Mitigating methane: emerging technologies to combat climate change's second leading contributor

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Dedication – Sadly, Dr Kevin Tate passed away during the final stages of producing this manuscript. Kevin will be remembered as an outstanding scientist, inspiring others with his unwavering commitment to tackling climate change.

Abstract

Methane (CH₄) is the second greatest contributor to anthropogenic climate change. Emissions have tripled since pre-industrial times and continue to rise rapidly, given the fact that the key sources – food production, energy generation and waste management – are inexorably tied to population growth. Until recently, the pursuit of CH₄ mitigation approaches has tended to align with opportunities for easy energy recovery through gas capture and flaring. Consequently, effective abatement has been largely restricted to confined high-concentration sources such as landfills and anaerobic digesters, which do not represent a major share of CH₄'s emission profile. However, in more recent years we have witnessed a quantum leap in the sophistication, diversity and affordability of CH₄ mitigation technologies on the back of rapid advances in molecular analytical techniques, developments in material sciences and increasingly efficient engineering processes. Here, we present some of the latest concepts, designs and applications in CH₄ mitigation, identifying a number of abatement synergies across multiple industries and sectors. We also propose novel ways to manipulate cutting-edge technology approaches for even more effective mitigation potential. The goal of this review is to stimulate the ongoing quest for and uptake of practicable CH₄ mitigation options; supplementing established and proven approaches with immature yet potentially high-impact technologies. There has arguably never been, and if we don't act soon nor will there be, a better opportunity to combat climate change's second most significant greenhouse gas.

Key words

Climate change, cross-sector, methane, mitigation

Introduction

Anthropogenic emissions of methane (CH₄) are second only to carbon dioxide (CO₂). According to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change ¹, annual anthropogenic CH₄ emissions total approximately 8 gigaton (Gt) on a carbon dioxide equivalent (CO₂-e) basis. Based on the IPCC's recently revised Global Warming Potential (GWP) factor of 28 for CH₄ ¹, CH₄ emissions likely contribute more than 20% towards total anthropogenic greenhouse gas (GHG) emissions, mainly from biomass burning, wetlands, rice paddies, enteric and waste emissions, and fossil fuel use ². There is also increasing evidence that the vast store of CH₄ in frozen soils in the Arctic is now being released as these soils thaw ³. Moreover, other stores of previously untapped CH₄, such as deep ocean sediment hydrates and clathrates, are currently being targeted as potential alternative energy sources ^{4, 5} and the risk of substantial indirect CH₄ emissions from these activities has triggered concern ⁶. This contribution to climate change from direct and indirect anthropogenic CH₄ emissions is clearly being recognised within the science and engineering spheres, as evidenced by the recent development of organisations such as the Global Methane Initiative and the Global Research Alliance.

Anthropogenic CH₄ sources vary in magnitude and are produced across a diverse range of industries and sectors. Yet, despite CH₄'s importance to climate change, few articles exist regarding technical mitigation opportunities across the key emission sectors. In 2012 there was a concentrated effort in this area with four overview papers published reporting emission estimates and mitigation options for the major CH₄ sources ⁷⁻¹⁰. The mitigation options discussed in the reviews by Höglund-Isaksson ⁷, Karakurt, et al. ⁸, and Yusuf, et al. ¹⁰ were presented from a high-level working principle perspective – i.e., logistics of gas recovery, collection and potential use as well as land use management practises to decrease emissions.

Moreover, these reviews focused on mitigation potential for individual sources. By contrast, Stolaroff, et al. ⁹ in their review recognised the potential for cross-sectoral CH₄ mitigation potential and presented a range of CH₄ mitigation options from a technology perspective. Since then, new scientific and engineering advances have paved the way for enhanced CH₄ mitigation technology development across several key contributing sectors. These technologies range in maturity from fledgling concepts, to field-tested systems that are now ready for full-scale deployment and validation, right through to commercially-viable practices.

In this review we springboard off the work of Stolaroff, et al. ⁹ and other earlier reviews to highlight the latest developments and opportunities in CH₄ mitigation technologies. Our review focuses on advances in this field over the past five years which have progressed on the back of three rapidly evolving disciplines: 1) material sciences; 2) microbial ecology; and 3) design and engineering. We largely avoid focusing on recent developments in enteric CH₄ mitigation using feed modifications, vaccines and animal selection – a wealth of thorough reviews evaluating the progress of these approaches can be readily found in the literature ¹¹⁻¹⁴. Rather, our emphasis is on emerging mitigation approaches for other key sectors, for which innovative options are also critically needed. As an integrated aspect of our review we also appraise the latest developments in emission estimates for the key sources.

Through presenting the underpinning concepts and practical considerations associated with emergent technologies we strive to stimulate opportunities for CH₄ mitigation on a cross-sectoral scale. Building on the effectiveness of existing conventional technologies and established management practises that have been demonstrated to abate CH₄ emissions, we envisage that the promotion of cross-sectoral emergent technologies will foster a more efficient approach for tackling global CH₄ emissions. Given the recent call for ‘aggressive CH₄

mitigation' measures issued by Kim, et al. ¹⁵, there has arguably never been a more pressing opportunity for appraising the state technology development in this field.

Contributions and trends for key emission sources

Current emission estimates for anthropogenic CH₄ sources are presented in Fig. 1, illustrating how CH₄ emissions compare with those of the other main GHGs (Fig. 1a). The magnitudes of emissions from the key anthropogenic CH₄ sources have previously been well-documented and are summarised in Fig. 1b, using a combination of literature values and IPCC estimates. It can be seen that total estimates vary between documented sources, resulting from uncertainties in emission estimates for each sector. The emission estimates in Fig.1b incorporate the IPCC's Second Assessment Report GWP of 21 for CH₄ which is currently adopted in most national inventories, although there is increasing momentum for adoption of the GWP of 28 proposed by the IPCC's AR5 metrics ¹⁶.

Two central concepts from Fig. 1b are pertinent to this review: 1) large-scale hydroelectric dams are a significant CH₄ source, despite not been accounted for as direct CH₄ emission sources by the IPCC or by most other inventory documents ¹⁷, and 2) the magnitudes of emissions are reasonably consistent across the key emission sources. From a mitigation perspective, the recognition of large-scale hydroelectric dams as a primary emission source is important because there are a number of inventive and practical approaches for abating these emissions, some of which might hold relevance to mitigating emissions from other sources.

Figure 1 – CH₄ emissions: a) in the context of the principal GHGs (*LUC = land use change); and b) as a function of the key CH₄ sources amounting to a total CH₄ emission estimate of 9.5 Gt CO₂-e yr⁻¹ in our study (compared with 7.8 Gt CO₂-e yr⁻¹ from the IPCC ¹, 8.5 Gt CO₂-e yr⁻¹ from Karakurt, et al. ⁸, and 6.2 Gt CO₂-e yr⁻¹ from Stolaroff, et al. ⁹); horizontal dashes in each category represent standard errors for mean emission estimates compiled from the following references (Enteric ^{18, 19, 12, 20, 21, 22, 23, 24, 25} – Hydroelectric dams ^{26, 27, 28, 29, 30, 31, 32, 33, 34, 35} – Oil and gas ^{9, 36, 8, 10, 24} – Rice farming ^{37, 10, 8, 38, 39, 40, 41, 42, 24} – Landfills ^{43, 10, 8, 44, 45, 24}, Permafrost thaw ^{9, 46, 47, 48} – Coal mines ^{9, 8, 10, 49, 50, 51, 24} Biomass burning – ^{9, 8, 10, 52, 53, 24} – Domestic and industrial wastewater ^{54, 10, 8, 55, 24} – Livestock manure ^{18, 8, 10, 24, 25}).

The fact that emission magnitudes are reasonably evenly distributed across the main sources is also telling, because it highlights the need for a cross-sectoral approach to achieve comprehensive CH₄ abatement. In Fig. 2 we project how the balance of direct CH₄ emissions from the major sectors will look in the coming decades. We emphasise that these projections are broad estimates based on current trends in the key CH₄-producing sectors and that several unavoidable caveats are associated with this type of exercise. This is particularly true for the energy sector where forecasting trends can be problematic. For example, Utgikar and Scott ⁵⁶

note that accurate estimates and forecasts of energy demand are hampered by barriers associated with technology adoption; socio-economic factors; and volatile economic policies. Moreover, as noted by Feng and Zhang⁵⁷ and Aydin, et al.⁵⁸ current trends in energy use may conceivably change drastically in coming years as some countries adopt widespread energy saving measures whereas other developing countries surge in energy demand to meet growing economies.

Figure 2 – trends in direct CH₄ emissions from key sectors: a) sector CH₄ emission estimates for 2020 and 2050, based on data in Fig. 1 and sector forecasts reported by: ⁶¹, ³⁴, and ⁶² for Energy; ⁶³, ⁶⁴, and ⁶⁵ for Agriculture; and ⁶⁶, ⁶⁷ for Waste Management; b) changes in emission profiles for the main sectors from 2020 to 2050.

The agricultural and waste sectors, by comparison, are projected to continue to grow rapidly in the coming decades, potentially even to the point of where they could exceed sustainable boundaries⁵⁹, ⁶⁰. Yet, considering all of the factors above, our 2050 projections indicate that the relative contributions of the direct anthropogenic CH₄ emission sectors will remain reasonably consistent into the latter part of this century, albeit with a slight increase in the proportion of agricultural emissions resulting from a surge in food demand and a corresponding proportional decrease in energy-driven emissions (Fig. 2).

Recognition of a strong diversity of key CH₄ emission sources well into the 21st Century is important if we are to limit global mean temperature increases to 2°C or less by 2100, a target agreed to at the Paris summit in 2015. Sixteen of the first twenty publications generated by a Google Scholar search employing the terms ‘methane mitigation’ pertain to enteric abatement. While large strides are being made in providing solutions for decreasing ruminant CH₄, clearly these emissions are just one component of the total anthropogenic CH₄ budget. There is a real need for giving equal focus to the pursuance of mitigation strategies for other key CH₄ sources.

In the following sections we present some of the emerging and novel technologies in the field of anthropogenic CH₄ abatement. Rather than present the technologies on a source-by-source basis we discuss the processes and mechanisms involved in the technology and then elaborate where we see potential for cross-sectoral application.

Breaking the bond – advances in catalysts for low concentration and low temperature methane oxidation

Breaking the carbon-hydrogen bond in the CH₄ molecule is the first obstacle faced by all CH₄ mitigation approaches that aren’t based on avoiding CH₄ production. Although this reaction is lucrative from an energy production perspective with a standard enthalpy of combustion of – 882 kJ/mol, a substantial amount of activation energy is required for its initiation. In CH₄-rich gas such as biogas produced at landfills and in wastewater treatment systems energy recovery from gas combustion is technically feasible and widely-practised⁶⁸⁻⁷⁰. However, in gas streams that are <5% CH₄ concentration, continuous thermal combustion of CH₄ is not achievable. One option is to use catalytic oxidation to break the C–H bond in low CH₄-concentration streams, employing materials with a high activity for oxygen scavenging, adsorption and subsequent CH₄ oxidation to methanol and, ultimately, through to carbon dioxide and water. This approach

is well-established, stemming from early endeavours to purify CH₄-gas streams into more desirable and transportable energy sources^{71, 72}. Yet, while the underlying principle behind catalytic CH₄ oxidation is decades-old, recent developments in materials science and engineering has opened-up promising new opportunities for CH₄ mitigation across a variety of key diffuse sources.

The application of catalytic oxidation to mitigate CH₄ emissions has been mostly applied to ventilation air methane (VAM) in coal mines; a methane-poor gas stream representing about 70% of coal emissions^{51, 73-75}. Karakurt, et al.⁷⁵, in their comprehensive review of approaches for mitigating coal mine CH₄ emissions highlighted several catalytic technologies, such as catalytic reverse flow reactors and catalytic monolith reactors, as critical tools in efforts to manage this emission source.

In terms of effective catalytic materials Setiawan, et al.⁷⁶ note that the noble metals – particularly palladium – are the most commonly adopted catalysts for low-temperature CH₄ oxidation including applications relating to mitigating VAM in coal mines⁵¹. The noble metals are particularly suited to catalysing CH₄ oxidation because they are stable over a wide range of conditions and are reasonably resistant to fouling by interfering compounds in the gas stream, such as sulphur gases⁷⁷. The downside, however, with using noble metals for this type of application is their scarcity and considerably high cost.

Recent advances in materials science may spark novel initiatives in the application and practicality of catalytic-based CH₄ oxidation technologies. Not only are new materials continuously being produced and tested but methods for analysing the surfaces of these materials and their catalytic chemistry are becoming increasingly sophisticated and powerful,

such as electron dispersive spectroscopy (EDS), x-ray absorption near edge spectroscopy (XANES) and x-ray photon electron spectroscopy (XPS) ⁷⁸⁻⁸¹.

On the back of these advances, a suite of alternative catalytic materials have been tested for their capacity to oxidise low concentration CH₄ streams such as VAM. Thiruvengkatachari, et al. ⁸², for example, reported an approximate 98% decrease in concentration of CH₄ emissions from VAM using a novel honeycomb carbon nanofiber composite material without the presence of noble metals. In addition to synthetic carbon materials, effective CH₄ oxidation has been documented using copper-rich synthetic zeolites at temperatures down to 125°C, which is remarkably low for catalytic CH₄ oxidation ^{83, 84}. The use of materials like carbon fibres and zeolites to enhance catalytic oxidation has the potential to provide some exciting prospects for practical CH₄ mitigation, perhaps in the complete absence of noble metals. Advances in materials engineering are resulting in the delivery of extremely robust and carbon compounds with high surface area ⁸⁵⁻⁸⁷ while new synthetic zeolites with enormous surface areas are continuously being fabricated for a variety of industrial purposes ^{84, 88}. Olivos-Suarez, et al. ⁸⁹ in their review of emerging CH₄ catalytic materials, note that metal organic frameworks (MOFs) also exhibit highly promising properties for enhanced catalytic oxidation. MOFs are 3-dimensional structures comprising metal ions coordinated with organic ligands and according to Olivos-Suarez, et al. ⁸⁹ MOFs more closely mimic the active sites of naturally-occurring CH₄-oxidising enzymes than any other class of engineered material. Consequently, there is strong anticipation that this group of materials will feature prominently in this field of application in the coming years^{89, 90}.

The combined properties of durability and high surface area exhibited by all of these classes of emerging materials discussed above present real practical opportunities for developing increasingly efficient materials to catalyse low concentration CH_4 oxidation in VAM.

Figure 3 – conceptual depiction of how catalytic structures could be deployed in animal housing and feedlot infrastructure to help abate ruminant CH_4 emissions: a) a wintering barn in the south island of New Zealand where dairy cattle reside for several months in the year; b) a beef cattle feedlot in the Darling Downs of Australia where rotating batches of up to 30,000 head are ‘finished’ year-round prior to going to market.

Indeed, with these improvements in materials science and engineering, the opportunity now exists to take catalytic CH_4 oxidation beyond the coal industry. The approach could be applied across a variety of low concentration CH_4 sources including fugitive emissions associated with natural gas infrastructure and landfills. Catalysts could also be adopted to help tackle enteric CH_4 : a source of considerable emissions for which there are few proven mitigation solutions. In a recent example, clays were shown to offer a possible way forward ⁹¹. The global rise in intensification of beef and dairy cattle farming presents opportunities for catalytic-induced mitigation of low concentration enteric emissions in structures including animal shelters, standoff pads, wintering barns and feedlots (Fig. 3). Encouragingly, as noted by Stolaroff, et al. ⁹, a key advantage for all of these applications whereby GHG abatement is the primary objective is that complete oxidation of CH_4 right through to CO_2 and H_2O is not a problem. This contrasts with endeavours for optimising fuel extraction from CH_4 -derivatives (e.g.,

methanol, formaldehyde) via catalytic oxidation where avoidance of complete combustion remains a critical logistical challenge.

Engineered biofilters – novel design approaches and applications

Methanotrophs are proteobacterial CH₄-oxidising bacteria. These organisms are diversely distributed throughout a range of soil, sediment and aquatic environments ⁹² and play a critical role in biogenic carbon cycling ⁹³. The fundamentally important characteristic to CH₄ regulation that all methanotrophs exhibit is their possession of the enzyme methane monooxygenase (MMO), which catalyses CH₄ oxidation to methanol, thus beginning a series of biochemical oxidation reactions culminating in CO₂ and H₂O production. The MMO enzyme exists in soluble and particulate forms, with the activity of the soluble form reportedly inhibited and the particulate form stimulated by free copper ions ⁹⁴. Broadly, the methanotrophs are classified as three types (1, 2 and X) with the primary distinction being the basis of formaldehyde assimilation – Type 1 use the ribulose monophosphate (RuMP) pathway, Type 2 use the serine pathway, and Type X primarily use the RuMP pathway but also possess serine pathway capability). Broadly, Type 1 methanotrophs favour CH₄-poor/O₂-rich environments whereas Type 2 methanotrophs dominate in CH₄-rich/O₂ poor settings ⁹². Ultimately though, as noted by Strong, et al. ⁹⁵, differences between the main methanotroph classes are often nuanced and no one particular group of organisms has been isolated demonstrating notably superior CH₄ oxidation capability to all others.

The feasibility of using soil-based biofilters containing active methanotroph populations has been demonstrated for mitigating substantial CH₄ emissions from various sources, including landfills, and stored agricultural effluent ⁹⁶⁻⁹⁹. Earlier applications have also been reported for coal mine emissions ^{100, 101}. A summary of CH₄ biofilter performance is given in Table 1.

Some basic requirements for efficient removal of CH₄ using soil-based biofilters include: good aeration status; water storage capacity; and adequate nutrients, particularly phosphorus^{92, 93, 98, 102}. Recently, advances in tailoring these requirements for optimised methanotroph activity has resulted in improvements in filter efficiency and upscaling potential. For example, materials other than soil, e.g., biochar¹⁰³, have been tested in filters for their capacity to enhance CH₄ oxidation efficiency. Brandt, et al.¹⁰⁴ demonstrated that using expanded vermiculite in biofilter packing media increased the active methanotroph biomass by a factor of 3, resulting in a corresponding 100% increase in CH₄ oxidation efficiency.

Table 1 – performance of engineered methane biofilters, compiled from literature reports.

Emission source targeted	Removal rate (g m ⁻³ h ⁻¹)	Filter media	Author	Notes
Landfill	80	Composite: clay, gravel, humic topsoil and vegetation	Gebert and Gröngröft ¹⁰⁵	Maximum removal rate reported for field chambers
General	65	Perlite	Kim, et al. ¹⁰⁶	Average removal rate reported for laboratory study
Stored piggery effluent	8	Compost	Melse and van der Werf ¹⁰⁷	Maximum for a pilot-scale filter
Landfill	28	Compost	Haubrichs and Widmann ¹⁰⁸	Average removal rate reported for laboratory study
General	24	Volcanic pumice soil + perlite	Pratt, et al. ¹⁰⁹	Maximum removal rate reported for laboratory study
Dairy effluent lagoon	102	Volcanic pumice soil	Syed, et al. ⁹⁹	Maximum removal rate reported for field study
General	10	Polyurethane beads	Gómez-Cuervo, et al. ¹¹⁰	Average removal rate reported for laboratory study
Ventilation coal mine air	27	Coal	Limbri, et al. ¹¹¹	Maximum removal rate reported for laboratory study
General	45	Stone inorganic media	Ferdowsi, et al. ¹¹²	Maximum removal rate reported for laboratory study
General	35	Expanded vermiculite	Brandt, et al. ¹⁰⁴	Maximum removal rate reported for laboratory study
General	37	Compost with fungi	Lebrero, et al. ¹¹³	Maximum removal rate reported for laboratory study
General	55	Compost	Farrokhzadeh, et al. ¹¹⁴	Maximum removal rate reported for laboratory study

In addition to promoting methanotroph growth and activity via physical processes, as alluded to by Brandt, et al.¹⁰⁴, the expanded vermiculite may have helped maintain sufficient nitrogen

concentrations for microbial activity via ammonium sorption. Previous work by Hill, et al.¹¹⁵ and Pratt, et al.¹¹⁶ has shown that vermiculite, which is a high cation-exchange capacity clay, can effectively bind soil and solution ammonium through exchange processes. It might be possible to overcome nutrient deficiency to methanotrophs through other sustainable approaches, such as incorporating nitrogen-fixing bacterial strains into the filter or by trickling nutrient-rich waste streams over the filter in applications relating to agriculture, landfills and wastewater treatment.

Recent studies have shown that fungi could potentially play an important role in CH₄ oxidation^{113, 117}. The CH₄ removal rates reported for a fungi-based compost biofilter by Lebrero, et al.¹¹³ are relatively high compared with the other high-performing systems documented in Table 1. Moreover, it is possible that fungi+methanotroph-bacteria mediated oxidation might enhance biofilter performance through stimulating nutrient exchange in the filter media.

The engineered design of the biofilter can also be tailored to the source, with potential cost savings. For example, open systems offer the advantage that they can deliver oxygen passively to the methanotrophs without the need for pumps. An example is a biofilter with a large surface area-to-volume ratio so air can passively diffuse into the biofilter from above while CH₄ can diffuse up into the biofilter from the source below; similar to process that over in landfill cover soils. This ‘floating’ design was tested by Pratt, et al.¹¹⁸ at the laboratory-scale with success, and subsequently confirmed at the field-scale by Syed, et al.⁹⁹.

There is considerable cross-sectoral potential to reduce emissions based on improved design of such biofilters coupled with a better understanding of the ecology of methanotrophs and methanogens. Recent New Zealand *in vitro* studies suggest the use of primed (with CH₄) soils

with high organic matter contents adjacent to the animal shelter may offer the potential to mitigate both enteric and effluent CH₄ emissions ¹⁰³. A similar approach was shown to have potential for oxidising CH₄ emissions from stored animal manure in the Netherlands ¹¹⁹. Such research according to Stolaroff, et al. ⁹ might also lead to a better understanding of Arctic sources and the means to mitigate them. However, the current increasing release of CH₄ from Arctic sources threaten to overwhelm the utility of current proven technologies such as biofilters and bioreactors, and urgently needs technological intervention.

Tailoring soil rhizosphere community dynamics for enhanced methanotrophy

Recent investigations have revealed important insights into the suite of microorganisms that inhabit the soil rhizosphere termed ‘plant growth-promoting rhizobacteria’ or PGPR. Soil PGPR exhibit beneficial functions relating to many growth considerations, including: stimulating plant access to soil nutrients ¹²⁰; protecting plants from pathogen infection ¹²¹; providing tolerance for toxic trace metals ¹²²; optimising root architecture ¹²³; and negating the impacts of salinity ¹²⁴.

Recently, Singh and Strong ⁶⁵ recognised the importance of interactions between PGPR and CH₄-regulating organisms in the soil rhizosphere. Indeed, these authors noted that some microorganisms that convey well-established benefits to plant-soil interactions also promote methanotrophy (Fig. 4). Enhanced CH₄ oxidation has been noted in the presence of plant N-providers such as cyanobacteria (*Azolla*) and diazotrophs (*Ochrobactrum anthropic*) ⁶⁵. Singh and Strong ⁶⁵ also suggested that incorporation of another diazotroph, *Azospirillum*, in the rhizosphere could stimulate biological CH₄ oxidation via increasing root mass, thereby promoting O₂ exchange to the soil.

Figure 4 – schematic illustration of the role of bio-fertilisers in improving soil CH₄ oxidation activity. Modified from Singh and Strong ⁶⁵.

The potential impact of PGPR associated with CH₄ oxidation function might entail widespread agronomic and GHG mitigation benefits beyond rice production systems, which formed the basis of the review by Singh and Strong ⁶⁵. Indeed, globally there is extensive agricultural use of landscapes overlying organic-rich peaty soils ¹²⁵⁻¹²⁸. These landscapes are an interesting category with regards to IPCC reporting. Although their net emissions are considered neutral, there are certainly cases where agriculture has demonstrably resulted in a net increase in emissions from the underlying organic soils ¹²⁶. In these settings there is significant potential for GHG mitigation using microbes to stimulate CH₄ oxidation. At an even broader level, there is scope to introduce methanotroph-stimulating PGPR into engineered biofilters, which have been shown to have extremely diverse application potential across multiple sectors.

Whilst a promising concept, the use of organic inputs to stimulate methanotrophy in agricultural soils and/or engineered systems has potential obstacles that need to be addressed. Long-term effects on CH₄ oxidation efficiency is unknown and needs testing. Moreover, addition of organic-amendments to soil as carriers of the targeted microorganisms entails the

obvious problem of introducing more carbon into the soil which could be converted to CH₄. This limitation could be overcome by extracting isolates of the desired microorganisms and introducing them into the soil as blends with conventional or low-labile carbon fertilisers. Successes in similar areas have already been made in the development of products referred to as 'biofertilisers' which have been demonstrated to convey agronomic benefits to soils ^{129, 130}. These products, which are carefully constructed under controlled conditions, might hold the key to developing an effective CH₄ mitigation tool with applications cutting across several key emission sources including: rice production; agriculture in peaty soils; as well as in biofilters designed for landfills, wastewater treatment systems, coal mines and leaks surrounding natural gas infrastructure.

Transgenics: a viable way forward for rice methane abatement?

Rice is the staple for more than half of the global population with rapid growth in consumption occurring in Africa and Latin America ¹³¹. These trends could conceivably see rice production become the principal source of anthropogenic emissions in the coming years, surpassing its current upper annual emission estimate of 100 Mt by Su, et al. ¹³². While substantial reductions in CH₄ emissions have been achieved by mid-season drainage ¹³³, there is the potential that management approaches alone can increase net GHG emissions via stimulating N₂O production ¹³⁴. Hence, additional ways are needed to mitigate CH₄ emissions from paddy rice to tackle this burgeoning GHG source.

In addition to crop management methods that have been reported to help suppress rice CH₄ emissions ¹³⁵⁻¹³⁷, a new study by Su, et al. ¹³² demonstrated CH₄ reduction and crop yield benefits in rice crops via transgenic manipulation. Using a barley transcription gene, these authors observed significant increases in grain starch and a corresponding decrease in CH₄

emissions coupled with a diminished soil methanogen community. Lower root mass and root exudates in the soil rhizosphere were proposed as the mechanism responsible for the suppressed CH₄ emissions^{132, 138}.

The findings of this novel transgenics study are very encouraging, particularly given that, as Su, et al.¹³² note, farming-based abatement approaches are difficult to implement without adversely impacting crop production. However, it is important to stress that this transgenics approach is in its infancy. Many questions remain relating to its widespread and long-term effectiveness in achieving CH₄ mitigation as well as grain yield. Moreover, the CH₄ suppression mechanism proposed by Su, et al.¹³² needs to be conclusively verified in order to assess the impacts of the technology on long-term soil function and productivity. Bodelier¹³⁸, in their News and Views article in Nature, have already questioned the impact that lower root-derived carbon inputs might have on long-term soil community structure, particularly with regard to the PGPR bacterial and rhizo-fungal consortia. The functions and diversity of the PGPR are becoming better understood, with plant benefits relating to synthesis of key nutrients for root uptake as well as pathogen control in the rhizosphere¹³⁹⁻¹⁴¹. Moreover, we saw in the previous section the importance of maintaining a healthy soil PGPR community for boosting soil methanotroph activity. The effects of decreasing organic carbon supply to the soil rhizosphere on these delicate ecological dynamics needs to be examined. In short, the role of transgenics as a powerful CH₄ mitigation tool holds promise but certainly a suite of impact investigations are needed before this approach can be framed as a viable abatement technique.

Outcompeting methanogens – feasibility of microbial fuel cells to mitigate emissions

Microbial fuel cells (MFCs) can generate electric current by catalysing the oxidation of organic and inorganic matter to liberate electrons at an anode with their subsequent consumption at a

cathode. When both these processes are sustainable the result is a working MFC. This is a rapidly developing field of research, and MFCs have already been shown to produce electricity from a wide variety of substrates, including simple molecules like glucose ¹⁴² and acetate ¹⁴³, as well as more complex materials such as natural wastewaters ¹⁴⁴. A thorough review of the methodology and technology aspects of MFC was published by Logan, et al. ¹⁴⁵.

Recently, MFC feasibility has been tested in two main sectors responsible for CH₄ emissions: rice paddy fields and wastewater treatment systems. The technology has shown promise in both areas of application ¹⁴⁶⁻¹⁵⁰: with regards to wastewater treatment applications, the approach has the added benefit of organic removal, with up to 80% removal efficiency reported by Liu, et al. ¹⁴⁹. However, as noted by Pham, et al. ¹⁵¹ MFC technology still has limitations relating to up-scaling, costs and electrode fouling.

In MFCs applied to CH₄ emission sources, electricity production is achieved in tandem with decreased CH₄ emission as the bacteria responsible for current generation are able to outcompete methanogens for substrate. Wrighton, et al. ¹⁵² found that the bacterial phylum Firmicutes was instrumental in achieving high current output in MFCs. Interestingly, this phylum has been reported to deliver plant growth-promoting effects including pathogen resistance ¹⁵³ as well as salinity and drought tolerance ^{154, 155}. Thus, members of Firmicutes might be beneficial in delivering plant growth benefits as well as optimised electricity generation where MFC technology is employed in agricultural settings.

The above approaches relate to cases where MFCs are able to decrease emissions through suppressing methanogen activity. There is also the potential for MFC technology to mitigate emissions by harnessing CH₄ that has already been produced. Methane has not, until very

recently, been used with much success as a substrate in MFC to generate electrical current. Now McAnulty, et al. ¹⁵⁶ have successfully constructed a consortium that combines the metabolically engineered archaeal strain *Methanosarcina acetivorans* that produces methyl-coenzyme M reductase and grows on CH₄ with *Geobacter sulfurreducens* and CH₄-consuming sludge to generate a substantial electrical current from CH₄. This work represents a major breakthrough in MFC biotechnology and has the potential in future to produce substantial amounts of electricity while reducing CH₄ emissions from a wide range of sources beyond rice paddy fields and wastewater treatment plants, including landfills, coal mines and possibly even emission hotspots in the melting permafrost (Fig. 5).

Figure 5 – a) concept of established MFC technologies where microbial colonisation and electron deposition is forced at the anode from where electrons transfer across a resistor to a cathode, generating electrical current and water, b) how the findings of McAnulty, et al. ¹⁵⁶, whereby specialised microbes convert CH₄ to acetate, could open up MFC technology to abate CH₄ from a diverse range of emission sources.

Getting the carbon out of methane

Mitigation of CH₄ emissions can also lead to the production of useful environmentally-friendly products. An example is the production of PHA (polyhydroxyalkanoate) in soil microorganisms exposed to a high CH₄ source in a low nutrient environment (the white spot in the methanotroph cell shown in Fig. 6a, Pratt et al., 2013). Polyhydroxyalkanoate is a

biodegradable polymer with similar properties to polyethylene and it can be used as feedstock for the manufacture of bioplastic products (e.g., children's toys, food packaging). The result is the removal of a potent greenhouse gas – CH₄ – from the atmosphere while at the same time replacing fossil fuel-based plastic with a biodegradable substitute that will also reduce pollution of the oceans. A number of methanotrophs have been assessed for PHA^{157, 158}. This biotechnological approach to reducing fossil fuel use also has the potential to produce plastics and other products with a wide range of beneficial properties and uses⁹⁵. These include other biodegradable polymers such as PHBs (polyhydroxybutyrate) and polylactic acid (PLAa from lactic acid) that can also be produced by methanotrophs from biogas (mostly composed of CH₄ and CO₂), but under carefully controlled conditions. For example, PHBs can also be produced from CH₄ by methanotrophs¹⁵⁹. They are apparently only produced by Type II methanotrophs¹⁶⁰ under conditions where CH₄ is delivered intermittently at low levels and at pH levels of 4 to 5. Commercial application is currently limited by the high cost of production⁹⁵. These examples illustrate the earlier point that a better understanding of the microbial processes involved is needed if the production of bioplastics and other products from these precursors is to be optimised and made economical. In their excellent review of the range of other potential opportunities offered by methanotrophs, Strong, et al.⁹⁵ list single cell protein, components for nanotechnology applications, soluble metabolites (e.g., methanol, formaldehyde), lipids (biodiesel and health supplements) growth media and vitamin B12, using CH₄ as their carbon source.

In addition to harnessing carbon deposits in methanotrophs for commodity or fuel production, there is potential to align methanotroph carbon-storage with improving stable carbon stocks in soils. Effective retention and cycling of carbon in soils is accepted as one of the principal climate change mitigation approaches, reportedly offering the potential to offset up to 15% of

global anthropogenic emissions ¹⁶¹. Deposits of fatty biopolymers that accumulate in methanotroph cells (Fig. 6a) could conceivably lead to increased soil carbon stocks as organisms die. Fig. 6b highlights just how much soil carbon contents can increase after sustained high methanotroph activity, depicting carbon concentrations in soil biofilters treating dairy effluent CH₄ emissions. After just 1.5 years, the carbon content of the biofilters nearly doubled.

A short-term increase in soil carbon levels does not necessarily amount to effective carbon soil sequestration. However, key soil components have been demonstrated to enhance carbon stability from inorganic soil inputs. For example, Bolan, et al. ¹⁶² reported that clays have the capacity to promote carbon protection in soils through developing stable metal organo-complexes. Clemmensen, et al. ¹⁶³ noted that fungi play a critical role in regulating soil carbon properties. Further understanding of how these soil constituents interact with carbon turnover could underpin viable technologies for enhanced carbon sequestration achieved through methanotrophy (Fig. 6c).

Figure 6 – biological carbon ‘mining’: a) deposits of carbon-rich polymers in different methanotroph strains; b) depicting how these carbon deposits manifest in markedly increased soil carbon concentrations in 1) a laboratory ¹¹⁸ and 2) field pumice-soil ⁹⁸ biofilter treating dairy effluent pond CH₄ emissions; and c) potential applications for methanotroph-harvested carbon in the fields of i) bioplastics development and ii) long-term soil carbon sequestration through interacting with soil components.

Hydroelectric dams: an overlooked methane source with significant mitigation potential

Hydroelectric power generation has long been regarded as an environmentally-clean energy source, but evidence has been mounting that substantial amounts of CH₄ are produced from rotting vegetation in lake sediments trapped by dams ³². In fact, tropical hydroelectric dams are possibly the single largest anthropogenic CH₄ source, yet remarkably these emissions are either undercounted or completely overlooked in IPCC and inventory reports ¹⁷. The IPCC cannot currently ascertain whether hydroelectric dams are net GHG sources or sinks which Fearnside ¹⁷ attributes to various factors including overrepresentation of emissions from temperate dams which typically produce negligible CH₄ and an underestimation of turbine emissions which can be very large. Recent discussion has indicated that reservoir emissions may be included as an anthropogenic category in IPCC reporting in 2019 ¹⁶⁴.

Despite uncertainty over the net climate change impact of these dams they are clearly substantial CH₄ emitters in certain locations, as recognised by a number of recent reports ^{26, 165}. Concern is rising over expanding emissions in countries with new large-scale hydroelectric dams, such as China ¹⁶⁶.

Specific CH₄ emission points in hydroelectric dams include surfaces of reservoirs and secondary channels, spillways and turbines ¹⁶⁷. An important first step proposed by Deemer, et al. ³² for identifying possible management opportunities for reducing reservoir greenhouse gas emissions is to establish the relationship between reservoir eutrophication and the associated emissions of greenhouse gases. Simple mitigation and recovery procedures have been proposed for tropical hydroelectric dams by Bambace, et al. ¹⁶⁷ involving engineered systems to capture CH₄-depleted waters, thus eliminating potentially severe CH₄ emissions from turbine locations.

This engineering strategy to minimise dam emissions was also advocated by Beaulieu, et al.³⁵. Energy recovery from CH₄-saturated water deep in the reservoir could offer further abatement. These relatively simple yet effective technologies could be applied widely across the industry. These mitigation options were predicated on the fact that the amount of CH₄ emitted at the reservoir surface represents only a fraction of the total CH₄ produced in the reservoir. The balance of this CH₄ is either emitted by degassing as the CH₄ enriched water passes through the turbines and into spillways, or is oxidised. Bambace, et al.¹⁶⁷ argue that by recovering this CH₄ for subsequent energy production, it may be possible to reduce the need to build new reservoirs, thereby protecting pristine biomes such as the Amazon rainforest. In other environments, strategies can be adopted to prevent eutrophication and reduce greenhouse gas reservoirs by specifically reducing movement of phosphorus and nitrate from entering the reservoirs³².

At a cross-sectoral level, new understandings in the microbial assemblages in dam reservoir profiles might yield important information for biofilter design and application across other industries. Lima¹⁶⁸ discovered the importance of ‘deep’ zones in hydroelectric reservoir profiles for CH₄ oxidation – reporting the highest rate of activity at this layer down to approximately 20m below the reservoir surface. The methanotroph community composition in reservoir profiles has been shown to be diverse, comprising assemblages of extremely efficient oxidisers¹⁶⁹. In fact, Dumestre, et al.¹⁷⁰ reported the occurrence of a methanotroph community in a damned reservoir in South America that specialised in oxidation at the oxic-anoxic interface of the water profile which was able to oxidise the entire portion of surface-emitted CH₄. Isolating these efficient oxidisers and associated symbiotic microbiota might offer a viable way forward to optimise biofilter design and application in other sectors including landfills, natural gas production, coal mines and wastewater treatment.

Technology maturity and opportunities for adoption

The technologies discussed in this paper cover a maturity spectrum ranging from blue-sky concepts through to commercially-practised activities. In this final section, we attempt to present the various development states of the technologies assessed, highlighting links to commercial applications and other investment initiatives where available.

Immature technologies

Transgenic technologies for producing rice strains with inherently low CH₄-emitting traits is a promising approach but is clearly in its infancy with validation of long-term benefits needed. This research is very new so it will likely take several years to gauge whether this technology will have potential traction as an effective CH₄ abatement strategy. Similarly, the use of waste CH₄ emissions to feed microbial fuel cells, while an exciting concept, really needs development and verification to frame it as a viable mitigation technology.

The potential for soil-based methanotrophs to achieve GHG mitigation benefits through carbon sequestration (additional to CH₄ oxidation) is very real. Research into the compounds formed in methanotroph fatty tissue deposits may well reveal quantifiable benefits for settings such as landfill cover soils, rice paddies and agricultural practises in peatlands. Testing if amendments such as clays, iron oxides and fungi do in fact stimulate soil carbon protection may lay a pathway to even greater soil carbon storage efficiency. However, research into these processes is critically lacking.

For hydroelectric reservoirs, engineering strategies to collect CH₄-depleted waters for passing through turbines is potentially very achievable but, as noted by Beaulieu, et al. ³⁵, these types

of approaches remain unimplemented – largely due to a lack of drivers for GHG abatement for this source.

The concept of using CH₄-primed soils surrounding animal housing to abate enteric emissions may potentially underpin a highly novel and practical technology. Although targeting only the proportion of emissions that occur when animals are housed, a technology founded on this approach could make substantial inroads into a key emission source. This work is very preliminary with the New Zealand Agricultural Greenhouse Gas Research Centre recently investing in a field-scale feasibility study on the approach (<http://www.nzagrc.org.nz/capture.html>). If successful, the technology could be applied to other diffuse ‘leaky’ CH₄ sources such as coal mine gas extraction and natural gas infrastructure.

Technologies of intermediate maturity

Floating CH₄ biofilters are an attractive and potentially low-maintenance technology for a varied array of CH₄ emission sources, including manure storage lagoons; wastewater treatment plants; and small-scale reservoirs. This technology has been successfully demonstrated in the field ^{98, 99}, but scaled-up, longer-term testing is needed. The technology is particularly appealing as a low-cost alternative for small-scale CH₄ emitting ponds or lagoons which represent substantial additive emissions yet do not offer feasible energy at the individual pond scale. Even gas capture and flaring as a GHG abatement option alone entails significant maintenance and compliance investments – we are aware of a commercial piggery in eastern Australia that in 2017 decommissioned its biogas collection system due to prohibitive maintenance and compliance costs. Floating biofilters present an attractive alternative for these cases. Several companies are manufacturing floating biofilter wetlands globally, mainly for attenuation of nutrient and odorous compounds (<http://www.aquabiofilter.com/>,

<http://waterclean.co.nz/floating-wetlands/>). There is a real opportunity to incorporate CH₄ oxidation capacity into the engineered commercially-available systems.

The development of biofertiliser products containing specific bacterial isolates has progressed significantly in the past few years. Indeed, a suite of biofertiliser products containing plant growth promoting bacteria are available through government initiatives as well as private enterprises (<http://www.sosbio.com.au/wp-content/uploads/2017/05/SOSBio-Products-SMALL.compressed-1.pdf>, http://www.nationalfertilizers.com/index.php?option=com_content&view=article&id=140&Itemid=156&lang=en). The opportunity now exists to establish if these products can achieve enhanced methanotrophy, in addition to the other plant growth benefits they deliver.

Full-scale and commercially practised technologies

While ideas for implementing catalytic-based CH₄ approaches for sources such as animal housing are speculative, the use of catalytic technology for abating ventilation air methane (VAM) has been successfully tested at full-scale. For example, the VAMCAT device (a joint initiative between Australia's Commonwealth Scientific and Industrial Organisation/CSIRO and the Huainan Coal Mining Group) is based on a novel catalytic design and successfully oxidised VAM from an operational coal mine in China with an inlet CH₄ concentration of 0.8% (<https://www.csiro.au/en/Research/EF/Areas/Coal-mining/Mining-safety-and-automation/VAMCAT>). Other systems purporting novel catalytic principles are commercially-available (<http://www.aerison.com/environmental/greenhouse-gas-reduction/>). In combination with the advances in materials science and surface analysis methods discussed earlier, as well as novel approaches for gas flow configurations to achieve optimal CH₄ oxidation ¹⁷¹, it is likely that VAM catalytic technologies will become more commonplace in coming years.

Another novel CH₄ mitigation technology that has seen recent commercial success is the development of bioplastics from waste CH₄ streams. Mango MaterialsTM (<http://mangomaterials.com/>), established in 2010, is a US-based company that converts landfill-generated CH₄ into biopolymers for applications in clothing and packaging industries. The process undertaken by the company entails the combined GHG benefits of CH₄ mitigation as well as offsetting petroleum-based C sources in plastic manufacture. Advances in molecular understanding of methanotrophs will undoubtedly improve the efficiency of initiatives such as this, and open the opportunity for similar enterprises to start-up and prosper in the market place.

Outlook

Methane emissions are reasonably evenly distributed across the key anthropogenic production sources and will likely continue this trend through to at least 2050, highlighting that mitigation of this GHG can only be effectively achieved through multi-sectoral efforts. Novel technologies to mitigate CH₄ emissions across all the primary emission sectors are being rapidly developed and tested and many of these approaches have clear cross-sector potential. Advances in molecular techniques, materials sciences and process engineering are all clearly contributing to opening the feasibility of new ideas and improving efficiency of established concepts in this area. Some of these technologies are extremely preliminary while others are currently practised at commercial-scale. We now have at our disposal a wide array of technology approaches to combat CH₄ emissions which offer the very real prospect of associated benefits in the areas of agriculture, waste management and energy production.

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