*/*z*), and the xylene isomers (106 *m*/*z*), respectively.

other American shale plays, suggests that mechanical inefficiencies from producing UD well pad sites, and not necessarily the inherent nature of the complete UD process, results in contamination events that are currently contributing to ambient BTEX levels on a regional scale. For example, whereas individual H_2S scavenger units were sources of ambient BTEX and triggered notable organoleptic observations of malodors on pads sites 1 and 6, this was not observed on pad sites 2–5. The observed differences are likely attributed to varying levels of operational efficiency, and/or in this case, differences in the solvents that can used during the H_2S scavenging process. Similarly, the variance observed in BTEX emissions resulting from gas flares is likely a function of operational variables that can be adjusted and optimized.

The cumulative effect of these contamination events for citizens living in the impacted areas remains to be determined, as does the relationship between the affected air and other aspects of environmental quality, such as the potential for the concentration of airborne chemicals in surrounding plants and favorable soil types. Subsequent monitoring of ambient BTEX in conjunction with the review of pertinent medical records, through collaboration with local health care officials, will provide greater insight into the human health implications of UD-related air contamination events. Additionally, the use of mobile mass spectrometry to perform air quality monitoring can identify individual point sources of contamination, ultimately guiding better environmental stewardship while improving the efficiency of shale energy extraction.

Author contributions

ZLH, PMM, DDC, KAS, and GFV designed research; ZLH, PMM, EMM, and JTT performed research; ZLH, PMM, EMM, DDC, KAS, and GFV analyzed data; and ZLH, PMM, EMM, DDC, JMM, BEF, KAS, and GFV wrote the paper.

Disclaimer

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.08.118.

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