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## RESEARCH LETTER

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### Special Section:

Carbon and Weather: Results from the Atmospheric Carbon and Transport – America Mission

### Key Points:

- Methane and ethane observations from aircraft are used to quantify methane emissions from coal and natural gas production in Pennsylvania
- Methane emissions from coal production align with national estimates, whereas emissions from natural gas production are underestimated
- Energy produced through natural gas production in Pennsylvania has half the carbon footprint compared to energy from coal mining

### Supporting Information:

- Supporting Information S1

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## Estimating Methane Emissions From Underground Coal and Natural Gas Production in Southwestern Pennsylvania

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**Abstract** Production of coal and natural gas is responsible for one third of anthropogenic methane (CH<sub>4</sub>) emissions in the United States. Here we examine CH<sub>4</sub> emissions from coal and natural gas production in southwestern Pennsylvania. Using a top-down methodology combining measurements of CH<sub>4</sub> and ethane, we conclude that while Environmental Protection Agency inventories appear to report emissions from coal accurately, emissions from unconventional natural gas are underreported in the region by a factor of 5 (±3). However, production-scaled CH<sub>4</sub> emissions from unconventional gas production in the Marcellus remain small compared to other basins due to its large production per well. After normalizing emissions by energy produced, total greenhouse gas emissions from Pennsylvania unconventional natural gas production produce half the carbon footprint compared to regionally produced coal, with carbon dioxide emissions from combustion being the dominant source of greenhouse gas emissions for both sources.

## 1. Introduction

Natural gas and coal constitute nearly half of the U.S. total energy production in 2016 (U.S. Energy Information Administration, 2018c). In addition to carbon dioxide (CO<sub>2</sub>) created through the combustion process, these two energy sources release significant amounts of methane (CH<sub>4</sub>) during their production phase (U.S. Energy Information Administration, 2018e). CH<sub>4</sub> is a potent greenhouse gas with 28 times the warming potential of CO<sub>2</sub> over a 100-year period and 84 times the warming potential over a 20-year period (without climate-carbon feedbacks; Myhre et al., 2013). The U.S. Environmental Protection Agency (EPA) estimates that natural gas and coal were responsible for 25% and 8% of the country's anthropogenic CH<sub>4</sub> emissions, respectively, for the year 2016 (U.S. Environmental Protection Agency, 2018b). Over the last 20 years, EPA-estimated CH<sub>4</sub> emissions from the natural gas and coal sectors have decreased by 30- and 40-Tg carbon dioxide equivalent (CO<sub>2</sub>e), respectively (U.S. Environmental Protection Agency, 2018b). For coal, this decrease is driven by both a decrease in U.S. coal production and a shift from underground mining to surface mining, a method of coal mining that produces significantly less CH<sub>4</sub> per unit of coal produced (U.S. Environmental Protection Agency, 2017, 2018b). Contrary to coal, natural gas production in the U.S. has increased by 40% since 1995, but emissions have decreased in national inventories by 16% due to advancements in technology which have led to increased efficiencies in the natural gas extraction process (U.S. Environmental Protection Agency, 2018b; U.S. Energy Information Administration, 2018e). Despite these decreases, natural gas and coal production continue to be a significant source of CH<sub>4</sub> both in the United States and globally. In order to accurately consider the climate impacts from these two energy sources, it is necessary to verify these CH<sub>4</sub> emission inventory estimates.

As CH<sub>4</sub> emanates from numerous reported and unreported sources (Turner et al., 2017), atmospheric studies have played a significant role in evaluating CH<sub>4</sub> emissions (Cui et al., 2017; Ganesan et al., 2017;

Ren et al., 2018; Schwietzke et al., 2016). In comparison to site-level data collected from various anthropogenic sources, atmospheric measurements integrate emissions across large areas, from both natural and man-made sources, allowing for detection and quantification of sources that may be missed or underrepresented in bottom-up inventories (Levin et al., 2010). Recent atmospheric studies measuring CH<sub>4</sub> emissions from natural gas activities ranging from component-level (Zavala-Araiza et al., 2015) to entire gas production basins (Barkley et al., 2017; Peischl et al., 2016; Pétron et al., 2014; Schwietzke et al., 2017; Smith et al., 2015) to continental-scale inversions of emission inventories (Schwietzke et al., 2016) have shown large discrepancies between atmospheric and inventory-based approaches. Emission estimates from this broad range of work systematically find that the EPA consistently underestimates CH<sub>4</sub> emissions from natural gas systems (Alvarez et al., 2018; Brandt et al., 2014; Zavala-Araiza et al., 2015). One reason for this discrepancy may relate to the presence of high emitters responsible for a skewed distribution of CH<sub>4</sub> emissions along the supply chain (Brandt et al., 2014). Multiple studies have shown that a small percentage of components/facilities is responsible for a majority of the CH<sub>4</sub> emissions from natural gas (Mitchell et al., 2015; Omara et al., 2016; Zimmerle et al., 2015). If the EPA's bottom-up emission inventories do not adequately sample and represent these large emitters in their emission factors, the nationwide reported emissions will often underestimate the contribution of natural gas production in the CH<sub>4</sub> budget.

Contrary to the large uncertainties associated with emissions from natural gas, emissions from coal mines are thought to be better understood. Due to the potential safety hazard of CH<sub>4</sub> buildup inside underground mines, quarterly measurements of CH<sub>4</sub> emissions from ventilation shafts, the largest source of CH<sub>4</sub> emissions from underground coal mines, are required by the U.S. Mine Safety and Health Administration (U.S. Environmental Protection Agency, 2018a, 2018b). Smaller sources of coal-based CH<sub>4</sub> emissions, such as from methane drainage systems, have less precise information available (Kirchgessner et al., 2000). Despite the large number of site measurements provided for underground coal mines, there have been few studies performing top-down estimates of individual coal basins in the United States. Such studies using atmospheric measurements can be useful even when thorough site-level data are available, as they can estimate emissions across a large area and thus detect sources that may be missed or underrepresented in bottom-up inventories.

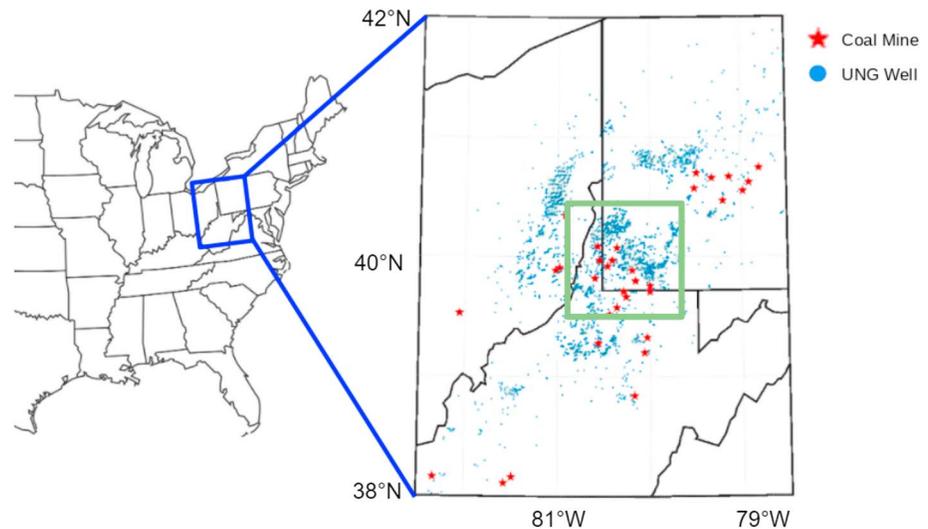
This study addresses the measurement gap associated with emissions from coal while also providing data on CH<sub>4</sub> emissions from the most productive gas basin in the United States by using aircraft data from six flights in 2015–2016 to estimate CH<sub>4</sub> emissions from coal and natural gas sources in the northern Appalachia. Atmospheric CH<sub>4</sub> observations are compared to modeled concentration fields, and emissions from coal and gas are adjusted within the model to create output that matches the observed plume. Additionally, ethane (C<sub>2</sub>H<sub>6</sub>) collected in the region is used to differentiate between coal and gas emissions, and a final range of possible emissions is provided for each source.

## 2. Materials and Methods

### 2.1. Observations

Observations used in this study come from a 2015 and 2016 aircraft campaign performed by the University of Maryland (UMD) over southwestern Pennsylvania and a small portion of northern West Virginia (Ren et al., 2018). Six flights used in this study were performed over the region, three in summer 2015 and three in summer 2016. These observations were broken down further into 19 segments that were downwind of the major coal and unconventional natural gas (UNG) plume. Continuous CH<sub>4</sub> observations from these flights were collected at 0.5 Hz using a Picarro cavity ring down spectrometer. Additionally, continuous C<sub>2</sub>H<sub>6</sub> measurements from two flights in the ACT-America campaign are used to help identify C<sub>2</sub>H<sub>6</sub>/CH<sub>4</sub> ratios during the flights (DiGangi et al., 2018; Dlugokencky et al., 2005).

Unlike a traditional mass-balance approach where the area between the upwind and downwind transects can be used to describe the emissions being quantified, the downwind transects from this study do not all have a corresponding upwind transect. In order to define the emissions and region characterized by the downwind transects, influence functions were generated for each transect using a Lagrangian particle dispersion model (Lauvaux et al., 2012; Uliasz, 1994). These influence functions provide information as to where the air that was measured by the aircraft along each transect was in contact with and influenced by the surface. The influence functions were averaged together for the 19 downwind transects and a centralized area of influence emerged, contained within the latitudes of 39.3–40.6°N and longitudes of 81.0–79.6°W (Figure 1; see supporting information S1 for details on influence functions). This domain was responsible



**Figure 1.** Location of the underground coal mines and UNG wells in the study region. The green square encloses the region downwind of the University of Maryland transects whose emissions are estimated. UNG = unconventional natural gas.

for roughly a quarter of all underground coal production and one third of natural gas production in the Marcellus shale for the year 2015. Because the majority of coal and gas production in the domain lies in and along the southwestern Pennsylvania boundary, for simplicity the study region will be referred to as southwest Pennsylvania (SWPA).

## 2.2. Model and Emissions Inventory

In this study, we use the Weather Research and Forecasting Model with chemistry enabled (WRF-Chem version 3.6.1) to model  $\text{CH}_4$  enhancements with the objective of adjusting regional emissions from coal and UNG sources to create the closest match between observed and modeled  $\text{CH}_4$ . A 3-km resolution domain containing tracers for different regional sources of  $\text{CH}_4$  is centered around the location of the flight campaign, and enhancements are projected from the various tracers for each of the flight days. For more information on model setup, see Barkley et al. (2017).

To project  $\text{CH}_4$  enhancements from WRF-Chem, a  $\text{CH}_4$  emissions inventory of the region was created. For anthropogenic sources other than natural gas production and processing, the EPA Gridded 2012 Methane Emissions Inventory was used as input (Maasackers et al., 2016). For  $\text{CH}_4$  emissions from natural gas production, well production data were first obtained from the Pennsylvania Department of Environmental Protection (Pennsylvania Department of Environmental Protection, 2018b), the Ohio Department of Natural Resources (Ohio Department of Natural Resources, 2018), and the West Virginia Department of Environmental Protection (West Virginia Department of Environmental Protection, 2018). Wells were sorted into either conventional or unconventional. Conventional wells were assigned a  $\text{CH}_4$  emission rate of 11% of production (Omara et al., 2016). Emissions from UNG wells are assigned a first-guess emission rate of 1% of production. This first-guess emission rate serves as a way to proportionally adjust emissions from UNG in the model and has no impact on the final, optimized emission rates. Potential emissions from abandoned wells were not considered in the inventory due to their negligible emission range compared to other sources within the domain (Kang et al., 2016).

## 2.3. Optimization Technique

The overall objective of the emissions optimization approach is to scale emissions from both coal and UNG sources such that the modeled enhancements produced by WRF-Chem match  $\text{CH}_4$  observations from the flight campaign. To do this,  $\text{CH}_4$  observations must first be converted to enhancements by subtracting off a background value unique to each flight

$$X_{\text{enh}} = X_{\text{obs}} - \text{BG} \quad (1)$$

where  $X_{\text{obs}}$  are the original  $\text{CH}_4$  observations, BG is the chosen background value for a given flight, and  $X_{\text{enh}}$  is the observed  $\text{CH}_4$  enhancement. The background value BG represents all  $\text{CH}_4$  from sources not accounted

for in the model (e.g., the overall regional atmospheric CH<sub>4</sub> mole fraction). To find this background value, we use CH<sub>4</sub> observational values in the boundary layer in areas where the observations are at their lowest values and the model shows little or no enhancement. These observations have minimal intrusion from sources within the model domain and thus best represent the background air mass not represented within the model. The mean value of these observations is subtracted from all boundary layer observations, resulting in a set of observed enhancements for each flight.

After the CH<sub>4</sub> enhancement is determined, the flights are dissected into individual transects that intersect the coal and UNG plume within the boundary layer. Using the model to forecast where the major plumes are located, we find 19 transects from the six flights which definitively intersect the major coal and UNG plume. For each of the 19 transects we solve for a range of coal and UNG emission rates through the following steps: First, model-projected CH<sub>4</sub> enhancements from sources unrelated to coal and UNG are subtracted from the observed enhancements using the equation

$$X_{\text{source}} = X_{\text{enh}} - Y_{\text{other}} \quad (2)$$

where  $Y_{\text{other}}$  are the total modeled enhancements at each observation from sources other than coal and UNG emissions (conventional gas wells and landfills; see supporting information S1), and  $X_{\text{source}}$  are the observed enhancements that are believed to originate from coal and UNG sources. Next, observed CH<sub>4</sub> enhancements are compared to the model-projected enhancements from coal and UNG. Modeled enhancements from coal and UNG are each adjusted by individual scaling factor to minimize a cost function given below.

$$J = \sum_{i=1}^n |X_{\text{source}_i} - C_{\text{coal}} Y_{\text{coal}_i} - C_{\text{ung}} Y_{\text{ung}_i}| \quad (3)$$

where  $i$  is the observation,  $n$  is the number of observations in the transect,  $Y_{\text{coal}}$  and  $Y_{\text{ung}}$  are the modeled coal and UNG enhancements at each observation,  $X_{\text{source}_i}$  is the total observed coal and UNG enhancement at observation  $i$ , and  $C_{\text{coal}}$  and  $C_{\text{ung}}$  are constants applied to the model enhancements to minimize the cost function  $J$ . In this study we use the absolute error between the observed and modeled enhancement as the basis for the cost function. The absolute error is chosen instead of the root-mean-square error because the latter emphasizes minimizing the difference between extreme values and puts less emphasis on smaller, broad enhancements, a trait not desired for this experiment. We note that using a root-mean-square error as the cost function produced a final emissions result that differed by <10% from results found using the absolute error.

Because modeled enhancements scale linearly with their associated emissions, the scaling factors used on the modeled coal and UNG enhancements to minimize the cost function are the scaling factors the emissions of each source need to be adjusted to achieve an optimal match between the observed and modeled enhancements. However, due to the collocation of coal and UNG sources, there may be multiple scaling combinations that produce similar cost function values for a given transect. To account for this range of possible solutions, we classify any combination of coal and UNG scaling factors that produces a cost function value within two times the minimum cost function value of the optimized solution to be a feasible solution. This optimization is done for all 19 transects, and the feasible range of emission rates for coal and UNG is overlapped to find which combinations satisfy the majority of transects.

#### 2.4. Optimization Using Ethane

Regional ethane (C<sub>2</sub>H<sub>6</sub>) measurements can be used as an additional tracer to solve for the ratio of contributed emissions from natural gas and coal sources (Peischl et al., 2016; Smith et al., 2015). Prior measurements of CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> from natural gas wells in the flight domain show an average C<sub>2</sub>H<sub>6</sub>/CH<sub>4</sub> source ratio of  $0.072 \pm 0.007$  (median: 0.055) (Román-Colón & Ruppert, 2016). Measurements of underground coal mines in SWPA have an average C<sub>2</sub>H<sub>6</sub>/CH<sub>4</sub> source ratio of  $0.0030 \pm 0.0027$  (Kim, 1973; Laughrey & Baldassare, 1998). Biogenic sources of CH<sub>4</sub> (landfills and animal agriculture) emit no C<sub>2</sub>H<sub>6</sub>. By multiplying these ratios by their corresponding source in the CH<sub>4</sub> emission inventory, the CH<sub>4</sub> emissions inventory can be transformed into a regional C<sub>2</sub>H<sub>6</sub> emissions inventory that can be used as model input to project C<sub>2</sub>H<sub>6</sub> plumes and model the C<sub>2</sub>H<sub>6</sub>/CH<sub>4</sub> ratio of the major coal and UNG plume in the study region.

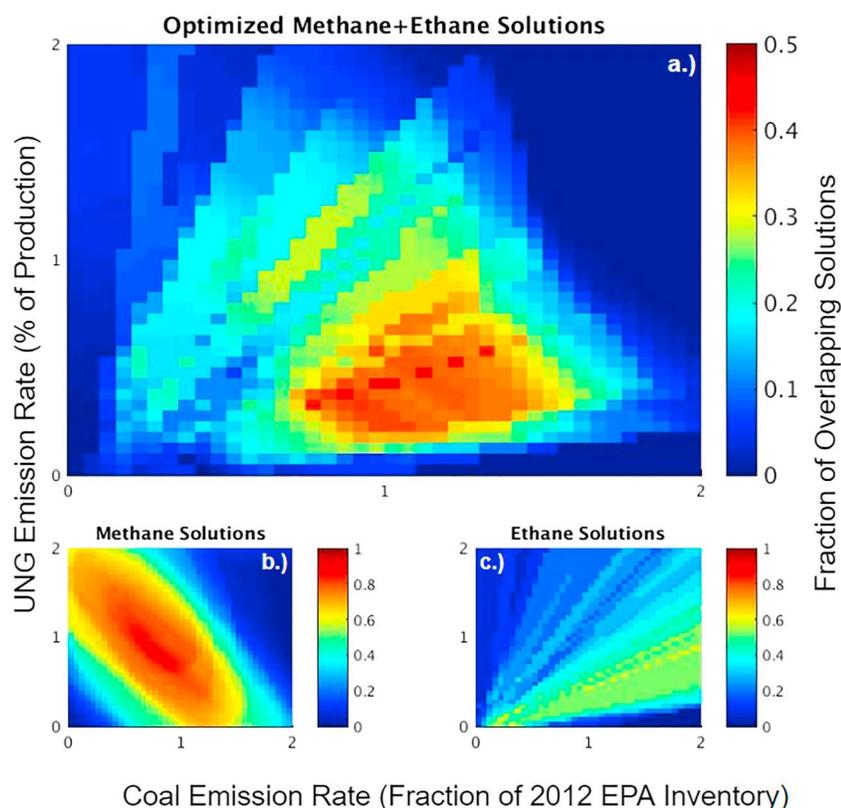
If the C<sub>2</sub>H<sub>6</sub>/CH<sub>4</sub> ratio of the coal and UNG plume is known through observations, the coal and UNG emission ranges can be scaled to create a modeled plume with the same ratio as the observed plume. In this

study we use flask samples from the UMD flights as well as continuous  $C_2H_6$  and  $CH_4$  measured using the CAMS-2 instrument from three flight segments that transect the region from the ACT-America mission. Lagrangian footprints were generated for the ACT-America flights to ensure that only measurements with footprints that overlapped the study region were used (see supporting information Figure S1). From these data sets, we determine the  $C_2H_6/CH_4$  ratio within the mixed coal and UNG plume lies between 0.010 and 0.028. Using this range and the source ratios measured from coal and gas sources in the region, a model analysis is performed to find coal and UNG emission rates for each transect that produced a  $C_2H_6/CH_4$  ratio that fell within the accepted range. The  $C_2H_6$  solutions for each transect are overlapped with their corresponding  $CH_4$  solution counterpart to find which solutions satisfy both criteria and thus best characterize the regional coal and UNG emissions.

### 3. Results and Discussion

Figure 2 presents the results of the joint  $CH_4$  and  $C_2H_6$  optimization in comparison to single-tracer optimization (i.e.,  $CH_4$ -only and  $C_2H_6$ -only optimization). Based on the  $CH_4$  optimization alone, emissions from both the coal and UNG sectors have multiple solutions due to difficulty in attributing the total enhancements observed to their respective sources, reflecting the limitations of a typical mass-balance calculation. The total regional  $CH_4$  emission rate is constrained but the lack of additional information produces an unconstrained set of coal and UNG solutions with a negative correlation between the potential rates of the two sources. When optimizing the sources with  $C_2H_6$  measurements, the opposite situation occurs. In this case, the set of solutions is constrained such that the ratio of coal-to-UNG emissions remains constant in order to match the observed  $C_2H_6/CH_4$  ratios. However, the total emissions are unconstrained with a positive correlation between coal and UNG emissions. Both approaches offer a wide range of solutions for each sector, underconstrained by the observations. For the joint optimization, and because of the characteristics of the individual solutions described above, the region of overlap between the  $CH_4$  and  $C_2H_6/CH_4$  solutions is small for any given transect (see Figure S5 for individual solutions). Over 19 transects collected during the UMD aircraft campaign, we performed a Monte Carlo analysis to optimize jointly coal and UNG emissions rates. A common set of solutions emerge (Figure 2) in which emissions from UNG production and gathering facilities lie within  $0.5 \pm 0.3\%$  of natural gas produced, and emissions from regional underground coal mines are found to be  $1.1 \pm 0.4$  times the EPA's 2012 gridded inventory estimate.

From the results of this study, we estimate emissions from UNG production and gathering facilities in SWPA to be equivalent to  $0.5 \pm 0.3\%$  of production, in agreement with published top-down emission estimates from northeast PA ( $0.36 \pm 0.09\%$ ) and SWPA (0.0–3.5%) and site-level measurements at well sites in northeast PA ( $0.44 \pm 0.15\%$ ) and SWPA ( $0.57 \pm 0.23\%$ ; Alvarez et al., 2018; Barkley et al., 2017; Omara et al., 2016; Ren et al., 2019). These emission rates as a percent of production are lower than rates found from top-down studies performed in other gas basins, with the next lowest rate measured in the Haynesville shale at 1.3% (Alvarez et al., 2018; Karion et al., 2015; Peischl et al., 2016; Pétron et al., 2014; Schwietzke et al., 2017; Smith et al., 2015). The low fractional emission rates in this region are likely due to Marcellus wells having the highest production per well in the United States (Barkley et al., 2017; U.S. Energy Information Administration, 2016), requiring fewer components to produce large amounts of gas and thus lowering the potential for leaks (Brantley et al., 2014; Mitchell et al., 2015; Omara et al., 2016). However, though the emission rate is low compared to top-down estimates performed in other regions, it is significantly higher than the inventory estimate reported by the Pennsylvania Department of Environmental Protection for the year 2015 (Pennsylvania Department of Environmental Protection, 2018a). Greene and Washington counties, the two counties responsible for the majority of UNG production in our study area, are reported by the Pennsylvania Department of Environmental Protection to have emissions equivalent to 0.10% of production for the year 2015 (Table S4). This number is lower than any peer-reviewed estimate for emissions from natural gas production and is outside of the error bounds for this study as well as a previous bottom-up study performed in the area (Omara et al., 2016). Thus, while UNG production in this region may be efficient compared to other gas basins in terms of  $CH_4$  emissions per production, the optimized emissions from our study shed light on a large underestimation of state inventory-based emission estimates from UNG sources in SWPA. Discrepancies between bottom-up inventory estimates of UNG emissions and independent verification have been observed in multiple studies prior to this one, with bottom-up inventories nearly always being lower than top-down studies (Brandt et al., 2014).



**Figure 2.** (a) Fraction of total number of Monte Carlo simulations that fulfills both the  $\text{CH}_4$  and  $\text{C}_2\text{H}_6/\text{CH}_4$  optimization criteria using different combinations of coal and UNG emission rates for all of the 19 transects. (b) Fraction of simulations that fulfills only the  $\text{CH}_4$  optimization criteria. (c) Fraction of simulations that fulfills only the  $\text{C}_2\text{H}_6/\text{CH}_4$  optimization criteria. UNG = unconventional natural gas.

In addition to solving for UNG emissions, this study finds emissions from underground coal mines in the region to be  $1.1 \pm 0.4$  times the EPA's 2012 gridded inventory. Given that Pennsylvania coal production in the region has changed by  $<10\%$  from 2012 to 2015, we assume that the 2012 gridded inventory remains an accurate estimate of the emissions for 2015 (U.S. Energy Information Administration, 2018a). Thus, the emission range found from this study indicates that EPA estimates of total emissions from underground coal mines in Pennsylvania and northern West Virginia are accurate to within 50%. Such a result is not unexpected. Emissions from ventilation shafts are measured four times each year and are believed to represent the majority of  $\text{CH}_4$  emissions from underground mines (U.S. Environmental Protection Agency, 2017).

Sources of uncertainty from the dual-optimization technique are addressed conservatively to ensure that the final emissions estimate for coal and UNG accurately represents the range of possible values. This uncertainty can be broken into five distinct categories: error in the chosen background value, error in the emissions inventory, error in the modeled wind speed/mixing height, error in the model transport, and error in the assumed  $\text{C}_2\text{H}_6/\text{CH}_4$  ratio of coal and gas emissions. Uncertainty in choosing the appropriate background  $\text{CH}_4$  value and potential errors from emissions not optimized in this study are addressed using a Monte Carlo approach, drawing a random error from a normal distribution with  $\mu = 0$  and  $\sigma = 2.5$  ppb for the background error and  $\mu = 0\%$  and  $\sigma = 25\%$  for the error in the emissions from unsolved  $\text{CH}_4$  sources. However, due to the magnitude of the major coal and UNG plume observed in each transect ( $>100$  ppb), these errors have near-negligible impacts on the overall range of solutions. Errors in the model wind speed and mixing height impact model-projected enhancements and are corrected based on the errors of each day's meteorology determined using vertical profile and reanalysis modeling data (Dee et al., 2011; Kalnay et al., 1996; Mesinger et al., 2006). Errors in the model transport are accounted for by allowing for small shifts in the modeled plume location along a transect, reducing mismatch between the observed and modeled plume location which would otherwise produce a low bias on the result. Lastly, potential errors in the assignment of a  $\text{C}_2\text{H}_6/\text{CH}_4$  ratio for coal and gas sources in the model are addressed by varying their source

**Table 1**  
Calculated CO<sub>2</sub>e From Coal and Natural Gas Sources in PA for the Year 2016

Source	Petajoules produced (2016)	Total CO <sub>2</sub> e (Tg)	CO <sub>2</sub> e per energy produced (g/MJ)	Contribution of CH <sub>4</sub> to CO <sub>2</sub> e
Coal	850	86.6	102	13%
UNG	5,200	274.0	53	5%
CvNG	130	12.7	97	52%

*Note.* Only CH<sub>4</sub> emissions from the production phase of each source are considered. Only CO<sub>2</sub> released through combustion is considered. CO<sub>2</sub>e is considered over a 100-year period using a conversion of 1 kg CH<sub>4</sub> = 28 kg CO<sub>2</sub>e. UNG = unconventional natural gas; CvNG = conventional natural gas.

ratios in a separate Monte Carlo analysis. For gas sources, a ratio is drawn from a normal distribution with  $\mu = 0.072$  and  $\sigma = 0.007$ , and for coal sources, a ratio is drawn from a normal distribution with  $\mu = 0.003$  and  $\sigma = 0.027$ . The resulting spread from these five sources of uncertainty can be seen in Figure 2 with a clear set of converging solutions. More details regarding the uncertainty assessment can be found in section S3 of the supporting information section.

Here we measured CH<sub>4</sub> emissions related to production from the two largest sources of energy production in Pennsylvania (U.S. Energy Information Administration, 2018d), but to understand the full climate impacts associated with coal and gas production in the state, the CO<sub>2</sub> released through combustion processes must be considered in addition to CH<sub>4</sub> emissions. To measure the potential implications of our findings, we consider the contribution CH<sub>4</sub> emissions have toward the carbon dioxide equivalent (CO<sub>2</sub>e) of these sources over a 100-year period after applying the rates found in this study for the state of Pennsylvania (Table 1; see supporting information for methods. For comparison over a 20-year time frame, increase all CH<sub>4</sub> contributions to CO<sub>2</sub>e by a factor of 3). From these calculations, a number of important conclusions can be drawn. First, we find that CH<sub>4</sub> contributes to only 13% of the CO<sub>2</sub>e associated with PA underground coal mines and only 5% for CH<sub>4</sub> emissions from UNG production; CO<sub>2</sub> emissions from combustion dominate the CO<sub>2</sub>e associated with both sources. Because of this, underground coal production has nearly twice as much of an impact on the climate compared to UNG produced in PA, driven by the cleaner combustion of natural gas compared to coal (U.S. Energy Information Administration, 2018b). Thus, any energy derived from PA underground mines that is directly replaced with energy derived from PA UNG production represents a decrease of approximately 50% in the CO<sub>2</sub>e released through production and energy generation. Second, the small contribution CH<sub>4</sub> has toward the CO<sub>2</sub>e from PA UNG production limits the potential reduction of the climate impacts of natural gas through mitigating CH<sub>4</sub> emissions from PA UNG production. For example, reducing CH<sub>4</sub> emissions from PA UNG production to half of their current value (0.50% to 0.25%) would only be reducing the overall CO<sub>2</sub>e from PA UNG production by 6.4 Tg CO<sub>2</sub>e. An equivalent reduction in CO<sub>2</sub>e while conserving total energy content could also be achieved by replacing 14% of coal production in PA with a 2.3% increase in UNG production (U.S. Energy Information Administration, 2016, 2018c) or replacing 2.5% of PAs energy production from UNG with a renewable energy source. We emphasize that these calculations do not consider potential loss rates of CH<sub>4</sub> from the natural gas storage and distribution sector. The EPA bottom-up inventory estimates these emissions to be less than 0.4% of total production (Alvarez et al., 2018), but recent studies have found discrepancies with distribution sector estimates (Lamb et al., 2016; McKain et al., 2015; Ren et al., 2018). More research is needed to quantify downstream emissions from natural gas on a nationwide scale.

The analysis above applies specifically to UNG wells in PA, whose high gas production per well results in a low emission rate when normalized to production. Low-producing wells, such as the older conventional natural gas (CvNG) wells in PA, have a much higher emission rate when normalized to production. Because of the higher loss rate of CH<sub>4</sub> per unit of energy produced, these CvNG wells have a more significant portion of their total CO<sub>2</sub>e contributions coming from CH<sub>4</sub> emissions (Alvarez et al., 2018; Omara et al., 2016). Emissions from CvNG wells were not solved for in this study, but a previous study estimated an emission rate from these wells equivalent to at least 11% of their production (Omara et al., 2016). At this leakage rate, the production and combustion of gas produced from these wells result in a CO<sub>2</sub>e per Joule equivalent to PAs underground coal mines. Replacing these 60,000 CvNG wells with less than 100 new UNG wells would effectively replace all natural gas produced from PA CvNG wells while halving the CO<sub>2</sub>e per Joule due to the much lower CH<sub>4</sub> emission rate from high-producing UNG wells. Such a concept is not limited to PA. Plugging inefficient, low-producing natural gas wells and replacing their energy with newer wells in

high-producing gas basins may be an effective means of lowering CH<sub>4</sub> emissions on a national scale, though the actual practicality of such methods is outside the scope of this study. Policy measures that incentivize reducing GHG emissions without prescribing the methodology to do so could expedite research toward the most effective means of emissions reduction.

#### 4. Conclusion

This study presents one of the first aircraft-based emission estimates of underground coal mines in the United States. Through the model optimization technique presented in this work, we find CH<sub>4</sub> emissions from underground coal mines in SWPA to be a factor of 0.7–1.5 times higher than values reported by the EPA and emissions from UNG sources to have an emission rate equivalent to  $0.5 \pm 0.3\%$  of production. Emissions from UNG in SWPA agree with other studies analyzing emission rates from the Marcellus shale, showing emission rates lower than the national average when scaled to production but higher than state reported estimates by a factor of 2 to 8. Despite this large discrepancy, CH<sub>4</sub> emissions from UNG sources with small emission rates contribute only a small fraction to their total greenhouse footprint compared to the CO<sub>2</sub> released through combustion process over a 100-year period.

This study shows that there is great potential in utilizing C<sub>2</sub>H<sub>6</sub> measurements in regions where multiple, colocated CH<sub>4</sub> sources exist with unique C<sub>2</sub>H<sub>6</sub>/CH<sub>4</sub> ratios. Large discrepancies were found between limited, single point C<sub>2</sub>H<sub>6</sub> measurements from flask samples and continuous C<sub>2</sub>H<sub>6</sub> measurements. Future studies planning on using C<sub>2</sub>H<sub>6</sub> as a tracer would benefit from high-quality, high-frequency C<sub>2</sub>H<sub>6</sub> measurements taken simultaneously with CH<sub>4</sub> measurements.

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