ADVANCING EARTH AND SPACE SCIENCE

Earth's Future

RESEARCH ARTICLE

10.1029/2022EF002877

Climate Impact Comparison of Electric and Gas-Powered End-User Appliances

Florian Dietrich[1], Jia Chen[1], Ankit Shekhar[2], Sebastian Lober[1], Konstantin Krämer[1], Graham Leggett[3], Carina van der Veen[4], Ilona Velzeboer[5], Hugo Denier van der Gon[6], and Thomas Röckmann[4]

[1]Environmental Sensing and Modeling, Technical University of Munich (TUM), Munich, Germany, [2]Department of Environmental Systems Science, ETH Zurich, Zurich, Switzerland, [3]LI-COR Biosciences, Lincoln, NE, USA, [4]Institute for Marine and Atmospheric research Utrecht, Utrecht University, Utrecht, The Netherlands, [5]Environmental Modelling, Sensing and Analysis (EMSA), TNO, Petten, The Netherlands, [6]Climate, Air and Sustainability, TNO, Utrecht, The Netherlands

Key Points:

- Methane emissions at Oktoberfest are measured and classified as natural gas-based using isotopic analysis and the ratio of ethane to methane
- Oktoberfest could save 87% of total carbon emissions from energy consumption if all gas-powered appliances were replaced with electric ones
- We aim to make people aware how the carbon footprint of electric and natural gas-driven end-user appliances compares and evolves over time

Supporting Information:

Supporting Information may be found in the online version of this article.

Correspondence to:

F. Dietrich and J. Chen,
flo.dietrich@tum.de;
jia.chen@tum.de

Citation:

Dietrich, F., Chen, J., Shekhar, A., Lober, S., Krämer, K., Leggett, G., et al. (2023). Climate impact comparison of electric and gas-powered end-user appliances. *Earth's Future*, *11*, e2022EF002877. https://doi.org/10.1029/2022EF002877

Received 10 JUL 2022
Accepted 19 DEC 2022

Author Contributions:

Conceptualization: Florian Dietrich, Jia Chen
Data curation: Florian Dietrich, Ankit Shekhar
Formal analysis: Florian Dietrich, Jia Chen, Ankit Shekhar, Sebastian Lober, Konstantin Krämer, Carina van der Veen, Ilona Velzeboer
Funding acquisition: Florian Dietrich, Jia Chen, Hugo Denier van der Gon, Thomas Röckmann

Abstract Natural gas is considered a bridging technology in the energy transition because it produces fewer carbon emissions than coal, for example. However, when leaks exist, methane is released into the atmosphere, leading to a dramatic increase in the carbon footprint of natural gas, as methane is a much stronger greenhouse gas than carbon dioxide. Therefore, we conducted a detailed study of methane emissions from gas-powered end-use appliances and then compared their climate impacts with those of electricity-powered appliances. We used the Munich Oktoberfest as a case study and then extended the study to 25 major natural gas consuming countries. This showed that electricity has been the more climate-friendly energy source at Oktoberfest since 2005, due to the extensive use of renewable electricity at the festival and the presence of methane emissions, particularly caused by the incomplete combustion and leakages of natural gas in cooking and heating appliances. By contrast, at the global level, our study shows that natural gas still produces lower carbon emissions for end-user appliances than electricity in 18 of the 25 countries studied. However, as the share of renewable energy in the electricity mix steadily increases in most countries, the carbon footprint of electricity will be lower than that of natural gas in these countries in the near future. These findings from our comparison of the total carbon emissions of electric and gas-powered end-use appliances can help inform the debate on how to effectively address climate change.

Plain Language Summary Although natural gas is considered a relatively climate-friendly energy source compared to coal, leakage of methane, the main component of natural gas, can significantly increase the climate impact of natural gas. This is because methane is a very strong greenhouse gas. In this study, we focused on methane leakage from end-use appliances used for cooking and heating. Using the Munich Oktoberfest as a case study, we found that these end-use appliances produce significant methane emissions. Therefore, we investigated at which leakage rates and which electricity mixes it would be better to use electric appliances for cooking and heating instead to reduce overall carbon emissions. We found that despite leakage rates, natural gas is still more climate-friendly than electricity in most countries around the world. However, as the share of renewable energy in the electricity mix increases in most countries, electricity is becoming a more climate-friendly energy source every year. With this study, we want to make people aware of how the climate friendliness of electricity compares to natural gas over time.

1. Introduction

To reach the goal of net-zero greenhouse gas (GHG) emissions, the usage of natural gas is considered to be a bridge technology in many countries, as it is promoted to be more climate-friendly than burning coal (Ladage et al., 2021). However, methane (CH_4), the main component of natural gas, has a much stronger warming potential (GWP_{20} of 86 with the consideration of climate-carbon feedback) than carbon dioxide (CO_2) and is released when natural gas enters the atmosphere incompletely burned (Myhre et al., 2013). Recent studies have shown that anthropogenic fossil CH_4 emissions are generally underestimated (Alvarez et al., 2018; Hmiel et al., 2020; Schwietzke et al., 2016) and that the targets set in the Paris Agreement can only be met if CH_4 emissions are drastically reduced (Nisbet et al., 2019).

To improve the quantification of CH_4 emissions, many studies around the world have focused on determining these CH_4 emissions using various measurement and modeling approaches including mobile street-level

10.1029/2022EF002877

Investigation: Florian Dietrich, Jia Chen, Ankit Shekhar, Sebastian Lober, Konstantin Krämer, Graham Leggett
Methodology: Florian Dietrich, Jia Chen, Ankit Shekhar
Project Administration: Florian Dietrich, Jia Chen
Resources: Jia Chen, Graham Leggett, Hugo Denier van der Gon, Thomas Röckmann
Software: Florian Dietrich, Jia Chen, Ankit Shekhar, Sebastian Lober, Konstantin Krämer
Supervision: Florian Dietrich, Jia Chen
Visualization: Florian Dietrich, Jia Chen, Ankit Shekhar, Sebastian Lober, Konstantin Krämer
Writing – original draft: Florian Dietrich, Jia Chen, Ankit Shekhar
Writing – review & editing: Hugo Denier van der Gon, Thomas Röckmann

measurements with fast in situ analyzers on vehicles (Gallagher et al., 2015; Jackson et al., 2014; Maazallahi et al., 2020; Phillips et al., 2013; von Fischer et al., 2017; Weller et al., 2018, 2020), interpretation of plume measurements utilizing the downwind tracer flux approach (Mitchell et al., 2015; Omara et al., 2016; Roscioli et al., 2015; Zimmerle et al., 2015), larger scale airborne measurements with analyzers on aircraft (Karion et al., 2013; Lyon et al., 2016), local eddy flux measurements (Gioli et al., 2012; Helfter et al., 2016), or FTIR sensor networks (Chen et al., 2016; Dietrich et al., 2021; Hase et al., 2015; Jones et al., 2021; Klappenbach et al., 2021; Makarova et al., 2021; Vogel et al., 2019; Zhao et al., 2019).

To attribute the source and determine the leakage rate, either isotopic signatures of the gas are measured (Beck et al., 2012; Chamberlain et al., 2016; Fisher et al., 2006; Lu et al., 2021; Menoud et al., 2020, 2021; Röckmann et al., 2016; Zimnoch et al., 2019), the ethane to methane ratio is determined, because ethane is a unique tracer for fossil fuel related methane emissions, or both methods are used simultaneously (Allen et al., 2013; Maazallahi et al., 2020; Yacovitch et al., 2014, 2015, 2018; Zavala-Araiza et al., 2015).

In our study, we focus specifically on total emissions from natural gas compared to electricity in the end-use sector, as these emissions are suspected of contributing considerably to the underestimates in current methane emissions inventories (McKain et al., 2015; Plant et al., 2019). We use the Munich Oktoberfest—a very large festival with more than 6 million visitors per year—as a case study where gas end-user appliances are highly concentrated. Overall, 40% of the energy demand at Oktoberfest is met by natural gas (mainly for heating and cooking). The event has already been identified as a significant source of CH_4 (Chen et al., 2020), but it has not been fully understood what portions of these methane emissions are due to natural gas leakages. Due to the extensive use of gas-powered cooking and heating appliances in a limited space, we believe Oktoberfest is a well-suited experimental setup where the climate impacts of using such appliances can be determined quite easily. In addition, Oktoberfest has an exemplary, steadily increasing share of renewable energy in electricity consumption (Landeshauptstadt München, 2019; Landeshauptstadt München Redaktion, 2020), making it a well-suited event where the climate impacts of gas-powered and electric end-user appliances can be compared in a representative way. Therefore, Oktoberfest is particularly suited for demonstrating the differences in total GHG emissions from natural gas compared to electricity for the case where the share of renewables increases over time.

Since emissions reductions only at Oktoberfest will not have a noticeable impact on a global scale, these findings that electricity could be the more climate-friendly alternative for cooking and heating, depending on the electricity mix and leakage rate, need to be transferred globally to achieve positive climate effects. While previous studies have focused either exclusively on leakage rates from gas-fired end-user appliances (Lebel et al., 2022) or on comparing emissions from coal to those from natural gas combustion (Fulton et al., 2011; Ladage et al., 2021; Qin et al., 2017; Tanaka et al., 2019), little consideration has been given to electricity generated by an ever-growing share of renewable energy. When considering how such end-user appliances can be operated in a climate-friendly way, for example, at other festivals, in restaurants or in private households, such a comparison is the basis for the right decision. That is why we also analyze the climate impact of electric and gas-powered appliances for 25 major natural gas-consuming countries to show where and when it is more climate-friendly to use one or the other energy source.

2. Materials and Methods

In the present study, we first determined the total CH_4 emissions from Oktoberfest based on mobile measurements combined with an atmospheric transport model, then used isotope and ethane analyses to assign emission sources and finally determined the point in time and the break-even share of renewables at which electric appliances are more climate-friendly than gas appliances at Oktoberfest. Afterward, we applied our newly developed methods to a global scale and compared the carbon footprint of natural gas and electricity as an energy source for end-user appliances around the world.

2.1. Mobile In Situ Measurements at Oktoberfest

We carried out mobile in situ measurements at Oktoberfest 2019. For that, we utilized the LI-7810 CH_4/CO_2/H_2O Trace Gas Analyzer from LI-COR, which uses the optical feedback cavity-enhanced absorption technique (OF-CEAS), as a mobile backpack instrument. Simultaneously to the measurements, the current position of each data point was recorded using a GPS application on a smartphone that was time-synchronized to the gas analyzer.

In contrast to our preceding study in 2018 (Chen et al., 2020), we were allowed to perform measurements both outside and inside the festival premises as well as inside the tents. To determine the emission strength, only the measurements around the perimeter of Oktoberfest (hereafter referred to as *outer rounds*) were used to allow for an easy comparison to our 2018 results. The measurements on the site (hereafter referred to as *inner rounds*) were mainly used to find emission hotspots.

To cover different days of the week and times of the day, we completed 56 rounds during Oktoberfest. For a comparison to CH_4 emissions outside the festival period, we also completed 15 measurement rounds after the end of Oktoberfest. We do not have any measurements before the start of the festival because the loaner gas analyzer did not arrive in time. However, we assume that the emissions before and after the Oktoberfest are comparable. The rounds during the festival period were divided into outer and inner rounds. Each inner round was always combined with at least one outer round to obtain background concentrations for CH_4 each time.

For the outer rounds, we chose the shortest possible walking distance around the perimeter of Oktoberfest, which is directly behind the security fences. The walking distance for such a round is about 2.6 km (see Figure S1 in Supporting Information S1) and took us on average about 40 min each. Combined with a gas analyzer sampling rate of 1 Hz, approximately 2,400 measurement points were recorded per round.

During our measurements inside the festival premises, we followed two routes that were predefined by us. Both of them were chosen to capture the emissions caused by the large tents and booths on the streets best. Therefore, they follow the streets between the tents that are mainly located in the northwest quarter (see Figure S1 in Supporting Information S1).

2.2. Modeling the CH_4 Emissions

To quantify the CH_4 emissions of Oktoberfest, we combined the measurements around the perimeter of Oktoberfest with the modeling approach developed in Chen et al. (2020). The model is based on a multiple Gaussian plume approach using the 17 largest beer tents as point sources, linearly weighted according to their size, and superimposed to model a continuous expected concentration signal at the sampling points around the perimeter of Oktoberfest. Since we used only the outer rounds for this approach to determining emissions, the sampling sites are at least 100 m away from the nearest sources, resulting in well-mixed concentrations. Since emissions are represented by the concentration differences between upwind and downwind sites, background concentrations were modeled linearly based on the 10% smallest measured values for each round and then subtracted. The entire model results in one emission number for each round. The final emission number is an averaged value of the emission numbers from the 38 individual outer rounds, taking into account the uncertainties of the four input parameters wind speed, wind direction, background concentration, and measured concentration values. For this purpose, these four input parameters were each modeled as an independent Gaussian distribution and the model was run 1,000 times. The resulting mean and standard deviation of the distribution of these 1,000 emissions represent the final emission number and its uncertainty.

The main differences to the investigations in Chen et al. (2020) were a different CH_4 analyzer (in 2018, the Picarro G4301 gas scouter, which is based on the cavity ring-down principle was used) and wind measurements closer to the festival premises, as the sensitivity study in Chen et al. (2020) indicated a strong influence of the wind measurements to the atmospheric model. Therefore, we established a wind sensor very close to the festival premises on top of a building, which is located approximately 150 m west of Oktoberfest (48.134°N, 11.545°E, 26 m a.g.l.). As a sensor, the Lufft WS200-UMB 2D ultrasonic wind sensor was utilized. The other model parameters such as the number of emitters, plume modeling algorithm, averaging approach, etc. were equal to Chen et al. (2020).

2.3. Air Sampling

In addition to the backpack measurements, we took samples of the environmental air at different locations, such as inside and outside the festival premises, inside the beer tents, next to possible emission hotspots at the festival, and in the subway. For this purpose, Standard FlexFoil air sampling bags from SKC Ltd. with a volume of 3 L were used. In total, we filled 30 bags and shipped them afterward to Utrecht University and TNO in the Netherlands, where they were analyzed in the lab. At Utrecht University, in 12 bags (two of them were background samples) $\delta^{13}C$ and δD were analyzed using Isotope Ratio Mass Spectrometry (IRMS) (Brass & Röckmann, 2010).

The device used was the spectrometer model Delta V Plus/Deltaplus XL from Thermo Fisher Scientific Inc. At TNO, the Δethane to Δmethane ratios of the remaining 18 bags (seven of them were background samples) were measured using the Quantum Cascade Laser (QCL) absorption spectrometer model QCL-TILDAS-76 from Aerodyne Research Inc.

2.4. Isotopic Analyses of Air Samples

To determine, whether the measured methane is anthropogenic or biogenic, analyses of the carbon isotopes were made. We used the $\delta^{13}C$ method, in which the ratio between ^{13}C and ^{12}C of the sample gas is compared to the ratio of a predefined standard. Similar to $\delta^{13}C$, we also looked at the ratio of deuterium to normal hydrogen using the δD method. The mathematical expressions for these two methods are shown in Section S2 in Supporting Information S1.

Since the sampled air also includes the unknown background isotopic signature of the gas, we utilized Keeling plots to determine the isotopic signature of the gas emitted exclusively by Oktoberfest. These plots linearize the relation between the $\delta^{13}C$ (or δD) value of the measured air sample and the methane concentration so that the $\delta^{13}C$ (or δD) portion added by the unknown source can be determined (Keeling, 1958).

2.5. Ethane to Methane Ratio of Air Samples

As a second kind of analysis to determine the origin of the sample gas, we examined the Δethane to Δmethane ratio (Allen et al., 2013; Maazallahi et al., 2020; McKain et al., 2015; Yacovitch et al., 2014, 2015, 2018; Zavala-Araiza et al., 2015). For this purpose, we subtract the background concentrations of methane ($CH_{4,bg}$) and ethane ($C_2H_{6,bg}$) from the measured concentrations ($CH_{4,sample}$ and $C_2H_{6,sample}$) to obtain the ratio of the gas added by the source:

$$\frac{C_2H_{6,\text{source}}}{CH_{4,\text{source}}} = \frac{\Delta C_2H_6}{\Delta CH_4} = \frac{C_2H_{6,\text{sample}}}{CH_{4,\text{sample}} - CH_{4,\text{bg}}} \tag{1}$$

Thereby, we assumed that the ethane concentration $C_2H_{6,bg}$ of the background can be set to zero, which is supported by our five background air samples that we took during the time of Oktoberfest 2019. For each sampling point, a Δethane to Δmethane ratio was determined and afterward compared with the ratio of the Munich gas network. Since the composition of Munich's natural gas is determined only once a month, a weighted average was calculated for the 16 days of Oktoberfest 2019, which took place 10 days in September and 6 days in October 2019. The uncertainties were calculated using the 99% confidence intervals of all gas samples measured in the tent combined with the minimum ($r_{\text{ethane, Sept}} = 3.04\%$) and maximum ($r_{\text{ethane,Oct}} = 3.07\%$) ethane share in September and October 2019, respectively (SWM Infrastruktur GmbH und Co. KG., 2019a, 2019b).

2.6. Calculation of the Climate Impact

To find out, whether gas or electric appliances for cooking and heating have a better carbon footprint, the total emission factors in CO_2 equivalents (CO_2eq) are calculated for the case of electric and gas use only. In our study, we did not focus only on end-user appliances, but looked at the entire supply chain. To this end, we included emission factors for the various energy sources, including power plant efficiencies and, for natural gas-related processes, methane leakage rates.

For the efficiency of the end-user appliances themselves, we have assumed that it is the same for electric and gas-powered appliances. This seems to be a reasonable assumption in terms of a mean value, as several prior studies have found a wide range of efficiencies, some stating that electric appliances (Hager & Morawicki, 2013) and some stating that gas-powered appliances (Adria & Bethge, 2013) require less energy. This wide range is due to different types of stoves and the time used for comparison. Gas-powered appliances, while generally less efficient, heat up much faster than electric appliances.

To calculate the emission factor $EF_{\text{elect}}(t)$, if only electricity would be used as an energy source, Equation 2 is utilized:

$$EF_{\text{elect}}(t) = \sum_{n=1}^{8} EF_n \cdot p_{\text{elect,n}}(t) \tag{2}$$

These emissions differ for each country and are time-dependent, as the proportions of fuel types used for electricity production $p_{\text{elect,n}}(t)$ vary over time. In this study, we considered four different types of non-renewable energy sources (coal, oil, gas, and nuclear power) and four different types of renewable energies (hydro, solar, wind, and geothermal/biomass power) with different emission factors EF_n obtained from Amponsah et al. (2014) (see Table S1 in Supporting Information S1). These emission factors represent global mean values and may vary from country to country due to technological progress. Therefore, the results for certain countries may be subject to uncertainties and should be examined more closely if a specific country is to be studied.

For the case, where we assumed that only natural gas is used for producing the same amount of energy, the emission factor $EF_{\mathrm{NG,\,total}}$ is calculated by adding the emission factors of combusting natural gas EF_3 (see Table S1 in Supporting Information S1) and leaking CH_4, as shown in Equation 3:

$$EF_{\mathrm{NG,total}} = EF_3 \cdot (1 - r_{\mathrm{leak}}) + \frac{\rho_{\mathrm{CH4}} \cdot \mathrm{GWP}_{20,\mathrm{CH4}}}{E_{\mathrm{d,NG}}} \cdot r_{\mathrm{leak}} \tag{3}$$

Where r_{leak} is the leakage rate of natural gas, ρ_{CH4} is the density of CH_4 (0.668 kg/m^3), $\mathrm{GWP}_{20,\mathrm{CH4}}$ the 20-year global warming potential of methane considering climate-carbon feedback (86 tCO_2eq/t) (Myhre et al., 2013) and $E_{\mathrm{d,NG}}$ the energy density of natural gas ($3.6 \cdot 10^{-5}$ TJ/m^3).

2.7. Country Specific Emission Data

Equations 2 and 3 are applied for different countries and years, resulting in a time-dependent country comparison of the carbon footprint of electrical versus gas-driven appliances. We examined the shares and types of non-renewables and renewables in the electricity mix only for countries that account for at least 0.5% of global natural gas consumption (40 countries in total). However, we excluded countries with a renewable energy share of less than 10% in 2019 (which primarily includes Middle Eastern countries), as we want to focus in this study primarily on how an increasing share of renewable energy can make electricity more climate-friendly compared to natural gas. The chosen 25 countries account for 75% of the world's natural gas consumption, with the United States alone accounting for about 21.7%, followed by Russia and China with 12.4% and 5.4%, respectively (WorldData.info, 2020). Similar to the Oktoberfest investigations, in this country comparison, we focused primarily on the climate impact of appliances at the end-user, namely cooking and heating appliances in the household sector.

The data on the electricity mix was taken from the *BP Statistical Review of World Energy, 69*th *Edition* (bp., 2020). The electricity mix data indicate the type and proportion of energy sources (coal, natural gas, oil, nuclear, and renewables) used to generate electricity from 1965 to 2019 (for some countries only from 2000 to 2019). In this study, we concentrate on the 21st century only. The data on the share of renewable energy was also cross-checked with *Trends in Renewable Energy* provided by the International Renewable Energy Agency (IRENA) (International Renewable Energy Agency, 2020). IRENA provides data from 2000 to 2018.

2.8. Phase Transition Plot

To show how the shares of renewable and non-renewable energy and the respective sources for these energies affect the climate friendliness of electricity and natural gas, we used phase transition diagrams. These 2D heat maps depict the standardized emissions difference between electricity and natural gas as a function of renewable energy shares of electricity generation and methane leakage rates. Red shaded areas indicate that natural gas is the more climate-friendly energy, while blue shaded areas indicate that electricity is more climate-friendly. Such a phase transition diagram is shown in Figure 4 on the left and is explained in more detail in the related results Section 3.3.

We used 2019 energy data for both Oktoberfest and each of the 25 countries to create these charts. To create the phase transition plots, we varied the methane leakage rate from 0% to 15% in 0.1% increments and the renewable energy share from 0% to 100% in 1% increments, and calculated the difference in carbon footprint between CO_2eq emissions from electricity and natural gas for each of these points. Then, the minimum percentage of renewables required to make electricity the more climate-friendly energy source is determined by intersecting the line representing all points at which natural gas and electricity are equally climate-friendly with the actual methane leakage rate. The y-value of this intersection represents the minimum percentage of renewable energy required to make electricity the more climate-friendly energy source, taking leakage into account. We then calculated the difference between this minimum share and the current (2019) share of renewables in each country and plotted the differences as a bar chart in Figure 6. Error bars were determined by using the lower and upper bounds

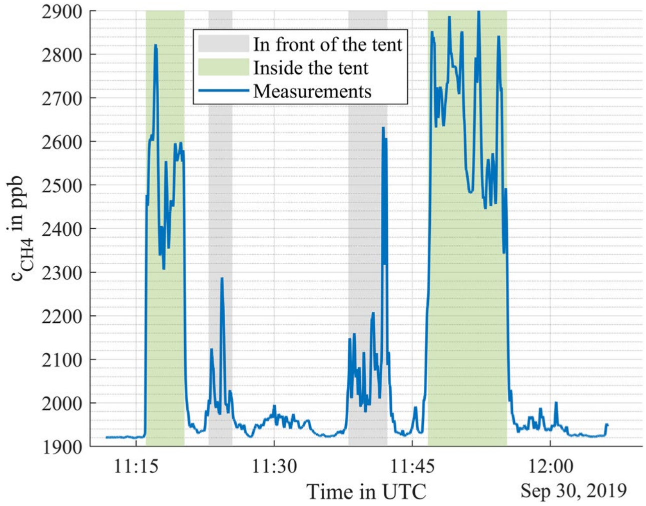

Figure 1. CH_4 mole fractions measured at the Oktoberfest premises. The concentrations measured during a tour at the Oktoberfest premises show a large heterogeneity and are especially enhanced inside (green shaded) and in front of (gray shaded) the beer tents.

of the 90% confidence interval of the leakage rate distribution, which correspond to 0.22% and 5.35%, respectively. The details are further explained in Section 3.4.1.

3. Results

3.1. CH_4 Emission Number

Utilizing all 38 outer rounds during Oktoberfest, we determined an emission number of $(8.5 \pm 0.5)\,\mu g\left(m^2 s\right)^{-1}$. The value is in the same order of magnitude as the one quantified in 2018: $(6.7 \pm 0.6)\,\mu g\left(m^2 s\right)^{-1}$ (Chen et al., 2020). Emissions identified for the period after the end of the festival also have a positive offset in 2019 ($2.5\,\mu g\left(m^2 s\right)^{-1}$ vs. $1.1\,\mu g\left(m^2 s\right)^{-1}$). The distributions of these two emission numbers are shown in Figure S2 in Supporting Information S1. Possible reasons for these slightly higher numbers in 2019 include more accurate wind measurements taken closer to the festival premises or real changes in emissions between the 2 years. Still our 2019 measurements confirm that Oktoberfest is a significant source of CH_4 that can be made more climate friendly if the emission sources can be precisely located and quantified and mitigation measures can thus be developed.

3.2. Source Attribution

To find emission sources on the large festival premises, measurements were made in the vicinity of possible sources and a categorization of the sources into biogenic and anthropogenic origin was carried out. For this purpose, we performed mobile in situ measurements inside the festival premises and determined the isotopic signature and the ethane to methane ratios of air samples taken at Oktoberfest.

3.2.1. Inside Measurements

During our measurements on the festival site, the measurements with our instrument did not detect any CH_4 enhancements in the vicinity of gas control stations and pipelines. This also confirms the statement of Stadtwerke München (SWM) that these stations are already carefully monitored and maintained. Therefore, we did not include these types of emissions in our modeling approach for Oktoberfest. CH_4 concentrations were significantly elevated especially next to the open doors of the beer tents (see Figure 1). In addition, we were allowed to enter one of the large beer tents with our backpack analyzer to verify our assumption further and localize the sources in more detail. Figure 1 shows that the CH_4 mixing ratios of up to 2,900 ppb inside the tents are even higher than in front of the entrance (approximately 2,000–2,600 ppb). Most of the high enhancements were detected when passing the tent kitchen, where cooking is done with gas appliances supplied with natural gas provided by the Munich gas network.

On the streets of the festival grounds, we discovered only two additional hotspots during our 18 tours at the site that were not associated with open tent doors and windows or tent chimneys. The first was close to one of the grilled chicken stalls that run on natural gas and the second was next to a place where fish were grilled over charcoal fires.

We conclude that mostly the 17 large beer tents contribute significantly to the CH_4 emissions of Oktoberfest to the atmosphere. This supports the statement of Chen et al. (2020) that beer tents are the major CH_4 source at Oktoberfest. Therefore, it is a valid approach to model only the large beer tents as sources in order to determine the overall emission strength of Oktoberfest. However, CH_4 is not only emitted by the chimneys but also by open doors and windows of the tents. This should be considered if a spatially higher resolved model is used.

To identify, whether these emissions are of biogenic origin produced by the human bodies or of anthropogenic origin caused by incomplete combustion and leakages of natural gas-driven appliances, air samples were taken and analyzed in the lab afterward.

3.2.2. Isotopic Composition

The results of the isotopic analyses of the samples taken at Oktoberfest are shown in Figure 2 (left and center) as a Keeling plot. The various types of sampling locations, such as in-tent, subway (inside the crowded train

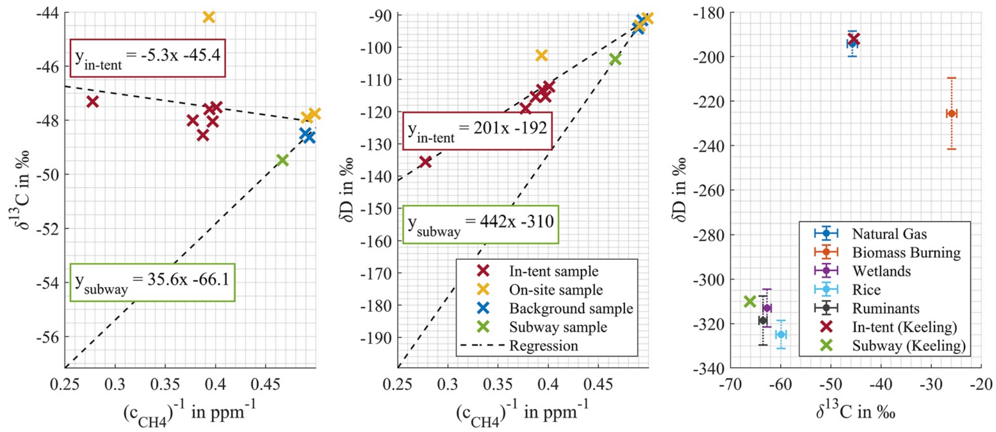

Figure 2. δ^{13}C (left) and δD (center) Keeling plot of the air samples taken at Oktoberfest. In addition, two regression lines are shown in both figures for the Oktoberfest and subway samples, respectively, to determine the isotopic signatures of the sources. Right: Isotopic fingerprint (δ^{13}C vs. δD) off different gas sources (dots with whiskers) based on results of Menoud et al. (2021) including source signatures of Oktoberfest, derived from the Keeling plots (crosses). While the signature of the subway measurement (green cross) is close to biogenic sources, the Oktoberfest measurements (red cross) show a comparable signature to natural gas. These results indicate that Oktoberfest emissions are primarily due to natural gas leakage.

between Oktoberfest and Munich Central Station), and background (outside the Oktoberfest premises) samples, are indicated by different colored crosses. To determine the isotopic signature of each of the two source types, a linear regression line is drawn through all sample points of each source type including the background samples for both δ^{13}C and δD. In this Keeling plot analysis, the intercept of the regression line with the *y*-axis represents the isotopic signature of the gas added by the unknown source. These intercepts are for [δ^{13}C; δD] at [−45.4‰; −192‰] for the in-tent samples and [−66.1‰; −310‰] for the subway sample.

In Figure 2 (right), these isotopic source signatures are compared to typical isotope signatures of different source types, such as natural gas, biomass burning, wetlands, rice, and ruminants. The subway sample (light green cross) shows a clear biogenic signature, which is the expected behavior of a crowd of people. In contrast, the in-tent signature (red cross) is very close to the signature of natural gas, suggesting that the methane emissions of Oktoberfest are primarily caused by fugitive natural gas leakages.

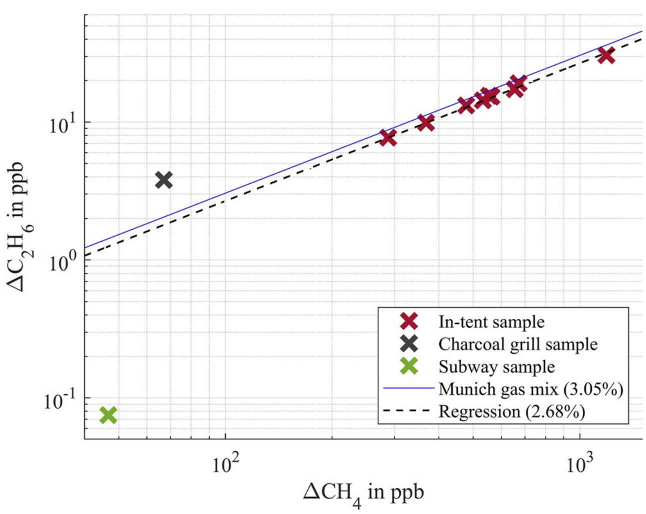

Figure 3. Correlation between Δethane (ΔC_2H_6) and Δmethane (ΔCH_4) of air sampled at various locations at Oktoberfest in a log-log plot. With the exception of the measurements for the subway (green) and the charcoal grill (gray), which show lower and higher ethane enhancement, respectively, all points lie on a line with slope 1, implying a linear relationship between Δethane and Δmethane. Since the slope of this regression line is very close to that of the Munich natural gas mixture, these results indicate that the high methane enhancement inside the tents is caused by natural gas.

3.2.3. Ethane to Methane Ratio

The results of the ethane analyses are shown in Figure 3, where the Δethane to Δmethane correlation is shown as a scatter plot using logarithmic axes. In addition to the two source types *in-tent* and *subway* that we also analyzed with respect to the isotopic fingerprint of the samples in Figure 2, another source type, namely air sampled in front of a large charcoal grill, was analyzed. These three source types exhibit significantly different behavior. The nine samples taken inside the tents (red crosses) show an almost constant Δethane to Δmethane ratio of 2.68% [2.57%, 2.78%] (99% CI). The number is very close to 3.05%, which is the reported averaged ethane to methane ratio of the natural gas used in Munich in September and October 2019 (SWM Infrastruktur GmbH und Co. KG., 2019a, 2019b). Together with the high concentrations measured inside the tents (see Figure 1), this result confirms our hypothesis that the elevated methane levels at Oktoberfest are primarily due to leaking natural gas. A distribution of these ratios is illustrated in Figure S3 in Supporting Information S1. In contrast, the subway sample (light green) has a much lower ethane content and the charcoal grill sample (gray) has a higher ethane content, indicating that small amounts of other methane emissions are present in addition to the natural gas leaks.

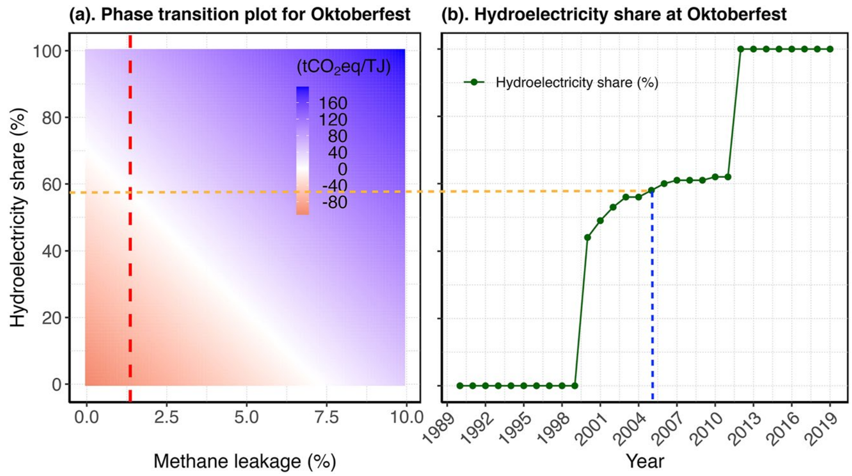

Figure 4. Left: phase transition diagram of hydropower shares in electricity generation versus methane leakage rates. It shows the difference in emissions (in tCO_2eq/TJ) between electricity and natural gas as the energy source for heating and cooking at Oktoberfest. For positive values (blue shaded areas), the use of electricity leads to lower emissions compared to natural gas; for negative values (red shaded areas), the opposite is true. The red vertical dashed line represents the leakage rate of 1.4% measured at Oktoberfest, while the orange horizontal dashed line represents the associated share of renewable energy, where electricity is the more climate-friendly energy source compared to natural gas (58%). Right: share of renewable energies in electricity consumption at Oktoberfest. The dashed orange line shows that the share of renewable energies at Oktoberfest reached the break-even point from 2005 onwards, which means that electricity has been the more climate-friendly energy source than natural gas at the festival ever since.

Dividing the ethane fractions of our Oktoberfest samples ($r_{\text{ethane,Okt}}$) by that of the Munich natural gas mix ($r_{\text{ethane,Muc}}$), we calculate the ratio r_{fugitive} between the ethane shares of these two gases to be

$$r_{\text{fugitive}} = \frac{r_{\text{ethane,Okt}}}{r_{\text{ethane,Muc}}} = \frac{2.68\%\ [2.57\%, 2.78\%]}{3.05\%\ [3.04\%, 3.07\%]} = 88\%\ [84\%, 91\%]. \quad (4)$$

Based on this calculation, we assume that about 88% of the methane emissions in the tents are attributable to fugitive natural gas. The remaining 12% are likely caused by biogenic processes. The values in squared brackets represent the 99% confidence intervals.

In summary, we conclude that the enhanced methane concentrations measured at Oktoberfest 2018 and 2019 are mainly due to natural gas that is either not fully combusted or leaking from natural gas-fueled equipment, such as heaters, grills, and ovens. According to our investigations, gas regulation stations and pipelines at Oktoberfest do not leak significantly and are, therefore, not the reason for the methane enhancements observed.

3.2.4. Leakage Rate

The leakage rate r_{leak} of CH_4 at Oktoberfest is determined as the ratio between the CH_4 loss measured with our instruments ($M_{CH4,\text{loss}}$) and the total CH_4 consumed at Oktoberfest 2019($M_{CH4,\text{total}}$), the calculation of which is explained more in detail in Section S3 in Supporting Information S1:

$$r_{\text{leak}} = \frac{M_{CH_4,\text{loss}}}{M_{CH_4,\text{total}}} = \frac{1.635 \cdot 10^3\text{kg}}{1.186 \cdot 10^5\text{kg}} = 1.4\% \quad (5)$$

The determined leakage rate is very close to the leakage rate determined by Chen et al. (2020) (1.1%) and lower than the leakage rates determined in the Boston area for all downstream leakage (2.7 ± 0.6%) (McKain et al., 2015) and the entire supply chain (3.3%–4.7%) (Sargent et al., 2021). Alvarez et al. (2018) suggested that methane losses in the U.S. oil and natural gas supply chain are equivalent to 2.3% of gross U.S. natural gas production. However, only end-use equipment was analyzed for Oktoberfest. All leakage in the upstream and midstream natural gas process is not captured by the measurements in this study. We, therefore, conclude that the leakage rate of end-use appliances at Oktoberfest appears to contribute significantly to the overall leakage rate of the natural gas chain. These results suggest that it is relatively easy to achieve a significant improvement in the carbon footprint of the natural gas chain by simply reducing the leakage rates of end-use appliances. This is likely true not only for Oktoberfest, but for many end-use gas appliances in the world.

3.3. Energy Consideration at Oktoberfest

Although the total energy demand of Oktoberfest has risen within the past 20 years, mainly due to an increase in electricity consumption, total carbon emissions have been drastically reduced. This effect is due to the steadily increasing proportion of renewable electricity used at the festival. Since 2011, only green electricity has been used, 100% of which is generated from hydropower, one of the cleanest renewable energy sources (Amponsah et al., 2014). It should be noted that the hydropower used is only an equivalent for purchased energy and does not represent actual time-of-use statistics. Therefore, emissions caused by electricity use at Oktoberfest could be greater than assumed in our study, depending on which paradigm is used to calculate emissions. We chose this approach of equivalent purchased energy over averaged or marginal emission factors because we believe that purchasing green power at higher rates than conventional power will encourage the expansion of renewable energy over time and should therefore be rewarded. A more detailed analysis of the energy development at Oktoberfest can be found in Section S4 in Supporting Information S1.

We incorporated all energy information determined for Oktoberfest 2019, such as fossil electricity composition, natural gas CO_2eq emissions, renewable energy type, and CH_4 leakage rate, into a phase transition diagram (see Figure 4). This identifies how a changing share of hydropower affects the climate friendliness of electricity compared to natural gas. From the intersection of the white line with the CH_4 leakage rate (red dashed line), it is possible to determine the fraction of hydropower from which electricity is the more climate-friendly energy source than natural gas with consideration of fugitive CH_4 leakages.

Assuming a methane leakage rate of 1.4% determined in our study, electricity with a renewable share greater than 58% is more climate-friendly than natural gas for Oktoberfest as demonstrated in the phase transition plot in Figure 4 (dashed horizontal orange line). Since the share of renewable energy at Oktoberfest exceeded the threshold of 58% in 2005 (see Figure 4, right), it would have been beneficial from a climate change perspective to replace all gas appliances at Oktoberfest with electric appliances starting in this year. In 2019 alone, this could have saved up to 450 tCO_2 emissions.

Such a reduction in emissions for an event that lasts only 2 weeks and is already quite climate-friendly is remarkable and gives us the opportunity to investigate on a larger scale how the type of energy source could help reduce carbon emissions worldwide.

3.4. Comparison of the Climate Impact in Different Countries

The Oktoberfest study showed that whether natural gas or electricity is the more climate-friendly energy source depends very much on the composition of the electricity mix as well as the leakage rate of natural gas. Since each country has its own electricity mix composition, we applied our approach developed for Oktoberfest to 25 major natural gas-consuming countries to understand which of the two energies is more climate-friendly for each of them. For these 25 countries, we studied the climatic impact of electric and natural gas energy sources in two ways. First, using their long-term (2000–2019) temporal trends (shown in Figure 5) and then through a more detailed analysis of the estimated renewable energy gap for the most current year 2019 (shown in Figure 6).

3.4.1. Comparison of Country Emission Over Time

Figure 5 shows the temporal trend of each countries' emission factor for both natural gas (shaded area) and electricity (green solid line). As shown in our Oktoberfest field study, the methane leakage rate has a significant impact on the emission factor of natural gas. Since it is beyond the scope of this paper to determine the leakage rate in each of these countries, we calculated leakage rates based on literature values instead. For this purpose, we calculated the ratio between the sum of all reported fugitive and vented CH_4 emissions of each country and the respective total consumption. Further details on the calculation of the leakage rate can be found in Section S5 in Supporting Information S1. These values only reflect the leakage rates in the respective countries, not the leakage rate in the entire natural gas supply chain. Therefore, upstream and downstream leakage rates are underestimated in countries that mainly consume natural gas and overestimated in countries that mainly produce natural gas. To compensate for these inconsistencies, we determined the distribution of all calculated leakage rates at the country

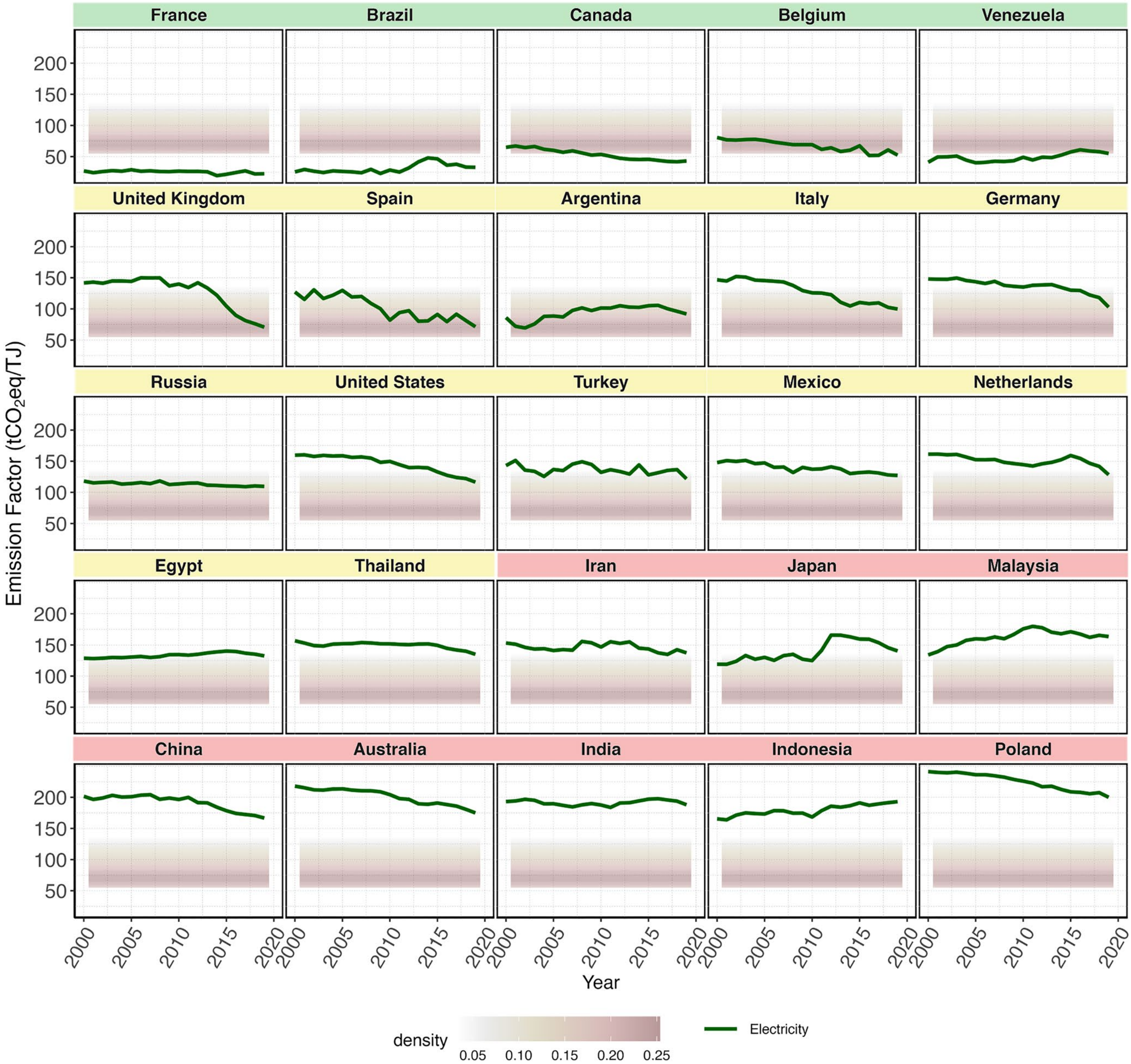

Figure 5. Comparison of CO_2eq emission factors from electricity and natural gas sources over the 20 years from 2000 to 2019 for 25 major natural gas-consuming countries. The emission factor of natural gas is represented by a distribution rather than a distinct line because of the methane leakage rate, which cannot be accurately determined for all countries studied. The countries are colored according to whether their current emission factor for electricity generation is below (green), within (yellow), or above (red) the 90% confidence interval of the distribution of natural gas emission factors.

level. This distribution is presented as a kernel density, with a 90% confidence interval considered, resulting in a lower bound of 0.22% and an upper bound of 5.35% for the leakage rate. This range represents the potential leakage rate throughout the supply chain for all combinations of natural gas producing, transit, and consuming countries. As a result of this leakage rate range, the comparison between the emission factors of electricity and natural gas also only provides upper and lower bounds as of which year electricity could be the more climate-friendly energy source for cooking and heating.

While the carbon footprint of natural gas has remained nearly constant over the years, the carbon emissions of electricity have fluctuated for most countries. This behavior is due to the widely varying emission factors for the different energy sources used to generate electricity (see Table S1 in Supporting Information S1) and is further

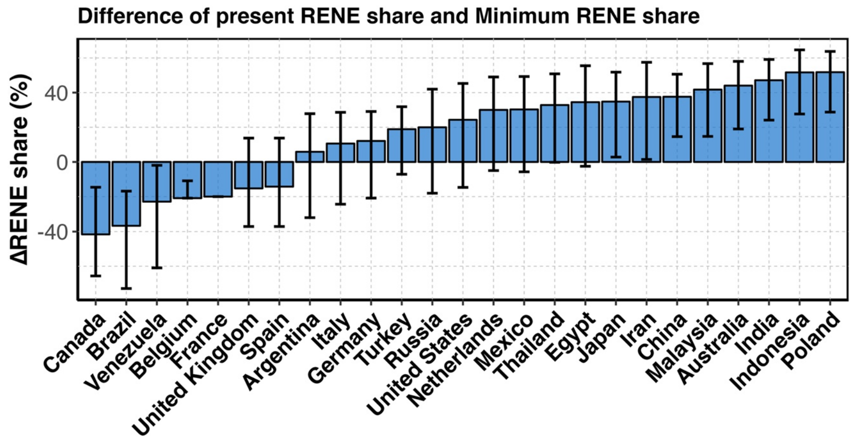

Figure 6. The difference in current renewable energy (RENE) share to reach the break-even point, where natural gas and electricity have the same carbon footprint for 25 countries. Negative values indicate that electricity is already the more climate-friendly energy source compared to natural gas. Values greater than zero represent the increase in RENE share required to make electricity more climate-friendly compared to natural gas. Error bars were determined by using the lower and upper bounds of the 90% confidence interval of the leakage rate distribution, which correspond to 0.22% and 5.35%, respectively.

analyzed in Section S6 in Supporting Information S1. According to the absolute carbon emission factors for electricity generation in 2019, these 25 major natural-gas consuming countries can be classified into three groups (see colored backgrounds in Figure 5).

The first group (green) consists of five countries where the emission factor of electricity in 2019 was below the lower limit of the natural gas emission factor (corresponding to a very small leakage rate of only 0.22%), making electricity very likely the more climate-friendly energy source compared to natural gas. Prominent examples of the first group are Brazil, which has a very high share of renewable energy, and France, which generates its electricity mainly through the extensive use of nuclear power. These results show that not only the share and type of renewable energy, but also the emission factor of non-renewable sources is decisive in determining whether electricity is the more climate-friendly energy source than natural gas.

The second group (yellow) consists of 12 countries where the absolute carbon emissions from electricity intersect the range between the lower and upper limits of the possible natural gas emission levels. These 11 countries have different characteristics that explain why their electricity emission factors are in the same order of magnitude as the natural gas emission factors. These are either the recent increase in the share of renewable energy in electricity generation (e.g., United Kingdom or Germany) or the transition from coal as energy source to electricity generation by natural gas (e.g., United States) (see Figure S7 in Supporting Information S1). More detailed studies are needed for these countries to definitively answer the question of which type of energy source for cooking and heating is the more climate-friendly now and in the near future.

The third group (red) is represented by eight countries where electricity is currently likely to be less climate-friendly than natural gas as the emission factor of electricity is higher than the upper bound of the emission factor of natural gas. These nine countries are characterized primarily by a fairly low share of renewables in electricity generation. Countries that use natural gas as a fossil fuel (e.g., Iran) have lower electricity emission values than countries with extensive use of coal (China, Australia, India, Indonesia, and Poland). For the countries of the third group, natural gas consumption could remain more climate-friendly compared to electricity even in the distant future. In fact, for the five countries with a high proportion of coal as an energy source large amounts of carbon emissions could be saved if natural gas were used as an energy source for end-use appliances instead, since natural gas is, in general, the more climate-friendly energy source compared to coal even if leakages are taken into account (Ladage et al., 2021).

All countries are sorted in ascending order according to their absolute carbon emission factors for electricity generation in 2019.

10.1029/2022EF002877

3.4.2. Country Comparison—Renewable Energy Gap

The results of the phase transition analysis for the 25 countries (see Section S7 in Supporting Information S1) are summarized in Figure 6. There, the 25 countries are sorted in ascending order of the percentage growth in renewable energy share required to reach the break-even point. A negative number means that electricity has a lower emission factor than the mean natural gas emission factor in 2019, so electricity is likely to be the more climate-friendly energy source for household cooking and heating in this country. However, the error bars resulting from the upper and lower bounds of the possible methane leakage rate are quite large, since the leakage rate, which is difficult to determine accurately, has a significant impact on the emission factor of natural gas.

When considering the mean leakage rate, for most of the countries, the share of renewable energy needs to be improved to make electricity a more climate-friendly energy source compared to natural gas. Only Canada, Brazil, Belgium, France, Venezuela, the United Kingdom, and Spain have already reached this point. For the other countries, the share of renewables in the overall electricity mix needs to rise further to make electricity the more climate-friendly alternative to natural gas. It should be noted that the share of renewable energy required to reach the break-even point varies greatly from country to country, depending on the energy mix used for power generation. It ranges from 0% to 67%, depending on the carbon emissions generated by non-renewable electricity generation.

3.4.3. Existing Obstacles for Such Carbon Reductions

Although, replacing natural gas with electric devices could save significant amounts of global carbon emissions, we recognize that it is not possible to immediately run all cooking and heating appliances on electricity instead of natural gas. First of all, there would not be enough electrical energy available or the electricity would have to be generated from non-renewable energy sources, which in turn would increase the carbon footprint. Furthermore, many appliances cannot be easily replaced due to the lack of electrical infrastructure. In addition, natural gas has been in most cases a significantly cheaper energy source than electricity. In Germany, for example, the price per kWh of natural gas in 2019 was only about half that of electricity (see Figure S11 in Supporting Information S1), making it unaffordable for many people to replace gas appliances with electric ones. However, such barriers could be removed by policymakers.

4. Conclusions

In this study, the climate impact of gas appliances used for cooking and heating including the effect of CH_4 leakages was investigated and compared with the carbon footprint of electric appliances. We used the Munich Oktoberfest, the largest beer festival in the world, as a case study and extended our findings to gas appliances around the world. To this end, the source signature of CH_4 enhancements at the festival was investigated utilizing a portable CH_4 gas analyzer combined with isotopic analyses of air samples to determine the δ^{13}C and δD ratios. In addition, the ethane share of the samples was examined.

Both isotopic and ethane analyses of the gas indicated that the CH_4 enhancements were predominately caused by natural gas used for cooking and heating at the festival premises and not by biogenic processes caused by visitors. Incomplete combustion and leakages in the appliances are much more likely the causes than leaks in pipelines. Since most of the cooking and heating takes place inside the beer tents, these tents are the main sources of CH_4 enhancements at Oktoberfest, which is supported by measurements inside the tents. However, food stalls on the street use natural gas driven appliances as well, so that they contribute to the overall CH_4 enhancements of the festival, too. Overall, the leakage rate at Oktoberfest 2019 is found to be 1.4%, which is slightly higher than the rate of 1.1% determined in 2018 (Chen et al., 2020).

Based on the knowledge of an existing leakage rate, we provide a possible solution to mitigate the climate impact of such large festivals by calculating the carbon footprint of natural gas driven appliances considering the leakage rate. Although, natural gas is considered a fairly climate friendly alternative to other fossil fuels, we found that electrical appliances at Oktoberfest have a much smaller carbon footprint than natural gas driven ones, since Oktoberfest is supplied by renewable electricity only. Replacing all natural gas driven appliances at Oktoberfest with electrical ones could have saved approximately 450 t of CO_2eq in 2019, equivalent to 87% of the carbon emissions caused by energy consumption on the festival premises.

Nevertheless, carbon emissions of Oktoberfest contribute only very little to the global carbon budget, making emission reductions at Oktoberfest not a solution to global climate problems. However, gas appliances are used not only at Oktoberfest but in many households around the world. Therefore, we extended our study to estimate whether replacing gas driven appliances with electric ones in specific countries would save global carbon emissions.

Since electricity is generated by different energy sources in each country, the carbon footprint of electricity generation differs significantly between them. To date, only in seven of these countries, electricity is likely the more climate-friendly energy source than natural gas for cooking and heating in the household sector. However, since the share of renewables is steadily increasing in many countries, electricity could become the more climate friendly energy source than natural gas in the near future.

We conclude that from a climate perspective, in countries with low carbon emissions from electricity generation, it would make sense now or in a few years to replace gas appliances for domestic cooking and heating with electric appliances to save overall carbon emissions. Nevertheless, we are aware of the fact that not all gas appliances worldwide can be replaced by electric appliances, especially since there would not be enough electrical energy available or the electricity would have to be generated from non-renewable energy sources, which in turn would increase the CO_2 footprint. Therefore, the share of renewable energies in electricity generation must be further increased. In addition, many countries around the world lack electrical infrastructure, and natural gas is the cheaper energy source compared to electricity in many countries, making it uneconomical for end users to switch from gas to electricity. However, some of these problems are more political in nature and could be solved by governments. In this study, we aim to rise people's awareness of how carbon emissions from electric and gas-powered end-use appliances compare, and identified an option that could reduce a significant amount of carbon emissions in the near future.

Data Availability Statement

The measurement data and scripts used for the Oktoberfest study is preserved at https://doi.org/10.14459/2022mp1663551, available via CC BY 4.0 license and developed openly at https://github.com/ankitshekhar99/Oktoberfest2019Study/tree/main (Dietrich et al., 2022).

Acknowledgments

We thank LI-COR Biosciences for the loan of the mobile gas analyzer; Pim van den Bulk (TNO) for helping with the QCL analyses; our students Xiao Bi and Xinxu Zhao for helping us with the measurements. This research has been supported by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) Grant CH 1792/2-1. Jia Chen is partially supported by the Technical University of Munich – Institute for Advanced Study, funded by the German Excellence Initiative and the European Union Seventh Framework Programme under grant agreement number 291763. Ankit Shekhar is supported by ETH Zürich project FEVER ETH-27 19-1. We would also like to thank the reviewers and the editor for their very helpful comments, which helped to improve the paper. Open Access funding enabled and organized by Projekt DEAL.

References

Adria, O., & Bethge, J. (2013). *What users can save with energy-efficient cooking stoves and ovens* (pp. 1–30). bigee.net - Wuppertal Institute for Climate, Environment and Energy. Retrieved from https://energypedia.info/images/2/26/Bigee_cookingstoves_user_savings.pdf

Allen, D. T., Torres, V. M., Thomas, J., Sullivan, D. W., Harrison, M., Hendler, A., et al. (2013). Measurements of methane emissions at natural gas production sites in the United States. *Proceedings of the National Academy of Sciences*, *110*(44), 17768–17773. https://doi.org/10.1073/pnas.1304880110

Alvarez, R. A., Zavala-Araiza, D., Lyon, D. R., Allen, D. T., Barkley, Z. R., Brandt, A. R., et al. (2018). Assessment of methane emissions from the U.S. oil and gas supply chain. *Science*, *361*(6398), 186–188. https://doi.org/10.1126/science.aar7204

Amponsah, N. Y., Troldborg, M., Kington, B., Aalders, I., & Hough, R. L. (2014). Greenhouse gas emissions from renewable energy sources: A review of lifecycle considerations. *Renewable and Sustainable Energy Reviews*, *39*(C), 461–475. https://doi.org/10.1016/j.rser.2014.07.087

Beck, V., Chen, H., Gerbig, C., Bergamaschi, P., Bruhwiler, L., Houweling, S., et al. (2012). Methane airborne measurements and comparison to global models during BARCA. *Journal of Geophysical Research*, *117*(D15). https://doi.org/10.1029/2011JD017345

bp (2020). Statistical review of world energy 2020. *bp Statistical Review of World Energy 2020*, *69*, 68. Retrieved from https://www.bp.com/content/dam/bp/business-sites/en/global/corporate/pdfs/energy-economics/statistical-review/bp-stats-review-2020-full-report.pdf

Brass, M., & Röckmann, T. (2010). Continuous-flow isotope ratio mass spectrometry method for carbon and hydrogen isotope measurements on atmospheric methane. *Atmospheric Measurement Techniques*, *3*(6), 1707–1721. https://doi.org/10.5194/amt-3-1707-2010

Chamberlain, S. D., Ingraffea, A. R., & Sparks, J. P. (2016). Sourcing methane and carbon dioxide emissions from a small city: Influence of natural gas leakage and combustion. *Environmental Pollution*, *218*, 102–110. https://doi.org/10.1016/j.envpol.2016.08.036

Chen, J., Dietrich, F., Maazallahi, H., Forstmaier, A., Winkler, D., Hofmann, M. E. G., et al. (2020). Methane emissions from the Munich Oktoberfest. *Atmospheric Chemistry and Physics*, *20*(6), 3683–3696. https://doi.org/10.5194/acp-20-3683-2020

Chen, J., Viatte, C., Hedelius, J. K., Jones, T., Franklin, J. E., Parker, H., et al. (2016). Differential column measurements using compact solar-tracking spectrometers. *Atmospheric Chemistry and Physics*, *16*(13), 8479–8498. https://doi.org/10.5194/acp-16-8479-2016

Dietrich, F., Chen, J., Shekhar, A., Lober, S., Krämer, K., Leggett, G., et al. (2022). Data sets and modeling software as part of our study titled "Climate impact comparison of electric and gas-powered end-user appliances" [Dataset]. Technical University of Munich. https://doi.org/10.14459/2022MP1663551

Dietrich, F., Chen, J., Voggenreiter, B., Aigner, P., Nachtigall, N., & Reger, B. (2021). MUCCnet: Munich urban carbon column network. *Atmospheric Measurement Techniques*, *14*(2), 1111–1126. https://doi.org/10.5194/amt-14-1111-2021

Fisher, R., Lowry, D., Wilkin, O., Sriskantharajah, S., & Nisbet, E. G. (2006). High-precision, automated stable isotope analysis of atmospheric methane and carbon dioxide using continuous-flow isotope-ratio mass spectrometry. *Rapid Communications in Mass Spectrometry*, *20*(2), 200–208. https://doi.org/10.1002/rcm.2300

Fulton, M., Mellquist, N., & Kitasei, S. (2011). *Comparing life cycle greenhouse gas emissions from natural gas and coal*. Worldwatch Institute.

Gallagher, M. E., Down, A., Ackley, R. C., Zhao, K., Phillips, N., & Jackson, R. B. (2015). Natural gas pipeline replacement programs reduce methane leaks and improve consumer safety. *Environmental Science and Technology Letters*, *2*(10), 286–291. https://doi.org/10.1021/acs.estlett.5b00213

Gioli, B., Toscano, P., Lugato, E., Matese, A., Miglietta, F., Zaldei, A., & Vaccari, F. P. (2012). Methane and carbon dioxide fluxes and source partitioning in urban areas: The case study of Florence, Italy. *Environmental Pollution*, *164*, 125–131. https://doi.org/10.1016/j.envpol.2012.01.019

Hager, T. J., & Morawicki, R. (2013). Energy consumption during cooking in the residential sector of developed nations: A review. *Food Policy*, *40*, 54–63. https://doi.org/10.1016/j.foodpol.2013.02.003

Hase, F., Frey, M., Blumenstock, T., Groß, J., Kiel, M., Kohlhepp, R., et al. (2015). Application of portable FTIR spectrometers for detecting greenhouse gas emissions of the major city Berlin. *Atmospheric Measurement Techniques*, *8*(7), 3059–3068. https://doi.org/10.5194/amt-8-3059-2015

Helfter, C., Tremper, A. H., Halios, C. H., Kotthaus, S., Bjorkegren, A., Grimmond, C. S. B., et al. (2016). Spatial and temporal variability of urban fluxes of methane, carbon monoxide and carbon dioxide above London, UK. *Atmospheric Chemistry and Physics*, *16*(16), 10543–10557. https://doi.org/10.5194/acp-16-10543-2016

Hmiel, B., Petrenko, V. V., Dyonisius, M. N., Buizert, C., Smith, A. M., Place, P. F., et al. (2020). Preindustrial 14 CH_4 indicates greater anthropogenic fossil CH_4 emissions. *Nature*, *578*(7795), 409–412. https://doi.org/10.1038/s41586-020-1991-8

International Renewable Energy Agency. (2020). Statistics time series. Retrieved from https://www.irena.org/Statistics/View-Data-by-Topic/Capacity-and-Generation/Statistics-Time-Series

Jackson, R. B., Down, A., Phillips, N. G., Ackley, R. C., Cook, C. W., Plata, D. L., & Zhao, K. (2014). Natural gas pipeline leaks across Washington, DC. *Environmental Science & Technology*, *48*(3), 2051–2058. https://doi.org/10.1021/es404474x

Jones, T. S., Franklin, J. E., Chen, J., Dietrich, F., Hajny, K. D., Paetzold, J. C., et al. (2021). Assessing urban methane emissions using column-observing portable Fourier transform infrared (FTIR) spectrometers and a novel Bayesian inversion framework. *Atmospheric Chemistry and Physics*, *21*, 13131–13147. https://doi.org/10.5194/acp-21-13131-2021

Karion, A., Sweeney, C., Pétron, G., Frost, G., Hardesty, R. M., Kofler, J., et al. (2013). Methane emissions estimate from airborne measurements over a Western United States natural gas field. *Geophysical Research Letters*, *40*(16), 4393–4397. https://doi.org/10.1002/grl.50811

Keeling, C. D. (1958). The concentration and isotopic abundances of atmospheric carbon dioxide in rural areas. *Geochimica et Cosmochimica Acta*, *13*(4), 322–334. https://doi.org/10.1016/0016-7037(58)90033-4

Klappenbach, F., Chen, J., Wenzel, A., Forstmaier, A., Dietrich, F., Zhao, X., & Fischer, M. (2021). Methane emission estimate using ground based remote sensing in complex terrain (Tech. Rep. No. EGU21-15406). *Copernicus Meetings*. https://doi.org/10.5194/egusphere-egu21-15406

Ladage, S., Blumenberg, M., Franke, D., Bahr, A., Lutz, R., & Schmidt, S. (2021). On the climate benefit of a coal-to-gas shift in Germany's electric power sector. *Scientific Reports*, *11*(1), 11453. https://doi.org/10.1038/s41598-021-90839-7

Landeshauptstadt München. (2019). Ratsinformationssystem der Landeshauptstadt München [text]. Retrieved from https://www.ris-muenchen.de/RII/RII/ris_fulltextsrch.jsp

Landeshauptstadt München Redaktion. (2020). M-Statistik München - Historische Berichte. Retrieved from https://www.mstatistik-muenchen.de/

Lebel, E. D., Finnegan, C. J., Ouyang, Z., & Jackson, R. B. (2022). Methane and nox emissions from natural gas stoves, cooktops, and ovens in residential homes. *Environmental Science & Technology*, *56*(4), 2529–2539. https://doi.org/10.1021/acs.est.1c04707

Lu, X., Harris, S. J., Fisher, R. E., France, J. L., Nisbet, E. G., Lowry, D., et al. (2021). Isotopic signatures of major methane sources in the coal seam gas fields and adjacent agricultural districts, Queensland, Australia. *Atmospheric Chemistry and Physics*, *21*(13), 10527–10555. https://doi.org/10.5194/acp-21-10527-2021

Lyon, D. R., Alvarez, R. A., Zavala-Araiza, D., Brandt, A. R., Jackson, R. B., & Hamburg, S. P. (2016). Aerial surveys of elevated hydrocarbon emissions from oil and gas production sites. *Environmental Science & Technology*, *50*(9), 4877–4886. https://doi.org/10.1021/acs.est.6b00705

Maazallahi, H., Fernandez, J. M., Menoud, M., Zavala-Araiza, D., Weller, Z. D., Schwietzke, S., et al. (2020). Methane mapping, emission quantification, and attribution in two European cities: Utrecht (NL) and Hamburg (DE). *Atmospheric Chemistry and Physics*, *20*(23), 14717–14740. https://doi.org/10.5194/acp-20-14717-2020

Makarova, M. V., Alberti, C., Ionov, D. V., Hase, F., Foka, S. C., Blumenstock, T., et al. (2021). Emission Monitoring Mobile Experiment (EMME): An overview and first results of the St. Petersburg megacity campaign 2019. *Atmospheric Measurement Techniques*, *14*(2), 1047–1073. https://doi.org/10.5194/amt-14-1047-2021

McKain, K., Down, A., Raciti, S. M., Budney, J., Hutyra, L. R., Floerchinger, C., et al. (2015). Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts. *Proceedings of the National Academy of Sciences*, *112*(7), 1941–1946. https://doi.org/10.1073/pnas.1416261112

Menoud, M., van der Veen, C., Necki, J., Bartyzel, J., Szénási, B., Stanisavljević, M., et al. (2021). Methane (CH_4) sources in Krakow, Poland: Insights from isotope analysis. *Atmospheric Chemistry and Physics*, *21*, 13167–13185. https://doi.org/10.5194/acp-21-13167-2021

Menoud, M., van der Veen, C., Scheeren, B., Chen, H., Szénási, B., Morales, R. P., et al. (2020). Characterisation of methane sources in Lutjewad, The Netherlands, using quasi-continuous isotopic composition measurements. *Tellus B: Chemical and Physical Meteorology*, *72*(1), 1–20. https://doi.org/10.1080/16000889.2020.1823733

Mitchell, A. L., Tkacik, D. S., Roscioli, J. R., Herndon, S. C., Yacovitch, T. I., Martinez, D. M., et al. (2015). Measurements of methane emissions from natural gas gathering facilities and processing plants: Measurement results. *Environmental Science & Technology*, *49*(5), 3219–3227. https://doi.org/10.1021/es5052809

Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestvedt, J., Huang, J., et al. (2013). Anthropogenic and natural radiative forcing. In T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex, & P. M. Midgley (Eds.), *Climate change 2013: The physical science basis. Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change* (pp. 659–740). Cambridge University Press. https://doi.org/10.1017/CBO9781107415324.018

Nisbet, E. G., Manning, M. R., Dlugokencky, E. J., Fisher, R. E., Lowry, D., Michel, S. E., et al. (2019). Very strong atmospheric methane growth in the 4 years 2014–2017: Implications for the Paris agreement. *Global Biogeochemical Cycles*, *33*(3), 318–342. https://doi.org/10.1029/2018GB006009

Omara, M., Sullivan, M. R., Li, X., Subramanian, R., Robinson, A. L., & Presto, A. A. (2016). Methane emissions from conventional and unconventional natural gas production sites in the Marcellus Shale Basin. *Environmental Science & Technology*, *50*(4), 2099–2107. https://doi.org/10.1021/acs.est.5b05503

Phillips, N. G., Ackley, R., Crosson, E. R., Down, A., Hutyra, L. R., Brondfield, M., et al. (2013). Mapping urban pipeline leaks: Methane leaks across Boston. *Environmental Pollution*, *173*, 1–4. https://doi.org/10.1016/j.envpol.2012.11.003

Plant, G., Kort, E. A., Floerchinger, C., Gvakharia, A., Vimont, I., & Sweeney, C. (2019). Large fugitive methane emissions from urban centers along the U.S. East Coast. *Geophysical Research Letters*, *46*(14), 8500–8507. https://doi.org/10.1029/2019GL082635

Qin, Y., Edwards, R., Tong, F., & Mauzerall, D. L. (2017). Can switching from coal to shale gas bring net carbon reductions to China? *Environmental Science & Technology*, *51*(5), 2554–2562. https://doi.org/10.1021/acs.est.6b04072

Röckmann, T., Eyer, S., van der Veen, C., Popa, M. E., Tuzson, B., Monteil, G., et al. (2016). In situ observations of the isotopic composition of methane at the Cabauw tall tower site. *Atmospheric Chemistry and Physics*, *16*(16), 10469–10487. https://doi.org/10.5194/acp-16-10469-2016

Roscioli, J. R., Yacovitch, T. I., Floerchinger, C., Mitchell, A. L., Tkacik, D. S., Subramanian, R., et al. (2015). Measurements of methane emissions from natural gas gathering facilities and processing plants: Measurement methods. *Atmospheric Measurement Techniques*, *8*(5), 2017–2035. https://doi.org/10.5194/amt-8-2017-2015

Sargent, M. R., Floerchinger, C., McKain, K., Budney, J., Gottlieb, E. W., Hutyra, L. R., et al. (2021). Majority of US urban natural gas emissions unaccounted for in inventories. *Proceedings of the National Academy of Sciences*, *118*(44). https://doi.org/10.1073/pnas.2105804118

Schwietzke, S., Sherwood, O. A., Bruhwiler, L. M. P., Miller, J. B., Etiope, G., Dlugokencky, E. J., et al. (2016). Upward revision of global fossil fuel methane emissions based on isotope database. *Nature*, *538*(7623), 88–91. https://doi.org/10.1038/nature19797

SWM Infrastruktur GmbH und Co. KG. (2019a). Erdgasbeschaffenheit: Monatsdurchschnitt Oktober 2019. Retrieved from https://www.swm-infrastruktur.de/dam/swm-infrastruktur/dokumente/gas/netzstrukturdaten/gasanalyse-oktober-2019.pdf

SWM Infrastruktur GmbH und Co. KG. (2019b). Erdgasbeschaffenheit: Monatsdurchschnitt September 2019. Retrieved from https://www.swm-infrastruktur.de/dam/swm-infrastruktur/dokumente/gas/netzstrukturdaten/gasanalyse-september-2019.pdf

Tanaka, K., Cavalett, O., Collins, W. J., & Cherubini, F. (2019). Asserting the climate benefits of the coal-to-gas shift across temporal and spatial scales. *Nature Climate Change*, *9*(5), 389–396. https://doi.org/10.1038/s41558-019-0457-1

Vogel, F. R., Frey, M., Staufer, J., Hase, F., Broquet, G., Xueref-Remy, I., et al. (2019). XCO_2 in an emission hot-spot region: The COCCON Paris campaign 2015. *Atmospheric Chemistry and Physics*, *19*(5), 3271–3285. https://doi.org/10.5194/acp-19-3271-2019

von Fischer, J. C., Cooley, D., Chamberlain, S., Gaylord, A., Griebenow, C. J., Hamburg, S. P., et al. (2017). Rapid, vehicle-based identification of location and magnitude of urban natural gas pipeline leaks. *Environmental Science & Technology*, *51*(7), 4091–4099. https://doi.org/10.1021/acs.est.6b06095

Weller, Z. D., Hamburg, S. P., & von Fischer, J. C. (2020). A national estimate of methane leakage from pipeline mains in natural gas local distribution systems. *Environmental Science & Technology*, *54*(14), 8958–8967. https://doi.org/10.1021/acs.est.0c00437

Weller, Z. D., Roscioli, J. R., Daube, W. C., Lamb, B. K., Ferrara, T. W., Brewer, P. E., & von Fischer, J. C. (2018). Vehicle-based methane surveys for finding natural gas leaks and estimating their size: Validation and uncertainty. *Environmental Science & Technology*, *52*(20), 11922–11930. https://doi.org/10.1021/acs.est.8b03135

WorldData.info. (2020). Energy consumption in Germany. Retrieved from https://www.worlddata.info/europe/germany/energy-consumption.php

Yacovitch, T. I., Herndon, S. C., Pétron, G., Kofler, J., Lyon, D., Zahniser, M. S., & Kolb, C. E. (2015). Mobile laboratory observations of methane emissions in the Barnett Shale region. *Environmental Science & Technology*, *49*(13), 7889–7895. https://doi.org/10.1021/es506352j

Yacovitch, T. I., Herndon, S. C., Roscioli, J. R., Floerchinger, C., McGovern, R. M., Agnese, M., et al. (2014). Demonstration of an ethane spectrometer for methane source identification. *Environmental Science & Technology*, *48*(14), 8028–8034. https://doi.org/10.1021/es501475q

Yacovitch, T. I., Neininger, B., Herndon, S. C., van der Gon, H. D., Jonkers, S., Hulskotte, J., et al. (2018). Methane emissions in The Netherlands: The Groningen field. *Elementa: Science of the Anthropocene*, *6*(57). https://doi.org/10.1525/elementa.308

Zavala-Araiza, D., Lyon, D. R., Alvarez, R. A., Davis, K. J., Harriss, R., Herndon, S. C., et al. (2015). Reconciling divergent estimates of oil and gas methane emissions. *Proceedings of the National Academy of Sciences*, *112*(51), 15597–15602. https://doi.org/10.1073/pnas.1522126112

Zhao, X., Marshall, J., Hachinger, S., Gerbig, C., Frey, M., Hase, F., & Chen, J. (2019). Analysis of total column CO_2 and CH_4 measurements in Berlin with WR_F-GHG. *Atmospheric Chemistry and Physics*, *19*(17), 11279–11302. https://doi.org/10.5194/acp-19-11279-2019

Zimmerle, D. J., Williams, L. L., Vaughn, T. L., Quinn, C., Subramanian, R., Duggan, G. P., et al. (2015). Methane emissions from the natural gas transmission and storage system in the United States. *Environmental Science & Technology*, *49*(15), 9374–9383. https://doi.org/10.1021/acs.est.5b01669

Zimnoch, M., Necki, J., Chmura, L., Jasek, A., Jelen, D., Galkowski, M., et al. (2019). Quantification of carbon dioxide and methane emissions in urban areas: Source apportionment based on atmospheric observations. *Mitigation and Adaptation Strategies for Global Change*, *24*(6), 1051–1071. https://doi.org/10.1007/s11027-018-9821-0