



## RESEARCH LETTER

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## Key Points:

- The Bakken shale in North Dakota accounted for 1–3% total global ethane emissions in 2014
- These findings highlight the importance of shale production in global atmospheric ethane shift
- These emissions impact air quality and influence interpretations of recent global methane changes

## Supporting Information:

- Supporting Information S1
- Movie S1

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## Fugitive emissions from the Bakken shale illustrate role of shale production in global ethane shift

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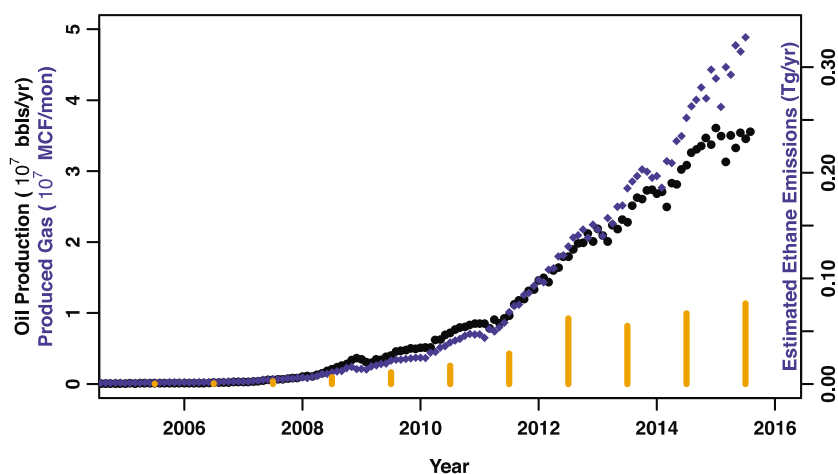
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**Abstract** Ethane is the second most abundant atmospheric hydrocarbon, exerts a strong influence on tropospheric ozone, and reduces the atmosphere's oxidative capacity. Global observations showed declining ethane abundances from 1984 to 2010, while a regional measurement indicated increasing levels since 2009, with the reason for this subject to speculation. The Bakken shale is an oil and gas-producing formation centered in North Dakota that experienced a rapid increase in production beginning in 2010. We use airborne data collected over the North Dakota portion of the Bakken shale in 2014 to calculate ethane emissions of  $0.23 \pm 0.07$  ( $2\sigma$ ) Tg/yr, equivalent to 1–3% of total global sources. Emissions of this magnitude impact air quality via concurrent increases in tropospheric ozone. This recently developed large ethane source from one location illustrates the key role of shale oil and gas production in rising global ethane levels.

### 1. Introduction

Fossil fuels are the primary source of ethane ( $C_2H_6$ ) to the atmosphere due to emissions during extraction, processing, and distribution [Blake and Rowland, 1986; Rudolph, 1995; Xiao et al., 2008; Simpson et al., 2012]. Additional sources include biofuels, biomass burning, and smaller contributions from biogenic and geologic sources [Etiope and Ciccioli, 2009]. The atmospheric abundance of ethane results from a balance between emissions and removal via reaction with the hydroxyl radical (OH). This reaction consumes OH, reducing atmospheric oxidative capacity and, depending on the chemical environment, can subsequently worsen air quality by enhancing tropospheric ozone ( $O_3$ ) formation through multiple pathways [Aikin et al., 1982]. Ethane's consumption of OH also increases the atmospheric lifetime of methane ( $CH_4$ ). Ethane thus acts as both a direct and indirect (via  $O_3$  and  $CH_4$ ) greenhouse gas, with modest global impact [Highwood et al., 1999; Collins et al., 2002]. The lifetime of ethane in the atmosphere is ~2 months, though it strongly varies seasonally at middle and high latitudes [Goldstein et al., 1995]. Concentrations in the remote atmosphere typically range from less than 1 ppb to 2 ppb [Simpson et al., 2012], with much higher levels encountered in the vicinity of sources [Smith et al., 2015]. Total global emissions are estimated at 11.3 Tg  $C_2H_6$ /yr in 2010 [Simpson et al., 2012]. While global observations suggest that emissions declined from 1984 to 2010 (14.3 to 11.3 Tg/yr), one remote mountaintop location in Europe reported an increase in regional atmospheric levels suggesting increased emissions again from 2009 [Franco et al., 2015]. While the decline in ethane emissions was attributed to reduced fossil fuel sources [Simpson et al., 2012], the more recent increase since 2009 was hypothesized to possibly be caused by the increase in shale gas production in the U.S., though without observations made in the U.S. [Franco et al., 2015].

The idea that the U.S. shale gas contribution is responsible for the recent global increase in ethane is plausible given the rapid increase in U.S. oil and gas production in the past decade. Much attention from the public and atmospheric community has focused on the greenhouse gas impacts of expanding shale gas production [Brandt et al., 2014; Peischl et al., 2015; Karion et al., 2015; Caulton et al., 2014], whereas the rapid increase in shale oil production has received less attention. The Bakken Formation of western North Dakota, primarily an oil-producing region where natural gas is a by-product, has seen substantial increases over the last decade with production levels in 2014 exceeding 2005 by a factor in excess of 3500 for oil and 180 for gas (Figure 1,



**Figure 1.** Oil (black circles) and gas (blue diamonds) production and estimated emissions (blue) and estimated emissions growth rate (orange) for the Bakken Shale in North Dakota. Production data from the North Dakota Department of Mineral Resources. On the right axis we show estimated ethane emission assuming the emissions rate observed in 2014 scales linearly with natural gas production. Emissions growth rate is shown in orange. Notice the very large increase in production (and likely emissions) since 2010.

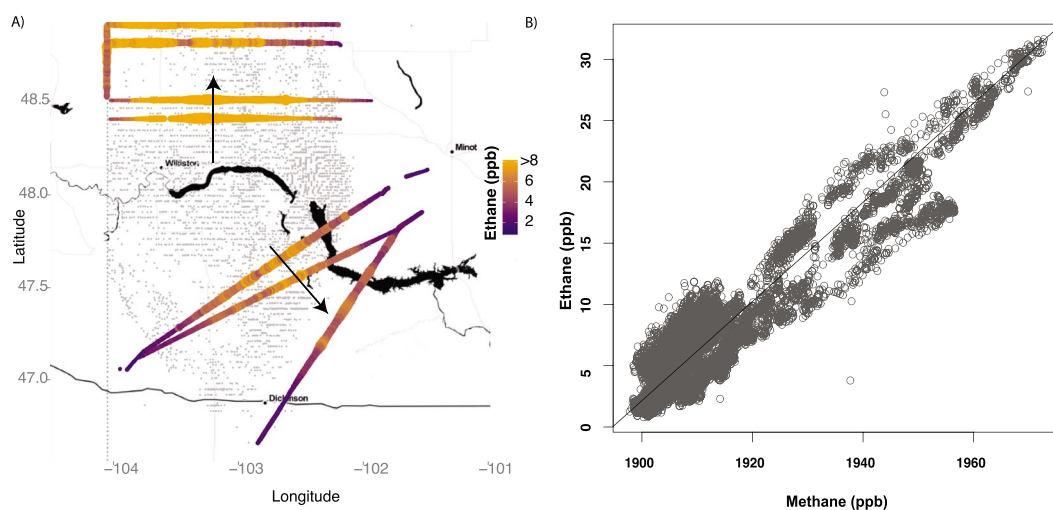
North Dakota Department of Mineral Resources). This tremendous increase in productivity has the potential to significantly impact the atmosphere if hydrocarbons are leaked or vented to the atmosphere prior to combustion. Here we present and analyze observations collected over the North Dakota portion of the Bakken shale during a National Oceanic and Atmospheric Administration (NOAA) airborne study conducted in Spring of 2014.

## 2. Materials and Methods

### 2.1. Aircraft Flights and Instrumentation

Flights were conducted in May 2014 with the National Oceanic and Atmospheric Administration (NOAA) DHC-6 Twin Otter aircraft. This study focused on assessing the atmospheric impact of oil and gas production in the Bakken with continuous measurements of methane, ethane, carbon dioxide, water vapor, carbon monoxide, ozone, black carbon, wind, pressure, and temperature as well as whole-air flask samples for analysis of dozens of other compounds. Flights consisted of horizontal transects in the daytime boundary layer to characterize emissions from the entire Bakken field, along with vertical profiles to quantify the depth of the mixed layer. The Twin Otter flew on 12 separate days. Flights typically were 3 to 3.5 h; on half of the flight days two research flights were flown (one 3–3.5 h flight, a stop for refueling, and then a second 3–3.5 h flight). Four different days (of the 12 research flight days) with steady winds appropriate for mass balance calculations are the focus of the analysis where we analyze downwind flights characterizing emissions from the entire field.

Airborne ethane measurements were made with an Aerodyne mini direct absorption spectrometer. The instrument has been described in detail previously [Smith *et al.*, 2015; Yacovitch *et al.*, 2014]. The instrument was deployed as in Smith *et al.* [2015] with the addition of hourly sampling of a standard gas to verify instrument stability. The instrument scans multiple ethane absorption lines centered at 3.3  $\mu\text{m}$ . Air is not dried prior to sampling; wet air mole fractions are observed and converted to dry mole fraction (values reported here) using the coincident water vapor observation made by a Picarro G2401-m. In-flight precision in typical conditions was  $< 0.1$  ppb. Assessment of in-flight standards indicates accuracy averages 0.5 ppb. All data reported here are dry air molar fraction, adjusted to be on the NOAA-2012 ethane scale by precampaign and postcampaign calibration (standard gas cylinders were prepared gravimetrically). Airborne methane measurements were made with a Picarro G2401-m. Sampling frequency is  $\sim 0.5$  Hz. In-flight calibrations ensure reported dry air mole fraction is on the World Meteorological Organization X2004A scale [Dlugokencky *et al.*, 2005]. Total uncertainty is estimated at  $\pm 1.0$  ppb. Wind speed and direction were measured at 1 Hz with estimated uncertainty at  $\pm 1$  m/s with a differential GPS approach as described by Conley *et al.* [2014]. Ozone was measured with a 2B Technologies analyzer. A Rosemount deiced total temperature



**Figure 2.** Ethane and methane observations over the Bakken Shale. (a) Mass balance flight tracks colored by  $C_2H_6$  for 13, 14, 21, and 22 May 2014. Approximate winds illustrated, from the NW for 13 and 14 May (flight paths below  $48^\circ$ ), and S to SW for 21 and 22 May (flight paths above  $48^\circ$  have been shifted for visual clarity). The locations of gas-producing wells are shown in gray dots. (b)  $C_2H_6:CH_4$  from legs illustrated in Figure 2a, exhibiting a slope of 40.5% (95% CI 40.2 to 40.7) as calculated accounting for variance in both  $CH_4$  and  $C_2H_6$  using a ranged major axis regression [Legendre and Legendre, 1998]. This closely matches the 42%  $C_2H_6:CH_4$  ratio present in raw gas.

sensor, model 102CP2AF, was used to measure ambient temperature. This was calibrated before and after the campaign and performed with estimated precision of  $\pm 0.2^\circ C$  and accuracy of  $\pm 1.0^\circ C$ .

## 2.2. Mass Balance Methodology

The mass balance approach provides an observation-based method for quantifying atmospheric fluxes of trace gases from a defined area. This approach assumes steady horizontal wind fields and a well-developed planetary boundary layer (PBL). We apply this method as has been extensively documented and demonstrated in the past for similar studies of oil- and gas-producing basins for methane, ethane, and black carbon [Smith *et al.*, 2015; Peischl *et al.*, 2015; Karion *et al.*, 2013, 2015; Petron *et al.*, 2014; Schwarz *et al.*, 2015]. The ethane flux is calculated as follows:

$$\text{flux}_{\text{ethane}} = v \int_{-b}^b X_{\text{ethane}} \int_{z_{\text{ground}}}^{z_1} n_{\text{air}} dz \cos(\theta) dx$$

Here  $X_{\text{ethane}}$  is the molar enrichment of ethane above background concentrations,  $v \cos(\theta)$  is the component of the horizontal wind perpendicular to the flight path,  $-b$  to  $b$  is the width of the downwind plume,  $n_{\text{air}}$  is the molar mass of dry air,  $z_{\text{ground}}$  is the ground level of the flight leg, and  $z_1$  represents the top of the mixing box. We calculate  $z_1$  as described by Peischl *et al.* [2015], where we account for ethane enhancements just above the PBL top by increasing the integrated mixing depth.

$$z_1 = (3z_{\text{pbl}} + z_e)/4$$

where  $z_{\text{pbl}}$  is defined as the top of the well-mixed layer and  $z_e$  is the top of the entrainment zone. This adjustment has a minimal impact in the study considered here. On average  $z_1$  is  $\sim 6\%$  larger than  $z_{\text{pbl}}$ .

## 3. Observations and Ethane Flux Estimate

The atmospheric  $C_2H_6:CH_4$  relationship observed over and downwind of the Bakken shale was consistent and elevated throughout the campaign. On downwind flight legs characterizing the total field emissions (Figure 2a), we observed a 40.5% molar  $C_2H_6:CH_4$  enhancement ratio (95% confidence interval (CI) 40.2 to 40.7; Figure 2b). This is a notably higher value than observed over Los Angeles [Wennberg *et al.*, 2012; Peischl *et al.*, 2013] or over the Barnett shale in Texas [Smith *et al.*, 2015], where molar enhancement ratios tended to remain below 15%. This very high  $C_2H_6:CH_4$  enhancement ratio in the Bakken is consistent with an oil-bearing

**Table 1.** Average Molar Composition of Natural Gas in the Bakken Shale (Means Normalized to 100%) as Reported in Brandt et al. [2015]<sup>a</sup>

C <sub>1</sub>	C <sub>2</sub>	C <sub>3</sub>	iC <sub>4</sub>	nC <sub>4</sub>	iC <sub>5</sub>	nC <sub>5</sub>	C <sub>6</sub>	O <sub>2</sub> /Ar	CO <sub>2</sub>	N <sub>2</sub>	H <sub>2</sub> S
47.0	19.8	14.0	1.6	4.7	0.8	1.3	1.7	0.1	1.2	7.5	0.2

<sup>a</sup>This corresponds to an ethane:methane (C<sub>2</sub>:C<sub>1</sub>) molar ratio of 42%.

reservoir rich in higher hydrocarbons and exhibiting a C<sub>2</sub>H<sub>6</sub>:CH<sub>4</sub> ratio of 42% based on 710 reported below-ground gas composition measurements (Table 1) [Brandt et al., 2015]. The close correspondence of atmospheric enhancement ratios to the reservoir gas composition indicates that emissions to the atmosphere in the Bakken shale are dominated by loss of raw gas rather than processed gas.

Flux estimates were made using the mass balance technique for seven downwind flight legs on 4 days. These flights encompass wind from both the NW and S (Figure 2a). The use of multiple downwind legs with differing wind directions supports the calculation of a robust, representative flux for the campaign period while simultaneously illustrating that out-of-field sources are negligible. Key observed values for calculating mass balance fluxes are summarized in supporting information Table S1. Average ethane emissions of  $27 \times 10^3$  kg/h were extrapolated to an annual emission of  $0.23 \pm 0.07$  ( $2\sigma$ ) Tg C<sub>2</sub>H<sub>6</sub>/yr. This extrapolation assumes constant emissions through the course of the year. We do not have observations at other times of year for the Bakken, but there is no reason to assume strong variance based on reported production [U.S. Energy Information Administration, 2016], and observations in other oil and gas basins have shown little seasonality [Smith et al., 2015; Karion et al., 2015; Kort et al., 2014].

Uncertainty for each individual flight flux estimate was calculated and propagated following Smith et al. [2015], Peischl et al. [2015], and Karion et al. [2015], with values and results summarized in supporting information Table S1. Uncertainty for the mean flux is calculated from the variance in the individual downwind legs and reported in the manuscript as  $2\sigma$ . We can also consider that the average single flight standard deviation from propagating uncertainty is  $\sim 40\%$  (supporting information Table S1). If we calculate the standard error of the mean using 40% as the standard deviation, we find that twice the standard error calculated this way is 0.07—exhibiting consistency between the individual derived flight uncertainty and the variance observed over the course of the campaign.

#### 4. Bakken Shale Contribution to Global Ethane Changes

Emissions of 0.23 Tg C<sub>2</sub>H<sub>6</sub>/yr represents 1–3% of total global ethane emissions from this single location. Annual ethane emissions are reported to have declined from 1986 to 2010 by 3.0 Tg in total [Simpson et al., 2012]. Assuming a linear change in annual emissions, this corresponds to a decrease in emissions rate of 0.12 Tg/yr/yr. To estimate the annual change in emissions from the Bakken, we assume that emissions of ethane track production of natural gas in the basin (equivalent to assuming a constant leak rate with time). Figure 1 illustrates this emissions estimate from the Bakken as well as the rate of ethane emissions increase. This estimate indicates that the growth rate in emissions from the Bakken alone reached a sustained level in 2012 (0.06 Tg/yr/yr) sufficient to cancel half the average long-term decline rate in global ethane emissions. This is consistent with the hypothesis that the large increase in U.S. oil and gas production has led to a reversal in the declining atmospheric ethane burden and highlights the disproportionate role played by the Bakken region, which represented only 2% of shale gas production in the U.S. in May 2014 [U.S. Energy Information Administration, 2016] and yet emitted 1–3% of total global ethane emissions.

The very heavy composition of raw gas in the Bakken shale (42% molar C<sub>2</sub>:C<sub>1</sub>) helps explain the relatively high emissions. Other shale plays in the U.S. have notably lower ratios (Table 2). Considering other basins' relative production and raw gas composition, it is reasonable to suspect that emissions of ethane from the Eagle Ford combines with the Bakken to represent a large fraction of the recent global shift in ethane, whereas very dry formations such as the Haynesville and Fayetteville likely play a modest role in ethane emissions in spite of their large gas production. The Marcellus, a very productive formation, has been observed to have a low C<sub>2</sub>:C<sub>1</sub> ratio [Peischl et al., 2015], though some composition data suggest higher ratios [Conder and Lawlor, 2014; Ghandi et al., 2015]. Observations in the Washington D. C. area in recent years have suggested increasing ethane emissions from the Marcellus [Vinciguerra et al., 2015].

**Table 2.** Ethane:Methane ( $C_2:C_1$ ) Molar Ratios of Major Shale Plays in the U.S. and Percentage of U.S. Shale Gas Production in May 2014<sup>a</sup>

Basin	Bakken	Eagle Ford	Marcellus	Barnett	Haynesville	Fayetteville	Utica
Molar % $C_2:C_1$	42 <sup>b</sup>	25 <sup>c,f</sup>	2 <sup>e</sup> 16 <sup>c,f</sup>	3 <sup>d</sup> (dry) 15 <sup>d</sup> (wet)	0.1 <sup>d</sup>	1 <sup>d</sup>	16 <sup>c,f</sup>
% U.S. shale gas production	2	12	35	12	12	8	3

<sup>a</sup>Production from the *U.S. Energy Information Administration* [2016]; gas composition from well measurements reported in the following.

<sup>b</sup>*Brandt et al.* [2015].

<sup>c</sup>*Conder and Lawlor* [2014].

<sup>d</sup>*Speight* [2013].

<sup>e</sup>*Peischl et al.* [2015].

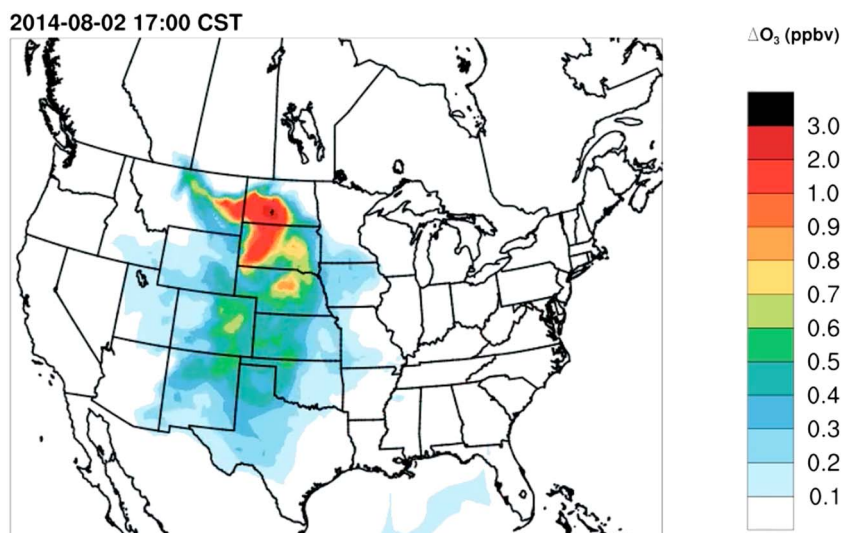
<sup>f</sup>*Ghandi et al.* [2015].

## 5. Implications for Tropospheric Ozone

Emissions of ethane, methane, and other VOCs from the Bakken have the potential to impact ozone formation on a variety of spatial scales. Local ozone enhancements observed during the Spring 2014 airborne study were relatively small due to low temperatures, large solar zenith angles, and generally high wind speeds. Initial Goddard Earth Observing System-Chem modeling (for details see supporting information Text S1) suggests that in summer, when surface ozone production peaks, up to 4 ppb of additional ozone are produced in plumes downwind of the Bakken region due to emissions of alkanes with composition as in Table 1 (Figure 3 and supporting information Movie S1). An increase of this magnitude could contribute to noncompliance with air quality regulations in affected areas downwind. More accurate modeling of the net impact of Bakken emissions on ozone formation would require additional measurement-based constraints on other reactive volatile organic compounds, including oxygenated compounds [*Edwards et al.*, 2015] and on nitrogen oxides [*Ahmadov et al.*, 2015] emitted from oil and gas activities in the region.

## 6. Conclusions

Much attention has focused on fugitive methane emissions from shale gas. This work demonstrates that we must also consider the impact of fugitive ethane emissions, particularly in basins where heavy gas composition leads to higher fugitive ethane efflux at similar volumetric leakage rates. The role of fugitive emissions with heavy gas composition also impacts assessments of global methane emissions from global ethane levels. Analyses that assume a temporally constant oil and gas production ethane:methane emission ratio



**Figure 3.** Simulation of change in surface ozone resulting from fugitive Bakken alkane ( $C_2+$ ) emissions, example for 2 August 2014 at 17:00 CST.

lower than present in the Bakken, or other productive basins, will erroneously conclude a large fossil methane emissions increase since 2010. Finally, the large, recently developed ethane source reported here has potential impacts on simulations of atmospheric composition in the last decade, as such a perturbation to ethane emissions are not presently represented in inventories and may impact representations of tropospheric ozone.

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