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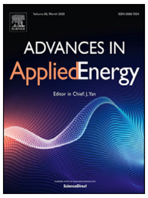

Perspectives on removal of atmospheric methane

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ABSTRACT

Methane's contribution to radiative forcing is second only to that of CO_2. Though previously neglected, methane is now gaining increasing public attention as a GHG. At the recent COP26 in Glasgow, 105 countries signed "the methane pledge" committing to a 30% reduction in emissions from oil and gas by 2030 compared to 2020 levels. Removal methods are complementary to such reduction, as they can deal with other sources of anthropogenic emissions as well as legacy emissions already accumulated in the troposphere. They can also provide future insurance in case biogenic emissions start rising significantly. This article reviews proposed methods for atmospheric methane removal at a climatically significant scale. These methods include enhancement of natural hydroxyl and chlorine sinks, photocatalysis in solar updraft towers, zeolite catalyst in direct air capture devices, and methanotrophic bacteria. Though these are still at an early stage of development, a comparison is provided with some carbon dioxide removal methods in terms of expected costs. The cheapest method is potentially enhancement of the chlorine natural sink, costing as little as $1.6 per ton CO_2-eq, but this should be carried out over remote areas to avoid endangering human health. Complementarity with methane emissions reduction is also discussed.

1. Introduction

1.1. A spotlight on methane versus carbon dioxide

While the atmospheric stock of carbon dioxide (CO_2) in the atmosphere has increased by about 50% since preindustrial time (417 vs 278ppm), that of methane (CH_4) has more than doubled (1879 vs 722 ppb) [1]. Although the importance of CH_4 as a greenhouse gas has been known about for many years, as reflected by the Kyoto protocol of 1997 [2], until recently public attention focused mainly on CO_2. Most mitigation and remediation proposals targeted CO_2.

Recently, however, more attention is being given to CH_4. Thus, in November 2021, at the UN Climate Change Conference (COP 26) held in Glasgow, 105 participating countries signed "*The Global Methane Pledge*" committing to a 30% reduction in emissions from oil and gas by 2030 relative to 2020 [3]. Moreover, the new contribution of Working Group 1 to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC WR1 AR6) released on August 2021 [4] highlights the need to quickly reduce global CH_4 emissions to slow warming [5,6] and "buy us time" [7].

Previously, on July 2021, a U.S.-Russia Joint Statement Addressing the Climate Challenge expressed their intent to work together bilaterally to address climate change, including emissions reductions from non-CO_2 greenhouse gasses (GHGs), including CH_4 [8]. Methane featured in the agenda of the recent U.S. Leaders' Climate Summit. Meanwhile, the Chinese 14th Five-Year Plan presented in March 2021 was expanded to include CH_4 and other non-CO_2 gasses [9], and China's biggest gas and oil producer is targeting a 50% reduction in CH_4 emission intensity by 2025 [10]. The United Nations Environment Program (UNEP) and the Climate and Clean Air Coalition in their "Global Methane Assessment" [11] as well as the International Energy Agency [12] are calling for urgent action to cut CH_4 emissions, and scientists and non-governmental organisations (NGOs) are calling for atmospheric CH_4 removal [13,14].

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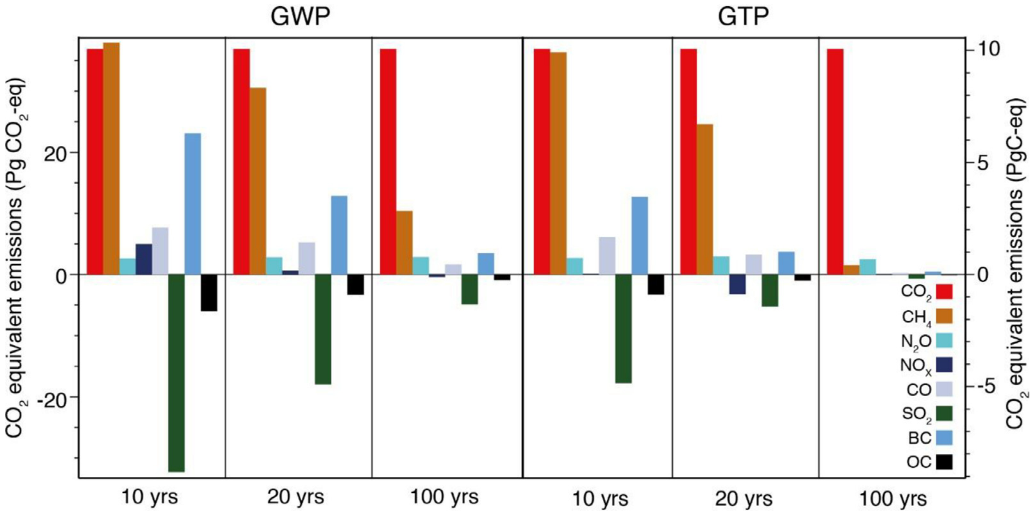

Fig. 1. Equivalent emissions of the principal GHGs, on a GWP and GTP basis, compared over a time horizon of 10, 20, and 100 years, from the IPCC [18].

These calls and decisions are timely. In the absence of further climate action, by the end of the century, global-mean warming due only to CH_4 emissions could contribute to about 0.9°C (±0.2°C), compared to a warming of about 0.5 °C (±0.1°C) currently, due to historical CH_4 emissions [15], and compared to the Paris agreement target of less than 2°C global warming including all GHGs.

1.2. *The atmospheric concentration of CH_4 is rising*

In 2020, despite Covid-19 shutdowns, the annual increase in atmospheric CH_4 was the largest recorded since 1983, when systematic measurements began [16]. Since the preindustrial era, tropospheric concentrations of CO_2 and CH_4 have increased by 47% and 167%, respectively [1]. Since 2007, tropospheric CH_4 has been rising rapidly, with an average annual growth rate of 9.3 parts per billion (ppb) (approximately 0.4% $year^{-1}$) between 2014 and 2019 [1]. Over a longer timescale, CH_4 concentrations have multiplied 3.28 times from a minimum of 570 ppb, reached 5000 years ago [17]. The IPCC [18] predicts that, over the next 10 to 20 years, CH_4 and CO_2 will have similar global warming impacts, as measured by heat absorbed (global warming potential, GWP) and temperature rise (Global Temperature change Potential, GTP). See Fig. 1.

Besides slowing global warming, CH_4 removal also can help protect the ozone layer, because one of the indirect effects of the rising concentrations of CH_4 is the increasing amounts of water in the stratosphere, which participates in ozone layer depletion [19].

Whereas the natural capacity of the atmosphere to remove GHGs remains roughly constant [20,21], CH_4 emissions from the major natural and anthropogenic sources are increasing [16]. The biogenic sources include tropical wetlands [22], lakes [23], ponds, hydroelectric reservoirs [24], and rivers [25]; and human-made sources include fossil fuels (coal mines, oil and gas wells); agriculture (livestock and rice cultivation); landfills; and some of the biomass burning due to intentional wildfires [26].

CH_4 emissions from the fossil fuel industry are uncertain but recently shown to be approximately 40% higher than previously estimated [27]. They include venting, flaring, and fugitive emissions of global diesel and gasoline [28], as well as leaks in gas distribution and use [29].

Due to warming oceans and surface air temperatures, several scientists perceive a risk of massive release of CH_4 by destabilization of seafloor methane-hydrates [30–32] and Arctic permafrost thaw [33,34]. Some observed CH_4 fluxes from 5 to 24 grams per square meter per day ($gm^{-2}day^{-1}$) were observed in 2013 [35]. However, some consider such risks to be lower, mainly thanks to microbial CH_4 consumption by methanotrophs [36–39] whose populations rise with the increasing abundance of CH_4. Meanwhile, increasing CH_4 emissions due to warming of wetlands and landfills, eutrophication of lakes [24], and fossil fuel extraction [40] are rising.

Most anthropogenic CH_4 emissions come from agriculture and waste management, which together constitute 60% of anthropogenic and 38% of total emissions. CH_4 emissions from oil and gas industries represent about 33% of anthropogenic emissions and about 17–19% of total CH_4 emissions, with extraction, processing and distribution accounting for about 2/3 and coal mining for 1/3 [26]. According to the UNEP, by fixing leaks and reduce venting and flaring, many CH_4 emissions reductions in the oil and gas industry can result in negative costs, as capturing CH_4 adds revenue [11].

The rapid rise of atmospheric CH_4 concentration requires action to try to limit global warming well below 2°C as targeted by the COP21 Paris Agreement, bearing in mind that the GWP of CH_4 is nearly 28 times higher than that of CO_2 on a 100-year basis, and 84 times higher on a 20-year basis [18]. Moreover, because CH_4 depletes the atmospheric reservoir of hydroxyl radicals responsible for removing CH_4 itself, large additions of CH_4 to the atmosphere extends the lifetime and GWP of the CH_4 already in the atmosphere [41,42].

In the next section the importance of focusing in CH_4 reduction is explained. The following sections review the infrastructure needed for large-scale CH_4 removal with the methods and strategies proposed so far, summarizing the advantages and disadvantages of those methods, and their potential costs compared to some CO_2 removal technologies. Then some expected co-benefits are discussed, before the concluding remarks.

2. Actions are needed against methane emissions and its atmospheric stock

Methods have been proposed to limit and reduce CH_4 emissions from several anthropogenic sources [43–45]: examples include better management of landfill (separate biodegradable waste) and coal, oil and gas fields (reduce leakage, recovering instead of flaring, capturing instead of venting, etc.) [11,15], as well as dietary changes to reduce emissions from livestock populations [46]. But few of these can be adapted to natural emissions, which are diffuse over large areas of thousands to millions of square kilometers and/or partly inaccessible. Unfortunately, most CH_4 emissions come from such diffuse natural biogenic sources [44], which by top-down estimations represent about 40% of total emissions [26]. Biogenic emissions come mainly from wetlands (31%) and a smaller part (7%) from freshwater systems, oceans, estuaries, permafrost, termites, wild animals and vegetation. Other estimates of the natural CH_4 emissions are of about 50% [47], as well as 50% by bottom-up estimates [26].

Removal or remediation methods seem even more technologically challenging, because they need to deplete CH_4 already released to the atmosphere, where it has been diluted about 0.5 million times in an air volume of some 1.4 billion cubic kilometers. In addition, the relatively short life cycle of atmospheric CH_4 (about 10 years) means that removal would have to be ongoing to reduce the concentration by a target amount. This contrasts with CO_2, for which a one-off removal achieves an almost permanent reduction. Further, CH_4 molecules are about 200 times scarcer in the atmosphere than CO_2 molecules. Unlike CO_2, however, CH_4 and other GHGs can be removed by in-situ oxidation, to products with no GWP or with much lower GWP than CH_4; for instance, CO_2 is obtained from CH_4 and nitrogen and oxygen from nitrous oxide (N_2O) without the need of capture, separation, or storage. The oxidation reaction is exothermic and exergonic, and as such requires no minimum energy input once the activation energy is overcome. This differs from CO_2, which requires at least 18 kJ mol^{-1} for its separation from the atmosphere [48], and more energy still if CO_2 is to be reduced to carbon or organic compounds such as industrial polymers [49]. Removal of CH_4 may be considered an acceleration of the natural oxidation processes, as once in the atmosphere it finally ends-up as CO_2.

3. Infrastructure needed for treating large volumes of air

As CH_4 is a well-mixed GHG, and its life expectancy in the troposphere is approximately 10 years, in order to have an impact on global warming, some authors have proposed that at least one-tenth of the atmosphere has to be processed every year, to compete with natural sinks [43]. In reality, any process of a portfolio of technologies allowing the atmospheric CH_4 concentration to start decreasing (while the imbalance between sources and sinks is currently increasing) will reduce its direct global warming impacts, as well as its indirect impacts (due to tropospheric ozone generation) on heath, food production, and primary productivity [50,51].

Boucher et al. first proposed direct atmospheric CH_4 removal in 2010 [52], but found that available technologies (zeolite minerals, adsorption filters, molecular sieves, and cryogenic separation) did not appear to be energetically or economically suiTable for large scale CH_4 capture from air. Consequently, instead of CH_4 capture, they proposed to directly oxidize it in-situ by a variety of possible methods, including bio-inspired aqueous-phase catalytic oxidation, bio-reactors containing methanogens, enzymatic systems, and catalysts made of precious metals. But these authors did not provide any specific details about how to process the very large volume of air in the atmosphere, concluding that these ideas were speculative.

Lockley [53] proposed several additional mitigation or removal techniques, such as: ignition of CH_4 at point sources, lake sealing with impermeable covers or with non-biodegradable foaming agents, ducting CH_4 bubble streams from underwater sources, and others. Again, in the absence of specific details, these ideas seem speculative.

Later, researchers proposed adding to the atmosphere CH_4 depleting agents such as chlorine (Cl) atoms generated by iron salt aerosols in the air [43,54] or using human made infrastructure devoted to another use. These could include:

- solar updraft chimneys (SUT) [55], which produce CO_2-free renewable electricity and each unit can process on the order of 6000 cubic kilometers (km^3) of air per year;
- direct air capture (DAC) systems [56] that are being developed to capture CO_2 from air [57,58].

Existing infrastructure could be used for CH_4 removal. For example, titanium dioxide in self-cleaning windows or other photocatalytic coatings such as paintings on buildings could contribute to CH_4 removal and attract carbon credits and certificates, thus helping to finance future removal infrastructure. Aircrafts, wind turbines, or other structures already in contact with large quantities of air might also provide reaction surfaces to oxidize CH_4.

4. Principal methods for enhancing atmospheric methane removal

4.1. Enhancing the main natural CH_4 sinks

Currently, in the troposphere, the principal natural CH_4 sinks are the hydroxyl radicals (which remove nearly 90% of the CH_4) [59], chlorine atoms (which remove about 2.5% of the CH_4) [60], minerals in soils and dust [61], soil microbes, plants and trees. Enhancing those natural sinks can be a strategy to increase atmospheric CH_4 removal.

4.2. Enhancing the hydroxyl radical $^{\circ}OH$ and targeting in majority point sources

Atmospheric natural self-cleansing and volatile organic compound (VOC) removal is mainly due to hydroxyl radicals [62]. Hydroxyl radical generators, as well as ozone generators, are commercially available, and indoor VOC pollution can be efficiently controlled by short-wave ultraviolet (UVC) light in closed systems [63]. Some coauthors of this article are working on methods to enhance the $^{\circ}OH$ sink of CH_4 and reduce its lifetime, although still more research is needed to be able to provide cost estimates and quantitative estimates of efficacy [64]. Still, the increasing efficiency and lifetime and the decreasing costs of ultra-violet light-emitting diodes (UV-LEDs) are promising. Research conducted at the University of Copenhagen has led to the creation of start-ups such as Infuser and AirLabs, which already apply this technology to point sources [65]. Several other possible strategies to generate $^{\circ}OH$ radicals and apply them in the open atmosphere might be possible, based on the numerous and complex mechanisms by which they are produced [66]. Knowing the intensity of sunlight UV, the $^{\circ}OH$ radical concentration can be predicted [67]. But care has to be taken not to expose human beings, animals, and plants to dangerous UV radiation and to ozone.

4.3. Enhancing the natural chlorine sink of CH_4–At the molecular level

In 2017, some of the coauthors of this article proposed to deplete the atmospheric CH_4 directly in the lower troposphere with Cl atoms [54,57], mainly but not exclusively under the marine boundary layer. Recent scientific research from 2015–2017 proved that Cl atoms can be generated in large amounts from the sodium chloride (NaCl) content of natural sea-spray aerosols, thanks to an iron(III)/iron(II) sunlight photo-catalyzed reaction [68,69]. Acidity (pH<3) found over coastal areas is naturally generated from NaCl by acid displacement with biogenic sulfate and nitrate [70]. However, it also can be enhanced by anthropogenic pollution due to combustion sources, where nitrogen oxides and sulfur oxides are further oxidized in the atmosphere into nitric and sulfuric acids that react with sea salt to generate hydrochloric acid and sodium nitrate and sodium sulfate salts. Fig. 2 illustrates a possible way by which CH_4 is already being removed by enhancing its chlorine sink.

Over polluted coastal areas, the chlorine sink destroys up to 11% of CH_4 [71]. It is worth noting that authors in favor of this method do not propose enhancing acid air pollution, and do not target the enhancement of the Cl atom generation over populated areas or coasts [54,72]. As CH_4 is a well-mixed GHG, it can be removed anywhere at atmospheric concentrations (about 1.9 ppm), consequently they propose to do so in remote unpopulated areas. They also plan to address point sources of CH_4, where it is more concentrated, to deplete it before it becomes diluted and mixed in the global atmosphere, and in this case the Cl atom generation will be carried out in closed systems such as the existing ventilation systems of coal mines, with very low risks.

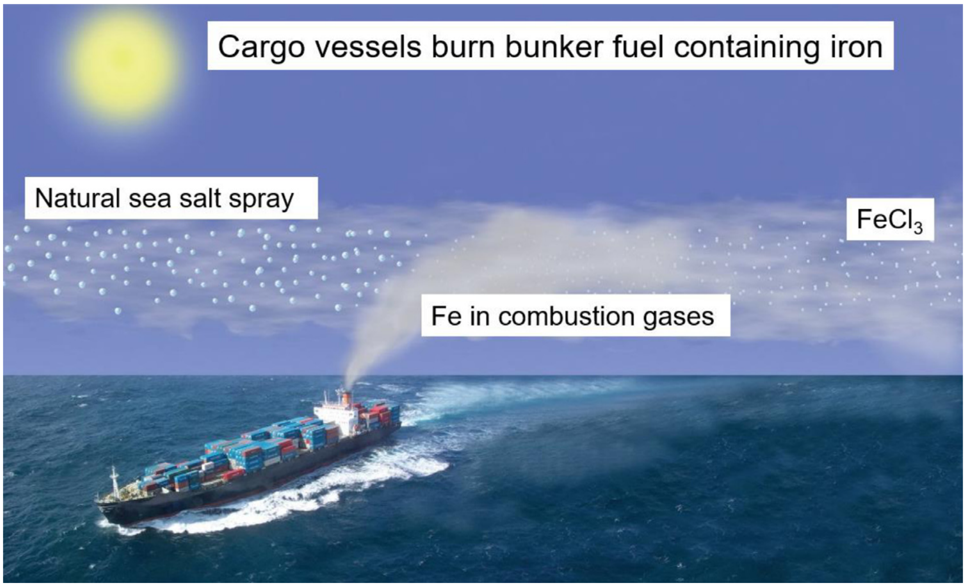

Fig. 2. A container ship powered by bunker fuel mixed with commercially available iron additives [73,74] which are sold to reduce black carbon and carbon monoxide (CO) emissions and to reduce fuel consumption. In the exhaust plume, the iron compounds react with sea salt to produce iron chloride ($FeCl_3$), which under sunlight generates Cl atoms [69] that oxidize CH_4 16 times faster than $^{\circ}OH$ radicals [71].

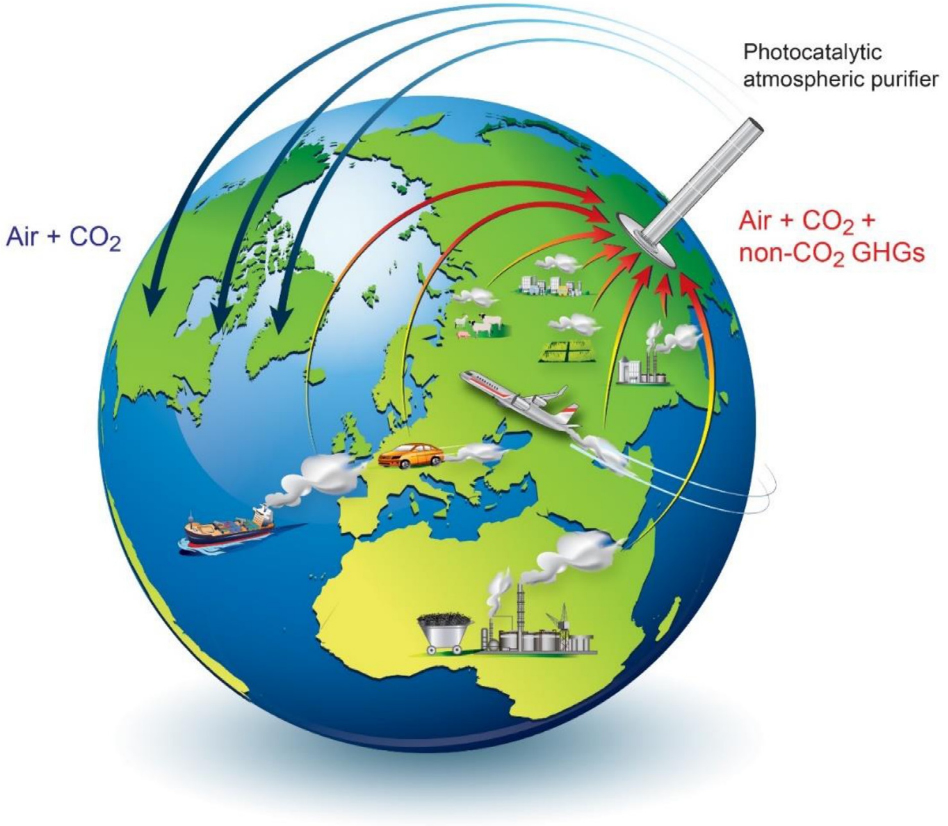

Fig. 3. A photocatalytic solar updraft tower concept to remove atmospheric CH_4, nitrous oxide (N_2O), and other GHGs, from [55].

4.4. Enhancing the natural mineral sink of CH_4–On surfaces

In 2017, it was proposed to perform CH_4 depletion by large scale photocatalysis [55] using solar updraft chimneys (SUT), which are structures able to process very large volumes of air, as illustrated by Fig. 3. As an example, a hypothetical 400-MW SUT would process 38,000 km^3 of air yearly [75,76].

The proposed photocatalyst for CH_4 depletion is a semiconductor metal oxide: a zinc oxide (ZnO) doped with 0.1% silver (Ag) [77]. It is expected to remove 50% of the CH_4 from the air processed thanks to sunlight at ambient temperatures [55].

Then in 2018, DAC devices were proposed to remove atmospheric CH_4 alongside CO_2, using the same ZnO-0.1%Ag photocatalyst illustrated by Fig. 4. The enormous energy costs of fan-driven DAC make it less attractive than passive generation of large airflows by the SUT devices proposed above. Nonetheless, it is anticipated that the DAC technology will develop rapidly to remove CO_2 from the atmosphere. Once DAC plants exist, profiting from this existing infrastructure by upgrading it to also remove CH_4 could enhance the capture yields in terms of

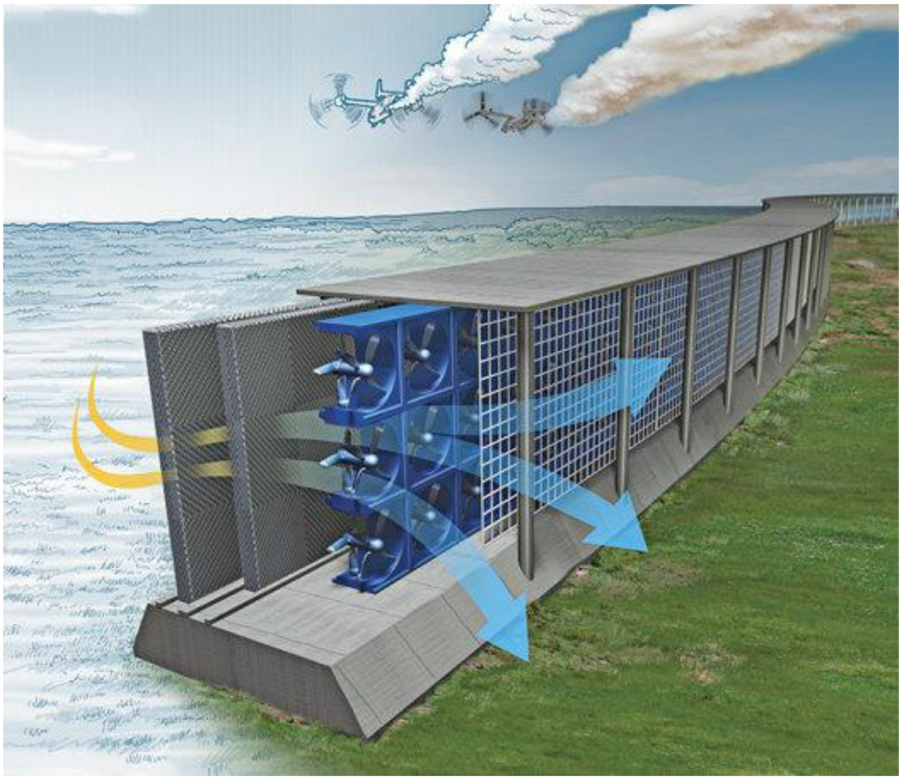

Fig. 4. A hypothetical industrial DAC device with added photocatalyst, proposed to oxidize CH_4 into CO_2 [57]. Reproduced from John Bradley [82].

CO_2-eq by 20%. Depending on the CH_4 oxidation yields and costs, it could be advantageous, especially since the removal of other GHGs also seems feasible [55,78,79] and does not introduce a pressure drop requiring more energy for the fans, and no additional CO_2 capture capacity would be required [55].

In 2019, other scientists proposed the use of DAC devices dedicated to CH_4 capture by zeolites and then removal of CH_4 using a thermal catalyst [58]. No published data was found on the pressure drop across the zeolite, making it difficult to know if the DAC device might be attractive to capture CO_2 also. Lackner [80] commented on this proposal by pointing to the extreme dilution of CH_4 in air (200 times more dilute than CO_2) which (based on the Sherwood law) may cause a three-order of magnitude energy penalty in using fan-driven DAC systems – thus suggesting that approaches should take advantage of natural air flow and use passive methods. Both the comment and the response [81] mention that it would be more interesting to remove N_2O, the third most important GHG by its radiative forcing, with an atmospheric lifetime estimated to 114 years and a GWP_{100} nearly 300 times higher than of CO_2. Such a proposal was made in 2016 [78].

Two proposals [55,57] also suggested removal of other GHGs (like N_2O, as well as many halogenated GHG gasses) with very high global warming potentials, which also damage the stratospheric ozone layer and are included in the Montreal Protocol. A long list of possible photocatalysts was proposed [78,79], mainly titanium dioxide derivatives, all acting at ambient temperature and activated by sunlight. Consequently, not only CH_4, but almost all non-CO_2 GHGs are targeted by the photocatalytic method [55].

In Table 1, the principal methods of CH_4 removal are summarized.

4.5. *Enhancing the sinks by the use of plants, trees, and microbes*

Mitigation of landfill CH_4 emissions using soil amendments such as biochar [94], and microbial CH_4 oxidation processes with bio-covers [90] or bio-trickling filters are well-established methods [95]. Adding methanotrophs to flooded paddy soil also mitigates CH_4 emissions [96]. Enhancing methanotrophic activity is among the mitigation methods proposed to prevent CH_4 from reaching the atmosphere [44,45]. Similar methods also might be possible for greenhouse gas removal (GGR), as it has been shown that an important CH_4 sink can be created by cropland reforestation [97]. One can probably imagine that for afforestation projects and for the “one trillion trees” initiative, planting trees that absorb tropospheric CH_4 [98] in addition to CO_2 (instead of plants and trees that emit CH_4 [98]), as recently observed in the seasonally flooded Amazon floodplain, is a good idea – especially if using local trees species, and if biodiversity is preserved or restored, without competing with agricultural land.

Airborne microbes are abundant in the atmosphere [99] and subject to long-range transport [100], so it might be possible to enhance the amount of CH_4 consuming microorganisms (methanotrophs) [101] for instance by enhancing the amount of methanotrophs already present on the bark of tree trunks [102]. Currently, plants, trees, and microbes represent 5%–6% of the sinks for atmospheric CH_4, but to our knowledge apart a brief mention [52], no large-scale strategy has yet been proposed to take advantage of these sinks.

5. Discussion

The mail benefit of returning to CH_4 pre-industrial levels will be to reduce global warming by up to 0.5°C [103], which can help reduce a temperature overshoot above 2°C by mid-century [104].

The expression “at a climatically significant scale” often appears in discussions about GHG removal, but lacks precise definition. Perhaps one benchmark could be the amount of GHG removal achieved up to now. According to the International Energy Agency [105], after 10 years of development, existing DAC installations captured just 9000 tons CO_2 yr^{-1} in 2019. The report of three U.S. National Academies [106] on negative emissions technologies (NETs) considers “coastal blue carbon” (mangroves) as being of interest, for a global potential of 0.13 Gt CO_2 removal per year and of only 0.02 Gt CO_2 yr^{-1} in the United States. After a half-century of development, solar photovoltaics

Table 1
Summary of the methane removal methods, technologies and estimated costs. For comparison, some methane mitigation strategies are also briefly described.

Method	Principal targets	Description, possible costs and comments	Ref.
Both for removal and mitigation of CH_4 [1]			
Zeolites + catalysis	Point sources such as ventilation systems; and global atmosphere	For removal, using air moving devices such as Direct CO_2 Air Capture plants associated with zeolites and a catalyst, if the DAC plant is only devoted to CH_4 capture and oxidation, costs might be slightly higher than for CO_2 capture [80] as tropospheric CH_4 is more than 200 times more diluted than CO_2 and air handling will consume more energy. But if passive systems are used and as on zeolites some catalysts can oxidize CH_4 at room temperature [83] and as no capture, no purification, no compression, no transport and no long-term storage is required in the case of CH_4 compared to CO_2, then global costs might be reduced, with a target cost-range of \$100 ton^{-1} of CO_2-eq [84]. In case of use of hybrid CO_2 DAC plants upgraded to also capture and oxidize CH4, the costs might be lower, as almost all the infrastructure and the air-flow already exist.	[85]
Photocatalysis on surfaces	Point sources such as ventilation systems; and global atmosphere	The estimated cost ton^{-1} CO_2-eq is \$166 by 2030 with a target of \$100 by 2040 [84].	[55]
Generation of hydroxyl radicals	Point sources such as ventilation systems; and global atmosphere	Estimations for the infrastructure requested, results in cost-range of \$200–1000 per ton of CO_2-eq [84]	[64]
Generation of Cl atoms	Point sources such as ventilation systems; and global atmosphere	For CH_4 removal using chlorine atoms, directly in the troposphere, mimicking natural processes costs estimates range from \$ 54 to as low as \$1.7 ton^{-1} CO_2-eq [86]. A startup targets costs of about \$1.6 ton^{-1} CO_2-eq [87]. Those estimations of the costs look very favorable but are uncertain prior to demonstration, waiting for field trails which cannot start before a full-scale environmental assessment has been conducted. For CH_4 mitigation at point sources, for instance in ventilation systems of coal mines where CH_4 is more concentrated, costs might even be lower as the infrastructure and the air-flow already exist and the generation of Cl atoms can be made by photolysis of Cl_2 gas, produced by the well-established chlor-alkali industrial process. Possible co-benefit: iron salt aerosols provide iron to depleted oceans, with possible CO_2 capture in the oceans at costs about \$1 ton^{-1} of CO_2 [72] based on the "Redfield ratio" of oceanic C-N-P-Fe stoichiometry and assuming 10% sequestration in the bottom of the oceans.	[54,72]
Methanotrophic bacteria	Point sources	(see next section) Spraying methanotrophs cultures on point sources such as trees which transfer CH_4 from underground to the atmosphere, or over large thawing permafrost areas and wetlands	[52]
CH_4 mitigation only[2]			
Food modification	Cattle breeding to reduce enteric CH_4 emissions	Among effective feed additives for beef–3-nitrooxypropanol and nitrates (respectively 22% and 14% CH_4 reduction) Many other feeds are effective, but less: chestnut, coconut, grape pomace, linseed, red seaweed… The practice may not be generalizable. Probably low global impact.	[46,88]
Biochar	Manure Sewage sludge	Co-composting can reduce CH_4 emissions by about 80%	[46]
Bio-covers For enhancing anaerobic oxidation of CH_4	Landfills with municipal solid waste	Several types of bio-covers exist, oxygenation of the soil is necessary. A better future strategy consists of removing all fermenTable organic matter from new landfills.	[89,90]
Leak repair	Oil & gas industry	The "methane pledge" signed at COP26 targets 30% reduction by 2030, from oil and gas industries.	[3,91]
Thermal catalysis	Coal mines (Ventilation systems of)	Usually applicable for CH_4 concentration about 0.5–1% as the reaction is exothermic it can be self-sustained. If CH_4 >1.5% energy generation is possible.	[92,93]

[1] Although it is yet too early in the development process to have accurate cost estimates for CH_4 removal directly in the troposphere, initial estimations have been provided

[2] Provided for illustration purposes only, as mitigation is out of the scope of this review and has been reviewed elsewhere [15,43-46].

and wind turbines are now cost-competitive with other electricity generating technologies. In 2018, PV and wind turbines avoided respectively 0.15 and 0.37 gigatons of CO_2-equivalent per year (Mt CO_2-eq yr^{-1}) [107,108], compared to only a few tons per year five decades ago. Even though these savings are still small compared to global CO_2 emissions (currently about 40 gigatons CO_2-eq yr^{-1}), many people would consider them significant. When "net-zero" is reached in the second half of this century, renewable energy will be preponderant.

Without knowing much about potential energy costs, and costs of scaling up, it is very difficult to understand and predict how feasible CH_4 removal technologies can be. But in our opinion, even if during the first decade following their invention, a very small-scale effective efficiency is obtained at very high cost, technologies able to avoid GHG emissions or able to remove GHGs already in the atmosphere deserve attention as soon as their scalability and globalization seems possible. Otherwise, the criterion of "at a climatically significant scale" may lead to too many options being dismissed and too few remaining, while the scale of the global warming problem requires a large portfolio of methods and technologies to be developed. A significant scale might be achieved as the sum of many contributions that are not individually very significant.

There are several direct and indirect co-benefits to reduced CH_4 atmospheric concentrations. The rapid climate benefits of reducing the concentration of CH_4 in the atmosphere are significant [15] for agriculture and the economy [109,110] as the tropospheric ozone burden enhanced by CH_4 will also be reduced [103]. Lower surface ozone concentrations will increase crops yields and global photosynthesis, potentially allowing some CO_2 removal [51]. The co-benefits for human health are numerous [111], as it will reduce hospitalizations, asthma and pulmonary diseases and premature deaths due to the linked ozone pollution [11]. Development of CH_4 removal methods is still in its infancy and requires more research, development and funding [85].

Some of the CH_4 enhanced oxidation methods proposed will have other co-benefits. As $^{\circ}OH$ and Cl atoms are very reactive and not very selective, by enhancing their generation several other GHGs and atmospheric pollutants will be removed faster than CH_4. This includes, for instance VOCs, whose removal will help reduce CH_4 lifetime [112]. Such

VOCs include also organo-halogens, human made hydrofluorocarbons and hydrochlorofluorocarbons, as well as natural biogenic halogenated-methane compounds produced mainly by oceanic plankton and bacteria [113]. The latter are not considered to be GHGs, but they participate in the stratospheric ozone layer natural cycle of destruction [114]. By reducing faster the amount of natural biogenic halogenated-methane compounds in the lower troposphere, as well as by reducing the water content of the stratosphere due to CH_4 oxidation [19], the ozone layer might recover faster.

Of course, the main benefit of returning to CH_4 pre-industrial levels will be to reduce global warming by up to about 0.5°C [103], which can help reduce a temperature overshoot above 2°C by mid-century [104].

6. Concluding remarks

This perspective article has discussed different strategies (some already proposed and several new ones) to accelerate the removal of already emitted CH_4, reducing its radiative forcing by direct and indirect effects. A reduction of the atmospheric CH_4 burden might help the ozone layer to recover faster and will have rapid climate benefits together with significant co-benefits for agriculture, human health and the economy.

Unlike CO_2 removal methods, the CH_4 removal methods described here do not require capture and long-term geological sequestration as for CO_2, as they only accelerate the natural oxidation processes that will anyway occur with the products remaining in the atmosphere. By returning to CH_4 pre-industrial levels, the increase of atmospheric CO_2 resulting from CH_4 oxidation is small compared to global CO_2 annual emissions, while the reduction of the radiative forcing could be significant. Net warming could be reduced by about 0.5°C.

Although CH_4 enhanced oxidation methods can be applied both to global tropospheric CH_4 and to some local concentrated sources, removal and mitigation strategies and methods do not necessarily target the same sources and are complementary. In case of an abrupt acceleration of CH_4 emissions from natural sources (e.g. submarine methane-hydrates, or wetlands), the availability of effective and proven techniques would constitute an assurance to avoid a rapid acceleration of global warming.

Those innovative methods deserve more attention from the scientific community to help evaluate their potential risks, costs, public acceptability, and societal appropriation. Together with CO_2 and CH_4 mitigation and with CO_2 removal, CH_4 removal methods can help fight climate change, win time by slowing down warming and thus meet the targets of the Paris Agreement with limited temperature overshoot.

Declaration of Competing Interests

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

CRediT authorship contribution statement

Tingzhen Ming: Conceptualization. **Wei Li:** Writing – original draft. **Qingchun Yuan:** Writing – original draft. **Philip Davies:** Writing – original draft. **Renaud de Richter:** Conceptualization. **Chong Peng:** Writing – original draft. **Qihong Deng:** Writing – original draft. **Yanping Yuan:** Writing – original draft. **Sylvain Caillol:** Writing – original draft. **Nan Zhou:** Formal analysis, Writing – original draft.

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