Exploring potential impacts from transitions in German and European energy on GHG and air pollutant emissions and on ozone air quality

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"Believe in the power of your own voice. The more noise you make, the more accountability you demand from your leaders, the more our world will change for the better." – Al Gore

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Abstract

Energy is at the heart of the climate crisis—but also at the heart of any efforts for climate change mitigation. Energy consumption is namely responsible for approximately three quarters of global anthropogenic greenhouse gas (GHG) emissions. Therefore, central to any serious plans to stave off a climate catastrophe is a major transformation of the world's energy system, which would move society away from fossil fuels and towards a net-zero energy future. Considering that fossil fuels are also a major source of air pollutant emissions, the energy transition has important implications for air quality as well, and thus also for human and environmental health. Both Europe and Germany have set the goal of becoming GHG neutral by 2050, and moreover have demonstrated their deep commitment to a comprehensive energy transition. Two of the most significant developments in energy policy over the past decade have been the interest in expansion of shale gas and hydrogen, which accordingly have garnered great interest and debate among public, private and political actors.

In this context, sound scientific information can play an important role by informing stakeholder dialogue and future research investments, and by supporting evidence-based decision-making. This thesis examines anticipated environmental impacts from possible, relevant changes in the European energy system, in order to impart valuable insight and fill critical gaps in knowledge. Specifically, it investigates possible future shale gas development in Germany and the United Kingdom (UK), as well as a hypothetical, complete transition to hydrogen mobility in Germany. Moreover, it assesses the impacts on GHG and air pollutant emissions, and on tropospheric ozone (O_3) air quality. The analysis is facilitated by constructing emission scenarios and performing air quality modeling via the Weather Research and Forecasting model coupled with chemistry (WRF-Chem). The work of this thesis is presented in three research papers.

The first paper finds that methane (CH_4) leakage rates from upstream shale gas development in Germany and the UK would range between 0.35 % and 1.36 % in a realistic, business-as-usual case, while they would be significantly lower – between 0.08 % and 0.15 % – in an optimistic, strict regulation and high compliance case, thus demonstrating the value and potential of measures to substantially reduce emissions. Yet, while the optimistic case is technically feasible, it is unlikely that the practices and technologies assumed would be applied and accomplished on a systematic, regular basis, owing to economics and limited monitoring resources. The realistic CH_4 leakage rates estimated in this study are comparable to values reported by studies carried out in the US and elsewhere. In contrast, the optimistic rates are similar to official CH_4 leakage data from upstream gas production

in Germany and in the UK. Considering that there is a lack of systematic, transparent and independent reports supporting the official values, this study further highlights the need for more research efforts in this direction. Compared with national energy sector emissions, this study suggests that shale gas emissions of volatile organic compounds (VOCs) could be significant, though relatively insignificant for other air pollutants. Similar to CH_4, measures could be effective for reducing VOCs emissions.

The second paper shows that VOC and nitrogen oxides (NO_x) emissions from a future shale gas industry in Germany and the UK have potentially harmful consequences for European O_3 air quality on both the local and regional scale. The results indicate a peak increase in maximum daily 8-hour average O_3 (MDA8) ranging from $3.7\,\mu g\,m^{-3}$ to $28.3\,\mu g\,m^{-3}$. Findings suggest that shale gas activities could result in additional exceedances of MDA8 at a substantial percentage of regulatory measurement stations both locally and in neighboring and distant countries, with up to circa one third of stations in the UK and one fifth of stations in Germany experiencing additional exceedances. Moreover, the results reveal that the shale gas impact on the cumulative health-related metric SOMO35 (annual Sum of Ozone Means Over 35 ppb) could be substantial, with a maximum increase of circa 28 %. Overall, the findings suggest that shale gas VOC emissions could play a critical role in O_3 enhancement, while NO_x emissions would contribute to a lesser extent. Thus, the results indicate that stringent regulation of VOC emissions would be important in the event of future European shale gas development to minimize deleterious health outcomes.

The third paper demonstrates that a hypothetical, complete transition of the German vehicle fleet to hydrogen fuel cell technology could contribute substantially to Germany's climate and air quality goals. The results indicate that if the hydrogen were to be produced via renewable-powered water electrolysis (green hydrogen), German carbon dioxide equivalent (CO_2eq) emissions would decrease by $179\,MtCO_2eq$ annually, though if electrolysis were powered by the current electricity mix, emissions would instead increase by $95\,MtCO_2eq$ annually. The findings generally reveal a notable anticipated decrease in German energy emissions of regulated air pollutants. The results suggest that vehicular hydrogen demand is 1000 PJ annually, which would require between 446 TWh and 525 TWh for electrolysis, hydrogen transport and storage. When only the heavy duty vehicle segment (HDVs) is shifted to green hydrogen, the results of this thesis show that vehicular hydrogen demand drops to 371 PJ, while a deep emissions cut is still realized ($-57\,MtCO_2eq$), suggesting that HDVs are a low-hanging fruit for contributing to decarbonization of the German road transport sector with hydrogen energy.

Zusammenfassung

Energie ist der Kern der Klimakrise—aber auch der Kern jeglicher Bemühungen zur Eindämmung des Klimawandels. Der Energieverbrauch ist heute für ungefähr drei Viertel der weltweiten Treibhausgasemissionen verantwortlich. Grundlegend für einen ernsthaft gemeinten Plan eine Klimakatastrophe abzuwenden ist daher eine umfassende Umgestaltung des weltweiten Energiesystems von fossilen Brennstoffen weg in Richtung zukünftige Netto-Null-Emissionen. Angesichts der Tatsache, dass fossile Brennstoffe auch eine Hauptquelle für Luftschadstoffemissionen sind, hat die Energiewende wichtige Auswirkungen auf die Luftqualität und damit auch auf die Gesundheit von Mensch und Umwelt. Sowohl Europa als auch Deutschland haben sich zum Ziel gesetzt, bis 2050 treibhausgasneutral zu werden und zeigen darüber hinaus ihr tiefes Engagement für eine umfassende Energiewende. Zwei der wichtigsten Entwicklungen in der Energiepolitik im letzten Jahrzehnt waren das Interesse an der Ausweitung von Schiefergas und Wasserstoff, das entsprechend großes Interesse und große Diskussionen in der Öffentlichkeit, im Privaten und in der Politik erzeugt hat.

In diesem Zusammenhang können fundierte wissenschaftliche Informationen eine wichtige Rolle spielen, indem sie Interessenvertreter und zukünftige Forschungsinvestitionen informieren und evidenzbasierte Entscheidungen unterstützen. Diese Doktorarbeit untersucht die Umweltauswirkungen möglicher, relevanter Veränderungen im europäischen Energiesystem, um wertvolle Erkenntnisse zu vermitteln und kritische Wissenslücken zu schließen. Insbesondere werden mögliche zukünftige Schiefergasentwicklungen in Deutschland und im Vereinigten Königreich (UK) sowie ein hypothetischer, vollständiger Übergang zur Wasserstoffmobilität in Deutschland untersucht. Darüber hinaus werden die Auswirkungen auf die Treibhausgas- und Luftschadstoffemissionen sowie auf die Luftqualität von troposphärischem Ozon (O_3) bewertet. Die Analyse wird durch die Erstellung von Emissionsszenarien und die Durchführung von Luftqualitätsmodellen über die Chemie-Version des „Weather Research and Forecasting Model" (WRF-Chem) erleichtert. Die Forschung dieser Doktorarbeit wird in drei wissenschaftlichen Artikeln vorgestellt.

Der erste Artikel beschreibt, dass die Methan (CH_4)-Leckraten aus einer vorgelagerten Schiefergasproduktion in Deutschland und Großbritannien in einem gewöhnlichen Fall zwischen 0,35 % und 1,36 % liegen würden, während sie in einem optimistischen, streng regulierten Fall signifikant zwischen 0,08 % und 0,15 % niedriger wären, und zeigt damit die Bedeutung und das Potenzial von Maßnahmen zur wesentlichen Reduzierung der Emissionen auf. Obwohl der optimistische Fall technisch machbar ist, ist es aufgrund

der Wirtschaftlichkeit und der begrenzten Überwachungsressourcen unwahrscheinlich, dass die angenommenen Praktiken und Technologien systematisch und regelmäßig angewendet und durchgeführt werden. Die in dieser Studie geschätzten realistischen CH_4-Leckraten sind vergleichbar mit Werten, die in Studien in den USA und anderswo angegeben wurden. Im Gegensatz dazu ähneln die optimistischen Raten den offiziellen CH_4-Leckraten aus der vorgelagerten Gasproduktion in Deutschland und Großbritannien. In Anbetracht des Mangels an systematischen, transparenten und unabhängigen Berichten, die die offiziellen Werte stützen, unterstreicht diese Studie die Notwendigkeit weiterer Forschungsanstrengungen in diese Richtung. Im Vergleich zu den Emissionen des nationalen Energiesektors deutet diese Studie darauf hin, dass die Schiefergasemissionen flüchtiger organischer Verbindungen (VOC) erheblich sein könnten, andere Luftschadstoffe jedoch relativ unbedeutend bleiben. Ähnlich wie bei CH_4 könnten Maßnahmen zur Reduzierung der VOC-Emissionen wirksam sein.

Der zweite Artikel beschreibt, dass VOC- und Stickoxidemissionen (NO_x) einer zukünftigen Schiefergasindustrie in Deutschland und Großbritannien potenziell schädliche Folgen für die europäische O_3-Luftqualität sowohl auf lokaler als auch auf regionaler Ebene haben. Die Ergebnisse zeigen einen Spitzenanstieg des maximalen täglichen 8-Stunden-Durchschnitts von O_3 (MDA8) im Bereich von $3{,}7\,\mu g\,m^{-3}$ bis $28{,}3\,\mu g\,m^{-3}$. Die Ergebnisse deuten darauf hin, dass Schiefergasaktivitäten zu zusätzlichen Grenzwertüberschreitungen des MDA8 bei einem erheblichen Prozentsatz der regulatorischen Messstationen sowohl vor Ort als auch in Nachbar- und entfernten Ländern führen können, wobei bei bis zu etwa einem Drittel der Stationen in Großbritannien und einem Fünftel der Stationen in Deutschland zusätzliche Überschreitungen auftreten. Darüber hinaus zeigen die Ergebnisse, dass die Auswirkungen von Schiefergas auf die kumulative gesundheitsbezogene Metrik SOMO35 (jährliche Summe des Ozonmittel über 35 ppb) mit einem maximalen Anstieg von ca. 28 % erheblich sein könnten. Insgesamt deuten die Ergebnisse darauf hin, dass die VOC-Emissionen von Schiefergas eine entscheidende Rolle bei der O_3-Erhöhung spielen könnten, während die NO_x-Emissionen in geringerem Maße dazu beitragen würden. Unsere Ergebnisse zeigen daher, dass eine strenge Regulierung der VOC-Emissionen im Falle einer künftigen europäischen Schiefergasentwicklung wichtig ist, um schädliche gesundheitliche Folgen zu minimieren.

Der dritte Artikel beschreibt, dass ein hypothetischer, vollständiger Übergang der deutschen Fahrzeugflotte zur Wasserstoff-Brennstoffzellentechnologie wesentlich zu Deutschlands Klima- und Luftqualitätszielen beitragen kann. Die Ergebnisse deuten darauf hin, dass bei einer Erzeugung des Wasserstoffs durch erneuerbare Wasserelektrolyse (grüner Wasserstoff) die Emissionen des deutschen Kohlendioxidäquivalents (CO_2eq) jährlich um $179\,MtCO_2eq$ sinken würden. Wenn die Elektrolyse jedoch mit dem aktuellen Strommix betrieben würde, würden sich die Emissionen stattdessen um jährlich $95\,MtCO_2eq$ erhöhen. Die Ergebnisse zeigen im Allgemeinen einen bemerkenswerten Rückgang der Luftschadstoffemissionen in Deutschland. Weiterhin legen sie nahe, dass der Wasserstoffbedarf von Fahrzeugen 1000 PJ pro Jahr beträgt, was zwischen

446 TWh und 525 TWh für Elektrolyse, Wasserstofftransport und -speicherung erfordern würde. Wenn nur das Segment der Schwerlastfahrzeuge (HDVs) auf grünen Wasserstoff umgestellt wird, zeigen unsere Ergebnisse, dass der Wasserstoffbedarf der Fahrzeuge auf 371 PJ sinkt, während immer noch eine tiefgreifende Emissionsreduzierung erzielt wird ($-57\,\mathrm{MtCO_2eq}$). Dies zeigt, dass die Umstellung der HDVs auf grünen Wasserstoff einen entscheidenden Beitrag zur Dekarbonisierung des deutschen Straßenverkehrs leisten kann.

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Abbreviations

AD	activity data
BADC	British Atmospheric Data Center
BC	boundary condition
bcm	billion cubic meter
BEV	battery electric vehicle
BGR	German Federal Institute for Geosciences and Natural Resources
BGS	British Geological Survey
BIGALK	$C > 3$ alkanes
conNO_x	concentrated NO_x scenario
CCS	carbon capture and storage
CG	coal gasification
CH_4	methane
CO	carbon monoxide
CO_2	carbon dioxide
CO_2eq	carbon dioxide equivalent
DE	Germany
EEA	European Environmental Agency
EF	emission factor
EI	emission intensity
EPA	US Environmental Protection Agency
EU	European Union
FCEV	fuel cell electric vehicle
GHG	greenhouse gas
GIP	gas-in-place
GWP	global warming potential
HDV	heavy duty vehicle
ICEV	internal combustion engine vehicle
IEA	International Energy Agency
IPCC	Intergovernmental Panel on Climate Change
JJA	June, July, August
LDV	light duty vehicle
LHV	lower heating value
MB	mean bias

MFB	mean fractional bias
MDA8	maximum daily 8-hour average O_3
MSLP	mean sea-level pressure
NIR	National Inventory Report
NMB	normalized mean bias
NMVOCs	non-methane volatile organic compounds
NO_x	nitrogen oxides
O_3	ozone
OEm	Optimistic emission scenario
PC	passenger car
P25/P50/P75	well productivity at the $25^{th}/50^{th}/75^{th}$ percentile of exceedance
PEM	proton exchange membrane
$PM/PM_{10}/PM_{2.5}$	particulate matter, $\leq 10\,\mu m$ and $2.5\,\mu m$ in diameter, respectively
r	temporal correlation coefficient
REC	Reduced Emissions Completion
REm	Realistic Emission scenario
SG1/SG2/SG3	scenario sets covering low, medium and high shale gas emissions
SMR	steam methane reforming
SOEC	solid oxide electrolysis cells
SOMO35	annual Sum of Ozone Means Over 35 ppb
tcm	trillion cubic meters
TRR	technically recoverable resource
TTW	tank-to-wheel
T2	2m temperature
UBA	German Environment Agency
UNFCCC	United Nation Framework Convention on Climate Change
US	United States
UK	United Kingdom
VOCs	volatile organic compounds
WD10	10m wind direction
WHO	World Health Organization
WS10	10m wind speed
WRF-Chem	Weather Research and Forecasting model coupled with chemistry

1 Introduction

1.1 Background and motivation

Climate change is one of the greatest challenges that humanity has ever faced. No longer is it a hypothetical situation of the future, climate change is already happening: the average global surface temperature has increased thus far by around 1 °C above pre-industrial levels (IPCC, 2018), ice and snow cover are diminishing, sea levels are rising, ocean temperature and acidity are increasing, and extreme weather events are becoming more common (IPCC, 2013). Many of the climatic changes we are experiencing are unprecedented over many millennia (IPCC, 2013), and have significant, dangerous consequences for biodiversity, ecosystems and human society. It is now essentially unrefuted scientifically that human activity has been the principal cause behind the warming increase since the mid-20$^{\text{th}}$ century, mainly through the vast emissions of greenhouse gases (GHGs) to the atmosphere (IPCC, 2013). Other anthropogenic forcings, such as land use change and black carbon contamination of ice and snow, are also contributing to climate change by altering Earth's albedo (surface reflectivity of solar energy). Radiative forcing quantifies the difference in the radiative energy balance of the Earth-atmosphere system relative to 1750. Radiative forcing can be both positive (warming the Earth's surface) and negative (cooling); the net anthropogenic radiative forcing is positive, resulting in the global warming that the Earth is experiencing. The most significant GHG contributing to climate change in terms of radiative forcing is carbon dioxide (CO_2), followed by methane (CH_4) (Myhre et al., 2013). A recent study found that atmospheric CO_2 is now at a level that is approximately as high as at any time since at least 3.3 million years ago—a period associated with global temperatures considerably higher than today (by about 2 °C to 3 °C during the Piacenzian, 2.6 Ma to 3.6 Ma) (de la Vega, Chalk, Wilson, Bysani, & Foster, 2020).

On 12 December 2015, 195 nations adopted the Paris Agreement, the first ever, universal climate change agreement with the target to limit the rise in global average temperature to well below 2 °C above pre-industrial levels, and to pursue efforts to limit it even further to 1.5 °C (UNFCCC, 2015a). Then, in response to an invitation under the Paris Agreement, the Intergovernmental Panel on Climate Change (IPCC) released in 2018 its landmark Special Report on Global Warming of 1.5°C. Illustrated within this report is the stark difference in impacts for human and natural systems between 1.5 °C and 2 °C of warming, underlining the importance of limiting the temperature rise to 1.5 °C, to avoid markedly worse outcomes that would be experienced at 2 °C (IPCC, 2018). To reach the 1.5 °C

target[1], emissions will have to drop by nearly half by 2030 compared to 2010 levels, and come to net zero by mid-century. This would require swift and widespread systematic changes in all sectors of society, at a scale unprecedented in human history.

The climate crisis is inextricably linked to energy: roughly three quarters of total anthropogenic GHG emissions stem from energy consumption[2] (Climate Watch, 2018). Fossil fuels, which continue to dominate global energy use, generate significant GHG emissions from their production and combustion. As a result, tackling climate change requires a major transformation of the worldwide energy system, away from fossil fuels and towards carbon neutral energy. This would offer a win-win situation by fighting climate change while simultaneously improving air quality. That is, producing and burning fossil fuels also generates emissions of air pollutants (and their precursors), e.g., particulate matter (PM), volatile organic compounds (VOCs), nitrogen oxides (NO_x), and carbon monoxide (CO). Moreover, the latter three species are precursors to tropospheric (ground-level) ozone (O_3). The harmful effects of poor air quality on health are well established, with the World Health Organization (WHO) listing ambient (outdoor) air pollution as a leading cause of premature death and disease globally (WHO, 2020). Finally, a low carbon energy transition can also strengthen domestic energy security.

At the end of 2019, the European Commission presented the European Green Deal, which provides an action plan to make the economy of the European Union (EU) sustainable (European Commission, 2019), and in early 2020 proposed the European Climate Law to ensure GHG neutrality by 2050 (European Commission, 2020a). Similarly, at the end of 2019 Germany enshrined the goal of becoming GHG neutral by 2050 in its first federal climate law (BMU, 2019a). Considering that air pollution is still a major health threat in Europe—being culpable for about a half million premature deaths annually—the low carbon energy transition can bring meaningful health benefits to the regional population as well (EEA, 2019). It remains to be seen how much the energy demand, and hence GHG and air pollutant emissions, will decrease in 2020 as worldwide economic activity has slowed down due to the Covid-19 pandemic and related countermeasures. While emissions are widely expected to rebound as the economy recovers, the International Energy Agency (IEA) recently emphasized that countries have a "once-in-a-lifetime opportunity" through their economic recovery packages to create jobs and drive the shift towards a sustainable energy future (IEA, 2020). Indeed, Germany's EUR 130 billion stimulus package and the European Commission's EUR 750 billion EU-wide stimulus package place the climate and sustainable energy transition in central focus (BMF, 2020; European Commission, 2020b).

About a decade ago, the outlook for shale gas development in Europe was promising. At the same time, the global wave of interest surrounding hydrogen was fading away. Of course, politics and industry can be transient and fickle: over the past decade, the situation has completely reversed between these energy types. Solid scientific information

[1] With no or little overshoot.

[2] The energy sector includes use of energy in buildings, electricity & heat, fugitive emissions, manufacturing & construction, other fuel combustion, and transportation.

is critical, both for directly informing decision-making to foster effective sustainable energy policy, as well as for providing input to transdisciplinary processes, which more closely connect science, policy and society. Given this, the motivation of this thesis is to strengthen and extend the knowledge base on relevant, potential environmental impacts from possible transitions and long-term changes in European energy, in the context of ongoing developments in energy discussions. Shale gas and hydrogen have been two of the most important energy breakthroughs in the past decade (IEA, 2012, 2019a), and thus are the foci of this thesis. Accordingly, this thesis begins by exploring possible future shale gas development in the United Kingdom (UK) and Germany, and then evolves with the present day energy discourse by exploring a possible shift towards hydrogen mobility in Germany. Given the importance and inherent connection between energy and emissions, and in turn air quality, examined here are the impacts on GHG and air pollutant emissions, as well as on O_3 pollution. In the next two sections, a brief overview of both energy types is provided, based partly on the introductions to the journal papers in Chapters 2, 3 and 4 of this thesis.

1.1.1 Shale gas

Natural gas emits less CO_2 per unit of energy compared with coal or oil. For this reason, it has been promoted by industry and other stakeholders over the years as a bridge fuel on the road to a decarbonized energy future (Paltsev et al., 2011; The White House, 2013). Shale gas is natural gas trapped in shale rock formations. It is referred to as unconventional natural gas owing to the advanced techniques required for its extraction compared with conventional gas. Regarding chemical composition, however, both gas forms are equivalent. Shale gas production has increased dramatically in the United States (US) since the early 2000s as a result of improvements in horizontal drilling and hydraulic fracturing ('fracking') technologies, which made previously uneconomical shale resources commercially developable (EIA, 2020). According to the US Energy Information Administration's 2013 assessment, world technically recoverable shale gas reserves amount to a considerable ∼200 trillion cubic meters (tcm), of which ∼13 tcm is attributed to Europe (EIA, 2013). To put this into perspective, shale gas represents approximately one third of the world's total technically recoverable natural gas reserves (EIA, 2013). The US shale gas revolution sparked interest for development of this resource in other regions around the world with shale reserves, including in European countries like the UK and Germany, with the hopes of harnessing, e.g., economic benefits, enhanced energy security, and the opportunity to reduce GHG emissions (IEA, 2012; JRC, 2012).

Nevertheless, shale gas has drawn criticism over potentially dangerous impacts it poses to the environment and human health. First, natural gas is composed primarily of CH_4, which is a highly potent GHG with a global warming potential (GWP; i.e., heat-trapping ability) 87 times that of CO_2 over a 20-year timescale, or 36 times CO_2 over a 100-year timescale (Myhre et al., 2013). Indeed, CH_4 is leaked throughout the stages of natural gas production and transmission. A number of studies in recent years indicate that CH_4

leakage from natural gas development (and especially from shale gas development on account of the intensive activities required) may be significantly underestimated, offsetting alleged climate benefits compared with oil and coal (Alvarez et al., 2018; Brandt et al., 2014; Howarth, 2014, 2019; Howarth, Santoro, & Ingraffea, 2011; Karion et al., 2013; Pétron et al., 2012, 2014). Second, VOCs are present in raw natural gas and thus co-emitted with CH_4, while NO_x and CO are generated throughout production from operation of heavy machinery. As these species can act as pollutants and are precursors to O_3, shale gas production may significantly worsen air quality. Given the close proximity of European shale gas reserves to urban areas combined with the region's high population density (JRC, 2012), a future shale gas industry in Europe may pose unacceptable risks to the health of its residents. Finally, a number of other environmental and health concerns remain, e.g., induced seismicity and surface and groundwater contamination (UBA, 2015).

Ground-level O_3 is formed through a complex series of reactions involving VOCs, CO and NO_x in the presence of sunlight (Sillman, 1999, 2003). O_3 pollution triggers a variety of deleterious health effects, most notably to the respiratory system (Amann et al., 2008; EPA, 2013; Horvath & McKee, 1993; Nuvolone, Petri, & Voller, 2018). Moreover, O_3 is harmful to ecosystems (Amann et al., 2008; EPA, 2013; Horvath & McKee, 1993), threatens food crops (Avnery, Mauzerall, Liu, & Horowitz, 2011; Sharma, Ojha, Pozzer, Beig, & Gunthe, 2019) and damages infrastructure (Kumar & Imam, 2013; Lee, Holland, & Falla, 1996). The EU target value for maximum daily 8-hour average O_3 (MDA8) is $120\,\mu g\,m^{-3}$, not to be exceeded on more than 25 days per year, averaged over 3 years (EP, 2002). On the other hand, the WHO sets a stricter guideline of $100\,\mu g\,m^{-3}$ on account of studies showing harmful health effects already below $120\,\mu g\,m^{-3}$ (WHO, 2005). Nevertheless, Europe is still suffering from unhealthy levels of O_3 pollution. For example, 96 % of the EU-28 urban population was exposed to levels of O_3 above the WHO guideline in 2017 (EEA, 2019). Accordingly, emissions from future shale gas operations in Europe have the potential to exacerbate O_3 issues in this region.

As of now, there is no shale gas industry in Europe. Poland has been the only country so far to run a proper geological investigation of shale gas (PGI, 2015), yet the test project was unsuccessful and hopes of a Polish shale gas industry were ultimately abandoned (LaBelle, 2018). On the other hand, strong opposition by the public, environmentalist groups and other stakeholders due to concerns over dangerous impacts have led to shale gas bans in several European countries. The German government currently has a strict ban on shale gas fracking activities, which may be reexamined in 2021, if specific action is taken to do so (German Federal Government, 2017). While the British government initially granted consent for hydraulic fracturing testing to the oil and gas company Cuadrilla in late 2018 (Perry, 2018), testing was riddled with numerous issues from inducing tremors. After the UK Oil and Gas Authority released a report in late 2019 finding that it was not possible to rule out unacceptable seismic activity impacts on local residents, the government announced the same day an indefinite moratorium on shale gas activities, in effect until compelling new data suggests otherwise (UK Government, 2019).

1.1.2 Hydrogen mobility

Hydrogen is a non-toxic, colorless, odorless gas, has high energy density per unit mass, and is considered a clean fuel as it generates zero carbon and pollutant emissions at the point of consumption[3]. For these reasons, hydrogen offers great potential to transform and decarbonize energy systems, hence the conception of the hydrogen economy. Hydrogen is also the most abundant element in the universe, though it is not found in nature on Earth in its pure form (UNEP, 2006). Thus it is an energy carrier and not a source, meaning that it must be produced from a hydrogen-rich raw material. Nevertheless, hydrogen offers a high level of flexibility because it can be made from a variety of energy sources (fossil fuels and renewable energy) and technologies. The most relevant hydrogen production methods today are steam methane reforming (SMR; using natural gas), coal gasification (CG), and water electrolysis; notably, the latter method offers the potential for zero-emissions hydrogen when renewable electricity is applied ('green' hydrogen). Additionally, hydrogen can be stored in pure form as gas (compressed) or liquid (cryogenic), in a blend with natural gas or bound with larger molecules, and transported by pipeline, truck or ship (IEA, 2019a).

Hydrogen is also flexible in the sense that it can be used in a wide array of energy applications, and is particularly promising to sectors that are difficult to decarbonize through electrification alone. Of especial relevance is transportation, considering that it is one of the chief polluting sectors in terms of GHGs and air pollutants (Ge & Friedrich, 2020; IEA, 2019b; WHO, 2020). For example, road transport currently accounts for nearly one-fifth of total GHG emissions in Germany[4], and is the country's only sector in which little to no progress has been made in terms of GHG emissions abatement (BMU, 2019b). Hydrogen fuel cell electric vehicles (FCEVs) offer attractive benefits over battery electric vehicles (BEVs), such as quick refueling ($\sim$3 min.) and longer driving ranges ($\geq$500 km) similar to conventional internal combustion engine vehicles (ICEVs) (Ehret, 2019; H2 Mobility, 2020). In particular, FCEVs are competitive over BEVs for long-range transport and heavy loads. Accordingly, hydrogen mobility is being considered as a possible complement in a low-carbon energy system, including in Germany. Indeed, passenger FCEVs have been in commercial production for many years now, with about 17,000 FCEVs sold globally at the end of 2019 (Mackenzie, 2020), of which 500 were registered in Germany (KBA, 2020), not to mention 432 hydrogen fueling stations operating globally, of which 87 were located in Germany ranking it the second largest network worldwide (H2stations.org, 2020).

The idea of the hydrogen economy is not new, and has gone through many cycles of built up hype followed by disillusion. One major challenge is the fact that the overwhelming majority of hydrogen today ($\sim$99 %) is produced by fossil fuels, generating 830 $MtCO_2$ annually (IEA, 2019a)—translating to roughly 2 % of total global anthropogenic CO_2

[3] When hydrogen is used with a fuel cell.

[4] The discussion here is focused on pre-COVID19, since energy changes during the pandemic are not the result of structural changes, without which emissions are expected to go back to normal.

emissions for 2019 (Friedlingstein et al., 2019). Moreover, the high cost of FCEVs, fueling stations and green hydrogen has been a lofty hurdle. Hydrogen also suffers energy losses along the chain from production to end use, which lowers its overall energy efficiency. For example, while hydrogen is highly energy dense by weight (roughly 3x the energy density of gasoline), its volumetric energy density is low, meaning that it must be compressed or cryogenically stored resulting in energy costs.

Nevertheless, there has been a new wave of unprecedented enthusiasm emerging for hydrogen by governments and businesses around the globe (IEA, 2019a). Progress with respect to renewable-based hydrogen is being made and it is extensively reported that major challenges such as cost can be overcome (as discussed in Chapter 4). In particular, the level of commitment appears different this time around on account of the urgency and earnestness to achieve deep emission reductions to avert irreversible impacts of climate change, as manifested by the 2015 Paris Agreement and the 2018 IPCC report as discussed above (IEA, 2019a; IPCC, 2018; IRENA, 2019; UNFCCC, 2015b). Both Germany and the EU have demonstrated eagerness to become global leaders in hydrogen technologies along with their commitment to establishing a hydrogen economy. Namely, in June 2020 Germany approved its ambitious National Hydrogen Strategy and dedicated EUR 9 billion of its pandemic economic stimulus package to hydrogen technologies (BMWi, 2020), and in July 2020 the EU adopted its hydrogen strategy (European Comission, 2020a, 2020b). Indeed it is widely expected that hydrogen will play a significant role in a low-carbon energy future.

1.2 Research questions

Given the background and motivation of this thesis as outlined in Section 1.1, the overarching question is formulated as follows and addressed through two research questions:

Overarching Question: What are the anticipated environmental impacts of shale gas and hydrogen as possible transition or long-term energy sources in Germany and the United Kingdom?

> *RQ1: What are the anticipated greenhouse gas and air pollutant emission impacts of shale gas in Germany and the United Kingdom and hydrogen in Germany?*
>
> *RQ2: How would shale gas emissions in Germany and the United Kingdom impact ozone air quality on the local and regional scale?*

1.3 Papers and personal contribution

The research questions in Section 1.2 are addressed through three research papers. All three papers address the overarching question, and specifically Papers I and III address RQ1, while Paper II addresses RQ2. The three papers and personal contribution to each are presented in Table 1 below.

Paper I	Cremonese, L., **Weger, L.B.**, van der Gon, H.D., Bartels, M.P. and Butler, T., 2019. Emission scenarios of a potential shale gas industry in Germany and the United Kingdom. Elem Sci Anth, 7(1), p.18. DOI: http://doi.org/10.1525/elementa.359
Personal contribution	*The conception and design of the paper were developed by Lorenzo Cremonese, Marianne Bartels, Tim Butler and myself. The drilling projections were performed by Lorenzo Cremonese. The data collection and execution of the emission scenarios were co-led by Lorenzo Cremonese and myself. The analysis and interpretation of the results were led by Lorenzo Cremonese, with contributions from Tim Butler on all parts and major contributions from myself on the emission scenarios (specifically for the manuscript sections: GHG emissions in shale gas scenarios, GHG emissions national contexts, CO_2 and CH_4 contribution to total GHG emissions, Methane leakage rates in the international context, and Other Pollutants). An additional sensitivity analysis was performed by Lorenzo Cremonese. The paper was mainly written by Lorenzo Cremonese, with substantial contributions from myself on the Introduction and the aforementioned manuscript sections, and edited by all co-authors.*
Paper II	**Weger, L.B.**, Lupascu, A., Cremonese, L. and Butler, T., 2019. Modeling the impact of a potential shale gas industry in Germany and the United Kingdom on ozone with WRF-Chem. Elem Sci Anth, 7(1), p.49. DOI: http://doi.org/10.1525/elementa.387
Personal contribution	*The conception and design of the paper, data collection, model simulations, analysis and interpretation of the results were led by myself, with contributions from the co-authors. The paper was written by myself and edited by all co-authors.*
Paper III	**Weger, L.B.**, Leitão, J., Lawrence, M.G., 2021. Expected impacts on greenhouse gas and air pollutant emissions due to a possible transition towards a hydrogen economy in German road transport. Int J Hydrogen Energ, 46(7), p.5875. DOI: https://doi.org/10.1016/j.ijhydene.2020.11.014
Personal contribution	*The conception and design of the paper, data collection, emission scenario execution, analysis and interpretation of the results were led by myself, with contributions from the co-authors. The paper was written by myself and edited by all co-authors.*

Table 1: Personal scientific contribution to papers on which this thesis is based.

1.4 Thesis overview

This thesis consists of five chapters, beginning with this introduction section, followed by three chapters consisting of the research papers, and concluding with a synthesis section. The introduction section provides the background and motivation of this work, along with the research questions, personal contribution and methodology (Chapter 1). The three research papers form the foundation of this thesis. The overarching research question is addressed by all three papers, and in particular research question RQ1 is addressed in Papers I and III (Chapter 2 and 4, respectively), and RQ2 in Paper II (Chapter 3). The synthesis section presents a discussion of the key findings of the papers with respect to the research questions and conclusions (Chapter 5).

1.5 Methodology

Emission scenarios and air quality modeling can serve as important tools in assessing how possible, future changes in the energy system may impact the environment, which can provide valuable insight to policymakers and other stakeholders. Accordingly, this thesis answers the research questions in Section 1.2 using these two methods. Specifically, RQ1 is addressed via emission scenarios, and RQ2 via air quality modeling with the online-coupled regional chemistry transport model WRF-Chem (Weather Research and Forecasting-Chemistry). The methodology is described in depth in the corresponding chapters of this thesis.

2 Paper I: Emission scenarios of a potential shale gas industry in Germany and the United Kingdom[5]

[5]Published as: Cremonese, L., Weger, L.B., van der Gon, H.D., Bartels, M.P. and Butler, T., 2019. Emission scenarios of a potential shale gas industry in Germany and the United Kingdom. Elem Sci Anth, 7(1), p.18. DOI: http://doi.org/10.1525/elementa.359

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RESEARCH ARTICLE

Emission scenarios of a potential shale gas industry in Germany and the United Kingdom

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The shale gas debate has taken center stage over the past decade in many European countries due to its purported climate advantages over coal and the implications for domestic energy security. Nevertheless, shale gas production generates greenhouse gas and air pollutant emissions including carbon dioxide, methane, carbon monoxide, nitrogen oxides, particulate matter and volatile organic compounds. In this study we develop three shale gas drilling projections in Germany and the United Kingdom based on estimated reservoir productivities and local capacity. For each projection, we define a set of emission scenarios in which gas losses are assigned to each stage of upstream gas production to quantify total emissions. The "realistic" (REm) and "optimistic" (OEm) scenarios investigated in this study describe, respectively, the potential emission range generated by business-as-usual activities, and the lowest emissions technically possible according to our settings. The latter scenario is based on the application of specific technologies and full compliance with a stringent regulatory framework described herein. Based on the median drilling projection, total annual methane emissions range between 150–294 Kt in REm and 28–42 Kt in OEm, while carbon dioxide emissions span from 5.55–7.21 Mt in REm to 3.11–3.96 Mt in OEm. Taking all drilling projections into consideration, methane leakage rates in REm range between 0.45 and 1.36% in Germany, and between 0.35 and 0.71% in the United Kingdom. The leakage rates are discussed in both the European (conventional gas) and international (shale gas) contexts. Further, the emission intensity of a potential European shale gas industry is estimated and compared to national inventories. Results from our science-based prospective scenarios can facilitate an informed discussion among the public and policy makers on the climate impact of a potential shale gas development in Europe, and on the appropriate role of natural gas in the worldwide energy transition.

Keywords: Shale gas; Unconventional gas; Methane leakage; O&G industry; Fracking; Natural gas

Introduction

Over the past decade there has been a rapid increase in natural gas production in the United States (US), mainly due to shale gas, which accounts for about 60% of current total production (WEO, 2017). As the name suggests, shale gas is natural gas that comes from shale reservoirs. Shale, a fine-grained, laminated, sedimentary rock, has an extremely low permeability which in the past made extraction of this gas type difficult and hence uneconomical. However, advancements in horizontal drilling and hydraulic fracturing in recent years have unleashed previously unrecoverable shale gas reserves to large-scale, commercial production (Jenkins and Boyer, 2008; Gregory et al., 2011).

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Natural gas is often described as a transition fuel on the road to a decarbonized global energy system. This is because natural gas generates less carbon dioxide (CO_2) emissions during combustion per unit of energy than coal or oil (WEO, 2017), and therefore enables continued fossil fuel use with an ostensibly smaller impact on the climate. However, methane (CH_4) – the main component of natural gas – is a powerful greenhouse gas (GHG). On a mass-to-mass basis, CH_4 warms the planet 87 times that of CO_2 over a 20-year timescale, and is 36 times more warming over a 100-year timescale (IPCC, 2014). Indeed CH_4 emissions (here also reported as losses) are generated during the various stages of natural gas production. In this study we distinguish emissions of CH_4 as follows: fugitive emissions (as a result of accidental leaks; e.g., damaged gaskets or pipes, incidents, etc.); gas venting (intentional design of machinery such as pneumatic device venting, equipment blowdowns, etc.), and associated emissions, such as CH_4 emitted by associated activities (e.g., trucks, indirect emissions induced by electricity usage, etc.).

CH_4 emissions additionally have a negative effect on public health due to the role of CH_4 as a precursor of ground-level ozone (O_3; Garcia et al., 2005). Furthermore, natural gas extraction and processing leads to emissions of air pollutants including volatile organic compounds (VOCs), nitrous oxides (NO_x), carbon monoxide (CO), and particulate matter (PM), which negatively affect human and environmental health (Dockery and Pope, 1994; Kampa and Castanas, 2008; Roy et al., 2014; Sweileh et al., 2018). The dramatic increase in shale gas exploitation has therefore raised concerns about the burden on the climate and air quality. Accordingly, many studies have been conducted over the past years to examine the influence of shale (also called unconventional) and conventional gas production on emissions, and on CH_4 emissions in particular. Especially in Europe, it is a shared belief among societal and political actors that emissions from conventional gas production are substantially lower than those from shale gas (DW, 2018; Energate, 2018; Zittel, 2015; Greenpeace, 2015; Howarth, 2014). Although this was probably the case at the onset of the shale gas boom when fracking operations were not properly regulated (e.g., open pits for storing flowback waters, improper well completion, etc.), rigid environmental standards are largely in place to date in the US. The latest scientific literature on this topic is still ambivalent, and the preliminary – despite insufficient – data available seems to not support this large discrepancy: as reported in Zavala-Araiza et al. (2015), about 50% of emissions investigated in their study are attributed to compressor stations and processing plants, and therefore sources unrelated to the production technique. The remaining share is generated at production sites extracting both conventional and shale gas. Therefore, in the extreme and unrealistic case where hydraulic fracturing (i.e., the recovery technique employed during shale gas extraction) were the only CH_4 source at production sites, the unconventional gas production chain would generate about three-fourths of total emissions associated with natural gas production in the US.

In support of this, hydraulic fracturing appears to not be responsible for larger emissions according to results by Allen et al. (2013), despite the fact that emission budgets here might be underestimated due to the bottom-up data method applied (Brandt et al., 2014). Although Reduced Emissions Completions (RECs) – a practice needed only at shale gas wells and able to cut emission by at least 90% during well completion (EPA, 2014a) – have been mandatory in the US since January 2015, studies still continue to measure very high losses from overall gas recovery activities. One explanation might be that gas released during well completions, often alleged to be responsible for augmented emissions at shale gas wells, have only a minor contribution to total budgets. For example, Alvarez et al. (2018) estimate an upstream leakage rate of 1.95% from about 30% of all existing oil and gas wells in the US without reporting any evident discrepancy between these two natural gas categories both present among the gas plays analyzed. Yet, the EDF chief scientist and co-author of the study stated that "most [of the emissions detected] are tied to hatches and vents in natural gas storage tanks at extraction wells",[1] sources that can occur at any stage along the production chain and are therefore not necessarily linked to fracking operations. Results from Omara et al. (2016) show a correlation between the CH_4 leakage rate and age of the wells rather than the nature of the gas, proving that impacts related to other factors may, at least occasionally, be greater than gas type. Data available for European gas plays is yet scarce. While US shale gas leakages reported in Howarth (2014) can be higher than 10%, data for conventional gas in countries like Germany and the UK (NIR 2017) shows instead leakage rate below 0.1%. This large emission discrepancy is widely applied to narratives on natural gas usage in the European context to oppose unconventional gas development. Nevertheless, Yacovitch et al. (2018) found high uncertainty in emission inventories from oil and gas wells in the Groningen Field in the Netherlands, and the occurrence of an unidentified offshore super-emitter source. Moreover, preliminary quantification of CH_4 losses at North Sea offshore oil and gas platforms suggest much higher estimates than those reported by the UK national emission inventory, up to 0.70% of the total gas produced (Riddick et al., 2019). All of these studies performed in the US and emission discrepancies with European datasets do not conclusively prove large offsets between emissions from shale gas and conventional gas activities, and specifically do not explain much more conservative emissions for the latter. At the same time, they neither prove the opposite. The emission contribution of shale gas and conventional gas to total gas losses remains unclear to date, and further research is needed to reconcile emission budgets and rates among these regions. This argument is further examined in the "Results and discussion" Section.

Notwithstanding that shale gas production has occurred primarily in the US, global shale gas resources are considerable, amounting to >200 tcm (trillion cubic meters) – or rather, about one third of the world total technically recoverable natural gas reserves (EIA, 2013). Several European countries, including Germany and the United Kingdom (UK), have expressed interest in recent years in utilizing domestic shale gas assets as part of their national energy agenda. Although shale gas reserves in Germany and the UK are substantially smaller than those found in, e.g., the US, production of shale gas has the potential to offset or slow down the decline in conventional gas production that these countries are experiencing. This would avoid increased dependency on foreign gas imports, as well as avoid a potential increase in coal use for electricity generation. However, opposition from the general public and environmental interest groups on account of potentially harmful effects from shale gas fracking activities – for example, surface and groundwater contamination (Osborn et al., 2011; Jackson et al., 2013; Darrah et al., 2014; Drollette et al., 2015), increased frequency of earthquakes (Ellsworth, 2013), as well as increased emissions as discussed above (Oltmans et al., 2014; Swarthout et al., 2015; Hildenbrand et al., 2016) – has led to moratoria and bans in various regions and countries like in France and Germany. In the latter, the government recently placed a ban on unconventional fracking at least until 2021 (Bundesregierung, 2017).

In the context of sustainability, a responsible energy strategy with regard to shale gas production in Europe requires sound scientific advice. Studies that explore what the range of impacts that a potential European shale gas industry would entail, as well as opportunities to reduce potentially harmful effects, are still missing although necessary to inform policy. Here we examine the impact of a potential shale gas industry in Germany and the UK – two countries where political and social discussion on shale gas has been intense over the last years – on GHG and pollutant emissions, including CH_4, CO_2, VOCs, NO_x, CO, PM_{10} and $PM_{2.5}$ (PM ≤10 μm and ≤2.5 μm in diameter, respectively) through emission scenarios. First, we give an overview of shale characteristics and examine the shale reservoirs considered in this work. Then, we discuss how the drilling projections and emission scenarios are developed for Germany and the UK. Next, we describe each of the scenarios that we designed, including the data that we incorporated and assumptions that we made. Subsequently we present the results, i.e., the impact of shale gas operations on emissions per each scenario. Finally, we analyze the impact on these two countries, putting the emissions into context with current inventories to develop and transfer findings to policy-makers. The aim of our scenarios is to understand what a shale gas industry in Europe may look like, to show how regulation and compliance (along with uncertainty ranges) may impact emissions, and to present opportunities for air quality and emission mitigation.

Other potential consequences of shale gas production, such as surface and water contamination, seismic activity, and an offsetting of emissions from coal in electricity generation due to availability of natural gas, are important but outside the scope of this study and are not be considered here. A follow-up study will explore the potential impact of shale gas emissions on local and regional air quality in Europe through atmospheric chemistry modelling.

Methodology

In this study we investigate realistic shale gas industrial developments in Germany and the UK, and quantify their associated GHG and air pollutant emissions. In order to do this, we first develop drilling projections in which we estimate the total number of "*wells under construction*" and "*producing wells*" required to achieve and maintain steady-state gas production in the two countries of reference, based on varying degrees of well productivity. After that, we quantify emissions associated with upstream production through a bottom-up approach in different scenarios covering a series of well productivity and technology/performance cases. The results presented here are plausible under specific geological and technological/performance conditions selected in this study and consistent with the existing scientific literature. Results and their interpretation reported in the discussion section, as well as their scientific relevance, have to be therefore evaluated taking such constraints into account. Additionally, a sensitivity analysis of the emission scenarios is performed and is provided in the SM, Text S1, Section S3. The purpose of this is to examine the contribution and influence of each varying parameter on the final results to guide the selection process of such parameters.

Shale characteristics and gas extraction

Shale is a sedimentary type of rock that is generated by the compaction of deposits containing silt- and clay-size particles. While shale is characterized by extremely low permeability, it possesses a high porosity. It is in these pores that the organic material and gas molecules are located, as free gas or adsorbed on organic remains (Glorioso and Rattia, 2012). During the shale gas extraction process, a vertical shaft is initially drilled. Then, when the vertical drill path reaches the target shale formation – usually between 1,000 and 4,000 m underground depending on local geological features – its direction is shifted horizontally to follow the shale plane (Elsner and Hoelzer, 2016). Afterwards, water, sand, and chemicals are injected at high pressure to create fractures in the rock during the hydraulic fracturing or "fracking" process, increasing permeability of the formation and thereby stimulating gas flow to the well (Gregory et al., 2011). Although most public attention tends to focus on the hydraulic fracturing, this recovery technique was performed experimentally in 1947 and has actually been in widespread use in Germany since the 1960s (LBEG, 2010; Wilson and Schwank, 2013). In fact, horizontal drilling is the more recent technology and game changer that has made commercial shale gas production possible. Horizontal wells – which can extend over several kilometers – maximize contact with the shale payzone which is typically spread out in narrow, horizontal bands, whereas vertical wells can only provide a small, insufficient portion of contact (Pearson et al., 2012; ACATEC, 2016). Furthermore, directional drilling is used to reach targets beneath adjacent lands, intersect fractures, and drill multiple wells from the same vertical borehole (Elsner and Hoelzer, 2016), thereby maximizing the shale gas yield while reducing the surface environmental footprint.

Shale reservoirs considered in this study

The shale gas reservoirs taken into account in the present study are based on recent studies which aimed to quantify the relevance of shale gas as a national energy asset by both the German and British governments. In its 2016 report, the Federal Institute for Geosciences and Natural Resources (BGR, 2016) found five shale basins in Germany to be promising for natural gas production: the *Fischschiefer*, *Wealden*, *Posidonien*, *Mittetrhät*, and *Unterkarbon* units. These basins are scattered across several federal states, covering a total area of more than 8,000 km^2, and are buried between 500 and 5,000 m underground. The technically Recoverable Resource (TRR) for these reservoirs ranges between 650 and 1,380 bcm (billion cubic meters), averaging at 940 bcm. By comparison, the UK's geological landscape is characterized primarily by one major shale basin, the Bowland-Hodder Carboniferous Unit. This basin spans an area of 14,000 km^2 underneath the regions of Yorkshire, North West, East and West Midlands and reaches a maximum depth of 4,750 m below ground. According to the British Geological Survey's (BGS) 2013

report, the total gas-in-place (GIP) buried in this formation is estimated at 37.6 tcm (trillion cubic meter), while the TRR is still unknown. Pilot exploration projects by Cuadrilla are planned and started again in late 2018, after a long break following the Blackpool Earthquake in 2011. The shale reservoir locations in Germany and the UK are shown in **Figure 1**. These basins are selected as the gas reservoirs to be exploited in our drilling projections.

Drilling projections

Three different projections of shale gas well populations are developed for Germany and the UK in this work, referred to henceforth as drilling projections. The drilling projections ultimately provide information on the number of *wells under construction* and *producing wells*, information that is necessary to quantify emissions from shale gas production in the emission scenarios. In the next paragraphs we describe the four main steps involved in building the drilling projections and the critical assumptions made.

Step 1: *Basin productivity.* We first define the extension of the shale gas prospective basins described in the previous section. Subsequently we considered the Technical Recoverable Resources (TRR), defined by national authorities as the volume of gas that can be produced with currently available technology and practices. The estimated TRR of the shale gas basins is typically provided in an uncertainty range described by three cases of productivity: 25th, 50th, and 75th percentile of exceedance, which we refer to as P25, P50 and P75. Following the approach adopted by the BGR, the TRR of the Bowland Basin was calculated as 10% of the total GIP range estimated by the BGS. In this study, P25 and P75 signify low- and high-basin productivities respectively, while P50 describes the "most likely" case. Due to the lack of data and to reduce complexity, we assume that each of the six basins contains a homogeneous gas density across their geographical extension (i.e., no hot spots are considered). The gas "density" is calculated for each basin and productivity case. Data are reported in SM Text S1, Table S1. TRR results are showed in **Table 1**.

Step 2: *Well Estimated Ultimate Recovery (EUR_{well}).* In order to assess the productivity of the wells (the total gas output from a single well during its lifetime) for each productivity case (P25, P50 and P75) and for each basin, we have to define the portion of reservoir that is exploited by a single horizontal well. To do this, we assume that each well pad exploited an area of 25 km^2, from which 30 horizontal wells are drilled (**Figure 2**; Pearson et al., 2012; Acatech, 2016). Based on the gas densities estimated in step 1, we are able to define the EUR_{well} that characterizes each population of wells. More information on the assumptions on which we base this well geometry is available in the SM, Text S1, Section S1.1.

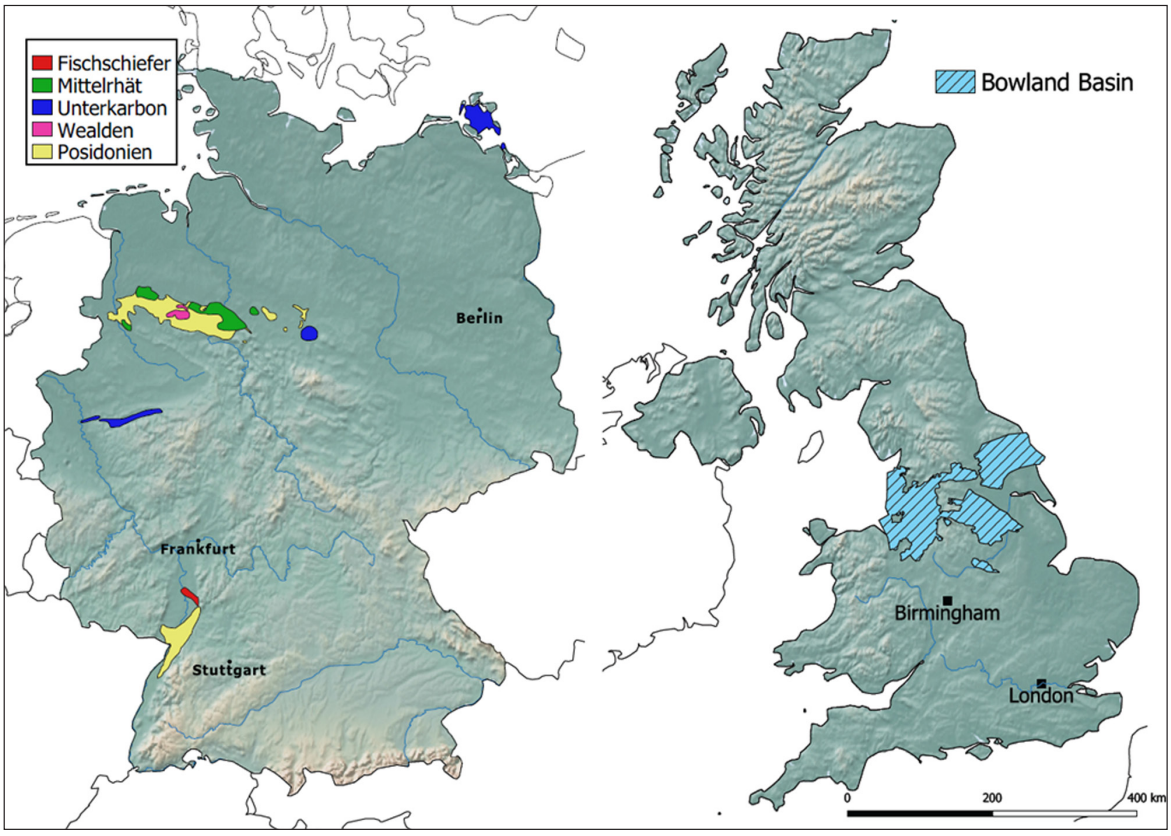

Figure 1: Shale gas basin areas for Germany (left) and the UK (right). Figures include legends with the basin names and their designated color on the map. DOI: https://doi.org/10.1525/elementa.359.f1

Table 1: Shale gas basin characteristics and well data at steady-state production. Area and TRR of all shale gas basins for both Germany and the UK. The number of years required to achieve the desired volume of gas for each productivity case under each basin productivity and emission scenario is also indicated. The ranges of *wells under construction* and *producing wells* represent the variance between the upper and lower boundary for each case. DOI: https://doi.org/10.1525/elementa.359.t1

Country	Productivity Case	Area [Km^2]	TRR [bcm]	Years to maturity	Wells at the steady-state production			
					Optimistic Emissions (OEm)		Realistic Emissions (REm)	
					Under construction	*Producing*	*Under construction*	*Producing*
Germany	P25	8,341	550	8	162	1,927–1,937	164–166	1,952–1,979
	P50		801	3	143	867–872	144–146	879–891
	P75		1,182	2	97–98	522–525	99–100	529–536
UK	P25	13,736	2,866	4	201–202	1,407–1,414	204–206	1,426–1,445
	P50		3,760	2	160–161	856–860	162–164	867–879
	P75		5,447	1	110	479–482	111–113	486–492

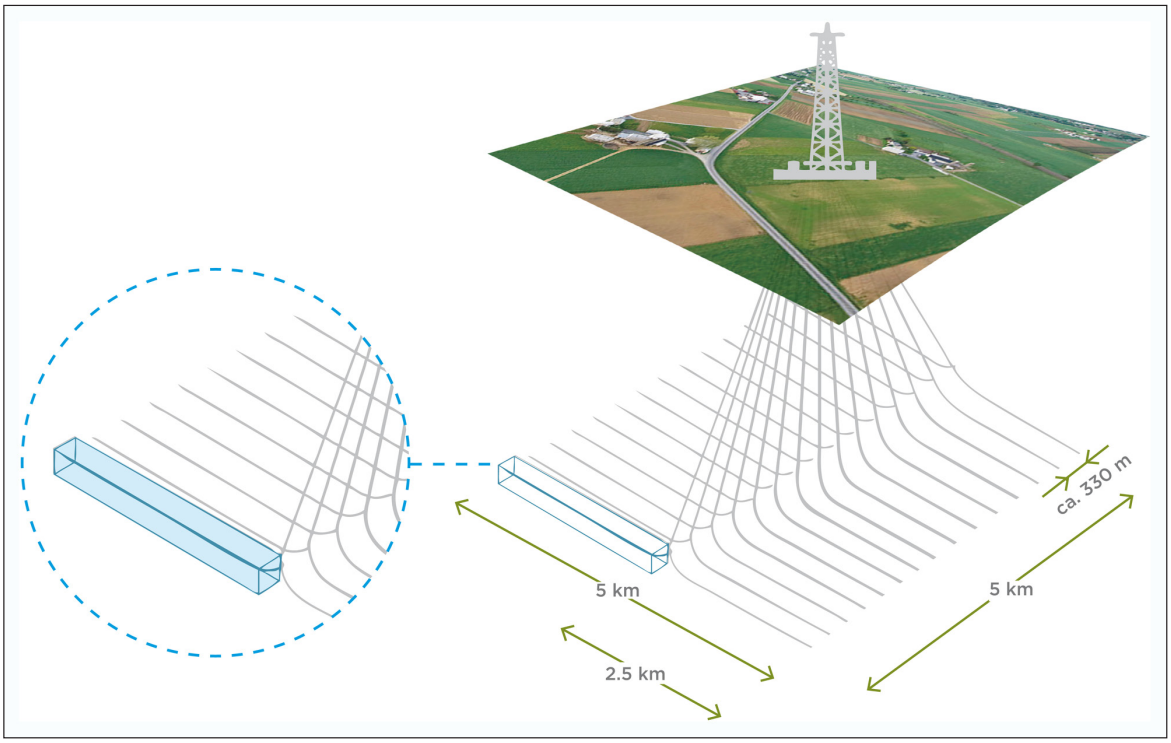

Figure 2: 3D underground view of well geometry. We assume that a total of thirty underground horizontal wells are drilled on a single well pad, which covers an area of 25 km^2, as shown in the figure. DOI: https://doi.org/10.1525/elementa.359.f2

Step 3: *Well productivity curve.* Shale gas wells present a steep production curve: After about one or two years of sustained production, their yield decreases substantially according to the geological characteristics of the shale reservoir (Patzek et al., 2013). This pattern is generally described by a curve declining asymptotically toward zero production (i.e., exhausted well). Here, we describe how the declining curve of the wells is determined. Once the EUR_{well} for each basin is defined, we estimate the Initial Production (IP_{well}) of each population of wells through the R_{ie} factor. This coefficient is based on the correlation between EUR_{well} and IP_{well} observed in the Barnett and the Eagle Ford plays, two US basins that show petrological similarities with the German Unterkarbon and the Posidonia shales (BGR, 2016). To reduce complexity, their IPs and production declining rates are averaged and

applied to all the German and UK basins investigated here. R_{ie} is defined as:

$$R_{ie} = \frac{IP_{well}}{EUR_{well}}$$

The resulting production decline curve is best described with an exponential trend in the first year (Patzek et al., 2013), and logarithmic trend for the following four years as shown in **Figure 3**.

For each basin, the same declining pattern describes the production variation over time, specific to each productivity case. After the fifth year, we assume that gas production remains constant because of our limited knowledge of long-term trends.

Step 4: *Estimating the population of wells under construction* and *producing wells.* These values are based on regional settings and comparisons with the development rates of US shale plays (Hughes, 2013). We estimated that 200 new wells are drilled each year in Germany and 280 in the UK. These estimates represent the final number of wells drilled, taking into account a failure rate of 20% (i.e., unsuccessful wells). Moreover, the wells are numerically distributed among the different German basins proportionally to the basins' extension. The drilling rates are kept constant in the two countries until a gas output of 11.58 bcm for the former and 36.62 bcm for the latter is achieved at industrial maturity. These two values are selected from among all the gas output results obtained by the three drilling projections developed for each country, since they best fit with historical data and realistic national goals of the region under observation (see discussion in SM Text S1, sections S1). Specifically, they are selected from the projections Germany P50 and UK P50. Once the gas flowing from *producing wells* – the population of which grows annually due to continuous drilling activity –reaches these volumes, we calculate the number of annual new wells required to maintain this production level for both Germany and the UK. In fact, this parameter varies over time since it depends on the number of existing *producing wells*, their age and declining rate, and the total gas output. Therefore, we considered the average over the following three years. This value, specific to each country and productivity case, is defined as *wells under construction.* The drilling projections also provide details of the number of active wells (i.e., *producing wells*) at steady state production in each country and productivity case. The output data from each drilling projection, namely *wells under construction* and *producing wells*, are used as input in the emission scenarios. The results are shown in **Table 1** and SM Text 1, Figure S1.

Emission scenarios

The emission scenarios are generated by compiling and aggregating emissions estimated at each stage of the supply chain (i.e., from well preparation to gas processing) at industrial maturity. By feeding the system with the outputs parameters of the drilling projections (*wells under construction* and *producing wells*) for both Germany and the UK, we quantify emissions for each country under diverse basin productivities and production settings (i.e., performance in recovery practices and different technologies). The category *wells under construction* is associated with the stages well pad development, trucks and water pipelines, drilling, fracking and well completion, while the *producing wells* is associated with the stages gas produc-

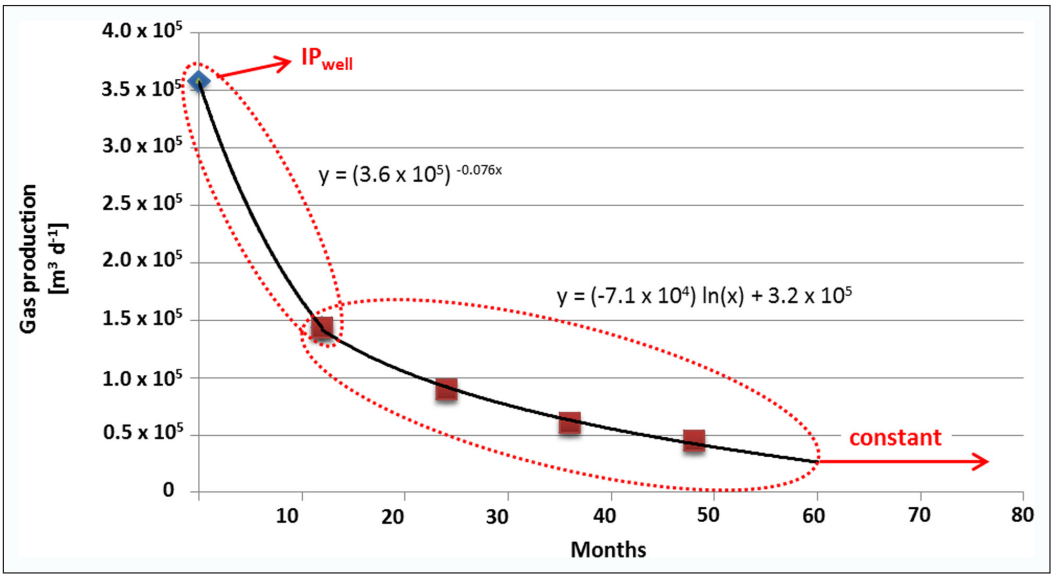

Figure 3: Example of well declining production curve in our scenarios – Unterkarbon (Germany), P50. The declining curve, extrapolated from the Eagle Ford and Barnett shale plays in the US and tailored to the EUR and IP of our case study reservoirs, follow an exponential trend in the first year, and a logarithmic trend in the following 4 years (see equations in the figure). From the 5th year onwards, we assume that production remains constant. DOI: https://doi.org/10.1525/elementa.359.f3

tion, wellhead compressor exhausts, liquids unloading, gas gathering and processing. Emissions are calculated by combining activity data and emission factors for each stage of shale gas production, depending on the technology and uncertainties associated with each specific scenario. Activity data represent the magnitude of activity that results in emissions, while emission factors represent the gas released per unit of a given activity, and are typically provided as a range. All input parameters are based on official reports, expert support and peer-reviewed publications, most of which focused on US shale gas plays since shale gas production has hitherto mostly occurred there. Additionally, the input parameters include our own critical assessments of how to best apply the data to the European cases proposed in this study (described in further detail in SM, Text S1 Section S2). Where available, we opted for large sample-size surveys, with a preference for results by accredited research groups such as the Environmental Defense Fund (EDF, 2019). A list of the parameters and variables that determine emissions as well as the reference literature is reproduced in **Table 2**. To realistically assess VOC emissions as a by-product of natural gas production, we also varied the VOC component of natural gas to examine the impact of both wet and dry gas on total VOC emissions according to gas composition reported by Faramawy et al. (2016).

Our emission scenarios are divided into two overarching categories based on varying technologies/performances at each stage of gas production, namely "realistic" and "optimistic" emission scenarios (abbreviated as REm and OEm). REm refers to practices and standard technologies used for gas exploitation and management which generate relatively high emissions (i.e., business as usual), and are still largely used in the US and Europe. This case is considered the realistic case that we expect for the two European countries examined. On the other hand, OEm refers to the challenging case where emission reduction technologies (e.g., electric motors instead of diesel-engines) and all best practices and monitoring services are in place and fully employed across the supply chain (e.g., no damages of any component, no malpractice and abatement of unwanted gas losses). This case is defined as the most optimistic case and represents the lowest technical emission boundary achievable according to the technologies and practices considered in this study and described in detail in **Table 3** and SM Text S1, Section S2. REm and OEm illustrate the degree to which these two different cases can affect emissions of the suite of pollutants and GHGs under study, and they provide a clear indication on possible mitigation potential of different options. Obsolete technologies or practices that we expect not to be permitted in Europe are not considered in any scenario: e.g., open-air pits, improper well completions (SM Text S1, Section S2.6), low number of wells per pad (SM Text S1, Section S1.1), insufficient environmental standards during the liquids unloading practice (SM Text S1, Section S2.9), lack of recycling of fracking/drilling waters (SM Text S1, Section S2.3), and so forth. These scenarios are informative for evaluating best recovery practices to outline new environmental regulations for drilling and producing. The main technologies and operations that differentiate REm and OEm are listed in **Table 3**, while a complete description is available in SM Text S1, Section S2.

Due to data uncertainty and unpredictable intrinsic variables (e.g., number of fracking stages, uncertainty in values reported by the source agency, etc.), a range of emissions are developed for REm and OEm. Therefore, both scenarios can be further broken down into "upper" (U) or "lower" (L) categories that define the ranges of uncertainty. "U" results define the high end of the emission range, while "L" results define the low end. Altogether, this produces four scenarios (from the lowest to the highest emissions):

Table 2: List of parameters and variables defining emission variations between REm and OEm. Sources of data are also provided. Note that EF stands for emission factor. DOI: https://doi.org/10.1525/elementa.359.t2

Activity	Parameters/variables defining emission scenarios	Source of reference
Well pad development	Length of operations, EF diesel motors	NYSDEC (2015); Helms et al. (2010); European Emission Standards.[2]
Truck Traffic	EF of truck motors, re-suspended particles, road type; materials, water and chemicals supply, waters recycle rate and "piped" vs. "trucked" rate, average well length, fracking stages.	IVT (2015), NYSDEC (2015); EMEP/EEA (2016); Denier Van Der Gon et al. (2018); Statista; CottonInfo (2015).
Drilling	Diesel generators vs. electricity; total wells length.	Pring et al. (2015); Helms et al. (2010).
Fracking operations	# fracking stages, length of operations, diesel engines EFs.	Roy et al. (2014); Helms et al. (2010).
Well completion	Emissions at operations.	Allen et al. (2013).
Production sites	Diesel vs. electric compressors, gas emissions at facility.	Omara et al. (2016); ICF (2014).
Wellhead compressors	Diesel/electric compressor.	NYSDEC (2015); Helms et al. (2010).
Liquids unloading	Automatic vs. manual plunger lifts, operations per well	Allen et al. (2015)
Gathering facilities and pipelines	Gas loss at facility, number of wells connected to the facility, gas loss from pipelines	Mitchell et al. (2015); Marchese et al. (2015); Helms et al. (2010);
Processing	Gas loss at facility, gas turbine efficiency	Mitchell et al. (2015); EPA (2000); Müller-Syring et al. (2016)

OEm-L, OEm-U, REm-L and REm-U. To visualize the breadth of our scenarios, we have represented them as a three-dimensional cube in **Figure 4**.

Results and discussion

GHG emissions in shale gas scenarios

In this section we examine annual emission results from all the scenarios developed in this study and extensively described in the methodology section and SM Text 1 sections S1 and S2. We discuss results under the two technological/performance settings employed during shale gas development (REm and OEm), under differing well productivities (P25, P50 and P75), for both wet and dry gas, for different GHGs and air pollutants, and for both countries under study (Germany and the UK). Both CH_4 and CO_2 emissions from our shale gas scenarios display significant differences in REm and OEm in both countries. In the following paragraphs we focus on wet gas scenarios, while we refer to dry gas scenarios only occasionally: emission trends from dry scenarios closely resemble those from wet scenarios, with the exception that VOCs make up a very small component of the gas composition. Total CH_4 released in REm ranges between 104 and 175 Kt in the UK, and between 46.4 and 78.5 Kt in Germany in the P50 cases (**Figure 5**).

Table 3: List of major differences in the technologies applied to REm and OEm scenarios. DOI: https://doi.org/10.1525/elementa.359.t3

Activity Data	REm	OEm
Motor type	Diesel-engines are used during all stages. Emission factors for non-road diesel machineries refer to the inventory from the German Environmental Agency (Helms, 2010).	Electrified motors applied at some of the production stages at gathering. Emission factors for the national electric grid are available from the German Environmental Agency (UBA, 2017) and are applied to Germany and the UK.
Fracking waters management	Fracking waters are transported to the well site via trucks, with low recycling rates (50%). Emission factors for trucks (Euro3/6) are available from the IVT database (IVT, 2015).	All fracking waters are piped to the well site, with high recycling rates (90%). Emission factors for trucks (Euro6) are available from the IVT database (IVT, 2015).
Turbines (processing)	The volume of gas combusted to fulfil energy requirements during processing is calculated according to the efficiency and performance of a simple cycle, uncontrolled turbines.	Volume of gas combusted to fulfil the energy requirement during processing is calculated according to the efficiency and performance of a combined cycle, water steam-injection turbines.
Emission factors	Emission factors for all engines categories are conservative.	Emission factors for all engines categories follow recent, strict national, legally-binding emission standards.
Well structures	Well ramifications (horizontal wells) at the bottom of each vertical well: 3.	Well ramifications (horizontal wells) at the bottom of each vertical well: 10.

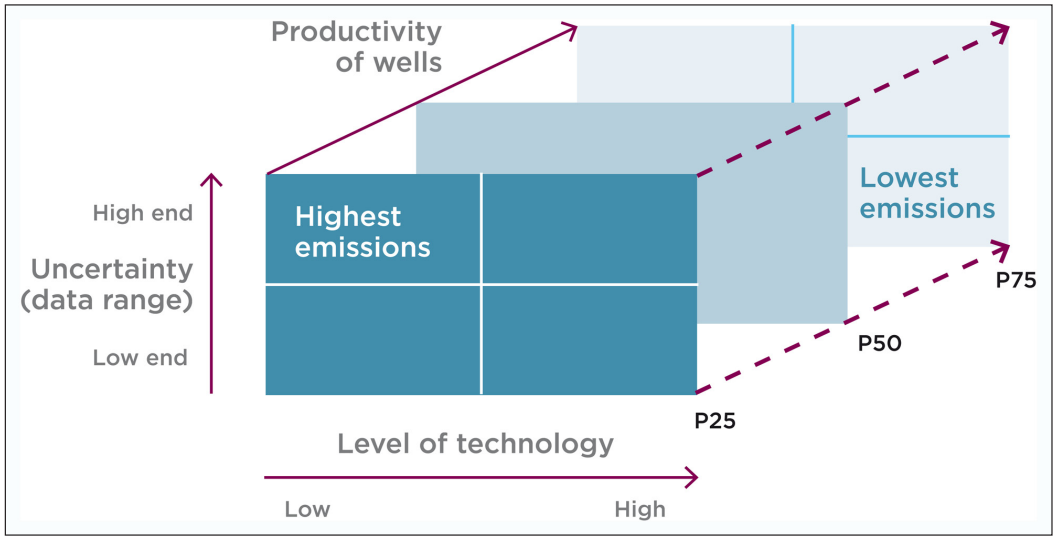

Figure 4: 3D cube representation of shale gas emission scenarios. On the x-axis is the level of technology which is based on the level of regulation; the y-axis represents the uncertainty range in the data, and the z-axis represents the productivity of the wells from the three drilling projections (i.e., P25, P50 and P75). DOI: https://doi.org/10.1525/elementa.359.f4

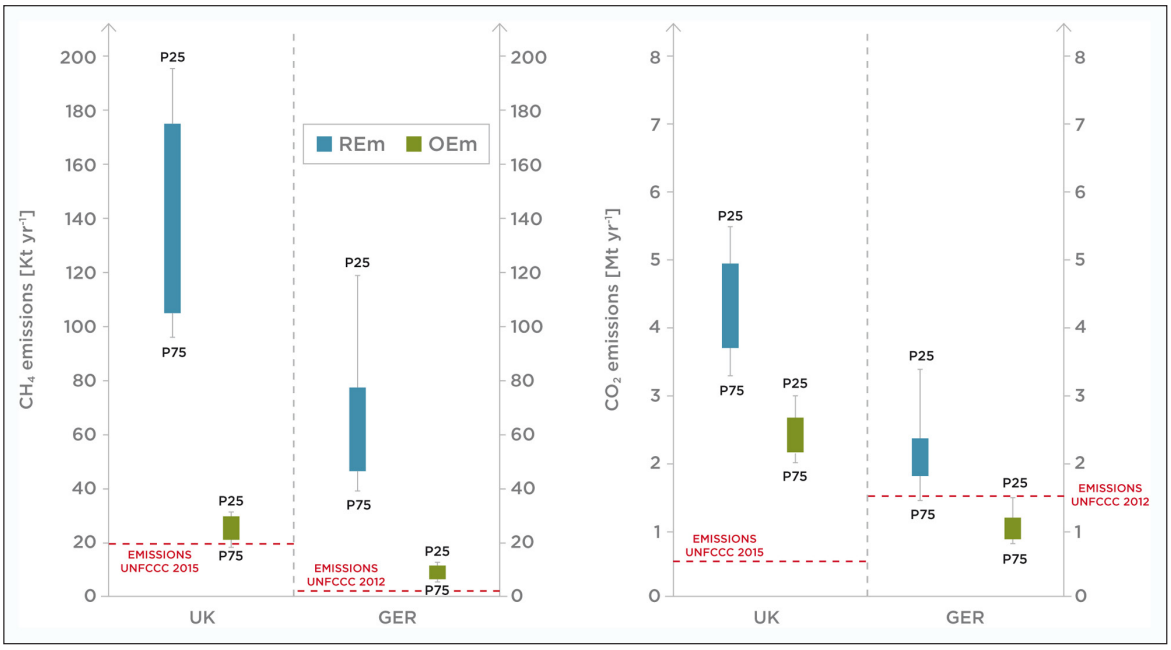

Figure 5: CH_4 and CO_2 annual emissions from our study for Germany and the UK. The dot lines in red represent the emission volumes as reported by the UNFCCC for the year reported. DOI: https://doi.org/10.1525/elementa.359.f5

On the other hand, under the P25 well productivity case maximum CH_4 emissions reach 196 and 119 Kt for the UK and Germany respectively, while in P75 values are not significantly lowered beyond the P50 case. Low well productivity therefore translates into significantly enhanced CH_4 emissions, especially in Germany. The wells' steep production curve characterizing shale reservoirs overall, along with lower volumes of recoverable gas in this specific case justify the higher drilling rate necessary to maintain production constant. The resulting larger population of active wells in both categories *producing wells* and *wells under construction* are ultimately responsible for augmented emissions. On the contrary, CH_4 losses generated by OEm are significantly lower and within a narrow range among the different productivity projections. Here the ranges are from 21.5 to 31.3 Kt for the UK, while from 7.4 to 11.0 Kt in Germany. CO_2 emissions in REm range from 3.7 to 4.9 Mt in the UK and 1.8 to 2.3 Mt in Germany under the P50 scenario case. While low well productivity in REm increases emissions in the UK to a maximum of 5.5 Mt, in Germany the increase is proportionally higher reaching almost 3.4 Mt. CO_2 emissions in OEm range between 2.2 and 2.7 Mt in the UK (P50), and between 0.9 and 1.2 Mt in Germany (P50). The distribution of GHG emissions across the production stages and their variability under different technological/performance cases and for each country are shown in **Figure 6**.

In the following discussion we focus on results from the P50 scenarios, with reference to the other productivity cases when significant emission variations warrants further analysis. Nevertheless, the emission boundaries for our scenarios are reported in all diagrams displayed in **Figure 6** to represent the range of variability in our results: OEm-L P75 and REm-U P25 for the lower and upper bounds, respectively. CH_4 released during well preparation stages (i.e., excavators, well pad configuration and construction) are trivial when compared with total emissions generated by the whole chain. From well construction up until hydraulic fracturing (i.e., drilling activities, water, sand and equipment moved by trucks, fracking and well completion) a maximum of 0.5 Kt CH_4 for both countries are lost over the entire year, mostly concentrated at well completion. Strict emission mitigation measures deployed in the OEm can facilitate reductions by a maximum of circa 50%. CH_4 leaked or vented by compressors, valves, joints and gaskets, represents an important contributor under REm (19 to 33 Kt in Germany, 62 to 108 Kt in the UK), while consolidation of wells onto centralized well pads (more horizontal wells per vertical well; see also Robertson et al., 2018) combined with substitution of diesel engines with electrically-powered ones as foreseen in OEm limits losses at production site to ca. 3 and 11 Kt in Germany and the UK, respectively. Since uncombusted gas and wet seals in diesel and natural gas-powered compressors are the main sources of fugitive CH_4, replacement with electric compressors can eliminate emissions (Kirchgessner et al., 1997; Marchese et al. 2015 Supporting Information; Mitchell et al., 2015 Supporting Information). Similar reductions (of ca. 95%) can be achieved by implementation of dry-seal compressors with flaring systems (EPA, 2014b).

Wellhead compressors and liquids unloading (both with manual or automatic plunger lifts) have a very limited impact on total figures. The former are employed to increase the gas yield from low-pressure reservoirs, while the latter is a practice necessary to unclog the wells when

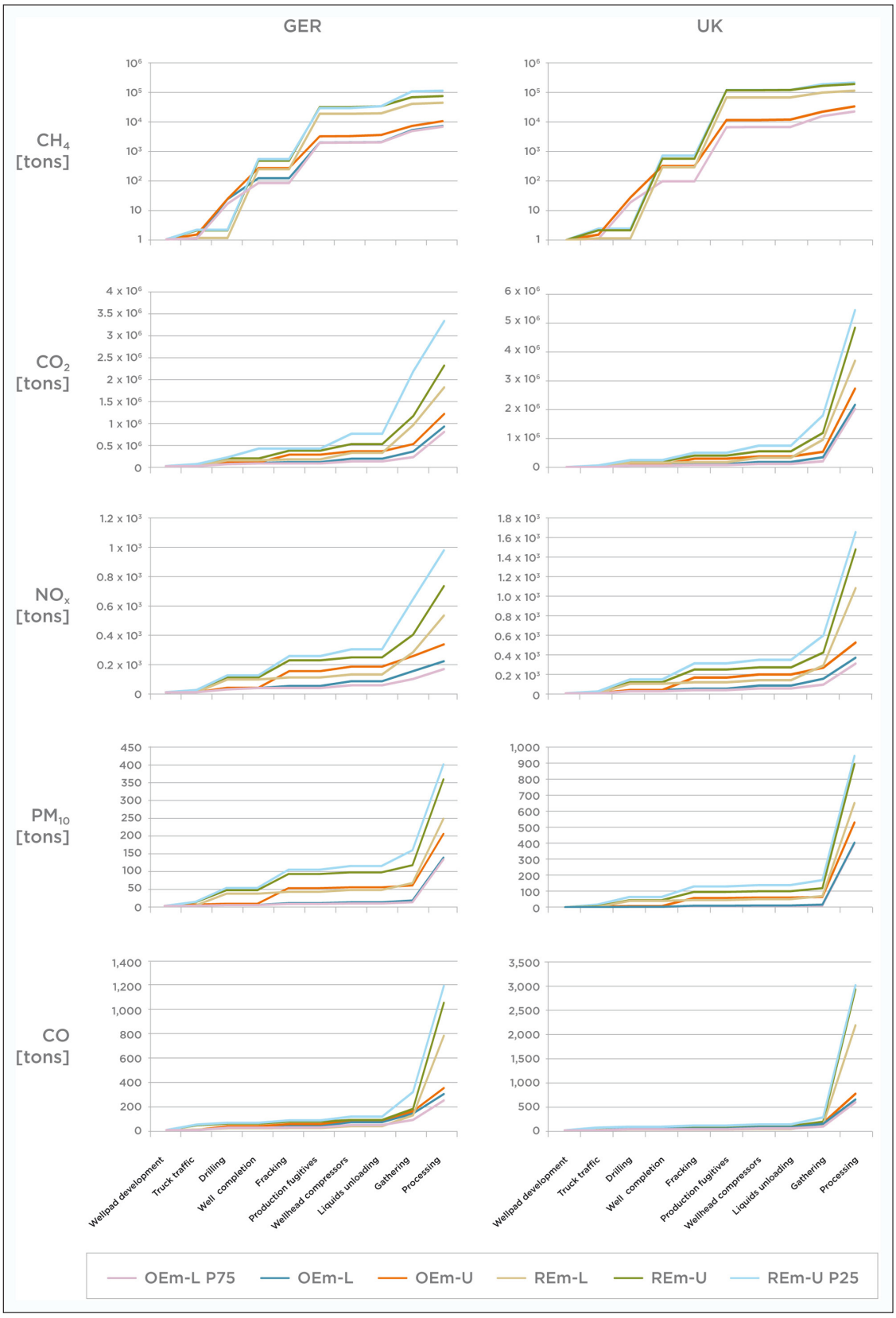

Figure 6: Annual cumulative emissions along the shale gas production chain. Results for CH_4, CO_2, NO_x and PM_{10} for Germany (GER, left) and the UK (right). REm and OEm are shown for the P50 case, while REm-U P25 and OEm-L P75 are also reported to show the emission boundaries. DOI: https://doi.org/10.1525/elementa.359.f6

large amount of liquids accumulate in the borehole. At the gathering stage, regardless of how compressors are operated, CH_4 losses (which include both gathering pipelines and gathering facilities, SM Text S1, Section S2.10) represent 35–48% of total emissions generated by the scenarios in Germany, and 24–41% in the UK. Here again, mitigation measures applied in OEm appear to be effective, decreasing CH_4 emissions by as much as 87% to 88% in Germany and 83% to 87% in the UK (low and high emissions boundaries, respectively). Because of the high emission factor associated with each gathering facility (Marchese et al., 2015; Mitchell et al., 2015) and also enforced in our emission exercise, reducing the number of gathering facilities collecting the gas from the production areas appear to contribute significantly to reduced gas losses. The number of these facilities is strictly linked to the geographical location of the *producing wells*, and to the technical feasibility and economic convenience to connect them to a single (large) or more (smaller) gathering plants. We assume one gathering facility for every 30 wells in REm, and one for every 80 wells in OEm (SM Text S1, Section S2.10.1). In the latter, emissions are mainly controlled by substitution of diesel to electric engines.

At gathering facilities, emission factor standards for diesel engines have negligible effects, while gas losses combined with the number of gathering facilities dominate this gas production stage contributing up to circa 90% in Germany and 70% in the UK of total CH_4 emissions. On the other hand, emissions from gathering facilities in OEm are significantly lower (circa 10% of CH_4 emitted in REm for Germany and between 20 and 30% for the UK), where gathering pipelines contribute to more than 65% in Germany and 87% in the UK of total CH_4 emissions in the gathering sector. It is worth noting that in REm-U P25 scenarios CH_4 emissions from gathering are particularly higher than the ones in P50 (50% higher in Germany and 35% in the UK), highlighting the impact that low well productivity (especially in Germany) has on gathering sector emissions. The prominent role of CH_4 emitted at gathering facilities and production sites finds confirmation in the literature (Zavala-Araiza et al., 2015; Balcombe et al., 2016; Littlefield et al., 2017). The gas processing stage, as characterized in our study (between 2.8% and 5.6% of gas burned for power production and turbines efficiency between 30% and 60%; SM Text S1, Section S2.11.2), comes only third in terms of CH_4 emissions contribution after the gathering and production stages in REm, while second in OEm. This is because best practices aiming to reduce the overall losses from processing plants are able to drive emissions down by about 50% overall. Other factors such as turbine efficiencies, combusted gas for energy needs and emission factors are, for this pollutant, irrelevant for both countries and scenarios. Gathering and processing of shale gas dominate total CO_2 emissions in all scenarios, spanning from 55% in OEm to 70% in REm. Mitigation measures such as electrification of all compressors and pumps applied in OEm at gathering facilities are particularly efficient to cut gas losses at this stage by ca. 75% in both countries. Similarly, the amount of gas burned to produce electricity and fulfil the energy needs at this stage – mainly driven by turbine efficiencies – can potentially reduce emissions between 30 and 40%. Wellhead compressors and fracking are next in order of importance although they only account for 2 to 15% of total CO_2 emissions in almost all scenarios for both countries.

GHG emissions national contexts

Here we compare our results of GHG emissions from shale gas with emission inventories supplied to the United Nation Framework Convention of Climate Change (UNFCCC) for the energy industrial system (i.e., power and heat production, petroleum refining and manufacture of solid fuels). For this scope, we select the year 2012 for Germany and 2015 for the UK as reported in the National Inventory Report (NIR) year 2017 submission, since the conventional gas domestically produced in these years is similar to the ones assumed in the scenarios: 10.7 bcm for Germany and 34.4 bcm for the UK.[3] Focusing on the UK, CH_4 and CO_2 generated by the well-preparation stage till processing (namely, upstream) of the current natural gas industry only contributed 7.0% (CH_4) and 0.4% (CO_2) of total emissions from the industrial energy sector for the UK. For the former, shares from the OEm P50 scenarios of total gas released from the energy sector are similar in magnitude, while emissions under REm P50 settings achieve 30 (REm-L) to 65% (REm-U) of reported current datasets for the UK (up until 70% under P25). All results are reported in **Table 4**. Most of the offshore gas produced in the UK requires processing (UNFCCC, NIR for the UK, year 2017 submission) due to its variable but still notable content of impurities like CO_2, nitrogen, ethane, and so on (Cowper et al., 2013). The conservative emission estimates from the current UNFCCC Report may be justified assuming that best practices for CH_4 capture are all in place and properly performed, keeping them down to a level comparable with the lowest depicted by our scenarios. On the other hand, the CO_2 relative contribution to emission from the energy industry raises from 0.4 to 1.6% when comparing results from the UNFCCC Report with OEm or until 3.5% with REm. Therefore, even under our most conservative scenario contemplating the highest combustion turbine efficiency and the lowest combustion rate of gas during high-emitting processing activities, CO_2 emissions reported by the UNFCCC are about one fourth of these. In Germany, CH_4 emitted from the natural gas upstream system contribute about 0.6% of the total emitted by the national energy system, while CO_2 only 0.4%. This is mostly due to the high consumption of solid fuels in the country that brings coal (and lignite in particular) far to the top of the list of emitters: CO_2 released from natural gas combustion are similar for the two countries, while those generated in Germany by solid fuels are four times more than in the UK (UNFCCC, NIR submission 2017 for both countries). CH_4 emissions produced by our scenarios raises contributions to a range of 1.6 to 2.4% of total energy from the industrial system emissions in OEm, and up to 10.3%–17.4% in REm. This means that the natural gas sector is an important contributor requiring appropriate attention by regulators when prescribing technologies, monitoring and verification systems, in Germany

Table 4: Annual CH_4 and CO_2 emissions generated by our shale gas industry for Germany (GER) and the UK. Results are compared with emissions of the current upstream natural gas chain and the energy (heat and power) sector as described by the UNFCCC. Please note that the term "fugitive" in the UNFCCC reference relates to all methane emissions associated to that specific stage. DOI: https://doi.org/10.1525/elementa.359.t4

Species	Fuel combustion emissions – Energy industry (UNFCCC, 2017). In Kt.	Fugitive emissions fossil fuels (UNFCCC, 2017). In Kt.	Emissions gas upstream (UNFCCC, 2017) and share	OEm P50 results as share of current emissions (Range boundaries). In Kt.	REm P50 results as share of current emissions (Range boundaries). In Kt.
GER CH_4	89.9	366	2.7 (0.6%)	7.4 (1.6%) to 11.0 (2.4%)	46.4 (10.3%) to 78.5 (17.4%)
GER CO_2	359,000	2960	1590 (0.4%)	924 (0.3%) to 1213.0 (0.3%)	1830 (0.5%) to 2330 (0.6%)
UK CH_4	12.1	258	19.5 (7.0%)	21.5 (7.9%) to 31.3 (11.6%)	104 (30%) to 175 (65%)
UK CO_2	133,000	4560	514 (0.4%)	2,190 (1.6%) to 2,750 (2.0%)	3,726 (2.7%) to 4,880 (3.5%)

as in the UK. On the other hand, CO_2 generated by the German shale gas industry maintain emissions from 0.2 to 0.6% (**Table 4**), to a maximum of 0.9% in the REm P25.

Of total gas produced in Germany, 40% has high sulfur content (sour gas) that has to be discarded by specific treatments before the gas can access transmission lines. In the UNFCCC NIR submissions 2017 for Germany, CO_2 and CH_4 emission factors for removing sulfur are 336 and 0.11 kg per 1,000 m^3 of treated gas respectively. The only CO_2 and CH_4 emissions associated with the processing stages of gas reported are those related to the treatment of sour gas, while no other emissions attributed to pre-treatments occurring at pumping stations (such as water, hydrocarbons and solid removals), are listed. We therefore assume that in the current gas extraction industry, no CO_2 or CH_4 emissions are associated with, or rather expected from, pre-treatments, an aspect for which we believe deserves further investigation into the reliability of such an assumption.

Despite the relevance of discussing our results in the context of national inventories, it is challenging to explain the inconsistency in the results for REm (and from the US) with the emissions reported for Germany and the UK under the UNFCCC. A study of CH_4 emitted from the Groningen field in the Netherlands by Yacovitch et al. (2018) also struggled to provide an explanation for the large emission discrepancy between their campaign observations and national inventories. The authors of the same study believe that major differences between North American and European estimates cannot be ascribed to the large-scale adoption of hydraulic fracturing in the former. Riddick et al. (2019) also report high and unreported CH_4 losses from oil and gas wells in the North Sea, criticizing bottom-up methods and self-reporting by operators as an improper practice. Our findings displayed in **Figure 6** also show that emissions generated at stages that are specific to shale gas activities (i.e., well completion and fracking) have only a minor effect on total CH_4 losses when RECs are in place (SM Text 1 Section S2.5 and S2.6). Most critical CH_4 and air pollutant sources across the gas chain have been attributed to above-ground malpractices, failures or malfunctions unrelated to the gas nature (i.e., conventional or shale gas), as reported by the studies produced by the Environmental Defense Fund initiative and others (Sauter et al., 2013; Elsner et al., 2015; Omara et al., 2016; Atherton et al., 2017). Based on the latest evidence, gas capture solutions, "detection and repair" services, as well as monitoring and early detection of super-emitters are the most likely key measures when it comes to effectively mitigate emissions for both gas sources (EPA, 2014a; Westaway et al., 2015; Ravikumar and Brandt, 2017; Zavala-Araiza et al., 2017; Konschnik and Jordaan, 2018). Unfortunately, surveys that investigate European CH_4 losses in a transparent and systematic way (e.g., peer-reviewed articles published by independent research bodies) do not exist or are not publicly available, raising doubts over the accuracy and objectivity of emission estimates provided to the UNFCCC (EC, 2015; Larsen et al., 2015; Cremonese and Gusev, 2016; Riddick et al., 2019). To facilitate identifying CH_4 emissions specifically from a future European shale gas industry, for instance Visschedijk et al. (2018) propose an atmospheric ethane monitoring system. Because of these research gaps, we find it hard to justify such a large discrepancy by citing technological, regulatory or geological factors alone. The results shown in OEm are instead much more similar to European national inventories. Namely, the leakage rate as calculated by data from the UNFCCC NIRs is as low as 0.02% for Germany and 0.08% for the UK. Based on our discussion and results, these estimates may be justified by systematic employment and application of best technologies/performances across each stage of the preparation and supply gas chain – a rather unlikely circumstance. Given the fact that there is no transparent information available on the quality of these estimates, we speculate that they could be based on very optimistic assumptions (i.e., as the ones we apply in OEm) instead of systematic and integrated monitoring campaigns (see also discussion in Riddick et al., 2019).

CO_2 and CH_4 contribution to total GHG emissions

The warming-related contribution of CH_4 – a much more potent GHG than CO_2 – is subject to the time frame of observation. This effect is controlled by the oxidation of CH_4 to CO_2 ($t_{1/2}$ ~ 12years; Myhre et al., 2013), so that its warming component in the atmosphere decreases

with time. Two Global Warming Potentials (GWPs) are commonly used in the scientific community: the GWP_{100} and GWP_{20}, with the numbers referring to the warming implications over those periods of time, respectively 20- and 100-years (IPCC, 2014). While these two parameters are complementary and can offer a comprehensive overview on the implications of this pollutant in the short- and long-term, one indicator may be preferred instead of the other according to the scope of a specific research exercise (Ocko et al., 2017; Balcombe et al., 2018). Total aggregated GHG emissions (CO_2-eq) from the shale gas upstream sector and covering all well productivity cases are shown in **Figure 7**, applying both 20- and 100-year periods.

On a mass-to-mass basis, CO_2 emissions are significantly higher than CH_4 because of the large amount of natural gas combusted to CO_2 at processing plants to fulfil the power demand (SM Text S1, Section S2.11.2). Our aggregated upstream shale gas industry displays much higher variability between REm and OEm, than between analysis under GWP_{20} and GWP_{100}. Results in OEm are comparable and between 3.8 and 5.3 Mt y^{-1} (minimum values of their ranges), while much higher in REm: between 19.6 and 36.0 Mt y^{-1} (maximum values of the ranges).

CH_4 represents a major contributor accounting for more than 50% of total CO_2-eq. when attributing a GWP_{20} under all scenarios (with a peak of 76%), while between 22 and 55% assuming a GWP_{100}. REm and OEm differ based on the performance of practices and implementation of diverse technologies to monitor and control CH_4 losses on the one hand, and to increase efficiency of engines and compressors so as to curb emissions of CO_2 on the other hand. It is evident here that, although CO_2 emissions can be cut up to ca. 60% (difference between the REm and OEm CO_2-bars in both GWP scenarios), CH_4 reduction measures are by far more effective and can technically reduce CO_2-eq. CH_4 emissions by 92%. Full compliance with the overall settings applied in OEm has notable climate benefits on reducing aggregated CO_2-eq. volumes by a factor of maximum 5 in the GWP_{100} case, and by a factor of maximum 7 in the GWP_{20} case.

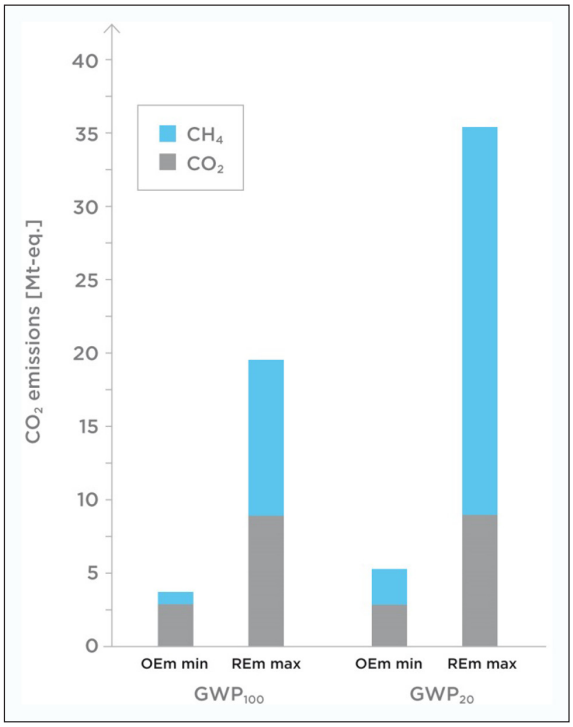

Figure 7: Annual aggregated CH_4 and CO_2 emissions in CO_2-eq. Maximum and minimum aggregated GHG emissions from REm and OEm are relative to all productivity cases (P25, P50 and P75) under different time horizons (GWP_{20} and GWP_{100}). Values refer to emissions produced from well preparation till processing (upstream sector). DOI: https://doi.org/10.1525/elementa.359.f7

Methane leakage rates in the international context

Figure 8 shows the CH_4 losses already reported in **Figure 5**, in relation to overall CH_4 production and expressed in percentage for all productivity and technological scenarios. Accordingly, CH_4 emissions from associated activities are not considered here. The Figure also illustrates the effect of different operation and technologies/performances on final leakage rates, the large variance within the OEm range, and the strong correlation between well productivity (the P-cases) and the extent of CH_4 emissions (see in particular Germany OEm P25). Although the leakage rates resulting from our emission scenarios vary considerably, results in REm are within the range of the estimates reported by the latest regional and nationwide studies carried out worldwide (Littlefield, 2017; WEO, 2017; EPA, 2018), or are of similar magnitude (Zavala-Araiza et al., 2015; Alvarez et al., 2018).

Studies based on single measurement campaigns (cross-sectional data) or focused on restricted areas may be inappropriate as a basis of comparison to nation-wide or nationwide emission estimates such as our results. Based on the results of Zavala-Araiza et al. (2017), a skewed emissions distribution generated by the irregular occurrence of super-emitters implies local leakage rates that are inconsistent (i.e., lower in the case that no super-emitters exist in a restricted area or higher if they are over-represented) with the mathematical mean representing a larger area under analysis. Skewed distribution might be caused by the age of a restricted population of wells (see, for example, Omara et al., 2016) or by lax state regulations and poor monitoring campaigns. For this reason, in the following we compare our results with regional or nationwide emission studies. Our scenario results are lower than most estimates from the US: for example, the EPA CH_4 emissions assigned to the gas upstream sector are slightly below the leakage rate of 0.9%, which is similar to the P50 upper value of REm for Germany, but higher than all the results produced by the UK scenarios (up to a maximum of 0.7%; **Figures 8** and **9**). It is here worth noting that the gas vented during well completion practices estimated by the EPA GHG Inventory 2016 are much higher than the amount we assign (SM, Text S1, Section S2.6). Littlefield et al. (2017) carried out a multi-basin analysis on old and new emission data in the US and processed

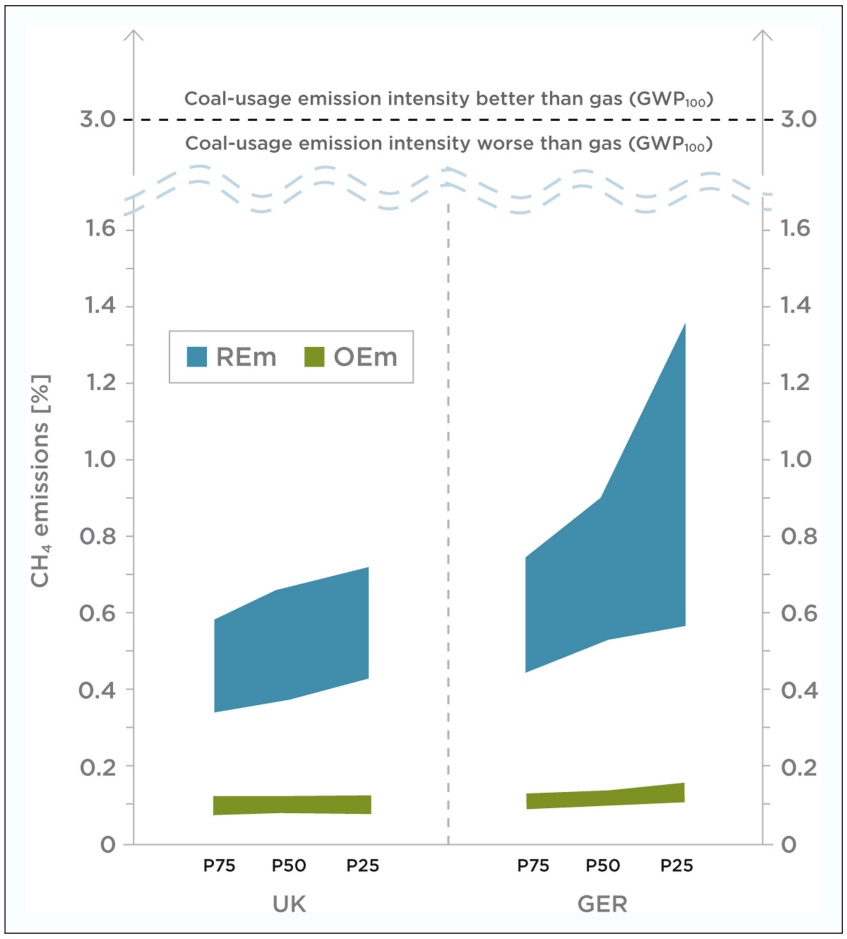

Figure 8: CH_4 leakage rates (in %) for all scenarios for Germany and the UK. The boundary depicted at % leakage: 3% defines the maximum gas leakage rate that still guarantees the climate benefits of gas over coal (WEO, 2017). DOI: https://doi.org/10.1525/elementa.359.f8

them in a Monte Carlo simulation to estimate the amount of CH_4 lost along the entire natural gas supply chain. Their study found a leakage rate of 1.23% (with a 95% confidence interval from 0.72 to 1.59%) of the total gas delivered from the wellhead up to the processing stage, a value close to our highest simulated leakage rate (Germany REm P25, **Figure 9**). Zavala-Araiza et al. (2015) estimated an even higher leakage rate at the Barnett gas field – the most investigated gas basin in terms of GHG and pollutant emissions in the entire US – up to 1.5% (confidence interval: 1.2 to 1.9%) as a result of the convergence of top-down and bottom-up measurements within the statistical confidence range.

A recent study that aggregated the results produced by a large number of investigations in the US (Alvarez et al., 2018) estimates national-wide CH_4 emissions from the natural gas supply chain to be 2.3%, with the upstream sector accounting for about 85% of this value (i.e., 1.9%; +0.4/−0.3%). To run the computational model and define nationwide estimates, Alvarez et al. (2018) also included emissions from abandoned wells and flares (with the latter only partially included in our activity data) as well as estimates from studies carried out in different regions. Their leakage rate is far higher than our highest estimate (i.e., REm-U Germany: 1.36%). In their study, production sites account for ~70% of total upstream CH_4 losses, compared to 30 to 40% in our scenarios. Worldwide, our estimates are consistent with official reports: According to the IEA (WEO, 2017), the average leakage rate in the upstream gas system amounts to 1.14% of total gas produced.

The bottom-up approach employed to develop the emission scenarios (i.e., aggregating process-level emissions) may lead to some relevant emitting sources being overlooked (Brandt et al., 2014), a factor that may partially explain the conservative nature of our results. The technological innovations discussed in the drilling projections, such as the aggregation of 30 wells on single producing pads (not observed in any case study at present), together with the exclusion of forbidden/unexpected gas recovery procedures in Europe can be of helps when it comes to interpreting results. Moreover, well productivity – as demonstrated by our results – shows to have a significant influence on total CH_4 emissions. The EUR_{well} across all cases is highly uncertain (i.e., gas-in-place and the TRR have never been empirically tested), and lower values (if

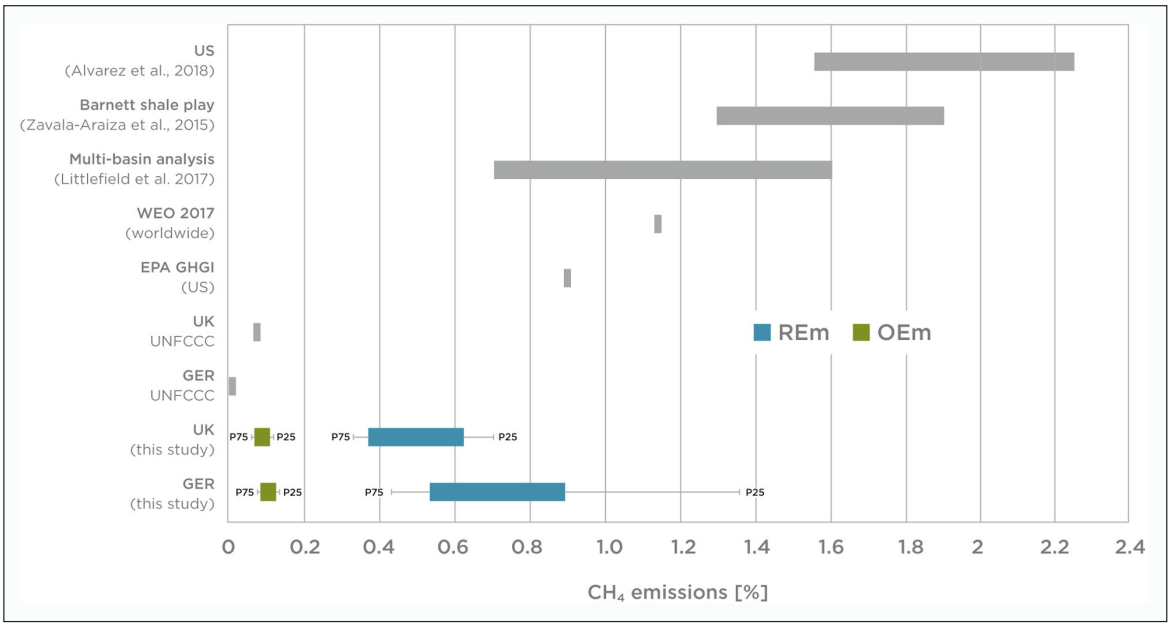

Figure 9: Collection of upstream CH_4 leakage rates of worldwide natural gas systems. The figure includes the most accredited studies investigating CH_4 leakages and compares them with the REm and OEm for shale gas scenarios in Germany (GER) and the UK (Zavala-Araiza et al., 2015; Littlefield et al., 2017; WEO, 2017; Alvarez et al., 2018; EPA, 2018; UNFCCC NIR submission 2018 for Germany and submission 2017 for the UK). All leakage estimates are normalized to the total gas produced, except for Littlefield's study (gas delivered). DOI: https://doi.org/10.1525/elementa.359.f9

determined) would surely increase the CH_4 leakage rates found in our study. It is also plausible that the source studies selected to define our emission factors do not wholly and/or comprehensively account for super-emitter contributions. Despite our tailored literature review criteria, this issue is realistic due to the random distribution in space and time of fat tail emissions.

Emission intensity

Despite secondary applications, e.g., the transport sector and homes, most natural gas in Europe is combusted at large power plants to fulfil the electricity and heating demand of households and industry. Its ultimate environmental impact is therefore bound to industrial performance during its extraction, processing, and transformation into accessible energy (e.g., power and heat) and final delivery to the consumers. Based on shale gas emitted from the different scenarios presented in the previous sections, here we determine their emission intensity (EI) in gCO_2-eq. kWh^{-1} of electricity produced and discuss how these relate to emissions generated by other sources such as coal, conventional natural gas utilization, and to power production distributed in the German and UK electricity grids (**Figure 10**).

Within a 100-year period, the electrical power EI of a shale gas industry ranges between 394 and 403 gCO_2-eq. kWh^{-1} in OEm (5.3% variability), and between 416 and 456 gCO_2-eq. kWh^{-1} in REm (12.6% variability). This discrepancy is more pronounced under a 20-year period: the same EI varies from 400 to 412 gCO_2-eq. kWh^{-1} in OEm (4.8% variability) and from 443 to 517 gCO_2-eq. kWh^{-1} in REm (16.7% variability). Gas databases from North America show that gas compositional ranges are unrelated to the gas nature (i.e., conventional vs. unconventional gas), so we can fairly assume the same energy potential when transforming these two types of gas to electrical grid power. Efficiency in overall German gas power plants is nowadays 53% after disaggregating the heat energy component (Icha and Kuhs, 2018). Our power EI results showed by REm are slightly lower than Life Cycle Assessment (LCA – a methodology aiming to assess environmental impacts of a product or service during its entire life cycle) data available in the literature on the US (Jang et al., 2011; Stephenson et al., 2011; Burnham et al., 2012; Laurenzi and Jersey, 2013; Skone et al., 2014) and the UK (Stamford and Azapagic, 2014). In fact, our results under REm overlap most of the lower GHG emission ranges reported by these studies. Differently, the emission intensity of shale gas-fired power plants in China is estimated at 625 gCO_2-eq. kWh^{-1} (Qin et al., 2017). Nevertheless, due to different impact categories analyzed (broader for LCA exercises) and the unclear EUR of European shale gas wells, the scientific significance of a direct comparison between our EI results and these studies is limited. In 2016, an emission factor of 382 gCO_2-eq. kWh^{-1} is attributed to the energy consumed and generated by natural gas in Germany, less than half of what is emitted per kWh by hard coal utilization (847 gCO_2-eq. kWh^{-1}) and just one-third of the value for lignite (1150 gCO_2-eq. kWh^{-1}; Icha and Kuhs, 2018). The EI offset between natural gas and coal observed in **Figure 10** well resembles the findings of the natural gas vs. coal intensity explanatory model

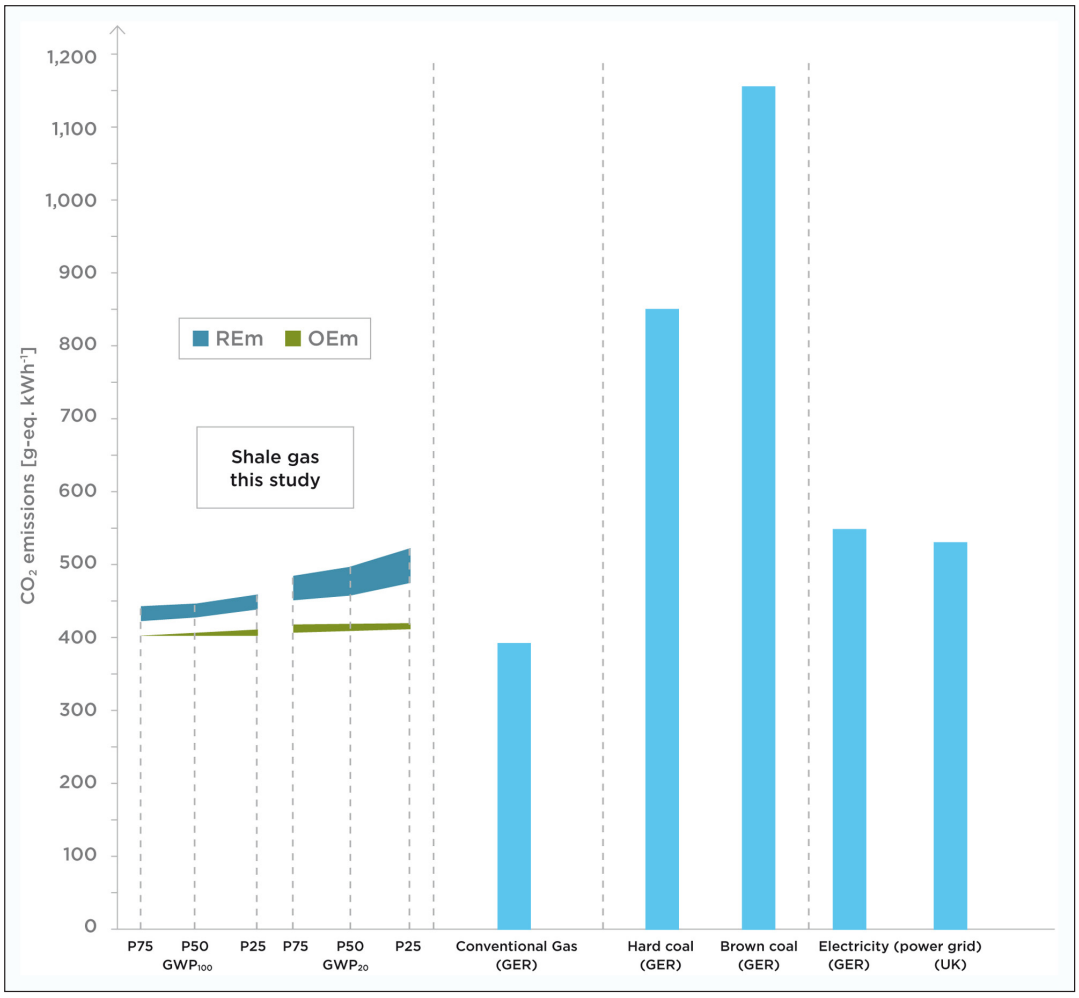

Figure 10: Comparison of emission intensities. Our results are compared with conventional gas (Germany, GER), hard coal (GER), brown coal (GER) and electricity production (GER and the UK). Results are shown in gCO_2-eq. kWh^{-1}. Grid losses are included in all cases. Natural gas energy density: 38.3 MJ m^{3-1}; Natural gas power turbine efficiency: 53% (Icha and Kuhs, 2018); Average indirect/WTT emission factors for fuels resulting from the production, transport and distribution: BEIS, 2016; Power grid losses: 9% own calculations based on results showed in BEIS (2016). DOI: https://doi.org/10.1525/elementa.359.f10

presented by the IEA (WEO, 2017), which illustrates how gas has a stronger impact on the climate compared with coal only at leakage rates above 3% (see **Figure 8**). In China instead, due to inadequate CH_4 recovery at coal power plants, this boundary is about 6% and 12% when attributing a GWP_{20} and a GWP_{100} respectively (Qin et al., 2017). Similarly, our results on the coal-gas comparison well resemble data from previous studies (Stamford and Azapagic, 2014). Nevertheless, as we critically discussed in a previous section, CH_4 losses associated with current gas production in Germany (see **Figure 5**) and in some of other countries exporting to it (i.e., Netherlands and Norway) are estimated as much lower than the ones forecasted in this study (Larsen et al., 2015; Cremonese and Gusev, 2016), a factor that alone drives the shale gas EI values beyond present national estimates of gas production. Accordingly, the EI of our European shale gas industry appears higher than the one from conventional gas combustion in Germany and the UK, spanning in a range from 3% (OEm-L P75, GWP_{100}) to 35% (REm-U P25, GWP_{20}). Moreover, it must be pointed out that CH_4 emissions at Russian producing fields – providing between 40% and 50% of natural gas consumed in Germany – and along Russian-European transmission lines are together responsible for a loss of about 1.3% of the total gas shipped (Cremonese and Gusev, 2016), an aspect that is only partially reflected in the German national emission databases and ultimately to EI estimations. Taking this into account would very likely imply higher natural gas emitted in Germany. For the same reason pertaining to low accuracy and reliability of data, CH_4 losses on transmission lines connecting processing plants and combustion sites in the future European shale gas scenario are not considered in our analysis. Moreover, a future European shale gas industry will necessitate the construction or refurbishment of new pipeline systems in conformity with best standards and environmental regulations available at that point in time. Under the assumption that power plants lie nearby

or within the producing shale gas regions in Germany and the UK, it is reasonable to expect minor losses along these structures that would therefore not affect the shale gas EI to any considerable degree. Taking these plausible assumptions into consideration, including inaccurate estimates of losses along natural gas transportation lines in our emission scenarios would not enhance its overall precision or better reflect reality.

Based on our comparison, EI of a shale gas industry is instead slightly below emissions associated with power production in Germany and the UK, indicating how carbon intensive fossil fuels still dominate emissions in this field despite the expansion of renewables and the still relevant share of nuclear power. At present, power produced in Germany and the UK by the national energy system and distributed by the electricity grid is currently equal to gCO_2-eq. kWh^{-1} and 528 gCO_2-eq. kWh^{-1}, respectively. These values include grid losses and are based on original datasets from the Department of Business, Energy & Industrial Strategy (BEIS, 2016) and Icha and Kuhs (2018).

Other pollutants

Volatile Organic Compounds (VOCs)

VOCs are highly-volatile carbon-based compounds which are generated by both natural (biogenic) and anthropogenic sources. Their emissions can be harmful to human health and the environment due to their role as a precursor in tropospheric (ground-level) O_3 formation. Natural unprocessed gas typically consists of 75–90% CH_4 by volume, while the rest is largely composed of VOCs (e.g., Baker et al., 2008, Gilman et al., 2013, Faramawy et al., 2016). Therefore, when CH_4 leakage occurs stemming from unprocessed gas, this also implies VOC leakage.

It is worth noting that the only difference between wet and dry gas scenario results presented in our study is the amount of VOCs released (i.e., CH_4:VOCs ratio in the raw gas), while all other species are unaffected. In the P50 wet scenarios, total VOCs emissions range from 3.8 to 38.5 Kt for Germany, and 10.7 to 85.8 Kt for the UK (**Figure 11**). Due to the notably lower well productivity in the P25 wet scenarios (especially for Germany), VOCs are circa 50% higher (58.3 Kt) in Germany and 12% (96.2 Kt) in the UK when compared with the P50 scenario. In the emissions scenario under P75, VOC losses decrease only minimally compared with the P50 case. As expected, VOC emissions are significantly lower under the dry gas scenarios: for P50, VOC loss ranges from 1.0 to 10.6 Kt in Germany, and 2.9 to 23.6 Kt in the UK (**Figure 11**).

Variations in VOC losses under the P25 the P75 cases for the dry gas scenario are proportional to the wet gas scenario.

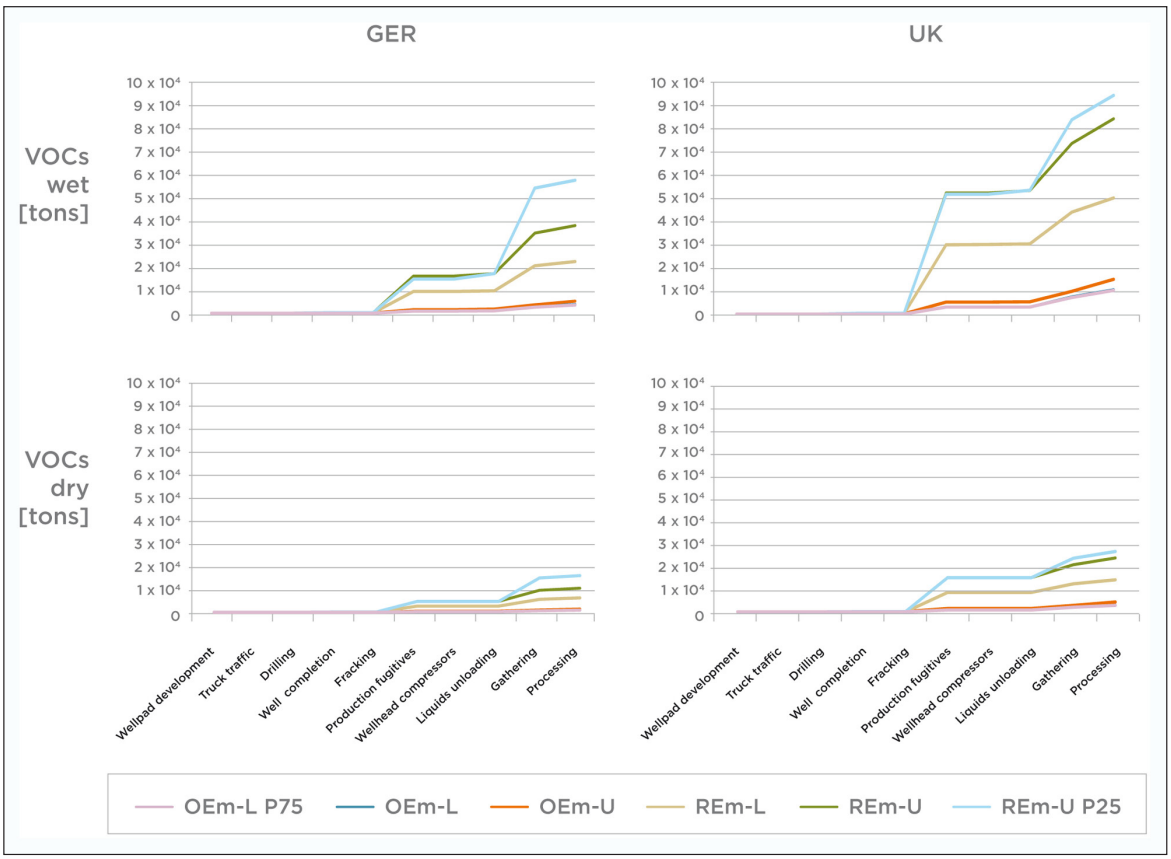

Figure 11: Cumulative annual emissions along the shale gas production chain. Results for Germany (left) and the UK (right) in tons. REm and OEm are shown for the P50 case, while OEm-U P25 and REm-L P75 are also reported to show the emission boundaries. DOI: https://doi.org/10.1525/elementa.359.f11

VOC emissions in both Germany and the UK have been decreasing over the past few decades (EEA, 2016). As reported in **Table 5**, total VOC emissions emitted by the energy sector were 83 Kt for Germany and 129 Kt in the UK for the year 2016 (NAEI, 2018; UBA, 2018c). This means that our results are equivalent to 4.5 to 46.2% of total annual VOCs emissions from the German energy system, and 8.3 to 66.5% for the UK (for all scenarios, including wet and dry). Nevertheless, it is worth emphasizing here the dominant role that VOC concentration in the natural gas can play when estimating total VOCs outputs. The sectors which are the most consequential for VOC leakage in shale gas production are the same as for CH_4, being these two compounds co-emitted, i.e., production, gathering and processing. These sectors vary in level of contribution depending on the country and scenario. For example, gathering is by far the most crucial sector for VOC emissions in Germany under the OEm-U P25 scenario. This is mostly due to the low productivity of German basins, which requires a greater number of wells to be drilled and in turn a greater number of gathering facilities and associated emissions. Based on these values, the VOCs emissions in these scenarios have the potential to notably impact local and regional air quality through O_3 production.

Nitrogen oxides (NO_x)

NO_x are exhaust by-product emitted by engines during combustion of fuels and a precursor to O_3 which negatively impacts human and environmental health. In our emission scenarios, NO_x are mostly produced during fracking, at gathering plants and during gas processing due to the extensive employment of diesel engines at these stages (**Figure 6**).

In the P50 scenarios, NO_x emissions range between 2.2 and 7.4 Kt in Germany and between 3.7 and 14.9 Kt in the UK. NO_x under P75 conditions display lower emissions by ca. 25% in Germany with respect to the lowest level reported under P50. Similarly, a lower value of about 25% is evidenced under P25 conditions with respect to the highest emissions under P50. In the UK, under P25 and P75 smaller variances are shown with respect to the P50 range boundaries, demonstrating a weaker dependence of emissions on well productivity. Our results show that the number of fracking stages necessary to exploit the shale reservoir under each well pad is mainly responsible for NO_x emissions at the well site, and a lower number of those can reduce emissions by up to a factor of five. Nevertheless, it is worth pointing out that the number of fracking stages is strictly dependent on the local geological characteristics of the reservoir which are unknown up until the drilling phase starts and cannot be controlled. Emission reductions systems employed in REm are particularly efficient at gathering stations, where the number of facilities is the main factor responsible for curbing NO_x release with respect to emission factors of engines or electricity-driven motors. During processing, high efficiency combustion (>50%) and high-performing water-steam injection gas turbines employed in OEm can together cut NO_x emissions up to 80%.

NO_x emitted from our scenarios account for only a small fraction of the national emissions associated with the energy sector. In both countries examined, NO_x emissions show a slow but constant decrease since the 1980s, with a parallel decrease in atmospheric concentrations (Minkos et al., 2018; NAEI, 2018). These trends follow more stringent regulations especially in the transport sector, which is the greatest contributor to total NO_x emissions. Energy production systems released 260 Kt of NO_x in Germany and 125 Kt in the UK in 2016, so that a potential shale gas industry as envisaged in this study does not have a significant impact on national inventories or on background levels (contributing 0.8/2.8% in Germany and 3/12% in the UK). A complete overview of NO_x national emissions and results from our study is reported in **Table 5**. Nevertheless, clustering of large emitters such as new well sites, gathering or processing plants may severely increase NO_x concentrations nearby so that it might have a considerable impact on air quality e.g., O_3 production, calling for smart industrial planning and appropriate prevention measures for these facilities. Gas flaring is also another important source of NO_x, and atmospheric concentration of these species can be significantly affected by this activity

Table 5: Annual air pollutant emissions from this study compared with national databases. All values are reported in Kt. The energy system includes public electricity and heating generation. [1]UBA (2018c); [2]NAEI (2018); [3]NAEI (2018) (stationary combustion sector); [4]Minkos et al. (2018); [5]UBA (2018a); [6]UBA (2018b). DOI: https://doi.org/10.1525/elementa.359.t5

Pollutant species		Emissions, this study (shale gas industry, P50 case)				National emissions (total)		National emissions (energy system)	
		GER		UK		GER [Kt]	UK [Kt]	GER [Kt]	UK [Kt]
		OEm [Kt]	REm [Kt]	OEm [Kt]	REm [Kt]				
VOCs	Wet	3.8–5.3	22.7–38.5	10.7–15.3	51.0–85.8	1,050[1]	821[2]	83[3]	129[2]
	Dry	1.0–1.5	6.3–10.6	2.9–4.2	14.1–23.6				
NO_x		2.2–3.3	5.3–7.4	3.7–5.3	10.9–14.9	1,220[4]	893[2]	260[2]	125[2]
PM_{10}		0.1–0.2	0.3–0.4	0.4–0.5	0.7–0.9	203[5]	170[2]	11[5]	3[2]
CO		0.3	0.8–1.0	0.6–0.8	2.2–2.9	2,840[6]	1,490[2]	148[6]	418[3]

(Duncan et al., 2015; Li et al., 2016). We did not attribute any emissions to flaring activities in our shale gas production system (with only a few exceptions, i.e., safety), although some of the top-down source studies selected to assign emissions may include this in their aggregate results. Our choice to exclude flaring in future scenarios is in accordance with worldwide initiatives such as the "Zero Routine Flaring by 2030" by the World Bank, and because of this CH_4 emissions presented here may lack a relevant source when comparing them to real case-studies.

Particulate matter (PM)

PM is an air pollutant which has negative implications for health and climate change. These particles are generated by several natural processes, while the industrial and transport sectors are the dominant anthropogenic sources (AQEG, 2012). In Germany, total annual PM_{10} emissions from the power and heat industry in 2016 registered at 11.1 Kt (UBA, 2018a), and 3.3 Kt in the UK (NAEI, 2018). An overview on PM10 emissions is reported in **Table 5**. The lower amount of coal usage in the UK energy mix may be responsible for the notable difference in PM emissions from this sector between the two countries: 8.5 Mtoe in the UK vs. 39.3 Mtoe in Germany (BMWi, 2018; GOV.UK, 2019). PM_{10} and $PM_{2.5}$ emissions are nearly identical in our study, because the majority of PM released by diesel engines and turbines are within the $PM_{2.5}$ range, which by definition is also within the PM_{10} range. Therefore, from now on we refer to PM_{10} only.

In our emission scenarios, PM_{10} is produced by diesel engines (locally) and indirectly by electricity consumption from machineries at all stages. As shown in **Figure 6**, PM_{10} emissions in the P50 scenarios fall within the range of 0.2–0.4 Kt (REm) and 0.1–0.2 Kt (OEm) in Germany, and 0.6–0.9 Kt (REm) and 0.4–0.5 Kt (OEm) in the UK. Results from the P25 and P75 scenarios do not show remarkable differences beyond this range. Although these values contribute less than 3.6% of total PM emitted by the German energy sector, in the UK they contribute up to 27%. Addressing their potential health and environmental implications identifying best shale gas production practices is therefore required. PM_{10} emissions are mainly produced during the gas processing stage (>70% of total volumes), and to a minor extent during drilling, fracking and gathering operations (in order of importance). PM_{10} emitted by the intensive truck movements from gas exhaust, tires, brakes and road wear affect total emissions only minimally, though they may have significant effects at the local level. High turbines efficiency at processing plants applied in OEm ameliorates PM_{10} emissions to a minor extent. Although drilling does not have a considerable impact on total emissions, it is worth noting that substitution of diesel engines with electrical motors powered by the electrical grid power has the potential to reduce emissions by a factor of 16 from this stage, which may signal significant implications for air quality at well sites. Any reduction of diesel engines usage at any stage of the production chain is linearly related to the decrease of total PM_{10} emissions. An overview or results is presented in **Table 5**.

Carbon Monoxide (CO)

CO is an odorless and colorless air pollutant produced by natural as well as anthropogenic processes (Khalil and Rasmussen, 1990; Guenther et al., 2000). CO emissions from all sectors in the UK have been steadily decreasing since 1990 (NAEI, 2018), and in the last years over half of its emissions are attributed to residential sector combustion (414 Kt in 2016) and stationary combustion (418 Kt, 28% of total emissions; NAEI, 2018). In Germany, total CO released in 2016 decreased by more than 70% compared with values reported in 1990, with the energy sector currently responsible for 148 Kt (5.2% of the total) (UBA, 2018b). All CO national emissions and our results are reported in **Table 5**.

Our results show that CO, likewise to NO_x and CO_2, is only emitted at production stages where engines, turbines or motors burning diesel or natural gas are employed. We assume that no CO is present in the shale gas based on the speciation we apply from Faramawy et al. (2016). Accordingly, the processing stage dominates CO emissions in Germany accounting for more than 50% in OEm and more than 80% in OEm, followed by gathering stage which accounts for less than 25% of CO released in all scenarios. These shares are more extreme in the UK, where processing accounts for more than 90% of total CO emissions in REm and almost 80% in the REm (see **Figure 6**). The large volume of gas combusted to provide power and in turn process the shale gas is therefore responsible for most of the CO released by the gas production industry. CO volumes as calculated in REm (between 2.2 and 2.9 Kt for the UK and between 0.8 and 1.0 Kt in Germany) and in OEm (between 0.6 and 0.8 Kt for the UK and 0.3 Kt for Germany) are considerably lower than values currently attributed to the power and heat production sectors in both countries (see above).

Conclusions

In this study we investigated: i) several development pathways of an upstream shale gas industry in Germany and the UK; ii) the GHG and air pollutant emissions resulting from these different pathways; and iii) the potential of different practice performances and technological solutions to control losses and ensure best climate standards. Based on shale gas reservoir estimates in these two countries, an industrial development that is able to maintain current gas production volumes for the next decades is within reach. Accordingly, our drilling projections show that a constant annual drilling rate of 480 wells in the two countries can lead to a flourishing and mature shale gas industry in one to eight years. Our investigation into the climate and atmospheric repercussions associated with such an industry quantifies emissions in the shale gas basin regions under business-as-usual (REm) and best-technical-case (OEm) frameworks, with the latter representing the minimum emissions achievable according to the technical boundaries. Well productivity, which cannot currently be predicted, has proven to be an important factor in the total emissions released. Although it is not possible to influence this variable through activity performance

and regulations, by considering all well productivities we have a complete overview of the extreme boundaries of our emission projections. Based on the drilling scenarios, the amount of shale gas produced annually is 11.58 bcm in Germany and 36.62 bcm in the UK. Between the scenarios, there is a wide emission variation between GHGs and air pollutants, with the OEm comparable to estimates reported in European national inventories. Although hypothetically feasible, it is very unlikely that all the conditions assumed in the optimistic scenario will be systematically met, i.e., application of best regulations, strict emission standards, best engine technology/performance, and lowest gas leakage estimates across all stages of the production chain. GHGs released under REm P50 scenarios range from 46.4 to 175.3 Kt for CH_4 (equivalent to 10.3 and 65%, respectively, of the total emissions from the current heat and power sector in the reference country) and between 1.8 and 4.9 Mt for CO_2 (equivalent to 0.5 and 3.5% of current total country emissions). Figures generated by OEm are significantly lower, ranging from 1.6 to 11.6% (CH_4) and from 0.9 to 2.0% (CO_2) of total country emissions. The CH_4 contribution to total CO_2-eq. emissions generated by our shale gas industry is ca. 25% in OEm (min) compared to ≥50% in all other cases (see **Figure 7**), indicating the climate effectiveness of measures aiming at curbing CH_4 emissions. In the broader carbon footprint perspective, our results depict scenarios where the shale gas EI for electricity generation is systematically higher than estimates of the current EI for gas in Germany, ranging from +3 to +35% when considering all well productivity and performance/technology cases. Conversely, the same results are below or similar to the CF assigned to the national power grid, and one half and one third of the EI indicated for hard coal and lignite, respectively.

CH_4 leakage in the shale gas upstream sector as a percentage of total gas production in REm ranges from 0.38 to 0.90% under the P50 case, and between 0.35 and 1.36% overall. Losses are much more constrained in OEm and range between 0.08 and 0.13% in the P50 case, and between 0.08 and 0.15% when the P25 and the P75 cases are considered. The emissions generated in OEm are comparable to official governmental figures in national inventories. On the other hand, CH_4 leakage rates reported in several field studies carried out in the US and elsewhere show leakage rates similar or slightly higher than those evidenced in REm: between 0.9 and 1.9%. The reason for the discrepancies between official estimates in Germany and the UK, and emissions reported by studies from the US are not fully understood, and may be caused by under-reporting of current emissions or by substantial differences in gas production practices, CH_4 monitoring, and maintenance of facilities and equipment. Further investigation in this direction is necessary.

The release of air pollutants along the entire shale gas production chain represents a significant health threat. Limiting our analysis to the P50 production case, we estimate that the release of CO, NO_x and PM_{10} to the atmosphere may be negligible in all scenarios when compared to national emissions. On the other hand, VOC losses can be relevant in REm, and strategies to abate them are applicable and explored in OEm. Air pollutants, as opposed to GHGs, have direct health repercussions at the local and regional levels. Our scenarios only focused on total annual emissions, while investigations of their implications for local and regional air quality are addressed in a follow-up study through the application of atmospheric chemistry modeling. The large variability of emissions of all air and climate pollutants described by REm and OEm testifies to the value and potential of implementing existing best technologies/practices across the different gas production stages. Based on the latest engineering innovations in this field and recent scientific findings, emissions can be ameliorated by a rigid and accurate regulatory scheme, keeping in mind that results under OEm are based on technically feasible emission reduction measures and technologies that are rather unlikely to be systematically employed or achieved. Our results suggest that this is the only chance for gas to represent a transitory solution when carbon-free technologies cannot be deployed. The development of new natural gas systems that are regulated by outdated or inappropriate legislation is – as largely proved by the existing literature – a missed opportunity. The same is true for existing gas systems, where other studies have shown how technical CH_4 leakage reduction strategies are mostly easy to implemented and are likewise cost effective, especially when the gas price is high (ICF 2015, 2014). In light of this and conscious of the climate crisis that the planet is currently facing as emphasized in the latest IPCC report released in 2018, addressing gas environmental hazards needs to swiftly take center stage in government policies and in negotiations with gas operators. Our findings on the potential risks of a future European shale gas industry also call for full environmental compliance to minimize inevitable shortcomings in the event that shale gas becomes a reality in Europe.

Data Accessibility Statement

The authors have no data accessibility statement to declare.

Notes

[1] Available at: http://www.sciencemag.org/news/2018/06/natural-gas-could-warm-planet-much-coal-short-term. Accessed 15 April 2019.

[2] Available at: https://www.dieselnet.com/standards/eu/nonroad.php#s3. Accessed 15 April 2019.

[3] Based on natural gas conversion standards provided by the International Gas Union (IGU).

Supplemental files

The supporting information describes how the drilling projections are constructed, the technical aspects of the O&G industry that are discussed, and the reservoir productivity cases (SM Text S1, Section S1). Activity data, emission factors and their application to each stage of the shale gas supply chain under different technology and productivity scenarios is provided in SM Text S1, Section S2. The sensitivity analysis of the emission scenarios is presented in the remaining SM Text S1, Section S3.

The supplemental files for this article can be found as follows:

- **Text S1.** Supplemental text. DOI: https://doi.org/10.1525/elementa.359.s1

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Competing interests

The authors have no competing interests to declare.

Author contributions

- Substantially contributed to conception and design: LC, LBW, TB
- Contributed to acquisition of data: LC, LBW
- Contributed to analysis and interpretation of data: LC, LBW, TB
- Drafted and/or revised the article: LC, LBW, TB, HDVDG, MPB
- Approved the submitted version for publication: LC, LBW

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3 Paper II: Modeling the impact of a potential shale gas industry in Germany and the United Kingdom on ozone with WRF-Chem[6]

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RESEARCH ARTICLE

Modeling the impact of a potential shale gas industry in Germany and the United Kingdom on ozone with WRF-Chem

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Germany and the United Kingdom have domestic shale gas reserves which they may exploit in the future to complement their national energy strategies. However gas production releases volatile organic compounds (VOC) and nitrogen oxides (NO_x), which through photochemical reaction form ground-level ozone, an air pollutant that can trigger adverse health effects e.g. on the respiratory system. This study explores the range of impacts of a potential shale gas industry in these two countries on local and regional ambient ozone. To this end, comprehensive emission scenarios are used as the basis for input to an online-coupled regional chemistry transport model (WRF-Chem). Here we simulate shale gas scenarios over summer (June, July, August) 2011, exploring the effects of varying VOC emissions, gas speciation, and concentration of NO_x emissions over space and time, on ozone formation. An evaluation of the model setup is performed, which exhibited the model's ability to predict surface meteorological and chemical variables well compared with observations, and consistent with other studies. When different shale gas scenarios were employed, the results show a peak increase in maximum daily 8-hour average ozone from 3.7 to 28.3 μg m^{-3}. In addition, we find that shale gas emissions can force ozone exceedances at a considerable percentage of regulatory measurement stations locally (up to 21% in Germany and 35% in the United Kingdom) and in distant countries through long-range transport, and increase the cumulative health-related metric SOMO35 (maximum percent increase of ~28%) throughout the region. Findings indicate that VOC emissions are important for ozone enhancement, and to a lesser extent NO_x, meaning that VOC regulation for a future European shale gas industry will be of especial importance to mitigate unfavorable health outcomes. Overall our findings demonstrate that shale gas production in Europe can worsen ozone air quality on both the local and regional scales.

Keywords: Shale gas; WRF-Chem; European air quality; Ozone; Methane leakage; Emission scenarios

Introduction

Natural gas production in the United States (US) has risen markedly over the past years: in 2018, US dry gas production was 30 trillion cubic feet (861 billion cubic meters; bcm), an increase of 69% over 2005 levels (EIA, 2019a). The majority of the US gas production increase since 2005 was afforded through shale resources (EIA, 2016). In fact, shale gas made up approximately 69% of total US natural gas production in the year 2018 (EIA, 2018). Commercial extraction of shale gas reservoirs has been made possible through relatively recent advancements in horizontal drilling and hydraulic fracturing technologies. While shale gas production has hitherto largely been an American phenomenon (BP, 2016), global shale gas resources[1] are vast totaling more than 200 trillion cubic meters (tcm; about one third of world total gas technically recoverable resources), with about 13 tcm in Europe (EIA, 2013). Though there is currently no European commercial shale gas industry, several countries there have considered using domestic supplies to complement their national energy strategy (EC, 2014; JRC, 2012). Shale gas has the potential to reduce dependency on foreign imports and to offset the decline in indigenous production of conventional gas that European countries have been experiencing (BP, 2017). While Poland was initially a promising location for commercial shale gas extraction, exploration efforts there ultimately failed (LaBelle, 2018). On the other hand, Germany and the United Kingdom (UK) have among the top European shale gas-in-place[2] volumes (Germany: 7.7 tcm; UK: 37.6 tcm), and have expressed interest in recent years in exploiting their reserves (BGR, 2016; BGS, 2013). Additionally many reports and papers

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have been published on European shale gas and for the UK and Germany in particular, e.g., ACATECH (2016), AQEG (2018), BGR (2012), Broomfield et al. (2014); Cotton et al. (2014), DECC (2013), DECC (2014), Hays et al. (2015), McGlade et al. (2014), MULNV NRW (2012), Pfunt (2016), Sauter et al. (2013), SGD (2013), Society (2012) SRU (2013), Stamford and Azapagic (2014), UBA (2013, 2014).

Methane (CH_4) is the main constituent of natural gas, and is leaked throughout the various stages of production. CH_4 is a potent greenhouse gas with a heat-trapping ability 87 times that of CO_2 over a 20-year time frame, or 36 over a 100-year time frame (Myhre et al., 2013). Accordingly numerous studies have focused in recent years on CH_4 loss from natural gas production, reporting CH_4 leakage rates[3] ranging from less than 1% to greater than 10% of natural gas production, e.g., Allen et al. (2013), Alvarez et al. (2018), Caulton et al. (2014), Howarth (2014), Karion et al. (2013), Peischl et al. (2015, 2013), Pétron et al. (2012, 2014), Schneising et al. (2014). Findings from studies suggest that oil and gas sector CH_4 emissions are higher than official inventory estimates, with superemitters likely responsible for a large fraction of the CH_4 leakage (Brandt et al., 2014; Johnson et al., 2017; Miller et al., 2013; Zavala-Araiza et al., 2015). For example Alvarez et al. (2018) estimated a CH_4 leakage rate of 2.3% for about one third of US gas production and distribution systems, roughly 60% higher than the US EPA inventory estimate. To put these values into perspective, Alvarez et al. (2012) found that a CH_4 leakage rate of 3.2% or greater would negate climate benefits gained by switching from coal to natural gas power plants. In addition, toxic pollutants are likewise released, including volatile organic compounds (VOCs), nitrogen oxides (NO_x), particulate matter (PM), and carbon monoxide (CO), e.g., Roy et al. (2014), Schade and Roest (2016).

In the presence of sunlight, VOCs, CH_4, and CO interact with NO_x through a complex series of reactions to form tropospheric ozone (O_3) (Sillman, 1999; Sillman, 2003). The relationship between O_3 and its precursors is nonlinear: in a NO_x-sensitive regime (high VOC/NO_x ratio), O_3 increases with increasing NO_x, while increasing VOCs will have little to no impact. In a VOC-sensitive regime (low VOC/NO_x ratio), O_3 increases with increasing VOCs, while the addition of NO_x decreases O_3 formation. VOC-sensitive regimes are often encountered in urban areas where emissions of NO_x from combustion (e.g., in road traffic) are high, while NO_x-sensitive cases are often found in rural areas.

Ground-level O_3 is a significant short-lived climate forcer (Myhre et al., 2013), and dangerous to human and environmental health (Amann et al., 2008). O_3 adversely affects the respiratory, cardiovascular and central nervous systems, reproduction and development (EPA, 2013; Horvath and McKee, 1993; Nuvolone et al., 2018), damages ecosystems (EPA, 2013; Horvath and McKee, 1993; Nuvolone et al., 2018), reduces crop yields (Avnery et al., 2011), and impairs infrastructure (Kumar and Imam, 2013; Lee et al., 1996). The European Union's (EU) air quality directive asserts that the maximum daily 8-hour average O_3 (MDA8) should not surpass 120 $\mu g\ m^{-3}$ (60 ppb[4]) as a long-term objective; as a target value it should not be exceeded on more than 25 days per year averaged over 3 years (EP, 2002). However, the EU's target value for O_3 is higher than the World Health Organization's (WHO) air quality guideline of 100 $\mu g\ m^{-3}$, which they recommend to adequately protect public health (WHO, 2005). In spite of extensive regulation of precursor emissions, O_3 pollution is still a problem in Europe: according to the European Environmental Agency (EEA), over the 2000–2015 period between 94–99% of the European urban population (EU-28) was exposed to O_3 levels exceeding the WHO guideline (EEA, 2017). Exposure to O_3 pollution is believed to cause thousands of premature deaths of Europeans annually, with 13,600 deaths estimated for 2014 (EU-28) (EEA, 2017). Unhealthy levels of ambient O_3 in Europe can be ascribed to rising hemispheric background O_3 (Monks et al., 2015), the transboundary nature of O_3 and its precursors i.e. they can be transported across national boundaries and regions (Bach et al., 2014), reduced NO_x titration in some urban areas whereby O_3 is scavenged through reaction with NO (Amann et al., 2008), and the nonlinear relationship between O_3 and its precursors (EEA, 2014).

Atmospheric chemistry transport modeling is a valuable tool for assessing air quality on the local and regional levels and assisting in air quality management. A widely used model is the regional scale air quality WRF-Chem model (Grell et al., 2004). WRF-Chem has frequently been used in European-based air quality studies in recent years, e.g., Brunner et al. (2015), Fallmann et al. (2016), Forkel et al. (2012), Im et al. (2015), Kuik et al. (2016, 2018), Mar et al. (2016), Solazzo et al. (2012a, 2012b), Tuccella et al. (2012), Zhang et al. (2013a, 2013b), among others.

Intensive natural gas production has motivated several modeling studies to assess the role of precursor emissions from this source in regional O_3 formation. Rodriguez et al. (2009) found that increased growth in oil and gas activities in the US Intermountain West could increase MDA8 by a maximum of 9.6 ppb in southwestern Colorado and northwestern New Mexico in the summertime. Kemball-Cook et al. (2010) modeled the impact of shale gas development in the Haynesville Shale play for 2012 and found increases in MDA8 up to 5 ppb within Northeast Texas and Northwest Louisiana. Carter and Seinfeld (2012) looked at the relative contributions of NO_x, individual VOCs and HONO (nitrous acid) in O_3 formation during episodes in the wintertime in the gas producing Upper Green River Basin region in Wyoming; they found that the locations varied in their sensitivity to VOC and NO_x emissions: one site for the 2008 episode was highly NO_x sensitive, while the other 2008 site and both sites for 2011 were highly VOC sensitive. Ahmadov et al. (2015) simulated high O_3 episodes during the winter of 2013 over the intensive oil and gas producing Uinta Basin in the Western US, finding that a simulation based on top-down oil and gas sector emissions was able to capture observations during high O_3 episodes, while a simulation using bottom-up emissions from the US Environmental Protection Agency's (EPA) National Emission Inventory did not. Fann et al. (2018) modeled the impact of oil and gas activities over the US for 2025, predicting increases in summertime MDA8 up

to 8.12 ppb in Western Texas. Archibald et al. (2018) modeled over the UK the potential impacts of VOC and NO_x emissions related to future shale gas fracking activities there for 2013 and generally found increases in MDA8 across the country throughout the year with a maximum increase of 2.3 ppb in June. In addition to these studies, much of the literature on US shale gas has focused on O_3, e.g., AACOG Natural Resources Department (2013, 2015), Chang et al. (2016), Edwards et al. (2014), Field et al. (2015), Helmig et al. (2014), Koss et al. (2015), NC Division of Air Quality (2015), Olaguer (2012), Schade (2017), Schnell et al. (2009). These works underline the value and need for studies that analyze how O_3 may be affected by oil and gas development.

Shale gas is a contentious topic among the public and political spheres in both Germany and the UK, with critics citing various concerns, e.g., adverse air quality impacts (Althaus, 2014; Cremonese et al., 2015; Vetter, 2016; Yeo, 2019). The German government currently has a moratorium on unconventional fracking activities, e.g. for shale, which can be reevaluated in 2021 (German Federal Government, 2017). On the other hand, the British government recently granted consent for hydraulic fracturing testing to the energy company Cuadrilla in late 2018 (Perry, 2018). However, the UK's safety regulations strictly limit the magnitude of seismic activity allowed by fracking activities, currently set to 0.5 on the Richter scale, which severely constrains the amount of gas that can be extracted (Thomas and Pickard, 2019). Indeed, tremors exceeding this limit have forced Cuadrilla to suspend work on numerous occasions since testing started. In any case, what role shale gas production will have in Europe's future, if any, is uncertain. In order to inform decisions and the greater debate on shale gas, scientifically-based knowledge on potentially dangerous impacts is critical. To this end, air quality modeling can be used to run scenarios and study how varying levels of precursor emissions from a future shale gas sector may impact regional European O_3 pollution.

This paper is a companion paper to Cremonese et al. (2019), who developed scenarios on a future shale gas industry in these two countries to quantify the potential impacts on greenhouse gas and pollutant emissions. The present study builds on Cremonese et al. (2019) by using their scenario work as the basis for emissions input to the WRF-Chem model to investigate the range of potential impacts from shale gas on local and regional O_3, with a major focus on O_3 health-related metrics MDA8 and SOMO35 (annual Sum of Ozone Means Over 35 ppb, daily maximum 8-hour). In addition to the CH_4 leakage rate of Cremonese et al. (2019), we also investigate higher CH_4 leakage rates from other studies up to 6%. Due to our interest in the European region, we base our WRF-Chem setup on Mar et al. (2016), whose evaluated setup over Europe performed well for both meteorology and chemistry. Our objective is to improve understanding on potential air quality impacts from a future shale gas industry in Europe for public and political discourse, and for supporting policymakers and other decision-makers. Further our work can inform regulation of a potentially emerging industry to enact emission control strategies and prevent potentially harmful levels of air pollution. To our knowledge, there have been no published studies of regional air quality impacts from future shale gas activities in Europe, apart from Archibald et al. (2018), which focused on the UK. In this paper, we first describe the methodology, detailing the model setup, initial and boundary conditions and emissions input, the shale gas scenarios, and the preprocessing of shale gas emissions. Subsequently we discuss the results, and finish with a summary and conclusions.

Methodology

Model description and emissions

We used the Weather Research and Forecasting model (WRF) version 3.8.1 (Skamarock et al., 2008), coupled with chemistry (WRF-Chem) (Fast et al., 2006; Grell et al., 2005). Additionally we employ the same principal WRF-Chem options as done by Mar et al. (2016), who performed an extensive evaluation over Europe for the entire year of 2007 with WRF-Chem version 3.5.1. Here we describe our setup, and the main options we use are summarized in **Table 1**. The namelist is provided in SM Text S1, Table S1.

The model domain is set over Western Europe and fully covers the countries of study (i.e., Germany and the

Table 1: Options used in WRF-Chem model simulations. DOI: https://doi.org/10.1525/elementa.387.t1

Atmospheric process	Option used
Cloud microphysics	Lin et al. scheme (Lin et al., 1983)
Longwave radiation	RRTMG (Iacono et al., 2008)
Shortwave radiation	Goddard shortwave scheme (Chou and Suarez, 1994)
Surface layer	MM5 Monin-Obukhov scheme (Jiménez et al., 2012)
Land-surface physics	Noah land surface model (Chen and Dudhia, 2001)
Urban surface physics	Urban canopy model (Kusaka and Kimura, 2004)
Planetary boundary layer	Yonsei University scheme (Hong et al., 2006)
Cumulus parameterization	Grell 3D scheme (Grell and Dévényi, 2002)
Chemistry	MOZART-4 chemistry, KPP solver
Photolysis	Madronich F-TUV photolysis

UK; **Figure 1**). The horizontal grid resolution was set to 15 km × 15 km, and the model domain was established with 150 grid points in both the west–east and south–north directions. We used 35 vertically-stretched levels in the model with the top layer at 50 hPa. Our simulation period spans from May 29, 2011 to September 1, 2011, where the days in May serve as model spin-up. We decided on JJA (June, July, August) as the period for which to run our simulations because meteorological conditions during the summer months, i.e., elevated temperatures, increased sunlight and slow-moving high pressure systems, are more favorable to production of high and potentially harmful levels of tropospheric O_3, and therefore this season commonly experiences adverse impacts on and air quality in terms of this pollutant, e.g., EEA (2017), Volz-Thomas and Ridley (1994).

Meteorological initial and lateral boundary conditions (BCs) were obtained from the ERA-Interim reanalysis dataset provided by the European Centre for Medium-range Weather Forecast (ECMWF; Dee et al., 2011) with a spatial resolution of ~80 km every 6 hours. We used nudging by four dimensional data assimilation (FDDA) to reduce model errors in meteorology that may be associated with deviation of simulated large-scale circulation from the observed synoptic conditions. We nudged temperature at all vertical levels, while horizontal winds are nudged above the planetary boundary layer (PBL). Mar et al. (2016) reported erroneous precipitation suppression over Europe in sensitivity studies compared with observations when water vapor nudging was applied; therefore we likewise did not nudge water vapor in our simulations, also following the approach of, e.g., Miguez-Macho et al. (2004) and Stegehuis et al. (2015). Space- and time-varying (i.e., dynamic) chemical initial and lateral BCs were implemented in this study and were provided by simulations from MOZART-4/GEOS-5 through NCAR at http://www.acom.ucar.edu/wrf-chem/mozart.shtml. Note that dynamic BC data can improve simulated O_3 (Gavidia-Calderón et al., 2018), and MOZART-4 global model data is frequently used as the BCs for simulations (Pfister et al., 2011).

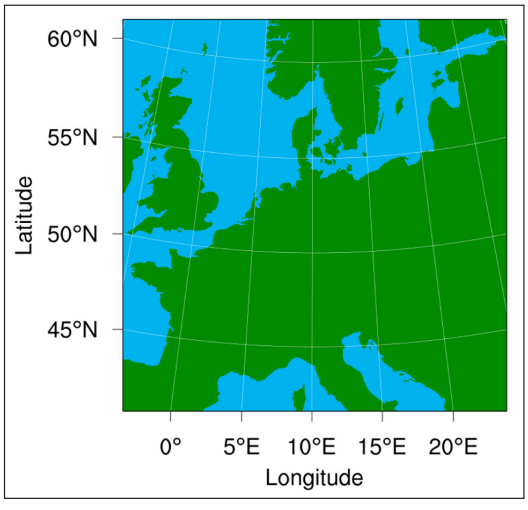

Figure 1: Domain of study. Model domain applied in WRF-Chem simulations, set over Western Europe and fully covering the countries under examination, Germany and the UK. DOI: https://doi.org/10.1525/elementa.387.f1

Based on the findings of Mar et al. (2016), we apply the MOZART-4 chemical mechanism in our simulations (Emmons et al., 2010). Of the natural gas-relevant VOC species that are considered in this work, ethane and propane are represented explicitly by the MOZART-4 mechanism, while the higher alkanes (i.e., butane, pentane, hexane and heptane) are lumped into the C > 3 group known as BIGALK. Note that VOC shale gas emissions in this study are very light and do not contain aromatics (**Table 4**), and NO_x emissions are likewise low compared with national inventories (**Figure 3**). This means that their effect on secondary aerosol formation would likely be insignificant, and as such we do not look at this in our study. Anthropogenic emissions of CO, NO_x, SO_2, VOCs, and NH_3 used in WRF-Chem were obtained from the TNO MACC III inventory provided at a resolution of 7 km × 7 km, for the year 2011 (Kuenen et al., 2014). In addition to the aforementioned anthropogenic emissions, shale gas emissions based on Cremonese et al. (2019) were included in our simulations (described in Scenario background).

Shale gas emission scenarios

Scenario background

The present study carries on the work of Cremonese et al. (2019), who developed a series of drilling and emission scenarios to study the effects of a potential shale gas industry in Europe on greenhouse gas and pollutant release. Specifically, they explore inter alia how different practices, uncertainty in data – i.e., activity data and emission factors, as well as the extent of well productivity, may impact emissions released annually from such an industry in Europe. The scope of Cremonese et al. (2019)'s scenarios covers upstream emissions, i.e., from well pad preparation up to gas processing, from shale gas development in Germany and the UK. Cremonese et al. (2019) assume in their scenarios that annual shale gas production will be equivalent to recent conventional production in these two countries (Germany: 11.6 bcm; UK: 36.6 bcm). All scenarios explored in the present study are based on the 'REm-U P25' scenario in Cremonese et al. (2019). REm stands for – 'Realistic Emissions' case, meaning that business as usual practices for US shale gas production are applied; -U stands for 'Upper' meaning that activity data and emission factors are on the high end of the uncertainty range. P25 means that well productivity is at the 25th percentile, i.e., the lower end of the spectrum of well productivity as determined by the authors. CH_4 leakage rates from REm-H P25 are up to 1.36%, which are well within the range predicted for average upstream natural gas leakage in the US and worldwide (Cremonese et al,. 2019). While European official CH_4 leakage estimates for natural gas systems are, comparatively, substantially lower, independent studies investigating European natural gas CH_4 leakage rates in a transparent way are unavailable (Cremonese et al. (2019) and references therein). Indeed recent studies suggest

that CH_4 emissions from European oil and gas systems are higher than indicated by inventories (Riddick et al., 2019; Yacovitch et al., 2018), while numerous studies carried out in the US have reported emissions from natural gas systems which are higher than national estimates (Introduction). Hence it is reasonable to explore higher leakages as unanticipated, yet plausible rates for large-scale gas production in Europe.

Present work

In total, we examine three scenario sets covering low, medium and high shale gas emissions which we refer to respectively as SG1, SG2, and SG3. Our interest is to see how varying emissions of VOCs with constant NO_x from shale gas activities may affect air quality in this region. Note that the shale gas emissions from Cremonese et al. (2019) included in our scenarios are NO_x, CO and VOCs. NO_x and CO are based on combustion processes, while VOC emissions are based on CH_4 leakage as the two species are co-emitted together in gas leaks. Due to its relevance to the literature on natural gas, we explore scenarios based on varying CH_4 leakage rates. Therefore NO_x and CO shale gas emissions remain constant in SG1-3, while VOC emissions (based on extent of CH_4 leakage) are the only ones that vary between the three scenario sets. It is necessary to note that changes in the CH_4 emissions are not used in this study due to model limitations as will be explained in Pre-processing shale gas emissions for WRF-Chem. In SG1 we use the CH_4 leakage rates directly from Cremonese et al. (2019), which are tailored specifically to Europe (**Table 2**). For SG2 medium level emissions, we chose a rate of 2% based on Alvarez et al. (2018) (the approximate value for the segment of the gas chain considered in our scenarios), as it is relatively high yet still representative of large-scale gas production in the US (Introduction). For SG3 high level emissions, we chose a conservative extreme value of 6% which is on the higher end of the literature for CH_4 leakage (typically ranging from <1.0 to 10%, e.g., Visschedijk et al., 2018). As reported in Saunois et al. (2016), the GAINS model adopted a CH_4 emission factor of 4.5% for shale gas mining with current technology, which also supports our choice of 6% as a conservative extreme rate.

Within each scenario set we explore three "sub" scenarios resulting in a total of nine simulated emission scenarios (**Table 2**), in addition to a baseline scenario for comparison. These three scenarios are wet gas composition, dry gas composition, and concentrated NO_x emissions, which we abbreviate to 'wet gas', 'dry gas', and 'conNO_x'. The first two are explored due to the critical impact that gas composition has on total VOC volumes released, and hence air quality. For example, unprocessed gas typically ranges from 75-90 vol% CH_4, with the rest mainly consisting of VOCs (Baker and Lokhandwala, 2008; Faramawy et al., 2016; Gilman et al., 2013). Wet gas is leaner in CH_4 and richer in VOCs, while the opposite case is true for dry gas. Because it is not known in advance what the gas composition will be, we explore scenarios that cover both wet and dry options. Furthermore, because NO_x emissions are unaffected by gas composition, wet and dry gas scenarios allow us to study the impact of varying levels of VOC loading to NO_x on O_3 production. In our scenarios we applied wet and dry gas compositions of 84.6/15.4 and 96/4 (vol% CH_4/VOCs), respectively, based on Faramawy et al. (2016) and displayed in **Table 3**. Finally, the interest of the latter scenario was to see if NO_x emissions from shale gas activities concentrated in space and time could lead to a significant impact on local O_3 production. Because the conNO_x scenario required further development, it is described in detail in the following section below.

conNO_x scenario design

The purpose of the conNO_x scenario is to explore the sensitivity of the simulation to concentrated NO_x emissions on O_3 production. It needs to be pointed out that absolute NO_x scenario emissions from Cremonese et al. (2019) are not *increased*, but rather *concentrated over space and time*. However, in order to avoid limiting the analysis to an arbitrary segment of the simulation, we run the concentrated NO_x emissions for the entire JJA simulation period. Because the NO_x emissions would be too high over the whole period to look at cumulative metric results, the analysis for the conNO_x scenario is restricted to peak daily values. In this way the model run shows results of concentrated NO_x emissions for any time during the study period, with the advantage being that days with more favorable

Table 3: Wet and dry gas compositions, in vol% (wt% in parentheses)[a]. DOI: https://doi.org/10.1525/elementa.387.t3

Species	Wet gas	Dry gas
CH_4	84.6 (67.1)	96.0 (88.1)
VOCs	15.4 (32.9)	4.0 (11.9)

[a] Data from Faramawy et al. (2016).

Table 2: Shale gas scenarios simulated in this study. DOI: https://doi.org/10.1525/elementa.387.t2

Scenario set	SG1	SG2	SG3
CH_4 leakage[a]	0.70% (UK) 1.36% (DE)	2%	6%
Reference	Cremonese et al. (2019)	Alvarez et al. (2018)	(see text)[b]
Sub-scenarios	dry gas, wet gas, conNO_x	dry gas, wet gas, conNO_x	dry gas, wet gas, conNO_x

[a] The UK and DE have different leakage rates in SG1 due to the design of the drilling projections in (Cremonese et al., 2019). In SG2 and SG3, a flat leakage rate is applied to gas production of both countries.

[b] Based on several different studies in the past years reporting very high CH_4 leakage rates from shale gas activities, as described in the text.

photochemical conditions for O_3 production will automatically be included in the results.

For conNO_x scenario development we first determined which sectors of shale gas exploitation from Cremonese et al. (2019) contribute to NO_x emissions in a manner that a high volume of emissions may be concentrated over space and time. Drilling and fracking activities fit these requirements: both activities require machinery with high horsepower and consequently result in high NO_x emissions, while operation time thereof is relatively short on the order of several days up to a few weeks. Furthermore it is plausible that a contractor would elect to drill/frack multiple wells within the same area, e.g., due to target sweet spots, regulation permits or restrictions, or favorable conditions of a location such as close proximity to roads, pre-existence of pipeline systems etc., so that total NO_x emissions from drilling and fracking activities may be concentrated in space.

According to Cremonese et al. (2019)'s shale gas emission scenarios, drilling and fracking combined constitute approximately 25% of total NO_x emissions for Germany and 20% for the UK. Therefore we concentrate this percentage of NO_x emissions for each respective country, while the rest of the NO_x emissions were treated as in the other scenarios (described in Pre-processing shale gas emissions for WRF-Chem). In our conNO_x scenario we assumed that one well is drilled and one well is fracked per well pad. Based on our calculations, NO_x emissions from drilling and fracking activities are concentrated over an area of 980 km^2 and 1225 km^2, or 8.0% and 7.2% of the total shale gas basin area, for Germany and the UK, respectively. With this information and the time over which the emissions occur, we determined the concentrated NO_x emissions flux. In order to place an upper bound on the effect of concentrated emissions of NO_x, we chose a location for these emissions based on which area exhibited sensitivity to added NO_x emissions for O_3 production. We achieved this through a sensitivity study in which we looked for coordinates within the shale gas basin region which displayed an increase in MDA8 when a simulation included both shale gas VOC and NO_x emissions over a simulation which included shale gas VOCs only (conNO_x regions displayed in **Figure 2**). Nevertheless it is worth noting that the increase displayed by added NO_x emissions was low (<4 μg m^{-3}). Additionally, it is important to note that the conNO_x scenarios are based on wet gas composition like the wet gas scenario (**Table 3**). Further information on the development of and calculations for the conNO_x scenario is provided in SM Text S1, Text S1.

Scenario emissions in context

In order to put our scenario emissions into context, we compare them with TNO MACC III inventory values for year 2011 (**Figure 3**). This offers a useful comparison since the TNO MACC III values are likewise used as anthropo-

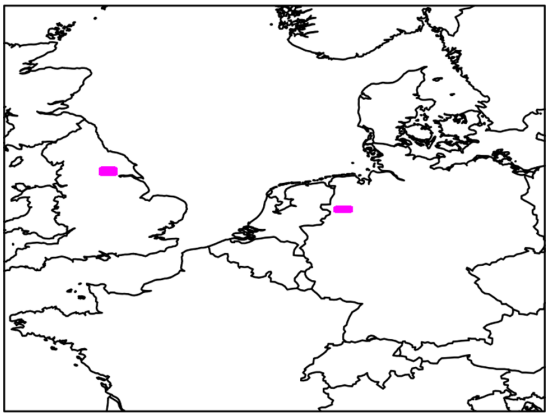

Figure 2: Concentrated NO_x (conNO_x) locations. Masks (in magenta) indicate the locations where NO_x emissions are concentrated in the UK and Germany under the conNO_x scenario. DOI: https://doi.org/10.1525/elementa.387.f2

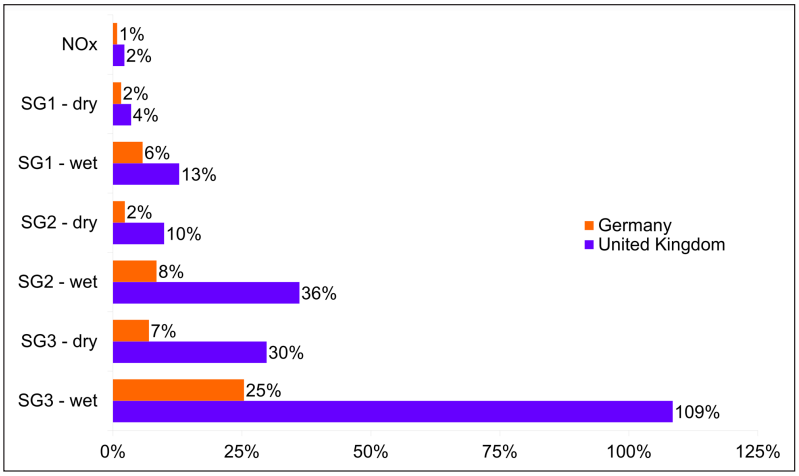

Figure 3: Shale gas scenario emissions compared with country total annual emissions. Scenario emissions are compared as a % equivalent to annual national total emissions from the TNO MACC III inventory, for year 2011. NO_x emissions are presented per country. VOC emissions are presented per scenario set per country, for both 'wet' and 'dry' VOC speciation. DOI: https://doi.org/10.1525/elementa.387.f3

genic emissions input in WRF-Chem (see Model description and emissions). Scenario emissions are equivalent to a higher proportion of country total emissions for the UK than Germany. This is primarily due to British gas production being greater than that of Germany in the scenario storyline. VOC emissions between the scenarios increase greatly from SG1 up to SG3. The importance of gas composition on total VOC emission volumes is evident, where wet gas emissions are about 3–4 times greater than dry gas emissions (more information on wet and dry gas is provided in Present work). With the leakage rate assumed in SG3 under the wet gas scenario, VOC emissions are equivalent to 109% of British country total emissions, and 25% for Germany. In contrast, VOC emissions under SG1 dry gas are equivalent to 4 and 2% of British and German country total emissions.

Pre-processing shale gas emissions for WRF-Chem

The shale gas activities examined in Cremonese et al. (2019) primarily occur within the shale gas basin regions of each respective country. Shale gas emissions from Cremonese et al. (2019) were adapted to the TNO MACC III grid for emissions pre-processing for WRF-Chem (**Figure 4**). The shale gas basin maps for Germany and the UK were obtained from reports by national institutions of each respective country, i.e., the Institute for Geosciences and Natural Resources (*Bundesanstalt für Geowissenschaften und Rohstoffe*) for Germany and the British Geological Survey for the UK (BGR, 2016; BGS, 2013). Because there is currently no commercial shale gas production in these countries, it is not known at which locations shale gas activities will occur and consequently where corresponding emissions will be released; therefore we averaged total scenario emissions over the shale gas basin area for each respective country. The impact of averaging emissions is not expected to be significant since VOCs are relatively long-lived (Atkinson, 2000). Furthermore no considerable diurnal variability in shale gas activities is expected, and accordingly total emissions for each respective country are assumed to occur at a constant rate. The only exception to this is that we concentrated some NO_x emissions over a subset of the shale gas basin region and JJA period, as described in conNO_x scenario design. Averaging the scenario emissions over space and time provided the emission flux for each shale gas species used in our simulations (VOCs, NO_x, and CO from Cremonese et al. (2019). While CH_4 is an important component of shale gas emissions and valuable to the scenario storylines explored here, shale gas CH_4 is not included in our simulations: WRF-Chem treats CH_4 as a BC on account of CH_4's relatively long atmospheric lifetime of circa 12 years (Myhre et al., 2013).

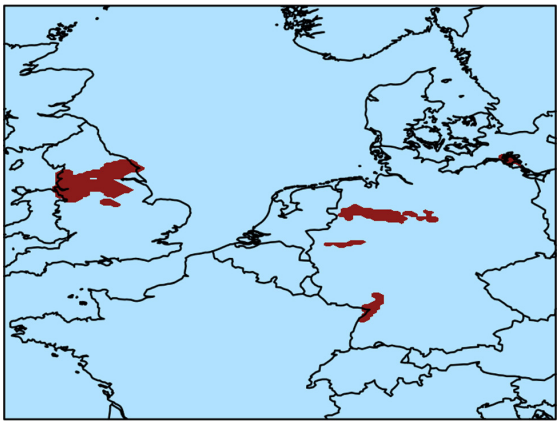

Figure 4: Shale gas basin areas. The basins (maroon) for the UK and Germany represent the areas over which scenario emissions were averaged by country. Corresponding emission fluxes were added to the basin masks and run in scenario simulations with WRF-Chem. Basin areas are based on reports by national institutions, i.e., BGS (UK) and BGR (Germany) (BGR, 2016; BGS, 2013). DOI: https://doi.org/10.1525/elementa.387.f4

In the scenarios explored here, >99.9% of the VOC emissions result directly from natural gas loss. For this reason we applied a typical natural gas VOC composition as the speciation for all VOC emissions from the shale gas scenarios that we pre-processed into WRF-Chem. The VOC speciation for natural gas was obtained from Faramawy et al. (2016) (**Table 4**). Since the MOZART-4 chemical mechanism used in our setup lumps C > 3 alkanes into one group (BIGALK), the actual speciation we applied includes 29% ethane, 35% propane and 36% BIGALK, by weight. We applied the wet gas VOC speciation to all scenarios to avoid confounding the results between wet and dry gas scenarios, i.e., so that wet and dry gas scenarios differ only in their extent of VOC emissions and not in their speciation. It also needs to be pointed out that natural gas is indeed a lightweight mixture of alkanes; nevertheless, in reality gas is often found in the reservoir in association with some oil which contains higher alkanes, alkenes, aromatics, etc. Ahmadov et al. (2015) found that aromatic VOCs have a disproportionately greater contribution to O_3 formation relative to all other VOC emissions. Additionally Carter and Seinfeld (2012) reported that both aromatics and alkenes were the most significant contributors to O_3 formation in spite of their comparatively small

Table 4: Natural gas VOC speciation applied to emissions during pre-processing for WRF-Chem, in % wt[a]. DOI: https://doi.org/10.1525/elementa.387.t4

Species	Faramawy et al. (2016)	MOZART-4
Ethane	29%	29%
Propane	35%	35%
Isobutane	10%	36% (BIGALK)
n-butane	12%	–
Isopentane	4%	–
Pentane	2%	–
Hexanes	5%	–
Heptanes	2%	–

[a] Values displayed adapted from the Faramawy et al. (2016) composition for wet gas for use with MOZART-4.

VOC contribution. Therefore if oil is present, the impact of VOCs on O_3 production may be even greater.

Results and discussion

Evaluation

Due to the similarity of our setups, we use the same approach and offer a comparison of our evaluation results to those of Mar et al. (2016). Notable differences between our setup and Mar et al. (2016) include the following (our setup/Mar et al., 2016): WRF-Chem versions (3.8.1/3.5.1), simulation years (2011/2007), anthropogenic emissions inventory (TNO MACC version III/TNO MACC version II), horizontal resolutions (15 km × 15 km/45 × 45 km), and European domain coverage (Western Europe/whole of Europe). It is worth emphasizing that the principle goal of this study is to quantify the impacts of shale gas production on air quality by means of a reasonably working setup; because an in-depth evaluation is provided by Mar et al. (2016), we focus on main findings here.

The meteorological observations used for evaluation are taken from the Global Weather Observation dataset provided by the British Atmospheric Data Center (BADC) (Met Office, 2006). The chemical observations are taken from AirBase, the European air quality database of the European Environmental Agency (EEA, 2013). Like Mar et al. (2016) we compare our data to rural background stations since our horizontal grid resolution is likewise relatively course and therefore more representative of rural conditions. We evaluate our model-simulated results against observations for the following statistics: mean model, mean observations, mean bias (MB), normalized mean bias (NMB), mean fractional bias (MFB), and the temporal correlation coefficient (*r*). Definitions of the statistical calculations are provided in Mar et al. (2016).

Meteorology

The meteorological evaluation was carried out for the following variables at a 3-hourly temporal resolution: mean sea-level pressure (MSLP), 2m temperature (T2), and 10m wind speed and direction (WS10 and WD10, respectively). A summary of the domain-wide statistical performance for meteorology of the base run setup with WRF-Chem against the observations at rural background stations is shown below in **Table 5**.

MSLP was reproduced over the domain with a high degree of accuracy. The bias for MSLP is negligible, where both NMB and MFB are 0, and the *r* value is 0.99. Likewise T2 was found to be reproduced with a high degree of accuracy by WRF-Chem, with a low MB of –0.08°C averaged over the entire domain. The seasonal average T2 spatial distribution statistics are displayed in **Figure 5**. In general the absolute values of MB in T2 (**Figure 5C**) were <1°C, where larger biases are found in the Alps. This greater bias over mountainous regions was also found in Zhang et al. (2013a) and Mar et al. (2016), which the latter notes is likely due to the complex mountain terrain and related unresolved local dynamics. Moreover the *r* values for T2 (**Figure 5D**) are generally >0.9 and do not display considerable geographical variation, which demonstrates the models ability to reproduce this parameter well. Furthermore it was found that the model represents wind speed well, with a domain-wide average MB of +0.07 $m \cdot s^{-1}$ and *r* value of 0.67. Averaged wind direction over the domain was found to originate from the south-west, with a MB of about 20 degrees (a wind rose diagram for model results over JJA has been provided in SM Text S1, Figure S1). In total, it was found that our WRF-Chem set-up is capable of reproducing meteorological conditions and their spatial and temporal variations in Europe reasonably well, and notably our values are consistent with those reported in, e.g., Zhang et al. (2013a) and Mar et al. (2016).

Chemistry

We performed an evaluation of the model chemistry with hourly observations of the following species: O_3, NO_x, NO_2 and NO. The domain-wide statistical performance for chemistry of the base run setup with WRF-Chem against the observations at rural background stations is shown below in **Table 6**.

In **Figure 6** we see that the lowest modeled surface O_3 concentrations in our domain are concentrated over the UK, Belgium, the Netherlands and the North Rhine-Westphalia region of Germany, with averaged values around 50–60 μg m^{-3}, while highest over the Mediterranean region, where values exceed 110 μg m^{-3}. O_3 is overpredicted throughout most of Italy; similarly, high O_3 was predicted over the Mediterranean with WRF-Chem in Mar et al. (2016) and with various models in Im et al. (2015). This finding may be due to the relatively coarse model resolution causing an underestimation of NO_x and in turn an overestimation of O_3, or causing an excessive diffusion of O_3 from the sea to the land. Predicted O_3 concentrations reproduce the north-south gradient shown in the observations. In **Table 6**, we see that our WRF-Chem setup overpredicts O_3 with a MB

Table 5: Domain-wide statistics of WRF-Chem base run setup against BADC 3-hourly meteorological observations, over JJA. DOI: https://doi.org/10.1525/elementa.387.t5

Meteorological variables	Mean-Obs[a]	Mean-Mod[a]	MB[a]	NMB[b]	MFB[b]	*r*[b]	No. stations
MSLP (hPa)	1013.43	1013.52	0.09	0.00	0.00	0.99	1332
T2 (°C)	16.93	16.85	–0.08	0.00	0.01	0.91	1629
WS10 ($m \cdot s^{-1}$)	3.45	3.52	0.07	0.02	0.14	0.67	1631
WD10 (°)	210.00	229.89	19.89	0.11	0.21	0.5	1619

[a] Means and MB are in units indicated next to meteorological variable.
[b] NMB, MFB and *r* are unitless.

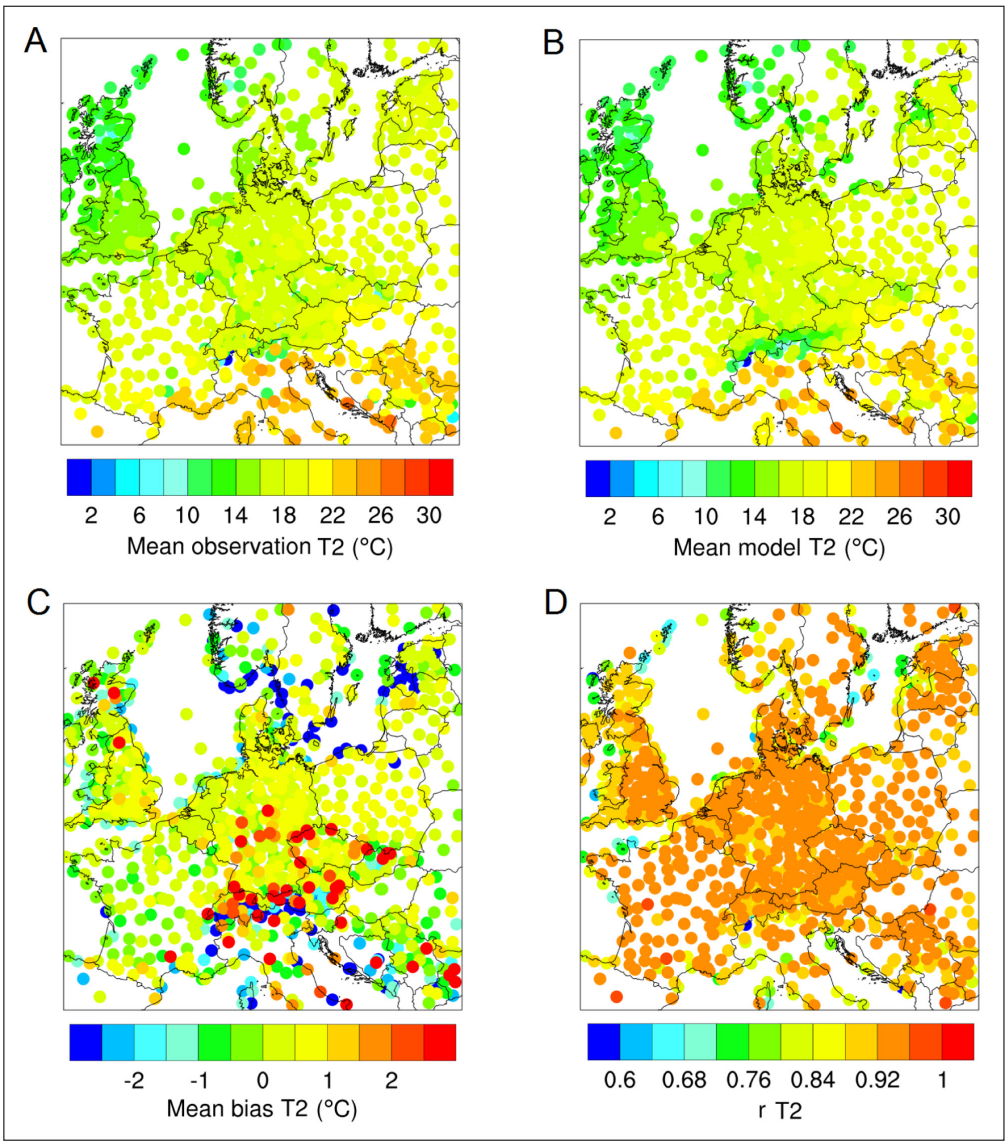

Figure 5: Seasonal average (JJA) 2m temperature (T2) spatial distribution statistics. Model values and statistics: **(A)** mean observation, **(B)** mean model, **(C)** mean bias, and **(D)** temporal correlation coefficient, are shown at the location of the observations. Results for means and MB are in °C, while *r* is unitless. DOI: https://doi.org/10.1525/elementa.387.f5

Table 6: Domain-wide statistics of WRF-Chem base run setup against AirBase hourly chemical observations, over JJA. DOI: https://doi.org/10.1525/elementa.387.t6

Species	Mean-Obs[a]	Mean-Mod[a]	MB[a]	NMB[b]	MFB[b]	$r^{b,c}$	No. stations
O_3	70.21	80.11	9.90	0.14	0.19	0.52	429
MDA8	91.64	96.97	5.33	0.06	0.07	0.63	429
NO_x	8.65	8.38	–0.27	–0.03	–0.20	0.22	283
NO_2	6.59	7.6	1.01	0.15	–0.12	0.27	298
NO	1.47	0.54	–0.93	–0.63	–1.07	0.23	216

[a] Means and MB are in units of μg m^{-3}.
[b] NMB, MFB and *r* are unitless.
[c] *r* represents the hourly temporal correlation coefficient for species O_3, NO_x, NO_2, and NO, while for MDA8 it represents the daily temporal correlation coefficient.

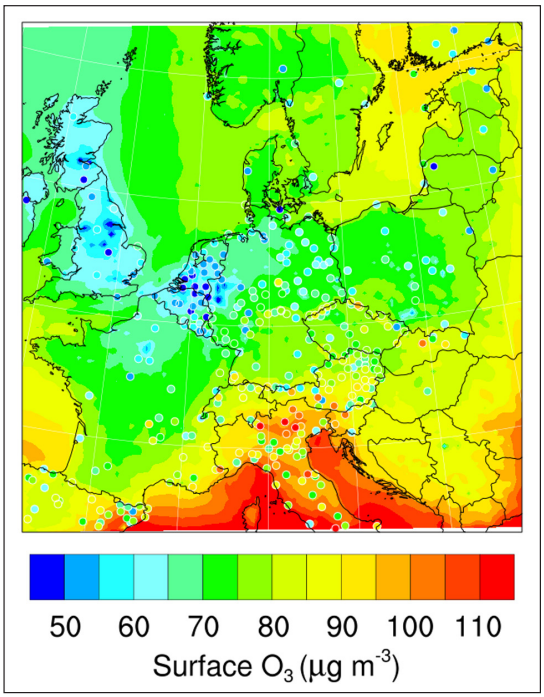

Figure 6: Seasonal surface-averaged O_3. Contours are modeled values and dots represent observational values measured at station locations, over JJA, in units of µg m^{-3}. DOI: https://doi.org/10.1525/elementa.387.f6

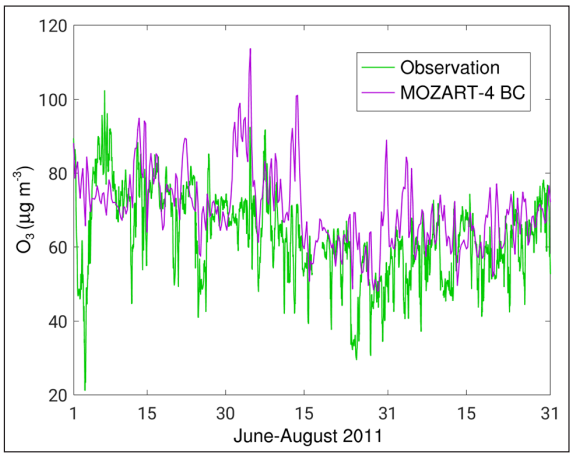

Figure 7: Time series of O_3 for boundary condition assessment at Mace Head Station. Hourly BADC observational data (green) are compared with 6-hourly MOZART-4 boundary condition modeled data (purple), over JJA, in units of µg m^{-3}. DOI: https://doi.org/10.1525/elementa.387.f7

of 9.90 µg m^{-3} and NMB of 0.14, similar to those reported in Mar et al. (2016) for the JJA season (MB: 9.92 µg m^{-3}; NMB: 0.14). Moreover, our results are consistent with other regional modeling studies for Europe, e.g., the absolute NMB values for O_3 in ensemble modeling studies of the Air Quality Model Evaluation International Initiative (AQMEII), e.g., Solazzo et al. (2012b) for the summertime and Im et al. (2015) for year 2010. Additionally the NMB value is within the model performance criteria by Russell and Dennis (2000) for O_3, for which normalized mean bias is suggested to be within a range of ±5 to 15%. Our temporal correlation value for O_3 is 0.52, which is consistent with Mar et al. (2016)'s value of 0.55 and Tuccella et al. (2012), who reported an hourly correlation value of 0.62 averaged over the year 2007. Also in **Table 6** we see that our setup slightly overpredicts MDA8, though our MB for MDA8 is lower than that for O_3. Since MDA8 is essentially a measure of daytime O_3, this indicates that our setup is performing very well predicting O_3 values during the day, but overpredicts O_3 nighttime values to a greater extent, possibly due to NO_x titration during the nighttime not being as well resolved by the model.

Due to the model overestimation of O_3, we provide a brief assessment of the O_3 BC (boundary condition) used in this study provided by the MOZART-4 global model (described in Model description and emissions) at Mace Head station. Mace Head is located on the west coast of Ireland making it less likely to be influenced by European emissions, and therefore representative of the background O_3 flowing into Europe. In **Figure 7** is depicted a time series plot comparing hourly observational (BADC) data with 6-hour O_3 BC data, over JJA. The O_3 BC slightly overpredicts background O_3, and occasionally overestimates O_3 peaks. The O_3 BC does not capture the drops in the observed O_3 which can be caused by the coarse resolution of the model (1.9 × 2.5 degrees). This finding is consistent with previous work of Pfister et al. (2011): in their study looking at summertime pollution inflow into California, they found that the O_3 BC from the MOZART-4 global model overpredicts measured background O_3. On the other hand, the large drops in observed O_3 not seen in the O_3 BC could be due in part to the influence of local NO_x sources, since the observational data at Mace Head station have not been filtered for background air. The O_3 BC likely contributes to the positive bias in O_3 found in statistical evaluation results for O_3 and MDA8 (**Table 6**) through transport of background O_3 via the westerly winds (HTAP, 2010). Overall, however, the O_3 BC compares well with the observational time series trend.

In **Figure 8** high NO_x concentrations (~30 µg m^{-3} and greater) are visible over parts of the UK, the Netherlands, Belgium, Germany, Poland, and large urban conglomerates like Paris, Barcelona, and Belgrade, as well as the coastal region where the English channel meets the North Sea, resulting from high emission activities in these areas. In **Table 6** we see that our setup slightly underestimates domain-average NO_x concentrations (MB: –0.27 µg m^{-3}) over JJA due to the balancing of underestimated NO (MB: –0.93 µg m^{-3}) and overestimated NO_2 (MB: 1.01 µg m^{-3}). Our results for JJA are consistent with NO_2 (over-) and NO/NO_x (underprediction) trends for the whole year reported in Mar et al. (2016). These biases are likely a combination of uncertainty in NO_x emissions (Kuenen et al., 2014) and in the model representation. Reported causes of the latter include deficiencies in mixing in the planetary boundary layer (Kuik et al., 2018), overestimating

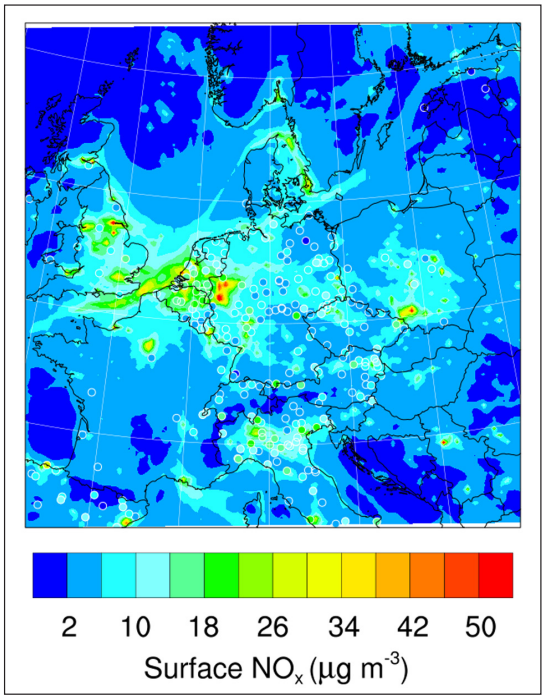

Figure 8: Seasonal surface-averaged NO_x. Contours are modeled values and dots represent observational values measured at station locations, over JJA, in units of µg m^{-3}. DOI: https://doi.org/10.1525/elementa.387.f8

nighttime NO_2 (Im et al., 2015), coarse grid resolution (Kuik et al., 2018), and that the atmospheric lifetime may be too long relative to deposition or chemical loss mechanisms (Stern et al., 2008). In the AQMEII project Im et al. (2015) found NO_2 to be overestimated by one model by 15%, while underestimated by the rest of the models by 9 to 45% for the European domain, where the WRF-Chem simulations used RADM2, CBMZ (Zaveri and Peters, 1999), RACM (Stockwell et al., 1997) mechanisms. In their model inter-comparison study on Central Europe, Stern et al. (2008) found both over- and underpredictions for NO_2 from 15 January 2003 to 5 April 2003. (Note that the aforementioned studies do not perform a validation for NO or NO_x). The domain average JJA temporal correlation coefficients against hourly NO_x, NO_2 and NO measurements are 0.22, 0.27 and 0.23, respectively. These *r* values are lower than that of O_3, but consistent with Mar et al. (2016) who reported values of 0.16, 0.22 and 0.19, respectively. Low *r* values as seen here indicate that the model is not suitable for predicting exceedances of NO_2. The generally low *r* values for NO_x are likely the result of model resolution, strong temporal variation of NO_x emission sources, and unreliable model inputs such as emissions (Karlický et al., 2017).

Overall, we find that the model performance for chemistry is consistent with Mar et al. (2016) on which we based our setup, and furthermore in line with biases of other studies for Europe, e.g., Solazzo et al. (2012b), Tuccella et al. (2012) and Im et al. (2015). Based on these aspects we find that our setup is performing at a reasonable level for modeling the impact of shale gas industry emissions on European air quality.

Quantification of O_3 impacts from shale gas activities

The maximum difference in daily MDA8 between scenario and base case, over the entire simulation period for each grid cell (referred to as ΔMDA8), is depicted in **Figure 9**; statistical data is provided in SM Text S1, Table S2. Note that in the following discussion we generally leave out SG2 to be concise as its impacts fall between the SG1 and SG3 cases.

In the SG1 wet gas scenario, predicted ΔMDA8 generally ranges between 2–4 µg m^{-3} and is restricted to relatively small areas located within the vicinity of the shale gas basin regions. Concentrating NO_x emissions leads to prominent differences compared with averaging NO_x emissions under SG1: ΔMDA8 values >2 µg m^{-3} stretch over a larger area for $conNO_x$, especially over Germany and within the vicinity where NO_x emissions are concentrated. The peak values likewise show a stark difference, where peak ΔMDA8 for wet gas is 4.5 µg m^{-3} and for $conNO_x$ is 9.5 µg m^{-3}. This indicates model O_3 sensitivity to NO_x, especially in Germany. In the dry gas scenario, ΔMDA8 values are lower compared with wet gas and are more strictly located over the shale gas regions, showing the importance of VOCs in model O_3 formation.

Under SG3 with wet gas speciation the largest peak ΔMDA8 occurs, reaching 28.3 µg m^{-3} and located over the North Sea off the British coast. This result is consistent with Archibald et al. 2018, where the largest maximum increase in MDA8 occurs under the scenario with the greatest VOC emissions. Also notable is that peak ΔMDA8 under SG3 is seen in the wet gas scenario; this is in contrast to SG1, where peak ΔMDA8 occurs in the $conNO_x$ scenario. Nevertheless, $conNO_x$ and wet gas are relatively similar under SG3, which also stands in contrast to SG1. ΔMDA8 values >2 µg m^{-3} extend over a considerably greater area of the domain for the wet gas and $conNO_x$ scenarios under SG3 compared with their SG1 counterparts, reaching Scandinavia, Eastern Europe, and the Mediterranean countries. ΔMDA8 values are especially prominent in marine areas including the Baltic Sea, the Mediterranean Sea, and the Atlantic Ocean west of the UK (in addition to the North Sea). Further under SG3 wet gas and $conNO_x$ scenarios, most of the UK experiences ΔMDA8 values >6 µg m^{-3} near the coast, with the inner portion of the country seeing enhancements from 14 up to about 22 µg m^{-3}. In Germany, the majority of the country experiences ΔMDA8 values >2 µg m^{-3}, with Northern Germany experiencing the highest values from 8 to 12 µg m^{-3} (SG3), in part due to the majority of the shale gas basin area being located in the upper half of the country. The much greater impact on MDA8 in the UK compared with Germany is likely due to VOC shale gas emissions being much higher in the UK (**Figure 3**); this is a result of the British shale gas industry being about three times larger than Germany in terms of gas production according to the scenarios. Additionally, under SG3 many of the neighboring countries experience high O_3 enhancement maxima likely due to long-range

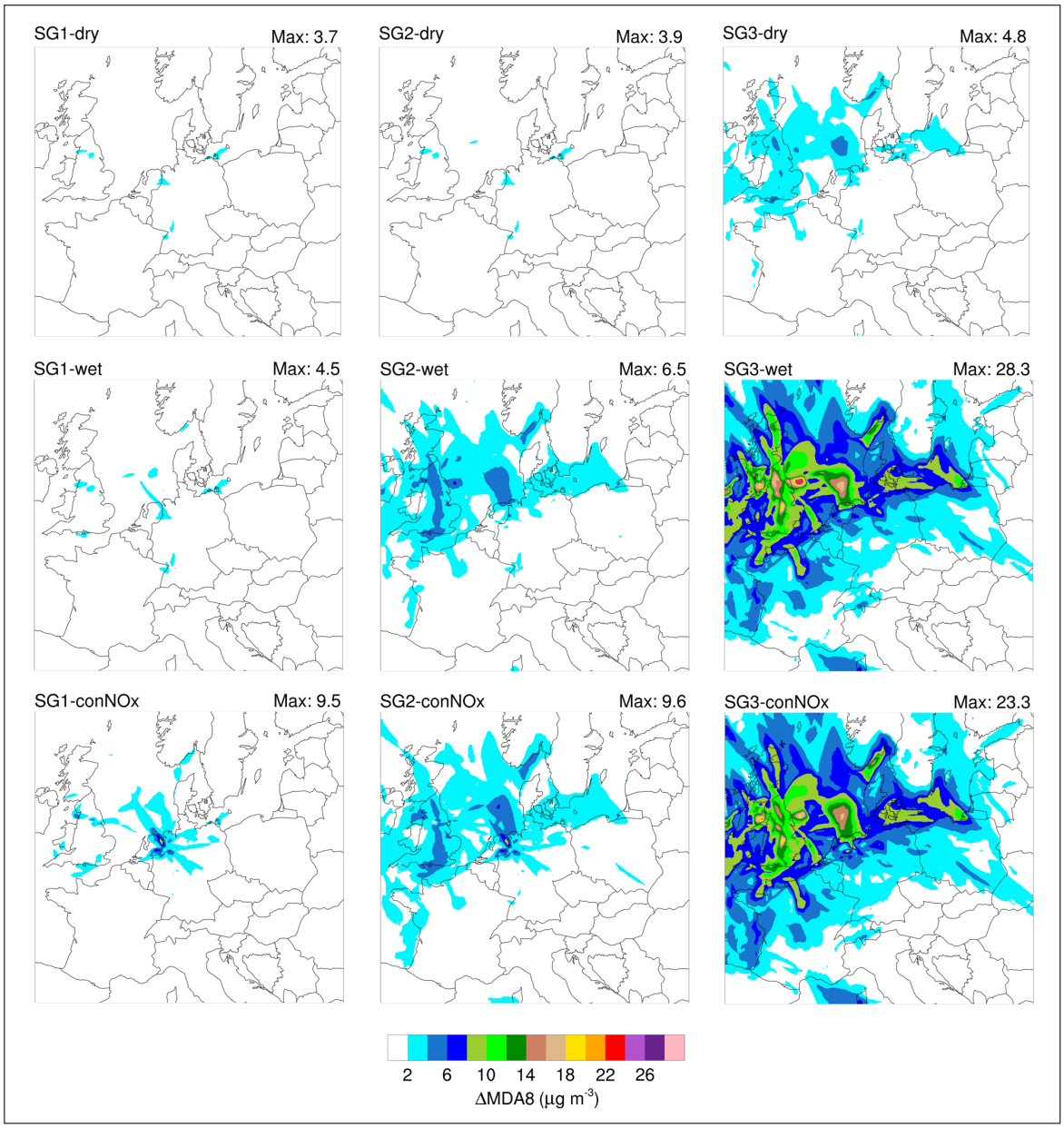

Figure 9: Maximum seasonal enhancement of scenarios on MDA8 (maximum daily 8-hour average O_3). Plots represent ΔMDA8, defined here as the maximum difference in MDA8 between the shale gas scenarios and base case over JJA, for each cell, in units of μg m^{-3}. The top left-hand corner of each plot indicates the particular scenario, and the top right-hand corner displays the peak ΔMDA8 value experienced over the domain and simulation period. DOI: https://doi.org/10.1525/elementa.387.f9

transport: Belgium, Denmark, and Norway have ΔMDA8 values up to 8–10 μg m^{-3}, France and Northern Ireland 8–12 μg m^{-3}, Kalingrad Oblast region of Russia 12–14 μg m^{-3}, and Ireland and the Netherlands 12–16 μg m^{-3}. Notably, the portion of Italy in our domain experiences ΔMDA8 values of only ~2–4 μg m^{-3}. It is interesting to note here that the weather over Europe during the simulation period was dominated by cyclonic fields (low pressure systems). Low pressure systems are generally associated with clouds, storms, and wind. In this case it means that the shale gas O_3 impacts would be more widespread, as seen in the results here. Finally, ΔMDA8 values for dry gas under SG3 are markedly less in extent and magnitude compared with wet speciation (i.e., wet gas and conNO$_x$ scenarios).

These results show that the O_3 enhancement resulting from shale gas activities taking place inside the UK and Germany can be significant, with the potential to negatively affect air quality on the local and regional scales in Europe. The general trend of model O_3 here is that as shale gas VOC emissions increase from SG1 up to SG3 and from dry gas to wet gas speciation (wet gas and conNO$_x$),

O_3 maxima increase considerably in magnitude as well as in extent over the domain. Total VOC emissions from shale gas activities (as a result of gas composition and leakage) are critical in increasing local and regional O_3 over the base case. Interestingly, when VOC emissions are relatively low (SG1), concentrating NO_x emissions leads to greater O_3 increase in magnitude and extent than when NO_x emissions are averaged; however, when VOC emissions are relatively high (SG3) O_3 maxima outcomes are similar when NO_x emissions are concentrated or averaged. This variance in model O_3 sensitivity to NO_x under different VOC loadings is likely the result of the nonlinear relationship between O_3 precursors and its formation in the atmosphere. Further the fact that concentrating NO_x emissions leads to similar or slightly greater O_3 than averaged NO_x emissions indicates a mixed regime of sensitivity to both NO_x and VOC; though, varying NO_x leads to a lower impact overall on O_3 than varying VOC emissions does. In sum, the most critical factor on increasing local and regional O_3 over the base case is the amount of VOC emissions, while concentration of NO_x emissions plays a further, albeit minor, role.

Next we explore extra exceedances of MDA8 at the locations of regulatory measurement stations, namely those which otherwise would not occur but do so as a result of shale gas activities. Here we explore the EU threshold due to its relevance for regional regulations of the study area, and examine the WHO guidelines due to the more comprehensive scope of health as well as international relevance. On account of inherent model bias in MDA8, which in our case is low yet nevertheless present, we calculate extra exceedances by the following: where an observational value is below the threshold, the difference in MDA8 between the base case and scenario at the measurement location is added to the observational value, and, when these two values together surpass the threshold, this is treated as an extra exceedance. Henceforth, we refer to these as exceedances for ease of discussion. AirBase measurement stations considered within our domain have at least 75% temporal data coverage for the JJA simulation period ("valid stations"). In the interest of clarity, it is worth noting that for countries not fully covered by the domain (e.g., Italy), only the stations of that country that are located within the domain are included in our analysis.

Figure 10 depicts the total exceedance counts, summed over the full set of valid stations, under the WHO and EU thresholds. The number of exceedances increases

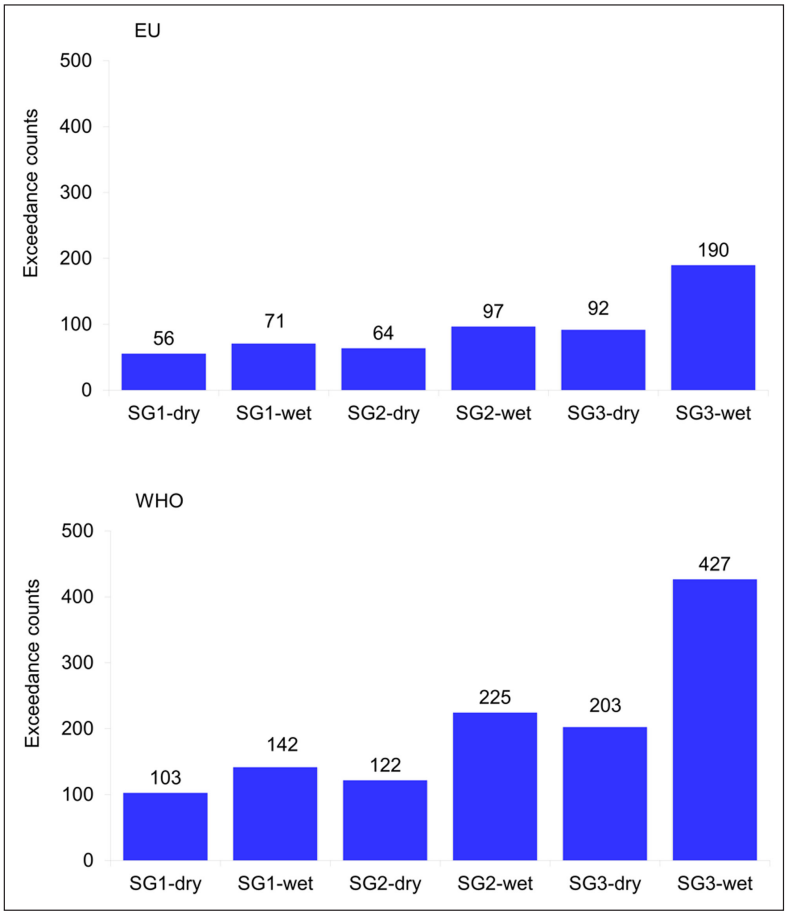

Figure 10: Total O_3 exceedance counts per scenario. The number of extra exceedances predicted as a result of O_3 enhancement from shale gas scenario emissions, over JJA, applying the EU and WHO thresholds for O_3 (120 and 100 μg m^{-3} respectively), summed over all AirBase stations within our domain that have at least 75% temporal data coverage. DOI: https://doi.org/10.1525/elementa.387.f10

substantially over the lower WHO threshold value compared with the EU threshold value, ranging from 56 to 190 (EU threshold) and from 103 to 427 (WHO threshold); this highlights the impact of the threshold value on the total number of exceedances registered. Further we see that exceedances increase considerably as VOC emissions increase: under both thresholds, the lowest number of exceedances occurs in the lowest VOC scenario (SG1-dry gas) while the greatest number of exceedances in the highest VOC scenario (SG3-wet gas). In order to put these exceedance counts into context, we examine the number of exceedances per station and then in **Table 7** present data on the percentage of stations in each country which have exceedances. The exceedances caused by the scenario emissions are spread out among the stations, rather than a few stations experiencing the majority of exceedances. The mean number of exceedances per station under all scenarios is 1 (not including stations which exhibit zero exceedances), where the maximum exceedance experienced at any station is 2–3. This demonstrates the widespread, episodic nature of shale gas emissions on station exceedances. Additionally it shows that while shale gas activities, even under extreme emission cases, have the potential to cause considerably more exceedances over large areas, the frequency of this effect on any particular station or area is low in this regard. Due to the similarity in trends when applying the EU and WHO thresholds, we show exceedances from here on applying the WHO threshold for the sake of brevity; data for the EU threshold is provided in SM Text S1, Table S3.

The number of stations with valid data over our simulation period varies strongly from country to country within our domain (**Table 7**). France has the highest number of valid stations (386) followed by Italy (244), of which 8–33% and 9–18% show exceedances, respectively (representing SG1-dry gas at the low end of the range and SG3-wet gas at the high end). Germany has the third highest number of valid stations (234), of which 6 to 21% of these stations have exceedances (SG1-dry gas to SG3-wet gas). 80 valid stations are located in the UK, of which 4–35% encounter exceedances (SG1-dry gas to SG3-wet gas). The higher percentage of British stations having exceedances

Table 7: Stations per country and exceedance data per country with respect to the WHO threshold, over JJA. DOI: https://doi.org/10.1525/elementa.387.t7

Country	SG1					SG2				SG3			
	dry gas			wet gas		dry gas		wet gas		dry gas		wet gas	
	Σ_c[a]	Σ_e[b]	%[c]	Σ_e	%	Σ_e	%	Σ_e	%	Σ_e	%	Σ_e	%
France	386	29	8	44	11	39	10	76	20	69	18	127	33
Italy	244	23	9	26	11	23	9	32	13	29	12	45	18
Germany	234	13	6	17	7	15	6	29	12	27	12	50	21
Spain	129	5	4	5	4	5	4	7	5	7	5	13	10
Austria	111	4	4	4	4	4	4	6	5	4	4	14	13
U. Kingdom	80	3	4	6	8	3	4	12	15	11	14	28	35
Poland	61	6	10	7	12	6	10	11	18	10	16	18	30
Czech Rep.	60	6	10	9	15	8	13	11	18	10	17	22	37
Belgium	42	0	0	1	2	0	0	2	5	2	5	6	14
Switzerland	30	3	10	5	17	4	13	6	20	5	17	9	30
Hungary	17	2	12	2	12	2	12	3	18	2	12	3	18
Netherlands	15	0	0	1	7	0	0	2	13	1	7	6	40
Macedonia	12	0	0	0	0	0	0	0	0	0	0	1	8
Slovenia	12	2	17	2	17	2	17	2	17	2	17	3	25
Sweden	12	0	0	1	8	1	8	1	8	1	8	1	8
Finland	11	1	9	1	9	1	9	1	9	1	9	1	9
Slovakia	11	0	0	0	0	0	0	0	0	0	0	1	9
Latvia	8	0	0	0	0	0	0	0	0	0	0	1	13
Denmark	7	1	14	1	14	1	14	2	29	2	29	4	57
Luxembourg	6	0	0	0	0	0	0	0	0	0	0	1	17

[a] Number of stations with valid measurements per country. Only country stations which are located within the model domain are included in the analysis.
[b] Number of stations that experience exceedances per country.
[c] Percentage of stations per country that have an exceedance.

compared with German stations is the result of the majority of the UK having a relatively high ΔMDA8 compared with Germany from shale gas emissions (**Figure 9**). Again, as VOC emissions increase from SG1 to SG3 and from dry gas to wet gas, exceedances generally increase. Furthermore stations in distant countries have exceedances as a result of long-range transport, e.g., Macedonia, Slovakia and Latvia. Based on these findings shale gas activities have the potential to cause exceedances at a considerable percentage of country stations (locally and in distant countries), where VOC emissions are critical to the extent of exceedances.

Plots of exceedance magnitude are presented in **Figure 11** applying the WHO guideline, and in SM Text S1, Figure S2 applying the EU threshold. We define exceedance magnitude as the difference between the scenario and base case when an exceedance occurs. This provides meaningful insight because it shows whether shale gas activities only effected an exceedance because the background O_3 concentration was already very close to the limit value (i.e., when exceedance magnitude is low), or whether shale gas activities had a robust impact on MDA8 during an exceedance (i.e., when exceedance magnitude is high). The maximum exceedance magnitude spans between 0.8 to 15.1 μg m^{-3} under the WHO threshold. Exceedance magnitudes are low (mostly <1 μg m^{-3}) in SG1, indicating that low shale gas VOCs are able to force an exceedance only when the background O_3 level is already close to the threshold. These results again reflect that the lower VOC scenarios lead to a low impact, whereas the impact is stronger with the high VOC scenarios.

While Italy is one of the top exceedance locations among the scenarios, it exhibits a very low exceedance magnitude, where in all scenarios the majority of values are <1 μg m^{-3}, with only a few values exceeding this value yet are still below 2 μg m^{-3}. This implies that background MDA8 levels are already high in Italy and close to the limit value, so that only a slight increase in MDA8 from shale gas through long-range transport is required to push it over. Consequently this results in many exceedances occurring in Italy (**Table 7**) in spite of being further away from the shale gas activity area and experiencing a low overall

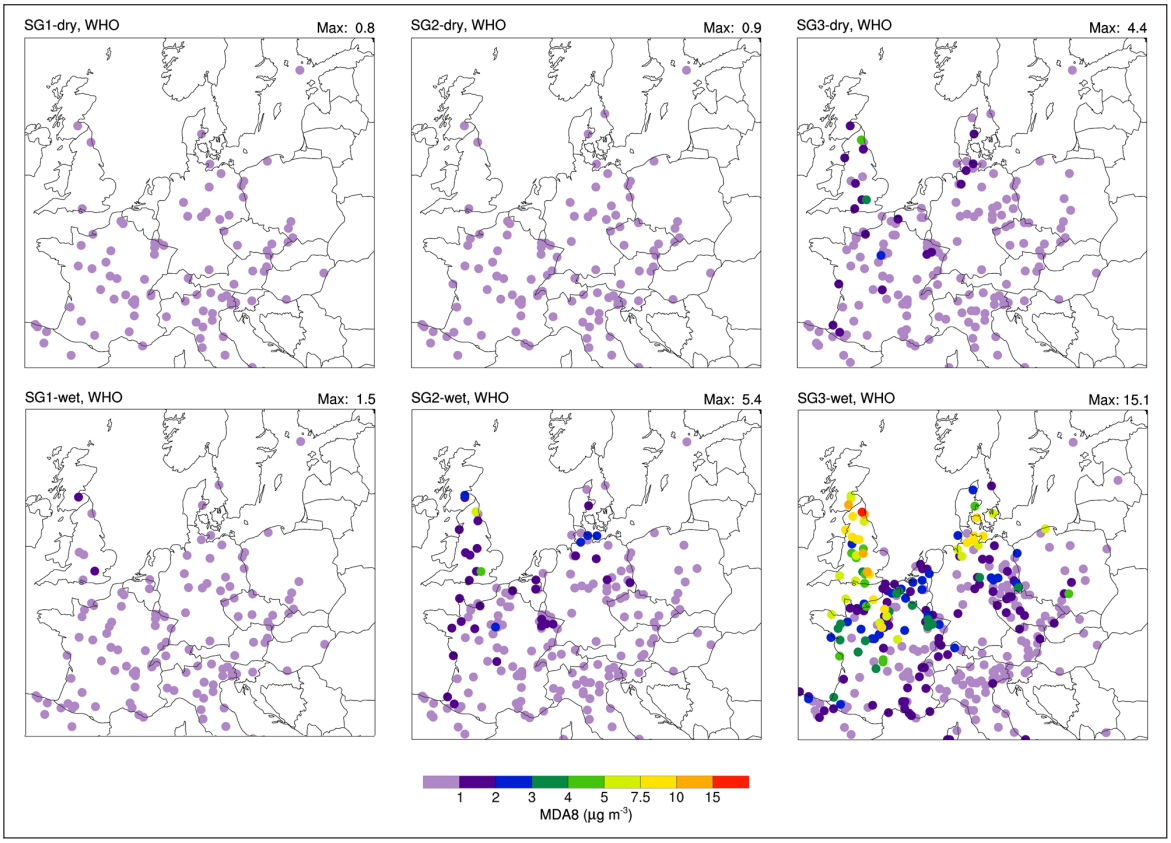

Figure 11: Spatial depiction of exceedances and corresponding exceedance magnitude. Exceedance magnitude is defined as the difference between the shale gas scenarios and base case when an exceedance occurs, and is an indicator of the robustness of shale gas emissions on an exceedance. Exceedances are displayed as filled dots at the station locations where they occur, in μg m^{-3}, over JJA, applying the WHO guideline for O_3 as the threshold (100 μg m^{-3}). For stations which experienced more than one exceedance, the maximum exceedance magnitude is shown. The top left-hand corner of each plot indicates the particular scenario, and the top right-hand corner displays the maximum exceedance magnitude value experienced over the domain and simulation period. DOI: https://doi.org/10.1525/elementa.387.f11

ΔMDA8 (**Figure 9**). The UK displays the highest exceedance magnitude of the countries in our domain, where maximum values reach >15 μg m^{-3} under SG3. This finding is in line with the surface plots displayed in **Figure 9**, which show that ΔMDA8 is highest in the UK among all countries in the domain; again, this is likely due in part to the shale gas industry and consequently VOC scenario emissions being highest in this country. The exceedance magnitude for France is relatively high under the greater VOC scenarios, which is not surprising because of its close proximity to the shale gas activity areas of both the UK and Germany and high ΔMDA8 (**Figure 9**). Additionally, France experiences a greater number of high magnitude exceedances than does Germany. This is not surprising considering that relatively high ΔMDA8 (4–6 ppb) covered a greater area in France than Germany (**Figure 9**).

SOMO35 is an indicator of accumulated O_3 exposure recommended by the WHO for use in health impact assessment, and is the sum of O_3 values exceeding an MDA8 level of 35 ppb or 70 μg m^{-3} (Equation 1) (Amann et al., 2008). The 35 ppb cutoff was chosen because the relationship between O_3 and negative health effects, as well as atmospheric models, is very uncertain below this threshold (WHO, 2013).

Equation 1

$$\mathrm{SOMO35} = \sum_{i\mathrm{day}} \max\left(0, C_{i\mathrm{day}} - 70\,\mu\mathrm{g\,m^{-3}}\right)$$

where $C_{i\mathrm{day}}$ is the maximum daily 8-hour average concentration and the summation is from i_{day} = 1 to 92 for the JJA simulation period.

Surface plots showing the percent impact of each scenario on SOMO35 levels are presented in **Figure 12**. A surface plot showing SOMO35 values for the base case is provided in SM Text S1, Figure S3. In the base run, a north-south gradient of increasing SOMO35 is apparent, where values reach as high as 7500 μg/m^3 days in some parts over the Mediterranean Sea. Under the SG1 scenario set, a low increase in SOMO35 is present over the entire domain (generally less than 1%), while slightly greater increases are localized to the German shale gas basin regions and some of the area north of the British basin. Largely in contrast with the German basins, over the British basin area

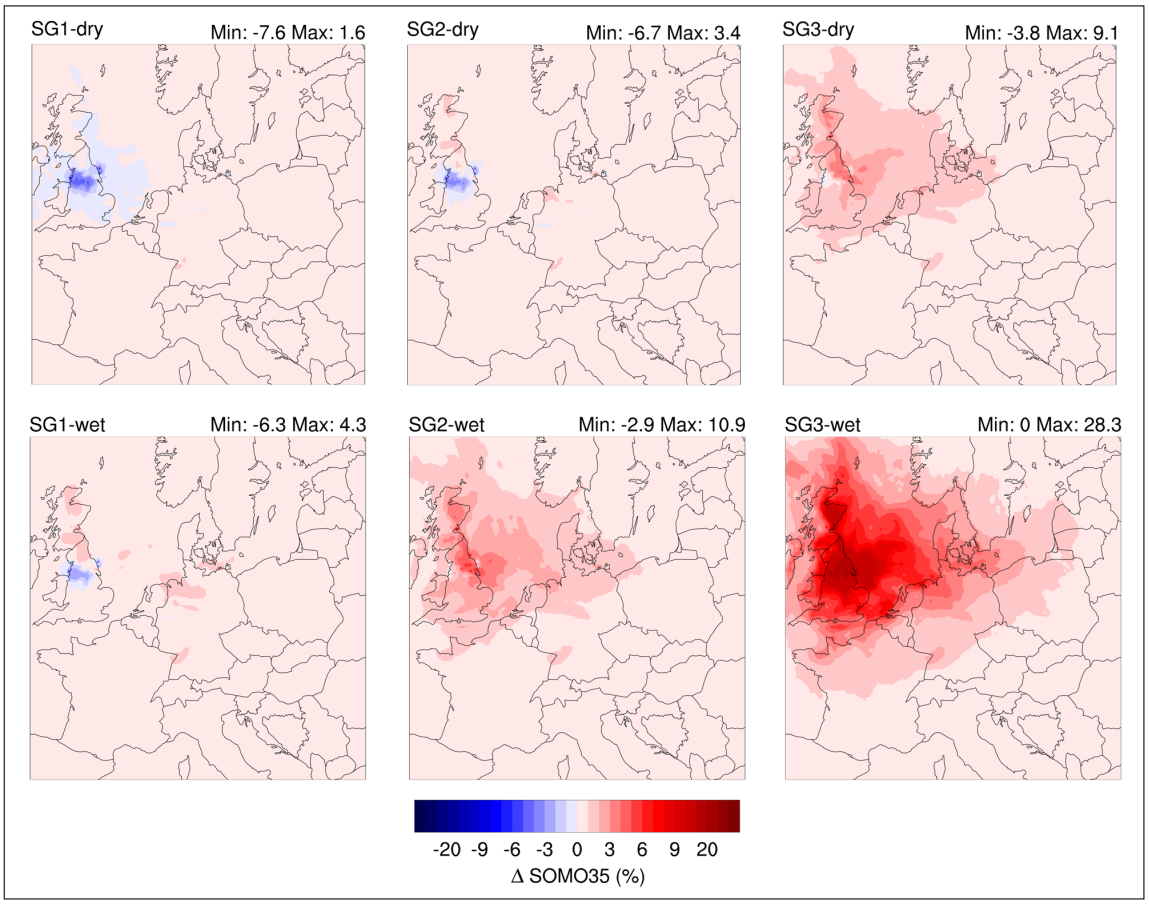

Figure 12: Percent change in SOMO35 (annual Sum of Ozone Means Over 35 ppb, daily maximum 8-hour) from scenarios compared with the base case, over JJA. SOMO35 is an indicator of accumulated O_3 exposure. The top left-hand corner of each plot indicates the particular scenario, and the top right-hand corner displays the minimum and maximum percent changes in SOMO35 values experienced over the domain. DOI: https://doi.org/10.1525/elementa.387.f12

there is a prominent decrease in SOMO35 as a result of NO_x titration. However, because the UK already experiences relatively low values of SOMO35 (SM Text S1, Figure S3), this is not expected to bring about substantial health benefits there through reduced O_3 exposure. The maximum percent increase in SOMO35 for dry gas under SG1 is only slightly less compared with wet gas, though the percent decrease covers a considerably greater area on account of greater NO_x titration due to less VOCs.

Under SG2 (wet gas) and SG3, greater increases in SOMO35 stretch over a more extensive area of the domain over the UK and Northern Germany and the surrounding region (with effects most concentrated over the UK and the North Sea). Notably, the southern portion of the domain experiences a very low percent increase meaning that VOC emissions are not expected to cause relatively worse health impacts on this region. Under SG3 wet gas, there is essentially no percent decrease in SOMO35 on account of less NO_x titration due to greater VOC emissions. The impact of VOC emissions on SOMO35 values is clear, which reach a maximum percent increase of about 28% for the wet gas scenario under SG3. On the other hand, dry gas under SG3 leads to a maximum increase of about 9%, and under SG2 still displays a percent decrease in SOMO35 that covers most of the British basin region (a maximum percent decrease in SOMO35 of circa –7%). These findings underline the important role of VOC emissions in increasing O_3 production and in turn worsening adverse health effects, and that effects are primarily localized to the countries where shale gas is being produced and the closely surrounding region.

Summary and conclusions

Our study offers the opportunity to understand and quantify potential implications from a future European shale gas industry on O_3 air quality. Here we use the WRF-Chem online-coupled regional chemistry transport model where our setup is based on Mar et al. (2016). We explore a total of nine comprehensive emission scenarios which are based on in-depth European shale gas scenarios from Cremonese et al. (2019), and which examine the effects of gas speciation, concentration of NO_x emissions over space and time, and a range of three CH_4 leakage rates on model O_3. Additionally, it is important to emphasize that our results depend on our scenario assumptions; because European shale gas does not yet exist as an industry, the results are not a predictor of what will happen in the future but rather a range of potential impacts, and further the results highlight what may be important for regulation if this industry were to come into existence.

Our results show that shale gas emissions are capable of significantly increasing O_3 concentrations (maximum of 28.4 $\mu g\ m^{-3}$ over the North Sea and maximum of about 22 $\mu g\ m^{-3}$ over land in the UK). Shale gas activities result in up to one third of all valid measurement stations in France and the UK having additional exceedances above the WHO threshold, up to about a fifth of stations in Germany, and a considerable percentage of measurement stations in neighboring and distant countries. Furthermore we find that values of SOMO35, an indicator of health impacts, can be considerable with a maximum percent increase of ~28%, which would further burden O_3-related health issues in Europe (Bell et al., 2014). This also poses concern for a future European shale gas industry where an ageing population is at greater risk to O_3-related health effects (Amann et al., 2008).

The overarching trend found in our results is that VOC emissions are critical to O_3 formation. Impacts were greatest for a scenario in which the VOC emissions are based on the assumptions that shale gas is wet and CH_4 leakage is relatively extreme. While in practice these assumptions together may be unlikely, high VOC emissions are also possible through greater shale gas production, wet gas with an even greater VOC component, and even higher gas leakage. Additionally the findings here demonstrate that concentrated NO_x emissions increase impacts on O_3. However these impacts from concentrating NO_x emissions are relatively low in comparison to impacts from increasing VOC emissions, and become less important as VOC emissions increase.

This study shows a clear potential for a future shale gas industry in Europe to adversely impact local and regional O_3 concentrations and to exacerbate already existing air quality issues in this region. Even in lower VOC emission scenarios, emissions are sufficiently high to effect a considerable number (minimum of 103 when applying the WHO threshold) of additional exceedances. Altogether these results show that future shale gas industries in Germany and the UK pose a threat to European O_3 air quality, and emission control strategies, especially for VOCs and to a lesser extent NO_x, are critical to mitigate impacts. While it is not possible to control the speciation of gas extracted, concerted effort would be required to reduce CH_4 leakage from gas production, and in turn associated VOC emissions. This would offer a three-pronged benefit by reducing detrimental outcomes for air quality, mitigating climate change, and improving the economics since less leakage means that more gas can be brought to market.

Data Accessibility Statement

All model results used in this study are available here:

Weger, Lindsey; Lupascu, Aurelia; Cremonese, Lorenzo; Butler, Tim (2019), Modeling the impact of a potential shale gas industry in Germany and the United Kingdom on ozone with WRF-Chem, v11, Dryad, Dataset, https://doi.org/10.5061/dryad.08kprr4xv

Notes

[1] (Technically recoverable) resources is the volume of gas that can be produced based on current technology. EIA. 2019. Oil and natural gas resource categories reflect varying degrees of certainty. Available at https://www.eia.gov/todayinenergy/detail.php?id=17151.

[2] Gas-in-place refers to the total amount of gas estimated in the reservoir, and not the amount that can be recovered. BGS. 2013. The Carboniferous Bowland Shale gas study: geology and resource estimation.

London, UK: British Geological Survey for Department of Energy and Climate Change.

[3] CH_4 leakage is expressed as total CH_4 emissions divided by total CH_4 (natural gas) produced, for the segment of the natural gas chain under study.

[4] Note that O_3 values are typically reported in units of parts per billion (ppb) for the US while in μg m^{-3} for the EU, where 1 ppb O_3 is equal to 2.00 μg m^{-3} at standard temperature and pressure (20°C and 1013.25 mbar). Due to the European focus of this study we will henceforth present our findings in μg m^{-3}, and list values from other studies in their original format.

Supplemental file

The supplemental file for this article can be found as follows:

- **Text S1.** Supplemental text. Text S1 includes the namelist used in the WRF-Chem simulations. A detailed description of the conNO_x scenario development is provided. Statistical data on MDA8 enhancement from shale gas scenarios are included. Exceedance data and plots resulting from scenarios applying the EU threshold for O_3 are also included. Additionally a wind rose diagram of the domain is included, as well as a surface plot of SOMO35 for the base case simulation. DOI: https://doi.org/10.1525/elementa.387.s1

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Competing interests

The authors have no competing interests to declare.

Author contributions

- Contributed to conception and design: LW, AL, LC, TB
- Contributed to acquisition of data: LW, AL, LC
- Contributed to analysis and interpretation of data: LW, AL, LC, TB
- Drafted and/or revised the article: LW, AL, LC, TB
- Approved the submitted version for publication: LW, AL, LC, TB

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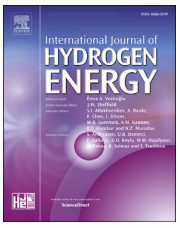

Expected impacts on greenhouse gas and air pollutant emissions due to a possible transition towards a hydrogen economy in German road transport

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HIGHLIGHTS

- Emission impacts from a full shift to German H_2 mobility are studied via scenarios.
- H_2 mobility could contribute significantly to Germany's climate & air quality goals.
- Total annual emissions could be cut by up to 179 $MtCO_2eq$ if green H_2 were used.
- Shifting only HDVs to green H_2 would also aid a deep emissions cut (−57 $MtCO_2eq$).
- HDVs represent a low-hanging fruit for road transport decarbonization with FCEVs.

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ABSTRACT

Transitioning German road transport partially to hydrogen energy is among the possibilities being discussed to help meet national climate targets. This study investigates impacts of a hypothetical, complete transition from conventionally-fueled to hydrogen-powered German transport through representative scenarios. Our results show that German emissions change between −179 and +95 $MtCO_2eq$ annually, depending on the scenario, with renewable-powered electrolysis leading to the greatest emissions reduction, while electrolysis using the fossil-intense current electricity mix leads to the greatest increase. German energy emissions of regulated pollutants decrease significantly, indicating the potential for simultaneous air quality improvements. Vehicular hydrogen demand is 1000 PJ annually, requiring 446–525 TWh for electrolysis, hydrogen transport and storage, which could be supplied by future German renewable generation, supporting the potential for CO_2-free hydrogen traffic and increased energy security. Thus hydrogen-powered transport could contribute significantly to climate and air quality goals, warranting further research and political discussion about this possibility.

Abbreviations: AD, activity data; BEV, battery electric vehicle; CCS, carbon capture and storage; CG, coal gasification; DE, Germany; EF, emission factor; FCEV, fuel cell electric vehicle; GWP, global warming potential; HDV, heavy duty vehicle; ICEV, internal combustion engine vehicle; IEA, International Energy Agency; LCA, life cycle assessment; LDV, light duty vehicle; LHV, lower heating value; NIR, National Inventory Report; PC, passenger car; PEM, proton exchange membrane; SMR, steam methane reforming; SOEC, solid oxide electrolysis cell; TTW, tank-to-wheel; UBA, Umweltbundesamt (German Environment Agency).

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Introduction

Transport is one of the most emission-intensive sectors for both climate forcers and air pollutants [1–3], yet meaningful mitigation of this source proves to be particularly challenging. In 2017, German transport was responsible for 18.4% of CO_2eq emissions, 96% of which comes from road traffic [4]. While Germany has decreased its emissions considerably in most areas of the economy since 1990, abatement of the transport sector has made little progress [4]. The major reasons for this are increasing kilometers traveled, the continued dominance of fossil fuels in transport, and high average vehicular CO_2 emissions [4]. The transport sector is in large part responsible for Germany failing to meet its target of a (lasting) 40% GHG emissions cut by 2020 compared to 1990 levels [5]; namely, it was originally estimated in 2019 that Germany would only achieve a total emissions reduction of about 33% [6]. Due to extraordinary circumstances, including countermeasures taken to contain the COVID-19 pandemic, Germany is now set to meet its original 2020 reduction target [7]. However, this is not expected to be a lasting reduction—for example, transport sector emissions were back to near normal levels already by mid-June.

Hydrogen energy not only offers the opportunity to decarbonize road transport, but also to strongly reduce local air pollution [8–13]. Hydrogen is a non-toxic, colorless, odorless gas, and has been safely produced and used in industry and space exploration for decades [14,15]. It possesses the highest energy density by mass among common fuels (though not by volume), and importantly its fueling infrastructure is comparable to that of conventional road fuels. It is also versatile: hydrogen can be produced from a wide array of energy forms, including renewable electricity; it can be easily stored, such as compressed or liquefied in pure form, in a blend with natural gas, or bound with larger molecules; and it can be easily transported by pipeline, truck or ship [9]. Moreover, hydrogen use in vehicles is safe [16] and in many ways even more so than gasoline and diesel, one of the important reasons being that hydrogen is 14 times lighter than air and thus disperses rapidly in the event of a leak, thereby lowering the risk of ignition (in contrast to gasoline and diesel) [15]. The most relevant powertrain for hydrogen energy is the fuel cell electric vehicle [FCEV]: this offers the advantage of high tank-to-wheel [TTW] efficiency, roughly two to three times greater than conventionally-fueled internal combustion engine vehicles [ICEVs] [17,18]. Additionally, FCEVs have zero tailpipe emissions with the only exhaust being water vapor, and produce virtually no noise when driving [8].

FCEVs also enjoy important advantages over battery electric vehicles [BEVs], in particular longer driving ranges (≥500 km) and shorter fueling times (approx. 3 min), both being comparable to ICEVs [19,20]. The longer range is afforded through higher energy density of compressed hydrogen compared with lithium-ion batteries [9]. If battery capacity is increased to extend BEV range, battery and vehicle mass (not to mention cost) likewise increase so that more energy is required to move the vehicle itself, which leads to diminished returns on range [21,22]. Indeed FCEV's greater on-board energy storage capability make it particularly competitive for segments of the fleet that require high payloads or extended range, i.e., heavy duty vehicles, long-haul transport, and passenger vehicles for long-distance travel [9,23,24]. Furthermore, a recent survey found that 78% of automotive industry executives believe that FCEVs will be the breakthrough for electric mobility—in large part due to their short fueling time—and that the long recharging time of BEVs will remain an insurmountable obstacle to their widespread acceptance [25]. This is likewise an important aspect for the trucking industry, for which long charging times may prove to be economically unacceptable [26]. Moreover quick fueling of the hydrogen tank does not impair FCEV lifetime, whereas high charging/discharging rates, in addition to overcharging, deep discharging and the climate all negatively impact BEV battery lifetime [13]. Hydrogen fueling stations can also service more vehicles than a BEV charging station, and a greater area on account of FCEV's longer range. Finally, the material manufacturing footprint of FCEVs (fuel cells, hydrogen tank and battery) could be lower than that of BEVs (battery) [27], though there is considerable uncertainty around such comparisons at present.

Transitioning to clean hydrogen energy in road transport nevertheless faces several challenges. While zero-carbon hydrogen is already possible via renewable-powered water electrolysis (green hydrogen), the vast majority of hydrogen production today is based on coal and natural gas (grey hydrogen) generating approximately 830 $MtCO_2$ per year [9]; to put this into perspective, this represents about 2% of total global anthropogenic CO_2 emissions for 2019 [28]. Cost represents another obstacle: FCEVs, fueling stations, and green hydrogen production—on account of renewable electricity and electrolyzers (i.e., electrolysis technology)—are all currently expensive [9,13,24,29]. For widespread FCEV adoption, hydrogen infrastructure is needed yet development has been slow thus far [9]; in contrast, BEVs are relatively mature in terms of lower capital and operating costs and readily-available infrastructure [13]. Electricity-based hydrogen for FCEVs involves the steps of converting electricity to hydrogen, transporting it (if made offsite), compressing or cryogenically storing it to obtain sufficient volumetric energy density, precooling it (if compressed) and converting it back to electricity with a fuel cell. This introduces energy losses that are avoided when using electricity directly via BEV. Moreover state-of-the-art electrolysis requires freshwater as input, which is a limited and valuable resource. While desalination can alternatively be employed to enable the use of seawater, this introduces energy and financial costs (albeit minor); research is currently exploring ways to use seawater in electrolysis directly [9,30]. Accordingly, the so-called hydrogen economy has seen waves of great expectation over the years that eventually came to naught.

Yet the International Energy Agency [IEA] recently announced that clean hydrogen is now experiencing "unprecedented political and business momentum" [9]. A multitude of countries, including those with the world's largest economies, have policies and projects in place for hydrogen energy [9]. In

fact, Germany approved its highly anticipated national hydrogen strategy in June 2020 [31]. In 2017 the Hydrogen Council was launched by a group of leading global energy, transport and industry companies to bring together political and private stakeholders, with the goal of fostering hydrogen as "a key element of the energy transition" [32]; among their members are several German companies. Commercial FCEVs have already been in large-scale production for several years, with approximately 500 passenger FCEVs on the road in Germany [33] and about 17,000 worldwide [34] at the end of 2019. While these figures are lower in comparison with BEVs (5.1 million worldwide in 2018) [35], Toyota has targeted to sell over 30,000 FCEVs annually starting from 2020 [36]. There are now well over 400 hydrogen fueling stations in operation globally as of 2019, with Germany boasting the second largest network with about 90 stations [37]. It is widely reported that through further research and development, continued declining costs of renewable power, economies of scale, and coordinated energy policy and investment, costs can be appreciably reduced and technological challenges overcome for renewable hydrogen [9,13,23,24,29,38–40]. For instance, a recent report by the Hydrogen Council found that green hydrogen will become cost-competitive with grey hydrogen over the coming decade, after which point its costs will continue to decline [24]. Moreover, range, load and fueling advantages as outlined above can make FCEVs competitive with BEVs. In any case, the major reason that hydrogen commitment may be different this time around is the increased sense of urgency to adequately address climate change and ambition to deeply reduce emissions, as evidenced by the 2015 Paris Agreement and the landmark 2018 IPCC report to limit warming to 1.5°C above pre-industrial levels [9,29,41,42].

It should be noted, however, that hydrogen is leaked along its utilization chain, which impacts both the climate and air pollution. For example, Derwent et al. [43] recently reported a global warming potential [GWP] for hydrogen of 5 over a 100-year timescale. This makes potentially rising tropospheric hydrogen emissions from a hydrogen economy an important consideration. It is nevertheless worth noting that the effects from hydrogen emissions are highly uncertain, and that any adverse impacts they cause are likely to be less than those caused by current fossil fuel usage which would be replaced by hydrogen fuel [44]. Yet there is a lack of data on hydrogen emissions, and to our knowledge no published data currently exists on hydrogen loss from commercial FCEVs.

In this context, hydrogen emerges as a viable means of decarbonizing hard-to-abate sectors like road transport in which electrification alone may be insufficient to help Germany achieve its ambitious climate targets, culminating in GHG neutrality by mid-century[1] [9,23,45,46]. Additionally, this would serve to improve air quality, promote energy security, economic growth, as well as technological leadership in a potentially core field of the future global energy system. Indeed, Germany sees hydrogen as a "central pillar" of its energy transition, and is working to maintain its reputation for technological leadership by securing itself as the global leader in hydrogen technologies [47–49]. It is important to note, however, that the future mobility mix is expected to be diverse rather than there being a winner-takes-all technology; FCEVs will likely be complemented with low-carbon technologies like BEVs.

Scenarios can serve as an important tool for assessing GHG and air pollutant emission impacts of possible, relevant transitions in the energy system to provide valuable insight and support informed dialogue. Over the years, several scenario studies quantifying emission impacts from hydrogen implementation in the mobility sector have been performed, from the city level up to the global scale, and with many focusing on the European region in particular, e.g., Ref. [50–56]. In terms of German-focused studies, Rocco et al. [57] carried out a life cycle assessment [LCA] including an analysis of GHG emission impacts from penetration of FCEVs in the German road transport sector in 2050. Additionally, Emonts et al. [58] executed a pathway analysis exploring renewable hydrogen penetration in the German passenger car transportation sector via FCEVs by the year 2050, with their investigation also including the CO_2 reduction potential of the transport sector. To our knowledge, however, there have been no published studies on GHG *and air pollutant* emission impacts from a widespread shift to hydrogen-powered traffic in Germany in the near-term. All of these aspects are valuable for investigation based on the discussion above (noting that a focus on the near-term is important given the current momentum behind hydrogen mobility).

This paper investigates the impacts of a possible, complete transition from conventional fossil fuels to hydrogen energy in German road transport on GHG and air pollutant emissions, through a variety of emission scenarios covering relevant hydrogen production choices and variables as described herein. Our emission scenarios are comprehensive as they encompass emissions incurred from hydrogen production and those avoided by replacing conventional road transport fuel. Emission results are presented and put into context by comparing changes in CO_2eq with German total emissions, and by comparing changes in air pollutants with German energy emissions, for the year 2016. Other important parameters are also examined including the maximum allowable hydrogen leakage from FCEVs to avoid a net increase in hydrogen emissions, road transport sector hydrogen demand, and energy required to achieve this level of hydrogen demand. This work is an exploratory study with the main objective of understanding overall emission impacts of such a potential transition, using illustrative scenarios rather than assessing a realistic implementation thereof and estimating precise outcomes, which are currently too dependent on extensive, unpredictable policy developments to be reliable or useful. The results from these scenario studies can support informed discussion among policymakers, the public and other relevant stakeholders on hydrogen mobility in Germany and beyond.

Methodology

Study

In the following we describe the scenario design, and provide a summary of the main points in Table 1. The scenarios

[1] Germany's climate targets include 40% GHG emissions reduction by 2020, 55% by 2030, 70% by 2040 and 80–95% by 2050, compared with 1990 levels.

Table 1 – Summary of the scenario design.

Scenario element	Element description
Data year[a]	2016
German road transport fuels replaced with hydrogen energy[b]	Gasoline, diesel
Road transport vehicle categories switched to hydrogen technology[c]	PCs, LDVs, HDVs, two-wheelers
Domains of activity in emission scenario model	Road transportation, hydrogen economy, natural gas production, steam methane reforming, gasoline production, diesel production, coal production, coal gasification, electricity generation, electrolysis, hydrogen transport and storage, LPG production, and biofuel production
Emission source segments	Energy production and use
Specie emissions quantified	CO_2eq (CH_4 & CO_2), NMVOCs, NO_x, $PM_{2.5}$ and PM_{10}, CO, SO_x, NH_3

[a] Data representing the year 2016 was used, where possible.
[b] Other forms of German 2016 road transport energy are left unaltered (LPG, CNG, biofuels, electricity).
[c] In the scenarios *SMR-ng1-C_HDV* and *Elec-renewable_HDV*, only HDVs are switched to hydrogen technology.

investigated in this study cover several assumed hydrogen production methods (Table 2). Conventional fuels replaced are gasoline and diesel, which together supply 94.2% of German road transport energy for the year 2016 [59,60]. Consumption of alternative transport energies, i.e., electricity, LPG, CNG, and biofuels, are unaltered as they fall within Germany's low-carbon transition strategy for transport [5]. Hydrogen replacement is applied to all road vehicle categories: passenger cars [PCs], light duty vehicles [LDVs], trucks and buses, here collectively referred to as heavy duty vehicles [HDVs], and motorcycles and mopeds, here collectively referred to as two-wheelers. Additionally, two scenarios focus solely on HDVs due to the fact that certain advantages of FCEVs (i.e., longer range, heavy loads, and quick fueling times) are particularly consequential to this vehicle category, as discussed in Section Introduction. It is assumed that hydrogen is produced in Germany, and based on the literature that hydrogen delivery is by pipeline [9] and storage by compression at 700 bar [20,61].

Both GHG and air pollutant species are examined, including: CH_4 and CO_2 (analyzed together as CO_2eq), and NMVOCs, NO_x, $PM_{2.5}$ and PM_{10}, CO, SO_x, and NH_3. These pollutants are particularly relevant as their emissions are regulated under the European Union's National Emission Ceilings Directive (noting that of the SO_x compounds, specifically SO_2 is regulated) [62]. In keeping with the German National Inventory Report [NIR] which follows the Revised UNFCCC Reporting Guidelines, CO_2eq emissions are calculated here using the IPCC AR4 100-year GWP for CH_4 of 25 [63]. It should be noted that in the more recent IPCC AR5, the corresponding GWP is 36; furthermore, the 20-year timescale is also commonly used and results in a higher GWP of 87 due to CH_4's comparatively short atmospheric lifetime (~12 years) [64]. As a result, using AR5 data and the 20-year timescale would lead to higher CO_2eq emissions for scenarios with CH_4 emissions than the results presented here.

Emissions are quantified by multiplying activity data [AD] with emission factors [EF]. The scenarios are constructed as a projected snapshot of the present-day situation, in which the proposed changes are enacted immediately. Additionally, the scenarios are built by quantifying and aggregating emissions associated with each domain of activity relevant to hydrogen production and German road transport. The domains and the information flow between them forming the basis of the emission scenario model used in this study are depicted in Fig. 1. The model is summarized and the domains are presented in detail in the Supplement (Section S1); the model methodology is based on that described in Ref. [65] which was published in Ref. [51].

Emissions examined are those associated with energy production and use. It is important to note that scenario emissions include those released from activities occurring within Germany and abroad. Namely, emissions from upstream activities of imported fossil fuels are not German emissions (described further in the Supplement, Section S1.14). Due to the focus of this paper, attention is specifically on German emissions rather than total values unless explicitly stated. As this work does not have the aim of being an LCA, emissions associated with, e.g., the manufacture of materials for or construction of fuel cells and power plants, are not considered. Being an exploratory study, limiting socio-economic aspects are likewise not considered.

Table 2 – Hydrogen production methods explored in this work.

Method	Abbreviation	Description
Steam methane reforming	SMR	Natural gas is reacted with steam producing syngas (mainly CO/hydrogen). This subsequently undergoes the water-gas shift reaction yielding more hydrogen (and CO_2) [66].
Water electrolysis	Elec	Electricity is used to separate water into hydrogen and oxygen in a unit known as an electrolyzer [67].
Coal gasification	CG	Coal is reacted with steam and oxygen at high temperatures and pressures forming a gaseous mixture, which is then scrubbed to remove impurities, producing syngas (mainly CO/hydrogen). This subsequently undergoes the water-gas shift reaction yielding more hydrogen (and CO_2) [68].

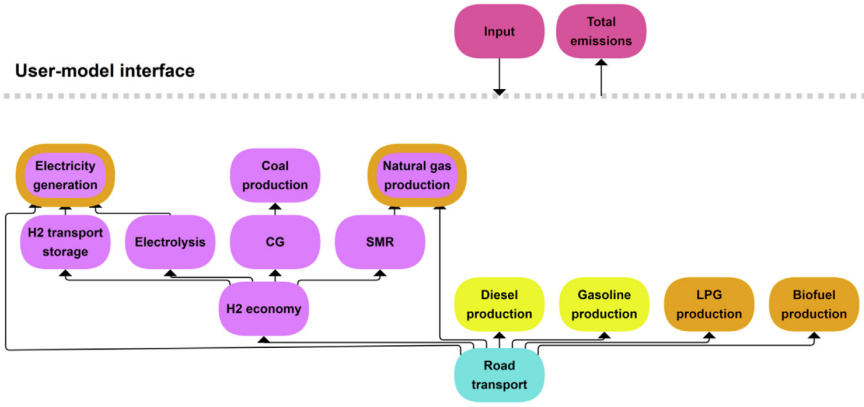

Fig. 1 – Schematic of information flow between domains forming the basis of the emission scenario model of this study. The arrows follow the flow of energy demand from end-use application, i.e., road transport, to the point of energy production. The main groups are hydrogen (purple), conventional fuels (yellow), and alternative fuels (orange).

All input data was obtained from peer-reviewed publications, official reports and institutions, and expert support (presented and described in detail in the Supplement, Section S1). The majority of EFs and AD represent the year 2016, which was the most recently available at the time when this study was conducted. However, hydrogen-related technology data (e.g., electrolysis efficiency) is generally based on present-day values. If German-specific data was not available, the best available data was used and adapted to Germany, where possible. Energy use data is based on the lower heating value [LHV], i.e., the condensing heat of vaporized water produced from combustion is not included. Finally, a sensitivity analysis has been performed to assess the impact of varying FCEV TTW efficiency on total emissions and is provided in the Supplement (Section S2).

Scenarios

Today, ~75% of global hydrogen production is based on natural gas, mainly via SMR, which is projected to remain the chief technology in the near-term [9]. While water electrolysis currently contributes <0.1% to the global supply [9], it enables green hydrogen (when powered by renewables), which is viewed as essential to the energy transition and is projected to have significant growth in the years ahead [40,69]. Thus SMR and electrolysis are highly pertinent and explored here. For these production types the impact of varying the following important parameters are also examined: CH_4 leakage rates from natural gas production (SMR), the current electricity mix vs. renewable electricity (electrolysis), electrolysis efficiencies (electrolysis), and centralized and decentralized hydrogen production (SMR and electrolysis).

Coal (via CG) accounts for nearly ~25% of today's global hydrogen production as a result of its predominance in China [9]. For completeness and in the interest of comparison with the other technologies, a scenario based on CG is explored due to its relevance to global hydrogen production and coal to German energy (e.g., coal will remain in Germany's electricity mix up to 2038 based on the recently adopted coal exit law [70]).

It is worth noting that carbon capture and storage [CCS] is a relevant technology with the potential to substantially lower the CO_2 emissions intensity of fossil fuel-based hydrogen (blue hydrogen) [9]. However, blue hydrogen is not emission-free: 5–15% of CO_2 remains uncaptured under optimal technological conditions [29], and CH_4 is still leaked throughout natural gas production and transport, while at the same time CCS necessitates more energy to run. Moreover, many open questions remain about its feasibility due to lack of progress [29], technological shortcomings, and Germany's low public acceptance of CCS [23], while the breadth and depth of technological options and lack of data clarity make it challenging to properly factor CCS into the emissions scenarios. For these reasons, blue hydrogen is not explored in this study.

Some emerging methods for low-carbon hydrogen production not considered in the present study include methane cracking and thermochemical water splitting [9,71–73]. The former technology involves the splitting of natural gas under high temperatures and in the absence of oxygen resulting in hydrogen and carbon black, and is currently at the pilot scale. The latter technology involves the splitting of water under high temperatures achieved by the concentration of solar energy, with the first pilot plants now in operation.

Finally, for each scenario, total hydrogen production is considered from one method alone and not a combination of different methods. This was done on account of the illustrative nature of the scenarios, and because the main goal here is to explore the benefits and trade-offs of each possibility. The scenarios are detailed in Table 3 and depicted in Fig. 2.

Results and discussion

Hydrogen demand

The total hydrogen demand required for replacing gasoline and diesel in road transport for all vehicle categories is

Table 3 – Description of emission scenarios explored in this study. The scenarios are grouped into sets by hydrogen production method, in addition to a baseline scenario for comparison. The SMR and electrolysis sets have multiple scenario variables which are combined, yielding four SMR scenarios, and five *Elec* scenarios (only one of which is based on renewable electricity because emissions are unaffected by the other variables given that renewable power generation assumes zero emissions); the CG set has no variables and therefore only has one scenario. The data used in the scenarios is provided in the Supplement (Section S1).

Scenario set	Variables	Variable description
Baseline	–	Present day (year 2016) emissions associated with the German road transport sector are quantified, including emissions from fuel combustion, gasoline evaporation, and energy production of road transport fuels.
Steam methane reforming: SMR	CH_4 leakage rate (natural gas production): *ng1*, *ng2*	Two sets of CH_4 leakage rates for natural gas production are examined: *ng1* (up-/downstream: 1.0%, 0.2%) and *ng2* (up-/downstream: 2.2%; 0.1%). Natural gas is mainly CH_4, a potent GHG, making leakage thereof an important consideration; thus this has been an area of intense research and discussion for many years, with studies reporting leakage rates from <1% to >10% of production [74]. *Ng1* is based on data from Ref. [75] and tailored to natural gas supply in Germany; as such it may be viewed as a standard estimate. *Ng2* is from a recent study [76] that found natural gas CH_4 leakage from about one third of production in the US to be 60% higher than official estimates; thus *ng2* represents a higher, yet plausible rate.
	Production site: *-C*, *-D*	Two cases of hydrogen production are examined: centralized ('*C*'; 100% at the plant) and decentralized ('*D*'; 100% at the hydrogen fueling station). The thermal efficiency of centralized SMR is higher ($\eta = 75\%$), it avoids downstream CH_4 emissions from distribution to the station, and the energy required for hydrogen compression is slightly lower; however, hydrogen must instead be transported which requires a low amount of energy. Decentralized SMR efficiency is slightly lower ($\eta = 67\%$), and energy needed for hydrogen compression slightly higher, but it avoids energy costs for hydrogen transportation; instead, CH_4 must be transported to the station which incurs downstream CH_4 emissions (though no additional energy).
Electrolysis: *Elec*	Electrolysis efficiency: *ef1*, *ef2*	Two efficiencies for low-temperature water electrolysis are examined: *ef1* ($\eta = 59\%$) and *ef2* ($\eta = 71\%$). Electrolysis efficiency is critical to the total electricity demand, and hence cost. Moreover, there is ongoing research to further improve the efficiency, making it a valuable parameter to explore. These efficiencies are based on proton exchange membrane [PEM] electrolysis, though values are similar for alkaline electrolysis (both technologies are among the most mature electrolysis methods today).
	Electricity supply: *cmx*, *renewable*	Two cases of the electricity supply are examined: 100% current mix ('*cmx*') and 100% renewable. *Cmx* EFs are averaged values of the current (2016) electricity mix in Germany. *Renewable* EFs are zero assuming wind and solar as the electricity sources.
	Production site: *-C*, *-D*	Like SMR, two cases of hydrogen production are examined: centralized ('*C*'; 100% at the plant) and decentralized ('*D*'; 100% at the hydrogen fueling station). The same tradeoffs between *C* and *D* production exist for electrolysis as for SMR, however the efficiency of water electrolysis does not differ between *C* and *D*.
Coal gasification: *CG*	–	Centralized CG is assumed with a thermal efficiency of 50.8%. It is important to emphasize that CG is not considered a realistic option due to coal's high emission and pollution intensity and that it is being phased out in Germany (thus no further variables are explored). Rather, CG is included in this study in the interest of completion and comparison with the other scenarios.

1000 PJ (Fig. 3). PCs and HDVs make up the majority of hydrogen demand in the scenarios (~95%), closely mirroring the share of total conventional fuel demand by these vehicle categories; thus the share required to replace conventional fuels for LDVs and two-wheelers is low (~5%). Hydrogen demand is about two thirds less (371 PJ) in the scenarios where only HDVs are targeted for fossil fuel replacement. To put these values into perspective, this work estimates 55 PJ hydrogen use in German refining of gasoline and diesel, one third of which is assumed as already achieved as a by-product through naphtha reforming. Accordingly, massive upscaling of hydrogen production will be necessary to meet the level of road transport demand in these scenarios. Also worth noting is that total hydrogen demand amounts to slightly less than half of gasoline and diesel energy demand in German 2016 road transport (2103 PJ) [59] on account of FCEV TTW efficiency being roughly twice that of conventionally-fueled ICEVs.

It is worth noting that hydrogen demand is affected by the assumed TTW (vehicle) efficiency. As discussed in the Supplement (Section S1.1), hydrogen demand may be slightly overestimated due to more recent (i.e., higher) ICEV TTW efficiencies applied to the entire auto fleet, which would imply a slightly less favorable emissions outcome from the shift to

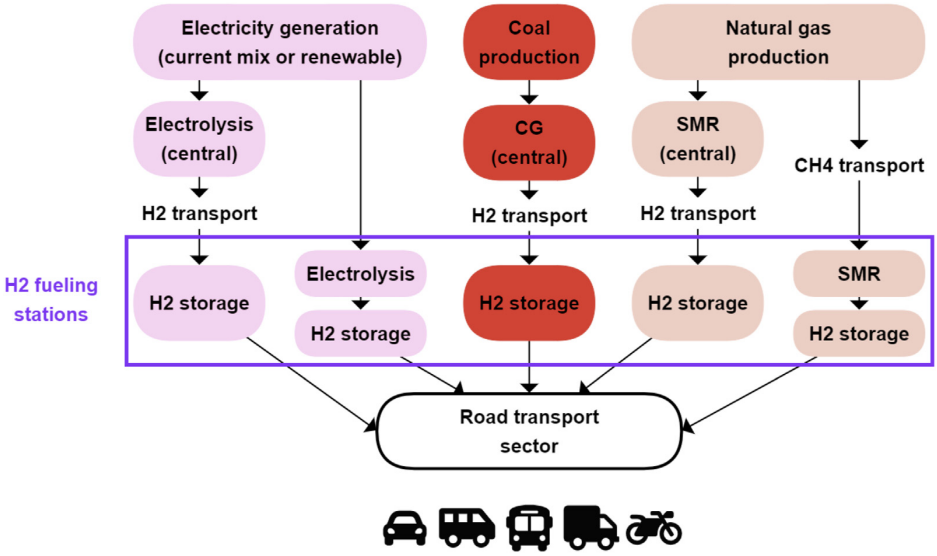

Fig. 2 – Steps in utilizing hydrogen energy in road transport for each scenario set. Scenario sets are Elec (pink), CG (red), and SMR (beige). In order to distinguish the scenarios sets of this study and avoid confusion, different colors are used than the standard hydrogen production color terminology. The average transport distance is about 6 km for hydrogen [61] and 2500 km for natural gas [77] based on the data used in this study (noting that the natural gas transport distance is based on CO_2 emissions data).

hydrogen transport than would be realized in reality. On the other hand, TTW efficiency data on FCEVs is rare and therefore a value representative of commercial PCs was applied to all vehicle categories in this study; this may over- or underestimate hydrogen demand and hence total emissions. Namely, the impact on total scenario CO_2eq emissions ranges from +10% to −5% when assumed lower and upper FCEV TTW efficiencies are applied, respectively, and based on the setup of our sensitivity analysis. However, the greatest sensitivity was seen among PCs, (i.e., for which the FCEV TTW efficiency was suited).

Production energy

Producing the level of hydrogen demand needed to cover all vehicle categories (i.e., 1000 PJ) by SMR requires between 1333 and 1492 PJ of annual natural gas for centralized and decentralized production, respectively (Fig. 4); this is equivalent to slightly less than half (44% and 49%) of 2016 German total natural gas primary energy consumption (3056 PJ) [78]. Coal demand for hydrogen production is higher (1968 PJ) on account of the lower thermal efficiency of CG (50.8%) compared with SMR (centralized: 75%; decentralized: 67%); this is equivalent to more than half (61%) of 2016 German total coal primary energy consumption (3204 PJ) [78].

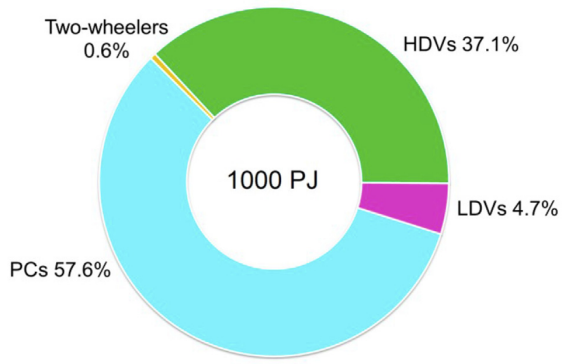

Fig. 3 – Hydrogen demand to power the 2016 German road transport sector to replace gasoline and diesel.

Based on the electrolysis efficiency, annual electricity demand for hydrogen production is between 391 and 466 TWh (*ef2* = 71%$_{LHV}$ and *ef1* = 59%$_{LHV}$, respectively), which equals 64% and 76% of 2016 German net electricity generation (614 TWh) [79]. It will be critical to optimize electrolysis efficiency to reduce the burden on renewable electricity demand. Yet the efficiencies of mature electrolysis technologies (i.e., PEM and alkaline) are not expected to improve significantly beyond the *ef2* explored in this work (note that *ef2* represents PEM efficiency estimated for 2030, as described in the Supplement, Section S1.10). For example, in the IEA's The Future of Hydrogen report [9], a long-term efficiency of up to 74%$_{LHV}$ for PEM and of up to 80%$_{LHV}$ for alkaline electrolysis are projected. On the other hand, solid oxide electrolysis cells [SOECs], the least developed technique, can offer higher efficiencies of up to ~80%$_{LHV}$ today and 90%$_{LHV}$ long-term [9].

The amount of electricity required for hydrogen transport and storage (including fueling) is 55 and 59 TWh for decentralized and centralized hydrogen production, respectively; the electricity demand for centralized production is slightly higher on account of additional electricity required to deliver hydrogen via pipeline. According to Ref. [80], Germany has the potential to more than meet total scenario electricity demand

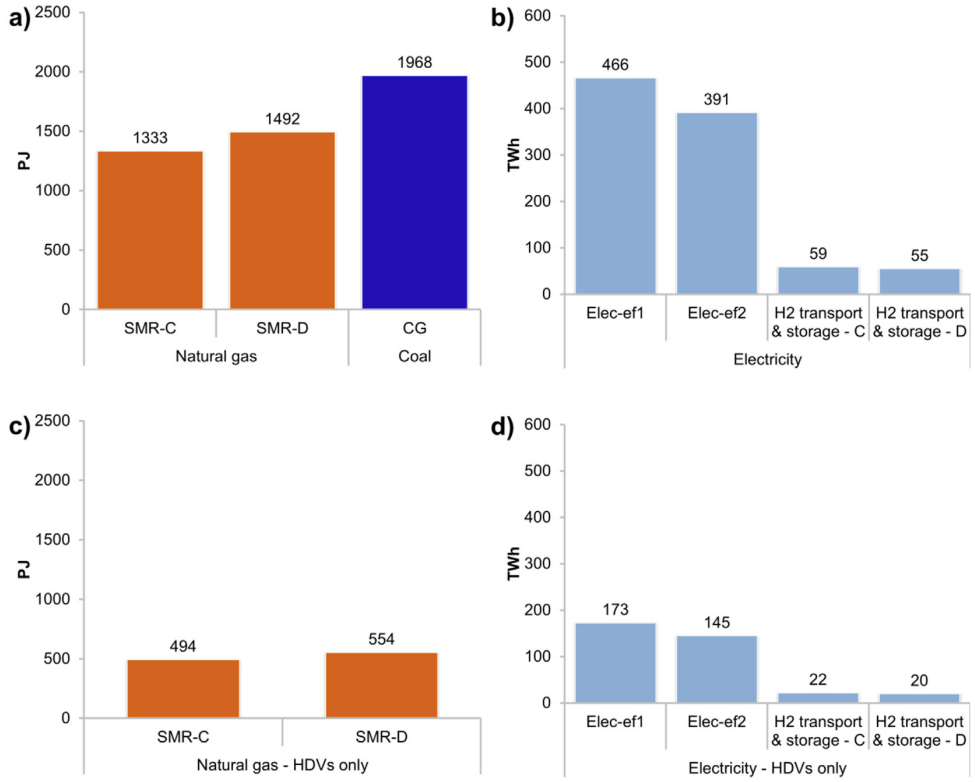

Fig. 4 – (a) Natural gas, coal and (b) electricity demand for hydrogen production to cover all vehicle categories, and (c) natural gas and (d) electricity demand for hydrogen production to cover HDVs only. The electricity requirements for hydrogen transport and storage are likewise displayed in (b) to cover all vehicle categories, and in (d) to cover HDVs only. SMR-D for HDVs was not explored as a scenario, and natural gas demand thereof is presented here for informational purposes only.

for road transport hydrogen (446–525 TWh)—covering hydrogen production, transport and storage—through domestic renewable power alone by means of solar and wind energy.

CO_2eq

The impacts of different scenario assumptions on CO_2eq emissions vary considerably, with German emissions ranging from +52% to −97% compared with the baseline (Table 4). To put these impacts into perspective, this translates to an increase of up to 11% and a decrease of as much as 21% of German country total emissions for the year 2016. All scenarios lead to a 99% decrease in CO_2eq emissions from the 2016 German road transport sector. In particular, the *Elec-renewable* scenario would contribute significantly to Germany getting back on track towards accomplishing its future emissions targets with a national emissions reduction of 179 $MtCO_2$eq. The *SMR*-based scenarios would also bring German emissions closer to this goal (up to −73 $MtCO_2$eq), though substantially less so than the former. On the other hand, the *CG* and *Elec-cmx*-based scenarios put Germany on a path further away from these targets (+50 and up to +95 $MtCO_2$eq, respectively). The range of emission impacts is extensive for the *Elec* scenarios, though narrower for *SMR* scenarios. This indicates that the assumed measures (variables) under the *SMR* set are less effective in their ability to reduce German emissions. Indeed the hydrogen production method is the most important factor of the variables examined in our study in influencing emissions outcomes. The exception to this is the electricity supply: switching from the current mix to renewable power under the *Elec* scenario set has the most significant effect on our results, flipping the highest emissions increase into the highest emissions decrease.

Among the fossil fuel-based hydrogen scenarios, the combustion of fuel in facilitating hydrogen production accounts for the overwhelming share of total CO_2eq emissions, while the contribution from producing fossil fuels (e.g., extraction, processing, and transport) is relatively low (Fig. 5). Accordingly, the *Elec-renewable* scenario set is able to achieve the greatest decrease among all scenarios by avoiding fuel combustion through the use of renewable electricity.

On the other hand, in *Elec* scenarios in which the current electricity mix ('*cmx*') is applied, the result is a significant increase in emissions. This is due to the high CO_2 intensity of the power supply (i.e., fossil fuels make up a robust share of 2016 German electricity generation, especially coal; see the Supplement, Section S1.9), the relatively low thermal efficiency of fossil fuel-powered generation (especially coal), and the fact that energy must be converted twice (i.e., first to generate

Table 4 – Absolute change in total and German CO_2eq emissions from scenarios relative to the baseline for the year 2016, in units of Mt, and percent change in total and German CO_2eq emissions from scenarios relative to the baseline, and percent change in German CO_2eq emissions from scenarios relative to official German country total emissions for the year 2016. Baseline and 2016 German total CO_2eq emissions are displayed at the top of the table, in units of Mt. The data used to calculate the scenario CO_2eq emission values comes from a wide range of sources, and is provided in the Supplement (Section S1).

	Total baseline		DE baseline		DE NIR 2016
$MtCO_2eq$ emissions	196		184		858[b]
Scenario	Δ_{Abs}[a]	$\%_{Baseline}$	Δ_{Abs}	$\%_{Baseline}$	$\%_{DE\_NIR\_2016}$
SMR-ng1-C	−71	−36	−73	−39	−8
SMR-ng1-D	−61	−31	−64	−35	−7
SMR-ng2-C	−62	−32	−72	−39	−8
SMR-ng2-D	−52	−27	−64	−35	−7
Elec-ef1-cmx-C	83	42	95	52	11
Elec-ef1-cmx-D	81	41	93	50	11
Elec-ef2-cmx-C	44	22	56	30	6
Elec-ef2-cmx-D	42	21	54	29	6
Elec-renewable	−191	−97	−179	−97	−21
CG	53	27	50	27	6
SMR-ng1-C_HDV	−16	−8	−18	−10	−2
Elec-renewable_HDV	−61	−31	−57	−31	−7

[a] The change in absolute emissions from the scenario relative to the baseline.
[b] Source: [63]; this value represents the total German NIR CO_2eq emissions for the year 2016 and does not include LULUCF (emissions and removals from land use, land use changes and forestry).

electricity, and then to convert it to hydrogen). The efficiency of the electrolysis process, i.e., electricity consumption, is likewise an important factor: when the efficiency increases from *ef1* to *ef2*, the increase in emissions among *cmx*–based scenarios is cut by about 40%. In contrast, the impact of centralized ('C') and decentralized ('D') hydrogen production on emissions is low under the *Elec* scenario set. This is because the thermal efficiency of electrolysis is assumed to be the same for *C*- and *D*-based scenarios, and they only differ slightly in the amount of electricity consumption required for transporting and storing hydrogen. It is worth mentioning that the *Elec-cmx* scenarios do not include upstream emissions of energy carriers used in electricity generation; if these were included, CO_2eq emissions would be somewhat higher (8% higher total emissions and <1% higher German emissions; see the Supplement, Section S1.9).

All *SMR* scenarios lead to substantial emission decreases, which are afforded by the relatively high thermal efficiency of SMR and the relatively low emissions from fuel combustion of natural gas. It is important to highlight, however, that because the energy source of *SMR* is still a fossil fuel, the emission reduction it achieves is significantly lower than that of *Elec-renewable* (up to −73 vs. −179 $MtCO_2$eq, respectively). It is also worth noting that because *ng1* and *ng2* affect CH_4 leakage, the

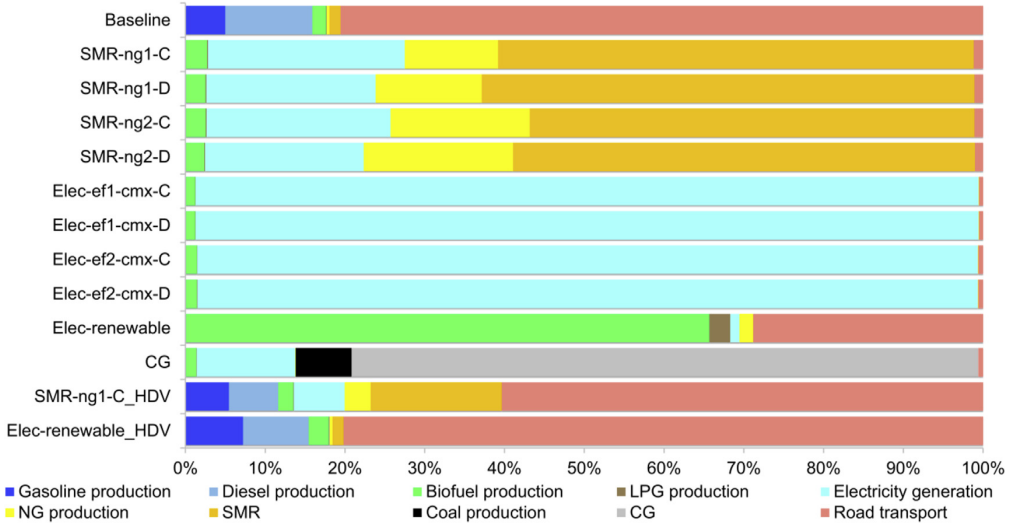

Fig. 5 – Contribution of domains to total CO_2eq emissions, per scenario, in %.

Table 5 – Percent change in 2016 German air pollutant road transport sector emissions from replacing conventional ICEVs, for all vehicle categories or HDVs only, with FCEVs, relative to the baseline.

Conventional ICEVs replaced with FCEVs	Change in German road transport emissions[a]						
	NMVOCs	NO_x	$PM_{2.5}$	PM_{10}	CO	SO_x	NH_3
All vehicle categories	−96%	−95%	−95%	−95%	−94%	−94%	−93%
HDVs only	−4%	−32%	−29%	−29%	−7%	−30%	−2%

[a] Sources [59]: and the *Umweltbundesamt* [UBA] (German Environment Agency) [M. Kotzulla and G. Gohlisch, written communications, 2019 and 2020]; further detail is provided in the Supplement, Section S1.1. Road transport emissions considered in this study include fuel combustion/exhaust emissions (and gasoline evaporation for NMVOCs) only; i.e., PM reduction does not include tire/brake wear and road abrasion emissions.

majority of which occurs upstream, and because the majority of German natural gas is produced outside of Germany, *ng1* and *ng2* have a small effect on German emissions. Considering, however, that the ultimate goal is to reduce global GHG emissions, CH_4 leakage is still an important factor for a hydrogen economy employing SMR.

The *CG* scenario leads to a high increase in CO_2eq emissions, and it is interesting to note that this increase is less than *Elec-ef1* and slightly less than *Elec-ef2* scenarios. This again emphasizes the importance of the electricity supply under the *Elec* set: if the current German mix (namely, the 2016 grid supply which was based heavily on fossil fuels) is employed, the emissions outcome can be more harmful to the climate than directly using coal under *CG*.

While HDVs only make up a small portion of the German vehicle fleet (namely, about one third thereof with respect to conventional fuel consumption), addressing this segment alone can achieve a strong emissions reduction under the *Elec-renewable-HDV* scenario (−57 $MtCO_2$eq); it is also notable that this reduction is only slightly less than the more extreme case of switching all vehicle categories to hydrogen under the *SMR*-based scenarios. On the other hand, the emissions reduction achieved under *SMR-ng1-C_HDV* amounts to about a third of that realized under *Elec-renewable-HDV*.

Air pollutants

Anthropogenic air pollutant emissions explored in this study—NMVOCs, NO_x, $PM_{2.5}$,[2] PM_{10}, CO, SO_x, and NH_3—mainly stem from incomplete fossil fuel combustion, though vaporization is also an important source of NMVOCs. Poor air quality is a serious problem in Europe, being responsible for about 500,000 premature European deaths in 2016 [81], in addition to posing a host of other serious hazards to human health [82,83], the environment [84,85], agriculture [86] and infrastructure [87]. Transport represents a major source of air pollution [3,88], meaning that a potential transition to a hydrogen economy in road transport has important implications for air quality. Since FCEVs have zero emissions at the tailpipe, replacing all gasoline and diesel ICEVs with fuel cell technology would drastically reduce pollutant emissions from the German road transport sector (−93% to −96%), depending on the species, for the year 2016 (Table 5). It is important to note that the remaining pollutant emissions result from use of alternative carbon-based fuels, i.e., CNG, LPG and biofuels. These reductions may also have important implications for secondary aerosol formation and ground-level O_3; e.g., the latter is a harmful air pollutant formed through reaction of NMVOCs and CO with NO_x in the presence of sunlight.

If HDVs are exclusively replaced with hydrogen energy, the emissions decrease for the road transport sector (Table 5) would still be high for most pollutants (approximately −30%), but for NMVOCs, CO and NH_3 the impact is lower (between −2% and −7%) on account of HDVs contributing a smaller share to these emissions in overall road transport. E.g., NMVOCs mainly stem from PCs, two-wheelers, and gasoline evaporation, while HDVs do not use gasoline, and the majority of CO and NH_3 emissions in road transport stem from PCs. Finally, the varying ratio of NMVOCs to NO_x emission reductions among *All vehicle categories* and *HDVs only* in Germany (Table 5) may lead to very different O_3 outcomes, on account of the non-linear relationship between these species in O_3 formation.

Shifting towards a hydrogen economy would not just avoid direct exhaust emissions from road transport, but would also incur emissions from activities related to hydrogen production (and avoid emissions associated with diesel and gasoline production). The impact of scenarios on German pollutant emissions are displayed in Table 6 (total pollutant emissions are provided in the Supplement, Section S3). In order to put the absolute emission numbers into perspective, changes relative to the German energy sector for the year 2016 are also presented in Table 6, and are focused on in the following discussion (caution is advised not to take these values out of context, since the relative changes depend on the reference year). The contributions of domains to scenario total air pollutant emissions are displayed in Fig. 6 (NH_3 is not displayed as its scenario emissions stem almost exclusively from road transport with a negligible fraction from diesel and gasoline production; i.e., the contributions to NH_3 do not differ between the hydrogen scenarios) (see Table 6).

The scenarios generally lead to reductions in German air pollutant emissions compared with 2016 energy sector emissions. *Elec-renewable* achieves the highest emission reduction among all species, followed by *SMR*. *Elec-cmx* reduces emissions for most pollutants, but the impact is low for PM and in fact increases for SO_x. CG achieves many decreases, but experiences increases in PM and a high increase in SO_x. Our results show that variables within scenario sets have a low potential to change pollutant emissions (with the exception of

[2] Only PM emissions from fuel combustion are considered in our study for the road transport sector, i.e., tire/brake wear and road abrasion emissions are not included.

Table 6 – Absolute change in German air pollutant emissions from scenarios relative to the baseline for the year 2016, in units of kt, and percent change in German air pollutant emissions from scenarios relative to German energy sector emissions for the year 2016. 2016 German energy sector air pollutant emissions are displayed at the top of the table, in units of kt. The data used to calculate the scenario air pollutant emission values comes from a wide range of sources, and is provided in the Supplement (Section S1).

	NMVOCs		NO_x		$PM_{2.5}$		PM_{10}		CO		SO_x		NH_3	
DE energy 2016 Abs[a]	255.3		1004.1		65.4		85.8		2037.9		278.7		17.3	
Scenario	Δ_{Abs}[b]	%	Δ_{Abs}	%	Δ_{Abs}	%	Δ_{Abs}	%	Δ_{Abs}	%	Δ_{Abs}	%	Δ_{Abs}	%
SMR-ng1-C	−105.9	−41	−341.4	−34	−6.4	−10	−6.6	−8	−658.1	−32	−14.6	−5	−11.0	−63
SMR-ng1-D	−105.9	−41	−336.7	−34	−6.5	−10	−6.6	−8	−657.4	−32	−15.8	−6	−11.0	−63
SMR-ng2-C	−105.6	−41	−341.4	−34	−6.4	−10	−6.6	−8	−658.1	−32	−14.6	−5	−11.0	−63
SMR-ng2-D	−105.6	−41	−336.7	−34	−6.5	−10	−6.6	−8	−657.4	−32	−15.8	−6	−11.0	−63
Elec-ef1-cmx-C	−98.2	−38	−191.2	−19	−0.2	0	0.1	0	−564.2	−28	120.4	43	−11.0	−63
Elec-ef1-cmx-D	−98.3	−38	−193.0	−19	−0.2	0	0.1	0	−565.1	−28	119.2	43	−11.0	−63
Elec-ef2-cmx-C	−99.5	−39	−224.2	−22	−1.2	−2	−1.0	−1	−581.4	−29	98.7	35	−11.0	−63
Elec-ef2-cmx-D	−99.5	−39	−226.0	−23	−1.3	−2	−1.0	−1	−582.3	−29	97.5	35	−11.0	−63
Elec-renewable	−107.1	−42	−422.3	−42	−7.5	−12	−7.7	−9	−685.0	−34	−31.9	−11	−11.0	−63
CG	−104.6	−41	−197.6	−20	2.2	3	6.1	7	−599.3	−29	345.8	124	−11.0	−63
SMR-ng1-C_HDV	−8.8	−3	−110.5	−11	−1.9	−3	−2.0	−2	−39.3	−2	−3.8	−1	−0.2	−1
Elec-renewable_HDV	−9.3	−4	−140.5	−14	−2.3	−4	−2.4	−3	−49.1	−2	−10.2	−4	−0.2	−1

[a] Source: UBA [M. Kotzulla and G. Gohlisch, written communication, April 15, 2019].
[b] The change in absolute emissions from the scenario relative to the baseline.

Elec-renewable vs. *Elec-cmx*). This is in part due to air pollutant emissions associated with current mix electricity generation being relatively low except for SO_x; accordingly, differences between *C* and *D* hydrogen production and *Elec-1* and *Elec-2* are relatively low. The most substantial relative reductions are seen for NMVOCs, NO_x, CO, and NH_3. The lowest relative reductions are seen for SO_x, PM_{10} and $PM_{2.5}$, with the latter two seeing no change or even a small relative increase in emissions for some scenarios, and the former a significant relative increase in emissions for some scenarios. This is because road transport is responsible for a significant portion of NMVOCs, NO_x, CO, and NH_3 emissions from the German energy system (35% and more), while for $PM_{2.5}$ and PM_{10} (12% and less) and SO_x (<1%) the contribution is lower. Despite the majority of emissions from road transport being avoided by replacing ICEVs for all vehicle categories with FCEVs (Table 5), this sector still represents one of the main sources of pollutant emissions in many of the hydrogen scenarios (Fig. 6) due to alternative road transport fuels (i.e., LPG, CNG and biofuels). Other main sources of pollutant emissions are SMR, CG and electricity generation, i.e., activities related to fossil fuel combustion. Energy production is an important source of total NMVOCs in the hydrogen scenarios (Fig. 6), as these compounds are typically found in association with fossil fuels. Yet German NMVOC emissions incurred from fossil fuel production are low because most fossil fuels are imported (though implying that some NMVOC burden will be carried elsewhere; German emissions allocation is described in the Supplement, Section S1.14); thus *SMR* and *CG* achieve similar NMVOC reductions to *Elec-renewable*. Due to the importance of fuel combustion for emissions of NO_x, CO and PM, the hydrogen production method—electrolysis (i.e., from electricity generation with *cmx*), SMR, and CG—and electricity generation for hydrogen transport and storage, are important sources for these species. Aside from *Elec-renewable*, the *SMR* scenario set achieves the greatest decrease in NO_x CO, and PM due to the higher thermal efficiency of SMR combined with the relatively lower NO_x, CO and PM emissions (approximately 60%, 70% and 100% lower, respectively) from natural gas stationary combustion compared with coal (see the Supplement, Sections S1.4 and S1.8). *Elec-cmx* and *CG* scenarios cause large increases in German SO_x emissions as a result of high SO_x release from current electricity generation and CG. NH_3 emissions in our study only stem from road transport and gasoline/diesel production; therefore, all scenarios experience the same NH_3 emission reductions (Table 6). It is worth noting that the majority of German NH_3 emissions (95% in 2016) stem from agriculture; as such, reducing road transport NH_3 emissions could have an important effect on urban emissions thereof.

Hydrogen emissions

Anthropogenic hydrogen emissions are released from incomplete fuel combustion and leakage throughout the hydrogen utilization chain [89]. Hydrogen functions as an indirect GHG by reacting with and thus reducing the abundance of the hydroxyl radical, the main oxidizing agent in the troposphere, which extends the atmospheric lifetimes of GHGs like CH_4 and hence their climate forcing and leads to O_3 production [43,90–93]. Since hydrogen emissions can influence O_3 concentrations, they can additionally impact air pollution and possibly contribute to depletion of the O_3 layer in the stratosphere (though any negative effects would likely be less than those from the fossil fuels to be replaced) [44]. Since, to the best of our knowledge, there is currently no data on hydrogen loss from commercial FCEVs (Introduction), scenario hydrogen emissions are not quantified in this work. Instead, the maximum allowable hydrogen loss rate from FCEVs that would be required to avoid a net increase in

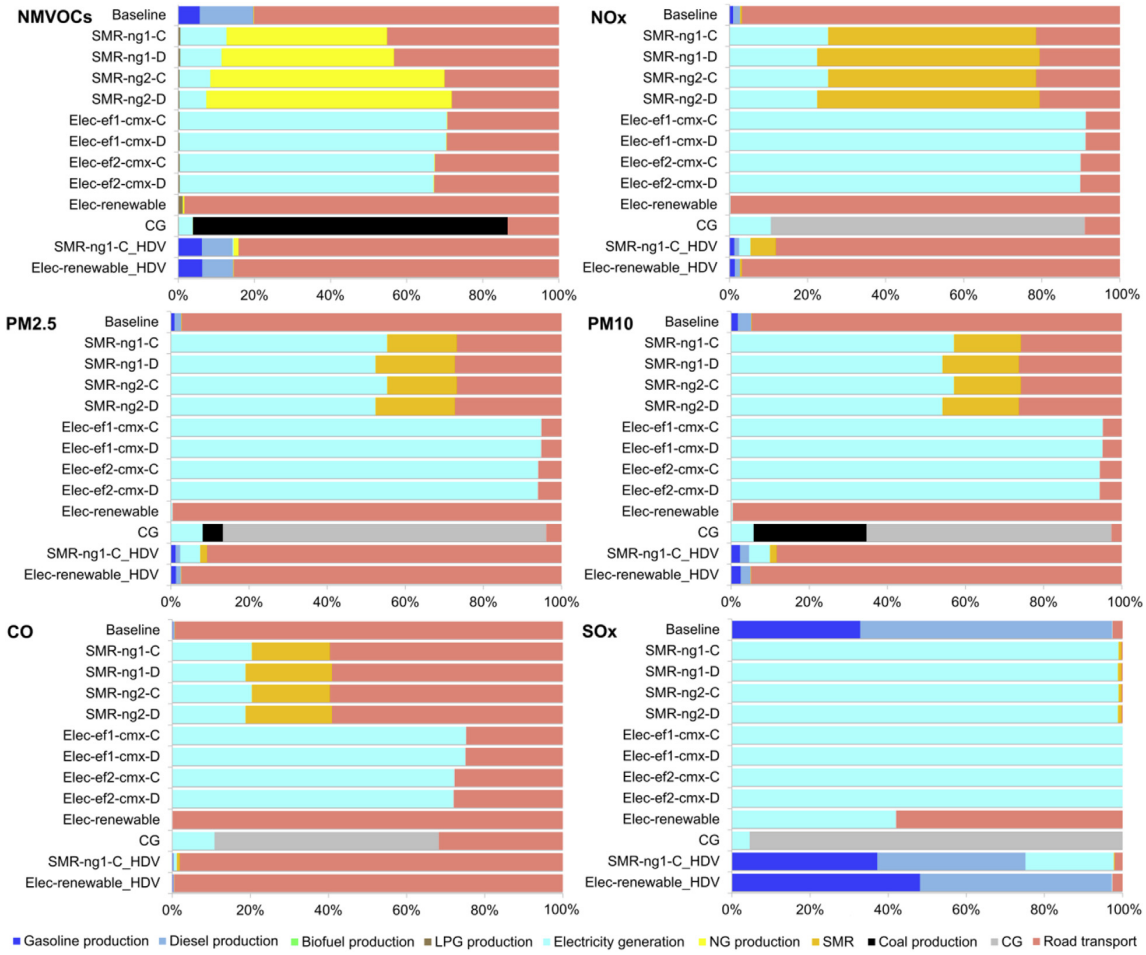

Fig. 6 – Contribution of domains to total air pollutant emissions, per scenario, in %.

German hydrogen emissions is explored here to provide a first estimate; this is done by assessing current hydrogen emissions from German road transport released based on our scenarios, i.e., stemming from incomplete fuel combustion and hydrogen refinery operations for gasoline and diesel (calculation described in the Supplement, Section S1.2). Based on this, we calculate a maximum allowable FCEV hydrogen loss rate of 106 mg/km for Germany when hydrogen fuel is implemented in all vehicle categories.

Conclusions

Our study shows that a deep transition of the German road transport sector from conventional fossil fuels to hydrogen energy can significantly reduce national CO_2eq emissions. However, the outcome depends strongly on the hydrogen production technology, and notably for electrolysis the electricity supply.

The scenario assuming renewable-powered electrolysis—that is, green hydrogen—has the greatest drop in emissions (−179 $MtCO_2$eq), and would contribute significantly towards achieving Germany's future GHG emissions reduction targets. According to our calculations on electricity requirements for hydrogen production (via electrolysis), transport and storage, the level of electricity demand (446–525 TWh) could be fully met through future German renewable power based on the domestic potential of solar and wind energy as estimated in the literature [80]. This highlights a clear potential for the transition of the German vehicle fleet to CO_2-free hydrogen and the opportunity to greatly enhance domestic energy security. Additionally, the green hydrogen scenario facilitates the largest reduction in regulated air pollutant emissions, with a decrease of up to 42% for NMVOCs, NO_x and CO, and up to 12% for PM and SO_x, compared with the German energy sector for the year 2016 (with all scenarios leading to the same decrease in NH_3 of 63%). Such changes will have important implications for

air quality which would be valuable to investigate in a follow-up modeling study.

Steam methane reforming [SMR] also decreases domestic emissions, though to a considerably lesser extent (between −64 and −73 $MtCO_2eq$) than renewable electrolysis on account of its energy source still being a fossil fuel. While combining SMR with CCS—namely, blue hydrogen—can substantially reduce direct emissions of CO_2, it is not emission-free and, due to several issues [29], open questions remain about its feasibility. Given these considerations in conjunction with the fact that green hydrogen is projected to be less expensive than its blue counterpart in the next 5–15 years [40], and that economies of scale are required to bring down costs of green hydrogen, it seems hard to justify diverting limited capital away from renewable-based towards natural gas-based hydrogen, not to mention prolonging the fossil fuel economy and foreign energy dependency. It may at first be worth using (at least in part) the current electricity mix with electrolysis in the interim to build up a green hydrogen economy infrastructure. However, our findings indicate that electrolysis powered by the current electricity supply would lead to the largest increase in national GHG emissions (up to +95 $MtCO_2eq$) along with an increase in SO_x emissions and no effect on PM (and hence no benefit through its reduction). Thus if electrolysis with grid electricity using the current mix were to be employed over a longer term, rather than as a bridge technology, then other measures would need to be implemented by policymakers in order to ensure that longer-term climate objectives can still be met. On the other hand, it is important to note that as the CO_2 intensity of German electricity generation continues to decrease (generally through an increase in renewables and a decrease in fossil fuels), employing electrolysis with future grid electricity (assuming that this current trend continues) would lead to lower emissions than found in our results, for which the 2016 German electricity supply is assumed. Unsurprisingly, coal gasification [CG] leads to a strong increase in domestic GHG emissions (+50 $MtCO_2eq$), along with SO_x and PM, supporting the exclusion of this technology from future German hydrogen production.

We find a hydrogen loss rate of less than 106 mg/km from FCEVs when shifting the German vehicle fleet to this technology is required to avoid a net increase in domestic hydrogen emissions. It would be interesting to explore hydrogen emissions from commercial FCEVs to understand potential changes to the hydrogen budget in a future road transport hydrogen economy.

By only shifting HDVs to green hydrogen, a deep cut in emissions can already be achieved (−57 $MtCO_2eq$), which is only slightly less than all vehicle categories being replaced with SMR-based hydrogen. We also find that the burden of hydrogen demand to fuel the vehicle fleet would be nearly two thirds less if limited to HDVs (from 1000 to 371 PJ). Accordingly, HDVs represent a low-hanging fruit for FCEVs on the path to road transport decarbonization; this is notable considering that the competing technology, i.e., the BEV, has major challenges with heavy load, long-range, and short recharging requirements associated with this vehicle segment.

It is important to note that some uncertainty is associated with the estimations of scenario results presented here due to lack of data on FCEV TTW efficiency, especially for HDVs, LDVs and two-wheelers. Therefore further research on this parameter would facilitate more robust estimates for vehicular hydrogen demand and hence emissions. Additionally, it is worth emphasizing that the scenarios explored here are illustrative, considering extremes. Namely, the entire conventionally-fueled German vehicle fleet is assumed to be replaced by hydrogen that is produced by a particular method. In reality, the future vehicle fleet will likely be diverse with FCEVs complemented by other technologies, and hydrogen may be produced by a combination of methods and may also be imported. Based on the results here and as the direction of Germany's hydrogen plans unfold, it would be valuable to perform follow-up studies based on more realistic scenarios, looking at the benefits and trade-offs from each case.

The exploratory nature of this analysis and the type of data that is available would make an extensive statistical (uncertainty) analysis not very meaningful, and could actually be rather misleading, since applying standard statistical techniques on the limited data could lead to the misimpression that the uncertainties are much smaller than they are in reality. This would, however, be an important aspect for future development, if a shift towards expanded hydrogen usage in the transport sector is being more seriously considered politically, so that bounds on the anticipated impacts can be estimated based on the uncertainty in the estimates of the most important parameters.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijhydene.2020.11.014.

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5 Synthesis

5.1 Discussion of the main findings

Climate change, air pollution and energy use are profoundly connected. To this end, emission scenarios and air quality modeling can play an important role in assessing environmental impacts from possible, future energy system changes to inform policy. This thesis contributes to current knowledge on relevant aspects of the energy transition in Europe. In particular, the potential of future shale gas development and hydrogen energy in Europe have been the subject of great interest and debate over the span of the past decade. In the following, the overarching question and research questions as outlined in the Introduction are presented, and subsequently addressed by discussing the main findings of this thesis:

Overarching Question: What are the anticipated environmental impacts of shale gas and hydrogen as possible transition or long-term energy sources in Germany and the United Kingdom?

> *RQ1: What are the anticipated greenhouse gas and air pollutant emission impacts of shale gas in Germany and the United Kingdom and hydrogen in Germany?*
>
> *RQ2: How would shale gas emissions in Germany and the United Kingdom impact ozone air quality on the local and regional scale?*

5.1.1 What are the anticipated greenhouse gas and air pollutant emission impacts of shale gas in Germany and the United Kingdom and hydrogen in Germany?

RQ1 is addressed by Papers I and III (Chapters 2 and 3, respectively) through the construction and application of comprehensive emission scenarios. While scenarios can be a valuable tool in providing insights on possible, future energy system changes, an important critique is their lack of transparency (Quarton et al., 2020; Rosen, 2016). This is especially troubling when considering that they can be influential to political discourse and policy decisions. Papers I and III seriously address this issue by providing a thorough documentation of assumptions, all input data and calculations for the reader to understand the results and retrace the scenario development. The findings of both studies are discussed in the following sub-sections.

Paper I: Emission impacts from shale gas in Germany and the United Kingdom

There is a commonly held perception among the public and other stakeholders, especially those in Europe, that shale gas emissions are considerably higher than those stemming from conventional gas production (discussed in Chapter 2). For example, US shale gas leakage has been found in some cases to reach or even exceed 10 %, while official data for Germany and the UK indicates conventional gas leakage rates below 0.1 % (as discussed therein). Consequently, this has led in part to opposition against shale resource development in Europe. Yet, research on the subject (i.e., future European shale gas impacts) has hitherto been scarce. Thus Paper I contributes valuably to understanding the range of emission effects from a possible future European shale gas industry, and moreover it reveals opportunities for mitigation of potentially dangerous impacts, which in turn can be used to inform policy. Due to the heightened importance of CH_4 leakage to shale gas emissions (as discussed therein), it is of focus in the following discussion, while a brief overview of the contributions to CO_2 and air pollutants is also given.

The results reveal that overall CH_4 leakage rates from upstream shale gas development as a percentage of total gas production would range between 0.35 % and 1.36 % for a realistic case ('REm') and between 0.08 % and 0.15 % for an optimistic case ('OEm')[8]. *Based on the large difference in leakage between the two cases, the results of this thesis demonstrate the value and potential of abatement measures employed in OEm to substantially reduce emissions. At the same time, while the practices and technologies applied in OEm are technically feasible, they are unlikely to be accomplished on a systematic, regular basis on account of economic factors and limited human resources for monitoring.* The most consequential sources for CH_4 emissions within the gas production chain in REm are gathering, production, and gas processing. Various measures were found to effectively mitigate these emissions in OEm, such as reduced number of gathering facilities and electrification of devices, e.g., compressors. Indeed, several papers have indicated the important role of gathering facilities and production sites to CH_4 emissions (Balcombe, Anderson, Speirs, Brandon, & Hawkes, 2017; Littlefield, Marriott, Schivley, & Skone, 2017; Zavala-Araiza et al., 2015). On the other hand, the results indicate that REm and OEm emissions associated with activities required for shale gas only (i.e., well completion and fracking) would only make a minor contribution to total upstream CH_4 emissions. This is due to tighter regulation assumed in the scenarios, particularly Reduced Emission Completions which have been mandatory in the US since January 2015. *Hence, the findings suggest that the difference in CH_4 leakage between European shale gas production and conventional gas production would be insignificant.* The results also demonstrate that low well productivity (an effect especially seen in Germany) is an important factor leading to significantly increased CH_4 emissions, on account of the overall population of active

[8]The realistic emission scenario category ('REm') assumes business-as-usual activities common in the US and Europe, i.e., it is the realistic case that would be expected for shale gas development in Germany and the UK. The optimistic emission scenario category ('OEm') assumes emission reduction technologies and full compliance with a strict regulatory framework for best practices along the supply chain.

wells being increased; however, this factor cannot be predicted, nor can it be controlled by regulation.

REm results are within the range of upstream CH_4 leakage rates estimated in regional and nationwide studies across the globe, and somewhat lower than many estimates in the US (0.9 % to 1.9 %; discussed and referenced therein). The slightly conservative nature of Paper I results may be explained by the bottom-up approach used in the study, which may potentially result in some relevant emissions sources being excluded. On the other hand, *upstream conventional gas leakage rates based on official data for Germany and the UK (*0.02 % *and* 0.08 %*, respectively) (UNFCCC, 2017) more closely resemble optimistic values for OEm, which, as noted above, are unlikely to be achieved on a systematic, regular basis.* Yet transparent reports on CH_4 leakage from European gas production unfortunately do not exist or are not available to the public, moreover, the numbers are not confirmed by independent studies, raising questions over the accuracy and objectivity of official emissions data (discussed therein). Indeed, the discrepancy between official estimates for Germany and the UK and values reported in the US warrants further investigation.

Overall CO_2 emissions range from 4.8–8.9 Mt in REm to 2.8–4.5 Mt in OEm (including emissions from both countries), with the greatest sources along the shale gas production chain being gathering and gas processing. In terms of CO_2-eq emissions, the findings of this thesis reveal that CH_4 reduction measures are the most effective for emissions mitigation compared with CO_2.

The results show that the main sources of VOC emissions are the same as for CH_4, i.e., production, gathering and processing, since these species are co-emitted. *Compared with national energy sector emissions, shale gas VOCs emissions could be significant, being equivalent to* 4.5 % *to* 46.2 % *for Germany and* 8.3 % *to* 66.5 % *for the UK. Similar to* CH_4*, the large difference in VOC leakage shows the effectiveness of emission reduction measures.* Additionally, speciation of natural gas is an important factor to VOC emissions (i.e., percentage of VOCs present in gas), though this can neither be predicted nor controlled. On the other hand, emissions of the other pollutants (NO_x, CO, and PM) were found to be relatively insignificant compared to national emissions.

Paper III: Emission impacts from hydrogen in Germany

Hydrogen is highly relevant to current energy transition discussions, with enthusiasm gaining an unprecedented level of momentum around the globe in recent years due to the urgency to reduce emissions and hydrogen's ability to deliver zero emissions and hence foster a decarbonized energy system (IEA, 2019a). Moreover, Europe, and especially Germany, have expressed strong interest of late in global leadership on hydrogen technologies (BMWi, 2020; European Comission, 2020a). One of the promising applications for hydrogen is the mobility sector. By examining the range of emission effects from a complete shift in German road transport from conventionally-fueled to hydrogen-

powered vehicles, the analysis in Chapter 4 contributes to advancement of knowledge in this field, which can in turn inform research investment and policy.

The study reveals that if the hydrogen were to be produced by renewable-powered electrolysis (i.e., green hydrogen), this would result in a substantial contribution towards Germany meeting its future GHG emissions reduction targets and air quality goals. Specifically, it would facilitate an annual German emissions reduction of 179 $MtCO_2eq$ (the greatest drop among all scenarios), which, to put into perspective, represents a 21 % abatement of total German CO_2eq emissions for the year 2016. The findings show that the annual electricity demand for hydrogen production (via electrolysis), transport and storage, would range between 446 TWh and 525 TWh[9], which, according to published estimates on Germany's potential of solar and wind energy (Ruiz et al., 2019), could be met through future domestic renewable power alone. Thus the results reveal the opportunity to deeply strengthen domestic energy security with green hydrogen mobility. Green hydrogen also would result in the greatest emissions reduction of regulated air pollutants among all scenarios: up to 42 % emissions reduction for NMVOCs, NO_x and CO, and up to 12 % for PM and SO_x, compared with the German energy sector for the year 2016. On the other hand, all hydrogen production methods would lead to a 63 % reduction in NH_3 emissions compared with the 2016 German energy sector. Notably, the largest source of total German NH_3 emissions is agriculture (95 % in 2016), whereas the NH_3 emissions in this study stem almost exclusively from road transport. Hence, the results indicate a strong drop in road transport NH_3 emissions, which may have important implications for urban emissions of this species.

Hydrogen produced via steam methane reforming (SMR) would result in an annual domestic GHG emissions reduction between 64 and 73 $MtCO_2eq$, substantially less than green hydrogen due to SMR being facilitated by natural gas. Notably, hydrogen produced by electrolysis powered by the current electricity supply would result in the greatest domestic GHG emissions increase (up to +95 $MtCO_2eq$), and moreover, an increase in SO_x emissions and no effect on PM (i.e., no benefit). Accordingly, the findings of this thesis suggest that *if the future strategy were to deploy electrolysis facilitated (at least in part) with the current electricity mix until enough renewable energy would be available, it will be valuable to thoroughly examine the overall GHG emissions impact to ensure that such a strategy does not negate longer-term climate objectives.* Additionally, the analysis shows that hydrogen produced by coal gasification (CG) would result in +50 $MtCO_2eq$ annual domestic GHG emissions and an increase in SO_x and PM (it is worth noting that CG is not a realistic case in Germany on account of the high emissions and pollution intensity and considering that coal is being phased out there, and rather is included here for comparison).

On the other hand, if only HDVs were to be shifted to green hydrogen, annual German emissions would still decrease by a substantial 57 $MtCO_2eq$, and the findings show that the burden of vehicular hydrogen demand would drop by almost two thirds (from 1000 PJ

[9]This value would be in addition to the renewable power already produced in Germany.

for powering the entire German vehicle fleet, to 371 PJ for HDVs only). Hence the results indicate that *the HDV segment is a low-hanging fruit on the road to decarbonization of the German road transport sector with hydrogen, which is a particularly noteworthy finding when taking into account that the HDV fleet segment is especially challenging to electrify with BEVs (as discussed in Chapter 4).*

Hydrogen also represents an indirect GHG, with an estimated GWP of 5 over a 100-year time frame (Derwent et al., 2020), due to its role in the atmosphere of reducing OH concentrations, which in turn especially extends the lifetime of CH_4 and has other effects on climate-forcing gases and particles. Because no data exists to the best of the author's knowledge on hydrogen emissions from vehicles, this thesis provides a first estimate on the maximum allowable hydrogen loss rate from FCEVs in Germany. According to the results of this thesis, a hydrogen loss rate of less than 106 mg/km is required to avoid a net increase in German hydrogen emissions when transitioning the German vehicle fleet to fuel cell technology.

5.1.2 How would shale gas emissions in Germany and the United Kingdom impact ozone air quality on the local and regional scale?

RQ2 is addressed by Paper II (Chapter 3). Paper II is a follow-up study to Paper I, implementing the shale gas emissions output from that study in the air quality model WRF-Chem. To the author's knowledge, no other studies have been published on regional air quality risks from a future shale gas industry in Europe, aside from Archibald et al. 2018 (Archibald, Ordóñez, Brent, & Williams, 2018), who exclusively assessed impacts for the UK. In this way, Paper II contributes uniquely to the literature, while also providing valuable insight to public and political discourse on this potential industry in Europe.

The study finds that when shale gas VOC leakage is at its lowest, ΔMDA8[10] is confined to small areas of the domain around the shale gas basin regions and generally ranges between $2\,\mu g\,m^{-3}$ and $4\,\mu g\,m^{-3}$. When VOC leakage is highest, ΔMDA8 values above $2\,\mu g\,m^{-3}$ cover a much greater area of the domain due to long-range transport. Moreover, the results show the largest peak in ΔMDA8 ($28.3\,\mu g\,m^{-3}$) when VOC emissions are highest, consistent with Archibald et al. 2018's study (Archibald et al., 2018). Interestingly, the results demonstrate that concentrated NO_x emissions have an effect on increasing ΔMDA8 in terms of magnitude and extent when VOC leakage is at its lowest and minimal effect when VOC emissions are at their highest, indicating a mixed regime of sensitivity to both NO_x and VOCs.

The results reveal that shale gas activities in Europe have the potential to result in a large number of additional exceedances of MDA8 on the local and regional scale. Specifically, the total exceedance count is found to range between 103 and 427 (above the WHO threshold), with up to one third of measurement stations in France and the UK, up to circa one

[10] ΔMDA8 is defined in Paper II as the maximum difference in daily MDA8 between the scenario and base case, over the entire simulation period for each grid cell.

fifth of the stations in Germany, and a substantial percentage of stations in neighboring and distant countries found to have additional exceedances. *Exceedances were found to increase significantly with increasing VOC leakage.* Namely, the lowest exceedance count was associated with the lowest VOC emissions and vice-versa. However, exceedances were generally found to be spread out among stations, with the mean value[11] of exceedances per station being 1. Thus the results suggest that, even with extreme VOC leakage, the effect on any particular station (or area) is low in terms of exceedances.

Additionally it was found that the impact of shale gas emissions on SOMO35 (an indicator of health impacts) could be substantial. Interestingly, when VOC emissions are low, there is a notable percent decrease in SOMO35 over the English basins on account of NO_x titration; nevertheless, this is not expected to meaningfully improve health, considering that SOMO35 values are already low in the UK. In any case, as VOC emissions increase, the percent increase in SOMO35 grows in magnitude and extent over the UK, Northern Germany and the surrounding area. The maximum increase in SOMO35 is about 28 %, which occurs when VOC emissions are highest. *Overall, the results demonstrate the key role of VOCs in O_3 enhancement and that accumulated exposure (SOMO35) effects are greatest in the areas of shale gas operations and close surroundings.*

5.2 Conclusions and outlook

This thesis aimed to provide valuable insights and fill in critical knowledge gaps on anticipated environmental impacts of two highly relevant energy developments in the context of the energy transition in Europe: the potential expansion of shale gas and hydrogen. It is envisaged that this work will enrich stakeholder dialogue and inform future research projects and policy making. Based on the results of this thesis, the following conclusions can be made:

Paper I estimates CH_4 leakage rates from a possible, future shale gas industry in Europe, based on realistic developments in Germany and the UK. The results show that activities specifically related to shale gas (as opposed to conventional gas) only make a small additional contribution to overall CH_4 emissions within the natural gas production chain when applying current (US) mandated practices, suggesting that European shale gas would not necessarily lead to notably worse emissions outcomes compared with conventional gas production (noting that other distinct environmental impacts are still possible). However, an important finding of this study is that realistic CH_4 leakage rates from gas production as estimated herein—while similar to values reported by studies carried out in the US and elsewhere—are much higher than official CH_4 leakage estimates from gas production for Germany and the UK based on their national inventories. In fact, official estimates of these countries are similar to the optimistic CH_4 leakage rates of this study. In this way, the results of this thesis raise further questions on the relatively conservative nature of European CH_4 leakage estimates and the discrepancy between European values and those

[11] This excludes stations that exhibit zero exceedances.

of the US, which has been noted elsewhere. Thus, this work further emphasizes the need for more research efforts in order to better understand this issue. The results of this study clearly demonstrate the potential of regulatory measures to significantly mitigate European shale gas emissions, especially for CH_4 and VOCs. However, it must be emphasized that the emission reduction measures in this study are unlikely to be applied and achieved on a systematic, regular basis. Nevertheless, if European shale gas were to come into existence, the results of this thesis suggest that political actors should establish a stringent regulatory framework with full compliance to minimize emissions and other environmental impacts. It is worth noting that in order to meet the Paris Agreement 1.5 °C target, nearly all existing fossil fuel projects (including natural gas extraction and use) would need to be curtailed in the next decade or two, rather than the development of alternate ones such as replacing coal with shale gas (IPCC, 2018). Finally, emission and air quality impacts aside, other harmful impacts to humans and the environment may still render European shale gas development unacceptable, e.g., the UK set a moratorium on shale gas activities on account of seismicity hazards.

Paper II assesses the impact of European shale gas emissions on O_3 air quality on the local and regional scale. The model simulation results suggest that future shale gas activities in Germany and the UK have the potential to significantly worsen European air quality locally and in distant countries through enhanced O_3 formation. This is considerable given that the region already suffers from O_3 health issues (Bell, Zanobetti, & Dominici, 2014), not to mention that it has an ageing population which is more susceptible to O_3-related health effects (Amann et al., 2008). Overall, the findings reveal that shale gas VOC emissions are the most important factor in O_3 formation, while concentrated NO_x emissions play a minor role. At the same time, it must be emphasized that the results here depend on scenario assumptions of an industry that does not yet exist in Europe. Moreover, some of the assumptions (VOC emissions) are rather extreme and may be unlikely to occur together in reality—though high VOC emissions are still possible by enhancing just one of the assumptions, e.g., increased shale gas production. Nevertheless, the results of this thesis suggest that enacting emissions control strategies, especially for VOCs, will be important to reduce potentially harmful impacts if this industry were to come into existence in Europe. The focus of the present study is on the June, July, August period, a time when meteorological conditions such as high temperature, strong solar radiation, and low horizontal and vertical dispersion favors O_3 production and ultimately leads to high concentrations of ground-level O_3. In order to gain a more comprehensive picture, future work is needed to extend the simulations to the other seasonal periods as well as to a different year to explore the O_3 impacts under differing meteorological and chemical conditions. Moreover, it would be valuable for future work to examine the impact of shale gas VOC and NO_x emissions on PM air quality, and to include PM emissions from shale gas development in Paper I as well. To better understand the health-related implications of these results, future studies could use the modeled shale gas air pollution impacts as input to assess the effect on premature mortality and associated cost estimates.

Paper III examines possible GHG and air pollutant emission impacts from a widespread shift to hydrogen-powered traffic in Germany. The results illustrate that a shift of the German road transport sector to hydrogen fuel cell technology via green hydrogen (i.e., renewable-powered water electrolysis) would deeply curtail German emissions of GHGs and regulated air pollutants, thus advancing the country's climate and air quality goals. On the other hand, the findings show that hydrogen production via electrolysis powered by the current electricity mix would lead to a substantial increase in emissions of GHGs and of the air pollutant SO_x. Thus the results of this thesis indicate that if policy were to opt for large-scale electrolysis—based at least partially on the current electricity mix until demand could be fully met with renewable generation—actors should thoroughly assess the anticipated net impact on GHG emissions to confirm alignment with longer-term climate goals. Another important finding of this study is that HDVs are a low-hanging fruit for decarbonization of German road transport through hydrogen energy. Specifically, this work reveals that by only shifting the HDV vehicle segment to green hydrogen, the burden of vehicular hydrogen demand would drop substantially and a deep reduction in GHG emissions would still be realized. This is notable considering that HDVs pose especial challenges for electrification via the competing technology, i.e., BEVs. Based on this finding, follow-up studies and political discourse should further assess the application of hydrogen for this vehicle segment. This work also provided a first estimate of the maximum allowable hydrogen loss rate from FCEVs. It would be valuable for future studies to explore this rate further for development of guidelines on FCEVs that aim to at least avoid a net increase in hydrogen emissions, on account of hydrogen's role as an indirect GHG.

It is important to note that the scenarios of Paper III are illustrative and consider extremes rather than a realistic implementation of hydrogen in German road transport. Specifically, all conventionally-fueled German vehicles are replaced by hydrogen fuel cell technology in the scenario, in which changes are enacted immediately, and where hydrogen is produced by a single method rather than a combination of methods. Further research is therefore needed on more realistic scenarios, especially as the potentially significant future role of hydrogen in German mobility continues to evolve. Moreover, it would be valuable to perform a life cycle analysis and to consider socio-economic aspects to provide a more comprehensive picture. Considering the inclusion, albeit minor, of blue hydrogen in Germany's national hydrogen strategy, it would be valuable to include this production method in future scenario work as well. Of course, it is important to emphasize that blue hydrogen is certainly not emissions-free, and on account of various issues (IRENA, 2019), open questions remain about the feasibility of this technology. To better understand the implications of these results on air quality, future work should investigate the air pollutant changes in a regional modeling study. Finally, it is worth mentioning that there is some uncertainty in the results of Paper III from lack of data on FCEV TTW (tank-to-wheel) efficiency. Thus, future research is needed to establish better data on this parameter, especially for HDVs, considering their potentially valuable role in hydrogen mobility.

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Appendix A: Supplement Paper I

Supplemental material

Emission scenarios on a potential shale gas industry in Germany and the United Kingdom

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1 Shale Gas Drilling Projections

1.1 Background information

The political discussion on a potential shale gas industry in Germany has been centered on the convenience of maintaining the domestic natural gas sector and know-how expertise in the country, as well as preserving a local production share of the domestic gas consumed. This is because natural gas production in Germany has been on the decline since the beginning of this century, decreasing from a previously stable production level of about 20 bcm (billion cubic meters) per year. For example, in 2016, roughly 7.2 bcm of natural gas was produced in Germany, which amounts to about 7% of its total consumption of natural gas (BVEG, 2018). Gas production saw an especially steep decline in the last five years mainly due to the majority of drilling projects being put on hold as a result of a revision of regulations placed on conventional fracking. In the UK, after experiencing a strong decline starting in 2000[1], offshore gas production has been stable at ~35 to 40 bcm per year, reaching 39.6 bcm in 2015, 58% of the total annual gas consumption of 68.1 bcm for that year (BP, 2017). Based on the historical data, we find it appropriate to develop projections targeting a shale gas output volume of about 10 bcm a year in Germany, and about 35 bcm in the UK.

In European countries where shale gas activity may take place in the future, it is plausible to expect engineering technologies capable to minimize the environmental footprint, similar or tighter than current ones. Therefore, these considerations are factored into our drilling and well specifications discussed here. In a 2012 report from the European Commission (Pearson et al., 2012), it was assumed that in the coming years a range between 15 and 36 wells per pad could be expected, each extending between 3,000 and 7,000 m horizontally. Based on this, we assume a value of 30 wells to be built per pad in our projections, with two groups of 15 wells running towards opposite horizontal directions and each extending for 2,500 m and in line with that reported by Acatech (2015), (Figure 2). This configuration can be achieved with a horizontal well-spacing of about 330 m, compatible with normal procedures performed in the US, as well as environmental standards (Díaz de Sousa et al., 2012; Harpel et al., 2012; Browning et al, 2013). The shale gas reservoir is reached from each well pad by three or ten vertical wells, according to the drilling settings choose in the emission scenarios. We assume that the vertical wells are drilled close to each other with a minimum distance of about 3 m, in accordance with industry practice to save space and reduce the environmental impact (DeMong and King, 2011).

1.2 Construction

The construction of the drilling projections is organized on a six-month (i.e., semester) basis. In the first semester, we assume that 100 wells in Germany and 140 in the UK start producing shale gas at a rate described by the production curve assigned to each basin. In the same semester, the same number of wells is under construction, and will constitute a new population that will enter the production phase in the following six-month period. In the second semester, two populations of producing wells determine the overall volume of gas produced, one at its first and the other at its

1 Available at: https://www.eia.gov/beta/international/analysis.cfm?iso=GBR

second semester of activity. In parallel, a new set of wells is under drilling, entering the production phase in semester three. This pattern of well evolution proceeds until the targeted annual production is reached (i.e., circa 10 bcm for Germany and circa 35 bcm fir the UK). At this point, we assume a new drilling rate in each country capable to maintain overall production constant (steady-state production). The average value of the drilling rates necessary to maintain production constant for the following three years is fed into the emission scenarios. For a given shale gas basin, a population of producing wells is defined as a cluster of wells at the same stage of production (i.e., same age).

The overall gas output is estimated as follows:

- To estimate the gas output for each population of producing wells in each basin, we refer to the production curves as explained in the main text;
- To reduce complexity, we assume that all the wells drilled over a semester enters production on the very first day of the following semester;
- The gas production rate changes over time, and consequently on a day-to-day basis. To reduce complexity in calculating monthly gas production, we applied gas production at day 15, which represents the median value for the month, to all the days of the month. The overall semester gas output is predicted by aggregating the gas produced over each month.

The volume of gas produced (*Vp*) by a given population of *producing wells* is calculated as follows:

$$V_p = \sum_{i=1}^{6} P_{day15_{(i)}} \; x\; 30$$

where:
P_{day15}= gas production at day 15 based on the production curve;
i= month (6 for each semester);
30= days of the month (average).

The national gas output (NG_o) for each country is estimated as follows:

$$NG_o = \sum_{b=1}^{n} \sum_{s=1}^{m} Vp_{(s)(b)}$$

where:
$Vp_{(s)}$ = volume of gas produced at semester (s);
m= age of the shale gas industry;
b= shale gas basin (b=5 for Germany and 1 for the UK).

1.3 Mishaps and adjustments

By developing the construction of the drilling projections on a semester-basis, it is not feasible to reach the same annual gas production in each well productivity case (see Figure S1). Due to the direct correlation between total gas produced and related emissions, a comparison of such drilling projections as originally calculated would be erroneous. To fix this incongruence, we operate in the following way. For each country, we select the productivity case that generates an annual gas

production closest to the targeted annual amount (see Section 1.1) as the reference case. All the parameters characterizing the reference case (i.e., total gas output, *wells under production* and *producing wells*, see the Methodology Section in the main text) are left unchanged. Differently, in order to obtain the same volume of gas as the reference case at the end of the supply chain for the other productivity cases, we linearly normalize their parameters based on their annual gas production offsets to the reference case. This operation is carried out for both countries. Such linear correction did not affect the qualitative significance of our emission results, and is required lest differing volumes of gas produced were largely responsible for emission discrepancies in the results (e.g., total CO_2 emissions are strongly correlated to the total methane flowing through the supply chain). Following the same approach, the gas combusted during the processing of the gas is also factored in to ensure the same CH_4 output at the end of the gas chain. Therefore, we add the amount of gas consumed at the processing stage at the beginning of the supply chain. In this way, we obtain a higher total amount of gas extracted by the producing well populations which accounts for the gas combusted during processing.

1.4 Results

In Table S1 we present the TRR_{basin} and EUR_{well} applied to the drilling scenarios. Our results are in line with EUR values of other shale gas basins in the US. For instance, the *Unterkarbon* P50 EUR_{well} of about 200 mcm (million cubic meters) is comparable to data from the Marcellus Shale play (WEO, 2015).

Table S1. TRR and EUR_{well} for all basins and productivity case considered in this study. TRR data are from BGR (2016) and BGS (2013).

	Productivity case	Unterkarbon	Mittelrhät	Posidonia Schiefer	Wealden	Fischschiefer	Bowland Basin
TRR_{basin} (bcm)	**P25**	220	30	270	30	0	2870
	P50	320	50	390	40	1	3760
	P75	480	70	570	60	2	5450
EUR_{well} (mcm)	**P25**	139	21	40	112	0	174
	P50	203	36	58	149	1	228
	P75	304	50	85	224	2	330

In Figure S1 we report results from the drilling projections for each country. The evolution of three parameters is shown: i) the drilling rates (number of wells drilled each semester, see main text, Methodology Section), ii) the total number of producing wells, and iii) the gas output for both countries under different well productivity scenarios. The total volume of gas produced is the amount as originally obtained by the drilling projections.

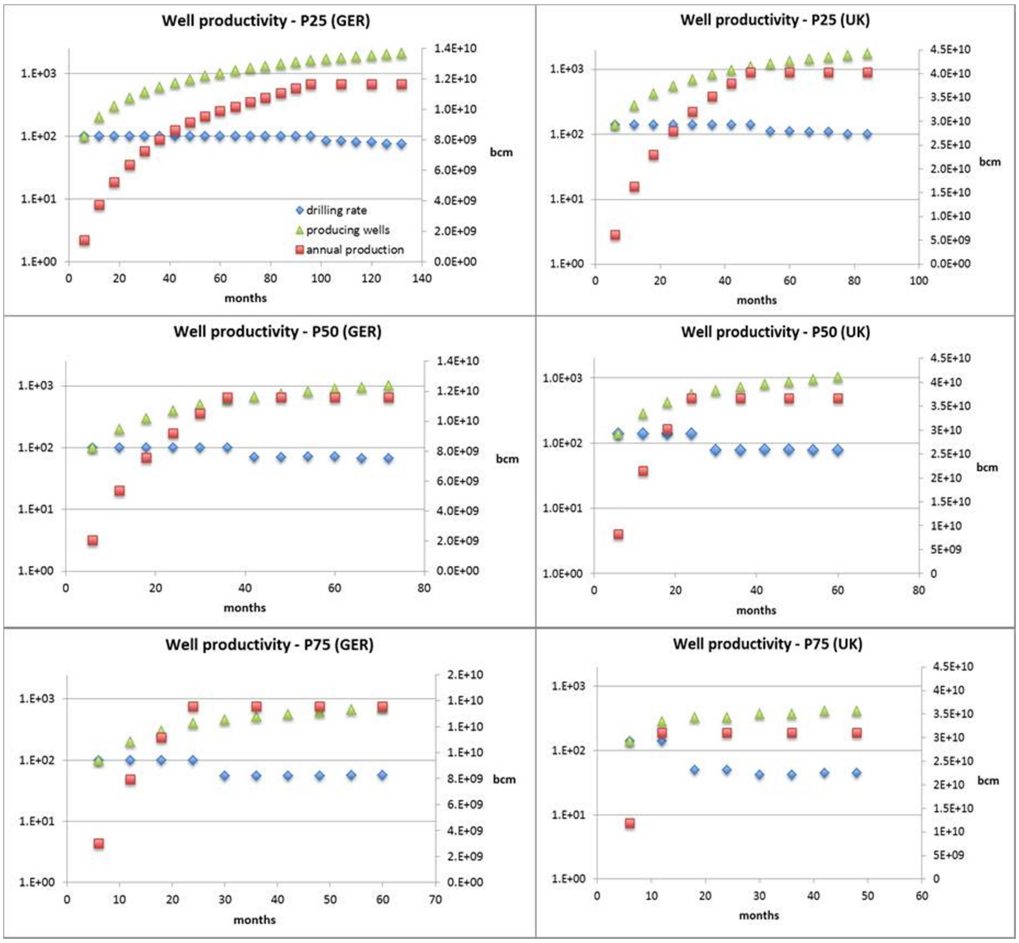

Figure S1. Drilling rate, producing wells and annual gas production for each scenario. The three productivity-cases (P25, P50 and P75) are shown for Germany (left) and the UK (right). For the drilling rate and number of producing wells refer to the left-hand y-axis on each figure.

2 Emission Scenarios

2.1 Gas composition

In the emission scenarios we consider two gas compositions defined as dry and wet compositions (low and high content of VOCs, respectively) representing the upper and lower boundaries of the shale gas compositional range. Concentrations are taken by data reported by Faramawy et al., 2016 (see Table S2). We also assume that no CO_2 is present in the raw wet gas although Faramawy et al. report a medium CO_2 concentration for the wet gas below 5%. This choice does not significantly affect our results since the volume of CO_2 emitted by all machineries and natural gas combustion along the production chain is by far higher than that lost from gas along the supply chain.

The emission scenarios exploring wet vs. dry gas production cases differ from each other only by the concentration of VOCs in the raw gas, respectively 15.4 and 4.0% v v^{-1} (see Table S2). The drilling scenarios are built based on the CH_4 content in the gas, and not total raw gas production. This means that the well parameters defined by the drilling projections represent the amount of wells necessary to produce the desired amount of CH_4 for each country, with the volume of other compounds like VOCs or CO_2 to be added to assess the total volumes of raw gas. In other words, wells active in the wet- and dry-gas cases extract the same amount of CH_4, but produce different volumes of other pollutants according to the composition of the remaining fraction of the raw gas.

Table S2. Gas composition in the raw and dry natural gas. Data from Faramawy et al. (2016).

Pollutant	Formula	Wet raw natural gas (% Vol)	Dry raw natural gas (% Vol)
Methane	CH_4	84.6%	96.0%
Carbon dioxide	CO_2	0%	0%
Ethane	C_2H_6	6.4%	2.0%
Propane	C_3H_8	5.3%	0.6%
Butane	C_4H_{10}	1.4%	0.1%
Pentane	C_5H_{12}	0.2%	0.1%
Hexane	C_6H_{14}	0.4%	0.1%
Heptane	C_7H_{16}	0.1%	0.8%
Isobutane	C_4H_{10}	1.2%	0.2%
Isopentane	C_5H_{12}	0.4%	0.1%

2.2 Well pad construction

We assume a well pad area of 5 acres (about 2 hectares) to accommodate the cemented drilling pad, other equipment and trucks. We envisage the utilization of two machines for road construction and one for well pad preparation over two-week periods (NYSDEC, 2015, p. 295-296). Bulldozers,

backhoes and graders are needed to build access roads and clear the area where drilling and related activities take place. We assume a net engine power of 150 kW for bulldozers, 170 kW for excavators and 190 kW for graders. Emission factors (EFs) and other estimates are based on data from Helms et al.(2010) and NYSDEC (2015).

Road preparation: 2 bulldozers, 2 excavators, 2 graders; Load Factor (LF; this parameter indicates the time share the machinery is in operation): 50%. Well pad configuration: 2 bulldozers, 1 excavator; LF: 50%.

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E(i) = \#_{wellpads}\ x\ EF(i)_{machines}\ x\ \#_{machines}\ x\ LF\ x\ h_{operations}\ x\ kW_{machine}$$

Where (i) is the specific pollutant.

Table S3. Values for REm and OEm applied to well pad construction.

REm-U	REm-L	OEm-U	REm-L
Road construction: 2 bulldozers, 2 excavators, 2 graders, 2-week operation **Wellsite configuration:** 2 bulldozers, 1 excavator, 1-week operation **LF**: 0.5 **EF**: stage IIIB/IV[2]	**Road construction:** 2 bulldozers, 2 excavators, 2 graders, 2-week operation **Wellsite configuration:** 2 bulldozers, 1 excavator, 1-week operations **LF**: 0.5 **EF**: stage IV	**Road construction:** 2 bulldozers, 2 excavators, 2 graders, 2-week operation **Wellsite configuration:** 2 bulldozers, 1 excavator, 2-week operations **LF**: 0.5 **EF:** stage IIIB/IV	**Road construction:** 2 bulldozers, 2 excavators, 2 graders, 2-week operation **Wellsite configuration:** 2 bulldozers, 1 excavator, 1 week operation **LF**: 0.5 **EF:** stage IV

2.3 Trucks and water supply

In this section we estimate the emissions generated by truck movements and electricity need to provide the well pads with materials required for i) well pad construction, ii) drilling the borehole, and iii) fracking activities. We consider the employment of trucks with a capacity of 20 m^3 for liquid transport (i.e., mainly chemicals and waters) and 30 m^3 for solids (i.e. drilling mud, sand, cement and proppants). A total number of 480 truck movements per well pad are assumed based on data reported from NYSDEC 2015 (p. 6-305). In our scenarios, the cement pad holding drilling operations is 30 x 30 m in REm and 10 x 30 m in OEm, half-meter thick and composed by a cement-sand-water mixture in the ratio 1:4:1.

Emissions associated with the trucks employed are based on the HBEFA report (IVT, 2015) considering an average speed of 40 km h^{-1}. The vehicle market in Europe and its emissions standard share is based on the KBA report (p. 27).[3] Here the % of each emission category is calculated only considering Euro II, III, IV, V (together with EEV, see Footnote 5 at p. 42 of the same report), and VI. "Sonstige", as it is not categorized, is excluded from our analysis. The trucks we consider, when loaded, belong to the group "12001 and more kg". Here values refer to trucks half-loaded. Because

[2] Emission standards for Nonroad Engines in the EU. More information available at: https://www.dieselnet.com/standards/eu/nonroad.php#s3. Accessed 15 April 2019.
[3] Report available at: http://www.kba.de/SharedDocs/Publikationen/DE/Statistik/Fahrzeuge/FZ/2016/fz13_2016_pdf.pdf?__blob=publicationFile&v=2. Accessed 15 April 2019.

our trucks are completely loaded on the way to the well site but mostly unloaded on their return, emissions related to 50%-loaded trucks can be fairly adopted. We estimate each truck drives about 100 km (including both ways), and the volume of water necessary for each fracking stage is kept constant at 2,000 m^3.

PM produced by tyre, brake, road wear combined and re-suspended material from truck movements are added to the emission scenarios according to EFs reported by the EMEP/EEA (2016) and Denier Van Der Gon et al. (2018). The share of km driven in highways vs. urban/rural roads is chosen at 70 and 90% in the high and low boundaries respectively. PM EFs fall to zero values during rain events and when the road surface is wet. To include this consideration into our scenarios we estimate the number of rainy days in Germany (186 d y^{-1}) and the UK (163 d y^{-1}), averaging it from major cities located in or close to the reservoir areas (Statista, data for 2008 and 2017 respectively, available online).

Most of the energy required to move a water mass is spent to lift the water, while in a horizontal tract the only resistance opposing the movement in the pipeline system is composed by frictional forces. In our case, distances and changes in altitude are unknown. Assuming that frictional forces are negligible, we estimate that (on average) our masses of water is lifted to a height of 50 m on their way to the production site. This assumption would therefore include cases where the water (e.g., from natural reservoirs like lakes, rivers, etc.) is delivered downhill without additional energy required, and cases where the energy required is higher (i.e., transporting the water uphill). Energy requirements are calculated using the following equation (CottonInfo, 2015). To calculate the electricity required to pump 1 Ml (1,000 t) of water, we apply the following equation:

$$Electricity = \frac{2.275\ x\ TDH}{EF_{pump}\ x\ EF_{drive}\ x\ EF_{motor}}$$

Where:

TDH: vertical difference between water source and delivery;
Eff_{pump}: efficiency of the pump (between 0.5 and 0.9, we choose 0.8);
Eff_{drive}: Efficiency of drives (between 0.95 and 1, we choose 1);
Eff_{motor}: Motor efficiency 0.9 (average from electrical motors).

Accordingly, the power requirement for 1 Ml is 190 kWh in our scenarios.

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E(i) = (\#_{truck}\ x\ EF(i)_{truck}\ x\ km_{truck}) + (kWh\ x\ EF\ (i)_{electr.})$$

Where:
(i) is the specific pollutant;
The electricity consumption is required for pipelining drilling and fracking waters.

Table S4. Values for REm and OEm applied to trucks utilization and water recycling.

REm-U	REm-L	OEm-U	OEm-L

Stage length: 60 m **Water piped:** 0% **Truck EFs:** Mixed Euro 3/6 **Well construction:** 10 verticals **Drilling water recycled:** 50% **Fracking water recycled:** 50% **Wear:** $PM_{2.5}$ and PM_{10} from upper 95% confidence interval **Road type:** Highway:Urban:Rural= 0.7:0.2:0.1	**Stage length:** 300 m **Water piped:** 100% **Truck EFs:** Mixed Euro 3/6 **Well construction:** 10 verticals **Drilling water recycled:** 50% **Fracking water recycled:** 50% **Wear:** $PM_{2.5}$ and PM_{10} from lower 95% confidence interval **Road type:** Highway:Urban:Rural= 0.9:0.05:0.05	**Stage length:** 60 m **Water piped:** 0% **Truck EFs:** Euro 6 **Well construction:** 3 verticals **Drilling water recycled:** 90% **Fracking water recycled:** 90% **Wear:** $PM_{2.5}$ and PM_{10} from upper 95% confidence interval **Road type:** Highway:Urban:Rural= 0.7:0.2:0.1	**Stage length:** 300 m **Water piped:** 100% **Truck EFs:** Euro 6 **Well construction:** 3 verticals **Drilling water recycled:** 90% **Fracking water recycled:** 90% **Wear:** $PM_{2.5}$ and PM_{10} from lower 95% confidence interval **Road type:** Highway:Urban:Rural= 0.9:0.05:0.05

2.4 Drilling

We envisage four drilling machines at each site: three in constant use and one as a back-up, so the assigned LF is 0.75 (as discussed with experts in the field) instead of 0.6 as reported by other authors (e.g., Pring et al., 2015). Four diesel electricity generators, 1000 kW each, are assumed to be installed at each well pad in REm, while in OEm electricity is always provided by the national grid power. Although the distance between remote wellsites and the electricity network might make this case rather unrealistic, a higher population density in Europe compared with the US must be accounted for (Kavalov and Pelletier, 2012). The time needed to complete the drilling operations is based on the drilling speed reported by Pring et al. (2015) and the total length of the wellbores, different in OEm and REm (3 and 10 vertical wells, respectively). Aggregation of several horizontal wells onto a single vertical well as implemented in our scenarios – enabled by recent engineering innovation - is a practice that has shown to control emissions (Robertson et al., 2017).

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E(i) = \#_{wells\ under\ construction}\ x\ EF(i)_{drilling}\ x\ \#_{machines}\ x\ LF\ x\ h_{operations}\ x\ kW_{machine}$$

Where (i) is the specific pollutant.

Table S5. Values for REm and OEm applied to the drilling stage.

REm-U	REm-L	OEm-U	OEm-L
Diesel-powered generators **EF:** Stage IV-IIIb 10 horizontal wells each vertical well wells each horizontal	Diesel-powered generators **EF:** Stage IV-IIIb 10 horizontal wells each vertical well wells each horizontal	Electric drilling 3 horizontal wells each vertical well wells each horizontal	Electric drilling 3 horizontal wells each vertical well wells each horizontal

2.5 Fracking

We assume a total pump power capacity required for each fracking stage between 35,000 and 45,000 hp with a LF of 0.5% (Roy et al., 2014). Electric pumps are not expected here due to the large amount of energy required that is not available from the national power grid under usual settings. Based on a fracking stage length between 60 and 300 m extrapolated from the literature and experts, we obtain a range of fracking stages for each 2,000 m-long horizontal well between 8.3 and 41.6

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E(i) = \#_{wells\ under\ construction}\ x\ EF(i)_{pumps}\ x\ \#_{machines}\ x\ LF\ x\ h_{operations}\ x\ kW_{machine} + \#_{stages}$$

Where (i) is the specific pollutant.

Table S6. Values for REm and OEm at the fracking stage.

REm-U	REm-L	OEm-U	OEm-L
41.6 stages wells^{-1} **Fracking operations:** 2.5 h **EF:** 50% stage IV – 50% stage IIIB	8.3 stages well^{-1} **Fracking operations:** 1.5 h **EF:** 50% stage IV – 50% stage IIIB	41.6 stages well^{-1} **Fracking operations:** 2.5 h **EF:** 50% stage IV	8.3 stages well^{-1} **Fracking operations:** 1.5 h **EF:** 50% stage IV

2.6 Well completion

To estimate CH_4 loss during the well completion stage, we refer to the findings reported by Allen et al. (2013). In their study, empirical data from a large population of wells resulted in an average emission of 1.7 Kt of CH_4 per well completion activity, significantly below the results shown by the EPA GHG Inventory 2016, Annex 3, Table A-134 (EPA, 2016), where volumes range between 3.2 and 36.8 Kt according to the technique adopted. Nevertheless, another EPA report (EPA, 2014a) states that a reduction of total emissions between 95 and 98% can be achieved through Reduced Emission Completions (REC also called "green" completion). We assume no existing limitations for REC such as absence of nearby pipelines or low pressure of the gas, also considering that this gas can be combusted in small site-turbines to produce electricity on-stage, a technology which is already deployed in Europe.

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E = \#_{wells\ under\ construction}\ x\ loss_{well\ site}$$

Table S7. Values for REm and OEm applied to well completion activities.

REm-U	REm-L	OEm-U	OEm-L
3.3 t CH_4 event^{-1} (high-boundary 95% confidence interval)	1.0 t CH_4 event^{-1}	1.7 t CH_4 event^{-1}	0.7 t CH_4 event^{-1} (low-boundary 95% confidence interval)

2.7 Production sites

Emissions of CH_4 and other pollutants at the production sites are chosen according to results from Omara et al. (2016). Here the CH_4 leakage rate average is estimated at 0.23% of total production, with a maximum of 0.40%. Production at wellsites analyzed by Omara et al. presents production volumes similar to our shale gas industry (between 40 x 10^6 cf d^{-1} in Germany P50 and 130 x 10^6 cf d^{-1} in UK P50 scenarios). Moreover, the study focused on the Marcellus play, a predominantly gas-producing area with several horizontal wells per pad at new sites. As reported by Marchese et al. (2015) and Mitchell et al. (2015), in the production sector compressor leaks are responsible for almost 90% of total CH_4 emissions. Substituting diesel with electric compressors would eliminate uncombusted fugitive CH_4 (Marchese et al. 2015 Supporting Information; Mitchell et al., 2015 Supporting Information). Nevertheless, in OEm (where we envisage the extreme case where all the sites are provided with electricity from the national grid) we assume an emission decrease by only 90% with respect to REm to account for accidental leaks from valves and joints.

Emissions reported by Omara et al. were measured during production activities including liquids unloading operations, the emissions of which are calculated separately in our scenarios. Therefore, to avoid double-counting, we reduce the volume of CH_4 from producing wells by the amount estimated during liquids unloading (see Section 2.9).

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E = gas_{\ produced} x\ leakage\ rate_{production\ site}$$

Table S8. Values for REm and OEm applied to the production sites stage.

REm-U	REm-L	OEm-U	OEm-L
Diesel compressors **Gas loss:** 0.40%	Diesel compressors **Gas loss:** 0.23%	Electric compressors **Gas loss:** 0.04% (10% of REm-U)	Electric compressors **Gas loss:** 0.02% (10% of REm-L)

2.8 Wellhead compressor exhaust

Wellhead compressors help to increase productivity from mature reservoirs where the natural gas pressure is not high enough to ensure economic production. We assume the engagement of 3 diesel compressors in REm (where 3 horizontal wells are connected to the same vertical well), and an electric compressor of 750 kW OEm, where the number of aggregated horizontal wells is 10 for each vertical well. The occurrence of wellhead compressors is assumed to be 25% of total producing sites (NYSDEC, 2015). Also for this stage of the shale gas supply chain, diesel compressors in REm are substituted with electric ones in OEm.

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E(i) = \#_{produing\ wells}\ x\ \%_{wells}\ x\ EF_{(i)}\ x\ h_{operations}\ x\ kW_{compressor}$$

Where:
(i) is the specific pollutant;
% wells is the share of wells we estimate are equipped with wellhead compressors.

Table S9. Values for REm and OEm applied to wellhead compressor exhaust.

REm-U	REm-L	OEm-U	OEm-L
Diesel compressors (300 kW) **EF:** stage IIIB/IV.	Diesel compressors (300 kW) **EF:** stage IIIB/IV.	Electric compressor (750 kW) - CH_4 accidental leaks are not accounted since they are already at losses at production sites	Electric compressor (750 kW) - CH_4 accidental leaks are not accounted since they are already at losses at production sites

2.9 Liquids unloading

Liquids unloading is an engineering practice that is required during gas production when the liquids co-produced with the gas clog the well and restrict or obstruct the free flow of gas. The frequency of this practice depends on the natural tendency of the well to produce liquids, which is strictly related to the geology of the target formation and the age of the well. Allen et al., 2015 SI (Table S5.2) reports that liquids unloading is performed at ca. 80% of the wells analyzed in their study, covering a population of different ages and nature (for both fracked and non-fracked wells). Plunger lifts are used to remove the liquids accumulated in the well and restore gas production, a valuable alternative to large VOC emissions during the blowing down of the well (EPA, 2014b). Both manual and automated plunger lifts are very effective in preventing emissions, although the former relies on onsite manual performance and is therefore less reliable than the automated one. It is important to note that the population of wells in our dry gas scenario does not require this procedure since dry gas is inherently low in VOCs. Since wells considered in our study are all relatively young (maximum 8-10 years old), we assume that between 5 and 10 liquids unloading activities take place at each well per year (Allen et al. 2015).

In OEm we assume implementation of automatic plunger lifts, which may become standard if strict regulations are in place. In REm, manually triggered plunger lifts are considered. Wells without plunger lifts have not been considered in our scenarios since this would not respect sufficient environmental standards. Uncertainties here are given by the low- and high-boundaries for emissions shown in Figure 5 of Allen's paper.

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E(i) = \#_{producing\ wells}\ x\ EF(i)_{wellsite}\ x\ \text{annual events}$$

Where (i) is the specific pollutant

Table S10. Values for REm and OEm applied to liquids unloading.

REm-U	REm-L	OEm-U	OEm-L
Manually triggered plunger lifts **Liquids unloading per well:** 10 events y^{-1} **CH_4 emissions:** 351 m^3 $well^{-1}$	Manually triggered plunger lifts **Liquids unloading per well:** 5 events y^{-1} **CH_4 emissions:** 195 m^3 $well^{-1}$	Automatic triggered plunger lifts **Liquids unloading per well:** 10 events y^{-1} **CH_4 emissions:** 59 m^3 $well^{-1}$	Automatic triggered plunger lifts **Liquids unloading per well:** 5 events y^{-1} **CH_4 emissions:** 14 m^3 $well^{-1}$

2.10 Gathering facilities and pipelines

Gathering is defined as the pipeline system connecting the wellhead compressor with the processing plant. Along this path several facilities and devices are employed to ensure a regular and safe flow of the gas, such as gathering compressors, separators for CO_2, water and condensate, and others. Here, emissions from gathering facilities and pipelines are calculated separately. Mitchell et al. (2015) provides data on CH_4 losses from this connecting system which we critically apply to assign appropriate EFs to the emission scenarios.

2.10.1 Gathering facilities

In order to define the amount of gas lost at gathering facilities (for leaks from pipelines, see below), we refer to a selection of stations analyzed by Mitchell et al. (2015), in which the gas throughput is comparable to the gas collected at gathering plants as described in our study (ranging between 12.7 to 217 t y^{-1}). The 25 and 75 FLER (Facility-Level Emission Rate) %tile averages of this selected population of plants is assigned in OEm (657 t $facility^{-1}$) and in REm (1110 t $facility^{-1}$) scenarios. Due to the fact that the number of electric compressors in operation is unknown since they were not listed by Mitchell et al. during sampling campaigns, and that some plumes were not correctly measured or systematically captured (as discussed in Mitchell's paper), there is the possibility that the data source to which we refer are overall biased slightly low.

As observed in the US gas plays and reported by Marchese et al. (2015) and Mitchell et al. (2015), gathering facilities generally collect gas from 10 to 100 horizontal wells. We assume that this parameter is regulated under state law and therefore we apply two different cases in our emission scenarios: 1) gas collected from 30 wells at gathering facilities in REm; and 2) gas collected from 80 wells in OEm.

2.10.2 Emissions from supplementary devices at facilities

Based on data reported in Marchese et al. (2015, SI), we assign the number of compressors, pneumatic controllers, pneumatic pumps, chemical pumps and Kimray pumps at each gathering station. Pneumatic devices are mechanically powered by the high-pressure of the gas, and are implemented anytime electricity supply cannot be provided. EFs for these devices are assigned according to data reported by Helms et al. (2010).

Other parameters that we associate with these devices are reported in Table S11.

Table S11. Number and operational characteristic of devices at gathering stations.

Device	Number $facility^{-1}$	Loading factor (LF)[4]	kW
compressors	35	60%	127[5]
pneumatic controllers	69	10%	0.03[6]

[4] Report available at: http://www.ourenergypolicy.org/wp-content/uploads/2014/04/epa-devices.pdf. Accessed 15 April 2019.
[5] Mitchell et al., 2015.
[6] See ref. 5.

pneumatic pumps	5	10%	1.7[7]
chemical pumps	12	10%	1.7[8]
Kimray pumps	1755	10%	3.7[9]

We assume that in REm all compressors are run via diesel engines, while controllers and pumps are pneumatic or activated by the national power grid (therefore assuming that connection to the national power grid is always possible). In OEm, all machines and controllers are supplied by the national power grid. As noted earlier, electric compressors have the potential to eliminate gas leaks. Emission ranges will therefore be representative for all types of compressors: diesel (highest emission case), electric (lowest emission case), and natural gas-powered (intermediate emission case).

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$\mathrm{E}(i) = \#_{gatherin\ \ plants}\ x\ [(losses_{gathering\ plant} + (\#_{devices}\ x\ kw_{device}\ x\ h_{operations}\ x\ LF_{device} x\ EF_{(i)})]$$

Where (i) is the specific pollutant.

Table S12. Values for REm and OEm applied to gathering facilities.

REm-U	REm-L	OEm-U	OEm-L
CH_4 loss at facility: 1110 t **Wells connecting to facilities:** 30 **EFs compressors:** Stage IIIB/IV	**CH_4 loss at facility:** 657 t **Wells connecting to facilities:** 30 **EFs compressors:** Stage IIIB/IV	**CH_4 loss at facility:** 5% of REm-U, plus emissions from electrical grid power **Wells connecting at facilities:** 80	**CH_4 loss at facility:** 5% of REm-L, plus emissions from electrical grid power **Wells connecting at facilities:** 80

2.10.3 Gathering pipelines

In order to assign gas emissions from gathering pipelines, we select data reported by Marchese et al. (2015, SI) in which they model the CH_4 loss rate in the US natural gas supply chain. Results show a gas leak of 0.035%, a value similar to the one reported by the EPA GHGI. We differentiate REm and OEm based on the fact that in the former more above-ground wellheads are planned (10 wellheads per well pad vs. 3 in OEm). In the absence of better data, we assume an arbitrary emission reduction in OEm by 15% when compared to REm that accounts for the reduced number of wellheads present on each well pad.

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E = gas_{produced}\ x\ leakage\ rate_{gathering\ pipeline}$$

[7] See ref. 5.
[8] Data available at: https://www.itc.es/. Accessed 15 April 2019.
[9] Data available at: https://kimray.com/Downloads/Marketing/Electric_Glycol_Pump/SSEG-001_Electric_Glycol_READER.pdf. Accessed 15 April 2019.

Table S13. Values for gas leakage rate for applied to gathering pipelines for REm and OEm.

REm-U	REm-L	OEm-U	OEm-L
Gas leakage: 0.035%	**Gas leakage:** 0.035%	**Gas leakage:** 0.030%	**Gas leakage:** 0.030%

2.11 Processing

2.11.1 Gas emissions

To determine natural gas and air pollutant emissions at natural gas processing stations we refer to the study by Mitchell et al. (2015) and to the AP-42 Report from the EPA[10]. The amount of gas produced at each well pad ranges between 12.7 and 217 t h^{-1} in our study, making it reasonable to assume that the amount of gas processed at each facility of our scenarios is comparable to data discussed in Mitchell et al. (here the gas amount ranges between 100 and 780 t h^{-1}). Based on these similarities, we assign the same leakage rate suggested by Mitchell's study at processing plants: 0.046% and 0.079% in OEm and REm, respectively.

By substituting diesel-engine compressors with electric ones in OEm, it would completely eliminate CH_4 leaks from these devices (Marchese et al. 2015 Supporting Information; Mitchell et al., 2015 Supporting Information) as well as most VOC emissions onsite: according to Marchese et al. (2015) venting and combustion from compressors represent 90% of all emissions (Table S5 in SI, Marchese et al.). Since we cannot rule out that emissions-reducing compressors (e.g., electric) were also deployed at the processing stations investigated by Mitchell et al., we only assume emission reductions of 50% in REm to avoid potential double counting.

2.11.2 Energy requirement

Different electricity-producing gas turbine typologies may be employed at processing plants (e.g., simple or with abatement measures for NO_x and CO like water-steam injection or heat-recovery systems). According to the AP-42 Report from EPA, simple cycle gas turbines are often used in the petroleum industry due to the low price and large availability of gas. In order to cover different turbine typologies, in our scenarios we assume implementation of uncontrolled gas turbines in REm and water-steam injection turbines in OEm. The latter is technologically more advanced and requires high quantities of fresh water which may make its adoption challenging in some areas. Therefore, its adoption is more consistent with an optimistic scenario. The AP-42 report indicates a gas combustion efficiency value for simple cycle turbines between 15 and 42%, while between 38 and 60% for combined cycle gas turbines. Very similar values are assigned to turbines for oil and gas applications by Siemens.[11] Accordingly, in our REm and OEm we apply turbines with a range of efficiency spanning from 30 to 60%.

In our scenarios, we estimate that 164 kWh of energy is required to process 1,000 m^3 of natural gas based on Müller-Syring et al. (2016) and consultants from the oil and gas industry. Assuming turbine

[10] Report available at: https://www3.epa.gov/ttnchie1/ap42/ch03/final/c03s01.pdf. Accessed 15 April 2019.
[11] An overview on different gas turbines available on the market is available at: https://www.siemens.com/global/en/home/products/energy/power-generation/gas-turbines/refining-petrochemical.html. Accessed 15 April 2019.

efficiencies of the range reported above, the amount of gas combusted on site ranges between 2.8 and 5.6% of total gas processed in the different scenarios.

Total emissions for each productivity case, emission scenario and country are calculated as follows:

$$E(i) = \left(leakage\ rate_{processing\ plants}\ x\ gas_{processed}\right) + \left(gas_{combusted} x\ EF_{(i)}\right)$$

Where:

(i) is the specific pollutant;
$gas_{combusted}$ depends on turbine efficiencies.

Table S14. Values for REm and OEm applied to gas processing.

REm-U	REm-L	OEm-U	OEm-L
Gas leakage: 0.079% Uncontrolled gas turbines with combustion efficiency of 30%.	**Gas leakage:** 0.046% Uncontrolled gas turbines with combustion efficiency of 40%.	**Gas leakage:** 0.039% Water-steam injection turbines with combustion efficiency of 50%.	**Gas leakage:** 0.023% Water-steam injection turbines with combustion efficiency of 60%.

2.12 National power grid

Power from the national electric grid is required in our scenarios to supply energy for operations at different stages. The amount of power required varies according to the technology involved in the different scenarios. EFs for the national electric grid are calculated according to emissions produced by the different energy carriers (mainly coal and natural gas) and to their share of energy generated. The German Environmental Agency (UBA) provides detailed and updated EFs for the national power grid[12], while data available for the UK are limited to CO_2 and CH_4. We therefore decide to apply EFs for Germany to both countries (data for Germany 2015, see Table S17), given the very similar gCO_2-eq. kWh^{-1} values associated with both countries (540 gCO_2-eq. kWh^{-1} for Germany and 528 gCO_2-eq kWh^{-1} for the UK in 2016, see Section *Emission Intensity* in the manuscript) and the negligible relative component that the power sector exerts on total CO_2 and CH_4 emissions (see discussion in the Sensitivity Analysis Section). The two countries examined have a similar energy mix: ca. 40% of the power generated is produced via nuclear and renewables, while the remaining share differs in gas and coal utilization: respectively 12 and 45% in Germany, while 42 and 22% in the UK.

Table S15. EFs for the national power grid applied for Germany and the UK. Data in g kWh^{-1}.

Pollutant	value
NO_x	0.454
PM_{10}	0.016
$PM_{2.5}$	0.014
CO	0.227
CO_2	534.000
N_2O	1.816
CH_4	0.167
VOCs	0.018

[12] Emission factors are available at: https://www.umweltbundesamt.de/themen/luft/emissionen-von-luftschadstoffen/spezifische-emissionsfaktoren-fuer-den-deutschen. Accessed 15 April 2019.

3 Sensitivity analysis

We carry out a sensitivity analysis (SA) for the scenarios presented in our manuscript to gain insight into the effects and implications that the input parameters of the system (i.e., independent variables) exert on final emissions (i.e., dependent variable). This procedure enables us to quantitatively characterize the sensitivity of the output values when each single input variable changes, so as to determine the influence of the input and therefore the accuracy it requires when defining the values' boundary. The SA is also a method to assess the robustness and limitations of the model, which in turn provides guidance on interpretation of results and restrictions of the results' applicability. The SA is carried out by systematically varying each input parameter while keeping the others constant, and observing the effect that this variation has on the output. The SA is run for the REm-P50 case for Germany, and investigates the following pollutants: CO_2, CH_4, VOCs and NO_x. We vary parameters that are constant through the scenarios, as well as variables that define and differentiate the scenarios. This is because the choice about variability of these coefficients is finalized at a later stage, and is also partially based on the SA results. A selected group of variables and parameters are increased from the lower end (REm-L) to the upper end of their uncertainty range (REm-U) to observe the variation generated on total emissions for each pollutant. When no range is described due to lack of data (i.e., kilometers driven by trucks), a flat increase of 50% is imposed. Results show a large impact variance associated with single emitting stages across the production system, and this distribution is characteristic and special for each pollutant. Variables which are shown to significantly affect final overall emissions undergo further investigation. Hereby we analyze variables and parameters based on two different aspects: on the one hand we measure the impact (in %) that a variation of the independent variables within its uncertainty range has on total emissions (i.e., effective impact, Table S16). On the other, we normalize this impact per single unit of variation of the independent variable, in order to define the "power" of a variable to affect final total emissions independent of the range of variability we impose (i.e., potential impact, Table S17). While through the first approach we obtain a sense of the real influence that each variable has in our study, the second one provides us with a qualitative assessment of the strength that such variables possess in affecting overall results.

CO_2 total emissions are largely influenced by variation in the utilization and performances of the numerous engines employed along the production chain, so that variables related to these during drilling, at wellhead compressors, at gathering facilities and during processing (i.e., efficiency of gas turbines during processing) strongly affect final emissions (Figure S2). Results show that, although the number of gathering facilities can strongly affect final CO_2 outputs, the very restricted variability within the OEm and REm range boundaries significantly limits its effective contribution. It is anyway worth noting that the range of this parameter varies significantly in the OEm and REm, so that its overall relevance in these emission cases is very different. A wide range characterizing the fracking stage time interval is responsible for an overall contribution up to 6.1% despite a low normalized potential (up until 2.6%). Gas turbine efficiency during processing is by far the key parameter capable of raising total emissions by 16.6% when varying within its uncertainty range. Likewise, it displays high normalized sensitivity. Figure S3 evidences how parameters controlling CH_4 emissions differ substantially from the results for CO_2. Gas losses at production sites and gathering facilities markedly dominate total emissions, each raising the final output by 31.6% and 32.7% respectively when varying within their corresponding uncertainty ranges. The normalized impact of the number

of gathering facilities necessary to streamline the gas register contribution on final emissions very similar to the ones displayed by CH_4 emissions at the same stage (both circa 41.6%), although its narrow confidence range preclude significant effects on total emissions. Gathering pipelines and processing activities (gas lost and turbine efficiency) show a normalized impact below 10% and an effective impact below 5%. As expected, VOC emissions (Figure S4) closely resemble CH_4 results: these two pollutants are (for the most part) linearly correlated, as they are co-emitted through natural gas losses. NO_x are emitted by hundreds of diesel engines employed across the gas production chain, so that the utilization of all these at each stage of gas production affect total emissions to different extents (Figure S5). Its highest impacts observed on total emissions are from the time interval of fracking operations, followed (in order) by gas turbine efficiency at processing stations, volume of water per fracking stage, as well as fracking stage length and drilling time operations. The influence of these stages on total NO_x emissions ranges from 8.3% to 19.3% per stage, while their normalized impacts range from 16.6% (drilling operations) up to 66.3% (gas turbine efficiency). Once again, the parameter "# gathering facility" has a considerable normalized impact but a negligible real influence on overall emissions. As observed for CO_2, the fracking stage length has an effective impact of 11.1% despite a much lower normalized potential.

Production site preparation and ancillary operations during drilling and fracking do not have any relevant repercussions on total emissions of the pollutants examined here. Similar results are evidenced by operations of liquids unloading as well as parameters associated with trucks such as kilometers driven, emission standards, and others. CH_4 and VOCs potentially lost during well completion operations or at the liquids unloading stage have a very irrelevant contribution to total volumes, although their effects at the local level may be more pronounced. EFs from the electrical power grid do not show any appreciable contribution to total volume for all pollutants, although showing a minimal effect on NO_x emissions (3.6% of effective impact against 7.1 of potential impact).

Table S16. Effective impact of independent variable variations on total emissions. The table summarizes the effect of variations of single pollutants within their range boundaries and at different stages of gas production on final emissions. The variation applied is reported in the final column.

Stage	Parameter	CO_2	CH_4	VOCs	NO_x	Variation
Well pad developmet	Duration of operations	<0.1%	-	-	<0.1%	REm-L to REm-U
Truck traffic	Driving distance	0.1%	-	-	0.1%	50%
Drilling	Duration of operations	3.7%	-	<0.1%	8.3%	50%
Fracking operations	Length fracking stage	5.2%	-	<0.1%	11.1%	REm-L to REm-U
	Vol. water per fracking stage	0.1%	-	-	11.1%	50%
	Duration of operations + # stages	0.8%	-	<0.1%	19.3%	REm-L to REm-U
Well completion	Emissions CH_4 per well	-	0.5%	0.5%	-	REm-L to REm-U
Production sites	loss (% of production)	-	31.6%	31.6%	-	REm-L to REm-U
Wellhead compressors	# Compressors	4.2%	-	<0.1%	2.0%	50%
Liquids unloading	Absolute emissions	-	-	-	-	REm-L to REm-U
Gathering facilities	# facilities	0.5%	0.6%	0.6%	0.4%	REm-L to REm-U
	Absolute emissions	-	28.8%	28.8%	-	RmM-L to REm-U
	Gas leaked (pipelines)	-	3.3%	3.3%	-	50%
Processing	Gas leaked (% of gas processed)	-	6.1%	6.1%	-	REm-L to REm-U

	Gas turbine efficiency	16.6%	1.4%	1.4%	16.6%	REm-L to REm-U
Electricity	EF electricity	2.4%	<0.1%	0.01%	3.6%	50%

Table S17. Potential impact of independent variable variations on total emissions. The table summarizes the effect of normalized variations of single pollutants at different stages of gas production on final emissions. The variation applied is reported in the final column.

Stage	Parameter	CO_2	CH_4	VOCs	NO_x	Variation
Well pad developmet	Duration of operations	<0.1%	-	-	<0.1%	REm-L to REm-U
Truck traffic	Driving distance	0.3%	-	-	0.3%	50%
Drilling	Duration of operations	7.4%	-	<0.1%	16.6%	50%
Fracking operations	Length fracking stage	1.3%	-	-	2.8%	REm-L to REm-U
	Vol. water per fracking stage	0.1%	-	-	22.2%	50%
	Duration of operations + # stages	1.2%	-	<0.1%	28.9%	REm-L to REm-U
Well completion	Emissions CH_4 per well	-	0.5%	0.5%	-	REm-L to REm-U
Production sites	loss (% of production)	-	42.7%	42.7%	-	REm-L to REm-U
Wellhead compressors	# Compressors	8.3%	-	<0.1%	4.0%	50%
Liquids unloading	Absolute emissions	-	-	-	-	REm-L to REm-U
Gathering facilities	# facilities	35.2%	41.6%	41.6%	29.0%	REm-L to REm-U
	Absolute emissions	-	41.5%	41.6%	-	REm-L to REm-U
	Gas loss (pipelines)	-	6.5%	6.5%	-	50%
Processing	Gas leaked (% of gas processed)	-	8.5%	8.5%	-	REm-L to REm-U
	Gas turbine efficiency	66.4%	5.5%	5.4%	66.3%	REm-L to REm-U
Electricity	EF electricity	4.9%	<0.1%	<0.1%	7.1%	50%

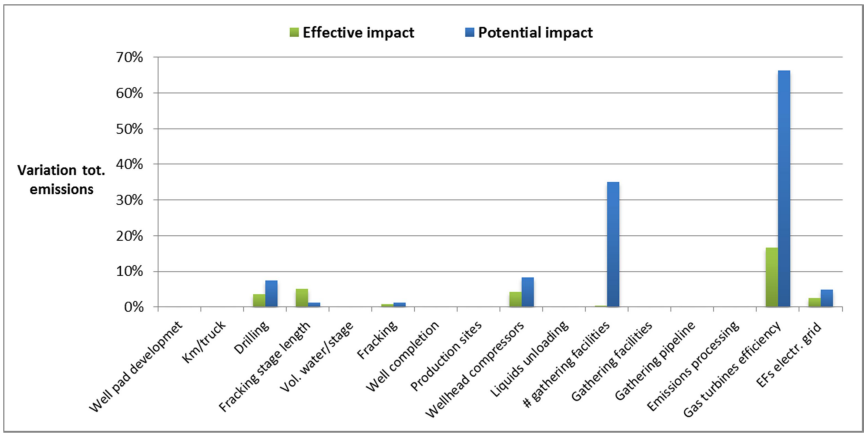

Figure S2. Sensitivity analysis results for CO_2. Visualization of the effective and potential impacts on final CO_2 emissions through variation of parameters at each stage of the upstream gas chain.

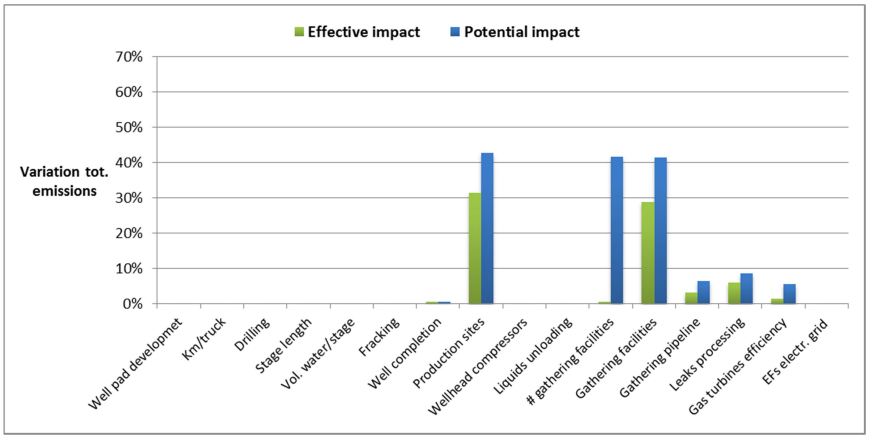

Figure S3. Sensitivity analysis results for CH_4. Visualization of the effective and potential impacts on final CH_4 emissions through variation of parameters at each stage of the upstream gas chain.

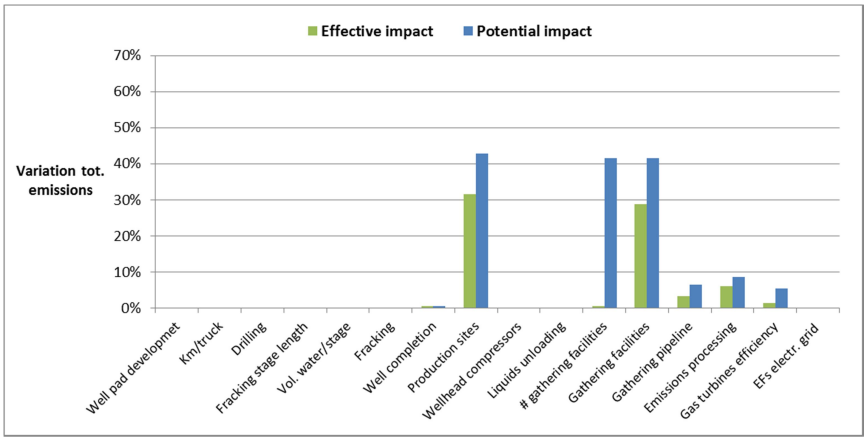

Figure S4. Sensitivity analysis results for VOCs. Visualization of the effective and potential impacts on final VOC emissions through variation of parameters at each stage of the upstream gas chain.

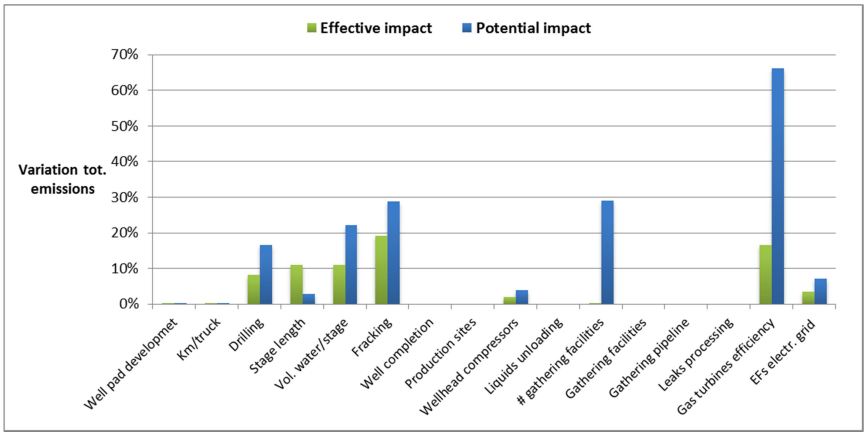

Figure S5. Sensitivity analysis results for NO_x. Visualization of the effective and potential impacts on final NO_X emissions through variation of parameters at each stage of the upstream gas chain.

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Appendix B: Supplement Paper II

Modeling the impact of a potential shale gas industry in Germany and the United Kingdom on ozone with WRF-Chem

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Supplemental material

Table S1. **Namelist used in simulations with WRF-Chem**

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start_month	= 05,
start_day	= 29,
start_hour	= 00,
start_minute	= 00,
start_second	= 00,
end_year	= 2011,
end_month	= 09,
end_day	= 01,
end_hour	= 00,
end_minute	= 00,
end_second	= 00,
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e_sn                                = 150,
e_vert                              = 35,
p_top_requested                     = 5000,
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eta_levels                          =       1.0,    0.993,    0.983,    0.97,
0.954,                              0.934,  0.909,    0.88,     0.845,
0.807,                              0.765,  0.719,    0.672,    0.622,
0.571,                              0.52,   0.468,    0.42,     0.376,
0.335,                              0.298,  0.263,    0.231,    0.202,
0.175,                              0.15,   0.127,    0.106,    0.088,
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parent_time_step_ratio      = 1,
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if_zfac_t                   = 0,
k_zfac_t                    = 8,
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gt                          = 0.0003,
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 v_mom_adv_order          = 3,
 h_sca_adv_order          = 5,
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 /

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/

&grib2
/

&namelist_quilt
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diagnostic_chem         = 0,
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/
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Text S1. conNO_x scenario development

In order to calculate the conNO_x scenario emission flux for emissions pre-processing, first the area over which to concentrate the NO_x emissions from drilling and fracking activities was calculated. Afterwards the NO_x emissions for drilling and fracking activities from the REm-U P25 scenario from Cremonese et al.

(2019), which make up 24% of total shale gas NO_x emissions for Germany and 18% for the UK, were divided by space and time values to get the emissions flux in units of kg m^{-2} s^{-1}. These calculations are described here.

Concentrated NO_x area

Displayed below are calculation steps (1-4) to convert number of wells pads (WP) in Germany (DE) and in the United Kingdom (UK) from concentrated NO_x activities to total area of concentrated NO_x emissions for the conNO_x scenario, adjusted to fit the pre-processing grid to which emissions are added. Note that, according to Cremonese et al. (2019)'s REm-U P25 scenario which are used as the basis for all simulations in this work, 166 and 206 wells are both drilled and fracked annually in DE and the UK, respectively. These values were averaged to 42 and 52 wells that are drilled and fracked over the JJA period respective to DE and the UK. In the conNO_x scenario one drilling and one fracking activity are applied per pad, so that a uniform emissions flux can be applied to the entire Concentrated NO_x area to reduce complexity (as opposed to say, half the concentrated NO_x area containing drilling activities only with one emissions flux and the other area containing fracking activities only with a different emissions flux). With one fracking and one drilling activity per pad, a total of 42 and 52 WPs are utilized in DE and the UK, respectively.

Calculation steps - Germany

1) $42\ pads_{DE} \times 25\ km^2_{WP\ area} = 1050\ km^2 total\ area_{WP,DE}$

2) $1050\ km^2_{WP,DE} / 49\ km^2_{PP\ area} = 21.43 \approx 20\ grid\ cells_{PP,DE}$

3) $20\ cells_{PP,DE} \times 49\ km^2_{PP\ area} = 980\ km^2 total\ area_{PP,DE}$

4) $980\ km^2 total\ area_{PP,DE} / 12250\ km^2 SG\ basin_{DE} = 8.0\%\ SG\ basin\ area$

Calculation steps - United Kingdom

1) $52\ pads_{UK} \times 25\ km^2_{WP\ area} = 1300\ km^2 total\ area_{WP,UK}$

2) $1300\ km^2_{WP,UK} / 49\ km^2_{PP\ area} = 26.53 \approx 25\ grid\ cells_{PP,UK}$

3) $25\ cells_{PP,UK} \times 49\ km^2_{PP\ area} = 1225\ km^2 total\ area_{PP,UK}$

4) $1225\ km^2 total\ area_{PP,UK} / 17052\ km^2 SG\ basin_{UK} = 7.2\%\ SG\ basin\ area$

Here we walk through the steps to calculate the concentrated NO_x area, for each country. In calculation step 1 the number of WPs is multiplied by the WP area used in Cremonese et al. (2019) (5kmx5km=25km^2) to calculate total WP area for concentrated NO_x emissions. In step 2, the WP area from step 1 is divided by the pre-processing (written as 'PP' in subscript) grid cell size (7kmx7km=49km^2, Kuenen et al. 2014) to adjust to the pre-processing grid, and subsequently rounded to produce a simple quadrilateral area. In step 3 the total concentrated NO_x area adjusted to the pre-processing grid is calculated. Finally in step 4 the percentage of shale gas (written as 'SG' in subscript) basin area consisting of concentrated NO_x emissions is calculated, based on the size of the shale gas basins of these two countries. Note that the shale gas basin area is based on the raster grid used in our simulations.

Concentrated NO_x time

According to Cremonese et al. (2019)'s REm-U P25 scenario, the time required to drill one well is 392 h, or approximately 16.33 days. On the other hand, the time required to frack one stage is 2.5 h, where there are a total of 41.7 stages per well. Because only one stage can be drilled at a time per well, this amounts to a total of 104.17 hours, or rather 4.34 days. In order to reduce complexity and have one emissions flux, all concentrated NO_x emissions are averaged over one period of time, based on the time required to carry out the longer activity, i.e., drilling. Since it is necessary to apply daily values in emissions pre-processing, this period of time is rounded up to 17 days.

Placement of Concentrated NO_x emissions based on sensitivity studies

In the sensitivity study to select the location for the concentrated NO_x emissions, the following steps were performed, for both Germany and the United Kingdom:

1. Shale gas basin mask netCDF file was converted from the emissions pre-processing grid to a mask adapted to the output grid, to analyze WRF-Chem output data in the shale gas regions only.
2. Areas classified as 'urban or built up land' in the USGS dataset were removed from the mask. This was done so as not to include areas which would be unrealistic for potential locations to add concentrated drilling and fracking activities.
3. The j, k, lat, lon, and $MDA8_{Diff,max}$ (scenario minus base) values were recorded per grid cell for each grid cell of the mask, and for each day of the simulation period. $MDA8_{Diff,max}$ is described in **Equation S1** below.
4. This data was obtained for two scenarios: SG1-wet gas, and SG1-wet gas *without* added shale gas NO_x emissions. Note that both scenarios contain added shale gas VOC emissions.
5. There are several shale gas basins for Germany, and so this data was filtered by removing coordinates and their corresponding values for German shale gas basins which were not large

enough to accommodate the concentrated NO_x area, i.e, Unterkarbon basins located in the Baltic Sea (near Rügen) and by the Ruhr Valley (see Cremonese et al. 2019) for depiction of basins and corresponding basin names). This was not performed for the UK, since it contains one large, continuous basin, i.e., the Bowland.

6. $MDA8_{Diff,max}$ values: SG1-wet gas (no NO_x) values were subtracted from SG1-wet gas (aptly referred to here as $MDA8_{Diff,max,NOx}$), to see what the effect of added NO_x emissions from shale gas was on MDA8 per grid cell and per day.
7. The top 30 highest $MDA8_{Diff,max,NOx}$ values for both Germany and the UK were recorded.
8. Of these top 30 values for each country, cells which formed a continuous area were filtered for. Note that in Germany a continuous area of about 4 grid cells was filtered for, and for the UK an area of about 5 grid cells, based on the size of the concentrated NO_x activities and the resolution, i.e., size of each grid cell of the output grid.
9. Based on these steps, a rough area that may be especially sensitive to concentrated NO_x emissions for increased MDA8 production was determined. Note that it was not possible to determine the exact area because of inherent and unavoidable differences between the output grid and pre-processing grid. That is to say, because the output grid and pre-processing grid contain differing horizontal resolutions, the grid cell lat/lon coordinates likewise differ.
10. After having determined a rough continuous area which displays a heightened sensitivity to added NO_x emissions for increased MDA8 production in the output grid, the closest lat and lon coordinates of the pre-processing grid matching the coordinates of the output grid were determined.
11. After determining this, a new mask was created for the concentrated NO_x area, for each country, adapted to the pre-processing grid. The mask was located roughly in the same area as the output grid.
12. Again, due to slight differences and discrepancies in coordinates as a result of converting from one grid to the other, the concentrated NO_x mask was checked against the shale gas basin mask of each country to ensure that the NO_x mask was roughly within the shale gas region; the mask was adjusted as necessary.

Equation S1

In order to provide statistical data (Table S2) on maximum difference in MDA8, the maximum of the difference in MDA8 between the scenario and base case at time t is calculated for every $x\ y$ coordinate over region R (ΔMDA8).

$$\Delta MDA8(t) = \max(MDA8_{scenario}(t,x,y) - MDA8_{Base}(t,x,y)) \forall\ xy\ \in R$$

Table S2. **Summary of ΔMDA8 statistical data over the whole domain in µg m^{-3}, over JJA**

	SG1			SG2			SG3		
Statistical data	dry gas	wet gas	conNO_x	dry gas	wet gas	conNO_x	dry gas	wet gas	conNO_x
Minimum	0.3	0.4	0.4	0.3	0.6	0.8	0.6	1.8	1.6
Q1	0.8	1.1	1.4	1.0	1.6	1.8	1.4	5.0	5.0
Median	1.2	1.4	2.1	1.3	2.5	2.9	2.1	6.7	6.3
Q3	1.6	2.0	3.2	1.8	3.2	4.0	2.7	10.2	9.7
Maximum	3.7	4.5	9.5	3.9	6.5	9.6	4.8	28.3	23.3
Average	1.4	1.6	2.6	1.5	2.6	3.2	2.2	8.0	7.5

Table S3. **Stations per country and exceedance data per country with respect to the EU threshold, over JJA**

	SG1					SG2				SG3			
Country		dry gas		wet gas		dry gas		wet gas		dry gas		wet gas	
	Σ[a]	Σ[b]	%[c]	Σ	%	Σ	%	Σ	%	Σ	%	Σ	%
France	386	27	7	32	8	30	8	41	11	40	10	67	17
Italy	244	17	7	20	8	18	7	23	9	21	9	37	15
Germany	234	4	2	4	2	4	2	6	3	5	2	16	7
Spain	129	1	1	2	2	1	1	3	2	3	2	5	4
Austria	111	2	2	4	4	4	4	4	4	4	4	6	5
U. Kingdom	80	1	1	3	4	3	4	5	6	5	6	15	19
Poland	61	1	2	1	2	1	2	2	3	2	3	2	3
Czech Rep.	60	0	0	0	0	0	0	2	3	2	3	5	8
Belgium	42	0	0	0	0	0	0	2	5	2	5	5	12
Hungary	17	0	0	1	6	0	0	1	6	1	6	2	12
Sweden	12	0	0	0	0	0	0	0	0	0	0	0	0
Netherlands	15	0	0	0	0	0	0	0	0	0	0	1	8
Slovakia	11	0	0	0	0	0	0	0	0	0	0	1	9
Denmark	7	1	14	1	14	1	14	1	14	1	14	2	29
Serbia	6	1	17	1	17	1	17	1	17	1	17	1	17

[a]Number of stations with valid measurements per country. Only country stations which are located within the model domain are included in the analysis.

[b]Number of stations that experience exceedances per country.

[c]Percentage of stations per country that have an exceedance.

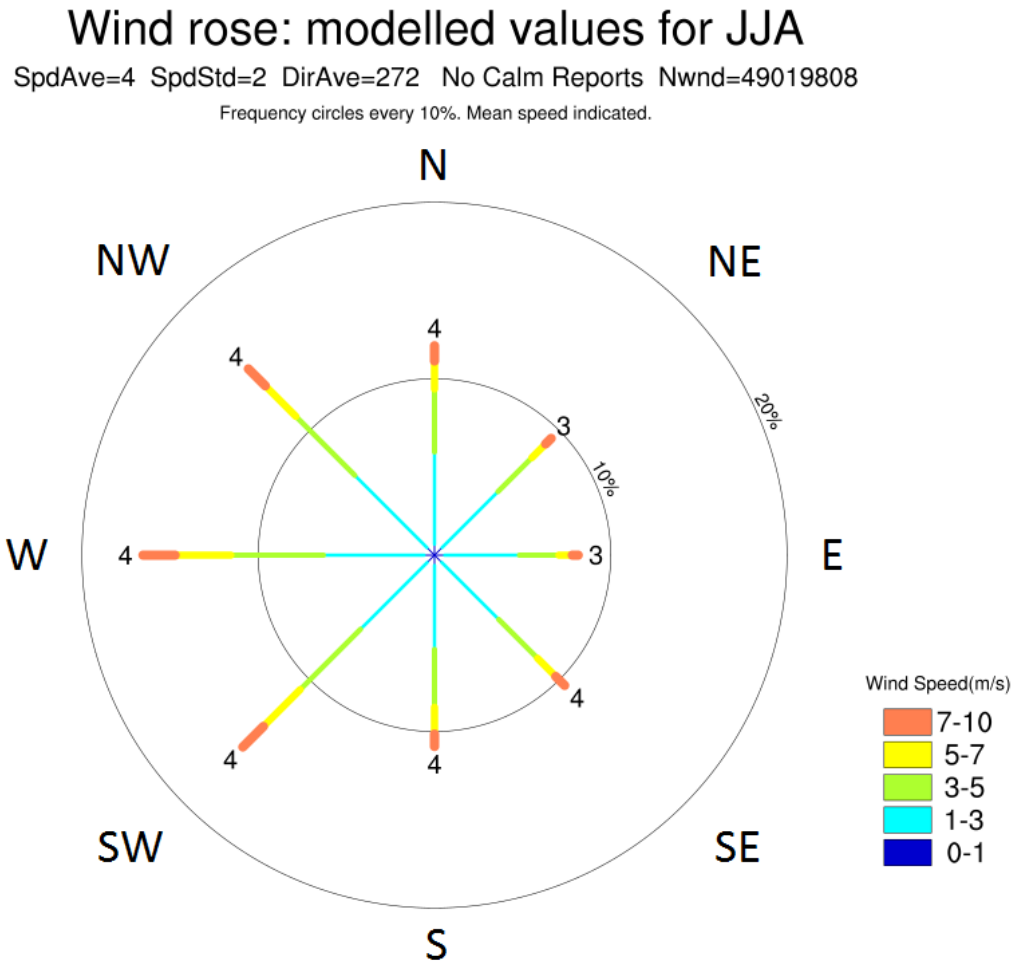

Figure S1. **Wind rose diagram of hourly modeled values over domain for JJA.** The length of each directional bar indicates the frequency (percentage of time) that wind blows from the respective direction, where each concentric circle represents a different frequency. The colors indicate the percentage of time that wind blows from a particular direction at a certain speed, in units of $m \cdot s^{-1}$. The number at the end of each directional bar indicates the average wind speed from that direction. Overall statistics for the entire data sample are included in the title, where SpdAve = wind speed average; SpdStd = standard deviation of the wind speed average; DirAve = directional average; Nwnd = number of modelled values. No calm reports means that there were no modeled data points where the wind speed was at exactly 0.

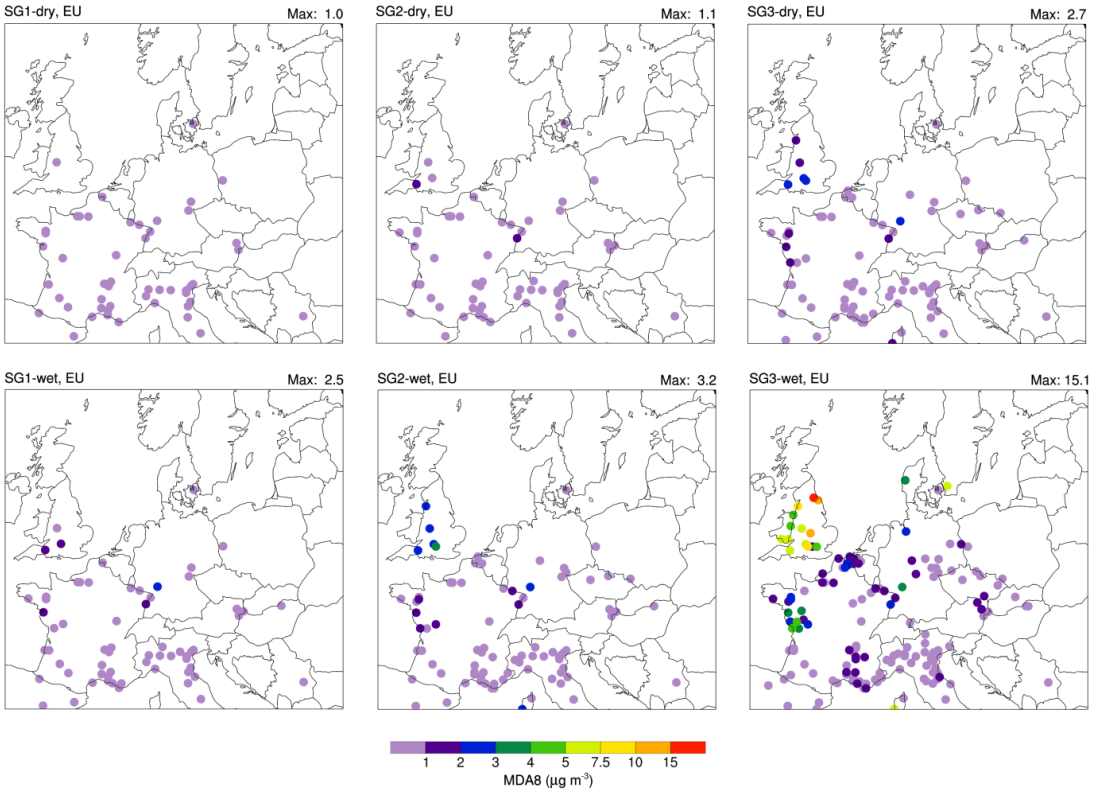

Figure S2. **Spatial depiction of exceedances and corresponding exceedance magnitude.** Exceedance magnitude is defined as the difference between the shale gas scenarios and base case when an exceedance occurs, and is an indicator of the robustness of shale gas emissions on an exceedance. Exceedances are displayed as filled dots at the station locations where they occur, in µg m^{-3}, over JJA, applying the EU guideline for O_3 as the threshold (120 µg m^{-3}). For stations which experienced more than one exceedance, the maximum exceedance magnitude is shown. The top left-hand corner of each plot indicates the particular scenario, and the top right-hand corner displays the maximum exceedance magnitude value experienced over the domain and simulation period.

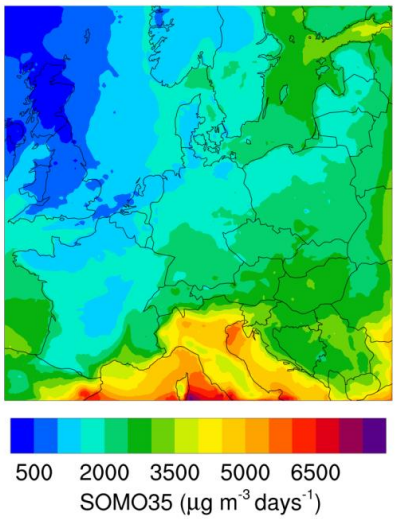

Figure S3. **Surface plot of SOMO35 (annual Sum of Ozone Means Over 35 ppb, daily maximum 8-hour) for base case simulation, in µg/m^3·days, over JJA**. SOMO35 is an indicator of accumulated O_3 exposure.

References

Cremonese L, Weger LB, Denier van der Gon HAC, Bartels M, Butler TM. 2019. Emission scenarios of a potential shale gas industry in Germany and the United Kingdom. *Elem Sci Anth* **7**(1): 18. doi:10.1525/elementa.359.

Appendix C: Supplement Paper III

Supplemental material

Expected impacts on greenhouse gas and air pollutant emissions due to a possible transition towards a hydrogen economy in German road transport

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Abbreviations

AD	activity data
CG	coal gasification
EF	emission factor
FCEV	fuel cell electric vehicle
HDV	heavy duty vehicle
HHV	higher heating value
ICEV	internal combustion engine vehicle
IEA WEB	International Energy Agency's World Energy Balances report
JEC WTT v4a	JEC Well-to-tank Report Version 4.a
LDV	light duty vehicle
LHV	lower heating value
Mop.	moped
Mot.	Motorcycle
NIR	National Inventory Report
NR	naphtha reforming
PC	passenger car
PEM	proton exchange membrane
PHEM	Passenger Car and Heavy Duty Emission Model
SMR	steam methane reforming
TE	theoretical energy
UBA	Umweltbundesamt (German Environment Agency)
TTW	tank-to-wheel
WLTP	Worldwide Harmonized Light Vehicles Test Procedure

S1 Emissions scenario model: domains and data

Total scenario emissions of GHG and air pollutants are quantified by using an emissions scenario model in Excel. Emissions are calculated by multiplying activity data [AD] (e.g., fuel consumption) with emission factors [EFs]. The model is made up of an array of input parameters and equations, which are organized into "domains" representing aspects of the hydrogen economy and road transport relevant to this study. Each domain covers a distinct area of emissions or AD quantification, and is represented by a sheet in Excel. The domains of this study include: road transportation, hydrogen economy, natural gas production, steam methane reforming, gasoline production, diesel production, coal production, coal gasification, electricity generation, electrolysis, hydrogen transport and storage, LPG production and biofuel production. Each domain, its purpose and input data are described in the sub-sections below (S1.1-S1.13). The model methodology and information flow is based on [1], in which a full description is provided, and which is published in [2].

Where possible and when applicable, data represents Germany and the year 2016 (the most recent data year when this study was carried out), though hydrogen-related technology data is generally based on present-day values. It is worth noting that when data represents Germany/Europe and/or year 2016, this is explicitly stated below; additionally, when a range of EF data was provided by a source, the default value within the range was employed unless otherwise stated. Emissions quantified include those produced from activities within and outside of Germany; explained in Section S1.14 is how values are limited to German emissions only. Finally, all energy use data is based on the lower heating value [LHV]. The LHV is more meaningful for small engines (automobiles) because the exhaust energy is not captured and water leaves as steam. It is also the European convention to use the LHV. Whether the LHV or higher heating value is used, it must be done consistently throughout the same work. E.g., in the case of LHV, the efficiency of electrolysis is lower and of the fuel cell higher, and vice-versa.

S1.1 Road transportation

Quantified here are exhaust emissions from fuel combustion and NMVOCs from gasoline evaporation. FCEVs do not have tailpipe emissions, i.e., no emissions from road transport. Also calculated here is the hydrogen demand to power the vehicle fleet (explained below in Section S1.1.1), which serves as AD to the hydrogen economy domain. All AD (fuel consumption) and EFs are from the *Umweltbundesamt* [UBA] (German Environment Agency), from [3] and [M. Kotzulla and G. Gohlisch, written communications, 2019 and 2020]. The exception to this is road transport electricity consumption (i.e., AD) which is from the International Energy Agency's World Energy Balances report [IEA WEB] 2018 [4]; EFs for electricity generation are located in Section S1.9, where associated emissions are likewise calculated. Species quantified include CH_4, CO_2, NMVOCs, NO_x, $PM_{2.5}$, PM_{10}, CO, SO_x, and NH_3. Vehicle categories include passenger cars [PCs], light duty vehicles [LDVs], trucks and buses collectively referred to here as heavy duty vehicles [HDVs], and mopeds [Mop.] and motorcycles [Mot.] collectively referred to here as two-wheelers. Fuel types include diesel, gasoline, LPG, CNG, biofuels (namely, biodiesel, bioethanol, and biogas), electricity and hydrogen. In the hydrogen scenarios, conventional fuels (i.e., diesel and gasoline) are replaced with hydrogen, while fuel consumption of alternative transport energies (i.e., electricity, LPG, CNG, and biofuels), are left unchanged because these are considered within Germany's low-carbon transition strategy for transport [5]. Following standard practice, CO_2 emissions from biofuel combustion are not counted based on the rationale that the carbon stems from atmospheric CO_2 that was absorbed

by biomass growth in the previous season. All values are representative of German road transport for the year 2016. Road transport data is extensive and therefore is not reproduced here.

Road transport emission results from the emission model used in this study were verified by comparing the baseline scenario emissions with the national emission values provided by UBA [M. Kotzulla and G. Gohlisch, written communications, 2019 and 2020]; emissions for all species were reproduced within 1% of UBA values.

S1.1.1 Tank-to-wheel efficiency

The amount of hydrogen energy needed to replace diesel and gasoline fuel in road transport is calculated based on the tank-to-wheel [TTW] efficiency of the propulsion systems (Table S1; demonstrated in Example S1). The TTW efficiency quantifies the performance of the drivetrain [6]. TTW efficiency data on internal combustion engine vehicles [ICEVs] and the hydrogen fuel cell electric vehicle [FCEV] used in this study is explained in the sub-sections below.

Powertrain	TTW efficiency					
	PC	LDV	Trucks	Buses	Mop.	Mot.
Diesel ICEV	29%	28%	33%	28%	-	-
Gasoline ICEV	23%	28%	-	-	18%	18%
Hydrogen FCEV	59%	59%	59%	59%	59%	59%

Sources: ICEV: the Institute of Internal Combustion Engines and Thermodynamics (Graz University of Technology) [publication in process by HyCentA]; FCEV: [7]

Table S1. Tank-to-wheel efficiencies based on the powertrain and vehicle category. In cases where gasoline/diesel are not relevant to the particular vehicle category in the present day (i.e., baseline scenario), values are omitted.

Example S1

Illustrated here is the calculation for determining the amount of hydrogen energy via FCEV required to replace 100 MJ gasoline via ICEV. In the first step, the amount of gasoline consumption is divided by the TTW efficiency (η) of the gasoline ICEV; this gives the "theoretical energy" [TE], defined here as the energy that would be required if η =100%. In the second step, TE is divided by the FCEV efficiency, resulting in the amount of hydrogen energy needed to replace gasoline. Both a relatively high and low ICEV efficiency are examined (where η=30% and 20%, respectively), while FCEV efficiency remains constant (η=50%); this shows the effect of varying ICEV efficiencies on final hydrogen demand, when starting fuel consumption remains constant.

Low ICEV efficiency

Step 1 $100\ MJ_{gasoline} \times 20\%\ \eta_{ICEV} = 20\ MJ_{TE}$

Step 2 $20\ MJ_{TE} \times \frac{1}{50\%\ \eta_{FCEV}} = 40\ MJ_{H2}$

High ICEV efficiency

Step 1 $100\ MJ_{gasoline} \times 30\%\ \eta_{ICEV} = 30\ MJ_{TE}$

Step 2 $30\ MJ_{TE} \times \frac{1}{50\%\ \eta_{FCEV}} = 60\ MJ_{H2}$

S1.1.1.1 Diesel and gasoline internal combustion engine vehicles

The TTW efficiency of diesel and gasoline ICEVs, for all vehicle categories, are based on simulated data and calculated using driving cycles based on real-world driving behavior provided by the Institute of Internal Combustion Engines and Thermodynamics (Graz University of Technology) [publication in process by HyCentA]. The efficiencies represent the Euro class with the greatest mileage share for Germany (year 2018 as this was the only data available) for each vehicle category (Table S2), and are based on the German vehicle average of the respective Euro class. In order to calculate the overall weighted efficiency per vehicle category, efficiencies of each individual Euro class and corresponding mileage share of the vehicle fleet would be required. Due to lack of data, these Euro class efficiencies are applied to the entire German fleet of the respective vehicle category. Nevertheless, because these more recent Euro classes have the largest share of mileage per vehicle category in the German fleet, the influence of earlier Euro classes on the overall vehicle fleet efficiency is lower since their mileage share is smaller. Additionally earlier Euro classes only have a slightly lower efficiency compared with the newer ones. It is worth noting, however, that this implies that the amount of hydrogen needed to replace diesel and gasoline in the scenarios may be slightly overestimated, resulting in a somewhat less favorable emissions outcome from hydrogen implementation than would be experienced in reality (exaggerated in Example S1 for emphasis).

Vehicle category	Euro class
PC	Euro 5
LDV	Euro 4
Trucks	Euro V
Buses	Euro V SCR
Mot.[1]	Euro 4

[1]No value was available for mopeds, thus it is not listed here.

Table S2. Euro class associated with the tank-to-wheel efficiency used in this study, per available vehicle category, for internal combustion engine vehicles.

For PCs with gasoline ICEV powertrains, a range of TTW efficiencies (22-24%) based on vehicle size was provided of which the middle value (23%) was applied in this study (Table S1). Note that for PCs with diesel ICEV powertrains, all vehicle sizes had the same TTW efficiency (29%). The LDV efficiency represents the vehicle category N1; it is only available for diesel and therefore is applied to gasoline; however the ratio of LDV fuel consumption of diesel to gasoline is approximately 95:4; hence this is not expected to have a significant impact on the results of this study. The truck efficiency is based on two values representing vehicle categories N2 and N3; it is weighted here based on the share of diesel trucks belonging to each category according to vehicle weight, for the year 2016 in Germany [8]. The bus efficiency is weighted based on the shares of city buses and coach buses; due to lack of data, it is assumed that city buses make up 90% and coach buses 10% of the German bus fleet. The motorcycle efficiency is for 4-stroke engines; no measurements are available for mopeds and therefore the motorcycle efficiency is applied.

S1.1.1.2 Hydrogen fuel cell vehicle

The FCEV TTW efficiency in this study is for the Toyota Mirai passenger vehicle and was obtained from Lohse-Busch et al. [7]. Several efficiencies were provided by [7] based on different driving cycles. The value applied in this study is based on the Worldwide Harmonized Light Vehicles Test Procedure [WLTP] driving cycle, which is representative of real driving profiles for PCs and is used by

the European Union [9]. Public data on the FCEV is especially rare; for this reason, the Toyota Mirai PC efficiency is applied to all vehicle categories in this study. Therefore, this value should be regarded as a best estimate and not the official efficiency of all FCEVs for all vehicle categories. Nevertheless, the more recent FCEV TTW efficiency used in this study (59%) is close in value to those listed in Helmers and Marx 2012 [6] (50% and 56%).

S1.1.2 Hydrogen in refinery operations

Hydrogen is used in refinery operations to transform crude oil into products such as diesel and gasoline. In order to account for the hydrogen that will be saved if diesel and gasoline are avoided, a conversion factor for refinery hydrogen is calculated and used. The conversion factor is 2.62% and represents the percent hydrogen energy required per unit of diesel/gasoline consumption in the road transport sector. The conversion factor is the product of annual global hydrogen used in refinery applications[1] [10] and the 2016 global share of road transport in oil refinery applications[2] [4], divided by the 2016 world road transport oil products energy [4]. It is assumed that LPG is found in association with natural gas at the fields; therefore the factor is not applied to LPG consumption in road transport.

S1.2 Hydrogen economy

Quantified here is the amount of hydrogen produced by technology type, which serves as AD to hydrogen production technologies. Also calculated is the maximum allowable hydrogen loss rate from FCEVs, defined here as the rate to avoid a net increase in H_2 emissions from FCEV implementation in the scenarios.

The hydrogen economy domain contains hydrogen production technology shares per application of hydrogen. The hydrogen production technologies considered are steam methane reforming [SMR], naphtha reforming [NR], coal gasification [CG] and water electrolysis. End-use applications of hydrogen in the model are hydrogen in refinery operations, and hydrogen fuel in road transport. Technology shares for refinery hydrogen are derived from the IEA's The Future of Hydrogen report [10]. According to the IEA's report [10], on average, in Europe, one-third of refinery hydrogen is produced on-site as a by-product, mostly through NR; the rest is often produced using natural gas as a feedstock. Based on this, the shares are adapted as displayed in Table S3. Shares are not shown for hydrogen fuel in road transport because these depend on the scenario pathway; note that each pathway simply has a 100% share for the technology under consideration. It is important to mention that emissions associated with NR are not quantified in the model because hydrogen is a by-product of this process; additionally, NR is not considered as a method for production of hydrogen fuel in road transport.

[1] The value for hydrogen in refinery applications represents the year 2018, as this was the only data available.
[2] The global share of road transport in refinery applications is calculated by dividing world road transport oil products energy by world oil refineries oil products energy in the IEA WEB 2018; i.e., it is assumed that hydrogen use is averaged evenly over all oil refinery applications.

Production technology	Share of refinery H_2
SMR	67%
NR	33%
CG	0%
Electrolysis	0%

Source: based on [10]

Table S3. Hydrogen technology production shares for hydrogen use in German refineries.

To calculate the maximum allowable hydrogen loss rate from FCEVs, the difference between 'conventional' hydrogen emissions (in the baseline scenario) and 'new' hydrogen emissions (in the hydrogen pathway scenarios) is divided by the total kilometers driven, based on the example from Bond et al. [11]. Hydrogen emissions are likewise based on data in Bond et al. [11] (hydrogen emissions calculations are explained in detail below). Kilometers driven by ICEVs represent Germany in 2016 [3], and are adjusted based on the share of diesel and gasoline in road transport fuel consumption. Conventional hydrogen emissions stem from fossil fuels in road transport (i.e., by incomplete combustion and released as vehicle exhaust), and from refinery operations hydrogen (i.e., by leakage). New hydrogen emissions result from road transport hydrogen fuel (i.e., by leakage, noting that only emissions upstream of transport are estimated).

Bond et al. [11] estimate annual hydrogen emissions of 4.51 and 2.33 Tg H_2 for the years 2010 and 2020 from incomplete combustion in global road transport. This implies, on average, an emissions decrease of 0.218 Tg H_2 per year from 2010 to 2020. Extrapolating this value to the year 2016 yields 3.20 Tg hydrogen; to adjust emissions to Germany, this value is multiplied by the German 2016 share of world road transport oil energy from IEA WEB 2018 [4]. This yields 85.24 Gg of conventional H_2 emissions from German road transportation for the year 2016.

A H_2 loss rate covering hydrogen production, distribution, storage and end-use (excluding transportation) is applied to hydrogen applications in this study. A value of 0.1% was chosen as a realistic value for a mature system with widespread hydrogen use, and considering that higher values would result in a significant monetary loss as described in Bond et al. [11], also noting that Schultz et al. [12] reported a H_2 loss rate in the order of 0.1% from the existing hydrogen distribution network in Germany. Due to the lack of detailed data available for loss rates, the loss rate is applied equally to all hydrogen use (i.e., in refinery applications and as fuel road transport), regardless of whether it is consumed onsite of production or requires distribution. With this loss rate the conventional hydrogen emissions from refinery applications is quantified, and new hydrogen emissions from hydrogen fuel in road transport. It is important to emphasize that hydrogen emissions from the latter source only cover losses in the hydrogen chain up until the FCEV. Indeed it is precisely the maximum allowable H_2 EF from FCEVs that is quantified here.

S1.3 Natural gas production

Emissions (CH_4, CO_2, and NMVOCs) associated with natural gas production are quantified. Natural gas energy demands stem from CNG consumption in road transport, and SMR for hydrogen production. EFs are presented in Table S4. Two sets of CH_4 EFs are considered in the SMR pathway: *ng1* (representing a standard estimate) and *ng2* (representing a higher, yet plausible rate). CH_4 EFs are divided into upstream (from the well site up to transmission and storage) and downstream (local distribution) processes of the natural gas chain. *Ng1* is adapted to Germany for the year 2016; the

CH_4 EF is based on Western and non-Western country data for source category 1.B.2.b Natural gas from the 2006 IPCC Guidelines [13], and weighted based on the 2016 share of German natural gas consumption originating from countries whose natural gas production operates according to Western and non-Western standards [14]. It was assumed that OECD countries operate based on Western standards and vice-versa. *Ng2* is from Alvarez et al. [15], who found that CH_4 leakage for a large portion of US natural gas production and distribution (approximately one third) is significantly higher (~60%) than official estimates. CO_2 EFs are based on the GMCG1 pathway from the JEC's Well-to-tank Report Version 4.a [JEC WTT v4a] [16]; this pathway represents the EU-mix natural gas supply, transported by pipeline at a distance of 2500 km (close to the approximate average distance of German natural gas sources); accordingly this pathway was selected as the most relevant for Germany. CO_2 'upstream' covers production, EU transport by pipeline, and distribution of the high pressure lines; note that no additional energy is required for downstream transport, and hence there is no 'downstream' CO_2 EF; additionally there is a CO_2 EF for compression and dispensing of CNG for vehicles at the fueling station. The NMVOC EF covers natural gas (1.B.2.b) [17]. The majority of NMVOC emissions from natural gas production occur upstream; accordingly, the EF is assumed here as upstream. CH_4 and NMVOCs are typically emitted together during natural gas production; therefore, the default NMVOC EF was assumed as the value for *ng1* and was scaled up for *ng2* based on the increase in upstream CH_4 leakage of *ng2* over *ng1*.

Species	Segment	Unit	EF	
			ng1	ng2
CH_4	Upstream	-	1.0%	2.2%
	Downstream		0.2%	0.1%
CO_2	Upstream	kg/TJ	5 400	
	CNG compression, dispensing		3 080	
NMVOCs	Upstream	kg/TJ	2.6	5.7

Sources: CH_4 (ng1): [13]; CH_4 (ng2): [15]; CO_2: [16]; NMVOCs: [17]; note that the EEA conversion factor of 38 MJ/m^3 [17] for natural gas was used to yield the value listed in the table.

Table S4. Emission factors for natural gas production. For CH_4, emission factors are expressed as a percentage of total natural gas production.

S1.4 Steam methane reforming

This segment quantifies the amount of natural gas primary energy needed for producing hydrogen by SMR and resulting emissions, based on the thermal efficiency and EFs associated with SMR. Both centralized (i.e., large-scale plant) and decentralized (i.e., fueling station) SMR production are considered. Respective thermal efficiencies are 75% and 67% based on JEC WTT v4a [16]; note that the slightly lower efficiency of decentralized production is on account of waste heat recycling not being practical at the smaller scale. The centralized production efficiency in this study falls within the range of values found in, e.g., Abánades et al. [18] (74%) and IAEA [19] (70-80%). EFs are presented in Table S5. The CH_4 EF is for source category 1.A.2 Stationary combustion in manufacturing industries and construction, Natural gas [13]. The CO_2 EF is for natural gas industry combustion, representing Germany 2016 [20]. The EFs of the other species are for source category 1.A.2.g viii Stationary combustion in manufacturing industries and construction: other production, Natural gas, for Germany [21].

Species	EF [kg/TJ]
CH_4	1
CO_2	55 800
NO_x	41.2
$PM_{2.5}$	0.2
PM_{10}	0.2
CO	10.0
SO_x	0.1

Sources: CH_4: [13]; CO_2: [20]; all other species: [21]

Table S5. Emission factors for steam methane reforming.

S1.5 Gasoline production

Emissions (CH_4, CO_2, and NMVOCs) associated with gasoline production are quantified. The gasoline energy requirement is based on consumption thereof in road transport. EFs are presented in Table S6. CO_2 and CH_4 EFs are based on the COG1 pathway from JEC WTT v4a [16]; this pathway represents crude oil from typical EU supply, transport by sea, refining in the EU, typical EU distribution and retail; it is the only gasoline pathway provided by the source. 'Upstream' covers crude oil production and transport; 'downstream' covers refining, distribution and dispensing at the retail site. For the NMVOC EF 'upstream' covers oil exploration, production, transport (1.B.2.a.i-iii) [17]; the 'downstream' EF for NMVOCs and other pollutants covers oil refining and storage (1.B.2.a.iv) [22]; note that NMVOC evaporation losses from distribution are already included in the CO_2 EF via degradation of NMVOCs into CO_2 in the atmosphere, and therefore not included in the downstream NMVOC EF to avoid double counting.

Species	Segment	EF [kg/TJ]
CH_4	Upstream	26
	Downstream	2
CO_2	Upstream	4 890
	Downstream	8 160
NMVOCs	Upstream	4.8
	Downstream	4.8
NO_x	Downstream	5.7
$PM_{2.5}$	Downstream	0.1
PM_{10}	Downstream	0.2
CO	Downstream	2.1
SO_x	Downstream	14.8
NH_3	Downstream	2.62E-2

Sources: CH_4 and CO_2: [16]; NMVOCs (upstream): [17]; NMVOCs (downstream) and other species: [22]; note that the EEA conversion factor of 42 GJ/Mg [17] for oil was used to yield the value listed in the table.

Table S6. Emission factors for gasoline production.

S1.6 Diesel production

Emissions (CH_4, CO_2, and NMVOCs) associated with diesel production are quantified. The diesel energy requirement is based on consumption thereof in road transport. EFs are presented in Table S7. CO_2 and CH_4 EFs are based on the COD1 pathway from JEC WTT v4a [16]; this pathway represents crude oil from typical EU supply, transport by sea, refining in the EU, typical EU distribution and retail; it is the only diesel pathway provided by the source. 'Upstream' covers crude oil production and transport; 'downstream' covers refining, distribution and dispensing at the retail site. For the NMVOC EF 'upstream' covers oil exploration, production, transport (1.B.2.a.i-iii) [17]; NMVOC

'downstream' is split into oil refining and storage (1.B.2.a.iv) [22], and distribution of diesel fuels (1.B.2.a.v) [23]. For the other pollutants, 'downstream' covers oil refining and storage (1.B.2.a.iv) [22].

Species	Segment	EF [kg/TJ]
CH_4	Upstream	27
	Downstream	2
CO_2	Upstream	4 970
	Downstream	9 650
NMVOCs	Upstream	4.8
	Downstream: refining and storage	4.8
	Downstream: diesel distribution	2.6
NO_x	Downstream	5.7
$PM_{2.5}$	Downstream	0.1
PM_{10}	Downstream	0.2
CO	Downstream	2.1
SO_x	Downstream	14.8
NH_3	Downstream	2.62E-2

Sources: CH_4 and CO_2: [16]; NMVOCs (upstream): [17]; NMVOCs (downstream refining and storage) and other species: [22]; NMVOCs (downstream distribution): [23]; note that the EEA conversion factor of 42 GJ/Mg [17] for oil was used to yield the value listed in the table).

Table S7. Emission factors for diesel production.

S1.7 Coal production

Emissions (CH_4, CO_2, NMVOCs and PM) associated with coal production are quantified. The coal energy requirement is based on CG for hydrogen production. EFs are presented in Table S8. The CH_4 EF covers mining, and is divided into underground and surface mining. It is assumed that hard coal is mined exclusively via underground mining and brown coal exclusively via surface mining based on the German National Inventory Report [NIR] [23]. The underground mining EF represents a global average taken from the IPCC 2006 Guidelines [13], since the overwhelming majority of German hard coal consumption in 2016 was imported [24]; the EF covers mining and post-mining. The surface mining EF represents lignite mining in Germany taken from the German NIR [23], since virtually all brown coal is produced domestically [24]; the EF covers extraction, though not storage since extracted brown coal is used directly, i.e., "mine-mouth". The CO_2 EF is based on the energy associated with the production of hard coal for the European coal mix [25]; note that in the same source emissions associated with coal transportation are listed as not applicable. The CO_2 EF for lignite is assumed here to be 20% of the value for hard coal, on account of surface mining being less emissions-intense than underground mining. The NMVOCs, $PM_{2.5}$ and PM_{10} EFs cover coal mining and handling (1.B.1.a) [26]. Note that the NMVOC EF is highly uncertain and is based on CH_4 leakage; because CH_4 emissions are significantly lower for surface (brown coal) mining compared with underground, the default NMVOC EF was assumed as the value for hard coal, and was scaled down for brown coal based on the fraction of brown coal CH_4 emissions compared with hard coal CH_4 emissions.

Species	Segment	Unit	EF[1]
CH_4	Underground mining	m^3/t	20.5
	Surface mining		0.016
CO_2	Hard coal production	kg/TJ	6 270
	Brown coal production		1 250
NMVOCs	Hard coal mining and handling	kg/t	0.8
	Brown coal mining and handling		6.24E-4
$PM_{2.5}$	Coal mining and handling		0.005
PM_{10}			0.042

Sources: CH_4 (underground mining): [13]; CH_4 (surface mining): [23]; CO_2: [25]; NMVOCs, $PM_{2.5}$ and PM_{10}: [26]

Table S8. Emission factors for coal production.

S1.8 Coal gasification

This segment quantifies the amount of coal primary energy needed for producing hydrogen by CG and resulting emissions, based on the thermal efficiency and EFs associated with CG. The thermal efficiency for coal gasification is 50.8% from [16]. EFs are presented in Table S9. EFs are weighted based on German primary energy consumption of lignite and hard coal, for the year 2016 [24], except for CH_4 as EF values are identical for lignite and hard coal. The CH_4 EF is for source category 1.A.2 Stationary combustion in manufacturing industries and construction [13]. The CO_2 EF is for industry combustion, representing Germany 2016 [20]. The EFs of the other species are for source category 1.A.2.g viii Stationary combustion in manufacturing industries and construction: other production, for Germany [21].

Species	EF [kg/TJ]
CH_4	10
CO_2	99 300
NO_x	101
$PM_{2.5}$	4.3
PM_{10}	4.9
CO	36.6
SO_x	183

Sources: CH_4: [13]; CO_2: [20]; all other species: [21]

Table S9. Emission factors for coal gasification.

S1.9 Electricity generation

Emissions associated with electricity generation are quantified using electricity consumption as AD. Electricity consumption stems from electrolysis, hydrogen transport and storage, and road transport (for BEVs). Electricity generation EFs are considered for two types of electricity supplies: the current (2016) electricity mix [27] and renewables only. EFs for the current mix are presented in Table S10, and the percent contribution of energy carriers for generation of the 2016 German mix [24] in Table S11. For the PM EF of the current mix, only a value was provided for PM_{10}; the value for $PM_{2.5}$ was calculated here based on the estimated fraction of $PM_{2.5}$ in PM_{10} for gaseous, liquid and solid fuels in public electricity production (1.A.1.a) [28], and the primary energy share of each of these fuels in 2016 German electricity generation [24]. EFs for the renewable mix are zero assuming wind and photovoltaics as the primary sources of renewable electricity. Note that electricity consumption for BEVs assumes the current (2016) electricity mix only in all scenarios; this is because alternative fuel consumption from 2016 German road transport is left unaltered in the study.

Species	EF [g/kWh]
CH_4	0.184
CO_2	516
NMVOCs	0.017
NO_2	0.44
$PM_{2.5}$	0.014
PM_{10}	0.015
CO	0.23
SO_2	0.29

Source: [27]; PM2.5 adapted based on [28]

Table S10. Emission factors for electricity generation for the year 2016 German electricity mix.

Energy carrier	%
Hard coal	19
Brown coal	28
Oil	1
Gas	13
Of that: natural gas	*10*
Renewables	19
Other	2
Electricity	1
Nuclear	19

Source: [24]

Table S11. Percent contribution of energy carriers to 2016 German electricity generation.

EFs of the current electricity mix cover direct emissions (i.e., fuel combustion) only. Upstream emissions from production of the energy carriers in German electricity generation are not included in the scenarios, reason being that data is not readily available for all energy carriers. However, because this type of data is already included in the present work for coal and natural gas production, upstream emissions are calculated from the contribution of these carriers only in order to understand the relative contribution and importance of upstream emissions to total emissions under scenario *Elec-ef1-C*, chosen as an example of the *Elec* scenario set in which electricity-based emissions play a considerable role in total CO_2eq emissions. To quantify upstream emissions, first the primary energy input per fuel type (i.e., coal and natural gas) for electricity generation is calculated based on data provided by AGEB 2019 [24] and the total net electricity generation in Germany for 2016 from BMWi 2019 [29]; then the primary energy input of coal and natural gas are multiplied by their respective production EFs. Based on these calculations, upstream coal and natural gas emissions increase total scenario CO_2eq emissions by 8.0%, using the 100-year global warming potential for CH_4 of 25 from IPCC AR4 following the German NIR and the Revised UNFCCC Reporting Guidelines [23]. However if upstream electricity emissions are considered for Germany only, they have less of an influence (<1% increase in German scenario CO_2eq emissions) since most natural gas and hard coal are imported. Considering that coal and natural gas make up 57% of primary energy input for electricity generation and represent 95% of fossil fuels used in electricity generation, this represents the majority of all upstream emissions [24]. Accordingly, upstream electricity emissions would increase overall CO_2eq emissions, but have a small contribution relative to those from fuel combustion in electricity generation, and a small impact on German CO_2eq emissions.

S1.10 Electrolysis

This segment quantifies the electricity consumption needed for producing hydrogen by low temperature water electrolysis (Table S12) [30]. Two electricity consumptions are used in the electrolysis pathway, *ef1* and *ef2*, which represent values for the years 2020 and 2030, respectively; note that these values were chosen because we are interested in looking into the potential of hydrogen technology (presently and) in the future. For 2030 it is assumed that some excess heat from PEM is recycled, which results in lower electricity consumption per kg of hydrogen produced. The electricity consumption covers all auxiliaries of the system and represents proton exchange membrane [PEM] electrolysis; however the efficiency for alkaline electrolysis is roughly the same as PEM.

	Unit	ef1	ef2
Electricity consumption	kWh/kg_{H2}	56	47
Efficiency LHV	-	59%	71%
Efficiency HHV	-	70%	84%

Source: [30]; note that the LHV value is used in this study and the corresponding higher heating value [HHV] is presented here for informational purposes only.

Table S12. Electricity consumption and efficiency for hydrogen production via water electrolysis.

S1.11 Hydrogen transport and storage

This segment quantifies the energy requirements for delivery, compression and pre-cooling of both centralized (i.e., offsite/large-scale plant) and decentralized (i.e., onsite/ fueling station) hydrogen production. The data was obtained from Schmidt et al. [30] and is displayed in Table S13. Energy is afforded by the electricity grid, and all values are for the year 2020. Hydrogen delivery via pipeline is assumed, based on the IEA's recent report The Future of Hydrogen [10]; this decision is also supported by a recent vision set forth by the German transmission system operators for a nationwide hydrogen network with a length of approximately 6000 km [31]. The energy requirement for pipeline delivery represents an average value derived from a modelled hydrogen pipeline grid for Germany. Due to hydrogen's low volumetric energy density under ambient conditions, it is necessary to increase it by, e.g., compression or liquefaction. Compression is the most well-established and prevailing method for hydrogen storage [32] with a standard tank pressure of 700 bar [30, 33]; accordingly this type of storage is assumed here. Note that for decentralized production, the temperature of the hydrogen leaving the electrolysis or SMR plant is higher than that leaving the pipeline; because a higher temperature of the gas requires more energy for compression, the electricity consumption is slightly greater for decentralized hydrogen. Compressing gas increases its temperature, and for this reason the hydrogen must be cooled prior to fueling to avoid the tank from overheating.

Segment	Unit	Electricity consumption	
		Decentralized	Centralized
Pipeline delivery	kWh/kWh_{H2}	0.000	0.018
Compression	kWh/kWh_{CGH2}	0.108	0.105
Pre-Cooling	kWh/kWh_{CGH2}	0.090	0.090

Source: [30]

Table S13. Electricity consumption for hydrogen delivery, compression and pre-cooling, for decentralized (onsite) and centralized (offsite) hydrogen production.

S1.12 LPG production

Emissions (CH_4, CO_2, and NMVOCs) associated with LPG production are quantified. The LPG energy requirement is based on consumption thereof in road transport. EFs are presented in Table S14. CO_2 and CH_4 EFs are based on the LRLP1 pathway from JEC WTT v4a [16]; this pathway represents LPG from a remote natural gas field, purification and liquefaction at source, long-distance transport by sea, distribution by road and retail; it is the only LPG pathway provided by the source. 'Upstream' covers LPG production, purification and liquefaction, and transport; 'downstream' covers distribution and dispensing at the retail site. The NMVOC EF covers natural gas (1.B.2.b) [17]; because the majority of NMVOC emissions from natural gas production occur upstream, the EF is assumed here as upstream.

Species	Segment	EF [kg/TJ]
CH_4	Upstream	16
	Downstream	2
CO_2	Upstream	5 810
	Downstream	1 780
NMVOCs	Upstream	2.6

Sources: CO_2 and CH_4: [16]; NMVOCs: [17]; note that the EEA conversion factor of 38 MJ/m^3 [17] for natural gas was used to yield the value listed in the table.

Table S14. Emission factors for LPG production.

S1.13 Biofuel production

Emissions (CH_4 and CO_2) associated with biofuel production are quantified. The biofuel energy requirement is based on consumption thereof in road transport. Energy requirements are examined for the production of each biofuel type relevant to road transport: biodiesel, bioethanol and biogas. EFs are presented in Table S15. EFs are based on biofuel production pathways from JEC WTT v4a [16], and generally cover crop cultivation, biofuel production, transport, distribution and retail. Note that the biofuel pathways do not include emissions associated with direct or indirect land use changes, as these currently cannot be estimated with confidence; nevertheless this is not expected to have a significant impact on the results. The specific pathway(s) chosen for each biofuel type are based on the feedstock contribution per biofuel type in Germany for the year 2016 [34], and taking actual practice into consideration. In some cases, feedstocks contributing to production of a particular biofuel in Germany were not available as pathways and thus were adapted. Additionally, some feedstocks with an insignificant contribution to German biofuel production were excluded. Therefore these EFs should be regarded as a best estimate. Nevertheless, biofuels have a relatively minor contribution to road transport energy (approximately a 4% share in German road transport fuel consumption for the year 2016); furthermore biofuel consumption is not being changed in the scenarios, nor are biofuels a focus of the work. For these reasons, it is not expected that the biofuel EFs have a negative impact on the overall results of this study. Adaptations and production pathways used to calculate the biofuel EFs are explained below. EFs of the other species considered in this work were not available and therefore are not included here.

Species	EF [kg/TJ]		
	Biodiesel	Bioethanol	Biogas
CH_4	134	112	37
CO_2	20 080	50 710	14 920

Sources: adapted using data from [16] and [34].

Table S15. Emission factors for biofuel production, based on German 2016 feedstocks.

S1.13.1 Biodiesel

German biodiesel – also known as fatty acid methyl ester (FAME) – was facilitated in 2016 through the following feedstocks, by share: waste (44%), palm oil (13%), rapeseed (43%), soy (<1%) and sunflowers (<1%) [34]. On account of the minor contribution of the latter two feedstocks, they are excluded here. The biodiesel EF in this study is weighted based on the proportional feedstock contribution, using the following JEC feedstock pathways for FAME production: WOFA3 (waste cooking oil), POFA3 (palm oil), and ROFA1/2/3 (rapeseed) [16]. Note that for POFA3, three variants (a-c) are provided, of which the average is used. ROFA has 5 options (1-5): an average based only on ROFA1/2/3 is applied, as these pathways reflect actual practice in which the rapeseed meal by-product is used for animal feed rather than for energy production (as assumed in ROFA4); ROFA5 covers hydrogen production and is not included here as hydrogen production is assessed elsewhere in the model. Only one pathway is provided for WOFA3.

S1.13.2 Bioethanol

German 2016 bioethanol was facilitated through the following feedstocks, by share: waste (<1%), barley (5%), maize (33%), rye (7%), triticale (8%), wheat (32%), sugar cane (8%), and sugar beet (7%) [34]. Due to the minor contribution of waste, it is excluded here. The bioethanol EF in this study is weighted based on the proportional feedstock contribution, using the following JEC feedstock pathways for bioethanol production: BRET2 (barley/rye), CRET2 (maize), WTET1 and WTET3 (wheat), SCET1 (sugar cane), and SBET1 (sugar beet) [16]. Note that several pathways based on wheat feedstock are provided; the average of pathways WTET1 and WTET3 are assumed here: WTET1 assumes the production process is facilitated by a conventional natural gas boiler (representative of the great majority of European installations and is also the least expensive option), and WTET3 by a lignite boiler with combined heat and power (prevalent in Eastern Germany). Additionally, two variants (a, b) are provided for these pathways; here WTET1a/3a are assumed as reflective of actual practices, in which DDGS (distiller's dried grain with solubles) – a by-product of ethanol production – is used for animal feed rather than for electricity generation. For pathway SBET1, three variants (a-c) are provided; assumed here is the average of SBET1a/b in which the pulp co-product is used for animal feed. Only one pathway is provided for the other feedstock pathways. Since feedstock pathways were not provided separately for barley and rye, BRET2 is applied to both. A pathway based on triticale – a hybrid crop of rye and wheat – was also not available; for this reason 50% of the triticale contribution is attributed to the BRET2 pathway and 50% to WTET1. The resulting contribution of each pathway to the weighted bioethanol EF used in this study is displayed in Table S16.

Pathway	Contribution
BRET2	15%
CRET2	33%
WTET1a/3a	36%
SCET	8%
SBET1a/b	7%

Sources: adapted using data from [16] and [34].

Table S16. Contribution of individual feedstock production pathway emission factors to the weighted total bioethanol emission factors used in this study, based on the feedstocks used for German bioethanol production in the year 2016.

S1.13.3 Biogas

German 2016 biogas was produced exclusively from waste, the vast majority of which was incurred from the distillation of fermented fruit, cereal and potato mash [34]. Accordingly the biogas EF in this study is solely based on the pathway in which (organic) waste serves as the feedstock in biogas production (OWCG1) [16].

S1.14 German emissions

In order to examine German emissions specifically, all emissions released from activities outside of Germany are excluded. These emissions stem from upstream activities of imported fossil fuels. To do this, the share of the fossil fuel produced in Germany [24] is multiplied by upstream emissions for that particular fossil fuel. For example, about 7% of German natural gas consumption was produced domestically in 2016; therefore, total upstream natural gas emissions are multiplied by this share. This approach assumes that the 2016 share of domestic production remains constant for simplification. Note that all coal production-related emissions are upstream. Additionally note that all biofuel emissions were allocated to Germany due to lack of data; nevertheless, biofuel emissions are unaltered in the scenarios.

S2 Sensitivity analysis

Data on vehicle efficiencies for FCEVs is rare; for this reason a vehicle efficiency based on PCs is applied to all vehicle categories (described in Section S1.1.1.2). In order to assess the impact of this on total emission results, a sensitivity analysis was performed by varying the FCEV efficiency for each vehicle category individually. Note that the *SMR-ng1-C* scenario was used as the basis. Lower (50%) and upper (65%) FCEV TTW efficiencies were chosen based on values from Helmers and Marx [6] and Lohse-Busch et al. [7], respectively. The impact on total CO_2eq emissions ranges from +10% and -5% based on the lower- and upper-end efficiencies, respectively (Table S17). The vehicle categories experiencing the highest to lowest sensitivity from varying this parameter are, in order, PCs, HDVs, LDVs, and two-wheelers. This is because PCs have the greatest share of total gasoline and diesel consumption in road transport, followed by HDVs, LDVs, and two-wheelers (the link between TTW efficiency and fuel consumption, and in effect emissions, is illustrated in Example S1). In conclusion, the sensitivity of other vehicle categories to FCEV efficiency is relatively low, which improves the confidence in our results.

Vehicle category	Variation[a]	CO_2eq	NMVOCs	NO_x	$PM_{2.5}$	PM_{10}	CO	SO_x
PC	Min.	10%	6%	8%	8%	8%	4%	10%
	Max.	-5%	-3%	-4%	-4%	-4%	-2%	-5%
LDV	Min.	1%	0%	1%	1%	1%	0%	1%
	Max.	0%	0%	0%	0%	0%	0%	0%
HDV	Min.	6%	4%	5%	5%	5%	3%	7%
	Max.	-3%	-2%	-3%	-3%	-3%	-1%	-3%
Two-wheelers	Min.	0%	0%	0%	0%	0%	0%	0%
	Max.	0%	0%	0%	0%	0%	0%	0%

[a]TTW efficiency: employed, i.e., value used in study = 59%; minimum = 50%; maximum= 65%.

Table S17. Impact of FCEV TTW variations, per vehicle category, on total emissions, in %.

S3 Total air pollutant emissions

Presented here are total scenario air pollutant emissions, i.e., those released within and outside of Germany (German values are provided in the main paper).

Scenario	NMVOCs		NO_x		$PM_{2.5}$		PM_{10}		CO		SO_x		NH_3	
Baseline	120.9		444.6		7.9		8.1		724.8		32.0		11.8	
	Δ_{Abs}[a]	%	Δ_{Abs}	%	Δ_{Abs}	%	Δ_{Abs}	%	Δ_{Abs}	%	Δ_{Abs}	%	Δ_{Abs}	%
SMR-ng1-C	-112.7	*-93*	-341.4	*-77*	-6.44	*-81*	-6.6	*-81*	-658.1	*-91*	-14.6	*-46*	-11.0	*-93*
SMR-ng1-D	-112.3	*-93*	-336.7	*-76*	-6.46	*-81*	-6.6	*-81*	-657.4	*-91*	-15.8	*-50*	-11.0	*-93*
SMR-ng2-C	-108.5	*-90*	-341.4	*-77*	-6.44	*-81*	-6.6	*-81*	-658.1	*-91*	-14.6	*-46*	-11.0	*-93*
SMR-ng2-D	-107.7	*-89*	-336.7	*-76*	-6.46	*-81*	-6.6	*-81*	-657.4	*-91*	-15.8	*-50*	-11.0	*-93*
Elec-ef1-cmx-C	-108.2	*-89*	-191.2	*-43*	-0.18	*-2*	0.1	*2*	-564.2	*-78*	120.4	*376*	-11.0	*-93*
Elec-ef1-cmx-D	-108.3	*-90*	-193.0	*-43*	-0.24	*-3*	0.1	*1*	-565.1	*-78*	119.2	*373*	-11.0	*-93*
Elec-ef2-cmx-C	-109.5	*-91*	-224.2	*-50*	-1.23	*-15*	-1.0	*-12*	-581.4	*-80*	98.7	*309*	-11.0	*-93*
Elec-ef2-cmx-D	-109.6	*-91*	-226.0	*-51*	-1.29	*-16*	-1.0	*-13*	-582.3	*-80*	97.5	*305*	-11.0	*-93*
Elec-renewable	-117.2	*-97*	-422.3	*-95*	-7.53	*-95*	-7.7	*-95*	-685.0	*-95*	-31.9	*-100*	-11.0	*-93*
CG	-93.3	*-77*	-197.6	*-44*	2.29	*29*	7.2	*89*	-599.3	*-83*	345.8	*1081*	-11.0	*-93*
SMR-ng1-C_HDV	-10.8	*-9*	-110.5	*-25*	-1.91	*-24*	-2.0	*-24*	-39.3	*-5*	-3.8	*-12*	-0.2	*-2*
Elec-renewable _HDV	-12.5	*-10*	-140.5	*-32*	-2.31	*-29*	-2.4	*-29*	-49.1	*-7*	-10.2	*-32*	-0.2	*-2*

[a]The change in absolute emissions from the scenario relative to the baseline

Table S18. Absolute and percent changes in total air pollutant emissions from scenarios relative to the baseline, in units of kt, for the year 2016. Baseline total air pollutant emissions are displayed at the top of the table, in units of kt.

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Declaration of Authorship

I, Lindsey Blaisdell Weger Coenen, declare that this dissertation entitled "Exploring potential impacts from transitions in German and European energy on GHG and air pollutant emissions and on ozone air quality" and the work presented in the thesis are my own. I prepared this dissertation without any illegal assistance. I confirm that the work is original except where it is mentioned by the reference in the text and no part of the dissertation has been submitted for any other degree. This dissertation has not been submitted to any other university for examination, neither in Germany nor in any other country.

Lindsey Weger Coenen

Potsdam, September 2020