# Identification and characterization of high methane-emitting abandoned oil and gas wells

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Recent measurements of methane emissions from abandoned oil/gas wells show that these wells can be a substantial source of methane to the atmosphere, particularly from a small proportion of highemitting wells. However, identifying high emitters remains a challenge. We couple 163 well measurements of methane flow rates; ethane, propane, and n-butane concentrations; isotopes of methane; and noble gas concentrations from 88 wells in Pennsylvania with synthesized data from historical documents, field investigations, and state databases. Using our databases, we (i) improve estimates of the number of abandoned wells in Pennsylvania; (ii) characterize key attributes that accompany high emitters, including depth, type, plugging status, and coal area designation; and (iii) estimate attribute-specific and overall methane emissions from abandoned wells. High emitters are best predicted as unplugged gas wells and plugged/vented gas wells in coal areas and appear to be unrelated to the presence of underground natural gas storage areas or unconventional oil/gas production. Repeat measurements over 2 years show that flow rates of high emitters are sustained through time. Our attribute-based methane emission data and our comprehensive estimate of 470,000–750,000 abandoned wells in Pennsylvania result in estimated state-wide emissions of 0.04–0.07 Mt  $(10^{12}$  g) CH<sub>4</sub> per year. This estimate represents 5–8% of annual anthropogenic methane emissions in Pennsylvania. Our methodology combining new field measurements with data mining of previously unavailable well attributes and numbers of wells can be used to improve methane emission estimates and prioritize cost-effective mitigation strategies for Pennsylvania and beyond.

abandoned wells | oil and gas development | methane emissions | high emitters | climate change

Methane is a potent greenhouse gas (GHG) with a global warming potential 86 times greater than carbon dioxide over a 20-y time horizon (1). A reduction of methane emissions can lead to substantial climate benefits, especially in the short term (2). Recent measurements of methane emissions from abandoned oil and gas wells in Pennsylvania indicate that these wells may be a significant source of methane to the atmosphere (3). Across the United States, the number of abandoned oil/gas wells is estimated to be 3 million or more (4, 5), and this number will continue to increase in the future. As of February of 2016, abandoned oil/gas wells remain outside of GHG emissions inventories, despite evidence that emissions may be substantial nationally. As interest in mitigation of GHG emissions increases, quantifying persistent and large emissions and mitigating them will be increasingly important.

Methane emissions from abandoned wells, as with other fugitive sources in the oil and gas sector, appear to be governed by relatively few high emitters (3, 6–8). It is important for current and future abandoned wells to identify the characteristics that lead to high emissions. This information can provide a rationale for prioritized mitigation.

The century-and-a-half-long history of oil and gas development in Pennsylvania and other US states, such as Texas and California,

has resulted in millions of abandoned wells, and in many cases, poorly documented or missing well records (3, 9–11). As a result, there is a lack of data to characterize abandoned oil and gas wells and the possible relationship between methane emissions and well attributes. Well attributes that may be correlated with methane emissions include depth, plugging status, well type, age, wellbore deviation, geographic location, oil/gas production, and abandonment method (9, 10, 12–14). Previous studies have been limited to wells and attributes with readily available data (12, 14). However, compilation and analysis of historical documents, modern digital databases, and field investigations can be used to infer well attributes of the many wells without data. In this work, we focus on Pennsylvania, which has the longest history of oil and gas development, to determine and explore the role of well attributes, mainly depth, plugging status, well type (e.g., gas or oil), and coal area designation as well as proximity to subsurface-based energy activities, on methane leakage.

Previously estimated numbers of abandoned wells in Pennsylvania range from 300,000 to 500,000 (3, 15) and are based on either incomplete databases or qualitative expert opinion. The Pennsylvania Department of Environmental Protection (DEP) manages oil and gas well data and has records of only 31,676 abandoned oil and gas wells for the state as of October of 2015. Only 5% of the wells in Pennsylvania measured in an earlier study (3) were on the DEP's list. Furthermore, because of changes in governing bodies and regulations over time, the quality of available records is likely to be poorer for older wells (15). To estimate the actual number of wells,

#### **Significance**

Millions of abandoned oil and gas wells exist across the United States and around the world. Our study analyzes historical and new field datasets to quantify the number of abandoned wells in Pennsylvania, individual and cumulative methane emissions, and the attributes that help explain these emissions. We show that (i) methane emissions from abandoned wells persist over multiple years and likely decades, (ii) high emitters appear to be unplugged gas wells and plugged/vented gas wells, as required in coal areas, and (iii) the number of abandoned wells may be as high as 750,000 in Pennsylvania alone. Knowing the attributes of high emitters will lead to cost-effective mitigation strategies that target high methane-emitting wells.

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historical documents and other data sources from oil and gas development need to supplement state records.

Pennsylvania, Ohio, West Virginia, and Kentucky, states through which the Appalachian Basin extends, are among the top 10 US states in terms of the number of inactive and total oil and gas wells (10). Questions remain about potential links between abandoned wells and other active subsurface-based energy activities commonly found in theses states, such as, underground natural gas storage and unconventional oil/gas production (9, 16). For example, could nearby unconventional gas production or underground gas storage reservoirs lead to larger methane leaks from abandoned wells? Previously available measurements and data are insufficient to explore these potential effects. Therefore, we conducted additional field measurement campaigns to fill the data gaps. In the process, we expanded the geographic coverage, previously limited to northwestern Pennsylvania (3), to cover much of the western portion of the state (Fig. 1).

Geochemical information including methane and noble gas isotopes is useful for understanding methane sources (16–18). To evaluate wellbore integrity and design effective mitigation strategies, it is important to identify the source of methane, including whether it is microbial or thermogenic, and if possible, the source formation and migration pathway. It is also important to know as many well attributes as possible and cross-check those attributes with geochemical data when possible. Here, we provide an expanded set of geochemical information including carbon and hydrogen isotopes of methane and concentrations of ethane, propane,  $n$ -butane, and noble gases.

To identify and characterize high methane-emitting abandoned oil/gas wells, we provide in this paper  $(i)$  a database of previously unavailable attributes of measured abandoned wells;  $(ii)$  122 additional field measurements over multiple seasons of methane flow rates and geochemical data, including previously unavailable hydrogen isotopes of methane and noble gas data; (iii) improved estimates of well numbers based on all available data sources; and (iv) an attribute-based methane emissions estimate for abandoned oil and gas wells in Pennsylvania. These data and the associated analysis framework will improve estimates of methane emissions from abandoned oil and gas wells and help develop mitigation strategies across Pennsylvania and beyond.

#### Results

Methane Flow Rates and Well Attributes. Methane flow rates span from below detection (BD) to  $10^5$  mg h<sup>-1</sup> well<sup>-1</sup> for positive methane flow rates (sources of methane to the atmosphere) and from BD to  $-10^1$  mg h<sup>-1</sup> well<sup>-1</sup> for negative methane flow rates (sinks of methane from the atmosphere) (Fig. 2). Most methane flow rates from abandoned wells (90%) are positive, and all negative numbers are small in magnitude.



Fig. 1. The 88 measured abandoned oil and gas wells in Pennsylvania overlaid with conventional oil and gas pools (34), underground natural gas storage fields (34), and workable coal seams within the study area (38).



Fig. 2. Methane flow rates of 88 abandoned wells in Pennsylvania and the coefficient of variation of methane flow rates measured from 2 to 10 times over 2 years (July of 2013 to June of 2015) at 27 wells. If more than one measurement has been made at the given well, the methane flow rates represent an average of all measurements taken. Plugging status is determined based on field observations, and the well type (gas vs. oil or combined oil and gas) is determined using our database-based estimates of well attributes. Methane flow rates below detection (BD) limits (P values  $> 0.2$ ) are shown in the gray portion of the plot between the plots of positive and negative flow rates.

Methane flow rates are measured from different categories of abandoned wells in Pennsylvania. For the measured wells without well records, plugging status is determined based on field observations, and the well type (gas vs. oil or combined oil and gas) is determined based on our estimates of well attributes from our assembled database ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)). Across the dataset, abandoned gas wells, specifically unplugged and plugged/vented wells (Pennsylvania Code, Chapter 78), have the highest observed rates of methane emissions (Fig. 2). Abandoned oil wells have consistently lower emissions compared with abandoned gas wells (Fig. 2). The highest measured methane flow rate is  $3.5 \times 10^5$  mg h<sup>-1</sup> well−<sup>1</sup> at an unplugged gas well in McKean County, and the second highest is 2.9 ×10<sup>5</sup> mg h<sup>-1</sup> well<sup>-1</sup> at a plugged but vented gas well in Clearfield County. Venting of plugged wells is required in coal areas, which in Pennsylvania, include regions where mineable coal seams exist ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)).

Methane flow rates are most strongly related to well type  $(W;$  gas vs. oil or combined oil and gas), plugging status  $(P)$ , and coal area designation  $(C)$  (Table 1 and *SI Appendix*[, Table S3\)](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf). No strong trends are observed between methane flow rates and well depth (d), distance to the nearest unconventional well  $(r_U)$ , or distance to the nearest underground natural gas storage field  $(r<sub>S</sub>)$ . A multilinear fit of d, W, P, C,  $r_U$ , and  $r_S$  to ln  $\dot{m}$ , where  $\dot{m}$  (mg hour<sup>-1</sup> well<sup>-1</sup>) is the methane flow rate, gives an  $R^2$  value of 0.44 and a P value of  $4.4 \times 10^{-8}$ . The P values for the intercept, C, P, and W are below 0.05 and range from  $2 \times 10^{-6}$  (for C) to 0.04 (for the intercept). The P values for  $d$ ,  $r_S$ , and  $r_U$  are high at 0.3, 0.8, and 0.4, respectively. The statistically significant well attributes ( $P$  values  $< 0.05$ ) based on the multilinear regression analysis (Table 1 and *SI Appendix*[, Table S3](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)) are used in methane emissions estimation. The methane emission factors for nine well categories defined by combinations of  $W, P$ , and C range from  $1.2 \times 10^{-2}$  to  $6.0 \times 10^{4}$  mg h<sup>-1</sup> well<sup>-1</sup> (Table 2).

Methane Flow Rates over Time. Repeat measurements of the same abandoned wells conducted 2–10 times (July of 2013 to June of 2015) (SI Appendix[, Table S2](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)) show that high emitters ( $\geq 10^4$  mg h<sup>-1</sup> well<sup>-1</sup>) have relatively low coefficients of variation, with values ranging from 0.04 to 0.3 (Fig. 2). This result implies that high emitters are emitting methane at consistent levels over multiple years. The coefficient of variation decreases with increasing methane flow rates, implying that lower emitters are more likely to be influenced by variable factors, such as seasonal impacts and measurement error. We also find that the coefficient of variation is





These results are for model L6b in [SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf), Table S3. The results of additional models are shown and discussed in [SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf). P values are noted (\* $P < 0.05$ ; \*\*\* $P < 0.001$ ).

unrelated to the number of repeat measurements ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf), [Fig. S2](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)).

Geochemistry. The origin of methane from high-emitting wells is predominantly thermogenic, with  $\delta^{13}$ C-CH<sub>4</sub> values ranging from −33 to −45‰ (Fig. 3). [Thermogenic methane typically has  $\delta^{13}$ C-CH<sub>4</sub> values greater than ~−40 to −50‰, whereas microbial methane typically has  $\delta^{13}$ C-CH<sub>4</sub> values below −50‰ (17, 19, 20); intermediate δ13C-CH4 values, around −50‰, can represent mixed thermogenic and microbial sources.] The ratio of  $C_{2-4}/C_1$  confirms the thermogenic source of high emitters, because the ratio ranges from 0.01 to 0.2. [Microbial sources of methane typically have ratios less than 0.0005 (19, 21).] A larger range in both  $\delta^{13}$ C-CH<sub>4</sub> and  $C_{2-4}/C_1$  values is observed for oil compared with gas wells, with oil wells more likely to emit methane in the microbial range. We do not observe a strong difference in methane isotopes or hydrocarbon ratios between plugged and unplugged wells, although we find that plugged/vented wells have narrower ranges in  $\delta^{13}$ C-CH<sub>4</sub> and C<sub>2−4</sub>/C<sub>1</sub> values. Wells in coal areas tend to have lower  $C_{2-4}/C_1$  ratios, regardless of their plugging status or well type, with ratios ranging from 0.001 to 0.04. For wells (in any area) where both  $\delta^{13}$ C-CH<sub>4</sub> and  $\delta^2$ H-CH<sub>4</sub> are analyzed, most are found to be within the thermogenic range for gases associated with oil reservoirs (17).

High methane-emitting gas wells are found to have the following noble gas ratios:  ${}^{3}\text{He} / {}^{4}\text{He} < 0.10 \text{R}_4$  (where R/R<sub>A</sub> is the

ratio of <sup>3</sup>He to <sup>4</sup>He in a sample compared with the ratio of those isotopes in air, and  $R_A$  nomenclature denotes the <sup>3</sup>He/<sup>4</sup>He ratios of samples with respect to air),  ${}^{4}$ He/<sup>22</sup>Ne > 100, and CH<sub>4</sub>/<sup>36</sup>Ar > 1,000 (Fig. 3 and  $\overline{SI}$  Appendix, Fig. S4). <sup>4</sup>He occurs in very low abundances in the atmosphere and is not produced in association with biogenic methane (22). By comparison, <sup>22</sup>Ne and <sup>36</sup>Ar are ubiquitous, well-mixed, and uniform in the atmosphere. As a result, the noble gases and specifically, elevated levels of <sup>4</sup>He or ratios of thermogenic gases  $(^{4}$ He or CH<sub>4</sub>) to atmospheric gases are able to identify high thermogenic methane-emitting gas wells, which cannot always be achieved with hydrocarbon-based geochemical information alone (23).

Number of Abandoned Wells. Using comprehensive databases (15, 24) and analysis of historical documents (25–28) ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)), we estimate the number of abandoned wells in Pennsylvania to be between 470,000 and 750,000 (SI Appendix[, Table S4\)](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf). The key difference between our well numbers and previous lower estimates is that we include additional wells drilled for enhanced recovery (ER) purposes ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)). Similar to oil and gas wells used for production, injection wells drilled for water flooding, a widely used enhanced oil recovery technique (26, 29), can also act as pathways for methane and other fluid migration. The data show that the inclusion of ER wells leads to an increase in estimated well numbers by multiplicative factors of 1.7–3.5. We base our estimate of ER wells using these factors for years before 1950, for which the number of ER wells is unknown. There also are discrepancies among the numerous data sources available in historical documents and modern digital datasets (Fig. 4). We compare the data sources to estimate the potential degree of error, which is included as multiplicative factors of 1.3–1.5 in the upper bound estimate ([SI](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf) Appendix[, Table S4](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)).

Methane Emission Estimates. The emission factors (Table 2) are combined with the number of wells in each well category in the Pennsylvania DEP database (24) (Fig. 5). The methane emissions contributed by gas wells and wells in coal areas are significantly larger than their share in well numbers. Considering each attribute independently, wells in coal areas represent 21% of the DEP database but 72% of the estimated methane emissions; similarly, gas wells represent 32% of the DEP database but 77% of the methane emissions (Fig. 5). Plugged wells, including those that are vented, represent an estimated 74% of the methane emissions, slightly

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The emission factors are averages of mean methane flow rate measurements per well (mg·hour<sup>−1</sup>·well<sup>−1</sup>). The corresponding numbers of wells and SEs are shown in the next columns. Coal areas are defined here as wells that overlap with one or more workable coal seams. n/a, not applicable.

\*The three highest emission factors are shown.

<sup>†</sup>The measured plugged wells in coal areas are vented as required by regulations.



Fig. 3. Carbon and hydrogen isotopes of methane  $(\delta^{13}C\text{-CH}_4$  and  $\delta^2H\text{-CH}_4$ ), hydrocarbon concentration ratios (C<sub>2−4</sub>/C<sub>1</sub>), noble gas data, and methane flow rate data shown colored by well type, circled by plugging status, and marked with green diamond outlines if in a coal area. For repeat measurements, the average of the data for the well is shown. The regions representing thermogenic methane associated and not associated with oil are from ref. 17.

higher than the number for plugged wells (70%) in the DEP database. The DEP database does not distinguish between plugged wells and plugged/vented wells; both are simply categorized as plugged. In our estimate, plugged/vented wells are those that are both plugged and in coal areas, following regulatory requirements in Pennsylvania. Therefore, the methane emissions for all plugged wells (Fig. 5) represent both a large contribution from high methane-emitting plugged/vented gas wells (in coal areas) and a smaller contribution from low methane-emitting plugged wells that are not vented.

Our attribute-based methane emissions estimates for Pennsylvania using improved well numbers range from 0.04 to 0.07 Mt CH4 per year, which correspond to 5–8% of estimated annual anthropogenic methane emissions for 2011 in Pennsylvania ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)).

#### **Discussion**

Methane Emissions. Well attributes determined for the measured wells in this paper likely remain unavailable for many wells across the United States. Therefore, well attribute estimation studies similar to this analysis may be valuable for many states. For example, West Virginia has at least 57,597 wells that were drilled before 1929 (34% of Pennsylvania wells over the same time period) (25), and records for many wells in the state are likely to be missing. Determining well attributes and numbers is as important as collecting additional measurements for estimating methane emissions. The attributes of high methane-emitting abandoned oil and gas wells identified here as plugging status  $(P)$ , well type  $(W)$ , and coal area designation  $(C)$  may also be indicative of high emitters elsewhere. In the United States, there are 31 oil-producing states, 33 natural gas-producing states, and 25 coal-producing states (30), with many states simultaneously producing oil, natural gas, and coal. Other well attributes, such as age, wellbore deviation, and operator (12), may also be predictors of methane flow rates. However, we do not explore these attributes here because of a lack of data. Efforts to collect and compile additional well attributes are needed to explore the role of attributes not considered in this study.

The total number of abandoned oil and gas wells remains uncertain in Pennsylvania and across the United States. Documented numbers of wells are more likely to represent lower bounds, because they may not include certain types of wells (e.g., injection wells for ER) and may be missing records. For example, the estimate of 3 million abandoned wells across the United States (4) does not include injection wells drilled for ER or undocumented wells. In addition, our upper limit in the number of abandoned wells in Pennsylvania of 750,000 may also be an underestimate because of uncertainties associated with differences in terminology among databases and the accuracy of modern digital databases, even in recent records ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)).

The uncertainties associated with well numbers may be addressed through the application of well-finding technologies (31), field verifications, and database updates. These activities can also help estimate well attributes. In addition, more field measurements of methane emissions are needed from abandoned wells with different attributes and in other geographical locations (i.e., states and countries) to reduce uncertainties in emission factors (32) (Table 2).

Mitigation. Targeting high emitters will lower mitigation costs per unit of methane emissions avoided. The identification of abandoned conventional gas wells and plugged/vented gas wells as the highest emitters allows government agencies to prioritize gas fields and coal areas in their mitigation efforts. Furthermore, explicit categorization of plugged/vented wells, which are found to be high emitters, in state databases may be useful. In addition to database analysis, noble gases, specifically low  ${}^{3}\text{He}^{A}\text{He}$  and high  ${}^{4}\text{He}^{22}\text{Ne}$  ratios, provide an independent approach to identify at  ${}^{4}$ He $/{}^{22}$ Ne ratios, provide an independent approach to identify attributes of high methane-emitting abandoned wells.

Because abandoned wells emit methane continuously, over multiple years and presumably many decades, mitigating their emissions will have a larger apparent benefit when longer time periods are considered. Our multiyear measurements show that the high emitters are likely to emit methane at consistently high levels. Such wells may have been emitting at these levels for many decades and will likely continue for decades into the future. A comparison of the benefits of methane emissions reductions from abandoned wells with reductions from intermittent, short-term sources, such as unconventional oil/gas well development, should be performed using emissions integrated over many years.

Well plugging, which is currently viewed as the main mitigation solution (5, 10), does not guarantee a reduction in methane emissions. Plugging was required originally to protect oil and gas reservoirs, reduce risks of explosions, and more recently, protect groundwater. Plugged wells that are vented, as required by regulations in coal areas in Pennsylvania, are very likely to be high



Fig. 4. Number of drilled and/or completed oil and gas wells in Pennsylvania from various historical documents and databases ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)). The thick black lines represent the 1929–2013 data used to estimate the total number of wells ([SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf), Table S4, second column). For 1859–1928, we use a total well number provided in ref. 25, and the curves shown here are not used to estimate well numbers.

emitters. There are many oil- and gas-producing states with geographically extensive coal layers (e.g., Colorado, Illinois, Indiana, Kentucky, Ohio, Oklahoma, Pennsylvania, West Virginia, and Wyoming). These states have special decommissioning or plugging requirements for coal areas (10). States that require venting in coal areas may want to consider alternatives that ensure safety while reducing methane emissions.

#### Conclusions

High methane-emitting abandoned wells are found to be unplugged gas wells in noncoal areas and plugged but vented gas wells in coal areas, and they seem to be unrelated to the presence of underground natural gas storage areas or unconventional oil/gas production. The identification of these high emitters provides an opportunity to target mitigation efforts and reduce mitigation costs.

Our attribute-based estimate of 5–8% of estimated annual anthropogenic methane emissions in Pennsylvania is higher than previous estimates, which were based on a single emission factor for all wells and a smaller well count (3, 8, 15). The methane flow rates characterized by well attributes may provide insight into potential emissions outside of Pennsylvania in the 33 oil- and gasproducing US states and other oil- and gas-producing countries. Using the analysis framework presented here, scientists and policymakers can better estimate methane emissions and develop costeffective mitigation strategies for the millions of abandoned oil and gas wells across the United States and abroad.

## Materials and Methods

Well Attributes and Numbers. To determine attributes of the measured wells and estimate the number of abandoned oil and gas wells, we combine information from different types of data sources: historical documents, published literature,

field investigations, and state databases. Historical documents include Pennsylvania agency reports (26–28) and books (25, 33). State databases, including geospatial data, were obtained from the Pennsylvania Department of Conservation and Natural Resources (DCNR) (34) and the Pennsylvania DEP (24), agencies that emerged in 1995 from the Pennsylvania Department of Environmental Resources (DER). We combine and analyze the information to estimate attributes of measured wells based mainly on their location with respect to nearby or overlying oil/gas wells, pools, and fields with attributes in the DCNR database. The attributes determined are depth  $(d)$ , coal area designation  $(C)$ , plugging status (P), well type (W), distance to nearest natural gas storage field  $(r<sub>s</sub>)$ , and distance to nearest unconventional oil and gas well  $(r<sub>U</sub>)$ . To estimate the number of abandoned wells, we sum the number of wells drilled annually compiled from multiple sources (15, 24, 25, 27, 28, 33, 35) and subtract the number of active wells from the total (24). We include wells drilled for ER purposes and estimate missing well numbers by scaling available well and production data. We also compare data sources to quantify uncertainties in well numbers. Details on the attribute estimation methodology and the well number estimation are provided in SI Appendix, [SI Materials and Methods](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf).

Field and Laboratory Methods. The measurements of methane flow rates and light hydrocarbon (ethane, propane, and n-butane) concentrations (January, March, and June of 2015 samples) followed methods presented in ref. 3. The measurements were performed across seven counties in Pennsylvania ([SI Ap](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)pendix[, Table S2](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf)). The measurements of methane isotopes were performed at Princeton University (3, 36) and Lawrence Berkeley National Laboratory (LBNL). At LBNL, we also analyzed hydrogen isotopes of methane if concentrations were sufficiently high (∼1,200 ppmv). For October of 2014 and January, March, and June of 2015, we analyzed the samples for the following noble gases, He, Ne, and Ar, at Ohio State University following methods presented in ref. 22. Additional information on the field sampling and the analysis procedures is provided in SI Appendix, [SI Materials and Methods](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf).

Multilinear Regression. We perform a multilinear regression using the following linear model expressed in Wilkinson notation (37):



Fig. 5. Number of wells in the PA DEP database (Left) and the corresponding relative methane emissions distribution (Right) based on plugging status, coal area designation, and well type. Each of three attributes is considered independently.

$$
\ln \dot{m} \sim 1 + d + C + P + W + r_{S} + r_{U}. \tag{1}
$$

Note that the categorical variables, C, P, and W, are denoted using uppercase letters. Multilinear regression is also performed on other linear models, which are summarized in SI Appendix, [SI Materials and Methods](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf).

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Methane Emission Estimates. Based on the multilinear regression results, we use C, P, and W as the key attributes for methane emission estimation:

$$
E_{\text{abandoned wells}} = \sum_{w} \sum_{p} \sum_{c} EF_{w,p,c} \cdot n_{w,p,c}, \tag{2}
$$

where  $E$  is the total methane emissions,  $EF$  is the emission factor,  $n$  is the number of wells, and subscripts  $w$ ,  $p$ , and  $c$  represent the appropriate values of W, P, and C, respectively. We consider two well types ( $w =$  oil or combined oil & gas and gas), two plugging statuses ( $P =$  plugged and unplugged), and two coal area designations ( $c =$  coal and noncoal area). We use the Pennsylvania DEP's wells database (24) and the above attributes to determine the proportion of wells in each category. Additional details, including discussions on uncertainties, are given in [SI Appendix](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf), SI Materials [and Methods](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1605913113/-/DCSupplemental/pnas.1605913113.sapp.pdf).

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# Supporting information

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## **Contents**



## Materials and Methods

Well attribute estimation. For the measured abandoned wells, we used reports and databases from the Pennsylvania Department of Environmental Protection (DEP)[1] and the Pennsylvania Department of Conservation and Natural Resources  $(DCNR)$  [2] to estimate well depths  $(d)$ , coal area designation  $(n_C, C, \text{ or } C_1)$ , plugging status  $(P)$ , and well type  $(W)$ and to calculate the distances to the nearest active unconventional oil/gas well  $(r_U)$  and the nearest underground natural gas storage field  $(r_S)$  (Table S1). We do not consider well attributes such as well age, wellbore deviation, and abandonment method because of a lack of data for these attributes. The previous study that considered the largest number of well attributes used data from Alberta, Canada [3]. In Alberta, the Energy Resources Control Board (ERCB) has required testing, prior to abandonment, for surface casing vent flow (SCVF) across the province and for gas migration (GM) in a test area since 1995 [3]. Using the SCVF and GM results reported by operators and a database of well attributes, the researchers found that geographic area, wellbore deviation, well type, abandonment method, oil price, regulatory changes, and SCVF/GM testing all had a major impact on wellbore leakage [3]. Their study also showed that well age, well-operational mode, completion interval, and  $H_2S$  or  $CO_2$  presence had no apparent impact on wellbore leakage [3]. However, the Alberta analysis explored the role of well attributes on the occurrence of SCVF/GM, not the magnitude of emission rates. In the U.K., elevated methane concentrations in the soil gas around "decommissioned" wells (plugged, capped, and buried) were found to occur within a decade of well decommissioning [4]. Although the U.K. study determined methane emissions rates, the study only considered age and geographic location (at the basin level) and did not include unplugged or not decommissioned wells, which are known to be prevalent in Pennsylvania and across the U.S. [5, 6, 7]. More recently, abandoned well measurements in four active production areas, Wyoming, Colorado, Utah, and Ohio, showed that plugging may be effective at reducing gas leakage [8]. Neither study considered plugged but vented wells.

We estimated  $d$  and  $W$  for the measured wells using the DCNR geospatial database [2] in ArcGIS. The DCNR database included 135,546 wells, 6,878 pools, and 676 fields. (Oil/gas fields consist of one or more oil/gas pools, from which oil/gas is produced.) The well dataset included both active and abandoned oil and gas wells. The pool dataset included the 2-dimensional outlines of geographical production limits for subsurface reservoirs containing oil, gas, or both. The

field dataset included the 2-dimensional geographical boundary outlines of groups of pools related to a single stratigraphic or structural feature. Our analysis framework involved a comparison of field, pool, and well shapefiles to measured well locations using the near distance analysis function in ArcGIS.

For  $W$ , we first compared the types of the nearest/intersecting well, pool and field to the measured well in consideration. If the well/field/pool type (oil, including combined oil and gas, or gas) was consistent across datasets, W was assigned the corresponding type. If the well/field/pool type was not consistent across all three datasets, we considered the distance from the measured well to the nearest well, then the nearest and/or intersecting pool, and finally the nearest and/or intersecting field. If the nearest DCNR well was  $<$ 50 m from the measured well, the DCNR well type was assigned to the measured well. We chose 50 m as the search radius based on our field experience of finding wells using a geographical coordinate system (e.g., GPS). (The accuracy of some DCNR well locations was uncertain because Pennsylvania's permitting requirements did not mandate precise survey location data until the mid-1980s [2].) If the nearest DCNR well was located >50 m from the measured well, the pool type was assigned if the measured well directly intersected the pool. Finally, when none of these approaches was successful, we considered the intersection of measured wells to fields, recognizing that multiple pools could be associated with a field and depths and other properties could vary within a field.

The depths of the nearest/intersecting field, pool, and well were used to assign d for each measured well. The pool depth was defined as the average of the producing formation depth provided in the DCNR database. The field depth was taken to be the depth of the pool with the largest number of wells within the field. The well depth was provided in DCNR's wells database. The depth of the measured well was taken to be the average of the nearest/intersecting well, pool, and field depths, unless: (1) there existed a well within 50 m of the measured well,  $(2)$  the nearest well depth was zero, or  $(3)$ the standard deviation of the three depths (well, pool, and field) was >1000 m. In 29 instances, the nearest well in the DCNR database was  $<50$  m from the measured well. In these cases, the nearest well depth was assigned as the measured well depth. There were 20 nearest wells with depths of zero, presumably unknown, in which case the average depth of the nearest pool and field was used. Finally, if the standard deviation of the three depths (the nearest field, pool, and well depths) were >1000 m, one or two of the following three approaches were taken: (1) if the nearest well was  $>1000$  m away, the well depth was omitted from the average; (2) if the measured well did not intersect a pool, the pool depth was omitted from the average; and (3) if the field displayed highly variable depths, the field depth was omitted.

We determined  $P$  of the measured wells based on the status specified in the DEP database, if available, and surface evidence (e.g., cementing or marker) [5]. The DEP database only indicates whether the wells are "plugged". Therefore, we are unable to differentiate between plugging techniques and other factors that may influence the effectiveness of the plug (e.g., casing integrity), which can vary, especially over time. We assumed wells that are "abandoned" or "orphaned" to be "unplugged". If the measured well could not be identified as matching a well on the DEP record (e.g., by the American Petroleum Institute number), we relied on field-based surface evidence to determine P. "Plugged/vented" wells were classified based on field investigation and were only found in coal areas as required in Pennsylvania. However, not all plugged wells that we determined to be in coal areas through our well attribute analysis were vented.

We assigned coal area designation by comparing well locations to mineable coal seams [9] and wells identified to be in coal areas by the DEP [1]. In Pennsylvania, a well is defined to be in a coal area if the well (1) overlies a mineable coal seam, (2) is  $\leq 1000$  ft (305 m) from the boundary of an area with a current Coal Mining Activity Permit, or (3) is in an area for which an underground coal mine permit application is under review [10]. We defined three different variables for coal area designation:  $n<sub>C</sub>$  represented the number of mineable coals seams that the well intersects, C was an indicator of whether a well intersects any mineable coal seam, and  $C_1$ was an indicator of whether a well intersects any mineable coal seams and/or was <50 m from a well identified to be in a coal area by the DEP. Coal seams were considered mineable if they were greater than 28 inches thick, greater than 100 feet from the surface, and laterally extensive [10]. Historical maps [9] showed coal seams that were located deeper than 100 feet from the surface and laterally extensive. These historical maps were manually digitized into ArcGIS shapefiles for coal seam areas greater than 28 inches thick. There were 13 maps, each showing different mineable coal seams occurring at different depths. We counted the number of coal seams that each measured well intersects based on locational information to obtain  $n_C$ . If  $n_C > 0$ , C was set as "coal" area, while if  $n_C = 0$ , C was set as "non-coal" area. Additionally, the DEP dataset of wells was used as another indicator for estimating whether a measured well was in a coal area. There were 31,676 wells in the DEP dataset and 6,740 wells were identified as coal wells. A near distance analysis was performed to determine how many coal wells in the DEP database were within 50 m of each measured well. If there were one or more DEP-designated coal wells within 50 m of a measured well and/or the  $n_C > 0$ ,  $C_1$  of the measured well was set as "coal" area. There were measured wells with no DEP wells within 50 m and thus, we could not rely on the DEP dataset alone to determine coal area designation.

For  $r<sub>S</sub>$ , we determined the distance from the measured well to the closest point of the nearest underground natural gas storage field provided as shapefiles in the DCNR database [2]. For  $r_U$ , we computed the distance between the measured well and the nearest active unconventional oil/gas well in the DEP database [1].

Field measurements. We employed static chambers to measure methane flow rates at wells and control locations, following the methodology outlined in Ref. [5]. Uncertainties associated with the chamber measurements were discussed in Ref. [5]. In addition, we performed laboratory testing of the chambers at methane flow rates of 1 to 60 g/hr (in the order of high emitters) to study potential errors. We found that the chamber measurements underestimated the methane flow rate by an average of 13%. We adjusted the high emitters' methane flow rates for the final estimates (Figure 4) and the emission factors (Table 2).

Ten sampling campaigns were conducted from July 2013 to June 2015 (Table S2). The later campaigns were designed to broaden geographic coverage compared to earlier measurements and to obtain a representative sample based on well attributes, specifically  $P, C$ , and  $W$  (Table 1). The earlier sampling campaigns (July 2013 to January 2014) emphasized wells that were more easily accessible (e.g., closer to roads). For the later campaigns (March 2014 to June 2015), we prioritized well attributes over ease of access when selecting wells to measure. The objective was to have good estimates of emission factors for well categories identified as high emitters. Regions to focus our measurement efforts were determined using the DEP database and their oil and gas mapping tool [1]. We also repeated measurements at the same wells in McKean, Potter, Venango, Warren, and Lawrence Counties over multiple seasons.

Hydrocarbons. Methane, ethane, propane, and n-butane concentrations in field samples collected in 20 mL pre-evacuated glass vials were analyzed using gas chromatography (GC), as described in Ref. [5]. The methane concentrations were used to determine methane flow rates based on a linear regression of methane concentrations accumulated in a flux chamber over time [5, 11]. Slopes, upon which methane flow rates are based, with p-values greater than  $0.2$  were found to be essentially zero, or below detection level [5]. Ethane, propane, and nbutane concentrations were summed and presented with respect to methane concentrations (Figure 3).

Isotopes of methane. Carbon isotopes of methane for samples collected from March to October 2014 were analyzed using a cavity ring-down spectrometer (CRDS) at Princeton University, following the carbon isotope analysis procedure performed for July 2013 to January 2014 samples reported in Ref. [5]. The samples were collected in 125 mL pre-evacuated Wheaton<sup>™</sup> glass flasks and 2 L SamplePro<sup>™</sup> bags. We used a pure methane standard with  $\delta^{13}$ C of -43.0‰ $\pm$ 0.5‰ determined by Isotope Ratio Mass Spectrometry at the University of Toronto [12]. The standard was diluted to around 15 ppmv with Ultra Zero Air (Airgas, Inc.). The CRDS instrument determined the peak absorbance ratio of  $^{13}CH_4$  over  $^{12}CH_4$ for the standard methane and the samples. The pressure and temperature were maintained within  $\pm 0.1$  torr and  $\pm 0.02$ °C respectively. The average time for each analysis was approximately 20 minutes, corresponding to the optimal integration time of the instrument. Between each sample, we flushed the sampling cavity with Ultra Zero Air and evacuated it three times. For ambient air methane concentrations, the precision of the  $\delta^{13}$ C was estimated as  $\pm 2.0\%$  (1 $\sigma$ ) [12] based on the standard deviation of absorbance ratio of  $^{13}CH_4$  over  $^{12}CH_4$ .

Carbon and hydrogen isotopes of methane for samples collected in 125 mL Wheaton™ glass flasks in January, March, and June 2015 were analyzed at Lawrence Berkeley National Laboratory. For CH<sub>4</sub> concentrations lower than  $\sim$ 1200 ppmv, carbon isotope ratios of CH<sup>4</sup> were measured by flushing 160 mL serum bottles into a Tracegas™ pre-concentrator interfaced with a Micromass JA Series Isoprime isotope ratio mass spectrometer (Micromass, Manchester, UK). Repeated injections of laboratory standards associated with sample analysis yielded a standard error  $\pm 0.4\%$  (1 $\sigma$ ; n=9). For CH<sub>4</sub> concentrations larger than ∼1200 ppmv, hydrogen and carbon isotope ratios of CH<sup>4</sup> were determined separately using a gas chromatograph and an isotope ratio mass spectrometer interfaced with a pyrolysis reactor (GC-P-IRMS) and a combustion GC-C-IRMS, (Trace™ GC Ultra-Isolink™-DeltaV™ Plus system, Thermo Fisher Scientific, Bremen, Germany). CH<sup>4</sup> was separated chromatographically on an HP-molesieve fused silica capillary column (30 m x 0.320 mm). For hydrogen isotopes, after GC separation, CH<sup>4</sup> was pyrolyzed in an empty ceramic tube at 1450°C and the hydrogen isotope ratios were measured using the IRMS. For carbon isotopes, after GC separation, the CH<sub>4</sub> was combusted to  $CO<sub>2</sub>$  at 1000°C in a capillary ceramic tube and the carbon isotope ratio was measured in the IRMS. Repeated injections of  $CH<sub>4</sub>$  laboratory standard yielded a standard error of  $\pm 0.4\%$  (1 $\sigma$ ; n=10) for  $\delta^{13}$ C and  $\pm 2.6\%$  (1 $\sigma$ ; n=14) for  $\delta^2$ H.

Carbon isotope ratios were reported in the conventional δ-notation relative to VPDB scale and hydrogen isotopes relative to SMOW.

Noble gases. Gas samples for noble gas analyses were collected in ∼8" (inch) long, 1/4" diameter refrigeration-grade copper tubing. Before sampling, the copper tubes were flushed inline with at least 50 volumes of sample gas prior to sealing by manually pumping on the downstream side of the copper tube with a 3-way Luer Lock syringe. After purging, samples were sealed with either a 30/1000 of an inch gap stainless steel refrigeration clamp or CHA Industries (Fremont, CA) cold weld refrigeration crimper [13, 14].

In the laboratory, the gases were extracted from the copper tube on an ultra-high vacuum line  $( $2x10^{-9}$  torr)$  with an MKS 0-20 torr and 0-1000 torr capacitance manometers that are calibrated daily using a water-vapor free, inter-laboratory validated air standard collected on the shore of Lake Erie. The sample was collected at 12.1°C at a latitude of 41.5931 N. The laboratory standard was cross-validated between two independent laboratories (USGS and U.Rochester). Total gas pressure was measured and an aliquot  $\left( \langle 10\% \rangle \right)$  of total gas volume was pipetted into an SRS quadrupole mass spectrometer (MS) and an SRI GC, fitted with a flame ionization detector (FID) and thermal conductivity detector (TCD), for scanning and measurement of major gas and trace gas components (e.g.,  $N_2$ , CH<sub>4</sub>, O<sub>2</sub>, H<sub>2</sub>, Ar, CO<sub>2</sub>, C<sub>2</sub>H<sub>6</sub>) [15, 16]. All samples were analyzed in triplicate. Standard analytical errors were all less than  $\pm 3.41\%$  for major gas concentrations above the detection limit. The average external precision of "known-unknown" standards were reported in the parentheses  $(CH_4 \ (2.14\%).$  $C_2H_6$  (2.78%), N<sub>2</sub> (1.25%), CO (3.41%), CO<sub>2</sub> (1.06%), H<sub>2</sub>  $(3.37\%),$   $O_2$   $(1.39\%),$  Ar  $(0.59\%)$ . These values were determined by measuring referenced and cross-validated laboratory standards including an atmospheric standard (Lake Erie Air) and a series of synthetic natural gas standards obtained from Praxair.

The isotopic analyses of noble gases were performed using a Thermo Fisher Helix SFT Noble Gas MS at The Ohio State University Noble Gas Laboratory following methods reported previously [15, 16, 17]. The average external precision based on "known-unknown" standards were all less than  $\pm 1.23\%$ for noble gas concentrations with values reported in parentheses ( $[{}^4\text{He}]$  (0.89%),  $[{}^{22}\text{Ne}]$  (1.23%), and  $[{}^{40}\text{Ar}]$  (0.49%)). These values were determined by measuring referenced and cross-validated laboratory standards including an established atmospheric standard (Lake Erie Air) and a series of synthetic natural gas standards obtained from Praxair. Noble gas isotopic standard errors were approximately  $\pm 0.0095$  times the ratio of air (or 1.27 x10<sup>-8</sup>) for <sup>3</sup>He/<sup>4</sup>He ratio,  $< \pm 0.681\%$  and  $< \pm 0.814\%$  for <sup>20</sup>Ne/<sup>22</sup>Ne and <sup>21</sup>Ne/<sup>22</sup>Ne, respectively, and less than  $\pm 0.916\%$  and 0.574% for <sup>38</sup>Ar/<sup>36</sup>Ar and <sup>40</sup>Ar/<sup>36</sup>Ar, respectively (higher than typical because of interferences from  $C_3H_8$  on mass=36 and 38).

Multilinear regression. We used multilinear regression to identify attributes that may predict methane flow rates  $(m)$ . Multilinear regression was performed using the fitlm function in Matlab. We considered numerous multilinear models but presented the results of six different models: L6a, L6b, L6c, L3, N6b, and N3. Model names have as their first letter an "L" if  $\ln \dot{m}$  was used or "N" if  $\dot{m}$  was used. The number indicates the number of variables in the multilinear regression model, where "6" means that six attribute variables are included. The last

letter differentiates among models with the same number of variables and  $\dot{m}$  representation.

Number of wells.We estimated the total number of wells drilled throughout the drilling history of Pennsylvania using all available data sources to obtain a range in possible well numbers. Because our goal was to determine the number of abandoned wells that may contribute to methane emissions, we considered all oil and gas wells drilled, including dry wells and those that were drilled for enhanced recovery. (Dry wells are wells incapable of producing enough oil or gas to justify completion. To enhance the recovery of oil, additional wells are drilled to inject fluids into the oil/gas bearing formation.) We did not include storage, test, observation, and waste disposal wells due to data limitations for years prior to 1957. For years after 1957, we assumed that these other well types are included in the modern digital records [18].

We compiled all available well numbers from data-based sources such as the published scientific literature [18], state databases [1], annual state reports [19, 20, 21, 22], and historical references detailing annual drilling and production [23, 24]. We did not include bulk estimates from references that did not provide estimation methods or annual data. For years for which reliable data were not available, we used relationships between oil production and the number of wells in the preceding or following 10-11 years. Historical oil production data were obtained from the 1993 Pennsylvania Department of Environmental Resources (DER) Progress Report [22].

Compiled annual well numbers were identified as including or excluding enhanced recovery wells. For years with well numbers that exclude enhanced recovery wells, we applied a factor based on the ratio of well numbers including and excluding enhanced recovery wells from other years to obtain our final estimate of well numbers.

To estimate the total number of abandoned wells, including those that have been orphaned and/or lost, we took the total number of wells drilled and subtracted from that total the number of active and inactive oil/gas wells in Pennsylvania as of March 1, 2014, which is 10,921. Our total number of abandoned wells represents wells drilled and abandoned, with and without plugging, from 1859 to 2013.

Methane emissions.We estimated the state-wide methane emissions from abandoned oil and gas wells in Pennsylvania using an attribute-based approach and our estimates of well numbers. We used the DEP database [1] to determine the proportion of wells with all combinations of  $W, C$ , and P (Table 2) and then scaled the numbers by the total estimated number of abandoned wells. There were 12 well types (including "undetermined"), which we aggregated into "oil", "gas", "other", and "undetermined". Our "oil" wells designation included "oil" wells and "combined oil & gas" wells. Our "gas" wells designation included "gas" wells and "coalbed methane" wells. "Other" wells were "injection", "dry holes", "storage", "observation", "multiple well bore type", "waste disposal", and "test" wells, to which we assigned the general average emission factor (Table 2). For "undetermined" wells and wells not assigned a type, we applied a weighted emission factor based on the distribution of well types in the DEP database [1]. For coal area designation and plugging status, we used the emission factors in Table 2 directly.

# Measurement Data and Well Attributes

 $R^2$  of methane flow rates at wells. Methane flow rates from abandoned oil and gas wells,  $\dot{m}$  (mg hr<sup>-1</sup> well<sup>-1</sup>), were based on linear fits of methane concentration accumulation over time [5]. The  $\mathbb{R}^2$  values for these linear fits showed that improved fits were more easily achieved for higher flow rates (Figure S1). Most methane flow rates had  $R^2$  values greater than 0.8. The distribution in  $R^2$  values remained similar for measurements conducted in all three years: 2013, 2014, and 2015.

Variation in methane flow rates at wells. Variances (and the coefficient of variation) in methane flow rates at wells were not related to the number of measurements at a given well site (Figure S2). Therefore, the relationship between variances and methane flow rates discussed in the main text (Figure 2) were not affected by the number of repeat measurements.

Methane flow rates at control locations. Methane flow rates at control locations (Figure S3) were generally orders of magnitude lower than those found at wells, similar to observations reported in Ref. [5]. Although methane flow rates could be similar to control flow rates at the lower end, control flow rates did not go above  $100 \text{ mg hr}^{-1}$  location<sup>-1</sup> while methane flow rates at abandoned wells frequently exceeded 100 mg  $hr^{-1}$  well<sup>-1</sup> and reached as high as  $10^5$  mg  $hr^{-1}$  well<sup>-1</sup>. The mean flow rate at controls was 0.6 mg hr<sup>-1</sup> location<sup>-1</sup>. Of the 193 control location measurements, 91 measurements (47%) showed methane emissions below detection level (i.e., no methane accumulation or update based on the p-value). There was a similar number of negative methane flow rates (i.e., methane sinks) (53 measurements or 27% of the control measurements) as positive methane flow rates (i.e., methane emissions to the atmosphere) (49 measurements or 25% of the control measurements).

Noble gas measurements at wells. The noble gas (e.g.,  ${}^{4}$ He) and methane concentrations were evaluated with  $22Ne$  and <sup>36</sup>Ar as environmental tracers of air and/or air-saturated water (Figures 3 and S4). High methane-emitting gas wells had  ${}^{3}\text{He}/{}^{4}\text{He}$  < 0.10R<sub>A</sub> (where R/R<sub>A</sub> is the ratio of <sup>3</sup>He to <sup>4</sup>He in a sample compared to the ratio of those isotopes in air and  $R_A$  nomenclature denotes the  ${}^3\textrm{He}/{}^4\textrm{He}$  ratios of samples with respect to air),  ${}^{4}\text{He}/{}^{22}\text{Ne}$ >100,  $|{}^{4}\text{He}|$ >10<sup>-4</sup> ccSTP/cc, and  $CH_4^{36}\text{Ar} > 5 \times 10^3$ . Selecting ratios of key gas parameters to  $22$ <sup>N</sup>e and  $36$ Ar provided a comparison between deep, thermogenic, crustal sources (<sup>4</sup>He and CH<sub>4</sub>) vs. atmospheric/airsaturated water  $(^{22}$ Ne and  $^{36}$ Ar). Because the atmosphere is extremely well mixed, <sup>22</sup>Ne and <sup>36</sup>Ar in air and air-saturated water are ubiquitous on the Earth's surface and in meteoric water.  $^{22}$ Ne and  $^{36}$ Ar are nearly constant in air and in meteoric water, and are very well constrained. As a result, minor variations in the <sup>4</sup>He/<sup>22</sup>Ne or CH<sub>4</sub>/<sup>36</sup>Ar ratios readily record minor contributions from deep crustal fluids at the surface. These ratios are specifically sensitive to gas leakage from natural gas wells in Pennsylvania, which contain relatively dry natural gas (i.e., low  $C_{2-4}/C_1$ ). In hydrocarbon fluids that have achieved lower relative thermal maturities throughout their geological histories (i.e., fluids that contain more wet  $(C_2 C_4$ ) gases and/or oil-associated gases), there is typically lower gas-water ratios (e.g.,  ${}^{4}$ He/<sup>22</sup>Ne,  ${}^{4}$ He/<sup>36</sup>Ar, or CH<sub>4</sub>/<sup>36</sup>Ar) [13, 14, 15]. Hence, wet and oil-associated methane typically has progressively lower  ${}^{4}$ He/<sup>22</sup>Ne, CH<sub>4</sub>/<sup>36</sup>Ar, or  ${}^{4}$ He/<sup>36</sup>Ar than non-oil associated dry natural gas. For these reasons, the <sup>4</sup>He/<sup>22</sup>Ne, CH<sub>4</sub>/<sup>36</sup>Ar, or <sup>4</sup>He/<sup>36</sup>Ar was relatively high in natural gas wells as opposed to oil-associated gases. The relative increases in <sup>4</sup>He $\tilde{}/^{22}$ Ne (and CH<sub>4</sub> $/$ <sup>36</sup>Ar) effectively served as a proxy for the original thermal maturity at which hydrocarbon gases were produced in the source rocks [13, 14, 15]. Therefore,  ${}^{3}\text{He}/{}^{4}\text{He}$ ,  ${}^{4}\text{He}/{}^{22}\text{Ne}$ , and CH<sub>4</sub> $/{}^{36}\text{Ar}$  were able to differentiate between gas and oil or combined oil & gas wells.

Noble gases did not differentiate between plugging statuses or coal area designation.

The impact of  $d$ ,  $n_C$ ,  $r_U$ , and  $r_S$  on methane flow rates. Here, we evaluated the role of d,  $n_C$ ,  $r_U$ , and  $r_S$  on  $\dot{m}$  (Figure S5), which were not presented in detail in the main text. Visually, there appeared to be a relationship between d and  $\dot{m}$ . However, much of the dependence of  $\dot{m}$  on d could be explained by the dependence of  $\dot{m}$  on  $W$ , as  $\dot{m} > 10^3$  mg  $hr^{-1}$  well<sup>-1</sup> were dominated by gas wells. Gas wells were drilled to both shallow and deep depths, whereas oil wells tended to be relatively shallow. For wells in coal areas,  $\dot{m}$  appeared to increase with the number of intersecting workable coal seams  $(n<sub>C</sub>)$ . However, several high emitters had no intersecting workable coal seams, suggesting that other factors were also important. Finally, high methane-emitting wells were up to ∼20 km away from the nearest active unconventional well and the nearest underground natural gas storage field, more than the average distance of all wells (18 km for storage fields and 8 km for unconventional wells).

Multilinear regression. Multilinear regression analysis of six different models showed that Model L6b was the best predictor of  $\dot{m}$  (Table S3). However, the p-values of the coefficients for d,  $r<sub>S</sub>$ , and  $r<sub>U</sub>$  were above 0.05 and these terms did not play a significant role in determining  $\dot{m}$ . In fact, Model L3 was able to provide a similar fit to Model L6b. The two models, N6b and N3, that were not based on the logarithmic values of  $\dot{m}$ performed significantly worse than the models based on  $\ln \dot{m}$ .

For upscaling methane emissions, it is important to get the correct order of magnitude of the methane flow rates of high emitters. None of the models was able to reproduce  $\dot{m}$  at the order of magnitude level (Figure S6). Therefore, we did not use the models to estimate methane emissions.

The multilinear regression analysis showed that  $W, C$ , and P were the best predictors of  $\dot{m}$ . For coal area designation, we used C instead of  $n<sub>C</sub>$  and  $C<sub>1</sub>$ , both of which had p-values greater than 0.05 (Figure S6).

# Estimate of Well Numbers

A brief history of oil and gas development in Pennsylvania. Pennsylvania has the longest history of oil and gas production in the U.S. The first commercial oil well in the U.S., the "Drake Well", was drilled in 1859 in Titusville, Pennsylvania. In 1881, the Bradford Oil Field in northwest Pennsylvania produced 83% of America's oil output [25]. After production in the Bradford Oil Field peaked in 1881, the field continued to produce significant quantities of oil using enhanced, or "secondary", recovery methods, mainly involving waterflooding [26].

Waterflooding involves drilling additional wells to inject water in oil formations and increase the flow of oil to producing wells [27]. It differs from primary oil production that uses natural pressures or pumping without any injection wells. These enhanced recovery (ER) wells, which are not producing oil or gas wells, can also act as conduits for subsurface fluid migration and gas emissions at the surface.

The large potential number of ER wells and the lack of historical reporting of these wells make them both important and challenging to quantify. The first use of water flooding occurred in 1880 in the Venango Oil Field in Pennsylvania [28]. Water flooding was illegal in Pennsylvania until 1921 [26] and water flooding wells drilled prior to 1921 were unlikely to have been reported or recorded. Even after 1921, ER wells may not have be considered as oil and gas wells and we could not find records of injection wells until 1950, when Pennsylvania's Department of Environmental Resources (DER) began publishing progress reports for oil and gas development in the state [19]. The "five-spot" method, a popular water flooding technique developed in 1927, involves drilling four additional injection wells for each producing oil well [26, 28]. Another popular method used was the "seven-spot" pattern with six injection wells per producing well [26]. Because producing and injection wells are likely to be in a grid pattern, there will likely be one additional row of injection wells per row of producing wells for the five-spot method. Therefore, inclusion of injection wells will increase the estimated number of wells by a factor of at least two.

Substantial secondary oil production, mainly through waterflooding, occurred in Pennsylvania in the 1930s and 1940s [22]. Oil production and enhanced oil recovery in Pennsylvania decreased steadily after the 1930s/1940s peak [22].

Natural gas production in Pennsylvania began in the late 1800s. Between 1882 and 1928, Pennsylvania's natural gas production was the second highest in the Appalachian Basin, after West Virginia [24]. In the early 1950s, discoveries of conventional natural gas reserves in deep gas fields led to growth in natural gas production [21]. More recently, Pennsylvania has experienced significant growth in natural gas production attributable to horizontal drilling and hydraulic fracturing of shale formations (e.g., the Marcellus formation) and has been the focus of scientific and media attention from both economic and environmental standpoints [6, 29].

Compilation of data sources. Information on the number of wells drilled from 1859 to 2013 requires compilation of different data sources, each covering different time periods (Table S4 and Figure 5). For 1859-1929, we obtained numbers of wells drilled from two historical books on oil and gas production [23, 24]. For 1930-1949, we estimated well numbers based on oil production and trends in preceding and following years. For the 1950-1991 time period, the Pennsylvania DER published annual to bi-annual progress reports on oil and gas development [19, 20, 21, 22]. (The discontinuation of these reports is likely due to the split of DER into the DCNR and the DEP in 1995.) The DCNR now manages a digital database known as the Pennsylvania Internet Record Imaging System/Well Information System (PA\*IRIS/WIS), which is being modernized and renamed as EDWIN (Exploration and Development Well Information Network). This database was used to estimate well numbers in Ref. [18]. The PA\*IRIS/WIS and EDWIN database of wells were different from the DCNR wells dataset [2] used in our attribute estimation framework. The DEP also maintains data on wells drilled, which was publicly available on their website [1]. In addition, since 2009, the number of wells drilled as reported by operators to the DEP was also available [30].

The total number of wells drilled in Pennsylvania from the start of drilling in 1859 until 1928 was reported to be 168,190 [24], which corresponded to an average of 2403 wells per year. Figure 5 showed the number of wells drilled per year from 1889 to 1920 in Pennsylvania [23] and from 1859 to 1928 in the Pennsylvania and New York (NY) portion of the Appalachian Basin [24]. During this time period, 89% of the total production was in Pennsylvania with the New York portion of the Appalachian Basin becoming only marginally significant in 1876 [24]. The two historical sources [23, 24] show that the first peak in well drilling occurred in the 1890s when approximately 6000 to 7000 wells were drilled annually. There was no mention of ER well counts in either of these historical references.

We estimated the number of wells drilled from 1929 to 1949, excluding ER wells drilled before 1938, to be 68,000.

For 1929, we estimated a well number of 3600, which was determined by scaling the combined NY and PA well number of 4009 by 0.89 (see previous paragraph). For most years from 1930 to 1976, well numbers were available from Oil Weekly annual activity reports [18], Minerals Yearbooks by the U.S. Geological Survey [18], and PA DER Progress Reports [19, 20, 21, 22]. No data for all of Pennsylvania were available for 1931, 1932, 1936, 1947, 1950, and 1953 and previous estimates used linear interpolation of wells numbers from the preceding and following years [18]. The well numbers given in Ref. [18] for 1930 to 1949 appeared to be an underestimate since the numbers conflict with other data sources and trends. For one, the well numbers in Ref. [18] for all of Pennsylvania were lower than those reported for the Bradford Field alone [26]. In addition, there was a drop in well numbers in Ref. [18] in a period of increasing production (1930-1937). Analysis of well numbers and oil production data from other time periods showed that well numbers generally increased with production (Figure S7). Data from 25 water flooding projects in northern Pennsylvania that began between 1927 and 1946 showed that an additional 1.5 "water-intake" wells were drilled for every new producing well drilled [26]. Therefore, the low numbers in Ref. [18] could not be explained by assuming that most of the additional wells were injection wells, not producing wells. Because the well numbers given in Ref. [18] appeared to be incomplete and inconsistent with our understanding of the oil and gas history in the region, we used oil production data to estimate well numbers across Pennsylvania for 1930 to 1949.

The oil production-well numbers relationship was dependent on whether production was increasing or decreasing. Oil production increased significantly from less than 8 million barrels in 1920 to 19 million barrels in 1937 (Figure 5) [22]. In the years preceding 1930, we saw a linear relationship between oil production and well numbers  $(R^2 = 0.63)$  as oil production increased (Figure S7). Using this relationship, the number of wells drilled in 1930 to 1937 was estimated to be 32,000. Oil production began to decrease in 1938. Data from the Pennsylvania DER Progress Reports also showed a linear relationship between oil production and well numbers  $(R^2 = 0.81)$  for 1950 to 1959, a period of decreasing oil production. The number of wells for 1938-1949 using this second relationship was 32,000.

The DCNR's PA\*IRIS/WIS data produced a number for well completions of 114,154 for 1957 to 2012 [18] (Table S4). The DCNR well number, which includes ER wells, was assumed to be accurate because previous research found the database to be the most "complete and internally consistent digital data record of documented wells in Pennsylvania" [18].

To consider the role of ER wells, we reviewed well numbers from the Pennsylvania DER Progress Reports and the DEP database. The DEP database contained 1738 "injection" wells, which represented 5% of the wells on the database. In contrast, the total number of wells drilled from 1950 to 1991 based on the Pennsylvania DER Progress Reports was 55,516 excluding ER wells and 65,286 including ER wells, which corresponded to 18% of the wells being ER wells. The annual DER well numbers including and excluding ER wells showed that the relative proportion of wells drilled for ER was the highest in the early 1950s. In 1951, the inclusion of ER wells increased the number of wells by a factor of 4 (Producing & Injection Wells / Producing Well). Using the 1950 to 1952 data, we calculated ratios of total wells including ER wells to producing wells (excluding ER wells) and obtained an average of 3.5. These ratios were in line with data from 25 water flooding projects in the Bradford Field, which have factors ranging from 1.7 to 3.3 with a mean of 2.5 [26]. We used the total well numbers including both oil and gas for these ratios because we did not have a reliable breakdown of oil and gas wells for the 1930-1937 time period.

Drilled well numbers were also available on the DEP's website beginning from 1940 (Figure 5). However, the DEP's well numbers prior to 1956 ranged from 1 to 22. These low numbers were inconsistent with our understanding of oil and gas development in Pennsylvania and we did not use the DEP numbers in our estimates. We used the DEP numbers for 2013 only because Ref. [18] did not provide well numbers after 2012.

For most years with data available, the DER and DEP numbers were underestimates when compared to the DCNR numbers (Figure 5). Nonetheless, the general trends in all three sources were similar. In all three data sources, there was a peak in the number of wells of up to 6500 wells drilled per year in the 1980s, which was followed by a steep decline to ∼1000-2000 wells drilled per year by the 1990s. The DEP and DCNR numbers also showed a 2007 peak in the number of wells drilled in Pennsylvania of ∼5000-6200 wells drilled per year. By 2012, the number of wells drilled per year dropped to ∼2000-3000.

Uncertainties. Modern digital records managed by state agencies are known to have poor records of wells drilled before 1957 [18]. We compared the DCNR numbers, which were assumed to be correct for 1957 onwards [18], to two historical data sources: the Pennsylvania DER Progress reports and the Minerals Yearbook. Considering data from 1957 to 1976, we found that the DCNR numbers were 1.3 times larger than the numbers from the PA DER Progress reports and 1.6 times larger than the number from the Minerals Yearbook. For years that we used historical data sources (i.e., the DER Progress Reports or Ref. [24]), we conservatively used a factor of 1.3 to account for underestimation due to missing data and other uncertainties for all years before 1957 (Table S4). Overall, we estimated ∼150,000 wells as unaccounted for due to underreporting and lack of documentation (Table S4).

A comparison of the annual drilled well numbers showed that there are discrepancies between data sources even for recent years. For 1992-2012, the DCNR numbers were on average 1.5 times larger than the DEP numbers. For 2009-2013, the number of wells reported by operators and the number of wells on the DEP's website were similar but not equal. Therefore, for 2013, a year for which there are no DCNR numbers, we used a factor of 1.5 times the DEP number to estimate the upper limit in the well number.

Another major source of uncertainty was the inconsistency in terminology. Well numbers given in Ref. [18] including oil, gas, and dry wells were stated to be the number of well completions. However, dry wells are typically not completed. It was unclear if the wrong terminology was used or if they represented some subset of dry wells that were completed. Here, we assumed the former and that all dry wells, both completed and not completed, were included in the well numbers given in Ref. [18]. However, this assumption may lead to an underestimate in our 1957-2012 well numbers.

Although we used data-based methods where possible, uncertainty remains in extrapolating data from different time periods to periods without data. This applied both to factors used to represent ER wells and underreporting.

Well numbers for years prior to 1957 may not have included oil/gas wells such as observation and test wells. Available historical data sources specifically stated numbers for oil, gas, and dry wells and made no mention of other oil/gas well types [24].

Total number of wells. Based on the trends and the history of oil and gas in Pennsylvania, we assumed that ER via waterflooding played a significant role in the years prior to the 1950s. We scaled the well numbers for 1859 to 1929 by 1.5 or 2.0 and the well numbers from 1930 to 1937 by 2.0 or 3.5 (Table S4). Given the uncertainty in the number of potential ER wells before 1921, we reduced the factor of 2.0 applicable for the five-spot method to a factor of 1.5 for 1859-1928 to obtain a lower bound estimate. The bulk of the wells in Pennsylvania were drilled after 1875 (Figure 5), shortly before the first ER wells were likely to be drilled. Therefore, a factor of 2.0 may also be reasonable for 1859-1928, and this factor was used as the upper bound estimate for well numbers including ER wells. For the upper bound number for 1930-1937, we used a factor of 3.5 determined using the average of well numbers from the Pennsylvania DER annual reports for 1950- 1952, when ER activities were more likely to resemble earlier periods. From 1938 onwards, we directly used the well numbers based on the DER progress reports or the DCNR well numbers [18] as they included ER wells. The total number of ER wells that were previously unaccounted for was estimated to be 110,000 to 250,000.

Adding ER wells and accounting for potential underreporting, we calculated the number of abandoned wells to be 470,000 to 750,000 for the state of Pennsylvania (Table S4). These numbers also included dry wells for all years and other well types (e.g., observation and test wells) for 1957 onwards.

Number of wells by attribute. It is important to estimate not only the total well numbers but also the numbers by attribute, especially for the three factors identified to be important predictors of high methane-emitting wells (well type, plugging status, and coal area designation).

We compiled historical data from various sources to estimate the proportion of wells that were oil and gas (Table S5). Considering only oil and gas wells (not dry, test, or other wells) to determine proportions, we found that oil wells may represent 65% to 76% of abandoned wells; while gas wells may represent 24% to 35% of abandoned wells. In contrast, the DEP database showed a breakdown of 50% oil and 50% gas wells [1].

Unfortunately, historical data to estimate the number of wells by plugging status or coal area designation were not available. In the DEP database, 70% of abandoned wells were plugged, leaving 30% unplugged [1]. Based on the long history of oil/gas development in Pennsylvania and poor historical records, the actual number of unplugged wells was likely to be higher. As for coal area designation, the DEP database showed that 21% of wells were in coal areas. Of these wells in coal areas, 86% were plugged and could be assumed to be plugged/vented.

# Methane Emission Estimates

Methane emissions from abandoned oil and gas wells was estimated to be 0.040 to 0.066 Mt  $(10^{12} \text{ g})$  CH<sub>4</sub> per year in Pennsylvania using well numbers of 470,000 to 750,000 (Table S4). These emissions represented 5% to 8% of total anthropogenic methane emissions for Pennsylvania in 2011, which was estimated by the World Resources Institute (WRI) [31] to be 15.26 Mt  $CO<sub>2</sub>e$  per year (0.73 Mt CH<sub>4</sub> per year). WRI used a global warming potential (GWP) of 21 following the second assessment report of the Intergovernmental Panel on Climate Change and used the State Inventory Tool (SIT) of the U.S. Environmental Protection Agency (EPA) [31]. The use of the GWP of 21 does not impact our percentages since they are in terms of mass of methane. The source categories included in the WRI estimates were energy, agriculture, industrial processes, waste, land use and forest, and bunker fuels. The WRI estimates contained uncertainties and may have underestimated total state-wide GHG emissions (including  $CO<sub>2</sub>$ ) by a few Mt  $CO<sub>2</sub>e$  per year [31]. Furthermore, there were year-to-year variabilities. Considering the period from 2001 to 2011, the minimum and maximum methane emission estimates were 13.1 and 17.6 Mt  $CO<sub>2</sub>e$  per year [31].

The above methane emission estimates were based on the distribution of attributes in the DEP database, which may not be representative of actual distributions of all abandoned wells in Pennsylvania. Nonetheless, it was the only source that provided a breakdown of wells by attributes (including well type, plugging status, and coal area designation). Although we could estimate the number of oil vs. gas wells, we still needed to rely on the DEP database for the proportion of plugged wells and the proportion of wells in coal areas. If we scaled the well numbers by the highest percentage of oil wells estimated using historical data (our Estimate 3 in Table S5), methane emissions from abandoned oil and gas wells went down to  $0.02$  to  $0.04$  Mt CH<sub>4</sub> per year in Pennsylvania, which corresponded to 3% to 5% of total anthropogenic methane emissions in 2011 for the state. However, this estimate assumed that the distribution of plugging status and coal area designation in the DEP database was correct. A change in these distributions could both increase and decrease the total methane emissions from abandoned wells. For example, increasing the percentage of wells in coal areas from 21% to 31% increased methane emissions to 0.05 to 0.08 Mt CH<sup>4</sup> per year in Pennsylvania, which corresponded to 7% to 12% of total anthropogenic methane emissions in 2011 for the state. Overall, more studies are needed to better estimate the distribution of plugged wells and wells in coal areas to further improve methane emission estimates.

Uncertainties in the methane emission estimates could be addressed with additional data, including information on estimate emission factors, well attributes, and well numbers. For example, field data obtained using geophysical methods could be used to improve estimates of the depths of measured wells, in addition to assessing plugging and casing conditions. Production or other well data that were not publicly available may be collected from industry and used to estimate the well attributes, which could then be used to verify the well attribute estimation approach. Well-finding methods including magnetometry surveys and field visits could be used to estimate errors in well numbers. Finally, additional field measurements of abandoned wells with various well attributes, especially undersampled categories such as plugged oil wells in coal areas and unplugged gas wells in noncoal areas (Table 2), could improve emission factors. These data collection and analysis efforts are needed not just in Pennsylvania, but also in the many other states (e.g., West Virginia, Texas, and California) and other countries with a long history of oil and gas development.

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The number in parentheses for categorial variables indicates the number of categories. \* Intersection with one or more mineable coal seams [9].

\*\* Intersection with one or more mineable coal seams [9] and within 50 m of a well designated to be in a coal area by Pennsylvania's DEP [1].

	<b>Sampling Campaigns</b>	Number of Well	
Year	Month	<b>Measurements</b>	Counties
2013	July-August	14	McKean
2013	October	13	McKean
2014	January	14	McKean, Potter
2014	March	11	McKean, Potter
2014	June	15	McKean, Potter
2014	July	17	Venango, Lawrence, Allgheny
2014	October	22	McKean, Potter, Venango, Lawrence
2015	January	4	McKean, Potter
2015	March	26	McKean, Potter, Warren
2015	June	27	McKean, Clearfield, Venango, Warren

Table S2. Sampling rounds





Models L6a, L6b, L6c, and L3 are based on  $\log \dot{m}$ ; while Models N6b and N3 are based on  $\dot{m}$ .  $*$  p-values $<$ 0.05.

 $**$  p-values $<$ 0.01.

\*\*\* p-values<0.001.



Fig. S1.  $\mathbb{R}^2$  values of methane flow rate measurements  $(m)$ .



Fig. S2. Coefficient of variation vs. number of measurements at 27 well sites.



Fig. S3. Methane flow rates measured at 193 control locations, which are generally <10 m from a measured well site. Each bar represents a methane flow rate measurement, of which 53 are negative and 49 are positive. The large region of the plot with no bars represents the 91 measurements with flow rates below detection level.



Fig.  $S4.$  Noble gas data with respect to  $C, W$ , and  $P.$  For repeat measurements, the average of the data for the well are shown.



Fig. S5. Methane flow rates  $(m)$  vs. depth  $(d)$ , number of intersecting workable coal seams  $(n_C)$ [9], distance to active unconventional oil and gas wells  $(r_U)$ , and distance to underground natural gas storage fields  $(r_S)$ .



Fig. S6. Modeled and measured methane flow rates,  $\dot{m}$  (mg hr $^{-1}$  well $^{-1}$ ).



Fig. S7. Oil production [22] and well numbers from two historical sources.



## Table S4. Number of oil/gas wells in Pennsylvania including enhanced recovery (ER) wells and potential underreporting.

Our estimates are rounded to two significant digits. Numbers from other sources are not rounded.

<sup>†</sup> Factors represent (number of producing & injection wells) / (producing wells).

\* We multiply well numbers that do not include ER wells by the specified factor, determined based on data and/or the assumption that the five-spot method is used.

\*\* We multiply well numbers including ER wells by the specified factor, determined based on comparison of historical and modern digital records [18].

Table S5. Number of oil and gas wells drilled

Of Oil & Gas Only					
Years	Oil	Gas	$%$ Oil	$%$ Gas	Source
1859-1956*	172750	11720	94%	6%	Table 1, Dilmore et al. [18]
1859-1928 <sup>†‡<math>\diamond</math></sup>	134575	3859	97%	$3\%$	Proved Fields, P. 63, Arnold & Kemnitzer [24]
1859-1928 **	178	120	60%	40%	Wildcat, P. 63, Arnold & Kemnitzer [24]
$1929^{\dagger\ddagger\Diamond}$	2808	770	78%	22%	Scaled from Arnold & Kemnitzer [24]
1929-1956	3059	3857	44%	56%	Incl. Injection, Table SI-6 & SI-5, Dilmore et al. [18]
1929-1956	2026	3857	34%	66%	Excl. Injection, Table SI-5, Dilmore et al. [18]
1929-1956	37995	7723	83%	17%	Table SI-8, Dilmore et al. [18]
$1930 - 1937$ <sup>‡</sup>	21618	10243	68%	32%	Scaled from Arnold & Kemnitzer [24] and PA DER Progress Reports [22]
1938-1949 <sup>‡</sup>	21752	10307	68%	32%	Scaled from Arnold & Kemnitzer [24] and PA DER Progress Reports [22]
1930-1937 <sup>†<math>\diamond</math></sup>	14092	17769	44%	56%	Excl. Injection, Table SI-5, Dilmore et al. [18]
1938-1949 <sup>†<math>\diamond</math></sup>	14180	17879	44%	56%	Excl. Injection, Table SI-5, Dilmore et al. [18]
1950-1956 ***	1160	9283	11%	89%	PA DER Progress Reports [22]
1957-2012* <sup>†‡<math>\diamond</math></sup>	38732	63010	38%	62%	Table 1, Dilmore et al. [18]
1950-1991	18890	21640	47%	53%	PA DER Progress Reports [22]
1992-2013	646	1339	33%	67%	PA DEP Oil and Gas Reports [1]
1859-2012	211482	74730	74%	26%	Table 1, Dilmore et al. $[18]^{*}$
1859-2012	205726	112690	65%	35%	Our Estimate $1^{\dagger}$
1859-2012	263903	113029	70%	$30\%$	Our Estimate $2^{\ddagger}$
1859-2012 .	357379 .	112690 $\cdots$	76%	24%	Our Estimate $3^\circ$

Note that dry and other well types are not included in the above numbers.

\* Numbers used in estimate by Dilmore et al. [18].

† Numbers used in our Estimate 1.

‡ Numbers used in our Estimate 2.

 $\diamond$  Numbers used in our Estimate 3, which includes a factor of 2.0 for oil wells drilled prior to 1938.