Quantifying atmospheric methane emissions from oil and natural gas production in the Bakken shale region of North Dakota

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Key Points

1. CH_4 emissions from the Bakken region of North Dakota quantified

This is the author manuscript accepted for publication and has undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the Version of Record. Please cite this article as doi: 10.1002/2015JD024631

2. First emission estimate using in situ CH_4 measurements

3. CH_4 sources dominated by oil- and gas-related activities

Abstract

We present in situ airborne measurements of methane (CH_4) and ethane (C_2H_6) taken aboard a NOAA DHC-6 Twin Otter research aircraft in May 2014 over the Williston Basin in northwestern North Dakota, a region of rapidly growing oil and natural gas production. The Williston Basin is best known for the Bakken shale formation, from which a significant increase in oil and gas extraction has occurred since 2009. We derive a CH_4 emission rate from this region using airborne data by calculating the CH_4 enhancement flux through the planetary boundary layer downwind of the region. We calculate CH_4 emissions of (36 ± 13), (27 ± 13), (27 ± 12), (27 ± 12), and (25 ± 10) × 10^3 kg/hr from five transects on three days in May 2014 downwind of the Bakken shale region of North Dakota. The average emission, (28 ± 5) × 10^3 kg/hr, extrapolates to 0.25 ± 0.05 Tg/yr, which is significantly lower than a previous estimate of CH_4 emissions from northwestern North Dakota and southeastern Saskatchewan using satellite remote sensing data. We attribute the majority of CH_4 emissions in the region to oil and gas operations in the Bakken based on the similarity between atmospheric C_2H_6 to CH_4 enhancement ratios and the composition of raw natural gas withdrawn from the region.

1. Introduction

From 1981 to 2003, North Dakota accounted for less than 1.5% of United States (U.S.) crude oil production (U.S. Energy Information Administration (EIA), www.eia.gov). However, the onset of directional drilling and hydraulic fracturing has led to a large increase in oil and natural gas extraction from shale formations in the U.S. in recent years. One such shale formation, the Bakken, is located in the Williston Basin of northwestern North Dakota. As a result, North Dakota accounted for 12.5% of the U.S. crude oil production in May 2014 (EIA), due mainly to oil extraction from the Bakken shale formation (Figure 1), surpassing the oil production from all U.S. regions except Texas and federal Gulf of Mexico (EIA).

In addition to crude oil, natural gas is also extracted from the Bakken shale formation. The region was responsible for 1.4% of U.S. natural gas production in May 2014 (EIA). Natural gas is a mixture primarily composed of methane (CH_4), with ethane (C_2H_6) typically the second largest component. Emissions of natural gas to the atmosphere may occur during various stages of oil and natural gas extraction and processing. Consequently, the U.S. Environmental Protection Agency (EPA) greenhouse gas (GHG) inventory estimates that CH_4 emissions to the atmosphere from the oil and gas industry account for 29% of 2013 U.S. anthropogenic CH_4 emissions (EPA 430-R-15-004, http://www3.epa.gov/climatechange/Downloads/ghgemissions/US-GHG-Inventory-2015-Main-Text.pdf). Two recent studies have shown measurement-based estimates of CH_4 emissions from the oil and gas industry are in agreement with the EPA GHG inventory for large gas-producing

regions [*Peischl et al.*, 2015; *Karion et al.*, 2015]. However, other studies have shown that GHG inventories underestimate CH_4 emissions from other oil and natural gas producing regions [*Pétron et al.*, 2012; *Peischl et al.*, 2013; *Karion et al.*, 2013; *Pétron et al.*, 2014; *Brandt et al.*, 2014]. Therefore, studies that estimate CH_4 emissions from the oil and gas industry are necessary to constrain regional and national GHG emissions inventories, and ultimately inform decisions based on the climate impacts of U.S. fuel choices.

Schneising et al. [2014] reported an increase in CH_4 emissions from the Bakken region of northwestern North Dakota and southeastern Saskatchewan of 0.99 ± 0.65 Tg CH_4/yr between the years 2006-2008 and 2009-2011 based on measurements from the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument onboard the Envisat satellite. *Schneising et al.* [2014] further estimated an "energy content leak rate" for the Bakken, *i.e.*, the energy content of the natural gas emitted to the atmosphere divided by the sum of the energy content of the oil and natural gas extracted from the ground, of 10.1% ± 7.3% by comparing the increase in CH_4 emissions with the increase in oil and natural gas production from this region over the same time period. We interpret this as a loss of 44 ± 32% of the natural gas withdrawn, as discussed in section 5.5. Several studies using in situ airborne measurements and the mass balance method have recently quantified CH_4 emissions to the atmosphere from oil and natural gas producing regions such as the Uinta Basin of Utah [*Karion et al.*, 2013], the Denver-Julesburg Basin of northeastern Colorado [*Pétron et al.*, 2014], the Haynesville region of northwestern Louisiana and eastern Texas, the Arkoma Basin of Arkansas, the Marcellus region of northeastern Pennsylvania [*Peischl et al.*, 2015], and the Barnett region of central Texas

[*Karion et al.*, 2015]. These aircraft studies report a variety of atmospheric CH_4 emission rates, ranging from $(14 – 80) \times 10^3$ kg CH_4/hr, with rates of natural gas losses to the atmosphere ranging from 0.18%–11.7% of the natural gas extracted from the ground from these regions. The variability in emissions and loss rates from these regions likely stems from a variety of factors, including whether oil is extracted in addition to natural gas, the amount of above-ground fossil fuel processing that occurs, the well count, the fossil fuel production volume, the age of the infrastructure, the type of activity occurring on days when the aircraft studies occur, etc. For comparison, the average hourly emission increase found by *Schneising et al.* [2014] was $(110 \pm 70) \times 10^3$ kg CH_4/hr over the years 2006–2008 to 2009–2011. This CH_4 emission rate is the largest emission reported from all oil and gas producing regions studied to date, which warrants further examination. Here, we derive atmospheric CH_4 emission rates from the Bakken region of North Dakota using in situ airborne data and the aircraft mass balance technique and compare to the estimate by *Schneising et al.* [2014].

We estimate the total CH_4 emitted to the atmosphere from the Bakken region of North Dakota using measurements taken aboard a chemically-instrumented National Oceanic and Atmospheric Administration (NOAA) DHC-6 Twin Otter aircraft in May 2014 during the Twin Otter Projects Defining Oil/gas Well emissioNs (TOPDOWN) field project, based out of Minot, ND (Figure 2). The NOAA Twin Otter flew on 12 days in the Bakken region, with transects performed upwind, over, and downwind of this region. The Twin Otter typically flew at ~65 m/s, and full field transects normally took 45 minutes. A flight was typically 3 to 3.5 hours in duration. On six of the flight days, the Twin Otter refueled and took off again for a second

flight. Figure 2 shows all the flight tracks of the NOAA Twin Otter around the Bakken region in May 2014. Using the mass balance technique, we determine the CH_4 emissions from this region on three days with steady winds. We then attribute the CH_4 emissions in the region to oil and natural gas extraction operations based on the similarity of atmospheric C_2H_6 to CH_4 enhancement ratios during the mass balance transects with the abundances of C_2H_6 and CH_4 in natural gas extracted from the Bakken region. Finally, we estimate the natural gas loss rate to the atmosphere from oil and gas operations in the Bakken region by converting the CH_4 emission to a natural gas emission and dividing by the amount of natural gas extracted in the region in May 2014.

2. Instrumentation

CH_4, carbon dioxide (CO_2), carbon monoxide (CO), and water vapor (H_2O) were measured by wavelength-scanned cavity ringdown spectrometry using a Picarro 2401-m [*Karion et al.*, 2015]. CH_4 was measured once every 2 seconds with an estimated accuracy of ± 1.4 ppb (all uncertainties herein are 1-σ); the precision of this measurement was ±0.2 ppb. All CH_4 measurements herein are reported as dry air mole fractions expressed as nanomole/mole, or ppb. C_2H_6 was measured by tunable infrared laser direct absorption spectroscopy using an Aerodyne mini ethane analyzer [*Smith et al.*, 2015]. C_2H_6 was measured every second with an estimated accuracy of ± 7%; 1-second precision was ±(0.02 to 0.21) ppb, depending on flight conditions. Additionally, black carbon (BC) was measured using a single particle soot photometer [*Schwarz et al.*, 2015], ozone was measured using a 2B Technologies analyzer, and flasks of air samples

were later analyzed for hydrocarbons and other trace gases. Two differential GPS antennae were mounted above the fuselage of the Twin Otter and provided aircraft heading, altitude, latitude, longitude, ground speed, and course over ground. Ambient temperature was measured with a Rosemount de-iced total temperature sensor, model number 102CP2AF, calibrated before and after the field project, with a precision of ±0.2 °C and an estimated accuracy of ±1.0 °C. Meteorological and navigational measurements were made once per second by various sensors aboard the NOAA Twin Otter. Potential temperature, θ, is calculated using the following equation:

$$\theta = T \times \left(\frac{1000}{P}\right)^{0.286} \quad (1)$$

where T is ambient temperature in units of Kelvin and P is ambient pressure is in units of millibar. The ambient wind speed and wind direction were derived from the true air speed, heading, speed over ground, and course over ground measurements aboard the Twin Otter using the technique of *Conley et al.* [2014]. We estimate uncertainties in the wind speed of ±1 m/s and wind direction of ±6° during typical level flight conditions, based on an analysis of the wind calibration flight pattern data [*Conley et al.*, 2014].

3. Other Data

The following inventories are used in the current analysis: the EPA 2013 GHG Inventory (publication EPA 430-R-15-004, downloaded September 2015, http://www3.epa.gov/climatechange/Downloads/ghgemissions/US-GHG-Inventory-2015-Main-

Text.pdf), and the EPA GHG Reporting Program (GHGRP) inventory (downloaded November 2015, http://www2.epa.gov/ghgreporting/subpart-w-reported-data). The GHGRP data include the latitude, longitude, and estimated CH_4 emission of known point sources (Figure 2). Some latitude-longitude coordinates in the 2013 EPA GHGRP inventory dataset were modified to match the actual locations determined using satellite imagery.

Natural gas sample analysis data are used from the work of *Brandt et al.* [2015] by way of the North Dakota Industrial Commission, Department of Mineral Resources, Oil and Gas Division (https://www.dmr.nd.gov/oilgas/). Samples were taken from 710 wells across the Bakken region (Figure 2) and analyzed for the molar composition of C_1–C_6 alkanes, CO_2, and other trace gases. North Dakota well locations were downloaded from the same website (accessed September 2014). North Dakota well production data are from the North Dakota State Industrial Commission May 2014 Oil and Gas Production Report (https://www.dmr.nd.gov/oilgas/mprindex.asp). Bakken production data were obtained from the EIA Web site (http://www.eia.gov/petroleum/drilling/, downloaded October 2015).

Terrain height below the Twin Otter aircraft is estimated using a Digital Elevation Map (DEM) from the U.S. Geological Survey (GTOPO30, 1996, https://lta.cr.usgs.gov/GTOPO30). The DEM has a resolution of 30 arc seconds (~900 m), and is based on World Geodetic System 1984 data. Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model back-trajectories [*Stein et al.*, 2015; https://ready.arl.noaa.gov/HYSPLIT.php] are used to evaluate the evolution of wind fields over time, and are run with the following settings: Meteorology: North

American Mesoscale Forecast System, 12 km; Vertical Motion: Model vertical velocity (which uses the vertical velocity field from the meteorological data). County-level livestock populations were downloaded from the U.S. Department of Agriculture (USDA) National Agriculture Statistics Service (NASS) (http://www.nass.usda.gov). We assume an average emission of 67.9 kg CH_4/yr per head from enteric fermentation (EPA GHG inventory) and no emission of C_2H_6. Bison populations are estimated from a North Dakota Department of Agriculture website (http://www.nd.gov/aitc/agfacts/Livestock/). We assume that bison populations scale with cattle populations, and assume an average emission of 72 kg CH_4/yr per head [*Kelliher and Clark*, 2010]. County-level manure emissions estimates were downloaded from the National Renewable Energy Laboratory (NREL) (http://maps.nrel.gov/biomass), and were based on 2002 USDA data. In cases where the NOAA Twin Otter flight track transects a county, an apportionment based on geographical area is used to determine livestock and manure emissions within the study region.

4. Mass Balance Technique

The technique used to estimate CH_4 fluxes is the mass balance approach [*White et al.*, 1976]. This technique has been used extensively to estimate CH_4 emissions from oil and gas producing regions [*Karion et al.*, 2013; *Pétron et al.*, 2014; *Peischl et al.*, 2015; *Karion et al.*, 2015]. Further, the mass balance technique was used to estimate C_2H_6 emissions from an oil and gas producing region [*Smith et al.*, 2015], and black carbon and C_2H_6 emissions from the Bakken region during the 2014 TOPDOWN project [*Schwarz et al.*, 2015; *Kort et al., in review*].

The mass balance technique is used as follows. We calculate the horizontal flux of a species *X* through a vertical plane defined by an aircraft transect:

$$flux = v\cos(\alpha)\int_{z_0}^{z_1}\int_{-y}^{y}\left(X - X_{bg}\right)dy\,dz \quad (2)$$

where v cos(α) is the magnitude of the component of the wind velocity normal to the flight track averaged over the course of a transect, z_0 is the terrain height in meters above sea level (masl), z_1 is the adjusted mixing height in masl, $(X - X_{bg})$ is the CH_4 enhancement above the background, and *y* is the crosswind distance [*White et al.*, 1976]. We define the background CH_4 mixing ratios as those found immediately outside the plume downwind of the Bakken region. This is discussed in more detail, below. We define the adjusted mixing height in meters above ground level (magl) as described by *Peischl et al.* [2015]: $z_1 = (3z_{PBL} + z_e)/4$, where z_{PBL} is the well-mixed planetary boundary layer (PBL) depth, and z_e is the entrainment height, or the altitude at which mixing from the PBL has reached estimated free tropospheric CH_4 mixing ratios. Below, we compare this method of determining the adjusted mixing height with that of *Schwarz et al.* [2015]. All other calculations are similar to those reported by *Peischl et al.* [2015].

5. Results and Discussion

5.1. CH_4 emissions to the Atmosphere from the Bakken Study Region

The NOAA Twin Otter aircraft flew three flights dedicated to determining the CH_4 mass balance from the Bakken region on 13, 14, and 22 May (Figure 3). Eight additional flight days

were dedicated to characterizing emissions from CH_4 point sources, flaring, and general field emissions. One flight day, 21 May, was dedicated to mass balance, but because the wind speeds were light (~2–3 m/s), we cannot verify that they were steady over a full traverse of the Bakken region, and we therefore discard this day from our analysis. For the three mass balance flights used here, the winds measured aboard the Twin Otter were steady throughout the Bakken region, in agreement with HYSPLIT back-trajectories.

The 13, 14, and 22 May flight tracks sampled downwind of the majority of the highest-producing wells in the Bakken region (Figure 3). The flight tracks in Figure 3 are colored by the CH_4 mixing ratio measured within the PBL along the transects used in the mass balance analysis. Figure 4 shows the time series of CH_4 and aircraft altitude for the downwind transects highlighted in Figure 3. Also plotted in Figure 4 is the background CH_4 mixing ratio used in Equation 2. We use the C_2H_6 measurement, which typically has a greater plume signal to background variability ratio than the CH_4 measurement, to help define the plume background, especially on days such as 22 May, when the CH_4 plume enhancements were only a factor of 2–4 greater than the CH_4 background variability. The CH_4 background is further selected to be consistent with the C_2H_6 data, such that the ratio of their derived fluxes is equal to the molar enhancement ratio of C_2H_6 to CH_4 observed on the downwind transects.

Examples of the mixing height determination are shown with three vertical profiles in Figure 5. The left panel of Figure 5 shows the vertical profile immediately before the transect shown in Figure 4a; the center panel of Figure 5 shows the vertical profile immediately before the second transect shown in Figure 4b; the right panel of Figure 5 shows the vertical profile

immediately after the first transect shown in Figure 4c. We calculate the adjusted mixing height, z_1, following the method of *Peischl et al.* [2015] to account for any transport of CH_4 through the top of the PBL. We increase the effective PBL depth by one quarter the distance between the well-mixed PBL, defined by the dashed lines in Figure 5, and entrainment height, z_e, defined by the dotted lines in Figure 5, and include an uncertainty equal to this same distance. This adjusted mixing height and uncertainty encompasses a range of possible sources for the CH_4 enhancement above the free tropospheric levels immediately above the PBL: z_{PBL} if the CH_4 enhancements above the PBL are solely due to pre-existing layers aloft; and halfway between z_{PBL} and z_e if all CH_4 enhancements above the PBL are due to detrainment from the PBL. The resulting adjusted mixing heights and their uncertainties from each vertical profile during a flight are fit with a line. We use the value of this fit at the time of the transect, along with the corresponding 1-σ confidence interval of the fit, to determine z_1 for Equation 2, and plot the resulting adjusted mixing heights as filled yellow circles in Figure 5. For comparison, the mixed height determined by *Schwarz et al.* [2015] using a different method are shown with orange diamonds. For each mass balance transect in this analysis, the mixed height determinations are well within the uncertainties of the two different estimation techniques.

Table 1 lists the parameters and their uncertainties used in Equation 2 to calculate the CH_4 flux for each transect. The largest uncertainties for the 13 May flight were from the uncertainty of the background CH_4 value (±25%), the variability of the wind speed (±20%), and the uncertainty of z_1 (±18%). The largest uncertainties for the 14 May flight were from the variability of the wind speed (±34%), the uncertainty of the background CH_4 value (±30%), and

the uncertainty of z_1 (±21%). Finally, the largest uncertainties for the 22 May flight were from the uncertainty of the background CH_4 value (±27%), the variability of the wind speed (±24%), and the uncertainty of z_1 (±21%). The CH_4 fluxes calculated from the five downwind transects are (36 ± 13), (27 ± 13), (27 ± 12), (27 ± 12), and (25 ± 10) × 10^3 kg/hr. The two flux determinations from the 14 and 22 May flights did not depend on transect altitude, suggesting the emissions were mixed vertically within the PBL by the time they were sampled by the Twin Otter. The $1/\sigma^2$-weighted average of the five CH_4 emissions estimates for the Bakken region is 28 (± 5) × 10^3 kg CH_4/hr (Table 2), suggesting relatively small day-to-day variability in the total emissions rate for this short study. We therefore assume this emission rate represents the average May 2014 CH_4 emission from the Bakken region.

5.2. Oil and Natural Gas Production in the Bakken Study Region

Monthly oil and natural gas production from the Bakken region is estimated for May 2014 using both EIA and North Dakota state data. The EIA reports gross gas withdrawal volumes in thousands of cubic feet (MCF) at 60°F and 14.73 pounds per square inch. We therefore adjust the volumes reported by the EIA by factor of 0.9483 to convert to standard temperature and pressure of 0°C and 1 atm, then convert cubic feet to cubic meters. The Bakken natural gas production for May 2014 was 10.2 × 10^8 m^3 according to the EIA when converted to standard cubic meters. Similarly, the sum of individual well production data from the North Dakota Oil and Gas Division is 9.7 × 10^8 m^3, within 5% of the EIA estimate. Each airborne transect analyzed here was downwind of wells that accounted for greater than 97% of total

production. We therefore estimate natural gas production upwind of any given transect in the region to be $(9.9 \pm 0.3) \times 10^8$ m^3 for the month of May 2014 (Table 2).

5.3. Other Sources of CH_4 in the Bakken Study Region

The Bakken region encompasses landfills and livestock; these sources emit CH_4 but are not known to emit C_2H_6. The total CH_4 emission from point sources in the 2013 EPA GHG inventory not related to oil and natural gas processing in the Bakken region, two natural gas-fired electrical generating stations, is 0.23×10^3 kg/yr, which averages to 0.03 kg CH_4/hr, which we treat as negligible. Here, we use bottom-up methods to estimate CH_4 emissions from livestock in the Bakken region, and then compare these emissions to the total CH_4 emission from the region to confirm that livestock emissions are also not a major source of CH_4 to the atmosphere in the Bakken region. We estimate that the Twin Otter flight track encompassed 9% of the cattle and calves in North Dakota, based on a geographical apportionment of USDA NASS county data. This results in an estimated enteric fermentation emission from 161,000 cattle and calves of 1.3×10^3 kg CH_4/hr. CH_4 emissions from manure are estimated at 0.003×10^3 kg CH_4/hr. We estimate approximately 32,000 bison emit 0.3×10^3 kg CH_4/hr, based on statewide bison populations scaled by estimated cattle and calf populations in the Bakken region. The combined point source and livestock emission, 1.5×10^3 kg CH_4/hr (Table 2), accounts for approximately 5% of the 28×10^3 kg CH_4/hr emission calculated for May.

Coal mines, another possible source of CH_4, are located southeast of the Bakken region. The state of North Dakota has four active coal mines (EIA,

http://www.eia.gov/coal/annual/pdf/table2.pdf), all of which are surface mines. These mines are represented in the 2013 EPA GHGRP inventory, and are located between 47.0° and 47.5°N latitude, and 101° and 102°W longitude (Figure 2). CH_4 emissions from these mines were further downwind of the 13 and 14 May transects, and therefore do not affect the mass balance calculations on these days, but may have contributed to the CH_4 enhancements measured on the 22 May flight. This is discussed in more detail in the next section.

5.4. Source Attribution of CH_4 Emissions

The small contribution of livestock to the total CH_4 emission in the Bakken region suggested by the USDA inventory is confirmed using airborne data. Enhancements of C_2H_6 in the boundary layer along the mass balance transects correlate with enhancements of CH_4 similar to the composition ratios of C_2H_6 to CH_4 in natural gas extracted from the region (Figure 6). The lines representing the composition ratios in Figure 6 are offset to 1900.5 ppb CH_4 and 1.5 ppb C_2H_6, which represents the approximate background mixing ratios for the mass balance transects on the 13 and 14 May flights. For the 22 May flight, we plot the CH_4 enhancement above the sloping background in Figure 4, then offset the difference to the background values as for the composition ratios. The C_2H_6 to CH_4 enhancement ratio was on the order of 0.4 – 0.5, an atypically high ratio compared to previously studied oil and gas producing regions. As a result, this ratio provides a unique source fingerprint, and distinguishes Bakken region oil and gas emissions from natural gas in the supply chain or coal mine emissions, which typically have C_2H_6 to CH_4 ratios of less than 0.05. Finally, an approximately 10 km wide CH_4 plume on 22

May sampled shortly before 17:00 CST not highly correlated with C_2H_6 represents 6% of the CH_4 flux calculation for that day. These data were sampled along the eastern edge of the 22 May transects, where emissions from coal mines to the south-southeast may have influenced the CH_4 measurements.

We attribute CH_4 emissions to the oil and gas industry in the Bakken region as follows. We subtract the estimated livestock emission and the plume not correlated with C_2H_6 on the 22 May flight from the total CH_4 emission estimated for the region. Since we cannot confirm quantitatively with our measurements the impact that livestock and coal mine emissions may have had on the CH_4 emissions, we assign a 100% uncertainty to these subtractions. Consequently, CH_4 emissions attributed to the oil and gas industry have larger error bars than the estimate for the total CH_4 emissions from the region, but both emissions estimates share the same upper limit (Table 2).

Our estimate for oil and gas emissions of $(20 – 33) \times 10^3$ kg CH_4/hr is a factor of 5–8 greater than the 2013 EPA GHGRP estimate for the entire Williston Basin of 0.0047 Tg CH_4/yr, which averages to 4.1×10^3 kg CH_4/hr. However, under this program, emissions from many small facilities may not meet the threshold required to report (http://www2.epa.gov/sites/production/files/2015-07/documents/subpartwinformationsheet.pdf), so we would expect the GHGRP value to be a lower limit to the total emissions. However, the EPA also estimates U.S. CH_4 emissions from petroleum systems of 1.009 Tg CH_4/yr (2013 EPA GHG inventory, Table 3-37). Assuming inventory CH_4 emissions scale with oil production, the

Bakken region would account for 12.5% of the total U.S. emission, which averages to 14.4×10^3 kg CH_4/hr. Therefore, our estimate of CH_4 emissions from the Bakken region oil and gas industry is a factor of 1.4–2.3 greater than the 2013 EPA GHG inventory's emissions from petroleum systems scaled by oil production.

5.5. Loss Rates to the Atmosphere from Oil and Natural Gas Production in the Bakken Study Region

CH_4 may be emitted, or lost, to the atmosphere from oil and natural gas operations through a variety of ways: intentional venting, partially-combusted flaring, fugitive losses, etc. An atmospheric loss rate from oil and natural gas operations in the Bakken region for May 2014 is estimated by taking the CH_4 mass emission rate attributed to the oil and gas industry and dividing by the average CH_4 mass extraction rate, or production rate, of natural gas for the month. We use 22.414 L/mol (the EIA standard 23.636 L/mol times 0.9483 to convert to standard L) of natural gas in the conversion from volume to mass. The mean CH_4 abundance in the 710 Bakken natural gas samples is 47 ± 13% [*Brandt et al.*, 2015] (Table 2). The resulting estimated natural gas loss rate from the Bakken region is 4.2% – 8.4% based on this analysis of airborne in situ data. This loss rate is not significantly different than that calculated for another oil-producing region, the Denver-Julesburg Basin in northeastern Colorado, where *Pétron et al.* [2014] found loss rates of 2.6% – 5.6%.

We convert this loss rate to an "energy loss rate" in order to compare to that reported by *Schneising et al.* [2014]. We estimate an energy loss rate by converting the natural gas and oil

extracted from the Bakken formation to energy extracted using the following conversions: 5.8×10^7 J/m^3 for natural gas, and 3.464×10^{10} J/m^3 for oil [*Brandt et al.*, 2015]. We assume the change in density of oil into standard units is <1%, and therefore negligible. In May 2014, natural gas accounted for 25.4% of the energy extracted in the Bakken region [EIA], which results in an energy loss rate of 1.6 ± 0.5%, a factor of approximately 6 lower than that reported by *Schneising et al.* [2014] for 2006–2008 to 2009–2011. Alternatively, we convert the energy loss rate found by *Schneising et al.* [2014] to a natural gas loss rate. Natural gas in the North Dakota portion of the Bakken accounted for 23% of the energy extracted from 2009–2011. Assuming that this energy extraction ratio holds true for production in the Saskatchewan portion of the Bakken, the 10.1 ± 7.3% energy loss rate reported by *Schneising et al.* [2014] would imply an emission to the atmosphere of approximately 44 ± 32% of the natural gas extracted from the Bakken formation. Finally, where we derive a total CH_4 emission of $(28 \pm 5) \times 10^3$ kg/hr from the Bakken region, *Schneising et al.* [2014] found an *increase* in emissions of $(110 \pm 70) \times 10^3$ kg/hr from western North Dakota and southeastern Saskatchewan. We conclude the exceptionally high atmospheric loss rate of CH_4 reported by *Schneising et al.* [2014] for 2009–2011 is inconsistent with our airborne data from May 2014.

6. Conclusions

We report the first in situ measurements of CH_4 to estimate atmospheric CH_4 emissions from the Bakken oil and natural gas production region of northwestern North Dakota. We calculate CH_4 fluxes from the Bakken region for May 2014 from 3 flights and 5 transects

downwind of the region and find CH_4 emissions to the atmosphere of $(28 \pm 5) \times 10^3$ kg CH_4/hr. Both aircraft measurements of C_2H_6 and a bottom-up assessment derived here indicate that livestock are not a dominant source of CH_4 to the atmosphere in this region, nor are landfills and other CH_4 sources not related to the oil and gas industry. Instead, atmospheric enhancement ratios of C_2H_6 to CH_4 are consistent with the composition ratio of natural gas extracted in the Bakken region. We therefore attribute CH_4 emissions of $(20 – 33) \times 10^3$ kg CH_4/hr to the oil and gas production and distribution activities in this region. This is approximately a factor of 1.4–2.3 greater than the 2013 EPA GHG inventory emission rate from petroleum systems when scaled by oil production. Stated in terms of the natural gas extracted from the ground, we estimate atmospheric loss rates from natural gas production of 4.2 – 8.4%, similar to the rate found for the Denver-Julesburg oil-producing region [*Pétron et al.*, 2014].

Acknowledgements and Data

J. Peischl, K. Aikin, M. Trainer, and T. Ryerson were supported in part by the NOAA Climate Program Office. A. Karion, C. Sweeney, S. Wolter, E. Kort, M. Smith, and A. Gvakharia were supported in part by the NOAA Atmospheric Chemistry, Carbon Cycle, and Climate Program under grant #NA14OAR0110139. Data are publicly available at http://esrl.noaa.gov/csd/groups/csd7/measurements/2014topdown/.

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Tables

Table 1

Data Used in Equation 2 from Five Transects on the 13, 14, and 22 May 2014 Flights and the Resulting CH_4 Fluxes.

Transect	Terrain Height, masl	z_1, masl	Wind Direction, °	Wind Speed, m/s	CH_4 Flux (10^{26} molec./s)	CH_4 Flux (10^3 kg/hr)
13 May	680 ± 60	2810 ± 265	312 ± 9	13.3 ± 2.7	3.8 ± 1.4	36 ± 13
14 May, #1	695 ± 85	2545 ± 395	339 ± 14	7.7 ± 2.6	2.8 ± 1.4	27 ± 13
14 May, #2	705 ± 80	2655 ± 245	339 ± 8	8.0 ± 2.3	2.8 ± 1.3	27 ± 12
22 May, #1	655 ± 45	2340 ± 355	177 ± 10	7.5 ± 1.8	2.8 ± 1.2	27 ± 12
22 May, #2	660 ± 45	2320 ± 300	180 ± 9	7.4 ± 1.7	2.7 ± 1.1	25 ± 10

Table 2

Summary of CH_4 Emissions from the Bakken Study Region.

CH_4 flux	$(28 \pm 5) \times 10^3$ kg/hr
CH_4 estimated from non-oil and gas point sources and livestock	~1.5×10^3 kg/hr
CH_4 emissions attributed to the oil and gas industry	$(20 – 33) \times 10^3$ kg/hr
Natural gas production in May 2014	$(9.9 \pm 0.3) \times 10^8$ m^3
CH_4 abundance in natural gas[a]	47 ± 13%
CH_4 extraction rate in May 2014	$(420 \pm 120) \times 10^3$ kg/hr
Natural gas loss rate to the atmosphere	6.3 ± 2.1%

[a] *Brandt et al.* [2015]

Figure Captions

Figure 1. Monthly average daily oil and natural gas production from the Bakken shale play through May 2014. Source: U.S. Energy Information Administration (downloaded October 2015).

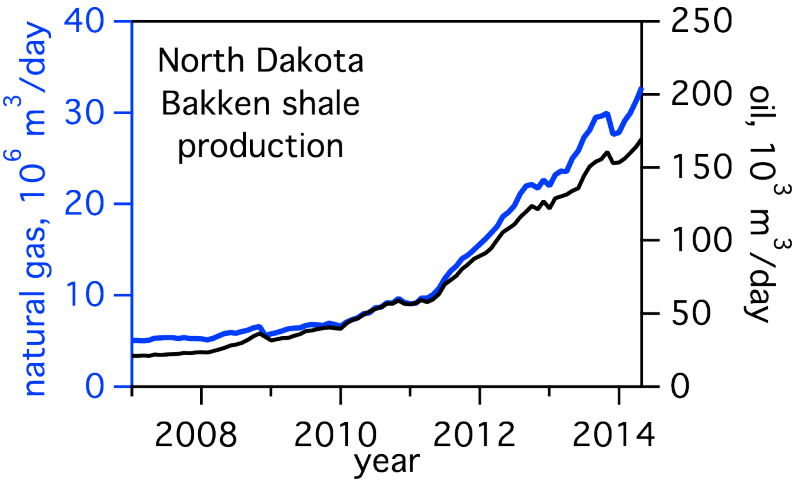
Figure 2. Map of the Bakken region. The NOAA Twin Otter flight tracks are shown with thin black lines. Active oil and gas wells in North Dakota (ND), Montana (MT), Saskatchewan (SK), and Manitoba (MB) are shown as light blue circles. Natural gas was sampled from 710 wells, colored mustard yellow, and analyzed for CH_4, C_2H_6, and other trace gas abundance [*Brandt et al.*, 2015]. Point sources from the 2013 EPA GHG Reporting Program inventory are shown as open dark blue circles sized by inventory CH_4 emissions. Urban area outlines, including Minot and Williston, ND, are colored orange. The approximate extent of the Williston Basin is shaded gray in the overview map.

Figure 3. Map showing the five transects downwind of the central Bakken shale formation from which CH_4 fluxes were calculated. North Dakota natural gas production data are summed into a ~5.6 × 5.6 km grid. Flight tracks are colored and sized by CH_4 mixing ratio for the transects shown in Figure 4. The arrow indicates the average wind speed and direction during the transects, where the length of the arrow is proportional to the wind speed. Figure 3a shows the 13 May transect at approximately 350 m agl. Figure 3b shows the 14 May northeast-to-southwest transect at approximately 350 m agl and southwest-to-northeast transect at approximately 650 m agl. Figure 3c shows the 22 May east-to-west transect at approximately 500 m agl. Figure 3d shows the 22 May west-to-east transect at approximately 1000 m agl.

Figure 4. Time series of CH_4 measurements (black) from the five transects downwind of the Bakken shale region shown in Figure 3. The blue trace shows the Twin Otter altitude agl. The red trace represents the background CH_4 used for the flux calculations; the dashed red traces represent the estimated $\pm 1\sigma$ uncertainty of the background. The shaded areas show the CH_4 enhancement above the background along the transects.

Figure 5. One vertical profile is plotted from each of the three flight days used for the mass balance. Dashed horizontal black lines indicate the well-mixed PBL depth. Dotted horizontal black lines indicate the entrainment height. The yellow circles indicate the average adjusted mixing heights used for the mass balance calculations. The orange diamonds indicate the average adjusted mixing heights ($h \times \beta$) used by *Schwarz et al.* [2015]. Solid vertical gray lines indicate the CH_4 background; dashed vertical gray lines represent the $\pm 1\sigma$ uncertainty of the background.

Figure 6. C_2H_6 and CH_4 are plotted for each of the five transects used to calculate a CH_4 flux downwind of the Bakken region. Also plotted are lines with a slope equal to the C_2H_6 to CH_4 molar composition ratio (C_2/C_1) from 710 samples of natural gas from the Bakken region [*Brandt et al.*, 2015], assuming a background of 1.5 ppb C_2H_6 and 1900.5 ppb CH_4. The slope of the yellow line represents the geometric mean C_2H_6 to CH_4 composition ratio of the samples.



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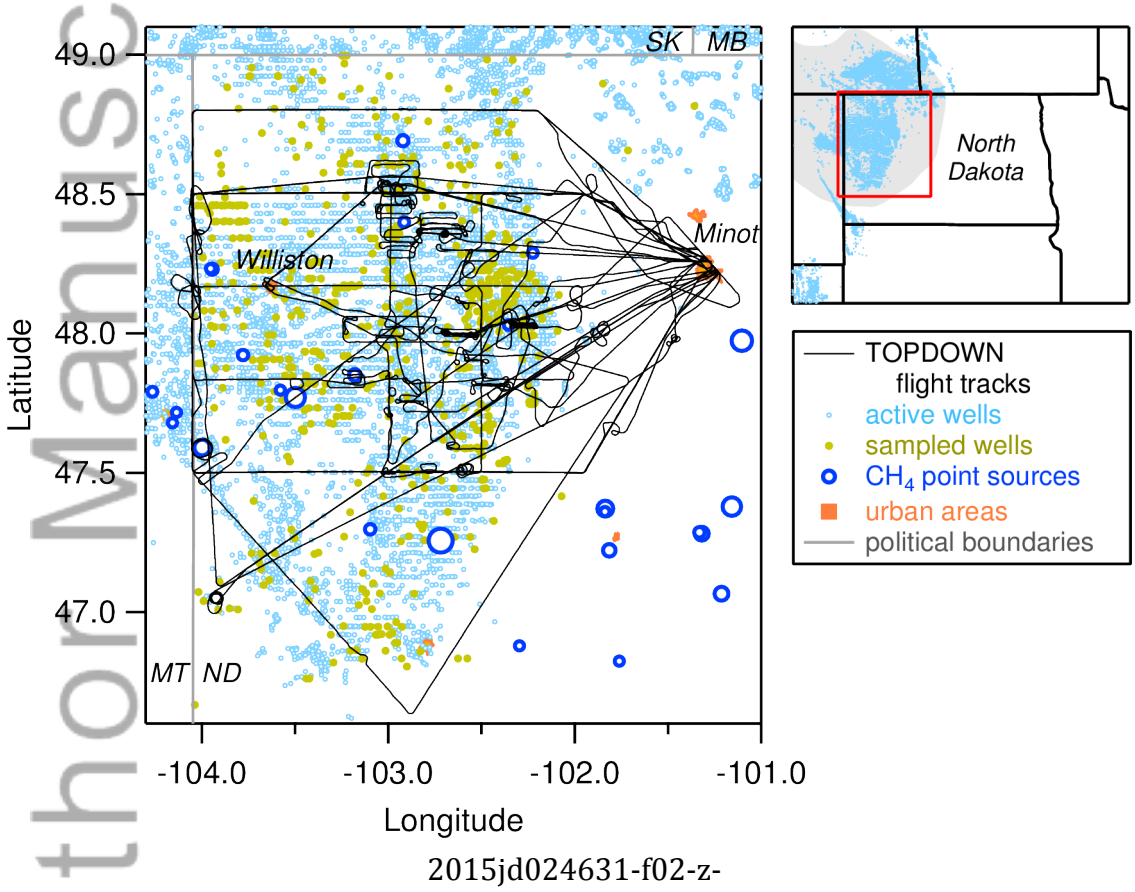

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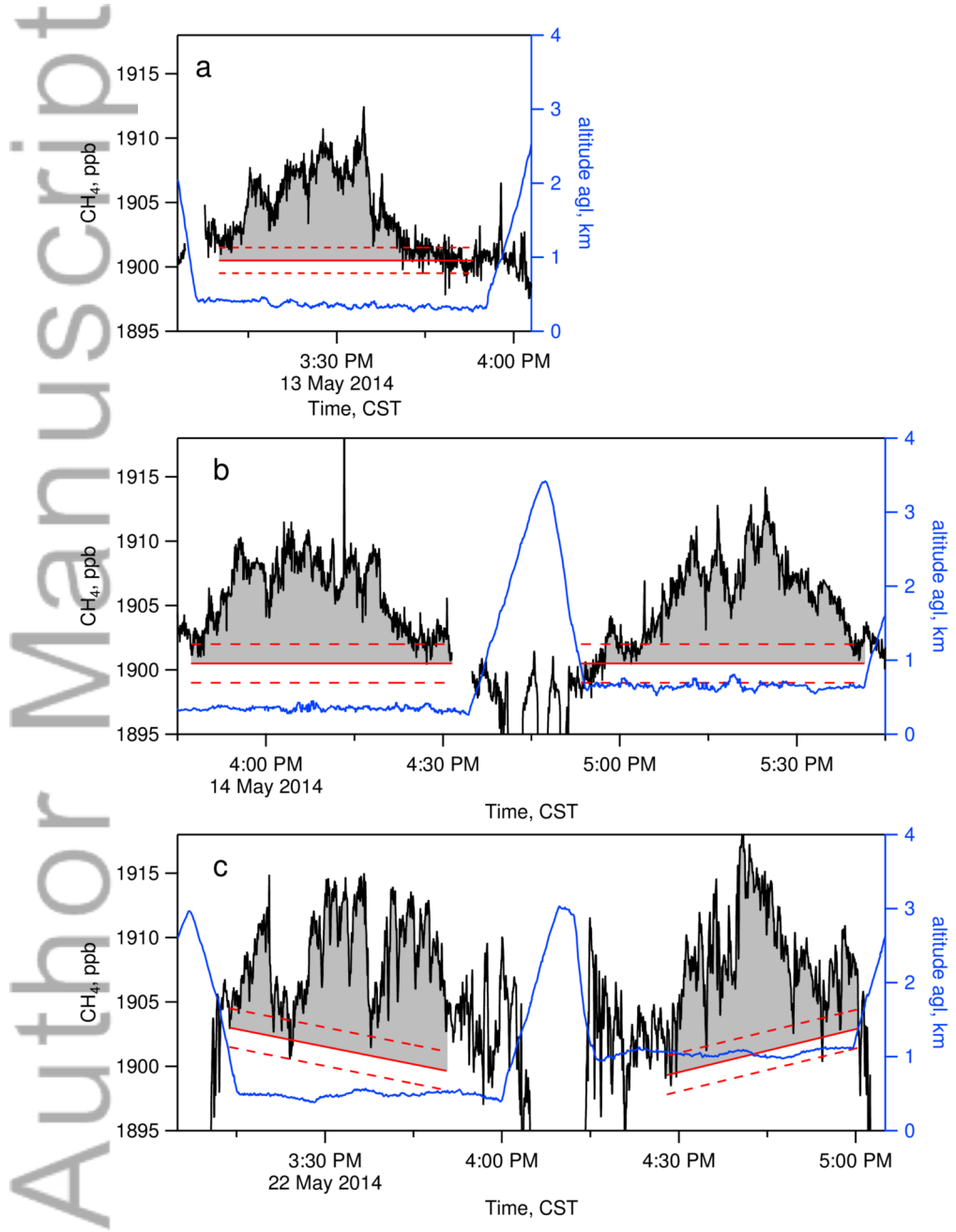

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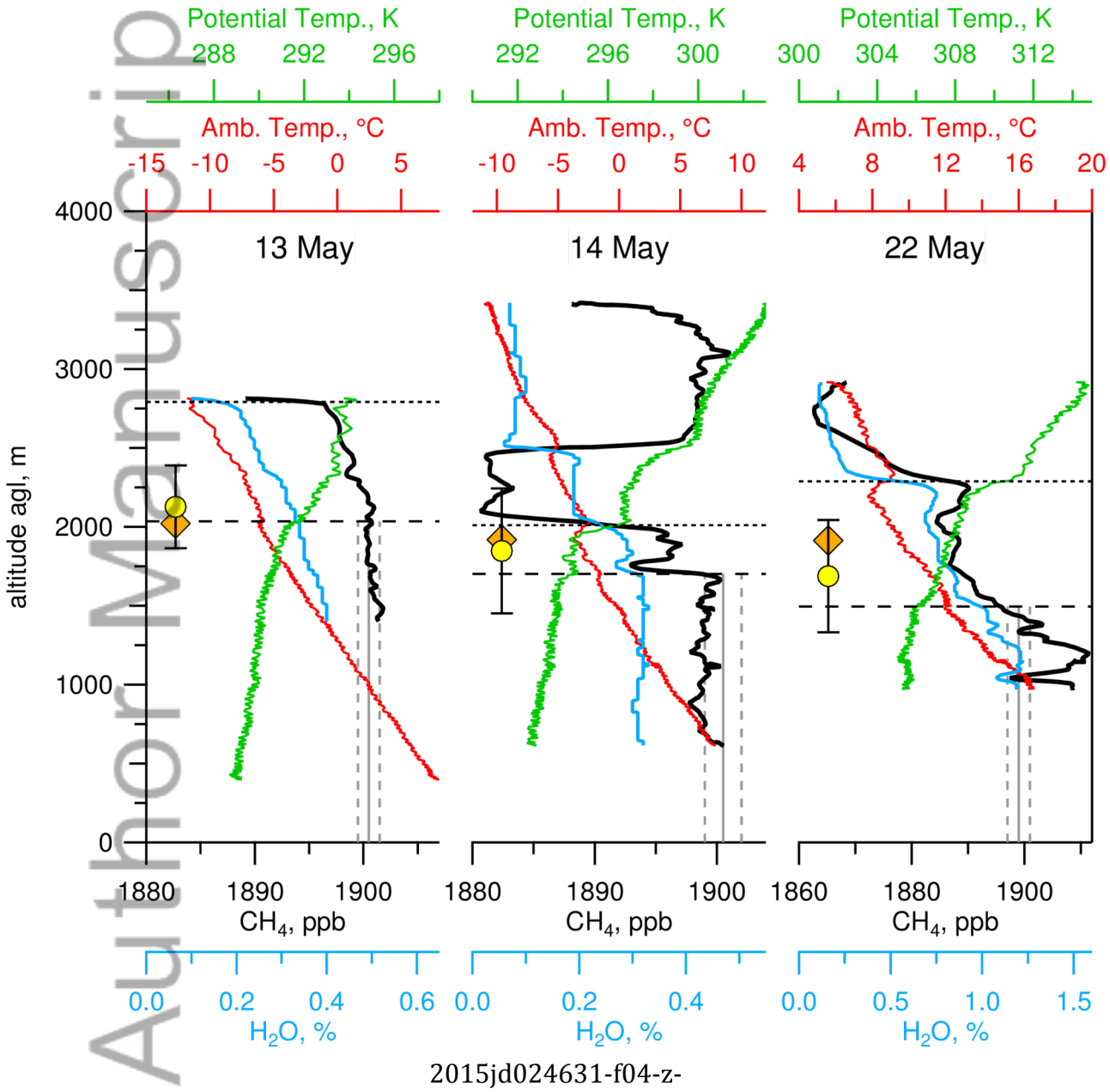

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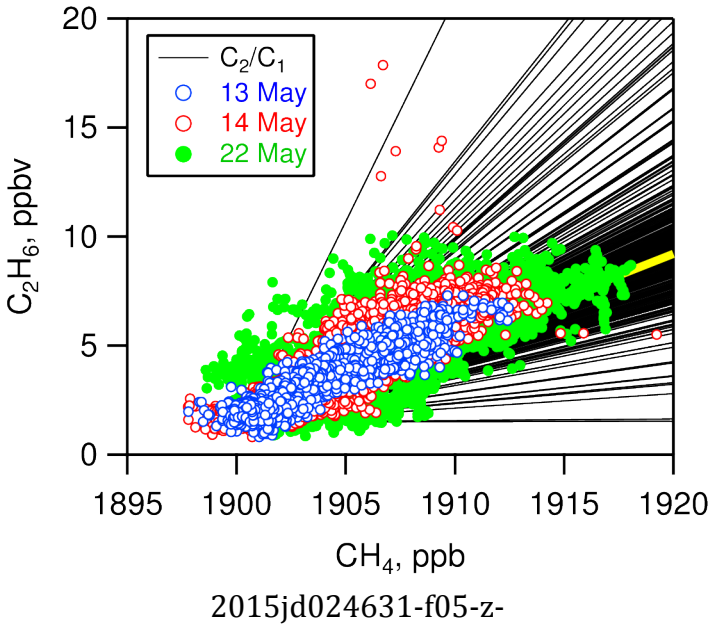

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