Supplemental Information

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SI Text

Definition of the Sampling Areas for Flux Calculations. The original sampling area (OSA) used for both aircraft flux calculations and a bottom-up inventory is defined by the bounding longitude coordinates −80.6405 to −79.9315 and the bounding latitude coordinates 40.1475 to 39.7217. This area encompasses all of Green County, PA, most of Washington County, PA, and parts of Fayette County, PA, Marshall County, WV, and Ohio County, WV, for a total area of 2,844 km^2. A bottom-up inventory was also conducted for the upwind accumulation area (UAA), an area represented by the extent of an 18-h back trajectory (Fig. S1) run from the dimensions of the OSA and comprises 14,597 km^2, extending into northwestern WV and southeastern OH. The counties included in the UAA are as follows: PA: Fayette, Greene, and Washington; WV: Calhoun, Doddridge, Gilmer, Harrison, Lewis, Marion, Marshall, Monongalia, Ohio, Pleasants, Ritchie, Tyler, Wetzel, Wirt, and Wood; OH: Athens, Belmont, Monroe, Morgan, Noble, and Washington.

Regional Bottom-up Emission and Flux Calculations. ***Natural gas and oil production.*** Well inventories and production data for active oil and gas wells in the counties of interest were downloaded from the Pennsylvania Department of Environmental Protection (PADEP),[*,†] the West Virginia Department of Environmental Protection (WVDEP),[‡,§] and the Ohio Department of Natural Resources (ODNR).[¶,‖] Duplicate entries were removed using well American Petroleum Institute numbers. We identified 57,673 wells across all counties of interest.

Wells were categorized by primary production type (gas, oil, or unknown) according to information provided in the state databases. Gas production considers only nonassociated gas production (i.e., gas produced only from wells specified as gas or coal-bed methane well types). Oil production is the total oil produced from wells specified as oil or combined oil and gas wells. Associated gas, a coproduct of oil production, is included in the transmission/distribution emission calculation but not production emissions. Accounting for emissions from associated gas production is assumed to occur via estimated emissions from oil wells. The PADEP dataset includes other well types, such as storage, injection, observational, and multiple wellbore type wells, which are not included in our analysis (356 wells). Five of these excluded wells fall within the bounding coordinates of both OSA and UAA, with a total reported gas production of 0.28 million $m^3\ d^{-1}$, or ∼1% of our estimated regional production.

Wells were mapped and clipped to the bounding coordinates of the sampling areas using the geographic coordinates provided in the state data. We found a total of 1,616 wells with no geographic coordinates (1,576 PA, 0 WV, 40 OH); production from these wells is not included in our inventories. The final count of wells within the bounding coordinates of the OSA and UAA are 3,438 (57.3% gas, 1.8% oil, 40.8% unknown) and 19,867 (65.6% gas, 13% oil, 21.4% unknown), respectively.

PADEP datasets for unconventional (January–June 2012 production) and conventional (January–December 2012 production) wells and the most recent production data available from ODNR (2011 production) provide total gas, condensate, and oil production over the given reporting period and days in production, making calculation of daily production straightforward. The latest production data available from WVDEP is also for 2011. WVDEP production data are available as a single zip file for all wells statewide. WV data report total oil and gas production on a monthly basis; WV daily production then is estimated by dividing the total annual production by the number of months in production and assuming 1 mo = 30 d. Production rates for all wells are allocated to emission categories (unassociated gas, oil) according to primary product allocation described above.

Minor adjustments to the PADEP data were necessary because of assumed errors in reporting. While validating the production data, we found a small subset of conventional wells permitted in the 1980s under a single operator (available upon request) that had reported 2011 productions 3 orders of magnitude higher than that reported in any previous year since the wells' inceptions; we adjust production on these wells assuming that cubic feet, rather than thousand cubic feet, was erroneously reported.

Natural gas pipeline throughput. We assume all gas produced (associated and nonassociated) within the sampling areas enters transmission lines within the respective areas. Local gas throughput is estimated at 23.03 million $m^3\ d^{-1}$ for the OSA and 37.67 million $m^3\ d^{-1}$ for the UAA. Additional gas enters the sampling areas through interstate pipelines that supply natural gas to regional urban centers. The Energy Information Administration (EIA) reports state-to-state capacities, providing inflow and outflow capacities at the state and county levels.[**]

Most pipelines do not operate at capacity, especially during the low-load summer periods when our sampling took place. We estimate the throughput into the sampling areas by comparing total system capacity reported by the EIA with Pipeline and Hazardous Materials Safety Administration (PHMSA)[††]-reported volumes moved in the region's major interstate pipelines. PHMSA data report 144 million $m^3\ d^{-1}$ of natural gas transmitted through 8,576 miles for the Texas Eastern Transmission pipeline, 82 million $m^3\ d^{-1}$ over 10,843 miles for the Columbia Gas Transmission line (another major line entering southern PA, but outside both of the sampling areas), and 73 million $m^3\ d^{-1}$ over 3,500 miles for the Dominion Transmission line, for a total of 300 million $m^3\ d^{-1}$. Data for the Equitrans line could not be found. Cumulative capacity for these three lines over their total mileage is 1,247 million $m^3\ d^{-1}$, indicating that the lines on average are running at roughly a quarter of capacity. Thus, our best estimate for interstate transmission is one quarter of the line capacities entering the sampling areas or 29.08 million $m^3\ d^{-1}$ for the OSA and 57.94 million $m^3\ d^{-1}$ for the UAA. A portion of the interstate pipeline throughput is already accounted by local production entering the interstate lines: 3.37 million $m^3\ d^{-1}$ in the OSA and 3.7 million $m^3\ d^{-1}$ in the UAA. Line volumes are, therefore, reduced by these locally produced inputs to

*PADEP oil and gas production: www.paoilandgasreporting.state.pa.us/publicreports/Modules/Production/ProductionHome.aspx.

†PADEP oil and gas well inventory: www.portal.state.pa.us/portal/server.pt/community/oil_and_gas_reports/20297.

‡2011 WV oil and gas production: www.dep.wv.gov/oil-and-gas/databaseinfo/Pages/default.aspx.

§WVDEP oil and gas inventory: http://tagis.dep.wv.gov/oog/.

¶OH DNR well inventory: www2.dnr.state.oh.us/mineral/OHRbdmsOnline/WebReportAccordion.aspx.

‖OH DNR oil and gas production: ohiodnr.com/mineral/production/tabid/15389/Default.aspx.

**EIA state-to-state pipeline data: www.eia.gov/naturalgas/data.cfm.

††PHMSA transmission line data: phmsa.dot.gov/portal/site/PHMSA/menuitem.ebdc7a8a7e39f2e55cf2031050248a0c/?vgnextoid = a872dfa122a1d110VgnVCM1000009ed07898RCRD&vgnextchannel = 3430fb649a2dc110VgnVCM1000009ed07898RCRD&vgnextfmt = print.

avoid double accounting. Total interstate pipeline transport through the OSA and UAA is estimated at 25.71 and 54.24 million $m^3\ d^{-1}$, respectively.

It should be noted that EIA reports outflow capacities from Washington County of 31 million $m^3\ d^{-1}$ that move gas north, and Fayette County capacity of 9 million $m^3\ d^{-1}$ moving gas east. Gas in these pipelines may or may not travel through both of the sampling regions. Because the majority of production for these counties occurs outside the sampling area, we assume these lines are outside of the sampling area.

Distribution throughput volumes are already included in the estimates of transmission throughput volumes and are not used in emission calculations.

Coal production. Active deep and surface coal mines within the boundaries of the study areas were determined from federal data from the Department of Labor.[‡‡] Our analysis uses 2012 production data from underground and surface mines. A total of seven coal mines are located in the OSA (five in PA and two in WV), and an additional eight mines are located in the UAA (five in WV and three in OH). With the exception of one surface mine in the UAA, all of the mines are underground operations. Annual coal production per mine as of 2012 was converted to daily rates assuming 365 d of operation.

Energy sector methane emissions. Regional expected emissions from fossil fuel production for the OSA and UAA are based on fugitive methane emission factors taken from refs. 1–3 for natural gas and ref. 1 for oil and coal. Ref. 1 presents methane emission factors for the natural gas life cycle as a percentage of lifetime well production and classifies emission factors by activity or operational sector: completion (e.g., flowback), routine losses at well site (e.g., production), liquids unloading, processing, and transmission/distribution. However, percent loss factors are not easily applied to all activities or sectors. Flowback and liquid unloading activities are isolated in time and space: thus the percent loss rate can only apply to the lifetime production of the individual well undergoing the activity, not the sum of production from all wells. Loss rates for the transmission/distribution sector reflect the portion of production lost over the total distance that a given unit of gas is moved, which may be greater than the pipeline distances contained within the sampling areas.

We apply the ref. 1 percentages to the total methane production from unassociated gas produced in the sampling areas for routine and continuous upstream activities (i.e., production and processing emissions). Unassociated natural gas production is converted to methane production assuming US default gas composition (4). Emissions from flowback activity and the transmission/distribution sector are estimated using alternative, but comparable, emission factors as described below. We assume no liquid unloading activity.

To assess well completion/flowback activity during the sampling times, we analyzed aerial photos taken during the sampling, PADEP well records and inspection reports, and compared production from Marcellus wells over the last 6 mo of 2011 with production through the end of June 2012. These analyses showed no evidence of well completion or unloading activity within the PA portion of the study area during the sampling dates. Similar data were not available for the OH and WV counties of interest. We searched these areas, as well as the PA counties of interest, using data from FracFocus.org (www.fracfocusdata.org/DisclosureSearch/MapSearch.aspx). Data submitted to FracFocus.org is voluntarily submitted by participating oil and gas operators and is not complete. The database does, however, provide publically accessible data on well stimulation start dates. A search for wells stimulated within the target counties from January 1, 2012 to June 22, 2012 returned a list of 88 wells. We assume a schedule of 2 d for stimulation, 1 d for rigdown, and 7–14 d flowback. Assuming this work schedule, we determined that 11 wells were potentially in flowback in the counties of interest at the time of the sampling, although only one of these was within the bounding coordinates of the OSA and not near any of the pads sampled.

As previously noted, emissions from flowback cannot be estimated from regional production rates. We instead use the Environmental Protection Agency (EPA) emission factor reported in ref. 2 of 37.1 thousand $m^3\ CH_4\ d^{-1}$, which on a per-well basis agrees reasonably well with the estimated loss rate reported in ref. 1. The EPA factor reflects potential methane emissions assuming 100% venting. Actual methane emissions may be reduced by flaring or capture. We note that there were no lit flares sighted during the flyover in the original study area of Fig. 1. We present a range reflecting 100% venting (high) and 49% venting (low) and estimate an additional 0.05–0.1 and 0.01–0.02 g $CH_4\ s^{-1}\ km^{-2}$ due to potential flowback activity during the time of the sampling for the OSA and UAA, respectively.

Emission estimates for the transmission/distribution sector are based on total methane pipeline throughputs (i.e., the sum of pipeline throughputs converted to methane assuming US default pipeline gas composition) and the total pipeline length contained within the sampling areas. Pipeline length is estimated by digitizing and clipping pipeline maps for the eastern United States available from the EIA to the boundaries of the sampling areas. Total length for the OSA and UAA are 809.7 km and 2,571 km, respectively. NETL (2011) provides a mass x distance – based emission factor for the natural gas transmission system of 5.37×10^{-6} kg km^{-1}. Combined transmission/distribution emissions are assumed to be twice the emission factor for the transmission alone.

Howarth et al. (2011) loss rates assume dry gas (>96% methane) as is often found in the northeastern Marcellus production areas. Our sampling area is dominated by wet gas production and a lower methane fraction. Not accounting for this change in gas composition will over-estimate regional methane emissions. Thus, we assume US default gas compositions to convert the volume of gas loss to a volume of methane loss in production, processing, and transmission/distribution sectors (4). Tables S1 and S2 summarize emissions from nonassociated natural gas production by operational phase using Howarth et al. (2011) emission factors.

The emission factor for oil production is estimated at 0.7 g C MJ^{-1} oil or, 0.76 $m^3\ bbl^{-1}$ oil produced (1). Estimated emission factors for deep coal range from 7.8 to 9 m^3 per ton of coal. In this analysis, we use the mean of 8.4 $m^3\ ton^{-1}$ of coal produced to estimate emissions from area coal mines. The methane emission factor from surface-mined coal is estimated at $2.3 \pm 1.0\ m^3$ per ton of coal (1). Emissions from oil and coal are reported in Tables S1 and S2.

Other methane emissions. In addition to energy production related methane emission, we also investigated other significant methane sources such as landfills and animal feeding operations (AFOs). We used county reported animal counts of dairy cattle, other cattle, pigs, chickens and turkeys. As some counties withhold animal counts to protect the privacy of individual farms, the animal counts for withheld counties were estimated as described by Hong et al., 2011 in ref. 5. Emission factors for CH_4 from enteric fermentation and manure management were taken from refs. 6 and 7. The availability of only county scale animal counts affected our ability to filter the data specific to the original or extended sampling area. To account for this we used county animal counts for every county the sampling area crossed which extends to an area much greater than our study regions. The total CH_4 emission for all of the counties was then divided by the total area of the counties and this resulting average across this area is expected to be representative of our smaller study regions. Tables S1 and S2 summarize emission factors, animal counts and the resulting emissions. We estimate an AFO emission of 0.015 g $CH_4\ s^{-1}\ km^{-2}$ for both the OSA and UAA.

‡‡Department of Labor 2010 employment/production data set: catalog.data.gov/dataset/employmentproduction-data-set-yearly.

A 2010 EPA summary and report of direct methane emissions in the region was used to identify 3 and 29 potential sources in the OSA and UAA, respectively (8). Of these, 2 and 5 sources in the OSA and UAA, respectively, overlapped with our analysis of methane emissions from energy production and were excluded. The remaining 1 source in the OSA contributed a negligible amount of methane. In the UAA the remaining 24 sources include a landfill, which produces 85% of the CH_4 emission from this subset of emission sources, various production facilities, power plants and incineration plants. The combined emissions for the UAA are equal to 0.019 g CH_4 s^{-1} km^{-2}.

Comparison with other inventories. For comparison, we calculated expected emissions from regional nonassociated natural gas production for the OSA using emission factors inferred from the National Energy Technology Laboratory (NETL) as reported in 2011(2). Converting from mass of methane to mass of C in methane and from higher heating values to lower heating values, the NETL (2) estimates of fugitive methane emissions are 0.29 g C MJ^{-1} for production and processing and 0.08 g CMJ^{-1} for transmission. The NETL (2) analysis does not include emissions from the distribution sector; we assume that fugitive emission rates for the distribution sector are equal to those from the transmission sector and simply double the emission factor. Emission intensity is converted to a percent loss according to calculations in ref. 1 (0.2% loss in production and processing, 1.1% loss in transmission/distribution). We estimate 4.44–4.49 g CH_4 s^{-1} km^{-2} on the basis of the NETL (2) emission factors and our own analysis of other methane emitters. Table 3 summarizes emissions using the NETL (2) approach in comparison with our estimates using emission factors from ref. 1.

1. Howarth RW, Santoro RL, Ingraffea A (2011) Methane and the greenhouse-gas footprint of natural gas from shale formations. *Clim Change* 106(4):679–690.
2. National Energy Technology Laboratory (2011) *Life Cycle Greenhouse Gas Inventory of Natural Gas Extraction, Delivery and Electricity Production* (DOE/NETL Publication 2011/1522). Available at www.netl.doe.gov/File%20Library/Research/Energy%20Analysis/Life%20Cycle%20Analysis/NG-GHG-LCI.pdf. Accessed December 11, 2012.
3. US Environmental Protection Agency (2010) Greenhouse gas emissions reporting from the petroleum and natural gas industry: Background technical support document. Available at www.epa.gov/ghgreporting/documents/pdf/2010/Subpart-W_TSD.pdf. Accessed December 15, 2011.
4. American Petroleum Institute (2009) Compendium of greenhouse gas emissions methodologies for the oil and natural gas industry. Available at www.api.org/ehs/climate/new/upload/2009_ghg_compendium.pdf. Accessed December 14, 2012.
5. Hong B, Swaney DP, Howarth RW (2011) A toolbox for calculating net anthropogenic nitrogen inputs (NANI). *Environ Model Softw* 26(5):623–633.
6. Jorgensen H (2007) Methane emission by growing pigs and adult sows as influenced by fermentation. *Livest Sci* 109(1-3):216–219.
7. Zhou JB, Jiang MM, Chen GQ (2007) Estimation of methane and nitrous oxide emission from livestock and poultry in China during 1949-2003. *Energy Policy* 35(7):3759–3767.
8. US Environmental Protection Agency (2010) EPA greenhouse gas reporting program 2010 summary. Available at www.epa.gov/ghgreporting/ghgdata/2010data.html. Accessed May 8, 2013.

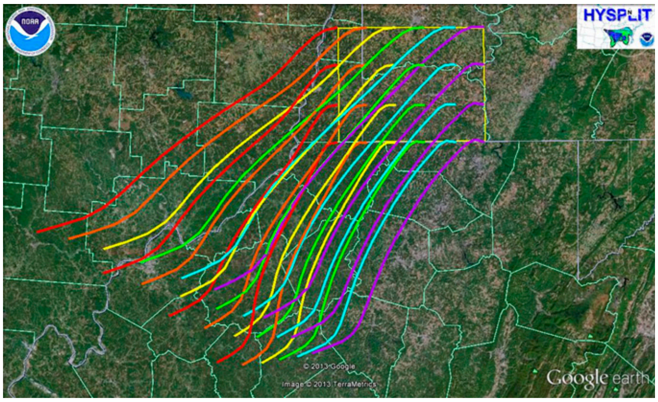

Fig. S1. Eighteen-hour isobaric back trajectory computed by HYSPLIT at 50 m for June 20, 2012.

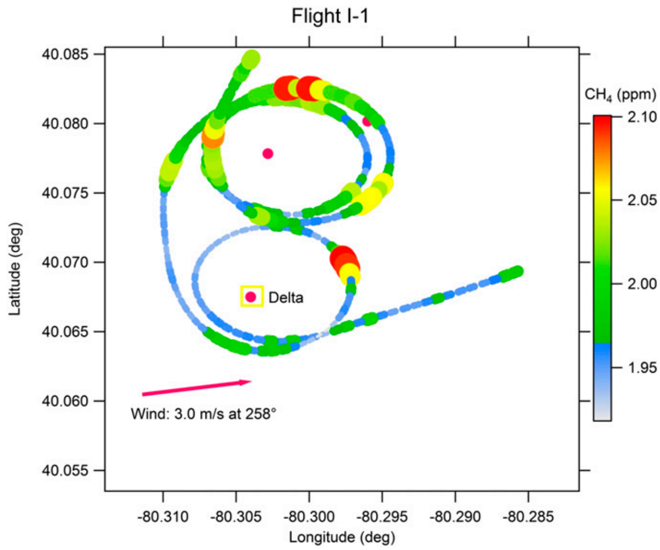

Fig. S2. Flight path between 12:25 and 12:30 EDT around Delta on June 20, 2012, showing a downwind plume of CH_4 and background concentrations directly upwind.

Fig. S3. Aerial photograph of pad Delta showing features of interest that indicate it is in the drilling phase: (a) drill rig, (b) unlit but venting flare stack, (c) air compressors, (d) main high-pressure air line, (e) flow line, (f) separator unit, and (g) water tanks.

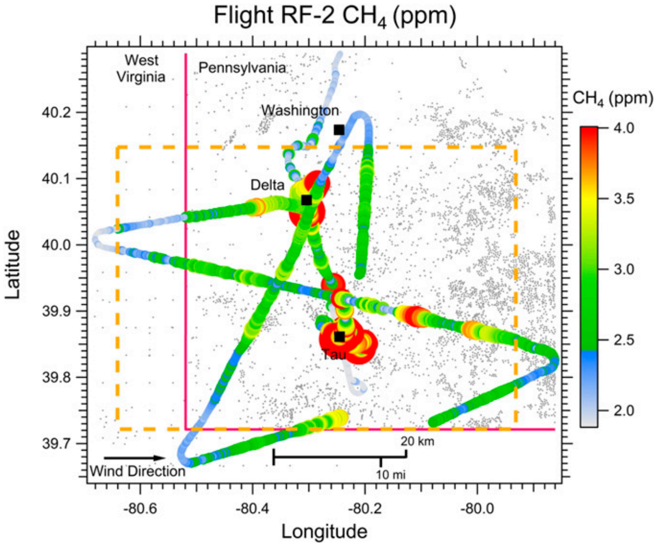

Fig. S4. Regional enhancement of methane at ~240 m above ground level (AGL) on the morning of June 21. The dashed orange box represents the OSA, and the gray dots show well locations.

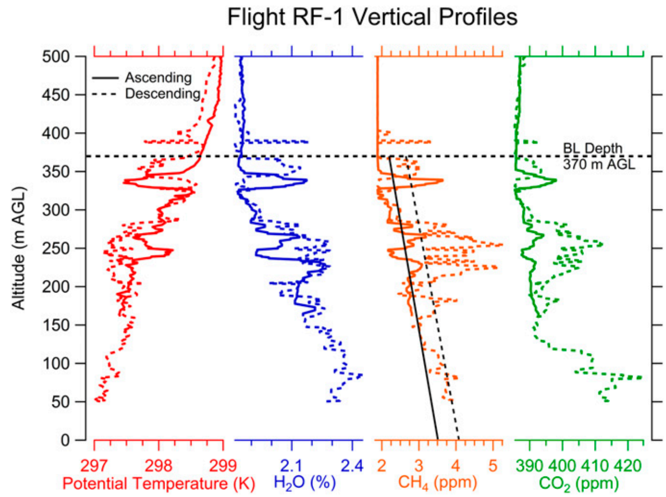

Fig. S5. Vertical profiles of potential temperature, H_2O, CH_4, and CO_2 during flight RF-1 at ~10:00 AM local time. Ascending profiles are denoted by solid lines and descending by dashed lines. The linear extrapolation of CH_4 is denoted by corresponding solid or dashed black lines superimposed on the CH_4 profile.

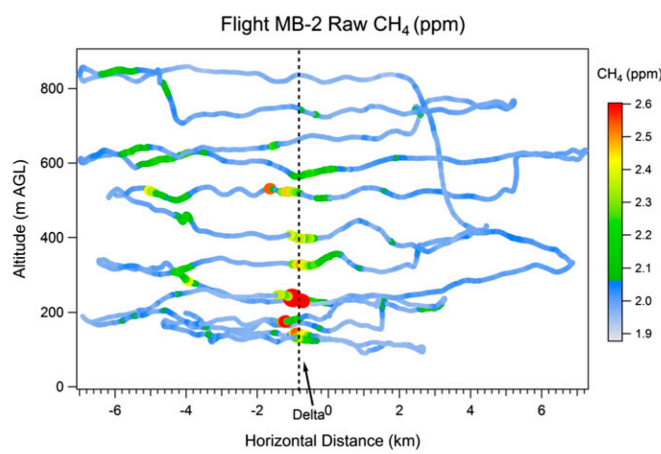

Fig. S6. Raw CH_4 data from flight MB-2 on June 20, 2012 ~1.1 km downwind of pad Delta.

Table S1. Detailed subtotals of emission in the OSA, including expected emissions from local natural gas production and interstate pipeline inputs assuming Howarth et al. (1) fugitive losses, expected emissions from local oil and coal production, and emissions from AFOs

Source (unit)	Production/throughput, unit d^{-1}	Fugitive loss, %		Estimated emissions, g CH_4 s^{-1} km^{-2}	
		Low	High	Low	High
Gas (Gg CH_4)					
Production*	12.31	0.3	1.9	0.15	0.95
Processing†	13.59	0.19	0.19	0.11	0.11
Local Trans/Distr‡	14.60	0.47	0.93	0.28	0.55
Interstate Trans/Distr§	16.30	0.47	0.93	0.31	0.62
Total				0.85	2.23
		Emission factor, m^3 $unit^{-1}$			
Oil (bbl)	31.7	0.8		0.00	
Coal, deep (ton)	127,700	8.4		2.96	
Total				2.96	
Animal		Emission factor, kg CH_4 $count^{-1}$ y^{-1}			
Animal	Animal count	Enteric fermentation	Manure		
Dairy cattle	7,827	65.25	8.95	2.10 E-3	
Other cattle	63,052	54.21	0.92	1.25 E-2	
Pig	8,455	2.43	1.53	1.21 E-4	
Chicken	19,045	0	0.015	1.03 E-6	
Turkey	24,373	0	0.11	9.68 E-6	
Total			1.48 E-2		

*Daily regional nonassociated natural gas production of 22.1 million m^3 converted to mass basis assuming EPA standard condition (1 mole of gas = 23.63 L) and US generic production gas composition (78.8% CH4).

†Daily regional nonassociated natural gas production of 22.1 million m^3 converted to mass basis assuming EPA standard conditions (1 mole of gas = 23.63 L) and US generic natural gas composition for processing (87% CH4).

‡Total daily regional natural gas production (associated and nonassociated) of 22.2 million m^3 converted to mass basis assuming EPA standard conditions (1 mole of gas = 23.63 L) and US generic natural gas composition for pipelines (93.04% CH4). Total pipeline length equals 809.7 km.

§Estimated interstate transports based on 25% of pipeline capacity minus Marshall County WV gas production (associated and nonassociated) accounted for in local production (28.1 million m^3 d^{-1}) converted to mass basis assuming EPA standard conditions (1 mole of gas = 23.63 L) and US generic natural gas composition for pipelines (93.04% CH4). Total pipeline length equals 809.7 km.

Table S2. Detailed subtotals of emission in the UAA including expected emissions from local natural gas production and interstate pipeline inputs assuming Howarth et al. (1) fugitive losses, expected emissions from local oil and coal production, and emissions from AFOs

Source (unit)	Production/throughput, unit d^{-1}	Fugitive loss, % Low	Fugitive loss, % High	Estimated emissions, g CH_4 s^{-1} km^{-2} Low	Estimated emissions, g CH_4 s^{-1} km^{-2} High
Gas (Gg CH_4)					
Production*	20.06	0.3	1.9	0.05	0.30
Processing†	22.15	0.19	0.19	0.03	0.03
Local Trans/Distr‡	23.88	1.48	2.96	0.28	0.58
Interstate Trans/Distr§	34.39	1.48	2.96	0.40	0.81
Total				0.76	1.7
		Emission factor, m^3 $unit^{-1}$			
Oil (bbl)	1,380	0.8		0.00	
Coal, surface (ton)	1,350	2.3		0.00	
Coal, deep (ton)	222,000	8.4		1.00	
Total				1.01	
		Emission factor, kg CH_4 $count^{-1}$ y^{-1}			
Animal	Animal count	Enteric fermentation	Manure		
Dairy cattle	16,044	65.25	8.95	1.47 E-3	
Other cattle	201,536	54.21	0.92	1.38 E-2	
Pig	15,583	2.43	1.53	7.64 E-5	
Chicken	82,730	0	0.015	1.54 E-6	
Turkey	82,625	0	0.11	1.13 E-5	
Total				1.54 E-2	

*Daily regional nonassociated natural gas production of 37.5 million m^3 converted to mass basis assuming EPA standard condition (1 mole of gas = 23.63 L) and US generic production gas composition (78.8% CH_4).
†Daily regional nonassociated natural gas production of 37.5 million m^3 converted to mass basis assuming EPA standard conditions (1 mole of gas = 23.63 L) and US generic natural gas composition for processing (87% CH_4).
‡Total daily regional natural gas production (associated and nonassociated) of 37.5 million m^3 converted to mass basis assuming EPA standard conditions (1 mole of gas = 23.63 L) and US generic natural gas composition for pipelines (93.04% CH_4). Total pipeline length equals 2,571 km.
§Estimated interstate transports based on 25% of pipeline capacity minus Marshall County WV gas production (associated and nonassociated) accounted for in local production (28.1 million m^3 d^{-1}) converted to mass basis assuming EPA standard conditions (1 mole of gas = 23.63 L) and US generic natural gas composition for pipelines (93.04% CH_4). Total pipeline length equals 2,571 km.