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Key Points:

- The atmospheric methane burden is rising fast; this growth is an increasing threat to the Paris Agreement of the UN Framework Convention on Climate Change (UNFCCC)
- New gas measurement technologies, on the ground and in the air, have greatly improved our ability to locate and quantify emissions and to identify mitigation targets
- Emissions can be cut by ending fossil fuel emissions, cutting biomass burning, improving landfills, especially in the tropics, and changing cattle farming practice

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Methane Mitigation: Methods to Reduce Emissions, on the Path to the Paris Agreement

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Abstract The atmospheric methane burden is increasing rapidly, contrary to pathways compatible with the goals of the 2015 United Nations Framework Convention on Climate Change Paris Agreement. Urgent action is required to bring methane back to a pathway more in line with the Paris goals. Emission reduction from “tractable” (easier to mitigate) anthropogenic sources such as the fossil fuel industries and landfills is being much facilitated by technical advances in the past decade, which have radically improved our ability to locate, identify, quantify, and reduce emissions. Measures to reduce emissions from “intractable” (harder to mitigate) anthropogenic sources such as agriculture and biomass burning have received less attention and are also becoming more feasible, including removal from elevated-methane ambient air near to sources. The wider effort to use microbiological and dietary intervention to reduce emissions from cattle (and humans) is not addressed in detail in this essentially geophysical review. Though they cannot replace the need to reach “net-zero” emissions of CO₂, significant reductions in the methane burden will ease the timescales needed to reach required CO₂ reduction targets for any particular future temperature limit. There is no single magic bullet, but implementation of a wide array of mitigation and emission reduction strategies could substantially cut the global methane burden, at a cost that is relatively low compared to the parallel and necessary measures to reduce CO₂, and thereby reduce the atmospheric methane burden back toward pathways consistent with the goals of the Paris Agreement.

Plain Language Summary Methane is a powerful climate warmer, and the amount of methane in the air is growing rapidly. Reducing human-caused methane emissions is urgent if the 2015 United Nations Paris Agreement to limit climate warming is to succeed. There is hope, though the problem of methane mitigation is very wide and complex. Much of the task is in finding, identifying, and quantifying emissions. Rapid technical advances are making it much easier to locate and thus cut emissions from fossil fuel industries (gas, coal, and oil). Assessing emissions from landfill and sewage facilities is also becoming easier. In particular, poorly regulated landfills in fast-growing tropical megacities need attention. Agricultural emissions are less tractable but may also be reduced to some extent, especially by improving manure management. Many methane mitigation options offer cost-effective approaches to cut global warming and bring the amount of methane in the air back to a pathway that is consistent with the aims of the Paris Agreement.

1. Introduction

1.1. Methane's Post-2007 Rise and Growing Distance From the Paris Target

Methane is a remarkably attractive target for reducing atmospheric greenhouse warming. It is the second-most important anthropogenic greenhouse gas, and a major contributor to the increase in tropospheric ozone, also a major greenhouse gas. Methane has a 100-yr global warming potential of 32 compared to

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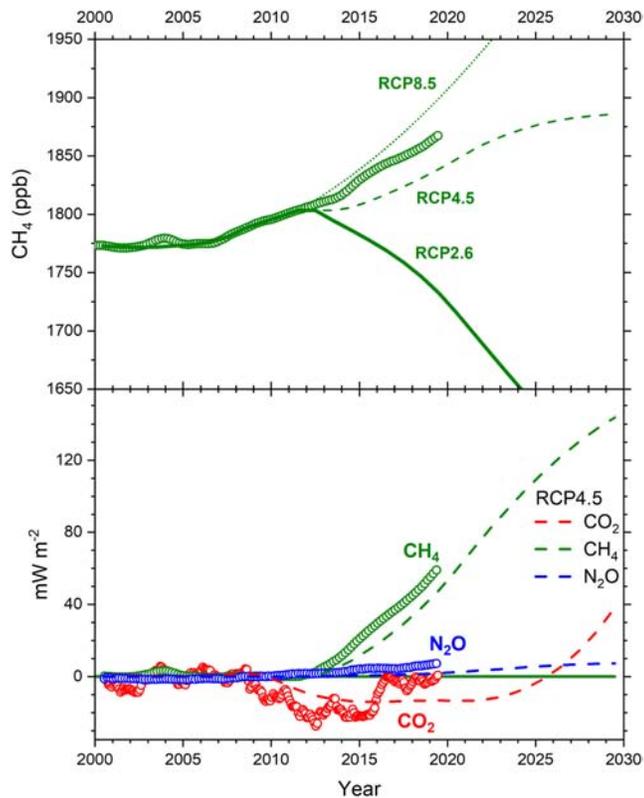


Figure 1. Methane and the Paris Agreement. Top panel shows evolution of the mean global atmospheric mixing ratio (green open circles) compared to Representative Concentration Pathway RCP2.6 (Collins et al., 2013) as used in the last IPCC Assessment and consistent with the Paris Agreement. RCP2.6 concentrations peaked in 2012, whereas those for the RCP 4.5 (dashed) pathway peak around 2045, while RCP 8.5 (dotted) mole fractions continue to rise throughout the 21st century. Lower panel shows differences in radiative forcing from the RCP2.6 pathway and actual evolution of methane (green open circles), CO₂ (red circles), and nitrous oxide (blue circles). Dashed lines show RCP 4.5 relative to RCP 2.6. Radiative forcing used here is from Etminan et al. (2016), and the figure is updated from that in Nisbet et al. (2019) using NOAA mole fraction data to June 2019.

the atmospheric methane mixing ratio was over 100 ppb above the RCP 2.6 path (Nisbet et al., 2019). If added to the atmosphere as a single pulse, this would equate to an input of about 300 Tg of methane (1 ppb is globally equivalent to about 2.8 Tg CH₄).

The amount of methane in the atmosphere above preindustrial levels directly affects the allowable cumulative carbon emissions budget (i.e., the total anthropogenic CO₂ emitted before reaching a temperature threshold, usually 1.5 or 2 °C). The rise thus has major implications for the UN Paris Agreement's goal to limit climate warming to 2 °C and especially to the 2018 Intergovernmental Panel on Climate Change (IPCC) advice to limit warming to 1.5 °C (IPCC, 2018). The global warming consequences of this rise, relative to the Paris-compliant path, are shown in Figure 1 from Nisbet et al. (2019). Moreover, the unexpected growth in methane has already significantly negated the expected impact of progress in controlling CO₂ emissions (Nisbet et al., 2019). Hausteine et al. (2017) suggest that methane is contributing to the accelerating global warming rate at a time when CO₂ emissions may be stabilizing. If growth in the methane burden continues at current rates in the coming decades, it will become impossible to meet the Paris target, especially in view of the recent upward reevaluation of the global warming potential of methane (Etminan et al., 2016).

The Paris Agreement, drafted in 2015 and effective in 2016, is clear in its call for better technology to reduce methane emissions. Article 10.2 and 10.5 of the agreement (UNFCCC, 2015) states that

CO₂ (Etminan et al., 2016); thus, removing 1 Mg (a ton) of methane has real impact (Collins et al., 2018). Moreover, with an average tropospheric lifetime of about 9–10 yr (Dlugokencky et al., 2011), successful moves to reduce methane emissions can have rapid effectiveness.

Since 2007 the atmospheric methane burden has risen sharply. It began growing by about 6 ppb/yr in 2007, after a sustained pause at the start of the millennium. The growth rate accelerated in 2014 and is currently around 10 ppb/yr (Nisbet et al., 2014; Nisbet et al., 2016; Nisbet et al., 2019). Note that the words “concentration” and “level,” terms derived from wet chemistry, are often used in popular media when discussing methane. The gas-specific terms “mole fraction” and “mixing ratio” are preferred for use in this report.

The rise in atmospheric methane has been accompanied by a significant shift in the C-isotope ratio of atmospheric methane to values more depleted in ¹³C (Nisbet et al., 2016, 2019). The post-2007 growth and isotopic shift has now been sustained for over a decade, but the reasons for the change are still not well understood. Possible explanations for this growth and the concurrent isotopic shift (for isotopic values of sources, see Sherwood et al., 2017); may include increases in biogenic emissions (especially in the tropics and subtropics), changes in the chemical sinks of methane by atmospheric OH or Cl, increased fossil fuel emissions coupled with declining biomass burning (Nisbet et al., 2019, 2016; Schaefer et al., 2016; Schwietzke, Sherwood et al., 2017; Turner et al., 2017, 2019; Rigby et al., 2017; Worden et al., 2017), more oxidation of methane by methanotrophy in forest soils (Ni & Groffman, 2018), or, more likely, some combination of all these factors.

One illustrative pathway that would lead to compliance with the 2015 United Nations (UN) Paris Agreement of the United Nations Framework Convention on Climate Change (UNFCCC, 2015) is Representative Concentration Pathway 2.6 (RCP 2.6) (Collins et al., 2013; Rogelj et al., 2012) (Figure 1). This pathway envisaged an immediate and significant fall in methane, allowing time for progress on the more difficult task of reducing CO₂. This is not a unique pathway to achievement of the Paris goals, but being Paris compliant is thus commonly cited as an exemplar of the reduction pathways needed. Yet, by 2019, as a result of the unexpected rise,

“Parties, noting the importance of technology for the implementation of mitigation and adaptation actions under this Agreement and recognizing existing technology deployment and dissemination efforts, shall strengthen cooperative action on technology development and transfer.” And that “Accelerating, encouraging and enabling innovation is critical for an effective, long-term global response to climate change and promoting economic growth and sustainable development.”

In Article 13.7, the Agreement places requirements on quantification of emissions:

“Each Party shall regularly provide the following information:

(a) A national inventory report of anthropogenic emissions by sources and removals by sinks of greenhouse gases, prepared using good practice methodologies accepted by the Intergovernmental Panel on Climate Change and agreed upon by the Conference of the Parties serving as the meeting of the Parties to this Agreement; and

(b) Information necessary to track progress made in implementing and achieving its nationally determined contribution under Article 4.”

There is thus a requirement for signatory nations to act effectively and urgently to reduce methane emissions.

1.2. The Challenge—Low-Hanging Fruit or Tough Target?

Cutting methane emissions has long been identified as a low-hanging fruit for climate action (Hansen et al., 2000; UNEP, 2018). Shindell et al. (2017) considered as much as 110 Tg CH₄ per year of abatement was possible by scaling up existing technology and industrial best practice and policy options, to provide societal benefits that outweigh implementation cost. A significant cut in anthropogenic methane emissions would be sufficient to reduce the overall methane burden within decades or less, to thus return to the RCP2.6 pathway. There is wide benefit in reducing methane emissions (Boucher & Folberth, 2010) including major reduction opportunities at less than zero cost (Warner et al., 2015).

Globally, the rate of growth in fossil fuel methane emissions may be slowing (Schaefer et al., 2016). Nisbet et al. (2016, 2019) used isotopic evidence to infer that fossil fuel emissions have declined as a proportion of the total methane budget, though they could not rule out an increase in absolute terms. But it is also possible (within the uncertainties of the observations) that a decline in biomass burning may be masking a rise in fossil fuel emissions (Worden et al., 2017).

Many jurisdictions have brought in legislation to control methane emissions (Iacobuta et al., 2018). In 2019, the Net Zero report of the U.K.’s Committee on Climate Change led to a national commitment to reduce net national greenhouse emissions to zero by 2050 (UK CCC, 2019). New Zealand has passed a similar promise. Though these commitments focus on CO₂, most nations also intend to reduce methane emissions in addition to cutting CO₂ emissions. The focus has been on point sources where very high mole fractions of methane can be measured in the nearby air—such as gas leaks, or gas emissions around landfills. Such sources can typically be detected easily, precisely located and then mitigated.

For example, Canada has committed to reduce methane emissions from the oil and gas sector by 40–45% by 2025 relative to 2012 levels (Canada, 2016). To take another example, in the United Kingdom where landfill emissions have been vigorously reduced, the national atmospheric emissions inventory suggests methane emissions have dropped by 61% since 1990 (UK NAEI, 2019; UK NIR, 2019), a pattern seen in many developed nations. In these specific sectors, there has been much progress in abatement of emissions, at relatively low cost by improved industrial practices to detect and prevent leaks, or to capture and use methane before it is emitted to atmosphere. But for many sources with more diffuse emissions that are less easily detected (as local air has low incremental methane over background), there has been little progress.

A challenge to policy-making is that there is a major discrepancy (Leip et al., 2018; Saunio et al., 2016, 2019) between “top-down” estimates (Nisbet & Weiss, 2010) of the annual global methane emission, assessed from measuring the atmosphere, and “bottom-up” totals summing emissions estimates based on national data (e.g., number of cows and area of rice fields): the “bottom-up” numbers are typically much higher (e.g. EDGAR- European Commission, Joint Research Centre / Netherlands Environmental Assessment Agency, 2011). Top-down results can cause major revisions to previous emission inventories, for example after the work of Bergamaschi et al. (2010). Press reports of an earlier study by this team show that it

Table 1

Estimates of Global Annual CH₄ Budget in 2012 (Range) (Tg/yr; Saunois et al., 2016; See Also Saunois et al., 2019 for Slightly Different Updates)

Sources	Top-down	Bottom-up
Natural sources total	221 (192–302)	386 (259–532)
Natural wetlands	172 (155–201)	187 (155–235)
Other Natural sources	49 (22–137)	
Agriculture and waste	200 (122–213)	197 (183–211)
Biomass burning, etc.	35 (28–51)	30 (25–36)
Fossil fuels	112 (90–137)	134 (123–141)
<i>Sum of sinks</i>		555

caused substantial revisions (<https://www.theguardian.com/environment/2006/jun/22/climatechange.climatechangeenvironment>). Chang et al. (2019) also illustrate this problem: they found, compared to previous estimates, a larger than expected contribution of ruminant enteric emissions to the increasing trend in global methane emissions between 2000 and 2012. This discrepancy may point to errors in either or both top-down and bottom-up assessment methodologies, a problem that does not instill confidence in using bottom-up emissions estimates to identify the optimum (least cost, most benefit) targets for reduction efforts.

The global challenge is clear from Table 1. The need is to cut emissions from fossil fuels and from biomass and biofuel burning and reduce the methane footprint of agriculture and waste. But within this global envelope, each nation has its own spectrum of emissions. Figure 2 illustrates the impact of a sustained effort to reduce U.K. national emissions. Earlier emission cuts were very significant, but recently, the reduction trend has slowed or ceased. In 2017, the National Atmospheric Emissions Inventory (UK NAEI) estimated total U.K. emissions from waste as about 756 kilotons (kt) (quoted in tons by NAEI with no errors given: 1 kt = 0.001 Tg), fugitive emissions from energy fuels as 217 kt, 63.5 kt from combustion, and 187 kt from agriculture. If emissions from waste, fuels, and combustion were removed, the national total could be cut a further 80% or more.

Reducing emissions from ongoing anthropogenic activities such as fossil fuel production and intensive ruminant farming is essentially palliative: it helps but does not cure. More substantial reductions can be achieved by ending or substantially reducing the activities themselves, for example, by shifting entirely away from coal and gas as energy sources and by reducing ruminant emissions. But such actions require political debate and major social adjustments, which are outside the scope of this journal. The more limited mitigation options discussed here are less contentious and can be implemented within the framework of existing commitments to the UNFCCC Paris Agreement.

2. Methodology

2.1. General Methodology

We need to develop cost-effective methodologies for cutting emissions and, where they cannot be cut, for removing the emitted methane from air with elevated methane around the sources. Methane reduction is a technical challenge demanding source detection and identification, source-specific emission flux quantification, and effective reduction methodologies and targeting.

The task of reducing the methane burden is discussed in the following sections:

- Section 2: *Location of emissions and Identification of sources*,
- Section 3: *Quantification of emissions*,
- Section 4: *Practical emission reduction—tractable emissions*,
- Section 5: *Emission removal—intractable emissions*,
- Section 6: *Reducing agricultural methane production*,
- Section 7: *Summary of priorities*,
- Section 8: *Outlook*.

Some of these tasks are straightforward but most are not simple. For example, *location* of emissions, especially major point sources, should intuitively be expected to be an easy task but in practice may be complex as what initially appears to be a point source may actually comprise many small sub-sources. For example, in a gasfield, leakage from producing wells may be relatively minor, while major emissions may be discovered from unexpected sites such as water processing facilities (e.g., Iverach et al., 2015; Kelly et al., 2015; Schwietzke, Pétron et al., 2017), or forgotten old (legacy) uncapped exploration wells (Day et al., 2015).

Source *identification* can be a complex task: For example, very different major sources can be closely collocated, such as cattle feedlots within gasfields, or urban landfills near biodigesters, sewage facilities, and gas distribution plants. Industrial area planning and land use often means that such facilities are located near to one another. Urban source types may also be multiple, yet geographically close. Attribution of emissions to specific sources thus can be a challenging task, demanding high-precision measurement of methane

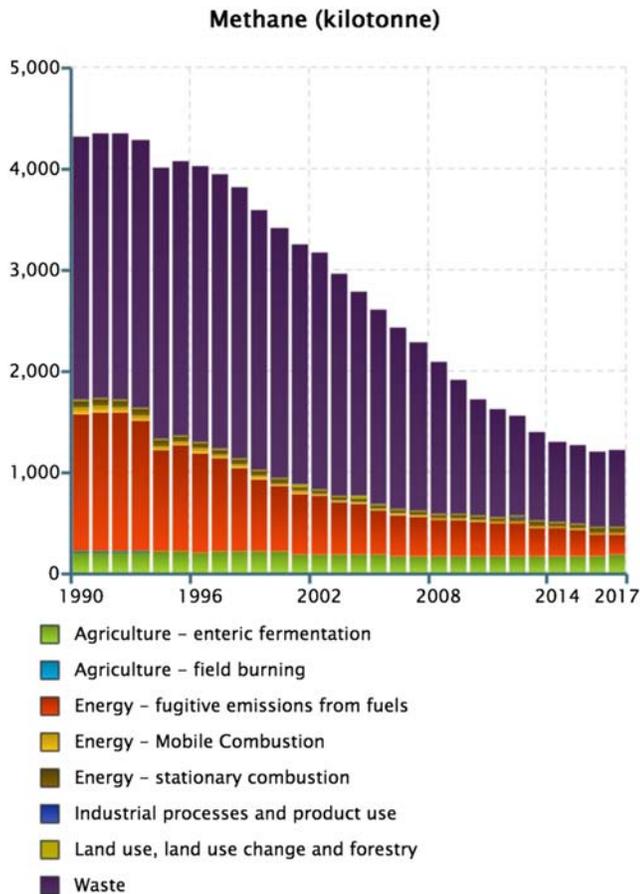


Figure 2. U.K. national inventory of methane emissions by source sector. Note the significance of the decline in emissions from waste and energy sources in the late 1990s, but the relative lack of progress in recent years. From UK NAEI (2019).

and also winds and boundary layer mixing heights, and often isotopic analysis as well as measurement of other proxy tracer species such as ethane or carbon dioxide (Allen, 2016).

Accurate *quantification* of emissions is necessary if the most cost-effective reduction strategies are to be targeted, but quantification of emission flux in plumes remains an imprecise art. Kang et al. (2016) showed the disproportionate impact of major gasfield leaks—the so-called “superemitters.” In the United States Omara et al. (2018) found the top 5% of sites accounted for 50% of cumulative emissions. Identifying, quantifying, and stopping these superleaks is the first step in emission reduction. More generally, jurisdictions need to quantify, declare, and verify emission declarations at all levels, from local emitter to nation state, but this remains very limited outside Europe (e.g., see Bergamaschi et al., 2018) and North America.

Even if major emissions are correctly identified, located, and quantified, then targeting the most cost-effective ways to *reduce* emissions is not simple. For example, Kuwait has a major hydrocarbon industry, but field campaigns carried out there by the Royal Holloway (RHUL) group there suggest, counterintuitively, that cutting Kuwait’s landfill methane output (Figure 3) may be a cost-effective first-choice target for cutting overall national emissions (al-Shalaan, 2019), in part perhaps because Kuwait’s oil industry already operates a program for emissions reduction.

Removal of emissions from the air, *after* they have been emitted, is a major part of the CO₂ carbon capture and storage discussion but has been little discussed for methane. This enquiry is a task that deserves attention and technological development.

In this synopsis for *Reviews of Geophysics* we focus on essentially geophysical methods to reduce emissions. Sustained reduction of the atmospheric methane burden will almost certainly demand much wider actions, both in management of ruminants and also perhaps societal change including demand-reduction for food derived from ruminants.

There is a broad literature on these major topics. The focus here is tactical: what can be done now. The wider strategic issues are briefly discussed later (section 8).

2.2. Locating Emissions

Rapid technical progress is being made, both in pinpointing emissions more accurately and quickly, and in finding smaller point sources. Various vehicle, drone, and aircraft-mounted technologies can be used. Although most of the simpler current technologies are still only able to provide rudimentary information on flux quantification, they are generally effective in finding leaks and suitable for localization of emission sources (Ravikumar et al., 2019).

Much progress is being made in leak detection and repair. The U.S. ARPA-E-MONITOR program (<https://arpa-e.energy.gov/?q=program-projects/MONITOR>) has the ambitious goal to “*detect and measure methane leaks as small as 1 ton per year from a site 10m x 10m in area with a certainty that would allow 90% reduction in methane loss for an annual site cost of \$3,000.*” [1 U.S. ton = 907.1 kg]. If this is achieved, it will radically improve our ability to cut emissions not only in the United States but worldwide. Currently, much leak detection in gasfields is by optical gas imaging using passive infrared cameras. For example, over the Bakken Formation in the Williston Basin of North Dakota, USA, Englander et al. (2018) used helicopter-borne infrared optical gas imaging to survey about 1,000 well pads, in conjunction with use of a light aircraft to quantify fluxes in a randomly selected sample of 33 plumes. However, the effectiveness of cameras depends on the backdrop, and their usefulness declines with distance, a problem because it limits their usefulness in surprise near- but off-site drive-by inspections by regulatory bodies. Cameras are effective close to large known sources, such as superemitters, and are excellent for detecting large leaks at gasfields from distances of 10–



Figure 3. Kuwait landfill; with $\delta^{13}\text{C}_{\text{CH}_4}$ of -57% , landfills may produce up to half of Kuwait's methane emissions (al-Shalaan, 2019). Photo: D. Lowry.

100 m (Ravikumar et al., 2016) and for big emissions over leaking street gas mains, but cameras are less able to find abundant smaller leaks in gas collection, transport and urban distribution networks.

In contrast to camera detection, mobile instruments that map by optical spectroscopy are much more sensitive. They can detect smaller leaks, with good precision and flux quantification (Weller et al., 2018) downwind from suspected sources. In the past decade, cavity-based instruments have radically improved the precision and mobility of in-the-field measurement of methane and other associated species (Baer et al., 2012; Caulton et al., 2018; Crosson, 2008). In particular, compared to older gas chromatography, with its low portability and calibration requirements, modern cavity-enhanced absorption spectrometry instruments have much better precision, robust portability, and more stable calibration in challenging environments (von Fischer et al., 2017; Zazzeri et al., 2015). Other techniques are also available for methane measurement—for example quantum cascade lasers and open path analyzers (McDermitt et al., 2011; Nelson et al., 2004). Other species such as ethane can be measured simultaneously, to differentiate between gas supply leaks and sewer emissions. Isotopic analysis can also be used to determine the nature of the source (e.g., gas leaks or sewage pipes) (Fries et al., 2018; Lowry et al., 2019, 2009; Phillips et al., 2013).

2.2.1. Location by Vehicle- and Boat-Mounted Mapping

It is becoming increasingly inexpensive and rapid to monitor leaks in both production gasfields and urban reticulation systems by routine drive-by surveys. Rapid precise mapping of methane emissions is now increasingly possible with offsite systems coupled to real-time global positioning system (GPS) location (Bamberger et al., 2014; Phillips et al., 2013, Rella et al., 2015, von Fischer et al., 2017). Such instruments can be deployed at remote places, and can be mounted on small domestic SUV vehicles (Zavala-Araiza et al., 2017; Zazzeri et al., 2015). Vehicle surveys can be supplemented by flying small unmanned aerial vehicles (UAVs) equipped with lightweight sensors (see section 2.2.2 below).

Continuously measuring GPS-linked mobile systems operate with anemometers to measure wind directions and speeds. As roads typically occur within 100 m to 1 km upwind and downwind of industrial, landfill, and cattle feedlot sources, this allows accurate location of emissions. Thus, in many cases biogas and broad landfill emission peaks can be separated by plume shape and wind direction analysis. In many cases, the likely source can immediately be pinpointed. Figure 4 illustrates a typical vehicle-mounted system. The costs of vehicle-mounted campaigns are low. The instrument and associated equipment costs can be below U.S. \$100,000. Common SUV vehicles can easily be adapted for the task. They can also deploy air hoses on pop-up balloons, to take air samples (e.g., for isotopic analysis) (Steiger et al., 2015).

The instruments map out methane mole fraction in the ambient air around the vehicle or above the vehicle's roof. Assuming a steady background air flow, the local increment over background can then be measured while the vehicle moves at speeds that can be up to ~ 80 km/hr, though typically optimized for plume capture at 20–30 km/hr. Methane peaks identify emission sources: then the sources can be located and mapped carefully using local access roads. Similarly, boats can be used to map emissions from offshore gas platforms (Riddick et al., 2019), though here the problem is that the emissions may be from locations such as offshore platform flares that are above the height of the boat's mast intake.

Helfter et al. (2019) used measurements from a North Sea ferry to estimate methane emissions from the United Kingdom and Irish republic. They mounted a cavity-ringdown analyzer on the bow of a commercial freight ferry routinely sailing between Scotland and Belgium and compared the results with measurements made at Mace Head on the west coast of Ireland, using a mass balance approach. They found that estimated annual methane emission from the study region was 2.55 ± 0.48 Tg. This result is comparable with the 2.29 Tg/yr total of estimates submitted to the UNFCCC.

Mapping by mobile analyzer is rapid, and field campaign costs can be modest, apart from the capital costs of the instrument and vehicle. In the Royal Holloway laboratory, located SW of the London urban area, Greater London's leaks are being mapped in the duration of a PhD thesis. Hong Kong sources were mapped within a 5-day working week during a period of steady methane background set by prevailing marine SE Monsoon winds, arriving from the South China Sea. Such campaigns are not in themselves quantitative, nor do they characterize sources as the surveys merely locate major "hot spots". But in most cases that is enough. Emissions sources can be pinpointed quickly and then remediation or mitigation can be considered. In some cases, halting emissions can be *profitable* (e.g., from broken pipes or leaking production wells). However, in other cases, where leaks do not pose a significant safety risk, repair may have low priority. In such cases, regulatory intervention may be needed: Thus, there is a strong argument that regulatory authorities should have the capacity for independent discovery of leaks and the power to impose financial penalties.

2.2.2. Location by Drone (UAV) Surveys.

Drones (UAVs) offer powerful opportunities for rapid identification and hence reduction of concentrated methane leaks (Fox et al., 2019). Aerial measurement and sampling have the potential to map out emission plumes in three spatial dimensions, as well as time. Moreover, if the drones carry anemometers, eddy flux calculations may be carried out to determine emission fluxes.

Small drones can carry instruments to map methane plumes close to sources and use coincident wind measurements to derive flux using a range of approaches including Gaussian plume modeling and mass balancing. Already it is possible to use these inexpensive light sensors to patrol gas installations. For typical small quadcopter and octocopter drones, the lifting ability is sufficient to carry small sensors able to achieve methane measurement to moderate precision. Most low-cost low-weight sensors are capable of measuring methane to 2-ppm precision (Collier-Oxandale et al., 2018; Shah et al., 2019), which should be capable of detecting significant emissions around known candidate sites. However the precision and accuracy of light-weight small sensors is currently poor compared to vehicle-mounted or static ground-mounted instruments. Larger UAVs using inexpensive lasers in the near infrared can achieve better precision, to ~100 ppb.

Recently, more expensive midinfrared lasers have been used, exploiting methane's strong absorption band around $3.3 \mu\text{m}$, and have demonstrated performance in the 1- to 10-ppb level (Golston et al., 2017; Shah et al., 2019) when mounted on drones weighing 5 kg or less. The technology is changing rapidly. Better than 10 ppb (at 1 Hz) precision instrumentation is now available and in use on commercially available UAV platforms (Shah et al., 2019), and 1 ppm precision is likely soon. Further marked improvement in the precision, price, and availability of UAV-mounted sensors should be expected in the near future. Small drone-carried sensors have much promise for safe close-up mapping of air with high methane mole fractions around major leaks, for example, in mapping the 3-D distribution of a plume from a leaking gas installation, and hence quantifying the flux (Emran et al., 2017).

UAV surveys are useful in large-scale screening of gas infrastructure for major point sources (e.g., over a spread-out gasfield or landfill site) to find and quantify large leaks quickly (Allen et al., 2019; Barchyn et al., 2017; Yang et al., 2018). Many leaks are episodic "one-offs"—the result of errors such as faulty or forgotten valves, ill-fitted connections or equipment failure. Sustained drone patrols, carrying low-cost sensors close to known gas systems, can identify these transient events quickly and cheaply. Such systems have great potential and are likely soon to become a high priority for industry and regulatory bodies (e.g., by the U.K. Environment Agency, see Allen et al., 2018). As equipment costs are low, it should be possible and inexpensive to ensure each gas installation has frequent (daily or potentially multiple times a day) routine inspection by automated drones. This may even be profitable, both in terms of less leakage and in safety improvements, averting the risk of major problems and reducing insurance costs.

For isotopic measurements, drones can be used to take grab samples of air in Tedlar or aluminum foil bags (Figure 5) to be analyzed in a laboratory. These bags only weigh a few grams and can be carried by small

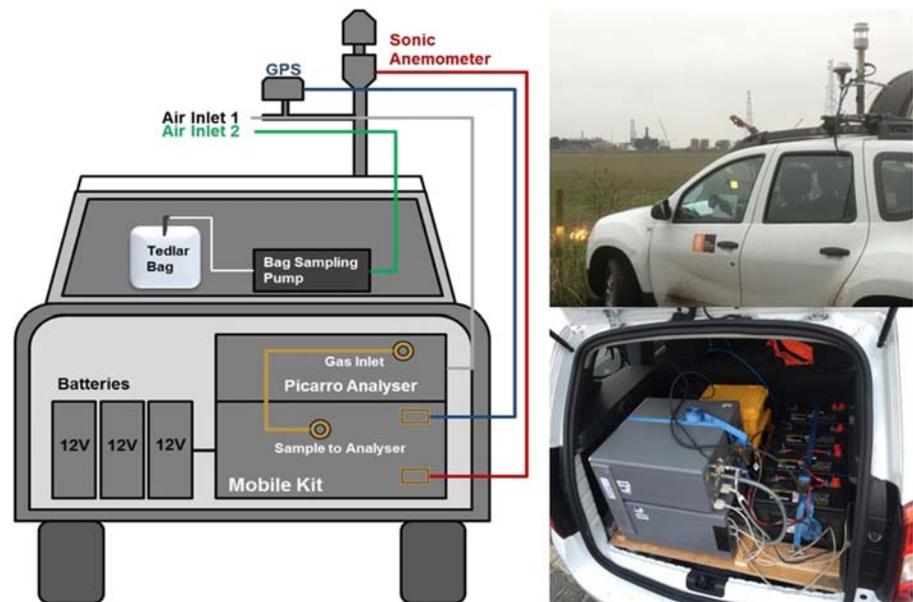


Figure 4. (left) Schematic set up of the Royal Holloway Picarro mobile measurement system (from Zazzeri et al., 2015). (right upper and lower) Vehicle mounting. (right) Deployment at Bacton gas terminal, Norfolk, UK.

quadcopters. Aircores, which are air samples collected in coiled tubes for later analysis in the laboratory, can also be used to sample air with UAVs (Andersen et al., 2018).

On Ascension Island in the equatorial South Atlantic, Brownlow et al. (2017) and Greatwood et al. (2017) report bag-sampling air from as high as 2,700 m in the midtroposphere, well above the height of the local South Atlantic Trade Wind Inversion at about 1,200–1,500 m that defines the top of the marine boundary trade winds. This allowed Brownlow et al. (2017) to sample air that had traveled from the otherwise difficult-to-access Congo basin in equatorial Africa, a potential future tool for validating national and regional emissions inventories.

In most locations, UAVs are permitted a maximum altitude of 100–200 m, confining them within the lower part of the boundary layer. This is sufficient for proximal surveys on sources, but in most countries higher altitudes are barred to drones and can only be accessed by aircraft. It should be noted however that in many tropical nations UAV restrictions are extremely severe and in some cases wholly barred, particularly near military bases. Moreover, in nations like the United Kingdom legal frameworks for UAV operation are increasingly complex and obtaining permits may become challenging to small academic teams, especially close to sensitive installations such as gas industry facilities, thus may limit the usefulness of UAV techniques for leak studies, and especially the development of novel applications by university research.

2.2.3. Location by Aircraft Surveys.

Piloted aircraft capable of carrying high-precision spectroscopic instruments are still needed for precise measurement of regional fluxes and major emissions sources. To assess emissions from further afield, where mixing with ambient air has taken place and the incremental methane signal is small, for example, some kilometers downstream from a coal mine or large cattle feedlot, or to characterize bulk emissions from a city or a major gasfield, high-precision optical cavity-based fast methane analyzers are needed. Currently, instrument and battery packages typically weigh >10 kg for mole fraction precision better than 1 ppb, and the required battery capacity for a useful survey is high (of the order of 100 amp-hour). These are too heavy to fly on cheap drones and require commercially available UAVs with a takeoff weights typically >20 kg. Moreover, as mentioned above, drones typically have a height limit in most jurisdictions, depending on airspace classification.

Karion et al. (2013, 2015) and Caulton et al. (2014) used aircraft-based measurements to map out emissions from unconventional oil and gas fields in the United States, while Peischl et al. (2015, 2018) and Schwietzke et al. (2017) used aircraft equipped with a high-precision analyzer to study several major shale gas fields.



Figure 5. Example of drone-carried Tedlar bag sampling for methane mole fraction and $\delta^{13}\text{C}_{\text{CH}_4}$ (Brownlow et al., 2017; Greatwood et al., 2017).

Similarly, in the Netherlands, Yacovitch et al. (2018) used a light aircraft equipped with a high-precision gas analyzer and flask sampling capability to determine emissions from the giant Groningen gasfield and the heavily agricultural surrounding area. They demonstrated that production volume from a specific production site is not a good guide to emissions from that site and that applying inventory emission factors (i.e., production-based weighting) does a poor job in assessing emissions.

Aircraft are useful in measuring agricultural emissions also. Using a light aircraft flying at 50–500 m above ground, Hiller et al. (2014) tested the Swiss methane emissions inventory over an agricultural valley in rural Switzerland. They measured methane mole fractions during the flights and then used two approaches, eddy flux covariance and boundary layer budgeting, to assess fluxes. Flux estimates were higher than the Swiss national inventory, suggesting Swiss agricultural emissions had been underestimated. Studying a powerful, more focused source, Hacker et al. (2016) used a low flying aircraft over an Australian feedlot with 17,000 cattle. The aircraft was equipped with quantum cascade laser analyzers, and high resolution wind turbulence measurement. Precision was adequate to apportion emissions qualitatively to individual rows of cattle pens, effluent ponds and manure piles, and elevated methane mixing ratios were detected as far as 25 km from the feedlot.

2.2.4. Use of Satellites in Locating Methane Emissions.

By measuring solar backscatter in the shortwave infrared, satellites offer rapid and powerful methods of locating methane emissions (Jacob et al., 2016). This is potentially important in the land tropics where in situ observations are typically few and infrequent. Although the utility of satellite observation has hitherto been limited by low measurement frequency, retrieval accuracy, and vertical and pixel spatial resolution, newer satellites, which include commercial satellites that offer pixel-scene resolutions of the order of a few kilometers and greater sensitivities to the tropospheric column.

Many earlier studies used the European MIPAS and SCIAMACHY instruments on ENVISAT (Bergamaschi et al., 2013; Frankenberg et al., 2006; Houweling et al., 2014), which also retrieved CO_2 in the 1.65 μm band, and other IR-active gases, with a coarse pixel resolution. More recently, the Japanese GOSAT (Hu et al., 2018; Kuze et al., 2009) has provided global mapping of methane, albeit with averaging kernels in the mid-troposphere and limited sensitivity to methane in the lower troposphere (e.g., Lange & Landgraf, 2018). Although SCIAMACHY (2002–2012) and GOSAT (launched 2009) were capable of identifying regionally important high column measurement over hotspots, the spatial and boundary layer resolution was poor compared to in situ and aircraft mapping, and complicated by biases and poor accuracy of satellite measurement. Moreover, it is not easy to obtain flux quantification from satellite observations. Thus, for the purposes of reducing emissions, satellite observation to date has been of general but not specific value.

However, when used in the context of other inputs and chemical transport modeling, satellite results can be helpful in targeting reduction measures. Specific local enhancements can be detected using GOSAT observations, as shown by Sheng, Jacob, Turner, et al. (2018), who used satellite results, together with a gridded

inventory of emissions (Maasackers et al., 2016), to infer trends in North American emissions from different source types (oil and gas, livestock, and wetlands). In an influential study, Kort et al. (2014) coupled satellite observation with ground-based validation to identify the largest satellite-detected anomaly in the United States within the Four Corners region in the Southwest United States (particularly coalbed extraction), which they showed to have emissions up to 10% of the total U.S. methane emission inventory. In another example, Wecht et al. (2014) used satellite and aircraft observations, coupled with the local inventory of emissions, to constrain and attribute the geographical distribution of emissions in California. On an even larger scale, Miller et al. (2019) used observations from the Japanese GOSAT satellite to assess China's methane emissions.

Recent instruments are considerably advancing the power of satellite observation to help in locating emissions, with better resolution and more frequent observation (Sheng, Jacob, Maasackers, et al., 2018). Turner et al. (2018) investigated the capabilities of different satellite observing systems. They found the TROPOMI instrument, with 7×7 -km pixels, 11-ppb single-retrieval column-weighted precision, and daily frequency (Veeffkind et al., 2012), should be capable of giving useful information on a spatial resolution of about 30 km. The planned GeoCARB satellite, to be launched in the early 2020s, should give even better resolution (2.7×3 km) and precision (4 ppb), making it potentially capable of usefully studying methane emissions from large urban centers such as Shanghai (O'Brien et al., 2016), although aerosol pollution will pose a major problem in retrieving accurate methane mixing ratios from regions like China, India, and tropical Africa in biomass burning season. Cusworth et al. (2018) showed that TROPOMI and GeoCARB will be successful in locating major sources of emissions in spread-out gasfields but will be less successful where gas installations are densely packed.

A powerful example of the usefulness of satellite detection was given by Pandey et al. (2019) who used TROPOMI observations to discover a very large and hitherto little reported methane emission in the U.S. state of Ohio in February–March 2018. The blowout continued for 20 days, and was observed by TROPOMI on the thirteenth day, when emissions had likely declined from an initial peak rate. With an atmospheric tracer transport simulation, Pandey et al. investigated the methane plume dispersion and used both mass balance and cross-section flux approaches to quantify the emission rate. They found an emission rate of 120 ± 32 metric tons per hour, which was twice that of the much better reported Aliso Canyon event (Conley et al., 2016 and see below) and that the total methane emission in this little-discussed Ohio event was perhaps a quarter of Ohio's annual emission. This case demonstrates the power of satellite observation and its usefulness in tracking underreported events.

Finer detail is offered by instruments with pixel resolutions varying from 1–10 km down to as fine as 50×50 -m resolution. MethaneSAT, with high spatial resolution and very high precision, is designed to monitor emissions from about 50 oil and gas production regions accounting for about 80% of global production. It will measure methane and carbon dioxide over a ~ 200 -km wide swathe (Wofsy & Hamburg, 2019). The high-resolution GHGSat microsattellites designed for greenhouse gas detection, observing methane columns over selected 10×10 -km locations, give high effective pixel resolution of 50×50 m and 1–5% precision (Jacob et al., 2016; Varon et al., 2018). Next-generation versions with excellent spatial resolution and improvement in point source detection better than ~ 10 kt/yr (0.01 Tg/yr) should be valuable where likely emission loci are already known, for example, in identifying superemitters in gasfields, and in targeting locations of major leaks on gas industry sites.

A different satellite perspective was taken by Elvidge et al. (2009, 2016) who used low-light imaging data to estimate global natural gas flaring. This major source of both CO_2 and methane emissions remains poorly quantified, especially in regions like Nigeria and Russia. Elvidge et al. (2009) showed that in both these regions the flaring efficiency markedly improved over the 2005–2008 period. More recently, Deetz and Vogel (2017) used visible infrared imagery to show continued decrease in Nigerian emissions of CO_2 from flaring. If it is assumed that as flaring reduced, methane emissions also declined, this methodology becomes valuable in regional inventory construction.

2.3. Source Identification

Source identification depends on source flux, temperature, mixing layer height, wind speed, and wind direction. The problem is complex (Lowry et al., 2019). Often, major emissions from different source types are

closely juxtaposed. Facilities like waste handling and sewage treatment plants tend to be colocated in industrial districts, often with a gas facility, a landfill, and perhaps an incinerator close by, surrounded in many cities by dense suburbs. Sewage treatment plants can be located next to gas distribution installations, or nearby landfills can emit gas close to waste burning facilities. In Australia, the United States, and Canada, gasfields host major cattle feedlots, in some cases sharing water use and capable of emitting more methane than the gas wells. In these settings, source attribution can be challenging. There are so many candidates for methane emissions. Where multiply overlaid sources occur, help can come from key identifiers such as the presence or absence of associated species such as ethane, or isotopic signatures (Lowry et al., 2019). Methane/ethane ratios are powerful discriminants between separate source signals, enabling identification of specific emitters (e.g., Barkley et al., 2019; Feinberg et al., 2018; Kille et al., 2019; Mielke-Maday et al., 2019). Gas leaks contain significant amounts of ethane, while cows do not breathe out much. $\delta^{13}\text{C}_{\text{CH}_4}$ studies are powerful tools in pin-pointing and segregating sources where urban land planning has produced clusters of large but distinct point sources.

2.3.1. Identification by Methane/Ethane Ratio

In Los Angeles, Wennberg et al. (2012) observed the ambient air observations of methane, ethane, and carbon monoxide, in ambient air, and also measured ethane/methane enhancement ratios in the natural gas supply. Assuming nearly all the ethane came from fugitive emissions from the natural gas supply, they were able to attribute most of the excess methane measured in regional air to losses from the gas system, which may have been losing 2.5–6% of its gas to the atmosphere. Similarly, Smith et al. (2015) used ethane-to-methane correlations to quantify the fraction of CH_4 emissions derived from fossil and microbial sources in the Barnett Shale in Texas.

Using an aircraft, Peischl et al. (2015) measured multiple species in the planetary boundary layer over the major U.S. Haynesville, Fayetteville, and Marcellus gasfields and showed the ratios between methane and other alkanes in the regional atmospheric increment were the same as in local natural gas, thereby proving the increment came from the gasfields. Also, in studies of five U.S. gasfields, Peischl et al. (2018) used methane/ethane ratios to demonstrate emissions were from the gasfields and not from agricultural sources, given similar atmospheric C_2H_6 to CH_4 enhancement ratios and the composition of raw natural gas withdrawn from the region. However, Lan et al. (2019) found an increasing trend in ethane/methane emission ratios in U.S. sources, which may have led to significant overestimation of methane emissions in some studies.

2.3.2. Isotopic Characterization

Isotopic characterization demands high-precision measurement of the $^{12}\text{C}:^{13}\text{C}$ ratios (expressed as $\delta^{13}\text{C}_{\text{CH}_4}$) of enhanced and background methane in ambient air masses (Keeling, 1958; Pataki et al., 2003; Zazzeri et al., 2017). In settings where many sources can be geographically closely superposed, isotopic signatures are powerful discriminators between sources. To take an extreme example, an aircraft survey found a methane-rich plume downwind of the U.K.'s East Anglian offshore gasfields, but isotopic study showed that a significant part of the methane anomaly was not from the gas platforms but had blown there from remote on-land sources (Cain et al., 2017). They found a significant methane anomaly in a plume between 25 and 50 km wide, with elevated methane at 70 to 100 ppb above the background, which was detected during a low altitude (100 masl) overflight over the Leman field's gas platforms in the southern North Sea, off the coast of Norfolk, England. The methane plume was in close proximity above large gas platforms. Air samples were taken and surprisingly showed much of the methane anomaly had an isotopic signature $\delta^{13}\text{C}_{\text{CH}_4}$ of -55‰ , a characteristic ratio for land sources such as cattle or landfills, and not the -32‰ signature of these gasfields, showing that much of the source was not the gas platforms.

In ambient air relatively remote from sources, off-line lab-based isotope ratio mass spectrometry is required to reach the needed analytical precision, by analyzing air samples taken at the study sites. This is because typically it is necessary to determine $\delta^{13}\text{C}_{\text{CH}_4}$ to a precision of 0.05‰ or better (Fisher et al., 2006) in ambient air masses. Adequate precision is currently achievable by vehicle-mounted traveling optical analyzers only when very close to source in methane-enriched air. These instruments typically have isotopic precisions of around 2–3‰, though improved precision can be attained, for example, if the plume can be sampled by AirCore (Karion et al., 2010; Rella et al., 2015) for isotopic playback. An alternative approach (Lu et al., 2019) that shows promise is to use a moving Miller-Tans analysis (Miller & Tans, 2003) of time series data to characterize the population distribution of the isotopic signature for a given source.

Townsend-Small et al. (2012) used isotopic ratios to interpret inputs to air samples collected across the Los Angeles megacity. They used vehicles to access a variety of sources including oil well fields, oil refineries, power plants, traffic, cattle and cattle feedlots, landfills, and sewage facilities. In addition, they used the geography of the region to collect air from a local mountain (1,707 m above sea level). Methane in the ambient air samples was then analyzed for $\delta^{13}\text{C}_{\text{CH}_4}$ and $\delta\text{D}_{\text{CH}_4}$ as well as for $\Delta^{14}\text{C}_{\text{CH}_4}$. They found that in Los Angeles the main source of methane emission was leakage of gas from fossil fuel use, including geological sources, natural gas pipelines, oil refining, and power plants. In the Los Angeles study, $\delta\text{D}_{\text{CH}_4}$ was the strongest indicator of methane source.

This result from Los Angeles was in contrast to the older results of Lowry et al. (2001), also using stable isotopes, who found a major source of methane emission in the London area was landfills. Lowry et al. (2001) were reporting on London around the millennium: More recently, much better landfill practice has led to lower U.K. landfill emissions (UK NIR, 2019). Currently, Royal Holloway laboratory observations suggest leaking natural gas is the largest contributor to methane emissions around Egham, in western London. This is because all the landfills west of London in the 2001 study have now closed, and gas leaks now predominate.

Zazzeri et al. (2015) used Keeling plots to characterize sources, using bag samples. Figure 6a is from a landfill at Mucking in SE England, emitting methane with $\delta^{13}\text{C}_{\text{CH}_4} -56.1 \pm 0.5\text{‰}$ (2 SD) while the Keeling plot in Figure 6b is from a gas works emitting methane with $\delta^{13}\text{C}_{\text{CH}_4} -36.3 \pm 0.3\text{‰}$. The results show that these two sources are readily distinguishable. In contrast, Figure 6c shows results from three coal mines. The $\delta^{13}\text{C}_{\text{CH}_4}$ intercept from the Thoresby mine in Nottinghamshire, England, was $-51.2 \pm 0.3\text{‰}$, markedly different from the methane emitted by the Aberpergwm and Unity mines, which were neighbors 1.2 km apart in the South Wales coalfield. Emissions from the latter two were isotopically similar, with signatures around $\delta^{13}\text{C}_{\text{CH}_4} -32\text{‰}$ that are just within error and very different from the first mine. Intrinsically methane from these two coalmine emission sources may be separable, with a high enough sample population and also using careful directional measurement by a mobile system. Thus, these results show that even with closely similar sources, such as neighboring coal mines or gaswells, it may be routinely possible to identify emissions by off-site surface measurement from public access roads. A similar methodology has been used for CO_2 by Domenikos et al. (2019), while Fries et al. (2018) used carbon ($\delta^{13}\text{C}_{\text{CH}_4}$) and hydrogen ($\delta^2\text{H}_{\text{CH}_4}$) stable isotopic determination to distinguish between biogenic methane from sewer gas and thermogenic methane from leaking natural gas pipelines in Cincinnati, Ohio.

In these studies, for each emission source type, the isotopic signature and/or ethane:methane ratio must be known for the specific location being investigated. However, both isotopic signatures and ethane:methane ratios vary widely depending on the source, whether gas basin or farm location, and in some cases significant variation in these signatures can occur very locally.

Case study A: attribution of sources around Fylde, UK. Source attribution by isotopic measurements was used to identify the emissions measured around Fylde, Lancashire, northern England, UK (Figures 7 and 8). The measurement, reported here, was carried out as a baseline survey around a proposed site for gas extraction by fracking. Gas sources are identified by their methane/ethane ratios (Figure 9) and their different isotopic signatures. Farm signatures have $\delta^{13}\text{C}_{\text{CH}_4}$ close to -60‰ , manure piles -50‰ , and gas leaks -40‰ . In a similar study in the Vale of Pickering, also in northern England, gas leaks from the natural gas offtake station had $\delta^{13}\text{C}_{\text{CH}_4}$ of -42‰ , while farm cow manure piles had $\delta^{13}\text{C}_{\text{CH}_4}$ of around -50‰ , and methane from the cattle in the barns themselves and their waste had a combined $\delta^{13}\text{C}_{\text{CH}_4} -59\text{‰}$. However, methane from the local landfill had $\delta^{13}\text{C}_{\text{CH}_4}$ of -57‰ , closely similar to methane from cattle eating mixed C3/C4 fodder (e.g., grass plus maize). Thus, wind direction is needed to discriminate between landfill emissions and cattle breath.

Case Study B: unexpected events: Elgin gas leak. Aircraft are important when major unexpected events occur. An uncontrolled gas release took place from the Elgin platform in the U.K. North Sea from 25 March to 16 May 2012 (Lee, Mobbs, et al., 2018). When the gas blowout occurred, the U.K. FAAM (Facility for Airborne Atmospheric Measurement) BAe146 aircraft was fortuitously fully equipped as it had just returned from measuring methane in Scandinavia: It was immediately sent to fly over the gas platform (Figure 9). Flying at low altitude, FAAM was able to map out and sample the plume of methane-rich gas from the leak (Lee, Mobbs, et al., 2018). This study was carried out flying normal to wind direction. As a general rule

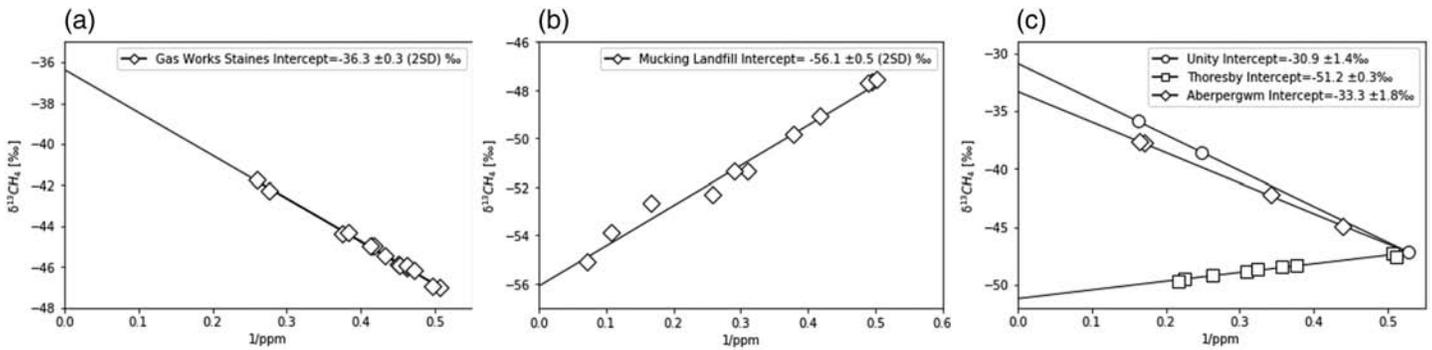


Figure 6. Keeling plots from (a) gas works in Staines (UK), (b) landfill (UK), and (c) three coal mines in the United Kingdom. Errors on the y axis are within 0.05 ‰ and on the x axis within 0.0001 ppm⁻¹ and are not noticeable on the graph. From Zazzeri et al. (2015).

however, where the source is known, it is better to fly at an angle (e.g., 30°) to wind direction in order to maximize the intersection.

Isotopic measurements (Figure 9) then helped pinpoint the source of the gas in the blowout. Keeling plots in air samples from flights transecting the gas plume (Figure 9) found the source gas had $\delta^{13}\text{C}_{\text{CH}_4}$ of $-42.3 \pm 0.7\text{‰}$. The results pointed to a source in a small gas pocket, geologically shallower than the main gasfield and that the potential volume of gas involved in the leak was limited. The hypothesis was borne out. The leak diminished rapidly. This FAAM aircraft study informed the platform operators, advising them that the gas



Figure 7. Fine detail of road circuit around a proposed U.K. shale gas drilling site (Fylde, Lancashire, UK) showing the main sources and their isotopic signatures, and the excess methane averaged across 18 days of surveys and aggregated into 10 m × 10 m grid squares. Farm $\delta^{13}\text{C}_{\text{CH}_4}$ signatures are close to -60‰ , manure piles -50‰ , and gas leaks -40‰ . RHUL results.

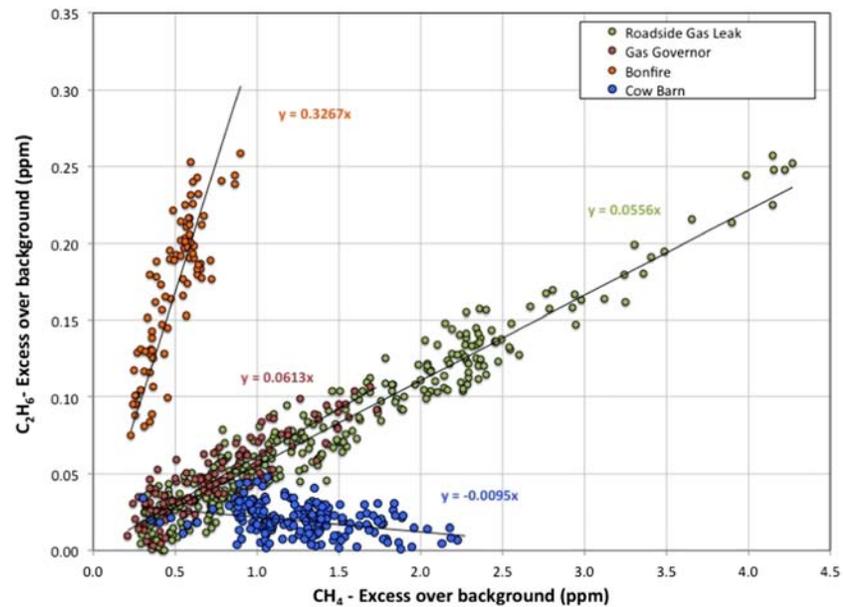


Figure 8. Distinct methane/ethane ratios for different sources, Fylde study, RHUL results.

flux from leak was reduced. The leak was then ended by accessing the platform for a top kill, rather than drilling down for a much slower bottom kill that had been discussed.

A much larger event followed 3 yr after Elgin, when a blowout took place from the Aliso Canyon underground storage facility in California in 2015. This emission was studied by a team which included participants both from the effort to quantify the Elgin leak and also from the study of fracking in U.S. gasfields (Conley et al., 2016). In an impressive campaign, they flew dozens of plume transects over the region, as well as taking whole air samples on the ground downwind. This successfully mapped out the dispersion of the leaked gas. However, in contrast to the Elgin study, which used $\delta^{13}\text{C}_{\text{CH}_4}$ in an attempt to identify the source reservoir, Conley et al. (2016) measured ethane, C_2H_6 , as the marker to distinguish the emissions from the natural gas leak from other sources of methane.

3. Quantification of Emissions

If a big leak occurs, it needs to be fixed. Rapid assessment that a leak is big is essential for safety. Unsafe leaks must be stopped immediately. Thus, in many cases, the only necessary regulatory quantification is the distinction between “large” or “small.” For mitigation however, flux quantification is important. The most obvious challenge is to find those neglected leaks that are large enough to be a significant emission, yet below the “unsafe” or “must-be-fixed” threshold. Smaller leaks are of course also important for mitigation, but how should mitigation efforts be targeted? To set priorities for cost-effective reduction, leak quantification is needed.

3.1. Direct Measurement

The simplest way to quantify an emission is to enclose it and measure it directly. Kang et al. (2014) and Lamb et al. (2015) placed enclosure chambers around gas leaks and then directly sampled the methane that was captured. Using this methodology, Kang et al. (2016) demonstrated that 5–8% of annual methane emissions in the U.S. state of Pennsylvania came from abandoned oil and gas wells, many of which were high emitters of methane. Many of the highest emitters are in the Appalachian basin, including Pennsylvania, Ohio, and West Virginia, and they are among the oldest oil and gas wells in the world, some over 100 or 150 yr old (Townsend-Small et al., 2016).

Abandoned wells are relatively straightforward (though not necessarily easy) to locate and once identified may be quickly quantifiable within broad error limits. Although unlined abandoned holes may be difficult

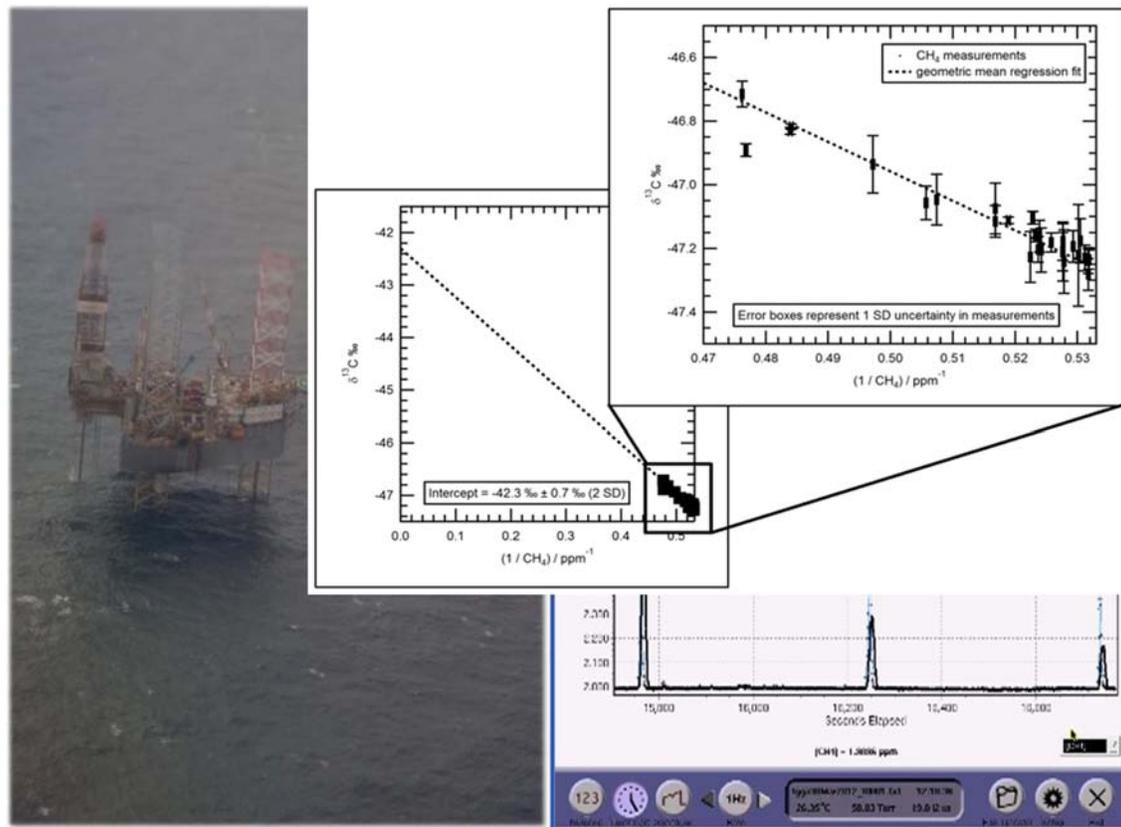


Figure 9. (left) Elgin gas production platform, from the window of the U.K. FAAM aircraft, taken during sampling on 2 April 2012. Photo: Mathias Lanoisellé (RHUL group). (lower right) 30 March onboard CH_4 plot for measurements 5 nautical miles (9.26 km) downwind of the leak; peaks are for measurements at different altitudes—from left 37, 153, and 233 m (see Lee, Mobbs, et al., 2018). (upper right) Keeling plot of air samples from Elgin methane plume. Inset—enlarged detail. $\delta^{13}C_{CH_4} - 42.3 \pm 0.7 \text{‰}$. (2σ error: geometric mean regression). Measurements of CH_4 and $\delta^{13}C_{CH_4}$ at RHUL (Lee, Mobbs, et al., 2018).

to plug, given the cost of both the leak and the explosion danger such super-emitters pose, they should usually be an obvious target for mitigation. However, many of these historic wells are “orphans” that do not have any permit or current owners, leaving the responsibility for plugging to the state or federal government, which is slow, and mitigation prioritization may not be based on methane emission rate (Townsend-Small et al., 2016).

3.2. Mass Balance and Tracer Quantification Methods

For most sources, fluxes need to be assessed by measuring the air into which the emissions are mixing. In most circumstances it is not possible to enclose sources in chambers, because the sources are unknown, physically too large or disseminated, or otherwise uncapturable. That poses the difficult problem: How does one quantify emissions that are input into moving air masses? The problem is exacerbated, especially in less developed nations, by the lack of access or skills to use sophisticated meteorological products. What may be possible on a flat North American prairie with funding for real-time local meteorology, good computing skill, and access to sophisticated modeling software may not be feasible for a low-income community trying to assess fluxes from a landfill located among complex topography under the intertropical convergence zone.

One of the simplest mass balance quantification methods was used by Lowry et al. (2001), who studied the London conurbation, which has a detailed CO_2 emissions inventory. They measured the excess CO_2 and CH_4 inputs to urban air, by subtracting the known mixing ratios in contemporary Atlantic background air, and then obtained the methane emissions as a ratio to the inventory CO_2 emissions, assuming the bottom-up CO_2 inventory to be valid. This allowed them to test the less well constrained London CH_4 emissions inventory for consistency against the better-constrained CO_2 emissions inventory.

Proxy tracer (e.g., CO, N₂O or acetylene, or additional CH₄) release methods, using deliberate emission of a tracer gas at a known mass/second rate to validate a dispersion model also offer important techniques to quantify fluxes (Bell et al., 2017). The method relies on the assumption that the released tracer gas will behave in the same way as methane emitting from the landfill. The methane emission is then proportional to the ratio of the integrated concentration of the emitted methane to the integrated concentration of the released tracer gas, as measured across the dispersion plume (Scheutz & Kjeldsen, 2019). Tracer release can be coupled with spectroscopy to calibrate fluxes from individual sites such as landfill and oil and gas facilities (Ars et al., 2017; Foster-Wittig et al., 2015; Rees-White et al., 2018; Roscioli et al., 2015; Scheutz et al., 2011). Indeed, Scheutz and Kjeldsen (2019) suggest that the tracer gas dispersion measuring technique should be a core methodology in monitoring to determine emissions.

The aircraft mass balance approach was used by Ren et al. (2019) to estimate emissions from unconventional gas and oil extraction from the Marcellus Shale in Pennsylvania and Virginia, over a 4,200-km² area. They found emission rates around 0.8% to 1.5% of total gas production, lower than some other studies but consistent with the national inventory assessment. In another study, over major U.S. East Coast cities, Plant et al. (2019) used aircraft observations of CH₄/CO₂ ratios and other species to test local emissions inventories. They found that emissions, predominantly attributed to fugitive natural gas, were very substantial and more than twice the recent gridded inventory estimates. Wunch et al. (2016) used a similar technique with CH₄/C₂H₆ ratios to estimate methane emissions in Southern California, another large heavily urbanized urban area. Using historic data, Wunch et al. (2016) analyzed records of total column abundances for ethane and methane and studied the historic evolution of ethane to methane ratios. They showed that more than half of the excess methane in the Southern California coast basin's air came from natural gas losses.

Pitt et al. (2019) quantified fluxes using a mass balance approach for the Greater London area using the U.K. FAAM research aircraft (www.faam.ac.uk) equipped with a cavity-enhanced absorption spectrometer. The same aircraft was used by Lee, Mobbs, et al. (2018), to quantify emission flux from an uncontrolled methane leak from an oil platform in the North Sea. Likewise, O'Shea et al. (2013) used a cavity-enhanced absorption spectrometer for airborne measurements over Arctic Scandinavia. With simple box modeling and boundary layer measurement to obtain methane fluxes from wetlands, they found results consistent with parallel in situ chamber-based measurements on the ground below (O'Shea, Allen, Gallagher et al., 2014). With a different approach in flights over England, Pitt et al. (2016) used an airborne quantum cascade laser absorption spectrometer to measure methane and N₂O, demonstrating the effectiveness of the method. Moreover, aircraft-based infrared remote sensing of methane (and other proxy tracers) can offer higher spatial resolution retrieval compared to satellites (e.g., Allen et al., 2014).

Airborne remote sensing with a visible/infrared imaging spectrometer is extremely powerful providing a regional assessment. In the California Methane Survey, Duren et al. (2019) measured ground-reflected solar radiation between 380 and 2,510 nm, from an aircraft at 3-km altitude, over oil and gas fields, as well as areas with manure and waste management activities. In their campaigns they covered 272,000 facilities, including over 200,000 oil and gas wells. They detected 564 strong point sources emitting methane, especially from landfills. For these distinct sources, measured at 250 facilities, they observed the local methane enhancement and then estimated emission fluxes using wind speed data from weather reanalysis products. They found a population with a heavy tail distribution, with 10% of the point sources responsible for 60% of the emissions. In particular, the largest category of super-emitters was landfills, especially a subset of superemitters, while dairy farms and oil and gas facilities also contributed.

3.3. Mathematical Modeling

An effective and simple quantification method uses the assumption that the source is a point and that the plume disperses downwind (Brandt et al., 2014; Robertson et al., 2017; White et al., 1976). When methane is emitted from a source, the input is entrained into the moving mass of ambient air. In this moving air mass, the input then disperses both horizontally and vertically, to form a cone of dispersion. If it is valid to assume the gas is well-mixed in this cone, then the mole fraction of the gas at any nearby point where it can be measured depends on the source strength and height, the wind speed, and the stability of the air mass (Seinfeld & Pandis, 2006; Pasquill, 1975).

Caulton et al. (2018) very successfully used a Gaussian methodology and a hierarchical measurement strategy involving both mobile vehicle-mounted measurement and small (2–3 m) deployable towers, as well as controlled releases, to quantify emissions from a field of over 1,000 gas wells in Pennsylvania, USA. They clearly demonstrated it is possible reliably to identify and roughly quantify extreme emission sites (i.e., superemitter mitigation targets).

Modeling tools are accessible via U.S. Environmental Protection Agency (EPA) “Other test method” OTM 33a (via <https://www3.epa.gov/ttnemc01/prelim/otm33a.pdf>). This is a way of identifying and roughly quantifying emissions from sources located by large spikes in the analyzer record as the vehicle drives past; for example, it is ideal for low-cost regulatory measurement of emissions from gas wells and pipe leaks.

Eddy covariance techniques have long been used to quantify emissions in natural settings (e.g., Detto et al., 2011; Rinne et al., 2007). In Florence, Italy, Gioli et al. (2012) sampled from a 3-m mast that was on a roof 14 m above average roof level and 33 m above ground. Methane fluxes from the footprint area were 189.2 ± 7.0 mg CH₄·m⁻²·day⁻¹ and did not show significant temporal variability. Similarly, Helfter et al. (2016) measured CH₄, CO₂, and CO for 3 yr from the BT Tower in central London, 190 m above street level, and from another rooftop site 2 km away, 50 m above street level. Methane emissions in London were similar to Florence, at 197 ± 8 mg CH₄·m⁻²·day⁻¹, but were found to be double inventory assessments and showed moderate seasonality (21% larger in winter). These studies demonstrate the power of the eddy covariance technique in urban settings and its usefulness in testing bottom-up inventory assessments. In principle, future drones fitted with better sensors and good anemometers will carry out eddy flux calculations directly.

3.4. Modeling Use of Tall Tower Networks

In densely populated urban areas, or in major producing regions with dense sources, such as many onshore gasfields, well-located measurement towers can be used to pinpoint emissions and to assess regional emissions using eddy covariance or optimized Lagrangian-inversion techniques. This can be applied on any scale, from city to continent or planet. For the US Los Angeles Megacity, Yadav et al. (2019) took data from a network of 15 in situ sites and used inverse modeling to estimate methane emissions from 3-km cells, over 4-day windows, across the megacity area. This method located emissions in enough detail to pinpoint major sources, for example correctly detecting the shutdown of emissions from a major landfill. However, for the very large and highly localized Aliso Canyon leak there were periods when the inversions did not capture fluxes well, especially in unhelpful prevailing wind conditions.

More widely in the United States, Lan et al. (2019) used long-term data from both aircraft and tall towers to analyze methane emissions from a variety of sites around the country, including in oil and gas production areas. Encouragingly, this important study found that in the period 2006–2015 the increase in emissions from North American oil and gas industry sources was much smaller than findings in many previous estimates.

In Europe, Bergamaschi et al. (2018) used data from measurement networks to validate regional emissions, and Wunch et al. (2019) used long-term, ground-based measurements of atmospheric total column methane abundance by remote sensing, to assess methane emissions from the wide northern European region from Poland to France. Their results implied that the European inventories were likely overestimated. The Integrated Carbon Observing System network maintains a long-term network of stations carrying out high-precision greenhouse gas measurement (<https://www.icos-ri.eu/icos-stations-network>). For the United Kingdom the GAUGE (*Greenhouse gAs Uk and Global Emissions*) Project used a mix of tall tower (Stanley et al., 2018; Stavert et al., 2018) and other ground-based measurements, supported by other airborne and ship-borne measurements and satellites with the aim of producing better understanding of U.K. emissions (Palmer et al., 2018).

While tall tower networks are appropriate as designed for regional budget estimates, their continuous records of methane mole fraction can intrinsically also be used by back-trajectory analysis to locate major nearby sources, especially if supplemented by air-sampling for high-precision isotopic or alkane ratio measurement to differentiate between source types such as landfills or gas leaks. Henne et al. (2016) validated the Swiss methane inventory using observations from a network of sampling sites in the Swiss plateau and

also in the Alps, as the basis of inverse modelling with high spatial resolution <10km over Switzerland. In Australia, Luhur et al. (2018) used two towers in conjunction with inverse modeling to assess regional methane emissions and quantify fluxes in the Surat Basin from coal seam gas production and processing, coal mining, cattle feedlots, piggeries, wetlands, and other anthropogenic activities. The model domain was 350×350 km, which demonstrates the cost-effectiveness of towers for making a top-down estimate check of bottom-up inventories.

3.5. Practical Methodology

In practice, each quantification problem is different, and measuring teams have different analytical assets. Ground vehicles are appropriate and widely used for studying locally sourced plumes (e.g., Baillie et al., 2019) and can be supplemented with UAV (drone) grab-bag sampling (Brownlow et al., 2016), or by tethered sampling tubes lifted either by UAVs (Allen et al., 2019), or pop-up balloons (Steiger et al., 2015). Such approaches can yield 3-D sampling for local-scale (e.g., site-specific) flux retrieval using mass balancing approaches to quantify fluxes accurately. In the cases where a measurement team has a mobile analyzer in a vehicle with a sonic anemometer and GPS location (as in Figure 3, left), and where low-level gas plumes can be measured within a few hundred meters of the source, methane emissions can be calculated by driving through the plume. If the source of the emission is known, tracer release can also be an accurate method for assessing methane emissions (Lamb et al., 1995, 2015). Isotopes are particularly helpful in apportioning emissions where various sources are present (Townsend-Small et al., 2015).

As well as cars, bicycles, and child buggies are useful for carrying instruments and batteries, as are backpacks. In a detailed study of emissions around the Munich Oktoberfest in Germany (a major beer festival), Chen et al. (2019) made in situ measurements by walking and biking around the perimeter of the Oktoberfest. They then applied a Gaussian plume dispersion model to assess emissions.

Aircraft are appropriate in assessing emissions from large conurbations and regions, to verify inventories from wider areas, where there are many sources, such as large gasfields or cities, or wide zones of natural emissions (Miller et al., 2016). For example, Cui et al. (2019) quantified emissions from the Haynesville-Bossier oil and gasfield by using inverse model calculations driven by aircraft measurements over the gasfield, and showed that emissions were probably larger than estimated in the national inventory. Over the U.S. city of Indianapolis, Cambaliza et al. (2015) and Heimburger et al. (2017) used a light aircraft and a mass-balance approach as well as methane:propane ratios in air samples to quantify urban emissions. O'Shea, Allen, Fleming, et al. (2014) used upwind and downwind airborne measurements and a mass budget box modeling approach to determine net regional flux of methane for Greater London. In an important study of the Fayetteville shale play, a major U.S. gas producing region, Schwietzke, Pétron et al. (2017) used aircraft measurement and a mass balance approach, integrating methane mole fraction measurements across plume widths in known wind velocities and direction, to assess methane emissions. The methodology of O'Shea et al. (2013), O'Shea, Allen, Fleming, et al., 2014. O'Shea, Allen, Gallagher, et al., 2014) and Hartery et al. (2018), though designed to locate and measure natural emissions, is well suited as a basis for verification of emissions inventories on a regional scale.

Frequent repeated surveys by light aircraft can track seasonal changes in emission patterns and inform local regulatory bodies of unexpected new inputs or successes in reduction policies. Though derived fluxes can be precise but highly inaccurate, when coupled with technological developments in sampling platforms and new advances in flux inversion modeling such as mass balancing, plume inversion, and gradient flux techniques, calculated fluxes can be used to identify and quantify individual sources and source types (Karion et al., 2019).

Schwietzke et al. (2018) and Englander et al. (2018) showed that in the U.S. context aerial surveys on light aircraft are an excellent tool, able to locate anomalously highly emitting sources and thus able to target on-the-ground vehicle-based identification and inspection. It is thus becoming feasible to identify leaks where fixing the emission is most cost-effective (Schwietzke et al., 2018). In some cases, it is likely the combination of aerial overpass and ground follow-up to mitigate emissions can be cost saving and thus profitable to the industry, in other cases, mitigation may have to be driven by regulatory pressure. Aircraft observation of sustained major methane emissions stimulates efforts to reduce unprofitable leakage.

The usefulness of aircraft in major events (see Case Studies A and B above) lends weight to the case for national bodies to maintain aircraft capability as a standing asset for dealing with major air pollution events, also useful in assessing dust clouds from volcanic eruptions, etc.

These intuitive points can be made:

1. For many companies, better aerial and ground identification of leaks should now become a priority as a source of profit and improved safety, in addition to greenhouse gas reduction.
2. For regulatory bodies, the costs of leak detection are declining and leak surveys are becoming more affordable as instruments improve. Leak detection is thus now becoming more affordable, especially when there is potential income from penalties and fines.
3. Methane surveys are likely also to detect sources of emissions that are not being captured in national inventories.
4. Where sudden accidental major leaks occur, urgent measurement is both essential and powerful in rapidly assessing the event and in helping to end it. Thus, wise policy is to maintain ongoing deployable capability for such events, including aircraft, at the national level.
5. There are persuasive grounds for regulatory jurisdictions to maintain aircraft capability for leaks as well as other air pollution events.
6. Hitherto, Drone/UAV measurements have lacked adequate precision to support accurate emission quantification. This is changing rapidly, for example, with new midinfrared lasers offering better sensor precision and/or lighter analyzers. However, it should be noted that with increasing regulation of UAV flying, especially near facilities such as gas installations it may become preferable to use pop-up balloons, for example, for sampling.
7. On a national or regional scale, various methodologies are available to quantify emissions, but many of these need access to prior inventories, whether CO₂ or CH₄, together with relatively sophisticated facilities (such as aircraft) and advanced modeling techniques. Such methodologies are useful in developed nations, but in many tropical nations may not be accessible. Here upwind/downwind simple mass balance techniques and single analyzer measurements (CO₂ and CH₄) may suffice. There is much need for better quantification methodologies that are more accessible to scientists and regulators in less-developed nations.

4. Practical Emission Reduction and Removal—Tractable emissions

“Tractable” emissions can be defined as those from easily located point sources, for which, once the points are found, mitigation measures can readily be undertaken. In most heavily populated regions the methane source mix is diverse (e.g., Lowry et al., 2001) but widely tractable to mitigation. Such emissions come from many sites—the gas industry, urban wastewater and sewage, landfills, etc. The development of low-cost methodological advances in leak detection, to identify superemitter leaks, should have a significant impact, especially if better UAV systems become widespread in the near future. It is bad management that leaks gas. It is much in corporate interest to eliminate super-emitters as they likely cause both loss of profit and also potentially expensive safety risks.

Obvious examples of tractable emissions are known deliberately vented emissions in gas fields and also many emissions in urban distribution systems, both deliberate vents and known leaks that are not large enough to be classed as safety hazards and hence neglected. At the other end of the tractable spectrum are leaks from abandoned installations, and small harder-to-find leaks from urban systems. In these cases, deliberate policy is needed to mitigate emissions. However, this is not necessarily very expensive. Kang et al. (2019) found that reducing methane emissions from abandoned oil and gas wells has comparable costs to other greenhouse gas mitigation options.

Typical examples in gasfields (Vaughn et al., 2017; Vaughn et al., 2018) would include methane emitted not only from readily discovered leaks around gaswells but also from deliberate operations such as flaring, venting, and pipe maintenance; unburned gas entrained in compressor exhausts during operation; and also emissions from known processes such as water handling, which can be redesigned or better contained. Episodic venting (Vaughn et al., 2018), which is deliberate, is particularly addressable. Leaks from urban gas

networks, landfills, and sewage treatment facility emissions, are widely tractable, as they are known, easily located, and predictable, as are emissions from underground coal mines, where shaft and ventilation outlet locations are well known.

In large gas production and urban distribution systems, discovery and quick approximate quantification is arguably nine tenths of mitigation: Once large sources are known, emissions can generally be ended quickly, often cheaply, even profitably, and with benefit to safety. Both deliberate venting and accidental fugitive releases are readily reducible. Linked to such mitigation measures, strong regulatory efforts can reduce venting and flaring (for U.S. data see https://www.eia.gov/dnav/ng/ng_prod_sum_a_EPG0_VGV_mmcf_a.htm). Flaring and venting gas not only does environmental damage; it also reduces the long-term energy security of the nation that tolerates the flaring.

4.1. Gasfield Superemitters

To cut emissions the most obviously tractable targets are the so-called “super-emitters” from the wells, pipelines, pumping stations, and urban distribution networks of natural gas systems. Brandt et al. (2014—see especially their Table S6) showed that in many U.S. gasfields and gas transmission and distribution systems, much of the mass of gas emitted comes from a few major leaks. Similarly, a recent synthesis of methane emissions from the oil and natural gas supply chain indicates that the most emissions come from production wells and that those are dominated by a few major sources (Alvarez et al., 2018). Mineral deposits provide a good analogy. They typically follow Zipf’s law of distributions (Guj et al., 2011): in gold mining, there are many small mineral deposits, but few of largest rank, and most of the extractable gold comes from a few very large gold mines. Intuitively, gas leaks would be expected to show similar power law patterns, with a few large leaks and many small, and with a significant proportion of the total amount of leaked gas coming from the few large sources.

Detection and monitoring of super-emitters is rapidly becoming simpler with rapid advances in vehicle-based sensors (Jackson et al., 2014; Zazzeri et al., 2015), aircraft measurement (Schwietzke, Pétron, et al., 2017) and satellite sensing (Jacob et al., 2016). Satellite observations can now attain high spatial resolution (50 m) at known sites, and are excellent for discovering large emission point sources in remote locations (Varon et al., 2019), but currently remain too sparse to constrain emissions by mapping extensive regions at fine local detail (Turner et al., 2018; Wecht et al., 2014). However, the detection and quantification of a major leakage event in Ohio by the TROPOMI instrument (Pandey et al., 2019; see section 2.2.4) demonstrates that satellite observation is rapidly becoming more able to find and quantify big leaks from superemitters.

Measurement by unmanned aerial vehicles (e.g., see the GHGmap project, <http://www.geosciencebc.com/projects/2016-065/>), is improving with the development of inexpensive, low-weight, yet more precise detectors. However, restrictions on UAV flying near “sensitive” facilities like gas installations, may impose significant limits on the future usefulness of UAV-mounted equipment to monitor facilities, detect leaks and hence mitigate emissions.

Aircraft, UAV, and vehicle surveys can now be augmented by direct on-site continuous monitoring, which is now feasible around production facilities, to watch superemitters and potential super-emitters across their life cycle, using low cost sensors such as those described by Collier-Oxandale et al. (2018). Similarly, to achieve higher precision on large offshore gas platforms, automated cavity-based analyzers can be linked to air inlets distributed around the platform (e.g., an “octopus” design, with air hoses feeding via a central multivalve to the analyzer).

In the Barnett Shale gasfield in Texas, superemitting sites accounted for roughly three fourths of total emissions (Zavala-Araiza, Lyon, Alvarez, Palacios, et al., 2015), which were 90% higher than inventory estimates and constituted 1.5% of natural gas production (Zavala-Araiza, Lyon, Alvarez, Davis, et al., 2015). Similarly, though with local variation, in four other major U.S. gasfields the bulk of emissions came from 20% of sites (Robertson et al., 2017). Gathering facilities are particularly important sources, especially the leakier installations: in the United States, Mitchell et al. (2015) found 30% of gathering facilities contribute 80% of the total emissions. Abandoned wells also contribute significantly in some regions (Kang et al., 2016; Townsend-Small et al., 2016). Similar patterns of superemission from abandoned wells and coal-mine vents were also found in Queensland and New South Wales, Australia (Day et al., 2015; Zazzeri et al., 2016). It will likely

need public intervention to locate and cap the multitude of leaking abandoned exploration holes in gas and coal extraction fields. This will at times demand deliberate significant public expenditure for old holes that are “orphans,” with no identifiable or solvent corporate owner.

Measurements by Pétron et al. (2012) and Karion et al. (2013) showed very high leakage rates in U.S. shale gasfields: In particular, in the Uintah gasfield in the western USA, Karion et al. (2013) showed emissions were as high as 6.2% to 11.7% of average gas production. These studies have been heavily cited in academic journals and were particularly effective in drawing attention to high loss rates. However, recent gasfield studies elsewhere (e.g., Schwietzke, Pétron, et al., 2017—see above) have found much lower leakage rates, typically well below 2%.

Leaks are not the only major cause of gasfield emissions. Schwietzke, Pétron, et al. (2017) and Vaughn et al. (2017) showed that in the Fayetteville Shale, in Arkansas, USA, deliberate manual actions drove major afternoon emission peaks. The western half of the gasfield had much higher emissions (1.8% of production) than the eastern part (0.8%): This was because the water content of the gas was higher in the west, and the higher but episodic emissions could be traced to manual unloading of accumulated water and other liquids. The liquid unloading was by deliberate action for water management.

Deliberate venting of unwanted gas is a major source of emission in many oilfields. O’Connell et al. (2019) studied heavy oil wells around Lloydminster, Canada. Over 40% of sampled well pads were emitting detectable methane, and of these, 40% emitted above the venting threshold beyond which mitigation was required by Canadian federal regulations. Similarly, in the Surat Basin, Australia, high-point vents on the coproduced water distribution lines were found to be important sources of emissions (Day et al., 2015). Major sources of methane emissions from venting and water handling (e.g., Figure 10; Iverach et al., 2015) should be feasible to control, as it should be easy for even ill-equipped local government authorities to detect and thus regulate them. Good design is generally better than post-installation mitigation. Thus, when developing new gasfields, consideration should also be given to reducing the number of high-point vents on the coproduced water distribution pipelines, and where such venting points are required the emissions from these points should be captured.

Development of effective low-cost leak detection and monitoring, coupled with targeting superemitters and working with operators to improve operational practices, particularly in water handling, can enable rapid, low-cost cuts in emissions (Mayfield et al., 2017). Mitchell et al. (2015) pointed to the benefits from simple vigilance: Gas processing plants were staffed and had comparatively low leakage; gathering facilities, typically unstaffed, had larger leaks. In major gasfield regions, even poorly resourced regulatory bodies should be able to afford vehicle-mounted detection systems of the type shown in Figure 3, which, by imposition of large fines for detected superemitters would likely pay for themselves.

Regulation, assisted by tax and penalty nudges incentives, is an obvious first step to reduce leaks and flaring from gasfield sources. In considering mitigation and safety (and perhaps profit), self-interest can be made to converge with greenhouse good. In some superemitter cases the economics of mitigating very large emissions are so attractive that high leakage simply denotes bad cost control and incompetent corporate leadership. If overall profit margins are, for example, 10%, a small investment to recapture a 10% leak will double profits, and enhance safety. Canada has introduced some controls, intending to cut methane emissions by 40–45% by 2025, but clearly ineffective as yet in the case of the Lloydminster emissions reported by O’Connell et al. (2019). Tyner and Johnson (2018), studying oil production sites in Alberta, Canada, used Monte Carlo simulations to show that a 45% reduction in methane emissions from flaring and venting would, per ton of CO₂ equivalent greenhouse warming, cost between about CAN \$2.5 to CAN \$–3 (i.e., a profit). Relatively small government tax incentives for mitigation and relatively large penalty disincentives imposed on leakage would likely serve to nudge the mitigation of emissions well beyond a 45% cut. In any nation, even such relatively lax requirements do help cut emissions, with the side impact of slightly enhancing national energy longevity and security of supply.

In 2016, as part of the United States’ plan to meet its Paris Climate goals, the U.S. EPA introduced the Oil and Natural Gas Sector New Source Performance Standards, with a variety of emissions reductions requirements wells and compressors constructed after September 2015, including regular leak detection and repair for wells and compressor stations. However, EPA has challenged this rule in court and has reduced the amount of monitoring required since the regulation was implemented. The U.S. Bureau of Land Management also

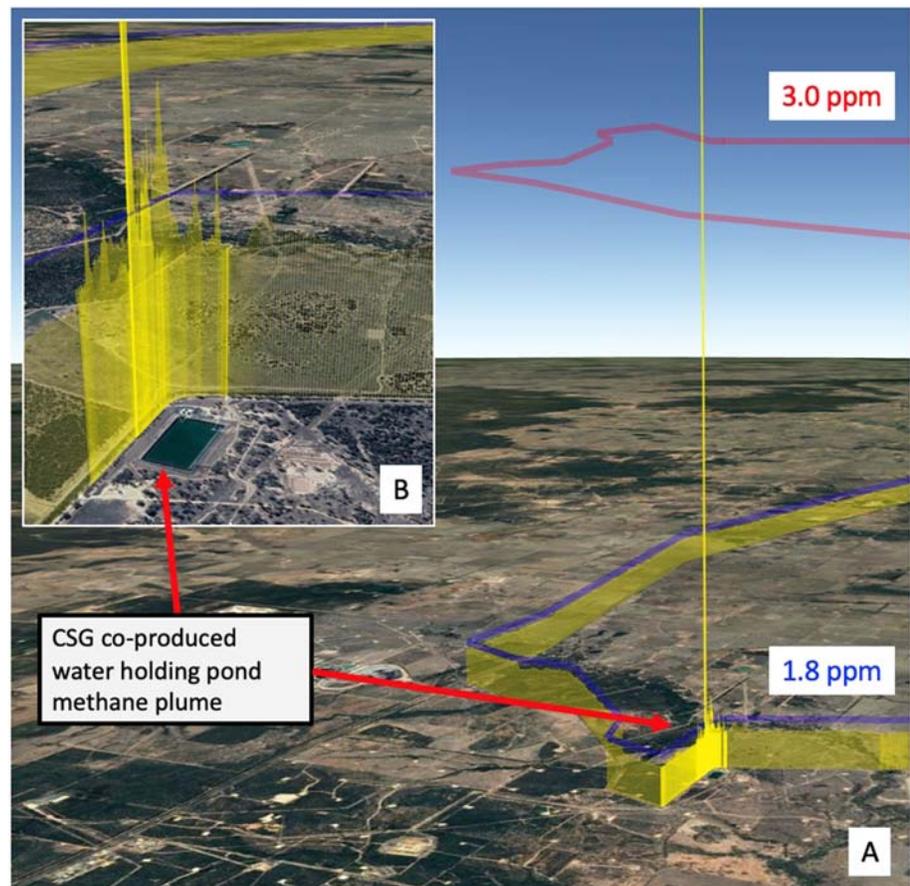


Figure 10. (a and b) Methane mole fraction plumes mapped during a mobile survey undertaken by UNSW and RHUL in the Surat Basin in 2014 and presented here. There are 22 similar water holding ponds throughout the Surat Basin. These ponds are required for managing the hundreds of megaliters of daily co-produced water associated with coal seam gas (CSG) production. Map data: Google DigitalGlobe.

implemented a Waste Prevention Rule in 2016 to reduce methane losses from oil and gas wells on federal land, but many aspects of that rule have also been repealed recently.

The intention of the 2016 EPA action was to *decrease* methane emissions by 300,000 short tons (unit from original citation – amount = 0.27 Tg) in 2020, and by 510,000 short tons in 2025 (0.46 Tg), compared to a baseline estimate. In contrast, under the revised 2018 EPA proposal, U.S. emissions were perversely expected to *increase* by 380,000 tons (0.34Tg) between 2019 and 2025 (Reuters, 2018a): In effect this emits future U.S. energy security to air. Interestingly, this 2018 EPA proposal was opposed by the industry, which is well aware of costs: An Exxon Vice-President stated “We support maintaining the key elements of the underlying regulation, such as leak detection and repair programs” (Reuters, 2018b).

4.2. Urban Gas Leaks

Urban gas leaks are major sources of methane emissions in most long-urbanized societies (e.g., Lowry et al., 2001; McKain et al., 2015; Zazzeri et al., 2017). Many cities in developed nations have built inventories of estimated methane emissions. However, using aircraft-mounted observations downwind of major U.S. cities, Plant et al. (2019) found that in current inventories, natural gas emissions were significantly underestimated. Moreover, in many places leak detection is still primarily by human smell. Although methane is odorless, this is effective as a safety measure because urban gas typically has the organosulfur compound mercaptan (CH_4S) added, which can be detected at a 0.14-ppb threshold by the human nose (Committee on Acute Exposure Guideline Levels, 2013).



Figure 11. Typical source facility: gas processing plant, Hong Kong. Photo: E.G. Nisbet.

In the Boston, USA, urban region, McKain et al. (2015) reported loss rates to the atmosphere of around 2.7%, as a proportion of the gas used. In the U.S. Los Angeles basin, He et al. (2019) found that about 1.4% of commercial and residential gas consumption is released into the air. Such loss rates are expensive, locally potentially dangerous, and an unnecessary greenhouse emission. Yet currently many jurisdictions pay little attention to mitigating urban gas leaks: for example, in the United States, Hopkins et al. (2016) concluded that “current mitigation approaches are absent or ineffective.” Even safety can be neglected. A recent example is the series of explosions and fires in the Merrimack valley, Massachusetts, USA, in September 2018 that killed one person and damaged over a hundred buildings (NTSB, 2018). Surveying the urban distribution network in Boston, USA, Hendrick et al. (2016) found that 15% of leaks surveyed were potentially explosive, and even small leaks could not be disregarded as “safely leaking.”

As with gasfields, there is strong evidence that a few major leaks constitute the bulk of emissions from urban distribution networks and that there is a “fat-tail” distribution of leaks. In Boston, 7% of leak sources contributed 50% of emissions (Hendrick et al., 2016) and in U.S. cities, it is estimated (von Fischer et al., 2017) that repairs to the largest 20% of pipeline leaks would cut losses by half. Leak prioritization by mobile measurement is now rapid, inexpensive, and effective (Weller et al., 2018).

Major leaks stand out strongly in vehicle surveys: Their identification does not need sophisticated quantification. Around the Royal Holloway location in outer London, gas governor stations, where pressures are controlled for domestic distribution, are particular point sources of emissions. It is likely that much could be done to cut emissions by identifying such specific “likely to leak” parts of the distribution network as mitigation

targets. There are strong grounds (Hausman & Muehlenbachs, 2016) for emphasizing leak reduction over pipeline replacement as a first option in mitigating emissions from a gas distribution system, especially with the marked advances in leak detection.

Leak quantification will help optimize mitigation: Identifying and repairing the worst emissions is an attractive and potentially profitable first step. There is indeed evidence that in the U.S. distribution pipeline leaks are being reduced (Lamb et al., 2015). However, it is difficult for gas distribution companies to make the jump between leak detection and leak quantification, and their criteria for repair or replacement may have different priorities (i.e., safety and proximity to structures, or cost, rather than greenhouse gas emissions). Currently, it is possible that widespread practice may be to mitigate methane leaks for safety reasons, but not to be overly concerned about greenhouse implications.

In addition to leak reduction, the replacement of century-old iron pipes is urgently and widely needed in Europe and the United States. This is necessary to cut greenhouse emissions and also to improve safety, by reducing unexpected catastrophic leaks, although the replacement process itself can be hazardous (NTSB, 2018). Robotic autonomous systems offer much (Robotics, 2017). The use of robotic systems removes the need to dig up the pipe to seal it. These systems can travel along pipes, detecting leaks and in principle also sealing many from the inside, including those in “live pipes filled with gas, and potentially giving longer lifetimes to old iron pipes.”

In the United Kingdom, replacement is typically achieved without major excavation by inserting new narrower-diameter higher-pressure piping within the older wider-diameter iron pipes. However, in countries with regulated local gas monopolies any investment in leak reduction increases the capital base of the gas industries and thus increases allowable gas prices. Hence proposals from industry to invest in new pipes may be blocked by regulators. For example, in the past the U.K.’s Office of Gas Supply (Ofgas) resisted environmental objectives (Danby, 1998). To keep prices down, U.K. regulators restricted corporate proposals for investment in leak reduction by pipeline renewal. While this may have been politically advantageous in

the short term, this extra leakage would have proportionately slightly shortened the useful life of the UK's domestic North Sea gas supplies, quite apart from the environmental damage.

4.3. “Habitual” Emissions From Domestic and Industrial Facilities

“Habitually” high methane air commonly exists in many places, ranging from near vents of domestic boilers and water heaters to the surrounds of industrial gas handling facilities such as compressor stations and gas distribution centers (Figure 11) (Schwietzke, Pétron, et al., 2017; Subramanian et al., 2015; Zavala-Araiza et al., 2017). In dealing with the high numbers of small leaks and emissions, low-cost sensors are becoming capable of detecting methane anomalies at the sub-ppm level (e.g., Collier-Oxandale et al., 2018). Just as smoke detectors are now ubiquitous, so methane sensors could be widely deployed to identify leaks rapidly.

Domestic heating systems emit methane on ignition because the first gas is only partly combusted. In the U.K. domestic boilers emit when they ignite for house heating and hot water. In some nations (e.g., Australia) “instant” hot water heaters that rely on burning gas are common: they emit methane each time the hot water tap is turned on. When a gas-burning system such as a domestic boiler or industrial compressor switches on or off, the combustion chamber is vented to prevent explosive build-up of unburnt gas. In addition, steady-state burning itself is inherently incomplete. In order to ensure all gas is oxidized and no stray gas leaks out, down to the ambient background level, the amount of oxygen in the system must be increased well above the stoichiometric level. The reduction in overall combustion chamber temperature caused by this excess airflow reduces the boiler efficiency by more than the energy cost of venting unburnt gas. Because unburned gas passing through can be significant when burners are first turned on, an alternative strategy to cut emissions may be to pass initial exhaust gas back into the burning chamber for a second combustion. To mitigate emissions, not all the methane needs to be removed: the challenge is to devise an inexpensive, robust system to reduce at least some of the excess methane in the exhaust.

Catalytic removal of a significant fraction of excess methane is feasible. Removing harmful trace gases is compulsory for vehicles: In a comparable way, in future methane removal from exhaust gases may become an attractive and feasible option for stationary gas-using installations. Catalytic reactors can be autothermal at methane mole fraction as low as 0.06% but for greenhouse mitigation the catalyst would need heating (Su & Agnew, 2006). Noble metal catalysts using noble metals such as Pt, Pd, or Rh are effective (Jiang et al., 2018) and in principle it should be possible to take advantages of economies of scale by using modified vehicle catalytic converters. But costs may be prohibitive. Gosiewski and Pawlaczyk (2014) estimate that 0.5 t of Pd would be needed to mitigate emissions from a single coal mine ventilation shaft. Unfortunately, currently, there has been little research on ultralean methane combustion using these catalysts (Jiang et al., 2018), though palladium-based zeolite catalysis is possible (Petrov et al., 2018). More generally, the use of zeolite-metal technologies to remove methane from air is promising (Jackson et al., 2019).

Inexpensive metal-oxide regeneratable catalysts (Buciuman et al., 1999; Döbber et al., 2004) have long been used in laboratory zero-air generators in reversible flow reactors. Typically, these reactors use catalysts such as hopcalite (a mixture of copper and manganese oxides, sometimes with cobalt or other metals), which work well at moderate temperatures (below 600 °C). These catalysts are typically rapidly made ineffective by water but can be easily regenerated by heating. Luo et al. (2012) showed full combustion of methane at 575 °C using biomorphic CuO-ZrO₂ catalysts synthesized on cotton templates. There is also great potential for the direct catalytic conversion of methane to methanol at low temperature (~220 °C or less) over zeolites that have been copper-exchanged (Narsimhan et al., 2016). UV-photocatalytic removal of methane may also be feasible (de Richter et al., 2017; Graetzel et al., 1989).

As methane catalysis only occurs at temperatures above several hundred degrees, a vital consideration in catalytic methane reduction is ensuring that the global warming caused by the extra energy demand needed to operate the catalyst is smaller than that of the mitigated gas, that is, that there is net benefit. For simple estimation the heat capacity of 1 m³ of air is about 1.2 kJ. If a catalytic converter raises the exhaust temperature by 250 °C before venting then the converter must reduce the exhaust methane concentration by well over 10,000 ppm (assuming heating at 0.1 g CO₂eq per kJ). This should be compared to exhaust concentrations from domestic solid-fuel boilers of 10–100 ppm (EU Ecodesign Directive 2009/125/EC).

One solution to this is to place the catalyst inside the combustion chamber where temperatures are already sufficient for catalysis and exhaust gas is still hot as it passes over. Vaillant and Gastec (1999) have

demonstrated such a system (Figure 12) and achieved 0-ppm exhaust emissions, an improvement even over ambient mole fractions. An alternative catalytic approach is to replicate the natural oxidation of atmospheric CH_4 by OH radicals—this is discussed further in section 6.

Finally, it is worth remarking that a more elegant solution to the problem of gas emissions from domestic boilers would be to eliminate the domestic gas boiler and its pipeline supply system entirely. Renewable energy sources such as wind and sunlight are sporadic. Inevitably, in power systems with high proportions of renewable generation, there are episodes of superabundant electricity, for example, at 3 am on a windy night, or on a quiet very sunny afternoon. As decarbonization proceeds, there will be many times of surplus supply at periods of low demand. Moreover, the costs of nuclear power are typically dominated by capital cost, and running costs are comparatively small. Thus, nuclear electricity can also be very inexpensive at 3 a.m.

Gas is primarily used either for domestic heating or for electricity generation. In European countries domestic central heating based on circulating hot water is common and wind, and solar power are widely and increasingly used, coupled with a baseload of nuclear power. Here, rather than using on-demand gas boilers, it is likely to become preferable to use surplus-period (e.g., postmidnight) electricity to heat water, and then release the heat into the home over the next day and evening. Hot water can easily be stored in an insulated tank for several days without losing much heat. Cold can obviously be stored also: In hot places it may become preferable to operate air conditioning by blowing air over a thermal mass such as ice chilled at a time when electricity was abundant. The intrinsically leaky urban gas pipeline network could be repurposed as cabling conduits both for telecommunication and, as vehicles turn electric, for electric power supply.

Closing the domestic gas network and switching to renewable-generated electric heating may have considerable synergy with the move to battery-powered electric cars. In the United Kingdom it would benefit long-term energy security if gas imports were halted and the declining local North Sea gas reserves were retained solely for use in peak-demand electricity generation, rather than used up for domestic heating.

4.4. Urban Wastewater and Sewage

Sewage systems host anaerobic methanogens and thus emit methane at all stages, from house drains to wastewater treatment plants (Liu, Ni, et al., 2015; Guisasola et al., 2008, 2009; Fries et al., 2018). Where biogas is extracted, it may be incompletely combusted. Drains are prone to methane explosions, while methane production in the sewage reduces the readily biodegradable chemical oxygen demand (Guisasola et al., 2008), detrimental to processes in the wastewater treatment plants. Thus, methane emissions from sewage constitute a safety and processing problem, as well as being a greenhouse concern. The wider problem of methane removal from sewers in the long gravity runs from buildings to wastewater treatment plants is potentially addressable by installing small bioreactors at intervals along the length of the sewers, and by extracting methane-rich air above the sewage for catalytic or soil methane oxidation.

Various techniques can be used to reduce emissions, but at present efforts have focused on the wastewater treatment plants. Here, methane recovery efficiencies over 50% can be achieved, for example, via a submerged underwater bioreactor (Giménez et al., 2012). Floating bioreactors on wastewater settling pools have achieved 67% methane oxidation in agricultural settings (Syed et al. (2017), and the technology may be transferrable to urban wastewater. Matsura et al. (2015) achieved over 99% removal of dissolved methane by a two-stage system using a downflow hanging sponge reactor to remove methane from an upflow of anaerobic sludge effluents. Similarly, using an upflow anaerobic sludge blanket reactor at ambient temperatures, Bandara et al. (2012) achieved good removal efficiencies of more than 50% in summer, as measured by chemical oxygen demand, though less than 40% in winter. No biogas was evolved. More generally, methane removal in wastewater treatment plants appears to be both feasible and inexpensive, partly self-financing in both reduction of safety risk and perhaps in providing methane for ancillary power.

4.5. Methane Emissions From Biodigesters and Biogas Generators

Biogas and oils are increasingly being produced by anaerobic digestion of various substrates such as domestic and industrial food waste, manure, or sewage sludge. There are significant benefits from this activity, including energy generation that replaces fossil fuel combustion, and mitigation of emission of methane from manure. However, as biogas is typically 50–70% CH_4 and 30–50% CO_2 , if part of the biogas is lost to the air, then there is a risk of cutting CO_2 emissions but increasing net greenhouse warming impact by the emission of methane.

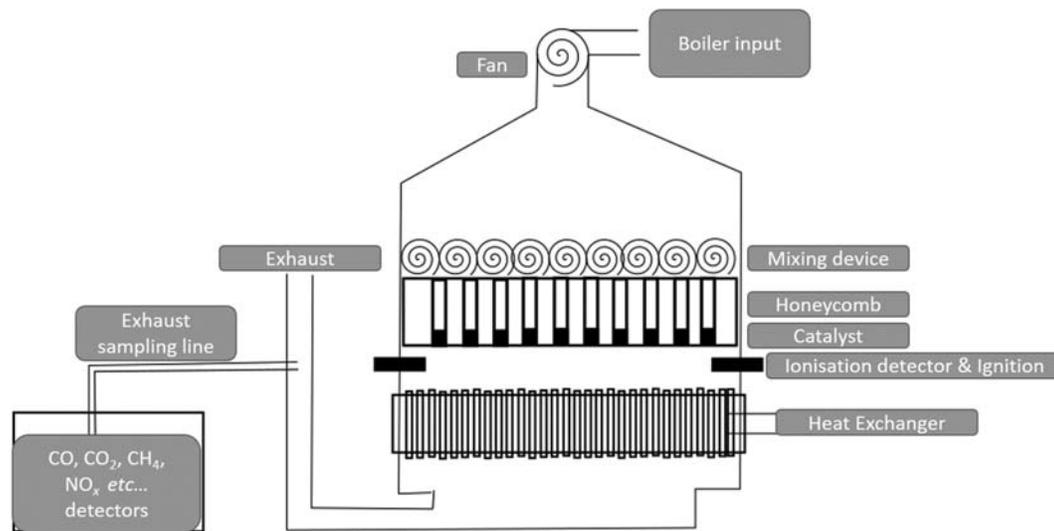


Figure 12. Concept for completely catalytic boiler system. The premixed gas air mixture is distributed over a heat exchanger. Modified on a concept from Vaillant and Gastec (1999).

The production of renewable energy from biogas plants is growing rapidly worldwide. In the United Kingdom, biogas plant numbers have increased almost 500% in the past 5 yr, with 607 operational biogas plants in 2018, and around 500 new plants being considered for construction (ADBA, 2018). The European Biogas Association anticipates that overall biogas production will be at least $50 \times 10^9 \text{ m}^3$ by 2030, capable of providing 2–4% of the EU's electricity needs by displacing methane from fossil fuels (EBA, 2019).

Unfortunately, biogas plants can have very high fugitive methane emissions. The emission rate depends on engine construction, plant design, and operation. Emissions can come from water handling facilities, gas engine exhausts, gas flaring, leaks from pipes, biogas upgrading units, tanks, etc., or from deliberate venting (Angelidaki et al., 2018; Duren et al., 2019; Fredenslund et al., 2018; Liebetau et al., 2018; Samuelsson et al., 2018; Scheutz & Fredenslund, 2019). Some emissions can be single large leaks or long-lasting bursts from pressure relief valves (Reinelt & Liebetau, 2019). In particular, Kvist and Aryal (2019) found that water scrubbers and pressure relief valves were especially significant sources of emissions, a finding similar to findings around many unconventional natural gaswells. To date biogas emissions have had comparatively little attention compared to leaks from other types of gas production facilities, even though biogas leakage rates are likely at times in excess of leakage from fracked gaswells. Typical methane emission rates range between 0.4% and 15.0% of the total methane production, and overall the methane leaks averaged 4.6% of methane production (Scheutz & Fredenslund, 2019). These loss rates are broadly comparable to the leakier end of the fracking range. Reducing methane emissions from biogas plants will probably take the type of painstaking attention to detail seen on better managed natural gas production facilities, for example, by reducing flaring, and restricting deliberate venting, and changing the way pressure relief valves operate. Kvist and Aryal (2019) showed that regenerative thermal oxidation had a major impact, in their study reducing the methane mixing ratio of from 6,900 ppm to an average of 13 ppm and suggested that in jurisdictions where methane emissions incurred financial penalties, this method of removal would become attractively advantageous to plant operators. Much can be done.

4.6. Landfill Emissions, Particularly in Less-Developed Countries

In addition to natural gas systems, large urban methane emissions also come from urban landfills. Saunio et al. (2016) cite bottom-up evidence for global emissions around 60 Tg annually. Landfill emissions can be readily mitigated, either by capture and piping, or simply by adding a thicker soil cover to act as host habitat for consortia of methanotrophic bacteria.

In some developed nations, particularly in Europe, landfill emissions are now heavily regulated. Figure 2 shows the effectiveness of this policy measure. Since 1990, U.K. annual emissions of methane from landfills

have been reduced to about a quarter of the previous amount (UK NIR, 2019). Worldwide, however, landfill regulation is weak, even in some jurisdictions with strong capabilities, such as parts of the United States. For example, Duren et al. (2019) found that landfills are the largest emitters of methane in California and dominate methane point-source emissions in the state (41% of total). Although such emissions are not well reported in state and national inventories (Duren et al., 2019), they are clearly targets for mitigation.

Ideally, waste inputs should be controlled, with organic wastes diverted to composters and biodigesters, and landfills themselves should be well lined and covered with geomembrane, and typically fitted with gas capture piping, delivering gas to fuel electricity generators. If the methane itself cannot be extracted and combusted to make power, biofiltration is effective in removal (Gebert & Gröngröft, 2006; Park et al., 2009). But such systems require both investment and ongoing commitment to management as pipes can frequently fracture and lead to large unidentified localized leaks. Moreover, linked facilities such as anaerobic biogesters can themselves be very high emitters of methane (see section 4.5). In particular, the working face of the landfill, which is where emissions are most active (Figure 13), necessarily remains uncovered, with the gas uncaptured.

Nevertheless, active mitigation of landfill emissions in developed nations is a long-established and effective priority and thus not the focus of the discussion here. In contrast, in many tropical nations, landfill methane emissions are wholly uncontrolled and are characteristically high, especially in India (e.g., Ghosh et al., 2019; Kumar et al., 2004; Lobert et al., 1990). Even in Kuwait, the landfill is a major source (al-Shalaan 2019) (Figure 3). There is often little interest in investment in methane emission control and there is poor management of the emissions in many cases. Fires often break out, either accidentally or deliberately. For example, South Africa, although it has strong landfill regulation on paper, has widespread illicit burning (Roberts, 2018). In less regulated tropical and subtropical locations waste is widely burned (Figure 14) and in many cities daily large scale burning of local waste piles equivalent to landfill occurs and is likely to emit both significant amounts of methane and much harmful air pollution.

Landfill soil covers host active consortia of methanotrophic bacteria, and thus, bacterial methane oxidation is an attractive option for low cost methane mitigation (Scheutz et al., 2009). In U.S. landfills with 0.3- to 1.2-m thick soil covers, typically clay, Chanton et al. (2011) showed that a third or more of the egressing methane ($37.5 \pm 3.5\%$) is oxidized by methanotrophs in these covers. While the uppermost soil crust may be dry, with depth the oxygen and nitrogen mole fraction decline and methane and CO_2 increase, especially in active faces and in landfills that are not membrane covered. Stable isotopes can be used to assess the impact of methanotrophy (Sparrow et al., 2019). Methane emitted from below passes upward into increasingly aerated soils, and methanotrophic oxidation rates in moist but not waterlogged soil can be extremely high (Whalen et al., 1990). Methanotrophic oxidation has been suggested by Stein and Hettiaratchi (2001) for mitigation around heavy oil wells and also sanitary landfills in Alberta, Canada. Moreover, soil oxidation can be effectively enhanced by simple techniques, such as enrichment with biochar (Reddy et al., 2014; Yargicoglu & Reddy, 2017).

The usefulness of such passive biocover systems was demonstrated by Scheutz et al. (2014), who studied an old unlined landfill, comparable to modern tropical landfills. They used a bioactive cover made of local compost materials such as garden and kitchen waste, and demonstrated an average mitigation efficiency of around 80%. Although the landfill they studied is in Denmark, even in freezing winter conditions the exothermic bioreactions maintained a high mitigation efficiency in the system, and optimal biosystem temperatures of 30–40 °C. This is because, although methane oxidation is strongly temperature dependent (Wang et al., 2011), landfills are strongly exothermic. Einola et al. (2007) found that exothermic methane consumption continues in landfill cover soil even in cold meteorological conditions, a finding that may have value for mitigation in cool-winter regions such as north China, central Asia, and high-altitude Bolivia and Peru.

Significant reduction in emissions, up to 96% in summer (Lee, Jung, et al., 2018), can be potentially achieved by inexpensive soil or compost coverings. Financially, Scheutz et al. (2014) showed that simple biocover mitigation was cost-effective: Indeed, it is possible that in some locations such as the Mediterranean it may be viable to use carbon credits to finance soil-cover landfill mitigation directly (Kormi et al., 2018). Moreover, soil management of emissions also contains unwanted smell and fire risks and is likely to gain local support (especially if the matured cover vegetation can then be given over to urban parks).

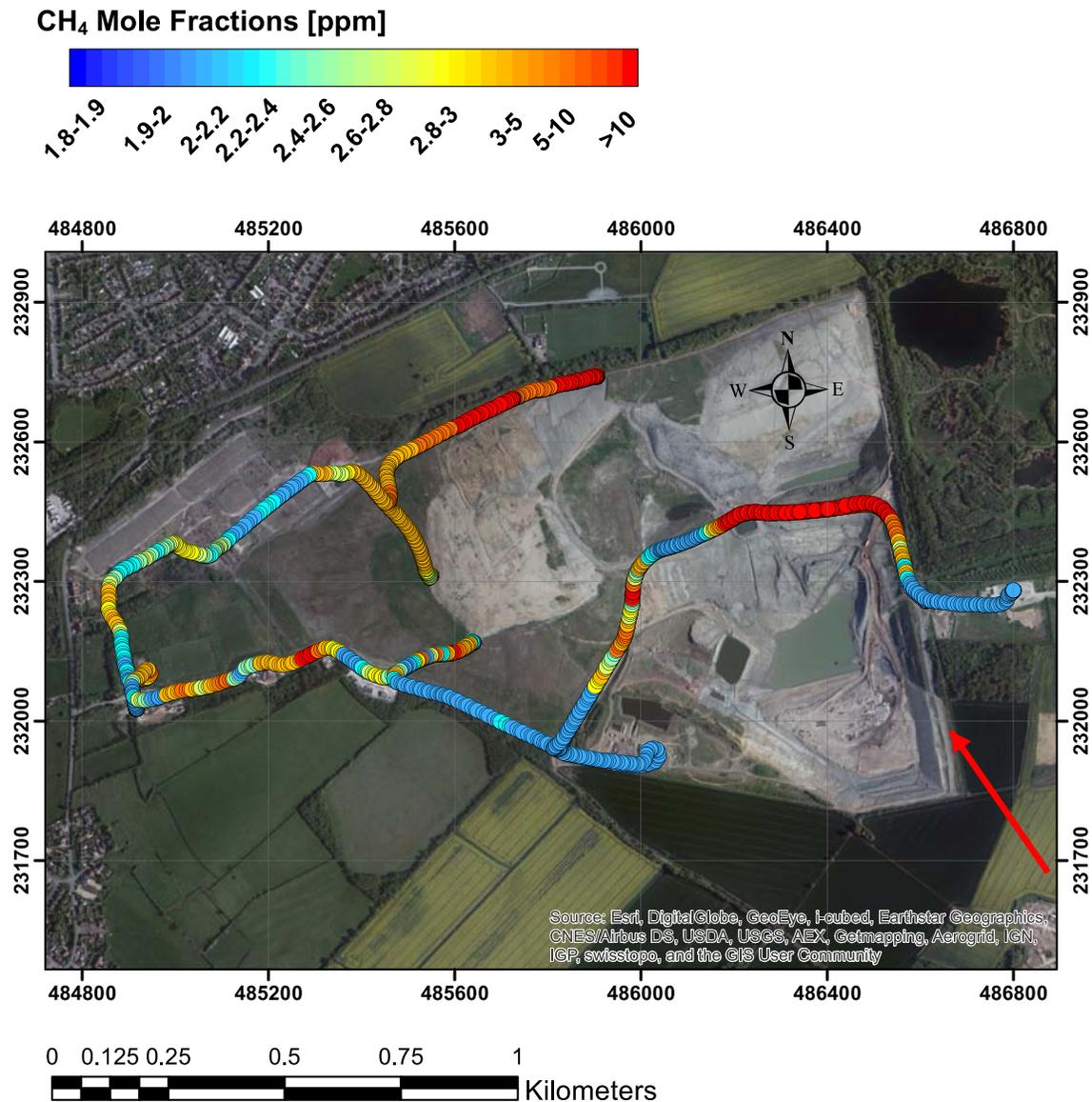


Figure 13. Vehicle-mounted mobile survey of U.K. landfill, superimposed on prefill topography. Note very high ambient methane downwind of active cells. Map data: Google DigitalGlobe.

Although methane mitigation in landfills in developed nations is typically complex and demands strong investment and monitoring, for less wealthy urban regions there is a wide array of simpler cheaper options to mitigate emissions that do not demand high capital investment or complex skills, and which can potentially be assisted within development aid budgets. Thus, where landfill gas extraction by complex piping is prohibitively capital intensive, soil biocovers teamed with careful attention to rapid covering of recently active faces offer high prospects of containing and mitigating emissions. Mitigation of emissions from composted heaps and from residual landfill can be achieved by methane reduction via bacterial oxidation in the soil covering the landfills (Abushammala et al., 2014; Boeckx et al., 1996; Sadasivam & Reddy, 2014; Serrano-Silva et al., 2014). In addition, in nations with high unemployment it is not expensive to adopt waste sorting for compostable materials.

With the rapid growth in tropical urbanization, methane emissions from tropical cities are probably now very large. Although they are very poorly studied and quantified, tropical urban emissions may be becoming significant in the global budget, and thus a mitigation target of growing importance. In particular, where



Figure 14. Waste heap fire in Pomona dump, Harare, Zimbabwe, November 2016. (Photo: T. and L. Broderick, Project MOYA).

regulatory capacity is limited, rather than attempt expensive and complex landfill gas recovery, as an initial mitigation method it may be more effective in newly urbanizing countries to adopt simpler strategies: cover the landfills with soil and sort the waste.

4.7. Deep Coal

Methane emissions from underground coal mining activities are important contributors to the global methane budget. They occur both when vented from coal mining and associated surface coal processing and from leaks from coalbed methane extraction (Moore, 2012). China, with the world's largest coal industry, is especially important (Thompson et al., 2015), with methane emissions dominantly from underground operations (Zhang & Chen, 2010). Ju et al. (2016) estimated China's underground coal mine emissions in the year 2007 at about 19 Tg.

Figure 15 shows mapped methane mole fractions around two U.K. coal mines, with well over 5-ppm methane widely present in places, especially coal piles and mine vents. Methane comes both from thermogenic (isotopically heavy) methane associated with the coal, and young microbial methane by reduction processes during and after mining (Krüger et al., 2008). Air ventilated from coal mines generally contains methane, often with mole fractions so high it is explosive or can be burned. Such methane-rich air, which can be removed by combustion in thermal reactors (Karacan, 2011), has always been of immediate safety concern to all coal mines.

Underground mine vents are emission hot spots, as shown in Figure 16, making such emissions a promising target for mitigation. Emissions estimates from Australian coalfields demonstrate that targeting the reduction of methane emissions from underground coal mine venting could yield significant mitigation benefits. In the State of New South Wales, Australia, there are 19 underground coal mines that produce 24% of the annual mined coal in the state (<https://www.coalservices.com.au/mining/statistics>). However, these vented emissions from underground coal mines account for 82.6% of the coal sector's reported emissions at the state level, 30.7% of the State's total methane emissions, and 8.4% of Australian national methane emissions (Australia's National Greenhouse Gas Accounts, 2017; <http://ageis.climatechange.gov.au>). They are an obvious target for mitigation.

Mitigating methane emissions from deep coal mines is not necessarily expensive. Holmes (2016) describes a 12-month trial involving comprehensive emission reduction in the Australian Hunter Valley coalfield, based on vigorous efforts to locate and seal leaks and to use measures such as pressure balancing to suppress egress. This work, entirely practical and using common sense, likely brought marked safety improvements and had little capital cost; it removed about 4 kt of methane or 80 kt of CO₂ equivalent (quantities and units cited as in

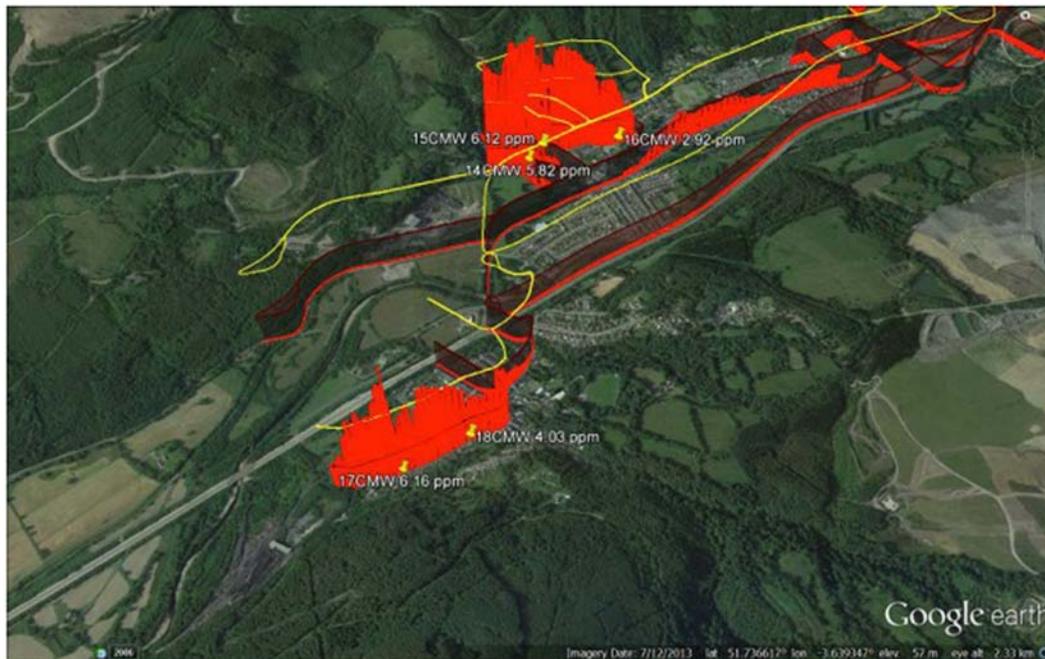


Figure 15. Google Earth view of methane mole fraction columns (red contours) measured along the transect downwind of Aberpergwm and Unity deep mines in Wales on 17 October 2013. The yellow line represents the 5 ppm mole fraction level, yellow markers samples location. Map data: Google, DigitalGlobe.

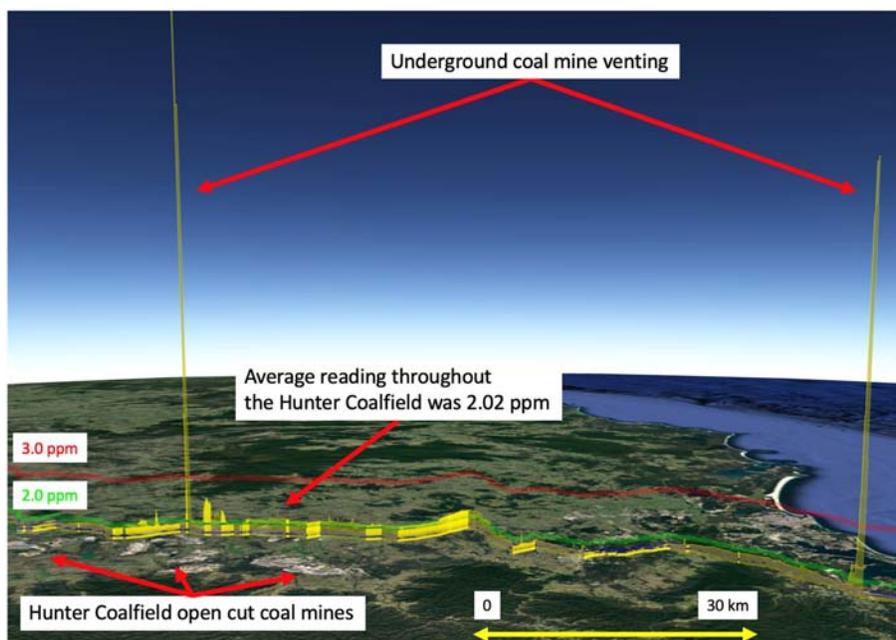


Figure 16. Mobile mole fraction methane measurements in the midcoast and Hunter coalfields of eastern Australia, recorded in 2014 (University of New South Wales and Royal Holloway, University of London). The background methane mole fraction for the region was 1.78 ppm. Emissions from the underground coal mines exceeded the measurement range of the analyzer (20 ppm), while throughout the Hunter coalfield the average methane mole fraction was 2.02 ppm. Map data: Google, DigitalGlobe.

the original source, Holmes, 2016, which used a Global Warming Potential of 21), for a cost of Aus\$1.28 (\$0.9USD) per ton of CO₂ equivalent. Such efforts are simple good management and should easily be implemented in a safety-conscious setting or with effective regulatory oversight.

Both working and closed coal mines also emit methane-rich air, well above ambient mole fractions but well below explosive-risk thresholds, which may not be of concern to mine managements on safety grounds but are important in terms of greenhouse gas emissions. This air has methane content that is too low to sustain a thermal reactor, but as with boilers and compressors, emissions can be mitigated in catalytic reactors. However, usually, the air is moist and the water degrades the catalytic effectiveness: Thus, the flow needs to be reversible to regenerate the catalyst regularly. For nonexplosive mixes, reverse flow reactors are effective in mitigating methane emissions from coal mines for air with ~0.3% methane and 5% water (Fernández et al., 2016). However, for mitigation purposes, air with methane as low as 5–10 ppm would be a target.

4.8. Open-Cast Coal

Open-cast (or open-pit or open-cut) coal mines are also a major source of emissions (Dontala et al., 2015). In contrast to deep coal, where the methane egresses from localized vents and openings, open cast mines have spatially spread-out emissions, which may be greatest near the active excavation face. Moreover, these emissions can be isotopically light, comparatively rich in ¹²C, indicating such methane is not thermogenic but is young, biologically made by in situ methanogens, in anaerobic settings (Zazzeri et al., 2016).

As with landfills, managing emissions from open cast coal mines should be possible. Many open cast mines currently remove the cover from many kilometer lengths of coal beds and then strip it in kilometers long benches, exposing large areas of uncovered organic-rich benches to develop anaerobic conditions just below the remaining surface. In contrast, landfill management seeks to expose the active face only, and rapidly to cover all inactive cells with aerobic soil that hosts methanotrophs. Changing open cast coal pits to short active benches and covering inactive surfaces quickly with methanotroph-hosting aerobic soil cover would likely reduce emissions substantially. More complex mitigations could be to ensure that the coal face is well aerated, and thus aerobic. It is also intuitively possible that arrays of high-powered UV-LEDs could be used to kill any surface methanogenic bacteria, but this has not been tested. Such changes would demand corporate cultural and management innovation and redesign, but not necessarily any major capital costs although the energy cost of the implementation must be taken in account to demonstrate net GWP reduction.

In the medium term, it is perhaps more likely that the entire coal industry will become uneconomic, except for coal destined for steel production.

4.9. Methane Created by Anthropogenic Biomass Burning in the Tropics

Emission of methane from deliberate biomass burning is globally important, mostly from tropical C4 savanna grasslands and crop waste and also from boreal and tropical forests. In managed boreal and dry tropical forests, fire suppression is controversial and the accumulation of leaf-fall and branch-fall fuel load may simply eventually generate larger fires. In the tropical C4 grasslands, however, under the seasonal shifts of the Intertropical Convergence Zone, fire suppression can be very advantageous, preventing valuable soil nutrients being lofted in the smoke plume and eventually deposited far away in the oceans.

Fire emissions are especially important in Africa (Andela et al., 2019; Andela & van der Werf, 2014; van der Werf et al., 2017; Zubkova et al., 2019), where burning occurs very widely in savanna grasslands (Figure 17), and in leaf litter fires in facultatively deciduous dry savanna woodlands. Burning can be accidental when crop waste fires spread into neighboring untended communal grazing land, or deliberate to bring on new green shoots of regrowing perennial grasses. Burning is also important in African cropland to clear crop residues, and as waste plastic burning.

As an example, at Bachok Marine Research Station (BMRS) on the NE coast of peninsular Malaysia, compelling evidence of burning emissions correlates with periods when the sea-breeze breaks down and land breezes dominate (Figure 17). During such episodes the methane mixing ratios at the station rapidly increase over a matter of minutes, along with increases in anthropogenic tracers such as CO₂, volatile organic compounds, and NO₂ (Dunmore et al., 2016), as well as combustion markers such as carbon monoxide (CO) and acetonitrile (CH₃CN) (e.g., Lobert et al., 1990). Although methane emission in Malaysia is more complex than this simplification (with isotopes of methane during land breezes suggesting a mixed source

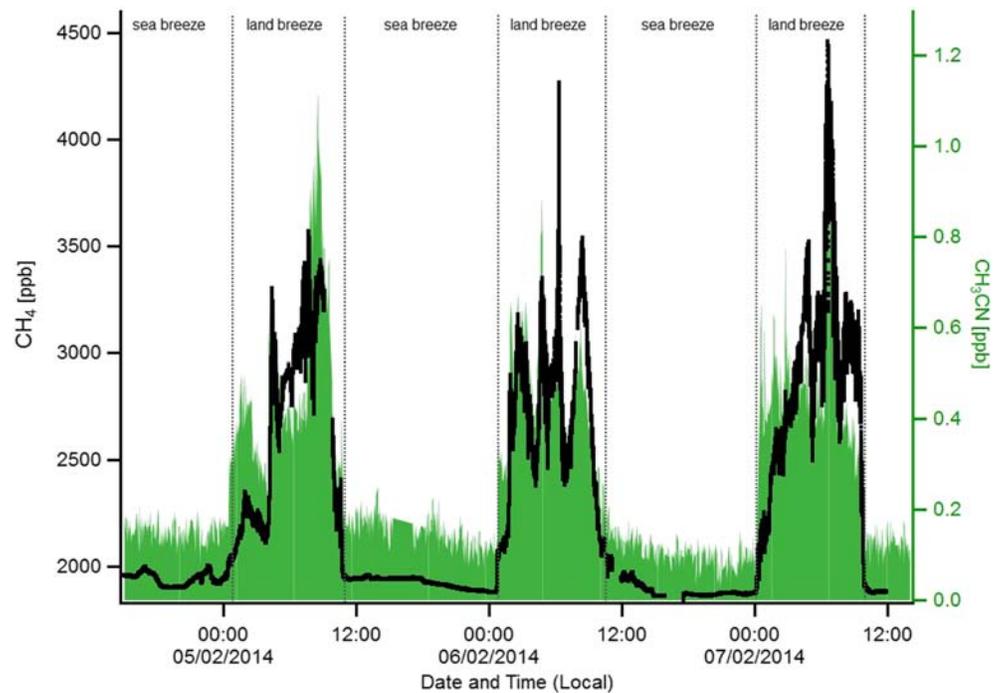


Figure 17. Continuous methane (CH_4) and acetonitrile (CH_3CN) measured over 3 days at Bachok station, Malaysia. The inlet height is 30 m, and the site is located 50 m inshore. The correlation between enhancements of CH_4 and CH_3CN is indicative of a strong burning contribution to the CH_4 enhancement (unpublished results, University of East Anglia/ University of Malaya).

including a biogenic source), it is clear that methane emission from all forms of waste disposal (rural agricultural crop waste, village debris, and urban waste) is widely underregulated, with unquantified amounts of methane entering the atmosphere. Such fires, in addition to producing methane, CO, particulates and black carbon, deplete essential soil nutrients such as nitrate and impoverish soil organic carbon, exporting them to the smoke.

Fire numbers vary strongly with meteorology and can peak during and after major *El Niño* events (Chen et al., 2017). Fuel loads are also strongly dependent on preceding meteorology (Zubkova et al., 2019). Satellite data sets may show a recent decline in global burnt area (Andela et al., 2017), and globally, Worden et al. (2017) showed evidence of a decline in biomass burning between the 2001–2007 and 2008–2014 time periods, which may reflect complex socio-economic changes as much as meteorology, though Forkel et al. (2019) found the trend is not significant. Zubkova et al. (2019) concluded that changing climate patterns and increased terrestrial moisture facilitated a decline in African fires in the 2002–2016 but pointed out that most African fires are human-caused and the fire-climate-human relationship is complex.

Wood and dung fuel burning is also widespread in the tropics, for cooking and heat, but slowly being replaced by electrification, which is progressing across most tropical nations. In India, widespread crop residue fires have been blamed for air pollution and may soon be controlled. In the savanna regions of Australia fire management is used to obtain carbon-offset units that may be sold to generate additional income, reducing the overall carbon emissions by doing carefully managed cool burns at the beginning of the dry season, rather than letting uncontrolled hot burn happen at the end of the dry season. Cool burns release less carbon dioxide, and the early dry season burns reduce the fuel available for uncontrolled hot burns that can naturally occur at the end of the dry season. This may reduce CO_2 emissions but not necessarily CH_4 , which is emitted under partial combustion conditions and may be increased with a moister fuel load (<http://www.cleanenergyregulator.gov.au/ERF/Choosing-a-project-type/Opportunities-for-the-land-sector/Savanna-burning-methods>).



Figure 18. Grass fire, August 2016, Harare, Zimbabwe. Photo: Lucy Broderick.

Land management is crucial in controlling burning. In Zimbabwe, for example (West, 1971), the commercial farmland, partly wooded and partly C4 savanna grasslands, was divided for many decades into “Intensive Conservation Areas” where vigorous fire suppression was practiced. As a result, soil nutrients were conserved and soil carbon built up. Savanna Africa has had tens if not hundreds of millennia of grass burning, both for crops in recent millennia and prior to that for hunting. Consequently, the region has developed a very widespread anthropogenic fire ecology (West, 1971). Suppressing burning clears the air and reduces methane emissions, but it changes the vegetation and the animals: fewer lion, zebra, and buffalo, more bush pigs, monkeys, and rats (West, 1971). With the abandonment of most commercial farming in the early 2000s, fire suppression halted in Zimbabwe. Interestingly, however, in recent years, regrowth of fire resistant trees has taken place in the unfarmed areas (own observations), and thus, C4 grass fires (Figure 18) may be eventually replaced by less intense C3 leaf litter fires.

Agricultural practice is widely changing, and increasing awareness of fire nutrient losses is likely. Widespread air pollution from smoke plumes that can be tens to hundreds of kilometers long can present major health hazards. Fire suppression is feasible and widely advantageous. Personal observation and discussion in Zimbabwe’s communally held lands (EGN) suggests that annually burnt areas may be substantially reduced by small-scale local reward programs that could reward schools or services in communities that had not burned their communal grazing land. Such programs could be validated inexpensively by end-of-season satellite observation. Public policy has also driven fires: For example, the European Union’s demand for palm oil biodiesel has played a role in burning forest in SE Asia to convert the land to palm oil plantations (Obidzinski et al., 2012; Susanti & Maryudi, 2016). Is it wise to reduce Europe’s CO₂ emissions by deforesting Indonesia? Thus, there is room for optimism that tropical biomass burning may be sharply reduced in the near future, relatively inexpensively, provided major climate events such as drought or heat waves do not intervene.

5. Emission Removal—Intractable Emissions

In many cases, emissions are regarded as “intractable” (i.e., cannot easily be stopped). Yet these need to be addressed. These emissions are typically disseminated, from many sources, some moving (e.g., animals). Such emissions are often regarded to be “inevitable” impacts of essential human activities like food production and occur where the mole fractions in ambient air are too low to consider combustion. Funding agencies have neglected the problem of methane extraction from ambient air, and where methane mitigation has been targeted, the focus has understandably been on emission reduction. In particular, the possibilities of methane removal on farms have had little attention. Currently, many agricultural emissions of methane can be seen as too intractable to consider for affordable mitigation.

In principle, as in landfills, methane removal around intractable sources can be achieved via soil methanotrophy. The movement of methane into the soil is controlled by gas diffusivity, which depends on the soil’s air-filled porosity, and the capacity of the soil for uptake of methane depends on soil moisture and aeration (Ball et al., 1997; Smith et al., 2018). Aerated soils such as sandy loams host methanotrophs, capable of living on the ~2 ppm methane mole fraction in ambient air, and thriving when the mole fraction is higher. Coupling biofiltration (Leson & Winer, 1991) with enhanced methanotrophy (Jiang et al., 2010; Yoon et al., 2009) could provide a powerful approach to a wide range of problems.

Afforestation has been much discussed as an attractive option for CO₂ capture (e.g., Humpenöder et al., 2014), but there has been less attention to the usefulness of afforestation in methane removal. Yet methanotrophy in forest soils can be very effective in taking up methane. Wu et al. (2018) showed that in central China the reforestation of cropland could create an important methane sink. They showed that recreating woodland nearly doubled methane uptake, and that shrubland, which took up ~37 $\mu\text{g}\cdot\text{m}^{-2}\cdot\text{hr}^{-1}$ (around 3.2 $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$), had nearly triple the methane removal of cropland. In natural settings, methane oxidation is highest in well-drained upland soils (Christiansen et al., 2016). More generally, coarse-textured moist but

not waterlogged forest soils show the most rapid methane oxidation rates (Levy et al., 2012; MacDonald et al., 1996), especially when the air can easily diffuse through the surface litter and the soil has a well-developed structure (Smith et al., 2003). Wide areas of the tropics, for example, in Africa, have been deforested in the past century and in many areas replaced by poorly yielding cropland. The primary reasons for reforestation are land restoration, improvement in perennial streams and carbon sequestration, to which the possibility of methane removal provides a small but useful added incentive.

6. Reducing Agricultural Methane Production

Agricultural methane emissions are very large, globally over 130 Tg/yr, with over 100 Tg/a from enteric fermentation and manure, around 30 Tg/a from rice cultivation, and in addition about 30 Tg/a from biomass burning, mostly deliberate (estimates from Saunois et al., 2016). This review journal focusses on specifically geophysical topics and the task of reducing agricultural methane emissions is a topic too large to be discussed here in detail: There is space only for a brief synopsis. Thus, the topic of methane reduction by changing pasture management and animal and human diets is not covered in detail here.

There is a very large literature on this (e.g., Benchaar et al., 2001; Buddle et al., 2011; Smith et al., 2008) but that is outside the scope of this review. The problem is challenging. Methane emissions from rural agricultural sources such as rice fields, cattle, and biomass burning (much of it deliberate to clear crop wastes or fields before planting) tends to be from widely disseminated or geographically spread out sources, sometimes in different locations from day to day or even hour to hour.

Integrating climate change mitigation goals into agricultural practice is urgent but poses great difficulties (Campbell et al., 2017; Fellmann et al., 2018). Cattle in the U.S. and Europe emit up to 400 g/day of methane (for an assessment see Niu et al., 2018). In particular, cattle feedlots and barns can be superemitters, as reported here using University of New South Wales/Royal Holloway, University of London measurements around Beef City, a 26,500 cattle feedlot in Australia (Figure 19). Similarly, Duren et al. (2019) found that dairies alone accounted for 26% of California's methane point-source emissions. Given the size of the total emissions, even small percentage reductions will have large impacts.

Methanogenic archaea are strictly anaerobic but find good habitats in the digestive systems of oxygen-breathing land ruminants. Methane is breathed out from all foregut fermenters—cattle, sheep, goats, and deer. Note that cattle primarily produce methane from the front end—bovine eructation dominates over flatulence. Moreover, it is not only the breath of animals that produce methane: If feces become anaerobic, then archaeal methanogenesis takes over. Thus, cattle pats in dry tropical grassland pastures are unlikely to produce much methane, nor do piles of elephant dung (RHUL observations), but large manure tanks in industrial farming are major sources (Veltman et al., 2018).

Some nonruminants are also methane emitters, although not on the scale of cattle. Hippos, camels, and llamas also process cellulose in similar ways, though not strictly ruminants. Hoatzin birds also ferment in the foregut, as presumably did dinosaurs in related clades (Wilkinson et al., 2012). Hind-gut fermenters, such as horses, elephants, and rhinos, appear to produce much less methane. Humans are a minor source of methane. Roughly 15–20% of humans breathe out methane (own observations; see also Polag & Keppler, 2019). This methane is presumably created in anaerobic parts of the digestive system and then transferred from blood to lungs.

6.1. Practical Mitigation of Methane Emissions From Farm Animals

Methane emissions from hard-to-mitigate sources, such as enteric fermentation, are very large (Rogelj et al., 2015; Reisinger & Clark, 2018), too important to ignore as intractable. For example, high methane mole fractions, often 10–100 ppm can occur in air around feed lots (Figure 19), cattle pens (Grainger et al., 2007), manure heaps, agricultural biodigesters (Flesch et al., 2011), and near active faces of landfills (Riddick et al., 2017; Zazzeri et al., 2015). Methane yield from manure is highly variable, depending on the diet fed to the cattle (Amon et al., 2007).

In U.S. cases, substantial reductions in methane emissions can be achieved by more sophisticated farm management (Dairy Coordinated Agricultural Project, 2019). Implementation of a beneficial whole farm management packages reduced carbon footprints substantially, by between roughly a third to more than 40%. Manure management is particularly effective, using improvements such as an anaerobic digester with

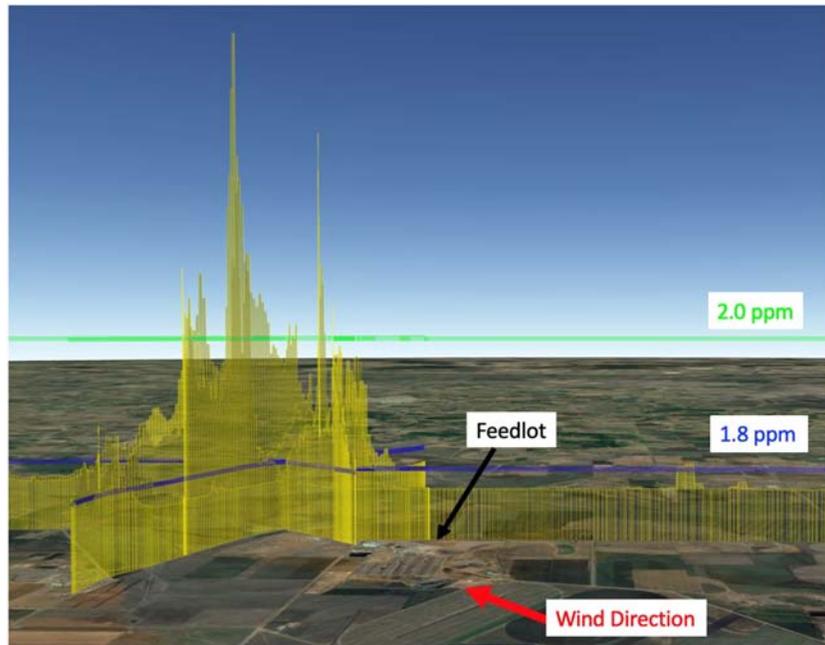


Figure 19. Beef City feedlot and processing facility downwind methane plume, Queensland Australia, mapped in 2014 (UNSW and RHUL). This facility processes more than 1,000 animals per day, while the feedlot can hold up to 26,500 head. Map data Google, DigitalGlobe.

methane capture for biogas, and sealed manure storage with gas flares (Veltman et al., 2018). This means manure tanks, lagoons, heaps, and stores are attractive first targets in efforts to mitigate agricultural emissions. Widely present, at sites like feedlots, barns and milking stations, they are localized and contained sources of high-methane air where emissions can be mitigated by careful manure management in oxidizing settings, including drying manure, using anaerobic digestion, or by catalytic oxidation (Pratt & Tate, 2018).

Van der Zaag et al. (2018) investigated two options for reducing emissions from manure—(1) separating solids and liquids and (2) anaerobic digestion. For solid-liquid separation they found an 81% average methane emission reduction compared to raw manure, on a per liter basis. For anaerobic digestion, the reduction was 59%, on average. More generally, in contrast to industrial-scale dairies with large numbers of animals in cattle courts, barns or feed lots, preferring “organic” grass-fed pasture-based dairy farming, with rapid oxidation of manure in the field, should also reduce methane emissions.

Globally, most cows live outside the United States and Europe, with the heaviest cattle populations in the tropics and subtropics, especially in India with 150 million small dairy farmers, and in small-scale beef and dairy farming in tropical Africa (Gilbert et al., 2018) where few technical resources may be available. Here management practices are very different and perhaps less amenable to emission reduction, but per-cow emissions may be lower than in industrial farming as cattle are largely free to roam, grass and browse-fed, and manure is typically left on grazing lands or collected and burned as fuel.

Although changes in cattle diet and management, such as open grazing on nitrogen-managed pastures (Warner et al., 2015) rather than stall feeding, can be effective in cutting emissions, these approaches (not discussed further here, but accessible in veterinary journals) are often costly, only partly effective, or demand high skill levels by the farmer. They may be difficult to apply in nations such as South Sudan, with very high cattle population densities but low governance capacity. Other innovative efforts to use microbiological intervention, including diet changes, to reduce emissions from cattle and sheep are also not discussed here as this very large topic is outside the scope of this essentially geophysical review (but see Smith et al., 2008).

Where animals are housed, particularly in winter, air is often extracted, and there is thus an opportunity for mitigation of methane emissions. Air extracted from intensive pig and poultry production can also generate

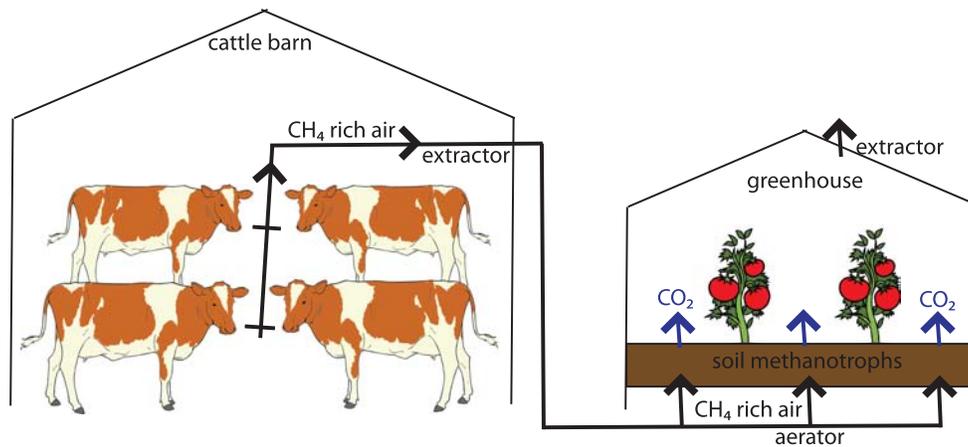


Figure 20. Conceptual model of low-cost reduction of emissions from cattle. Extractor fan removes air under mouths of cattle in milking shed or winter cattle court. Air is pumped to biofilter growth medium in a greenhouse, polytunnel, or under plastic field covers. Power from rooftop solar or wind.

high methane mole fractions (Van der Heyden et al., 2015). In Europe some facilities housing cattle also have air extraction systems installed. Note that methane removal would not have to be complete nor continuous, merely substantial. The ethical balances in industrial animal farming are not addressed here.

6.2. Practical Mitigation of Methane Emissions From Rice Paddies

Methanogenic habitats are created in wetland rice paddies. Zhang et al. (2016) found the magnitude of emissions depended mainly on the water management procedures. Studying the 2000s decade, they estimated annual global rice field methane emissions to be under 20 Tg under intermittent irrigation, but nearly 40 Tg under continuous flooding in the 2000s. It is in concept possible to reduce methane emissions from wetland rice production, both by directed water management and by selection of rice varieties and application of fertilizers and organic residues (e.g., Smith et al., 2008). However, as with diet-management for cattle, widespread reduction of methane emissions by deliberate management of rice production would likely demand high skill levels and strong governance, and perhaps also acceptance of lower yields. Moreover, in some cases there can be a trade-off between methane emission and N_2O emission (e.g., Naqvi et al., 2018; Smith et al., 2008). Net benefit to the greenhouse is not necessarily obtained by cutting methane output. Another interesting option is the use of biofertilizers to enhance methanotrophy in fields (Singh & Strong, 2016).

6.3. Conceptual Models—Future Possibilities

One approach for dealing with methane emissions from intensive animal farming may be abiotic removal via ambient-temperature UV-photocatalysis of methane and water using TiO_2 substrates, in a process analogous to the natural oxidation of atmospheric CH_4 by OH radicals (de Richter et al., 2017; Graetzel et al., 1989). For example, Costa et al. (2012) and Guarino et al. (2008) painted the walls of pig barns with TiO_2 paint and hung UV lights. They both measured significantly reduced methane in the barn exhaust. However, Maurer & Koziel (2019) and Liu, Maghirang, et al. (2015) ran exhaust air through external catalysis reactors, and neither of these saw a methane reduction. Thus, it is possible the pig barn experiments did not actually react any methane, but rather they may have UV irradiated and killed the methanogens in the manure on the floor. A further problem is that OH radicals will oxidize the most reactive molecules first, and thus methane last (Hay et al., 2015). This means that photocatalysis of methane in barn air laden with organic molecules is more likely to attack other species before methane: this is good for smell reduction but not methane removal. Photocatalysis in clean ambient air however is potentially more feasible (de Richter et al., 2017).

Biological options may in future be more attractive than abiotic approaches for methane removal. Most biofilter studies have investigated removal of methane at mole fractions in excess of 1,000 ppm (Melse & van der Werf, 2005; Ramirez et al., 2012), but Girard et al. (2011) report effective use of biofiltration in removing up to 45% (and potentially up to 65%) of methane emitted from swine slurry in Canada, at mole fractions as low as 250 ppm (Ramirez et al., 2012). However, significant extra study will be required due



Figure 21. High methane in ambient air around Zimbabwean cow. Current RHUL study. Photo: Lucy Broderick.

to the extremely different conditions under which a low methane concentration biofilter will operate, and there is a risk the biofilters may convert ammonia into nitrous oxide, another powerful greenhouse gas (Melse & Hol, 2017). Yoon et al. (2009) modeled the behavior of trickling biofilters using standard methanotrophic bacteria and found that operation at ~500 ppm was possible. Potentially, extraction should be feasible even down to ambient air mole fractions if soil microbes that can consume ambient concentrations are used; however, rates may be slow and some cells probably cannot survive in a standard trickling biofilter as the falling water shears cell walls and cell loss through shearing is greater than cell growth (Yoon et al., 2009). Passive airflow biofilters may offer a more favorable environment.

Unfortunately, despite their apparent promise, one significant concern with using biofilters at low methane concentrations is that their operation may inadvertently increase other greenhouse gas emissions, in particular emissions of N_2O (which has a lifetime of over a century and a global warming potential 10 times greater than methane over 100 yr). Specifically, a review of bio-filters in the swine industry has shown that biofilters *increase* the amount of N_2O present of up to 400% (Van der Heyden et al., 2015, and references therein). The proposed biochemical pathway is the conversion of NH_3-N with 10–40% efficiency, with long residence times of the air in the filter, necessary for low concentration methane removal, positively correlated with emission. It has been suggested that governments should be reluctant to permit biofilters to be

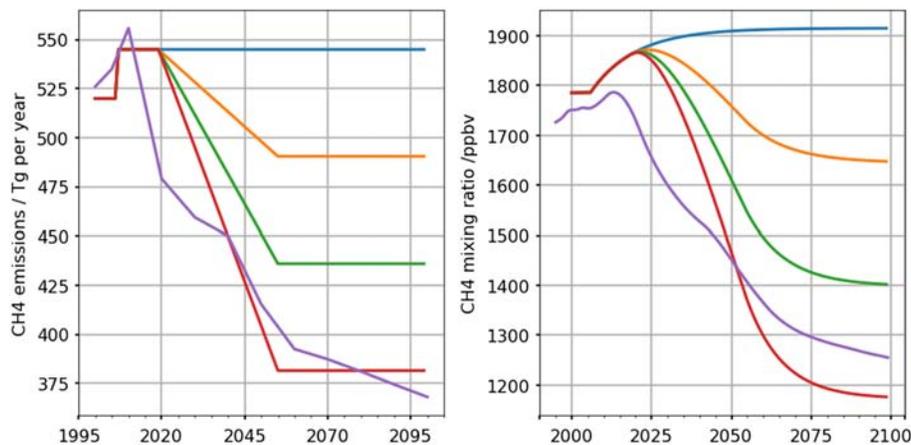


Figure 22. Simple box model to show the potential impact of mitigation. Left-hand panel shows modeled emissions. Right-hand panel shows the results in terms of the methane mixing ratio. Purple line approximates Pathway RCP2.6 (Collins et al., 2013; Rogelj et al., 2012), which is compliant with the Paris Agreement. Red line assumes starting global emissions of 520 Tg/a, increasing with a step change of 25 Tg/a in 2007–2020, to match the recent observational record (Nisbet et al., 2019). From 2020, the paths diverge. Blue line—no change in emissions after 2020. Orange line—10% cut in emissions spread linearly over the period 2020–2055, followed by stable emissions. Green line—20% cut over period to 2055, followed by stable emissions. Red line—30% cut in methane emissions to 380 Tg/a in 2055, followed by stable emissions, which cuts the atmospheric mixing ratio of methane to about 1,200 ppb by the Year 2100.

installed at livestock operations (Melse & Hol, 2017). Further study of N₂O production in biofilters is urgently required.

Figure 20 shows a conceptual model we have originated, suitable for winter cattle barns in northern Europe: air (~100 to 200 cows, with air having 10–100 ppm CH₄) is extracted from the barn via pipes laid at low level along food troughs. In the conceptual model, the air is then drawn through adjacent greenhouse plant soils or growth media inoculated with methanotrophs. Scheutz et al. (2009), reviewing studies of soil uptake performance, showed that consumption rates of 100–400 g·m⁻²·day⁻¹ or higher can potentially be achieved. Extractor fans and light heating could be powered by solar or wind electricity and operate intermittently when power is available. Thus, if the greenhouse is large enough (e.g., 50–100% the area of the cattle barn, assuming typical per-cow areas), then in principle it may be possible to remove a substantial part of the emitted methane, reducing the methane mole fraction toward ambient amounts in the air that is finally extracted to the atmosphere. But this attractive conceptual model hides a problem. Ammonia would be converted to N₂O, which is an important a long-lived greenhouse gas.

The viability of such conceptual model will depend not only on demonstrated effectiveness and size of the soil biofilter (or other material, such as gravel, woodchip or biochar) needed but also on demand (number of greenhouses installed) and financial subsidy. Careful consideration of the biofilter energy use is also required to ensure that the energy used to operate a filter with dilute ppm-level emissions does not have a higher warming potential than the emitted methane. Renewable energy sources should remove this concern. In northern temperate countries, viability of methane removal by twinning greenhouses with winter cattle barns could be enhanced by incentives favoring local greenhouse products, rather than long distance trucking or airfreighting of winter fruit and vegetables from warmer regions.

7. Summary of Priorities

The U.S. EPA's Greenhouse Gas Equivalencies Calculator (<https://www.epa.gov/energy/greenhouse-gas-equivalencies-calculator>) gives a good perspective on the options for reducing climate warming impacts.

The measures advocated here to reduce methane emissions are widely varied, and mostly specific to the sources considered. For methane mitigation, costs vary. Some are inexpensive but others will demand substantial investment or tax incentives, costly either for producers or governments. All rely on well-understood technology and are well proven, with the exception of some options discussed in section 6.3, such as the application of methanotrophy to remove ruminant emissions. For a review of frontier ideas and technology, many of which are likely to become economic to apply in the next decade, see Pratt & Tate (2018) and Jackson et al. (2019).

Mitigating gas emissions from the fossil fuel industry is an important target. In the gas industry, better and more comprehensive monitoring programs are needed (Fox et al., 2019). Many emissions are deliberate vents, that can be much reduced by better design and firmer regulation. Reduction of leaks at production sites can be driven by a mix of regulatory change, penalty fines, and small tax incentives, with little demand on the public fiscus. In the industrialized nations, the urban gas distribution network is leaky and antiquated. Cheap off-peak renewable or nuclear electricity may make it possible to abandon gas entirely for domestic purposes, and repurpose the pipeline networks for telecommunications or power delivery.

Cutting landfill emissions is a priority. In Europe there have been sharp reductions some years ago, but it is possible the progress is stalling. In the United States and China, there is much to be done. In the tropics, the work is barely starting. Uncontrolled emissions from huge landfills in cities like Delhi (Ghosh et al., 2019) not only damage the quality of life of local people but hurt the environment worldwide. The prime responsibility is with local jurisdictions, which should share in the burden of maintaining planetary health.

Mitigating methane emissions from agriculture (Smith et al., 2008) is an immense topic. The most obvious target is to cut emissions from manure tanks and lagoons around large cattle feed lots and barns. These sources are easily located and aerated, to limit anaerobic methanogenesis. More generally, there is a need for better comparative studies of grass-fed versus stall-fed cattle. Intuitively, grass-fed cattle with manure falling in open fields should be less methane-emitting than industrial cows (Figure 21).

Cutting agricultural ruminant populations by changing human diets reduces methane emissions (Poore & Nemecek, 2018). There is enough food for a substantial reduction if food is better shared and wasted less (Campbell et al., 2017). The broader impacts of going further are less clear. Cattle are essential to India, the world's ruminant super power, and in moist tropical Africa. The trade-offs between providing human food from local ruminants compared to importing arable crops are complex, even if cultural factors are ignored. Pasture lands are usually land that is too dry, too hilly, too stony, or too infertile for crops. Terminating production from the 22% of the planet's ice-free land surface that is pasture (Ramankutty et al., 2008), and replacing it with fully vegan (nonanimal) food would require more fertilizer-intensive farming of existing arable land and likely expansion of crop production into the remaining ploughable forest land in the moist tropics.

Such questions as yet remain open. Detailed whole-chain studies are needed that fully account for all impacts, both direct (e.g., greenhouse gas emissions, including CO₂ and N₂O) and indirect (biodiversity impact, etc.). Careful whole-impact thought is essential before advocating abandoning food production from grassland pastures and replacing it by intensive crop farming elsewhere.

8. Outlook

What will be the impact of successful mitigation? Is it worth even attempting?

The recent unexpected strong growth in the atmospheric methane burden challenges the feasibility of meeting the 1.5 °C target of the UNFCCC Paris Agreement (Nisbet et al., 2019). Collins et al. (2018) showed that, if the Paris target is to be met, there is a simple relationship between methane in the Year 2100 and “allowable” carbon emissions to that year: -0.27 ± 0.05 GtC per ppb methane. Thus, cutting methane sharply allows more relaxed CO₂ targets. Conversely, inaction on methane would imply stronger CO₂ (or other forcing agent) targets. However, Solomon et al. (2013) pointed out that although emphasis on a short-lived forcing agent like methane will indeed help to moderate warming, if that emphasis comes at the expense of efforts to reduce CO₂ emission, then short-term gain is paid for by greater long-term warming. Thus a “two-basket” policy is wiser—to cut *both* CO₂ and methane burdens. Cain et al. (2019) propose the use of CO₂-warming-equivalent emissions (*CO₂-we*) to assess how different greenhouse gases contribute to global warming. Using *CO₂-we* emissions allow methane and other greenhouse gases to be brought into the concept of a carbon budget and show how much each gas contributes to warming.

Figure 22 shows what can be achieved, showing results from an atmospheric box model using a simple CO, CH₄, and OH chemical mechanism, following Prather (1996). Inputs include a prescribed OH source, along with CH₄ and CO emissions. For a small (5%) perturbation in methane emissions, the box model gives a feedback factor (ratio of fractional change in concentration to fractional change in emissions) of ~1.5, agreeing well with values derived from other studies, in the range 1.3 to 1.7 (Holmes et al., 2013; Prather et al., 2001). Box model results for different possible methane emission reduction paths suggest that emission reductions of ~25 % are required to bring global methane mixing ratios back in line with RCP 2.6 projections by 2100.

Figure 22 implies that a cut of about 30% in total methane emissions—for example, 150 Tg/a or more—is needed to bring methane back to a pathway comparable to RCP2.6. To put this into context, the finding by Shindell et al. (2017), that as much as 110 Tg CH₄ per year of abatement is possible by scaling up existing technology and policy options, offers an attractive route. Note, however, that RCP2.6 is only illustrative, as one of many possible paths to the Paris Agreement goals. But if methane reduction does not happen, in order to achieve the Paris Agreement goals, other greenhouse gases will have to be reduced more substantially in place of methane. Are these large methane reductions feasible? They should be, if very substantial cuts in gas and coal industry losses and landfill outputs are partnered with significant reductions in agricultural emissions. Methane mitigation has no magic bullet: the sources are too diverse.

Science can only inform, not choose. Successful mitigation will require implementation of a wide array of differing approaches, each appropriate to specific circumstances and source types. Wise policy-making depends on judging the balance of cost and benefit: Given the overall international reluctance of democratic taxpayers to attack emissions, what is the most acceptable compromise? Information is vital—

would the 2018 suspension of methane rules by the U.S. EPA (EPA, 2018; Reuters, 2018a, 2018b) have happened if public information had been better? Given the existence of many “no regrets” options for methane mitigation, for example, eliminating leakage of methane that can otherwise be used, or consuming fewer products from intensive industrial-scale animal husbandry, it would be foolish not to act, but there is a danger in acting without considering full whole-impact consequences.

The challenge is considerable: Where should resources be allocated? How should global warming potential be assessed? How can local, regional, and global impacts be balanced? If the removal of methane demands energy, as is likely, it is also likely that the energy will have been generated by a process that emits CO₂. Will the mitigation of methane increase CO₂?

Striking a balance between methane reduction and CO₂ reduction involves political judgment. The evidence presented in this review suggests the methane burden could be reduced significantly by an investment in mitigation that is comparatively modest in comparison to the costs of parallel and necessary measures to reduce CO₂, and much less than the cost of climate change. Advances in detection, characterization and quantification of methane emissions mean that methane mitigation is becoming much easier, which potentially may tilt the near-term balance of priority in favor of methane reduction.

The task is that of Sisyphus, who pushed a boulder up a slope only to find it rolled back just as quickly. It is necessary to support the growing global human population while reducing the downward risk of breakdown in the natural atmospheric maintenance system. Yet, though Sisyphus made no progress in pushing the boulder up the hill, a better hero, Hercules, did succeed in a tougher task—he cleared the Augean stables by adopting new technology.

Mitigating methane will need a global effort. It is not only Europe, North America, Australia, and East Asia that need to act. Commitment is needed also from Asian and African tropical nations. Fire management and covering landfills are obvious immediate actions that will bring rapid air quality improvement as well as helping the global greenhouse. Burden sharing is important. International meetings on greenhouse gas reduction often focus on emissions from developed nations, but there are many actions, many at comparatively low cost (such as landfill cover) and with strong local benefits (such as fire reduction), that can be implemented by tropical nations.

The greatest long-term driver of change is human population growth and food needs, particularly in the tropics. Diets can change. Population growth slows wherever nations provide better education, especially female education, and improved living standards. Those factors are outside the scope of this discussion. Nevertheless, within the narrower focus of the world in 2020, application of the technical options discussed here could indeed slow methane growth, and soon. But if methane is not brought under control, the consequences are severe. The warming may feed the warming (Dean et al., 2018). Methane mitigation is important and not enormously costly. It is possible, and it can be done quickly. If the Paris Agreement is to succeed, methane needs attention.

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