Response to Reviewers

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.

The authors thank the reviewers for the positive comments regarding the manuscript. Here we respond to the comments from Reviewer 2 only, as there were no recommended corrections from Reviewer 1. Comments from the Reviewer are given in *italics*.

1.) It should be clarified in the introduction that while VOC and NOx from oil and gas operations can drive high ozone, this requires strong inversions and has not been observed in the Bakken.

The section has been reworded (page 4, lines 64-69) to clarify this point.

'Emissions of VOCs and NOx associated with oil and gas extraction can drive elevated ozone concentrations (Olaguer, 2012), which can impact national parks (Rodriguez et al., 2009) and other sensitive areas. High wintertime ozone concentrations have also been associated with oil and gas activities (Ahmadov et al., 2015;Edwards et al., 2014;Helmig et al., 2014;Schnell et al., 2009); however, these wintertime ozone episodes occur during strong inversions, which are not typically observed in the Bakken region.'

2.) The introduction references Howell et al. to say that oil and gas can impact particulate pollution. However, this reference is a study of oil sands, which is a very different process than that occurring in the Bakken, this should be noted.

This is a fair point. However, given that the Introduction is already quite long, rather than adding text to differentiate between processes related to oil extraction in the Bakken versus the Oil Sands, we have simply removed the reference to the *Howell et al.* paper here. The sentence is very general, and does not really need a reference, and the following sentences describe how particle concentrations might be impacted by oil and gas activities. We've also added a new reference regarding fugitive dust emissions, so that the section now reads (page 5, lines 89-94):

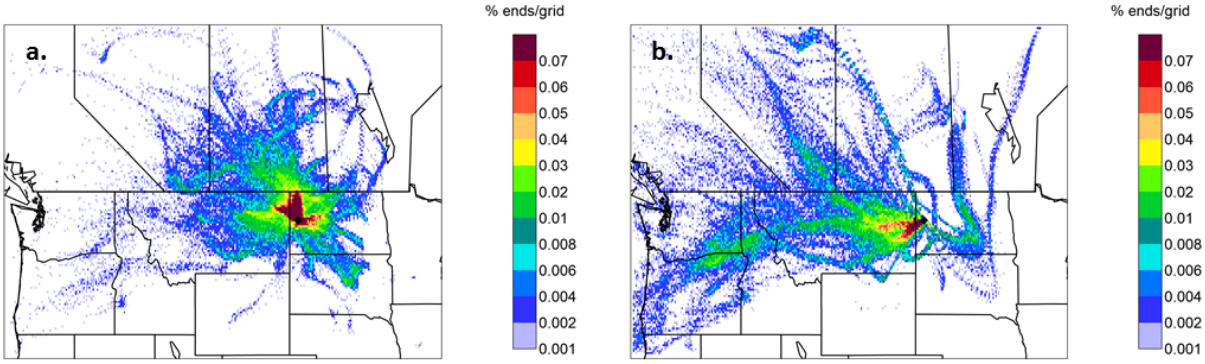
'Ambient particle concentrations also can be impacted by oil and gas activities. Increased particle loading has the potential to degrade visibility (Malm et al., 1994), a protected air quality related value in Class I areas, and can cause adverse health effects (e.g. (Laden et al., 2006)). These particles can be the result of direct emissions, such as fugitive dust from mobile sources (Ilan-Bar et al., 2011), or formed from reactions of precursor species such as SO_2, VOCs, and NO_x.'

3.) It would be good to have some type of figure or more quantitative measures demonstrating the results of the HYSPLIT analysis indicating that higher concentrations were from lower wind speeds (Section 3.1).

A figure has been added to the manuscript: Figure 3 in Section 3.1. All other figures have been renumbered accordingly. The text has been modified slightly to reflect this addition (page 14, lines 270-279).

'To better understand the cause for the elevated concentrations, hourly ensemble back trajectories with a maximum length of 5 days were generated using version 4.9 of the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998), as shown in Figure 3 for NO_2 data

collected during the two study periods. Gridded meteorological data from the 12-km North American Mesoscale Model (NAM12, http://www.emc.ncep.noaa.gov/NAM/.php) (Janjić, 2003) were used as input. During the study period, back trajectory analysis showed that the periods with highest concentrations (top 5%) for SO_2 and NO_2 corresponded to trajectories that were shorter (slower speeds) and were more likely to be impacted by closer sources. In contrast, the lower concentration days had higher wind speeds and winds were preferentially from the west.'



'Figure 3. Back trajectory residence times showing areas where air masses resided during the two days prior to arriving at THRO-N. a) Trajectories arriving when the hourly NO_2 concentration was at the 95th percentile (4.63 ppb) or higher for the two Bakken Studies combined. b) Trajectories arriving when the hourly NO_2 concentration was at the 5th percentile (0.57 ppb) or lower for the combined measurements from both studies. Trajectories were generated using the Hysplit Model in ensemble mode with a start height of 10 m. All trajectories have a duration of two days, so longer lengths correspond to high wind speeds. Note that transport patterns associated with the highest concentrations tend to arrive from a smaller area, indicating probable stagnation, while those associated with the lowest concentrations correspond to travel from more distant areas, indicating high wind speeds, and are more predominantly from the west.'

4.) For Figure 12, it needs to be made more clear if all the species are averaged between 8 a.m. and 4 p.m. measurements as ethane is or if the other species are a single measurement and when that measurement is made.

This point has been clarified in the text and in the Figure caption. In the text (pages 20-21, lines 421-423) it now states:

'NO_x, SO_2 and BC concentrations are daily average values; in contrast, ethane data are calculated as the average of two grab samples per day: one collected in the morning (typically 8 AM), and one collected in the afternoon (typically 4 PM).'

The figure caption has also been updated to read:

'Timeline of ethane, NO_x, SO_2, and BC during the second study period in 2013-2014. NO_x, SO_2 and BC concentrations are daily average values; ethane data are the average of two grab samples per day, one collected in the morning and one collected in the afternoon.'

Other changes

In creating the new figure in response to Comment 3, we found a minor error with the SO_2 dataset that was used to create Figure 3, specifically for the data from THRO-N. The data have been updated, and a revised Figure 3 (now Figure 4) has been uploaded. The revised data do not change the conclusions from the figure.

We have added two new references regarding the data analysis package used throughout the paper (page 16, lines 314-315). The text reads:

'Trend analysis throughout the paper was conducted using the Open Air package in R (Carslaw and Ropkins, 2012; Carslaw, 2014).'

Carslaw, D.C. and K. Ropkins, (2012). openair — an R package for air quality data analysis. Environmental Modelling & Software. Volume 27-28, 52-61.

Carslaw, D.C. (2014). The openair manual — open-source tools for analysing air pollution data. Manual for version 1.0, King's College London.

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

A.J. Prenni[*1], D.E. Day[2], A.R. Evanoski-Cole[3], B.C. Sive[1], A. Hecobian[3], Y. Zhou[3], K.A. Gebhart[4], J.L. Hand[2], A.P. Sullivan[3], Y. Li[3], M.I. Schurman[3], Y. Desyaterik[3], W.C. Malm[2], J.L. Collett Jr. [3]~~B.A. Schichtel[4]~~, and B.A. Schichtel[4]~~J.L. Collett Jr.[3]~~

[1]National Park Service, Air Resources Division, Lakewood, CO, USA

[2]Cooperative Institute for Research in the Atmosphere (CIRA), Colorado State University, Fort Collins, CO, USA

[3]Department of Atmospheric Science, Colorado State University, Fort Collins, CO, USA

[4]National Park Service, Air Resources Division, Fort Collins, CO, USA

*Corresponding author: anthony_prenni@nps.gov

970 491 8414 (Office 1)
303 969 2257 (Office 2)

Submitted *to Atmospheric Chemistry and Physics Discussions*, September 11, 2015

Revised: January 19, 2016

Abstract

The Bakken formation contains billions of barrels of oil and gas trapped in rock and shale. Horizontal drilling and hydraulic fracturing methods have allowed for extraction of these resources, leading to exponential growth of oil production in the region over the past decade. Along with this development has come an increase in associated emissions to the atmosphere. Concern about potential impacts of these emissions on federal lands in the region prompted the National Park Service to sponsor the Bakken Air Quality Study over two winters in 2013-2014. Here we provide an overview of the study and present some initial results aimed at better understanding the impact of local oil and gas emissions on regional air quality. Data from the study, along with long term monitoring data, suggest that while power plants are still an important emissions source in the region, emissions from oil and gas activities are impacting ambient concentrations of nitrogen oxides and black carbon and may dominate recent observed trends in pollutant concentrations at some of the study sites. Measurements of volatile organic compounds also definitively show that oil and gas emissions were present in almost every air mass sampled over a period of more than four months.

1. Introduction

The Williston Basin covers several hundred thousand square kilometers in parts of North Dakota (ND), Montana (MT), South Dakota (SD), Saskatchewan, and Manitoba. In the Bakken and Three Forks formations within the Williston Basin, it is estimated that there are more than 7 billion barrels of recoverable oil (http://www.eia.gov/), making it the largest tight oil play in the United States (US) (EIA, 2014). Despite these vast deposits, it is only in the past decade that horizontal drilling and hydraulic fracturing methods have allowed for cost-efficient extraction of these resources, which has led to exponential growth in the number of wells in the region. Much of this activity is in ND (Figure 1), where there are currently ~10,000 active wells producing over 1 million barrels of oil each day (https://www.dmr.nd.gov/oilgas/). These numbers are expected to grow, with associated increases in pollutant emissions.

Oil and gas development activities provide potential sources of air pollutants during all stages of well development and resource extraction (Brown et al., 2015;Field et al., 2014;Olaguer, 2012;Roy et al., 2014). Emissions come from a large number of small sources that can vary broadly both in terms of absolute amounts and compositions (Field et al., 2014). A large number of studies have focused on methane emissions (e.g. (Brandt et al., 2014;Howarth et al., 2011;Subramanian et al., 2015)), and associated greenhouse warming (e.g. (Jiang et al., 2011)). These issues are particularly significant in the Bakken, where it is estimated that methane emissions correspond to leakages of 9.1% ± 6.2% of energy content (Schneising et al., 2014). Along with methane, other pollutants such as volatile organic compounds (VOCs), including hazardous air pollutants, can be released (Helmig et al., 2014;Olaguer, 2012;Petron et al., 2012;Swarthout et al., 2015). Chemicals used in oil and gas extraction (Colborn et al., 2011)

55 are associated with a wide range of human health hazards, and potential health impacts have
56 been identified for communities near well pads (Bamberger and Oswald, 2015;McKenzie et al.,
57 2012;Steinzor et al., 2013).

58 There are also emissions from the equipment needed for oil and gas development (Roy et al.,
59 2014), including VOCs, nitrogen oxides (NO_x: NO + NO_2), elemental carbon (EC), particulate
60 matter (PM), and sulfur dioxide (SO_2). In 2011, NO_x emissions in the Williston Basin related to
61 oil and gas activities were estimated at 29,400 tons (Grant et al., 2014). NO_x emissions for
62 highway transportation were less than half of this value this same year (EPA National Emissions
63 Inventory), when considering the same counties in the Williston Basin (ND, SD and MT).
64 Emissions of VOCs and NO_x associated with oil and gas extraction can drive elevated ozone
65 concentrations (Olaguer, 2012), which can impact national parks (Rodriguez et al., 2009) and
66 other sensitive areas. High ~~, including in~~ wintertime ozone concentrations have also been
67 associated with oil and gas activities (Ahmadov et al., 2015;Edwards et al., 2014;Helmig et al.,
68 2014;Schnell et al., 2009); however, these wintertime ozone episodes occur during strong
69 inversions, which are not typically observed in the Bakken region.~~, which can impact national~~
70 ~~parks (Rodriguez et al., 2009) and other sensitive areas.~~

71 Support for drilling and operation of active wells has its own associated emissions. For
72 example, in the Marcellus shale region it is estimated that over one thousand diesel trucking
73 events are typical for every new well drilled (Roy et al., 2014). Average traffic counts on
74 Highway 85 in McKenzie County, which runs through the center of the oil and gas activities in
75 the Bakken and is adjacent to the North Unit of Theodore Roosevelt National Park, have more

than tripled from 2008 to 2014 (http://www.dot.nd.gov/road-map/traffic/). Along with increased traffic, increased population to support these activities adds to emissions. The region may also be impacted by pollutants originating in Canada, where there is extensive oil and gas activity regionally (see Figure 1), as well as potential for long range transport of pollutants from the Alberta Oil Sands (Bytnerowicz et al., 2010;Howell et al., 2014;Simpson et al., 2010).

Although oil is the primary commodity in the Bakken, there is also a large amount of associated natural gas. However, because the infrastructure to transport this natural gas is still being developed, until recently about one third of this gas was flared. In 2013, this amounted to over 100 billion cubic feet of natural gas flared or vented in ND (http://www.eia.gov/). These flares add to the pollution burden (Pederstad et al., 2015), producing CO_2, EC, CO and NO_x, particularly in the Bakken where flare efficiencies (Leahey et al., 2001) may be quite low owing to persistent high winds and the presence of surface/pit flares. The percentage of gas that is flared is now declining as a result of new regulations, with proposed limits on flaring of 10% of produced natural gas by 2020.

Ambient particle concentrations also can be impacted by oil and gas activities ~~(Howell et al., 2014)~~. Increased particle loading has the potential to degrade visibility (Malm et al., 1994), a protected air quality related value in Class I areas, and can cause adverse health effects (e.g. (Laden et al., 2006)). These particles can be the result of direct emissions, such as fugitive dust from mobile sources (Ilan-Bar et al., 2011), or formed from reactions of precursor species such as SO_2, VOCs, and NO_x. Monitoring data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) Program suggest that sites in the Bakken region have not experienced

the same improvements to air quality as many places in the United states since 2000. Across the region, annual trends in composition for sulfate, nitrate, and EC are generally decreasing (Figure S1 in Supplemental Material), although within the Bakken trends are mixed and nitrate and sulfate concentrations have increased in the Bakken region during December from 2000-2010 (Hand et al., 2012a). Increasing, although statistically insignificant, trends also were observed at IMPROVE sites in the Bakken region on the 20% haziest days from 2000-2011 (Hand et al., 2014), counter to national trends.

The rapid expansion of the oil and gas sector has the potential to impact four national park units in this region: Fort Union Trading Post National Historic Site (NHS) (FOUS), Knife River Indian Villages NHS (KNRI), and the north and south units of Theodore Roosevelt National Park (THRO-N and THRO-S). THRO is a Class I airshed, which provides for the highest level of federal protection of its air quality, while FOUS and KNRI are Class II airsheds. There are also two US Fish and Wildlife Class I areas nearby: Lostwood, ND (LOST) and Medicine Lake, MT (MELA). In 2011, McKenzie County in ND, where THRO-N is located, accounted for the highest emissions of NO_x, VOCs, PM, CO, and SO_2 from oil and gas in the Williston Basin (Grant et al., 2014), making THRO-N highly vulnerable to impacts from air pollutants related to oil and gas development.

The lack of progress toward the goals of the Regional Haze Rule (EPA, 2003) at THRO and increasing emissions and potential impacts of oil and gas development on air quality in these natural areas prompted the National Park Service to sponsor the Bakken Air Quality Study, carried out in two field deployments in 2013-2014. The locations of the field sites are shown in Figure 1, as well as the locations of long term monitoring sites. Here we provide an overview of

the measurements and determine their representativeness relative to the historical record. A summary of key results is presented, and we address the question of whether energy development in the Bakken region is impacting air quality in national parks and other federal lands in the region.

2. Experimental

2.1. Study Periods

The Bakken Air Quality Study (BAQS) was conducted to assess the mix of pollutants impacting national parks and Class I areas in the Bakken region. Although elevated pollutant levels can occur anytime of the year, measurements were focused primarily on winter months. The first BAQS study period was in 2013, with measurements from February 15 to April 6. The study was conducted at five field sites: FOUS, KNRI, MELA, THRO-N and THRO-S. THRO-N served as the core sampling site. At the core site, high time resolution measurements were made of NO_x, CO, Total Reactive Nitrogen, O_3, SO_2, black carbon and aerosol light scattering. More extensive data were also obtained at lower time resolution (6 hr – 1 week) of organic and inorganic composition of particles and gases. The other four sites were not as heavily instrumented. FOUS, MELA and KNRI had 48 hour integrated samples (6 days a week) of inorganic gas and particulate composition, real time ozone measurements, automated precipitation samplers and Radiello passive samplers, which measured weekly integrated concentrations of SO_2, NO_2, NH_3 and O_3. A nephelometer was also deployed at KNRI. Because THRO-S is heavily instrumented through state and federal monitoring programs, only passive samplers were deployed at this site. Meteorological data were available at all sites. In addition to the sampling sites, two days

of measurements of methane and VOCs were made using a mobile laboratory. A detailed list of measurements from the first study period is given in Table 1.

The second study period ran from November 23, 2013 through March 28, 2014, encompassing the largest increasing trends in sulfate and nitrate as determined from IMPROVE observations (Hand et al., 2012a). During the second study, measurements were limited to three sites, with increased emphasis on higher time resolution data collection. THRO-N remained the core site, while FOUS and MELA served as satellite sites. At THRO-N and FOUS, additional measurements of gas and particle concentrations and compositions were made, including VOC measurements (Table 2). The VOC data provide markers for many of the potential air pollutant sources in this region. Mobile measurement were also conducted (see Methods).

These study periods correspond to months when temperatures are typically below freezing, and where minimum winter temperatures can fall below -30 °C. Based on meteorological data collected during 2002-2013 at Watford City (near THRO-N), MELA, and LOST, the predominant wind direction in the study region was southwesterly and the second most common direction was northwesterly, though air masses can arrive from all directions. There are spatial, diurnal, and seasonal fluctuations around this predominant pattern. Seasonally, air masses from the northwest are most common during fall and winter; this was generally observed during BAQS. Transport from easterly directions is most likely during spring and summer. Average wind speeds in winter were in the range of 3-5 m/s at all of the study sites, with Watford City (near THRO-N) having the slowest and LOST the highest mean speeds.

2.2. Methods

Many of the measurements listed in Tables 1 and 2 will be described in detail in forthcoming publications, and so are not discussed further. Here we provide a brief description of measurements from the Results and Discussion section.

For real time measurements of NO_x and SO_2 during the first study period, sampling was from a common inlet ~3 m above ground level. The sampling line was 0.64 cm OD Teflon tubing. For SO_2 from the real time measurements, a calibration was performed prior to the study. For NO_x, calibrations were conducted daily using certified, traceable standards provided by Airgas (Prenni et al., 2014). Every calibration included zero air and a span concentration, with calibration gases introduced at the sample inlets.

NO_x measurements were made using a chemiluminescence instrument (Teledyne 201E). The technique alternately measures NO directly and measures NO_x by first converting NO_2 to NO using a molybdenum converter. NO is reacted with ozone forming NO_2 in an excited state which emits radiation while decaying to the ground state.

Real time SO_2 measurements were made during the first study period using a Thermo Scientific SO_2 Analyzer (Model 43C), which uses pulsed fluorescence and has a detection limit of 1 ppbv (60 s averaging). During the second study period, and during both studies at the satellite sites, SO_2 concentrations also were derived from University Research Glassware (URG) samplers.

Twenty-four hour samples were collected using URG annular denuder/filter-pack samplers from 8:00 AM to 8:00 AM local time at THRO-N during both study periods. During the first study period, 48 hour samples were also collected at FOUS, MELA and KNRI, covering 6 days per

week. During the second study, 24 hr samples were collected at FOUS, and weekly samples were collected at MELA. Extracted samples were analyzed for inorganic gas and particulate species using ion chromatography (IC). Sample collection and analysis procedures were similar to those described elsewhere (Benedict et al., 2013).

Real time black carbon (BC) data were collected using a multi-wavelength aethalometer at the core site (Magee Scientific AE-31). We follow the recommendation of *Petzold et al.* (2013) in designating aethalometer measurements as BC and measurements from the IMPROVE program as EC. The sample is collected on quartz fiber filter tape and absorption is measured at seven wavelengths from 370-950 nm. For this study, a $PM_{2.5}$ inlet was used and BC mass was determined as the mean of the masses measured from all wavelengths; no further corrections were implemented. Aethalometer data were logged as 5 min averages. The instrument was factory calibrated prior to the first study period and has a sensitivity of <0.1 $\mu g\ m^{-3}$.

The THRO-N site also had an IMPROVE particle monitor that collected 24-hour samples. Samples were collected daily, on the same schedule as the URG samplers (8 AM to 8 AM). Modules A, C, and D were used during the study. Modules A and C collect fine particles ($PM_{2.5}$), while module D collects both fine and coarse particles (PM_{10}). Module A is equipped with a Teflon® filter that is analyzed for $PM_{2.5}$ gravimetric fine mass, elemental concentration, and light absorption. Module C utilizes a quartz fiber filter that is analyzed by thermal optical reflectance (TOR) for organic carbon and EC. Module D utilizes a Teflon filter to determine PM_{10} aerosol mass concentrations gravimetrically. Module A was used during the first study,

and Modules A and C were used during the second study period. Module D was used for a limited time during the second measurement period.

For VOC measurements, whole air samples were collected at THRO-N, FOUS, and MELA, as well as at various locations throughout the Bakken region as part of the mobile measurements. During the second study period, samples were collected into evacuated 2 L passivated, stainless steel canisters. A total of 40 individual VOCs were quantified from the canister samples using a five-channel, three gas chromatograph (GC) analytical system which employed three flame ionization detectors (FIDs), one electron capture detector (ECD) and one mass spectrometer (MS). The gases analyzed included C_2-C_{10} nonmethane hydrocarbons (NMHCs), C_1-C_2 halocarbons, C_1-C_5 alkyl nitrates and reduced sulfur compounds. The analytical system and methodology are similar to those used in previous studies (Russo et al., 2010a;Russo et al., 2010b;Sive, 1998;Swarthout et al., 2013;Swarthout et al., 2015;Zhou et al., 2010). Multiple whole air standards were used during sample analysis (analyzed every 10 samples). The measurement precision, represented by the relative standard deviation of the peak areas for each compound in the whole air standards, was 1-8% for the NMHCs, 3-10% for the halocarbons, 3-8% for the alkyl nitrates and 3-5% for the sulfur compounds. For the second study period, a canister sample was collected twice per day at THRO-N, four times per week at FOUS, and once per week at MELA. For approximately one month of the study (December 19, 2013 – January 31, 2014), canisters were collected only once per week at FOUS.

Meteorological data were collected with a Climatronics All-In-One Weather Sensor (Part Number 102780), co-located with the gas measurements.

~~2.2.1.~~ *Mobile Measurements*

A Picarro A0941 mobile measurement kit combined with a Picarro G2203 analyzer was deployed inside a Chevrolet Tahoe Hybrid vehicle for mobile measurements of CH_4 and C_2H_2. The Tahoe SUV was deployed three times during BAQS for measurements of ambient concentrations of CH_4 and C_2H_2 near oil and gas activities, encompassing both study periods. Mobile nephelometer and BC data were collected during the second study period. Whole air canister grab samples also were collected and analyzed for VOCs as part of the mobile measurements in March 2014. Mobile measurements were conducted while driving ~50 km per hour on mostly main roads throughout the Bakken region. Measurement locations were chosen to represent a combination of areas of high oil and gas activity and locations where little or no oil and gas activities were present. When elevated concentrations of CH_4 were observed, nearby upwind sources were investigated. When a source was confirmed, based on elevated methane concentrations and wind direction, the vehicle was stopped and measurements were made downwind of the site. For the measurements in this manuscript, no vehicular traffic was observed within the operator's visual range.

A detailed description of the Picarro analyzer is presented by *Mønster et al.* (2014). Briefly, a Cavity Ringdown Spectroscopy (CRDS) instrument was used to quantify ambient concentrations of CH_4 and C_2H_2. The inlet of the system was located on a mast secured in front of the vehicle, at a height of 3 meters. Teflon tubing was used to direct the airflow from the inlet to the analyzer at 5 LPM. The A0941 mobile unit was equipped with a Climatronics sonic anemometer for wind speed and direction and a GPS unit for location. Data were collected at 3 Hz.

A microAeth Model AE51 (AethLabs) with a measurement wavelength of 880 nm was used for measuring ambient concentrations of BC. A more detailed description and characterization of the microAeth is presented by *Cai et al.* (2014). One minute data were collected at a flow of 200 ccm. The microAeth inlet was comprised of black conductive tubing (~20 cm long) which was located outside of the back passenger side window of the Tahoe SUV.

3. Results and Discussion

The objectives of the field studies were to provide initial information on the composition and properties of particulate and gaseous pollutants in national park units in the region and at MELA, a Class 1 US Fish and Wildlife area. In this paper we take a broad look at measurements from the study and explore the representativeness of the study time period. We also provide some highlights from the study, with a focus on measurements of NO_x, SO_2, EC/BC and some VOCs, all primary emissions from the energy industry, and address the question of whether energy development in the Bakken is impacting air quality in national parks and other federal lands in the region. More detailed results from all of the measurements, as well as source apportionment, will be presented in forthcoming publications.

3.1. Study Representativeness

Before presenting measurements from these studies, we first examine monitoring data during the two time periods of the intensive field campaigns to determine if the measurement periods were typical for the region. To this end, we use long term air monitoring data from the region (EPA AirData: https://ofmext.epa.gov/AQDMRS/aqdmrs.html). In Figure 2, box plots of mean daily values of SO_2 and NO_2 concentrations at THRO-N are presented for all available data since 2000. Data also are shown separately for each month of the BAQS study periods. As shown in

Figure 2, there was significant variability for SO_2 and NO_2 during the study periods; e.g. daily average concentrations of NO_2 ranged from near zero to 7 ppbv. March and December 2013 showed elevated concentrations of both species at THRO-N, with median values for both months falling above the 75th percentile, and December 2013 having the highest median concentrations for each of these species during the two intensive study periods. Using the Wilcoxon Rank Sum test, we determined that March and December 2013 were the only two months during the studies in which NO_2 and SO_2 were both significantly greater than the historical data ($p<0.05$). To better understand the cause for the elevated concentrations, hourly ensemble back trajectories with a maximum length of 5 days were generated using version 4.9 of the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998), as shown in Figure 3 for NO_2 data collected during the two study periods. Gridded meteorological data from the 12-km North American Mesoscale Model ~~(NAM12)~~ (NAM12, http://www.emc.ncep.noaa.gov/NAM/.php) (Janjić, 2003~~;National_Weather_Service~~) were used as input. During the study period, b~~B~~ack trajectory analysis showed that the periods with highest concentrations (top 5%) for SO_2 and NO_2~~2~~ corresponded to trajectories that were shorter (slower speeds) and were more likely to be impacted by closer sources. In contrast, the lower concentration days had higher wind speeds and winds were preferentially from the west~~northwest~~.

Although NO_2 and SO_2 were significantly higher at THRO-N in March and December 2013, EC concentrations from the IMPROVE network (http://views.cira.colostate.edu/fed/; (Malm et al., 1994)) at THRO-S were not elevated relative to historical data (Figure S2). EC typically peaks in summer, when wildfires influence much of the west. Further, THRO-S is at the southern end of

286 the oil and gas fields and winds at THRO-S are primarily out of the northwest and south, so that
287 THRO-S may be less influenced by oil and gas emissions. Comparing EC concentrations across
288 the region during the study period, we observed an increase in EC in going from THRO-S
289 northward to THRO-N and LOST (discussed further below). Thus aerosol concentrations at
290 THRO-S may be driven more by regional reductions in particulate matter (Figure S1), while sites
291 farther north appear to be impacted by local sources. Considering both THRO-N and THRO-S
292 measurements, we find no instances from the two study periods when the median values for all
293 three species fell outside of the interquartile range, indicating that regionally the study periods
294 were not anomalous relative to past years.

295 **3.2. Changing Emissions and Impacts on Regional Air Quality**

296 Across the United States, emissions from power plants have decreased dramatically in recent
297 decades as the result of legislatively mandated controls, leading to broad improvements in air
298 quality (Hand et al., 2014;Rieder et al., 2013;Sickles and Shadwick, 2015). In the region
299 surrounding the Bakken, annual power plant SO_2 emissions have decreased four-fold over the
300 past 20 years and NO_x emissions have been cut in half (http://ampd.epa.gov/ampd/). At the
301 same time, the number of producing oil and gas wells in the ND Bakken region increased by
302 nearly a factor of 50 (https://www.dmr.nd.gov/oilgas/) from January 2005 to January 2015. To
303 better understand the impact of these changing emissions, we again use long term monitoring
304 data.

305 Figure 4~~3~~ shows annually averaged SO_2 concentrations collected from monitoring sites at six
306 locations in western ND (EPA AirData; Figure 1). THRO-N, THRO-S, and LOST all fall within the
307 area with oil and gas activities, although, as noted above, THRO-S may be influenced more by

regional trends than local sources. Dunn falls on the outskirts of the Bakken region. Beulah and Hannover both lie to the east of most of the activity, near KNRI and several coal-fired power plants (see Figure 1), which represent major sources of SO_2 in the region. As shown in Figure 4~~3~~, SO_2 concentrations are declining throughout the region, particularly at sites closer to the power plants, consistent with observations in the eastern United States (Sickles and Shadwick, 2015;Hand et al., 2012b) and decreasing SO_2 emissions from power plants across the United States. These reductions were determined to be significant at all of the sites in Figure 4~~3~~ except LOST and THRO-N using the Theil-Sen method (Sen, 1968;Theil, 1950) for trend analysis ($p<0.001$; monthly averaged values). Trend analysis throughout the paper was conducted using the Open Air package in R (Carslaw and Ropkins, 2012; Carslaw, 2014)~~)~~.

Despite these reductions, power plants still represent a large source of SO_2 in the region, exceeding that from oil and gas development (http://ampd.epa.gov/ampd/; (Grant et al., 2014)). The influence of SO_2 emissions from regional power plants was observed during BAQS on multiple occasions. In Figure 5~~4~~, SO_2 concentrations from the URG samplers at four sites are shown for the first study period. Data are presented based on the time resolution at which they were collected (24 or 48 hr samples). Data collected using the real time SO_2 instrument at THRO-N were compared to the URG data, and showed reasonable agreement averaging over the same time periods (not shown; $R^2 = 0.87$; real time instrument produced higher values, slope = 1.19). Apparent in the figure are the higher concentrations observed at KNRI, which is located east of the Bakken and very near several power plants (Figure 1). We focus on two high SO_2 events at KNRI during the first measurement campaign: February 20-22, 2013 (8 AM to 8 AM) and March 27-29, 2013 (8 AM to 8 AM). On February 20-22, 48 hour average

concentrations at KNRI were ~8 ppbv, the highest concentrations observed during the study. During this event, none of the other sites had elevated SO_2. Comparing back-trajectories for each of the sites, we see that the air masses which impacted KNRI during this two day period passed directly over several coal-fired power plants (Figure 6~~5~~a), while the air masses reaching the other sites had very little influence from these same plants. Further, the air masses which reached THRO-N during this time period spent minimal time in the Bakken region, and NO_x and BC concentrations at THRO-N were relatively low during this event (Figure ~~6~~7).

During the episode on March 27-29 slower moving air masses with changing wind directions impacted THRO-N, as well as FOUS and MELA, and the airmasses spent more time over the Bakken region (Figure ~~5b~~6b), yielding considerably higher concentrations of NO_x and BC at the core site (Figure ~~6~~7). However, the air masses which reached THRO-N had only a minor influence from emissions sources east of the Bakken, and SO_2 concentrations were again low. While THRO-N, MELA and FOUS were minimally impacted by power plants, KNRI was impacted by several plants on these dates, and KNRI had elevated SO_2 concentrations at this time (Figure ~~4~~5). These observations are consistent with regional power plants largely influencing SO_2 concentrations, and emissions sources from within the Bakken, likely tied to the many sources associated with oil and gas activities, leading to the observed increases in NO_x and BC.

Long-term monitoring data for NO_2 (EPA AirData) are consistent with these observations and provide an interesting contrast to SO_2. Like SO_2, NO_2 concentrations have decreased in Hannover and Beulah (Figure ~~7~~8), east of the Bakken region ($p<0.001$; Theil-Sen method, monthly averaged values), likely driven by decreasing NO_x emissions from power plants. In

351 contrast, THRO-N and Dunn, within and at the outskirts of the oil and gas production region,
352 show no significant trends when considering the entire time period shown. However, when
353 limiting the data to the past 10 years (2005-2014), when oil and gas activities intensified,
354 significant increasing trends in NO_2 are observed at both THRO-N and Dunn ($p<0.001$; Theil-Sen
355 method, monthly averaged values). Finally, LOST shows a significant ($p<0.001$; Theil-Sen
356 method, monthly averaged values) trend of increasing NO_2 throughout the time period shown.
357 These changes are consistent with increasing NO_x emissions from oil and gas activities, which
358 more than doubled in the Williston Basin from 2009 to 2011 and are expected to continue to
359 increase (Grant et al., 2014). Unlike SO_2, NO_x emissions from oil and gas are similar in
360 magnitude to those from regional power plants ((Grant et al., 2014);
361 http://ampd.epa.gov/ampd/). Trends of increasing NO_2 have also been observed in the
362 Marcellus Shale region (Carlton et al., 2014) and the Canadian Oil Sands (McLinden et al., 2012),
363 with increases corresponding to increased activities related to oil and gas extraction.

364 To further explore the influence of oil and gas emissions, we consider monitoring data at LOST.
365 Focusing on NO_2 and segregating measurements by local wind direction, we find significant
366 ($p<0.01$; Theil-Sen method, monthly averaged values) trends of increasing NO_2 only when winds
367 are out of the W, SW and S (Figure ~~8~~9), areas with major oil and gas development. EC
368 concentrations from IMPROVE, which have many of the same sources as NO_2, showed an
369 identical pattern. In contrast, the only significant trend for SO_2 at LOST is for winds out of the
370 south, where concentrations have decreased, likely from decreasing power plant emissions.

371 **3.3. Oil and Gas Impacts on Regional Air Quality**

To better establish a connection between measured pollutants at the study sites and regional oil and gas activities, we consider data from the second study period, November 23, 2013 – March 28, 2014. These measurements included the use of canisters to collect air samples for analysis of key tracer species. Of particular interest for the Bakken are the light alkanes, which serve as markers for oil and gas activity (e.g. (Gilman et al., 2013;Petron et al., 2012;Swarthout et al., 2013;Swarthout et al., 2015)). Figure ~~9~~ 10 summarizes measurements of ethane, propane, n- butane, and n-pentane throughout the campaign at all three sites. The mean ethane and propane mixing ratios from all three sites were 16 and 15 ppbv, respectively, with maximum values approaching 100 ppbv for ethane and 150 ppbv for propane. The i-butane levels ranged from 0.1 ppbv - 22 ppbv, and n-butane peaked over 60 ppbv. The i-pentane and n-pentane had comparable mixing ratios with mean concentrations of ~1.2 ppbv (range <0.1 ppbv - 17 ppbv). These concentrations are significantly higher than typically observed in remote regions (Russo et al., 2010b) and are comparable to levels observed in urban areas known to be influenced by petrochemical industry emissions, as has been observed in other oil and gas basins (Swarthout et al., 2013;Swarthout et al., 2015). Despite variability in absolute concentrations throughout the study, these data provide evidence that emissions related to oil and gas activities were observed at THRO-S, FOUS, and MELA during the second study period.

To better characterize the extent of this impact, we focus on pentane measurements from all of the sites. Recent studies have used the ratio of pentane isomers to identify air masses that are influenced by oil and gas emissions. Although this ratio varies by basin, a ratio of i-pentane to n-pentane which falls at or below one is generally indicative of oil and gas emissions (Swarthout et al., 2013;Swarthout et al., 2015;Gilman et al., 2013), whereas higher ratios correspond to

394 background conditions, largely resulting from automobile emissions and fuel evaporation (e.g.
395 (Russo et al., 2010b)). The i-pentane to n-pentane ratios for all sites for the entire sampling
396 period are shown in Figure 1~~0~~1; the slope is 0.77. Although there is scatter in the data,
397 particularly at the lowest concentrations; only two out of 287 samples at THRO-N, FOUS and
398 MELA had i-pentane to n-pentane ratios that were consistent with background air; all other
399 samples indicated oil and gas influence. These data not only confirm that oil and gas emissions
400 are impacting the region, but also that this influence was present at nearly all times during the
401 second study period.

402 Mobile measurements collected throughout the Bakken region support these data.
403 Background concentrations for CH_4 observed in the Bakken region for the sampling period of
404 December 10-16, 2013 were 2.2 ± 0.4 ppmv, above expected background levels of <2 ppmv for
405 a remote location (Farrell et al., 2013;Wofsy et al., 2011), with peak measured concentrations
406 reaching 16.1 ppmv (1 min average). BC concentrations also were elevated for a remote region,
407 with average concentrations of 900 ± 100 ng m^{-3}. To better demonstrate the direct impact of
408 oil and gas activities on these species in the region, two mobile sampling periods from the
409 Bakken region are shown in Figure 1~~1~~2. One set of measurements was located on the Indian
410 Hill oil field, where there was an active flare at the time of the measurement. The other set of
411 measurements was located on the Painted Woods oil field, with no active flare. Figure 12~~1~~a
412 shows the data collected near the Indian Hill location (active flare) where an increase in CH_4
413 concentrations corresponded to high concentrations of BC. During the flaring, maximum BC
414 concentrations near the site were approximately 4 times higher than the regional BC
415 concentrations. The data collected from the Painted Woods oil field, with no active flare, are

shown in Figure 1~~1~~2b. Without flaring, these measurements show elevated CH_4 concentrations (~7 times above the regional background average), with no corresponding increase in the BC. These areas thus provide sources of VOCs, and, when flaring is present, BC.

Using the light alkanes as markers for local oil and gas activities, we compared alkane concentrations to measurements of NO_x, SO_2 and BC throughout the second study period. Timelines of all of these species are shown in Figure 1~~2~~3 for THRO-N. In the figure, we use ethane as a marker for oil and gas emissions, but all of the light alkanes showed similar results. NO_x, SO_2 and BC~~C~~ concentrations are ~~presented as~~ daily average values~~,~~, in contrast, ~~despite the fact that~~ ethane data are ~~based on~~calculated as the average of two grab samples per day: one collected in the morning (typically 8 AM), and one collected in the afternoon (typically 4 PM). Concentrations of NO_x and BC were correlated with ethane (correlation coefficients, r = 0.75 for NO_x and r = 0.70 for BC) throughout the study period. Although these measurements do not identify which emissions source drives the elevated concentrations for NO_x and BC, the data suggest that VOCs, NO_x and BC likely have collocated sources. SO_2 was not as strongly correlated with ethane (r = 0.42). The lower correlation is presumably because SO_2 comes largely from different sources, as discussed above.

Measurements collected at THRO-N as part of the first intensive study (February – April 2013) also showed that NO_x and EC were correlated (Figure 7~~6~~; r = 0.81 for 1 hour data) with elevated concentrations observed throughout the campaign. Although we did not make routine measurements of VOCs during the first study period, we expect that these measurements were likely impacted by similar sources as observed in the second study period; i.e. oil and gas

related emissions. Hourly NO_x and EC concentrations reached 10 ppbv and 1.3 μg m^{-3}, respectively, during the first study period; the maximum observed hourly SO_2 concentration was just over 9 ppbv. As discussed above, higher concentrations for all species typically were observed during periods of low wind speeds and changing wind directions. Such conditions allow pollutants to accumulate in the region, particularly those with local sources.

The data shown thus far suggest that emissions from oil and gas activities are impacting air quality in the region, raising ambient concentrations of VOCs, NO_x, and EC. Next, we use VOC measurements to estimate the amount of photochemical processing within the air masses that reached THRO-N during the study. To this end, we use the alkyl nitrate to parent hydrocarbon ratios ($R\text{-}ONO_2/R\text{-}H$) to estimate air mass age for measurements at THRO-N (e.g. (Bertman et al., 1995;Russo et al., 2010a;Simpson et al., 2003;Swarthout et al., 2013)). The modeled ratios of 2-pentyl nitrate to n-pentane versus 2-butyl nitrate to n-butane are presented in Figure 1~~4~~3 (solid line) along with the measurements of these species. For the model, a diurnally averaged OH concentration of 5×10^5 molec/cm^3 was assumed. The measured ratios fall on the modeled line, suggesting that the photochemical sources of the alkyl nitrates are reasonably well represented in the model. Results indicate a photochemical processing time of <2 days throughout the majority of the campaign. These results are similar to wintertime VOC measurements in the Denver-Julesburg basin in NE Colorado (Swarthout et al., 2013). For a windspeed of 1.44 m s^{-1}, the median windspeed observed at THRO-N during the second study, a processing time of 2 days corresponds to a transport distance of ~250 km. Because the absolute air mass age determined from the estimates are OH radical concentration dependent, these estimates are subject to uncertainty. However, the results show that processes are occurring

on relatively short time scales and are associated with fresh emissions, rather than aged airmasses, and so point to emissions within the Bakken, rather than long range transport from other oil and gas basins. NO_x and BC concentrations also are shown on the alkyl nitrate evolution plots in Figure 1~~3~~4a and 1~~3~~4b, respectively. These data show that the highest levels of NO_x and BC occur in air masses with short processing times (<12 hours), consistent with the data presented thus far, and further implicating local sources for NO_x and BC; this is particularly relevant for BC, which has a longer atmospheric lifetime. A similar plot is shown for SO_2 in Figure S3.

~~3.3.1.~~ EC Concentrations and Well Counts

If we assume that THRO-S is representative of background aerosol changes, as discussed above, then the ratio of concentrations from surrounding sites relative to THRO-S represents the influence of changing emissions from local sources. As such, we compared the ratio of concentrations from several sites relative to THRO-S, for species monitored as part of the IMPROVE network. For EC measurements made since 2000, all regional IMPROVE sites north of THRO-S show significant increases relative to THRO-S, while regional IMPROVE sites to the south show decreases relative to THRO-S (Figure S4 in the Supplemental Material). These data are consistent with improving regional air quality, and increasing EC sources north of THRO-S, likely from flaring, diesel traffic, and the many diesel engines used in oil and gas activities. THRO-S is likely impacted some from local emissions, but there is a clear gradient in EC trends (Figures S1 and S4). Nitrate shows a similar pattern to EC, with sites to the north increasing relative to THRO-S, and sites to the south decreasing relative to THRO-S. In the case of nitrate, however, not all of these trends are statistically significant. Unlike EC, the trends in nitrate are

confounded by the fact that nitrate is not a primary emission and the monitoring sites are very near the source of precursor NO_x. The ability of NO_x emitted from oil and gas activities to form ammonium nitrate particles, after being converted to HNO_3, also depends on the availability of background ammonia (Li et al., 2014).

Focusing on EC, Figure 15~~4~~ presents a timeline of the ratios of EC concentrations from Figure S4 for two sites north of THRO-S (LOST and MELA), plotted along with data corresponding to oil and gas activities in the region, represented by the number of wells within 100 km of the site of interest. A distance of 100 km was chosen to limit the comparison to development in the Bakken region (see Figure 1). Of the wells that were within 100 km, wells were weighted according to distance from the site as 1/distance, in km, to account for the greater contributions of wells nearer to the sampling sites. For example, a well that is located 1 km from the site would be weighted as 1 well/1 km = 1 well; whereas a well which is 100 km from the site would be weighted as 1 well/100 km = 0.010 well. Well data were downloaded in February 2015 from the relevant state and provincial websites for ND, SD, MT, Saskatchewan and Manitoba. These websites provide different milestone dates from which the years that the wells were completed were estimated. For example, ND includes spud date, while MT includes well completion date. Data also were filtered to include only wells that were active/producing at the time of the download; as such, wells that were active at an earlier date, but were plugged prior to 2015, were not included. Despite these shortcomings, these data provide a reasonable estimate for the year in which wells began operation. As indicated in Figure 14~~5~~, at both sites we observe an increase in EC concentrations at the given site, relative to THRO-S, corresponding to increases in regional oil and gas activities, as designated by well counts. This

503 increase is more evident at LOST, where there is more oil and gas development. For MELA,
504 there is a much smaller increase in EC, relative to THRO-S, corresponding to fewer wells. For
505 both sites, most of the changes occur after about 2008, when oil and gas activities accelerated,
506 further suggesting that oil and gas activities are impacting air quality in national parks and Class
507 1 areas in the region.

508 ## 4. Summary and Conclusions

509 Over the past 10 years, the Bakken region has seen a tremendous increase in oil and gas
510 extraction, such that North Dakota is now the second leading oil producing state in the United
511 States (www.eia.gov). Combined with decreasing emissions from power plants, new emissions
512 sources related to oil and gas activities are playing an increasingly important role in regional air
513 quality. In response to these changes, the Bakken Air Quality Study was conducted to better
514 characterize the impact of these changing emissions sources on federal lands in the region.
515 Measurements were carried out at multiple sites during two study periods (February - April
516 2013; November 2013 – March 2014), along with mobile measurements made throughout the
517 region during select time periods.

518 Results from BAQS demonstrate that oil and gas emissions are impacting air quality at THRO,
519 FOUS, MELA and LOST, with larger effects observed in those areas near the most extensive oil
520 and gas development. The impacts include higher ambient concentrations of VOCs, NO_x and
521 EC, offsetting some of the benefits from decreased power plant emissions. Although the
522 observed concentrations fall well below the National Ambient Air Quality Standards, they are
523 elevated for a remote area, and in some cases are increasing. Continued development is

524 expected to exacerbate these problems, particularly during periods when lower wind speeds
525 allow pollutants to accumulate and react in the atmosphere, forming secondary pollutants.
526 Stagnant air conditions have also been associated with health impacts in regions with
527 unconventional natural gas development (Brown et al., 2015).

528 New state regulations are in place to reduce emissions from flaring, a potentially major source
529 of pollutants in the area. However, even if flaring goals are met by 2020, up to 10% of the
530 produced gas will still be flared, far exceeding the national average. As such, efforts to identify
531 further reductions in emissions are needed to ensure that air quality in federal lands in the
532 region remains unimpaired for the enjoyment of future generations.

533 **Acknowledgements**

534 This project was funded by the National Park Service. The CSU portion of the work was funded
535 by Cooperative Agreement H2370094000, Task Agreement P13AC01187. The authors wish to
536 thank the staffs at THRO, FOUS, MELA, and KNRI for site access and logistical support
537 throughout the study. The assumptions, findings, conclusions, judgments, and views presented
538 herein are those of the authors and should not be interpreted as necessarily representing the
539 NPS. IMPROVE Data: IMPROVE is a collaborative association of state, tribal, and federal
540 agencies, and international partners. US Environmental Protection Agency is the primary
541 funding source, with contracting and research support from the National Park Service. The Air
542 Quality Group at the University of California, Davis is the central analytical laboratory, with ion
543 analysis provided by Research Triangle Institute, and carbon analysis provided by Desert
544 Research Institute. EPA AirData: US Environmental Protection Agency. Air Quality System Data

Mart [internet database] available at http://www.epa.gov/ttn/airs/aqsdatamart. Accessed February 17, 2015.

References

Ahmadov, R., McKeen, S., Trainer, M., Banta, R., Brewer, A., Brown, S., Edwards, P. M., de Gouw, J. A., Frost, G. J., Gilman, J., Helmig, D., Johnson, B., Karion, A., Koss, A., Langford, A., Lerner, B., Olson, J., Oltmans, S., Peischl, J., Petron, G., Pichugina, Y., Roberts, J. M., Ryerson, T., Schnell, R., Senff, C., Sweeney, C., Thompson, C., Veres, P. R., Warneke, C., Wild, R., Williams, E. J., Yuan, B., and Zamora, R.: Understanding high wintertime ozone pollution events in an oil- and natural gas-producing region of the western US, Atmos. Chem. Phys., 15, 411-429, 10.5194/acp-15-411-2015, 2015.

Bamberger, M., and Oswald, R. E.: Long-term impacts of unconventional drilling operations on human and animal health, J. Environ. Sci Heal. A, 50, 447-459, 10.1080/10934529.2015.992655, 2015.

Bar-Ilan, A., Grant, J., Parikh, R., Morris, R., and Henderer D: Oil and Gas Mobile Sources Pilot Study: Final report, pp. 41, 2011.

Benedict, K. B., Day, D., Schwandner, F. M., Kreidenweis, S. M., Schichtel, B., Malm, W. C., and Collett, J. L.: Observations of atmospheric reactive nitrogen species in Rocky Mountain National Park and across northern Colorado, Atmos. Environ., 64, 66-76, 10.1016/j.atmosenv.2012.08.066, 2013.

Bertman, S. B., Roberts, J. M., Parrish, D. D., Buhr, M. P., Goldan, P. D., Kuster, W. C., Fehsenfeld, F. C., Montzka, S. A., and Westberg, H.: Evolution of alkyl nitrates with air-mass age, J. Geophys. Res.-Atmos., 100, 22805-22813, 10.1029/95jd02030, 1995.

Brandt, A. R., Heath, G. A., Kort, E. A., O'Sullivan, F., Petron, G., Jordaan, S. M., Tans, P., Wilcox, J., Gopstein, A. M., Arent, D., Wofsy, S., Brown, N. J., Bradley, R., Stucky, G. D., Eardley, D., and Harriss, R.: Methane Leaks from North American Natural Gas Systems, Science, 343, 733-735, 10.1126/science.1247045, 2014.

Brown, D. R., Lewis, C., and Weinberger, B. I.: Human exposure to unconventional natural gas development: A public health demonstration of periodic high exposure to chemical mixtures in ambient air, J. Environ. Sci Heal. A, 50, 460-472, 10.1080/10934529.2015.992663, 2015.

Bytnerowicz, A., Fraczek, W., Schilling, S., and Alexander, D.: Spatial and temporal distribution of ambient nitric acid and ammonia in the Athabasca Oil Sands Region, Alberta, J. Limnol., 69, 11-21, doi:10.4081/jlimnol.2010.s1.11~~10.3274/jl10-69-s1-03~~, 2010.

Cai, J., Yan, B. Z., Ross, J., Zhang, D. N., Kinney, P. L., Perzanowski, M. S., Jung, K., Miller, R., and Chillrud, S. N.: Validation of MicroAeth (R) as a Black Carbon Monitor for Fixed-Site Measurement and Optimization for Personal Exposure Characterization, Aerosol Air Qual. Res., 14, 1-9, 10.4209/aaqr.2013.03.0088, 2014.

Carlton, A. G., Little, E., Moeller, M., Odoyo, S., and Shepson, P. B.: The data gap: Can a lack of monitors obscure loss of Clean Air Act benefits in fracking areas?, Environ. Sci. Technol., 48, 893-894, dx.doi.org/10.1021/es405672t, 2014.

Carslaw, D.C. and K. Ropkins, (2012). openair — an R package for air quality data analysis. Environmental Modelling & Software. Volume 27-28, 52-61.

Carslaw, D.C. (2014). The openair manual — open-source tools for analysing air pollution data. Manual for version 1.0, King's College London.

Colborn, T., Kwiatkowski, C., Schultz, K., and Bachran, M.: Natural Gas Operations from a Public Health Perspective, Hum. Ecol. Risk Assess., 17, 1039-1056, 10.1080/10807039.2011.605662, 2011.

Draxler, R. R., and Hess, G. D.: An overview of the HYSPLIT_4 modeling system of trajectories, dispersion, and deposition, Aust. Meteorol. Mag., 47, 295-308, 1998.

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., and Zamora, R.: High winter ozone pollution from carbonyl photolysis in an oil and gas basin, Nature, 514, 351-+, 10.1038/nature13767, 2014.

EIA: U.S. Crude Oil and Natural Gas Proved Reserves, 2013, edited by: US Department of Energy, U.S. Energy Information Administration, Washington, D.C., 42, available at: http://www.eia.gov/naturalgas/crudeoilreserves/pdf/usreserves.pdf (last access: April 2015), 2014.

~~in, edited by: Energy, U. S. D. o., U.S. Energy Information Administration, Washington, D.C., 42, 2014.~~

EPA: Guidance for tracking progress under the Regional Haze Rule EPA-454/B-03-004, 96, available at: http://www3.epa.gov/ttnamti1/files/ambient/visible/tracking.pdf (last access: January 2015), 2003~~, in, EPA-454/B-03-004, 96, 2003~~.

Farrell, P., Culling, D., and Leifer, I.: Transcontinental methane measurements: Part 1. A mobile surface platform for source investigations, Atmos. Environ., 74, 422-431, 10.1016/j.atmosenv.2013.02.014, 2013.

Field, R. A., Soltis, J., and Murphy, S.: Air quality concerns of unconventional oil and natural gas production, Environmental Science-Processes & Impacts, 16, 954-969, 10.1039/c4em00081a, 2014.

Gilman, J. B., Lerner, B. M., Kuster, W. C., and de Gouw, J. A.: Source Signature of Volatile Organic Compounds from Oil and Natural Gas Operations in Northeastern Colorado, Environ. Sci. Technol., 47, 1297-1305, 10.1021/es304119a, 2013.

Grant, J., Parikh, R., Bar-Ilan, A., and Morris, R.: Development of baseline 2011 and future year 2015 emissions from oil and gas activity in the Williston Basin: Final report, 103, available at: http://www.wrapair2.org/pdf/2011_2015_Williston_Basin_14Aug2014.pdf (last access: February 2015), 2014.~~in, 103, 2014.~~

Hand, J. L., Gebhart, K. A., Schichtel, B. A., and Malm, W. C.: Increasing trends in wintertime particulate sulfate and nitrate ion concentrations in the Great Plains of the United States (2000-2010), Atmos. Environ., 55, 107-110, 10.1016/j.atmosenv.2012.03.050, 2012a.

Hand, J. L., Schichtel, B. A., Malm, W. C., and Pitchford, M. L.: Particulate sulfate ion concentration and SO2 emission trends in the United States from the early 1990s through 2010, Atmos. Chem. Phys., 12, 10353-10365, 10.5194/acp-12-10353-2012, 2012b.

Hand, J. L., Schichtel, B. A., Malm, W. C., Copeland, S., Molenar, J. V., Frank, N., and Pitchford, M.: Widespread reductions in haze across the United States from the early 1990s through 2011, Atmos. Environ., 94, 671-679, 10.1016/j.atmosenv.2014.05.062, 2014.

Helmig, D., Thompson, C. R., Evans, J., Boylan, P., Hueber, J., and Park, J. H.: Highly Elevated Atmospheric Levels of Volatile Organic Compounds in the Uintah Basin, Utah, Environ. Sci. Technol., 48, 4707-4715, 10.1021/es405046r, 2014.

Howarth, R. W., Santoro, R., and Ingraffea, A.: Methane and the greenhouse-gas footprint of natural gas from shale formations, Climatic Change, 106, 679-690, 10.1007/s10584-011-0061-5, 2011.

Howell, S. G., Clarke, A. D., Freitag, S., McNaughton, C. S., Kapustin, V., Brekovskikh, V., Jimenez, J. L., and Cubison, M. J.: An airborne assessment of atmospheric particulate emissions from the processing of Athabasca oil sands, Atmos. Chem. Phys., 14, 5073-5087, 10.5194/acp-14-5073-2014, 2014.

Janjić, Z. I.: A nonhydrostatic model based on a new approach, Meteorol. Atmos. Phys., 82, 271–285, 2003.

Jiang, M., Griffin, W. M., Hendrickson, C., Jaramillo, P., VanBriesen, J., and Venkatesh, A.: Life cycle greenhouse gas emissions of Marcellus shale gas, Environ. Res. Lett., 6, 03414, doi: 10.1088/1748-9326/6/3/034014, 2011.

Laden, F., Schwartz, J., Speizer, F. E., and Dockery, D. W.: Reduction in fine particulate air pollution and mortality - Extended follow-up of the Harvard six cities study, Am. J. Resp. Crit. Care, 173, 667-672, 10.1164/rccm.200503-443OC, 2006.

Leahey, D. M., Preston, K., and Strosher, M.: Theoretical and observational assessments of flare efficiencies, J. Air Waste Manage., 51, 1610-1616, 2001.

Li, Y., Schwandner, F. M., Sewell, H. J., Zivkovich, A., Tigges, M., Raja, S., Holcomb, S., Molenar, J. V., Sherman, L., Archuleta, C., Lee, T., and Collett, J. L.: Observations of ammonia, nitric acid, and fine particles in a production region, Atmos. Environ., 83, 80-89, 10.1016/j.atmosenv.2013.10.007, 2014.

Malm, W. C., Sisler, J. F., Huffman, D., Eldred, R. A., and Cahill, T. A.: Spatial and seasonal trends in particle concentration and optical extinction in the United States, J. Geophys. Res.-Atmos., 99, 1347-1370, 10.1029/93jd02916, 1994.

McKenzie, L. M., Witter, R. Z., Newman, L. S., and Adgate, J. L.: Human health risk assessment of air emissions from development of unconventional natural gas resources, Sci. Total Environ., 424, 79-87, 10.1016/j.scitotenv.2012.02.018, 2012.

McLinden, C. A., Fioletov, V., Boersma, K. F., Krotkov, N., Sioris, C. E., Veefkind, J. P., and Yang, K.: Air quality over the Canadian oil sands: A first assessment using satellite observations, Geophys. Res. Lett., 39, L04804, 10.1029/2011gl050273, 2012.

Monster, J. G., Samuelsson, J., Kjeldsen, P., Rella, C. W., and Scheutz, C.: Quantifying methane emission from fugitive sources by combining tracer release and downwind measurements - A sensitivity analysis based on multiple field surveys, Waste Manage., 34, 1416-1428, 10.1016/j.wasman.2014.03.025, 2014.

~~North American Mesoscale Forecast System (NAM) access: accessed 26 Jul 2012, 2012.~~

Olaguer, E. P.: The potential near-source ozone impacts of upstream oil and gas industry emissions, J. Air Waste Manage., 62, 966-977, 10.1080/10962247.2012.688923, 2012.

Pederstad, A., Gallardo, M., and Saunier, S.: Improving utilization of associated gas in US tight oil fields, 67 pp., available at: http://www.catf.us/resources/publications/view/212, last access: April 2015.

~~http://www.catf.us/resources/publications/view/212, 67, 2015.~~

Petron, G., Frost, G., Miller, B. R., Hirsch, A. I., Montzka, S. A., Karion, A., Trainer, M., Sweeney, C., Andrews, A. E., Miller, L., Kofler, J., Bar-Ilan, A., Dlugokencky, E. J., Patrick, L., Moore, C. T., Jr., Ryerson, T. B., Siso, C., Kolodzey, W., Lang, P. M., Conway, T., Novelli, P., Masarie, K., Hall, B., Guenther, D., Kitzis, D., Miller, J., Welsh, D., Wolfe, D., Neff, W., and Tans, P.: Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study, J. Geophys. Res.-Atmos., 117, 10.1029/2011jd016360, 2012.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S. M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., and Zhang, X. Y.: Recommendations for reporting "black carbon" measurements, Atmos. Chem. Phys., 13, 8365-8379, 10.5194/acp-13-8365-2013, 2013.

Prenni, A. J., Levin, E. J. T., Benedict, K. B., Sullivan, A. P., Schurman, M. I., Gebhart, K. A., Day, D. E., Carrico, C. M., Malm, W. C., Schichtel, B. A., Collett, J. L., Jr., and Kreidenweis, S. M.: Gas-phase reactive nitrogen near Grand Teton National Park: Impacts of transport, anthropogenic emissions, and biomass burning, Atmos. Environ., 89, 749-756, 10.1016/j.atmosenv.2014.03.017, 2014.

Rieder, H. E., Fiore, A. M., Polvani, L. M., Lamarque, J. F., and Fang, Y.: Changes in the frequency and return level of high ozone pollution events over the eastern United States following emission controls, Environ. Res. Lett., 8, 014012, doi:10.1088/1748-9326/8/1/014012, 2013.

Rodriguez, M. A., Barna, M. G., and Moore, T.: Regional Impacts of Oil and Gas Development on Ozone Formation in the Western United States, J. Air Waste Manage., 59, 1111-1118, 10.3155/1047-3289.59.9.1111, 2009.

Roy, A. A., Adams, P. J., and Robinson, A. L.: Air pollutant emissions from the development, production, and processing of Marcellus Shale natural gas, J. Air Waste Manage., 64, 19-37, 10.1080/10962247.2013.826151, 2014.

Russo, R. S., Zhou, Y., Haase, K. B., Wingenter, O. W., Frinak, E. K., Mao, H., Talbot, R. W., and Sive, B. C.: Temporal variability, sources, and sinks of C-1-C-5 alkyl nitrates in coastal New England, Atmos. Chem. Phys., 10, 1865-1883, 2010a.

Russo, R. S., Zhou, Y., White, M. L., Mao, H., Talbot, R., and Sive, B. C.: Multi-year (2004-2008) record of nonmethane hydrocarbons and halocarbons in New England: seasonal variations and regional sources, Atmos. Chem. Phys., 10, 4909-4929, 10.5194/acp-10-4909-2010, 2010b.

691 Schneising, O., Burrows, J. P., Dickerson, R. R., Buchwitz, M., Reuter, M., and Bovensmann, H.: Remote
692 sensing of fugitive methane emissions from oil and gas production in North American tight geological
693 formations, Earth's Future, 1-11, 10.1002/2014EF000265, 2014.

694 Schnell, R. C., Oltmans, S. J., Neely, R. R., Endres, M. S., Molenar, J. V., and White, A. B.: Rapid
695 photochemical production of ozone at high concentrations in a rural site during winter, Nat. Geosci., 2,
696 120-122, 10.1038/ngeo415, 2009.

697 Sen, P. K.: Estimates of the regression coefficient based on Kendall's tau, J. Am. Stat. Assoc., 63, 1379-
698 1389, 1968.

699 Sickles, J. E., II, and Shadwick, D. S.: Air quality and atmospheric deposition in the eastern US: 20 years of
700 change, Atmos. Chem. Phys., 15, 173-197, 10.5194/acp-15-173-2015, 2015.

701 Simpson, I. J., Blake, N. J., Blake, D. R., Atlas, E., Flocke, F., Crawford, J. H., Fuelberg, H. E., Kiley, C. M.,
702 Meinardi, S., and Rowland, F. S.: Photochemical production and evolution of selected C-2-C-5 alkyl
703 nitrates in tropospheric air influenced by Asian outflow, J. Geophys. Res.-Atmos., 108,
704 10.1029/2002jd002830, 2003.

705 Simpson, I. J., Blake, N. J., Barletta, B., Diskin, G. S., Fuelberg, H. E., Gorham, K., Huey, L. G., Meinardi, S.,
706 Rowland, F. S., Vay, S. A., Weinheimer, A. J., Yang, M., and Blake, D. R.: Characterization of trace gases
707 measured over Alberta oil sands mining operations: 76 speciated C-2-C-10 volatile organic compounds
708 (VOCs), CO2, CH4, CO, NO, NO2, NOy, O-3 and SO2, Atmos. Chem. Phys., 10, 11931-11954, 10.5194/acp-
709 10-11931-2010, 2010.

710 Sive, B. C.: Atmospheric nonmethane hydrocarbons: Analytical methods and estimated hydroxyl radical
711 concentrations, University of California, Irvine, 1998.

712 Steinzor, N., Subra, W., and Sumi, L.: Investigating links between shale gas development and health
713 impacts through a community survey project in Pennsylvania, New Solutions, 23, 55-83, 2013.

714 Subramanian, R., Williams, L. L., Vaughn, T. L., Zimmerle, D., Roscioli, J. R., Herndon, S. C., Yacovitch, T. I.,
715 Floerchinger, C., Tkacik, D. S., Mitchell, A. L., Sullivan, M. R., Dallmann, T. R., and Robinson, A. L.:
716 Methane Emissions from Natural Gas Compressor Stations in the Transmission and Storage Sector:
717 Measurements and Comparisons with the EPA Greenhouse Gas Reporting Program Protocol, Environ.
718 Sci. Technol., 49, 3252-3261, doi:10.1021/es506025~~810.1021/e35060258~~, 2015.

719 Swarthout, R. F., Russo, R. S., Zhou, Y., Hart, A. H., and Sive, B. C.: Volatile organic compound
720 distributions during the NACHTT campaign at the Boulder Atmospheric Observatory: Influence of urban
721 and natural gas sources, J. Geophys. Res.-Atmos., 118, 10614-10637, 10.1002/jgrd.50722, 2013.

722 Swarthout, R. F., Russo, R. S., Zhou, Y., Miller, B. M., Mitchell, B., Horsman, E., Lipsky, E., McCabe, D. C.,
723 Baum, E., and Sive, B. C.: Impact of Marcellus Shale Natural Gas Development in Southwest Pennsylvania
724 on Volatile Organic Compound Emissions and Regional Air Quality, Environ. Sci. Technol., 49, 3175-3184,
725 10.1021/es504315f, 2015.

726 Theil, H.: A rank-invariant method of linear and polynomial regression analysis, I, II and III, Proceedings
727 of the Koninklijke Nederlandse Akademie Wetenschappen, Series A – Mathematical Sciences, Statistical

Department of the “Mathematisch Centrum”, Amsterdam, the 25 Netherlands, 50, 386–392, 521–525, 1397–1412, 1950.

~~50, 386–392, 521–525, 1397–1412, 1950.~~

Wofsy, S. C., Team, H. S., Cooperating Modellers, T., and Satellite, T.: HIAPER Pole-to-Pole Observations (HIPPO): fine-grained, global-scale measurements of climatically important atmospheric gases and aerosols, Philos. T. Roy. Soc. A, 369, 2073-2086, 10.1098/rsta.2010.0313, 2011.

Zhou, Y., Shively, D., Mao, H., Russo, R. S., Pape, B., Mower, R. N., Talbot, R., and Sive, B. C.: Air Toxic Emissions from Snowmobiles in Yellowstone National Park, Environ. Sci. Technol., 44, 222-228, 10.1021/es9018578, 2010.

Table 1. Measurements from the first field campaign: February 15 - April 6, 2013.

Measurement Method	Measured Species	Time Resolution of available data	Notes	THRO-S	THRO-N	FOUS	KNRI	MELA
URG annular denuder/ filter-pack sampler	$PM_{2.5}$ inorganic ions; NH_3, HNO_3, and SO_2	See Methods section	Analysis with Dionex IC system		X	X	X	X
IMPROVE module A	$PM_{2.5}$ mass, elemental composition	24 hr sample THRO-N: daily; Existing: every 3 days	Per IMPROVE protocol	Existing	X			Existing
Teledyne O_3 or portable ozone monitors (POMs)	Ozone	Teledyne: 1 min POMS: 1 hr	Teledyne 400E at THRO-N; 2B Technologies at other sites	Existing	X	X	X	X
Continuous gaseous samplers	NO_x, NO, NO_2, SO_2, CO	1 min	See Methods section		X			
Automated precipitation (rain/snow) sampler	Wet Deposition	THRO-N: Samples collected daily; Satellite Sites: Twice per week	NCON Atmospheric Deposition Sampler/ National Trends Network (ADS/NTN); Yankee Envir. TPC 3000	Existing	X	X	X	X
Nephelometer	Particle light scattering	5 min	THRO-N: Radiance Research; KNRI: Ecotech		X		X	
Aethalometer	Black Carbon	5 min	Magee Scientific 7 wavelength		X			
Passive samplers	SO_2, NO_2, NH_3 and O_3	1 week	Radiello	X	X	X	X	X
Meteorological station	Surface meteorology	1 min at THRO-N and FOUS	Climatronics All-In-One Weather Sensor	Existing	X	X	Existing	Existing
Mobile Sampling	Methane and acetylene	3 Hz	Picarro G2203 with mobile kit A0941					

Table 2. Measurements from the second field campaign: November 23, 2013 - March 28, 2014.

Measurement Method	Measured Species	Time Resolution of available data	Notes	THRO-S	THRO-N	FOUS	KNRI	MELA
URG annular denuder/ filter-pack sampler	$PM_{2.5}$ inorganic ions; NH_3, HNO_3, and SO_2	See Methods section	Analysis with Dionex IC system		X	X		X
IMPROVE module A	$PM_{2.5}$ mass, elemental composition	24 hr sample THRO-N: daily; Existing: every 3 days	Per IMPROVE protocol	Existing	X			Existing
IMPROVE module C	$PM_{2.5}$ OC and EC	24 hr sample THRO-N: daily; Existing sites: every 3 days	Per IMPROVE protocol	Existing	X			Existing
Aerosol Mass Spectrometer	PM_1 nitrate, sulfate, ammonium, organics	5 min	Aerodyne High Resolution Time of Flight		X			
MARGA (Monitor for Aerosol and Gases)	$PM_{2.5}$ Inorganic ions; Gaseous NH_3, HNO_3, and SO_2	1 hour	Applikon 1S		X			
Teledyne O_3 or portable ozone monitors (POMs)	Ozone	Teledyne: 1 min POMS: 1 hr	Teledyne 400E at THRO-N; 2B Technologies at other sites	Existing	X	X		X
Continuous gaseous samplers	NO_x, NO, NO_2, CO, NO_y	1 min	See Methods section		X			
Automated precipitation (rain/snow) sampler	Wet Deposition	THRO-N: Samples Collected after precipitation	NCON ADS/NTN Sampler	Existing	X			
Nephelometer	Particle light scattering	5 min	THRO-N: Radiance Research; FOUS: Optec; MELA: Ecotech		X	X		X

Aethalometer	Black Carbon	5 min	Magee Scientific 7 wavelength	X		
TEOM	$PM_{2.5}$ Mass	6 min	Thermo Scientific 1405-DF	X		
VOC canisters	VOCs	THRO-N: Twice per day; FOUS: 4 times per week; MELA: once per week	Analysis with 5-channel GC system; FID, ECD and MS	X	X	X
Proton Transfer Reaction-Quadrupole Mass Spectrometer (PTR-QMS)	VOCs	1-5 min Data available for ~5 weeks of the study	Ionic Analytik; Measurement site not collocated with core site measurements	X		
Meteorological station	Surface meteorology	1 min at THRO-N and FOUS	Climatronics All-In-One Weather Sensor	X	X	Existing
Mobile Measurements	VOC canisters; mini-aethalometer; nephelometer; acetylene; methane	Canisters: Grab Samples; MicroAeth: 1 min; Picarro: 3 Hz; Neph: 5 sec	AethLabs MicroAeth AE51; Radiance Research Nephelometer; Picarro G2203 with mobile kit A0941			

Figure Captions

Figure 1. Map of study area, including state monitoring sites and regional power plants. For oil and gas wells, shapefiles were filtered to only include active/producing wells. For reference, the blue line represents 250 km from THRO-N, the distance traveled by an airmass in 48 hours, based on the median wind speed at the site during the study. Green lines represent 100 km distance from THRO-S, LOST, and MELA.

Figure 2. Box plots of daily mean concentrations for (a) SO_2 and (b) NO_2 at THRO-N, shown in black, for all data available from these sites dating back to 2000. Also shown are daily averaged data collected during the study periods in 2013 and 2014~~-~~, with median concentrations shown as horizontal line segments.

Figure 3. Back trajectory residence times showing areas where air masses resided during the two days prior to arriving at THRO-N. a) Trajectories arriving when the hourly NO_2 concentration was at the 95th percentile (4.63 ppb) or higher for the two Bakken Studies combined. b) Trajectories arriving when the hourly NO_2 concentration was at the 5th percentile (0.57 ppb) or lower for the combined measurements from both studies. Trajectories were generated using the Hysplit Model in ensemble mode with a start height of 10 m. All trajectories have a duration of two days, so longer lengths correspond to high wind speeds. Note that transport patterns associated with the highest concentrations tend to arrive from a smaller area, indicating probable stagnation, while those associated with the lowest concentrations correspond to travel from more distant areas, indicating high wind speeds, and are more predominantly from the west.

Figures ~~3~~4. SO_2 monitoring data from the EPA AirData website. Data are annually averaged. Missing data points are for years which had less than 50% of the possible data.

Figure ~~4~~5. URG measurements of SO_2 from all of the field sites during the first study period.

Figure ~~5~~6. 48 hour back trajectories from THRO-N (black), MELA (blue), FOUS (red) and KNRI (green) for (a) February 20-February 22, 8 AM to 8 AM; and (b) March 27-March 29, 8 AM to 8 AM. Back trajectories were run four times per day. Each dot designates a 24 hour period. Field sites are shown as gold stars. Power plants are shown as triangles, with black triangles representing coal-powered plants, and green triangles representing gas-powered plants.

Figure ~~6~~7. Measurements during the first study period (Feb-April 2013) at THRO-N of hourly averaged (a) NO_x (blue), SO_2 (red), and BC (black); and (b) wind speed (red) and wind direction (black). Shaded areas indicate time periods discussed in the text and shown in Figure ~~6~~5.

Figures ~~7~~8. NO_2 monitoring data from the EPA AirData website. Data are annually averaged. Missing data points are for years which had less than 50% of the possible data.

Figure ~~8~~9. Trends in NO_2 data from Lostwood, segregated by wind direction. Slope is per year.

Figure ~~9~~10. Timeline of light alkane concentrations at THRO-N, FOUS and MELA during the second study period (November 2013 – March 2014). All data are from grab samples.

Figure 1~~1~~0. Ratio of iso- to n-pentane for canister samples collected at THRO-N, FOUS, and MELA throughout the second study period.

Figure 1~~2~~1. Concentrations of CH_4 (ppmv) and BC (ng m^{-3}). a) Measurements collected near a well with an active flare at Indian Hill; and b) measurements collected downwind of a site in Painted Woods oil field, with multiple well-heads and collection tanks, but no flare. Note the difference in scale for the two plots.

Figure 1~~3~~2. Timeline of ethane, NO_x, SO_2, and BC during the second study period in 2013-2014. NO_x, SO_2 and BC concentrations are daily average values; ethane data are the average of two grab samples per day, one collected in the morning and one collected in the afternoon.

Figure 1~~4~~3. A photochemical clock utilizing ratios of alkyl nitrates to n-alkanes. Modeled ratios are shown as the solid line, and measured data are given as points, colored and sized by daily averaged (a) NO_x and (b) BC concentrations.

Figure 1~~5~~4. Timeline of ratios of EC concentration at (a) LOST, and (b) MELA, relative to THRO-S. Also shown are data representing wells within 100 km of the given site, weighted by distance, and wells within 100 km of THRO-S. EC data are shown as monthly averages, while data for well counts are annual averages to better account for the uncertainty in the dates when wells began operation.