



Methane Removal

IGSD Background Note

R&D needed for removing methane from the atmosphere

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Institute for Governance & Sustainable Development

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About the Institute for Governance & Sustainable Development

IGSD's mission is to promote just and sustainable societies and to protect the environment by advancing the understanding, development, and implementation of effective and accountable systems of governance for sustainable development.

As part of its work, IGSD is pursuing “fast-action” climate mitigation strategies that will result in significant reductions of climate emissions to limit temperature increase and other climate impacts in the near-term. The focus is on strategies to reduce non-CO₂ climate pollutants, protect sinks, and enhance urban albedo with smart surfaces, as a complement to cuts in CO₂. It is essential to reduce both non-CO₂ pollutants and CO₂, as neither alone is sufficient to provide a safe climate.

IGSD's fast-action strategies include reducing emissions of the short-lived climate pollutants—black carbon, methane, tropospheric ozone, and hydrofluorocarbons (HFCs). Reducing HFCs starting with the Kigali Amendment to the Montreal Protocol has the potential to avoid up to 0.5 °C of warming by end of century. Parallel efforts to enhance energy efficiency of air conditioners and other cooling appliances during the phase down of HFCs can double the climate benefits at 2050, and by 2060 avoid the equivalent of up to 460 billion tonnes of CO₂.

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Background Note on Methane Removal

R&D needed to determine potential for removing methane from the atmosphere

30 September 2021

1. Introduction and summary

Learning how to remove methane from the atmosphere would complement strategies to mitigate methane emissions from anthropogenic sources. Methane removal technologies would be especially important because around 40% of atmospheric methane originates from natural sources,¹ and because some anthropogenic emissions are difficult to reduce. Natural methane emissions come primarily from wetlands, but also, increasingly, from thawing permafrost, and there is a risk of high-impact emissions from undersea methane hydrates.² Although these emissions come from natural sources, they are enhanced by global warming, and as warming accelerates these emissions will likely continue to grow.³

Both mitigation and removal technologies would provide crucial levers in the struggle to slow warming rapidly enough to limit irreversible and potentially catastrophic climate change.⁴ Indeed, reducing methane concentrations is the most effective way to rapidly limit warming. A new modelling study by a Stanford University-led team calculates that removing around three years' worth of human-caused methane emissions would reduce warming by 0.21 °C, two orders of magnitude greater than the temperature reductions from removing the same amount of CO₂ (1 petagram).⁵ These results compare with the 2021 [Global Methane Assessment](#), which concluded that reducing anthropogenic methane emissions by 45% could avoid nearly 0.3 °C of warming by 2045.⁶

In April 2021, the U.S. Department of Energy's Advanced Research Projects Agency–Energy (ARPA-E) announced a \$35 million program to reduce methane emissions in the oil, gas, and coal sectors.⁷ In developing this program, ARPA-E recognized the need for further research on methane capture in parallel with efforts to capture CO₂.⁸ While removing methane could provide more avoided warming than removing the same amount of CO₂, the fact that methane is 600 times more diluted than CO₂ in the general atmosphere presents its own challenge.⁹ Since 2003, studies have highlighted two potential pathways for research into anthropogenic methane removal: catalytic oxidation and methanotrophic bacterial filtration. Preventing natural methane formation is an additional pathway.

2. Catalytic oxidation

Most research has focused on catalytic surfaces, including platinum and palladium,¹⁰ and more recently iron-containing zeolites¹¹ and porous polymer networks.¹² Iron salt aerosols have been proposed as a cost-effective mechanism of potential climate intervention due to their higher surface area and catalytic efficiency. Soluble iron hydroxides react with the hydrogen chloride in ocean spray to form an iron salt aerosol. This aerosol acts as a photocatalyst, accelerating a natural

process of generating chlorine atoms that oxidize methane,¹³ as well as bromine atoms¹⁴ that destroy ozone.¹⁵ Although the hydroxyl radical is responsible for a far greater share of natural methane oxidation than chlorine is, chlorine has a reaction rate approximately 16 times higher.¹⁶ Iron salt aerosols occur naturally at large scale as airborne dust, but fossil-fuel combustion is also responsible for significant iron emissions.¹⁷ The availability of soluble iron is directly linked to oceanic phytoplankton growth.¹⁸ Groups advocating for iron salt catalysis have suggested that the emission of iron salts could be rapidly scaled up by modifying pre-existing sources of iron emissions, or that catalysis systems could be installed at methane point sources.¹⁹

3. Methanotrophic bacteria

The second major pathway for methane removal involves methanotrophic bacteria. Methanotrophs are “ubiquitous” in earth’s ecosystems, including anaerobic environments such as those where methane forms.²⁰ Experiments with methanotrophic filters have employed bacteria to process relatively concentrated methane, such as that found in coal mines, and above landfills and manure lagoons.²¹ Naturally occurring methanotrophs have been described as a “microbial lid” mitigating emissions from undersea methane deposits.²²

4. Other pathways

Alternatives to natural methane removal may include measures to prevent natural methane formation. Preliminary studies suggest the use of additives such as biochar, acids, straw, or technology based on calcium sulfate dihydrate (gypsum) could reduce methane emissions from manure lagoons, rice paddies, and other anthropogenic methane sources.²³ Similar methods might be applied to natural and semi-natural sources, such as wetlands and reservoirs.

5. Research agenda

Implementing a systematic methane removal research agenda would prepare policymakers to respond more flexibly to the challenge of atmospheric methane, particularly the significant methane emissions from natural sources. A recent paper proposes a research agenda for methane removal, including the creation of a Methane Removal Model Intercomparison Project.²⁴ The paper identifies six areas for further modelling, including how the timing, amount of methane removed, and location affects removal’s impacts, as well as how methane removal would interact with other mitigation approaches.²⁵ More research on technological design and scalability is also needed to test the effectiveness of methane removal techniques.²⁶

References

¹ United Nations Environment Programme & Climate & Clean Air Coalition (2021) [GLOBAL METHANE ASSESSMENT: BENEFITS AND COSTS OF MITIGATING METHANE EMISSIONS](#), 28 (“Approximately 60 per cent of total global methane emissions come from anthropogenic sources. Of these, more than 90 per cent originate from three sectors: fossil fuels, ~35 per cent; agriculture, ~40 per cent; and waste, ~20 per cent.”). *See also* Saunio M., *et al.* (2020) [The Global Methane Budget 2000-2017](#). *EARTH SYST. SCI. DATA* 12:1561-1623 (“For the 2008–2017 decade, global methane emissions are estimated by atmospheric inversions (a top-down approach) to be 576 Tg CH₄ yr⁻¹ (range 550–594, corresponding to the minimum and maximum estimates of the model ensemble). Of this total, 359 Tg CH₄ yr⁻¹ or ~ 60 % is attributed to anthropogenic sources, that is emissions caused by direct human activity (i.e. anthropogenic emissions; range 336–376 Tg CH₄ yr⁻¹ or 50 %–65 %).”).

² Intergovernmental Panel on Climate Change (2021) [CLIMATE CHANGE 2021: THE PHYSICAL SCIENCE BASIS](#), Masson-Delmotte V., *et al.* (eds.), 5-34, 5-67 (“The largest natural sources are from wetlands, freshwater and geological process, while the largest anthropogenic emissions are from enteric fermentation and manure treatment, landfills and waste treatment, rice cultivation and fossil fuel exploitation (Table 5.2). . . . In conclusion, thawing terrestrial permafrost will lead to carbon release under a warmer world (high confidence). However, there is low confidence on the timing, magnitude and linearity of the permafrost climate feedback owing to the wide range of published estimates and the incomplete knowledge and representation in models of drivers and relationships. It is projected that CO₂ released from permafrost will be 18 (3.1–41) PgC per 1°C by 2100 with the relative contribution of CO₂ vs CH₄ remaining poorly constrained. Permafrost carbon feedbacks are included among the underrepresented feedbacks quantified in Figure 5.29.”). *See also* Wadhams P. (2017) [A FAREWELL TO ICE: A REPORT FROM THE ARCTIC](#), Oxford University Press, 4 (“The consequences of the collapse of Arctic summer ice will be dramatic. . . . When the overlying ice is gone, the surface water can warm up by several degrees in summer (satellite observations have shown 7°C), and over the shallow continental shelves wind-induced mixing extends this heat down to the seabed. This then thaws the surface layer of the offshore permafrost, frozen seabed sediments which have lain there undisturbed since the last Ice Age. The thawing offshore permafrost will trigger the release of huge plumes of methane from the disintegration of methane hydrates trapped in the sediment.”).

³ Lan X., *et al.* (2021) [Improved Constraints on Global Methane Emissions and Sinks Using \$\delta^{13}\text{C-CH}_4\$](#) , *GLOBAL BIOGEOCHEM. CY.* 35(6): e2021GB007000 1 (“We find that the fossil fuel (FF) CH₄ emission trend from the Emissions Database for Global Atmospheric Research 4.3.2 inventory does not agree with observed $\delta^{13}\text{C-CH}_4$. Increased FF CH₄ emissions are unlikely to be the dominant driver for the post-2006 global CH₄ increase despite the possibility for a small FF emission increase. We also find that a significant decrease in the abundance of hydroxyl radicals (OH) cannot explain the post-2006 global CH₄ increase since it does not track the observed decrease in global mean $\delta^{13}\text{C-CH}_4$. Different CH₄ sinks have different fractionation factors for $\delta^{13}\text{C-CH}_4$, thus we can investigate the uncertainty introduced by the reaction of CH₄ with tropospheric chlorine (Cl), a CH₄ sink whose abundance, spatial distribution, and temporal changes remain uncertain. Our results show that including or excluding tropospheric Cl as a 13Tg/year CH₄ sink in our model changes the magnitude of estimated fossil emissions by ~20%. We also found that by using different wetland emissions based on a static versus a dynamic wetland area map, the partitioning between FF and microbial sources differs by 20 Tg/year, ~12% of estimated fossil emissions.”). *See also* Wik M., *et al.* (2016) [Climate-sensitive northern lakes and ponds are critical components of methane release](#), *NAT. GEOSCI.* 9(2): 99–105. (abstract) (“Lakes and ponds represent one of the largest natural sources of the greenhouse gas methane. By surface area, almost half of these waters are located in the boreal region and northwards. A synthesis of measurements of methane emissions from 733 lakes and ponds north of ~50° N, combined with new inventories of inland waters, reveals that emissions from these high latitudes amount to around 16.5 Tg CH₄ yr⁻¹ (12.4 Tg CH₄-C yr⁻¹). This estimate — from lakes and ponds alone — is equivalent to roughly two-thirds of the inverse model calculation of all natural methane sources in the region. Thermokarst water bodies have received attention for their high emission rates, but we find that post-glacial lakes are a larger regional source due to their larger areal extent. Water body depth, sediment type and ecoclimatic region are also important in explaining variation in methane fluxes. Depending on whether warming and permafrost thaw cause expansion or contraction of lake and pond areal coverage, we estimate that annual water body emissions will increase by 20–54% before the end of the century if ice-free seasons are extended by 20 days. We conclude that lakes and ponds are a dominant methane source at high northern latitudes.”); Schaefer K., Lantuit H., Romanovsky V. E., Schuur E. A. G., & Witt R. (2014) [The Impact of the Permafrost Carbon Feedback on Global Climate](#), *ENVIRON. RES. LETT.* 9: 1–9, 2 (“If temperatures rise and permafrost thaws, the organic

material will also thaw and begin to decay, releasing carbon dioxide (CO₂) and methane (CH₄) into the atmosphere and amplifying the warming due to anthropogenic greenhouse gas emissions ... The PCF is irreversible on human time scales because in a warming climate, the burial mechanisms described above slow down or stop, so there is no way to convert CO₂ into organic matter and freeze it back into the permafrost.”); Intergovernmental Panel on Climate Change (2021) *Technical Summary*, in *CLIMATE CHANGE 2021: THE PHYSICAL SCIENCE BASIS*, Masson-Delmotte V., *et al.* (eds.), TS-60 (“Natural sources and sinks of non-CO₂ greenhouse gases such as methane (CH₄) and nitrous oxide (N₂O) respond both directly and indirectly to atmospheric CO₂ concentration and climate change, and thereby give rise to additional biogeochemical feedbacks in the climate system. Many of these feedbacks are only partially understood and are not yet fully included in ESMs. There is medium confidence that the net response of natural ocean and land CH₄ and N₂O sources to future warming will be increased emissions, but the magnitude and timing of the responses of each individual process is known with low confidence. {5.4.7}”); Wadhams P. (2017) *A FAREWELL TO ICE: A REPORT FROM THE ARCTIC*, Oxford University Press; and Shakohva N., Semiletov I., & Chuvilin E. (2019) *Understanding the Permafrost-Hydrate System and Associated Methane Releases in the East Siberian Arctic Shelf*, *GEOSCI.* 9(251): 1–23. *See also* Mendoza-Pascual M. U., Itoh M., Aguilar J. I., Padilla K. S. A. R., Papa R. D. S., & Okuda N. (2021) *Controlling Factors of Methane in Tropical Lakes of Different Depths*, *J. GEOPHYS. RES. BIOGEOSCI.* 126(4): e2020JG005828; *as discussed in* Wheeling K. (30 April 2021) *Tropical Lakes May Emit More Methane*, *EOS* 102 (“Water temperature was also linked to methane production, and higher concentrations of methane can be found in the profundal, or deep, zones of tropical lakes compared to temperate or subtropical lakes. Overall, the study suggests that lakes with higher water temperatures may contribute more to methane emissions, with tropical lakes emitting between 58% and 400% more methane than lakes in more temperate zones.”).

⁴ Jackson R. B., Solomon E. I., Canadell J. G., Cargnello M., & Field C. B. (2019) *Methane removal and atmospheric restoration*, *NAT. SUSTAIN.* 2: 436–438, 436 (“In contrast to negative emissions scenarios for CO₂ that typically assume hundreds of billions of tonnes removed over decades and do not restore the atmosphere to preindustrial levels, methane concentrations could be restored to ~750 ppb by removing ~3.2 of the 5.3 Gt of CH₄ currently in the atmosphere. Rather than capturing and storing the methane, the 3.2 Gt of CH₄ could be oxidized to CO₂, a thermodynamically favourable reaction.... In total, the reaction would yield 8.2 additional Gt of atmospheric CO₂, equivalent to a few months of current industrial CO₂ emissions, but it would eliminate approximately one sixth of total radiative forcing. As a result, methane removal or conversion would strongly complement current CO₂ and CH₄ emissions-reduction activities. The reduction in short-term warming, attributable to methane’s high radiative forcing and relatively short lifetime, would also provide more time to adapt to warming from long-lived greenhouse gases such as CO₂ and N₂O.”). Klaus Lackner critiqued the Jackson *et al.* article in a published response, arguing that implementing zeolite mechanisms to facilitate CH₄ removal is not practical. Lackner noted CH₄ removal faces the challenge of extreme dilution in the atmosphere, so “the amount of air that would need to be moved [to facilitate CH₄ removal] would simply be too great” to be economically feasible. However, Lackner did note passive methods of CH₄ removal through the use of zeolites may still be a viable solution. Lackner further argues that N₂O may be a more worthy target for removal due to its long lifetime in the atmosphere. *See* Lackner K. S. (2020) *Practical Constraints on Atmospheric Methane Removal*, *NAT. SUSTAIN.* 3: 357. Jackson *et al.* published a response to Lackner, acknowledging his stature in the greenhouse gas removal field and his concerns about the feasibility and energy requirements of their proposed mechanism, offering additional explanation about alternative options for use of the captured methane instead of just converting it to CO₂ as suggested in the original study. *See* Jackson R. B., Solomon E. I., Canadell J. G., Cargnello M., Field C. B., & Abernethy S. (2020) *Reply to: Practical constraints on atmospheric methane removal*, *NAT. SUSTAIN.* 3: 358–359. Another study looking at removing non-CO₂ GHGs investigated the potential of using solar chimney power plants (SCPPs) with select photocatalysts (depending on what GHGs desired to be captured). While the SCPP serves as a source of renewable energy that could remove methane and nitrous oxide among other atmospheric pollutants, scaling up the prototype would require a massive amount of land area (roughly 23 times the size of the entire Beijing municipality) and a chimney stretching 1000–1500 m into the air, which limits how practical the existing technology may be. *See* de Richter R., Tingzhen M., Davies P., Wei L., & Caillol S. (2017) *Removal of non-CO₂ greenhouse gases by large-scale atmospheric solar photocatalysis*, *PROG. ENERGY COMBUST. SCI.* 60: 68–96 (“Large-scale atmospheric removal of greenhouse gases (GHGs) including methane, nitrous oxide and ozone-depleting halocarbons could reduce global warming more quickly than atmospheric removal of CO₂. Photocatalysis of methane oxidizes it to CO₂, effectively reducing its global warming potential (GWP) by at least 90%.”); Methane Action (16 April 2021) *Scientists’ Statement on Lowering Atmospheric Methane Concentrations* (“To deal with methane emissions that can’t otherwise be mitigated, to reduce the overall methane burden, and to get atmospheric methane levels to a range consistent with meeting climate goals, we must combine prevention and mitigation of new methane emissions with actively lowering the concentration of methane already in the

atmosphere.”); and Jackson R. B. & Wysham D. (28 September 2021) *Focus on methane is timely and appropriate*, THE HILL.

⁵ Abernethy S., O’Connor F. M., Jones C. D., & Jackson R. B. (2021) *Methane removal and the proportional reductions in surface temperature and ozone*, PHIL. TRANS. R. SOC. A 379: 1–13, 6 (“Due to the temporal nature of effective cumulative removal, comparisons between methane and carbon dioxide depend on the timescale of interest. The equivalent of MCR for carbon dioxide, the TCRE, is $0.00048 \pm 0.0001^\circ\text{C}$ per Pg CO₂ [38], two orders of magnitude smaller than our MCR estimate of $0.21 \pm 0.04^\circ\text{C}$ per effective Pg CH₄ removed (figure 2). Accounting for the time delay for carbon dioxide removal due to the lagged response of the deep ocean, the TCRE for CO₂ removal may be even lower [39]. If 1 year of anthropogenic emissions was removed (0.36 Pg CH₄ [3] and 41.4 Pg CO₂ [40]), the transient temperature impact would be almost four times larger for methane than for CO₂ (0.075°C compared to 0.02°C). Using this example, however, maintaining a steady-state response of 0.36 Pg CH₄ effectively removed would require the ongoing removal of roughly 0.03Pg CH₄ yr⁻¹, since a removal rate of E/τ is required to maintain an effective cumulative removal of E .”). *Discussed in* Jordan R. (26 September 2021) *Stanford-led research reveals potential of an overlooked climate change solution: Analyses lay out a blueprint for speeding development of atmospheric removal and modeling how the approach could improve human health and have an oversized effect on reducing future peak temperature*, STANFORD WOODS INSTITUTE FOR THE ENVIRONMENT (“The analyses, published Sept. 27 in *Philosophical Transactions of the Royal Society A*, reveal that removing about three years-worth of human-caused emissions of the potent greenhouse gas would reduce global surface temperatures by approximately 0.21 degrees Celsius while reducing ozone levels enough to prevent roughly 50,000 premature deaths annually. The findings open the door to direct comparisons with carbon dioxide removal – an approach that has received significantly more research and investment – and could help shape national and international climate policy in the future. [...] Under a high emissions scenario, the analysis showed that a 40 percent reduction in global methane emissions by 2050 would lead to a temperature reduction of approximately 0.4 degrees Celsius by 2050. Under a low emissions scenario where temperature peaks during the 21st century, methane removal of the same magnitude could reduce the peak temperature by up to 1 degree Celsius.”).

⁶ United Nations Environment Programme & Climate & Clean Air Coalition (2021) *GLOBAL METHANE ASSESSMENT: BENEFITS AND COSTS OF MITIGATING METHANE EMISSIONS*, 8 (“Reducing human-caused methane emissions is one of the most cost-effective strategies to rapidly reduce the rate of warming and contribute significantly to global efforts to limit temperature rise to 1.5°C. Available targeted methane measures, together with additional measures that contribute to priority development goals, can simultaneously reduce human-caused methane emissions by as much as 45 per cent, or 180 million tonnes a year (Mt/yr) by 2030. This will avoid nearly 0.3°C of global warming by the 2040s and complement all long-term climate change mitigation efforts. It would also, each year, prevent 255 000 premature deaths, 775 000 asthma related hospital visits, 73 billion hours of lost labour from extreme heat, and 26 million tonnes of crop losses globally.”).

⁷ Advanced Research Projects Agency-Energy (8 April 2021) *Reducing Emissions of Methane Every Day of the Year*, ARPA-E Programs (“Program Description: REMEDY (Reducing Emissions of Methane Every Day of the Year) is a three-year, \$35 million research program to reduce methane emissions from three sources in the oil, gas, and coal value chains: 1) Exhaust from 50,000 natural gas-fired lean-burn engines. These engines are used to drive compressors, generate electricity, and increasingly repower ships. 2) The estimated 300,000 flares required for safe operation of oil and gas facilities. 3) Coal mine ventilation air methane (VAM) exhausted from 250 operating underground mines. These sources are responsible for at least 10% of U.S. anthropogenic methane emissions. Reducing emissions of methane, which has a high greenhouse gas warming potential, will ameliorate climate change.”).

⁸ Advanced Research Projects Agency-Energy (30 September 2020) *Prevention and Abatement of Methane Emissions* (“We’re open to all options – but specifically are looking for solutions that: Prevent methane emissions from anthropogenic activities. In other words, solutions which intervene before anthropogenic emissions escape to the atmosphere. Abate methane emissions at their source. Sources include vents, leaks, and exhaust stacks. Remove methane from the air. As mentioned above, methane only lasts about 9 years in the atmosphere. Nature is very good at getting rid of methane using reactions in the atmosphere and methanotrophs in the soil. Maybe we can learn from Nature, and help her out.”). *See also* Lewnard J. (16 November 2020) *REMEDY – Reducing Emissions of Methane Every Day of the Year*, ARPA-E Presentation, Slide 7 (“Example Potential Approaches, Not Intended to Limit or Direct... “Geo-engineering”: Accelerate tropospheric reactions; Accelerate soil/methanotroph reactions”).

⁹ Lackner K. S. (2020) *Practical constraints on atmospheric methane removal*, NAT. SUSTAIN. 3: 357 (“Methane removal poses two challenges: extreme dilution and competition from natural processes. This raises the question of whether methane is really the best target for removal from the air. First, the dilute concentration of methane in the atmosphere challenges economical removal. On a mass basis, methane is currently 600 times more dilute in Earth’s atmosphere than carbon dioxide; in pre-industrial times it was 1,000 times more dilute.”).

¹⁰ Janbey A., *et al.* (2003) *Noble metal catalysts for methane removal*, CHEMOSPHERE 52: 1041–1046, 1041 (“Using a bench-scale rig, the activities of Pt, Pd and Pt + Pd catalysts supported on γ -Al₂O₃ and on TiO₂ (anatase) for the complete oxidation of methane (300 ppmv) in air have been measured as a function of temperature; values of T10, T50 and T90 together with the Arrhenius parameters (activation energy and pre-exponential factor) are reported. Pt is less active than Pd when deposited on the surface of the TiO₂, but more active when deposited on γ -Al₂O₃, however when combined, the Pt + Pd mixture is more active than either metal individually. The T10 for Pt + Pd/ γ -Al₂O₃ was being as low as 228 °C. The significance of the Arrhenius parameters, for metal containing catalysts is that they exhibit compensation with increasing activation energy, while securing a more rapid increase in conversion from 0% to 100% when the temperature is increased.”).

¹¹ See Snyder B. E. R., Vanelderden P., Bols M. L., Hallaert S. D., Böttger L. H., Ungur L., Pierloot K., Schoonheydt R. A., Sels B. F., & Solomon E. I. (2016) *The active site of low-temperature methane hydroxylation in iron-containing zeolites*, NATURE 536(7616): 317–321; and Jackson R. B., Solomon E. I., Canadell J. G., Cargnello M., & Field C. B. (2019) *Methane removal and atmospheric restoration*, NAT. SUSTAIN. 2: 436–438.

¹² de Richter R., Tingzhen M., Davies P., Wei L., & Caillol S. (2017) *Removal of non-CO₂ greenhouse gases by large-scale atmospheric solar photocatalysis*, PROG. ENERGY COMBUST. SCI. 60: 68–96.

¹³ Wittmer J., Bleicher S., & Zetzsch C. (2015) *Iron(III)-Induced Activation of Chloride and Bromide from Modeled Salt Pans*, J. PHYS. CHEM. A, 119(19): 4373–4385, 4382 (“In summary, our study demonstrates the photocatalytic effect of Fe(III) causing gaseous Cl production and the influence of inorganic and organic contaminants including the underlying mechanisms that explain the various observed Cl and Br sources.”). See also Wittmer J. & Zetzsch C. (2017) *Photochemical activation of chlorine by iron-oxide aerosol*, J. ATMOS. CHEM. 74: 187–204, 187 (“The photochemical activation of chlorine by dissolved iron in artificial sea-salt aerosol droplets and by highly dispersed iron oxide (Fe₂O₃) aerosol particles (mainly hematite, specific surface ~150 m² g⁻¹) exposed to gaseous HCl, was investigated in humidified air in a Teflon simulation chamber. Employing the radical-clock technique, we quantified the production of gaseous atomic chlorine (Cl) from the irradiated aerosol. . . . The resulting Cl production rates ranged from 8 × 10²⁰ Cl atoms cm⁻² h⁻¹ (at ~4 ppb HCl) to 5 × 10²² Cl atoms cm⁻² h⁻¹ (at ~350 ppb HCl) and confirmed the uptake and conversion of HCl to atomic Cl (at HCl to Cl conversion yields of 2–5 %, depending on the relative humidity.”); and Zetzsch C., Bleicher S., & Wittmer J. (2013) *Smog Chamber Investigation on the Iron-Catalyzed Activation of Chloride from Sea-salt for a Depletion of Tropospheric Methane*, AMERICAN GEOPHYSICAL UNION, Fall Meeting (“Investigations on NaCl salt pans in an environmental simulation chamber under artificial sunlight yield high concentrations of chlorine atoms in the gas phase if FeCl₃ is added to the salt mixture. The impact of chlorine atoms led in a direct experiment to a methane reduction of 8% within 24 h. This corresponds to a methane lifetime of only 12 days in a constantly illuminated chamber (or a half-life of 8 days). In quantitative measurements almost 1E6 Cl atoms per cm³ were observed as peak concentrations in the chamber above mixtures of NaCl and 0.5% FeCl₃. This would decrease the lifetime of methane under atmospheric conditions of usually 8-10 years down to a hundredth of it.”).

¹⁴ Wittmer J., Bleicher S., & Zetzsch C. (2015) *Iron(III)-Induced Activation of Chloride and Bromide from Modeled Salt Pans*, J. PHYS. CHEM. A, 119(19): 4373–4385, 4382 (“Chlorine is a strong oxidant and is highly reactive with organics having, for example, a 16 times higher reaction rate than OH toward the climate forcing greenhouse gas methane. While it reacts relatively slowly with organics, it leads to surface-level O₃ destruction, mainly initiated by the autocatalytic release of bromine from sea salt.”).

¹⁵ Thompson C. R., *et al.* (2015) *Interactions of bromine, chlorine, and iodine photochemistry during ozone depletions in Barrow, Alaska*, ATMOS. CHEM. PHYS. 15(16): 9651–9679, 9673 (“This work has demonstrated that bromine chemistry is clearly the dominant destruction pathway for ozone depletion episodes, but that chlorine and, especially, iodine, can contribute significantly to both the rate and timescale of ozone depletion.”).

¹⁶ Witmer J., Bleicher S., & Zetzsch C. (2015) *Iron(III)-Induced Activation of Chloride and Bromide from Modeled Salt Pans*, J. PHYS. CHEM. A, 119(19): 4373–4385, 4373 (“Among the VOCs, the short-lived greenhouse gas methane (CH₄) is of major interest in climate research to date. Atomic Cl has a 16 times faster reaction rate constant toward methane in comparison to hydroxyl radicals at 298 K.”).

¹⁷ Lin Y. C., Chen J.-P., Ho T.-Y., Tsai I.-C. (2015) *Atmospheric iron deposition in the north-western Pacific Ocean and its adjacent marginal seas: the importance of coal burning*, GLOBAL BIOGEOCHEM. CY. 29(2): 138–159, 153–154 (“Simulations revealed large amounts of iron being deposition in the NWPO and NSCS from both mineral dust and fly ash. The industrial coal burning was the largest source of iron associated with fly ash, among which steel/iron plants were the largest contributors, accounting for more than 50% and 60% of the fly ash iron deposited in the NWPO and NSCS, respectively. Iron from mineral dust remains the main source of iron over the NSCS and NWPO, contributing 85% and 92% of the total iron input, respectively. However, when taking into account the high solubility of combustion-related iron, the proportion of bioavailable iron from anthropogenic sources may well exceed that from natural sources.”). See also Sedwick P. N., Sholkovitz E. R., & Church T. M. (2007) *Impact of anthropogenic combustion emissions on the fractional solubility of aerosol iron: Evidence from the Sargasso Sea*, GEOCHEM. GEOPHYS. GEOSYST. 8(10): 1–21, 17 (“In terms of understanding the atmospheric input of biologically available iron to the surface ocean, the most significant finding of our study is a strong inverse relationship between the operational solubility of iron and the total atmospheric concentration of iron in aerosols over the Sargasso Sea. This relationship appears to reflect source-dependent differences in the solubility characteristics of aerosols carried to the Sargasso Sea, whereby the relative solubility of iron is elevated in aerosols borne by North American air masses, relative to Saharan dust. . . . On the basis of compositional trends observed in historical data for aerosols collected on Bermuda and in our aerosol data from the Sargasso Sea, we infer that the elevated solubility of iron in aerosols associated with North American air masses reflects the presence of anthropogenic fuel-combustion products, which contain iron that is highly soluble relative to Saharan dust. We thus propose that the source-dependent composition of aerosol particles, specifically, the relative proportion of anthropogenic combustion aerosols, is a primary determinant for the fractional solubility of aerosol iron over the Sargasso Sea. This hypothesis implies that anthropogenic combustion emissions may play a major role in determining the fractional solubility of iron in aerosols entering the surface ocean.”).

¹⁸ Ito A., Ye Y., Baldo C., & Shi Z. (2021) *Ocean fertilization by pyrogenic aerosol iron*, NPJ CLIM. ATMOS. SCI. 4(30): 1–20, 15 (“Pyrogenic aerosol potentially contributes to ocean fertilization by supplying DFe, especially in the HNLC regions where even a small addition of Fe can trigger large phytoplankton blooms. Most previous studies, however, focused on mineral dust and neglected pyrogenic aerosol since the total Fe supply by pyrogenic aerosol is significantly lower than by other aerosol sources. In the last decade, there is increasing observational evidence on the presence of pyrogenic sources to atmospheric DFe supply in the remote marine atmosphere and the dominant role of atmospheric processing (such as acid dissolution) in producing secondary DFe. In addition, more atmospheric models calculated deposition fluxes of DFe from pyrogenic sources, and the number of ocean biogeochemistry models considering pyrogenic DFe as a source of micronutrient is growing. Our understanding on pyrogenic Fe aerosols including their emission, chemical processing, deposition, and impact on ocean biogeochemistry (Fig. 1) has improved substantially in the past few years. However, there are still major challenges that hamper our ability to more quantitatively understand the highly variable pyrogenic Fe sources, the complex aerosol and seawater chemistry, and diverse marine responses to this Fe supply, and to predict their changes under ongoing climate change.”). See also Schoffman H., Lis H., Shaked Y., & Keren N. (2016) *Iron-Nutrient Interactions within Phytoplankton*, FRONT. PLANT SCI. 7(1223): 1–12, 7 (“Iron plays a central role in energy production and biochemical catalysis within phytoplankton cells, being involved in multiple metabolic pathways either directly, via its catalytic role in enzymes, or indirectly via its part in the production of energy rich molecules such as NADPH and ATP. Fe thus influences the requirements, acquisition and utilization of essential resources within phytoplankton cells. These same resources, in turn, modulate iron requirements, acquisition and use efficiency (summarized in Figure 3 and Table 1). Considering the core functions iron fulfills in cell metabolism and its relative scarcity in aquatic environments, it is little wonder that the availability of this trace metal exerts strong selective pressure on phytoplankton. Under iron-limiting conditions, phytoplankton must find a way to increase cellular iron on the one hand and decrease their Fe demands on the other.”); Bristow C. S., Hudson-Edwards K. A., & Chappell A. (2010) *Fertilizing the Amazon and equatorial Atlantic with West African dust*, GEOPHYS. RES. LETT. 37(14): L14807, 1–5, 1 (“Dust eroded from the Bodélé is blown across the Atlantic Ocean towards South America. The mineral dust contains micronutrients such as Fe and P that have the potential to act as a fertilizer, increasing primary productivity in the Amazon rain forest as well as the equatorial Atlantic Ocean, and thus leading to N₂ fixation and CO₂ drawdown. We present the results of chemical analysis of 28 dust samples collected from the source area, which indicate that up to 6.5 Tg of Fe and 0.12 Tg of P are exported from the Bodélé Depression

every year.”); and Street J. H. & Paytan A. (2005) *Iron, phytoplankton growth, and the carbon cycle*, MET. IONS BIOL. SYST. 43: 153–193, 153 (“Iron is an essential nutrient for all living organisms. Iron is required for the synthesis of chlorophyll and of several photosynthetic electron transport proteins and for the reduction of CO₂, SO₄(²⁻), and NO₃(⁻) during the photosynthetic production of organic compounds. Iron concentrations in vast areas of the ocean are very low (<1 nM) due to the low solubility of iron in oxic seawater. Low iron concentrations have been shown to limit primary production rates, biomass accumulation, and ecosystem structure in a variety of open-ocean environments, including the equatorial Pacific, the subarctic Pacific and the Southern Ocean and even in some coastal areas. Oceanic primary production, the transfer of carbon dioxide into organic carbon by photosynthetic plankton (phytoplankton), is one process by which atmospheric CO₂ can be transferred to the deep ocean and sequestered for long periods of time. Accordingly, iron limitation of primary producers likely plays a major role in the global carbon cycle.”).

¹⁹ de Richter R., *et al.* (11 September 2019) *Iron Salt Aerosol a natural method to remove methane & other greenhouse gases*, Institution of Mechanical Engineers Presentation, 21–24 (“Several other possible localized ISA applications / Underground coal mine aeration-ventilation systems / Tropical hydroelectric dams / Greenhouses dedicated to sludge drying / Livestock farms venting systems / Leakage at hydraulic fracturing sites to extract shale gas / Destabilization of oceanic methane-hydrates / Siberia and Northern Canada, if permafrost melts and massive CH₄ releases occur / Leakage of gas reservoirs / Open pit coal mines / Etc.”). *See also* Ito A. (2013) *Global modeling study of potentially bioavailable iron input from shipboard aerosol sources to the ocean*, GLOBAL BIOGEOCHEM. CY. 27: 1–10, 8–9 (“The model results suggest that highly soluble iron-containing aerosols from shipping may contribute about 40% of the SFe deposition into HNLC regions over the northeastern Pacific in summer, when the sporadic emission from Asian dust is low. The results presented in this article could have important implications for the parameterization of iron dissolution and highlight the necessity to improve the source chemical composition. . . . The anthropogenic emission changes from 2000 to 2100 (RCP4.5) were applied to the global chemical transport model to examine the relative importance of shipping sources of SFe, compared with those from the other sources. The simulation results suggest that the combustion sources from ships in 2100 may contribute 30–60% of the SFe deposition in the high-latitude North Atlantic and North Pacific. The SFe from shipboard aerosol sources is directly emitted over the surface ocean and mostly supplied to the open ocean, such as the northeastern Pacific. In contrast, the majority of the crust-like aerosols, such as dust and coal fly ash, are deposited near the source regions, such as the East China Sea, because of faster removal for larger particles. Given the continuing growth in global shipping, it may be important to evaluate the role of iron from shipping in regulating marine biogeochemical processes.”).

²⁰ Hanson R. S. & Hanson T. E. (1996) *Methanotrophic bacteria*, MICROBIOL. REV. 60(2): 439–471, 463 (“We have seen that methanotrophs are ubiquitously distributed and play a significant role in the global methane budget and therefore in moderating the impact of methane on global warming. They oxidize most of the methane produced in anaerobic environments before it reaches the atmosphere, and in unsaturated soils they oxidize significant amounts of atmospheric methane. Increased rates of methane oxidation and decreases in methane flux may occur as northern latitudes dry during global warming. Therefore, organic carbon would be mineralized and the impact of methane on global warming would be reduced. However, it is extremely difficult to predict climatic changes and the nature of the habitats that will exist in the future given our current state of knowledge.”). *See also* Einsiedl F., Wunderlich A., Sebilo M., Coskun Ö. K., Orsi W. D., & Mayer B. (2020) *Biogeochemical evidence of anaerobic methane oxidation and anaerobic ammonium oxidation in a stratified lake using stable isotopes*, BIOGEOSCI. 17(20): 5149–5161, 5149 (“At a water depth from 12 to 20 m, a methane–nitrate transition zone (NMTZ) was observed, where $\delta^{13}\text{C}$ values of methane and $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ of dissolved nitrate markedly increased in concert with decreasing concentrations of methane and nitrate. These data patterns, together with the results of a simple 1-D diffusion model linked with a degradation term, show that the nonlinear methane concentration profile cannot be explained by diffusion and that microbial oxidation of methane coupled with denitrification under anaerobic conditions is the most parsimonious explanation for these data trends. In the methane zone at the bottom of the NMTZ (20 to 22 m) $\delta^{15}\text{N}$ of ammonium increased by 4 ‰, while ammonium concentrations decreased. In addition, a strong ^{15}N enrichment of dissolved nitrate was observed at a water depth of 20 m, suggesting that anammox is occurring together with denitrification.”).

²¹ Yoon S., Carey J. N., & Semrau J. D. (2009) *Feasibility of atmospheric methane removal using methanotrophic biotrickling filters*, APPL. MICROBIOL. BIOTECHNOL. 83: 949–956, 949 (“Here, we describe the modeling of a biotrickling filtration system composed of methane-consuming bacteria, i.e., methanotrophs, to assess the utility of these systems in removing methane from the atmosphere. Model results indicate that assuming the global average atmospheric concentration of methane, 1.7 ppmv, methane removal is ineffective using these methanotrophic biofilters as the methane concentration is too low to enable cell survival. If the concentration is increased to 500–6,000 ppmv,

however, similar to that found above landfills and in concentrated animal feeding operations (factory farms), 4.98–35.7 tons of methane can be removed per biofilter per year assuming biotrickling filters of typical size (3.66 m in diameter and 11.5 m in height). . . . The use of methanotrophic biofilters for controlling methane emissions is technically feasible and, provided that either the costs of biofilter construction and operation are reduced or the value of CO₂ credits is increased, can also be economically attractive.”). See also Sly L. I., Bryant L. J., Cox J. M., & Anderson J. M. (1993) *Development of a biofilter for the removal of methane from coal mine ventilation atmospheres*, APPL. MICROBIOL. BIOTECHNOL. 39: 400–404, 400 (“The experimental biofilter utilising a biofilm of *M. fodinarum* was shown to reduce methane levels substantially provided the residence times were sufficiently long. In the range 0.25–1.0% methane in air, commonly experienced in coal mine atmospheres, more than 70% of the methane was removed with a residence time of 15 min, with a 90% reduction at 20 min. Even at a residence time of 5 min approximately 20% of the methane in air was removed. Equal quantities of O₂ are consumed during the bacterial oxidation of methane and 1% methane is converted to 0.7% CO₂. Scale-up and alternative biofilter packings are likely to reduce the residence times in the biofilter.”).

²² Thornton B. F. & Crill P. (2015) *Microbial lid on subsea methane*, NAT. CLIM. CHANGE 5: 723–724, 723 (“Submarine permafrost thaw in the Arctic has been suggested as a trigger for the release of large quantities of methane to the water column, and subsequently the atmosphere—with important implications for global warming. Now research shows that microbial oxidation of methane at the thaw front can effectively prevent its release.”).

²³ Kebreab E. & Feng X. (2021) *Strategies to Reduce Methane Emissions from Enteric and Lagoon Sources*, Prepared for State of California Air Resources Board, 69 (“In general, higher moisture contents in raw composting manure could enhance the CH₄ mitigation rates, however, the pH, and C/N content were not linearly related to CH₄ mitigation. Adding biochar, acids, and straw to manure could mitigate CH₄ emissions by 82.4%, 78.1%, and 47.7%, respectively. However, the data for straw is quite small so it should not be taken out of context as it may introduce a source of carbon into lagoons. The meta-analysis conducted with selected additives indicated manure additives were an effective method to reduce CH₄ emission, with biochar being the most effective. However, further studies of manure additives on CH₄ mitigation are required to support a more accurate quantitative analysis and potential impacts to water quality and crop yield after land application. Most of the research for biochar and straw is when used as additive to solid or semi solid manure so they should be interpreted in that context.”). See also Peterson C., El Mashad H. M., Zhao Y., Pan Y., & Mitloehner F. M. (2020) *Effects of SOP Lagoon Additive on Gaseous Emissions from Stored Liquid Dairy Manure*, SUSTAINABILITY 12(1393): 1–17, 14 (“A variety of additives have been applied to reduce emissions from manure. Although the composition and mechanism of the emission reduction of several additives are known, information on many other commercial additives is not available because of confidentiality and limits in the marketing literature. Calcium sulfate (gypsum) can be found abundantly in nature and has been used to improve soil properties. . . . Different forms of gypsum have been tested for the mitigation of GHG and ammonia emissions from livestock effluents. The results have had varying results: while some studies reported a decrease in ammonia emissions after the addition of gypsum, not all have demonstrated the efficacy of gypsum in reducing the release of GHGs. Many of the results were obtained using a considerable amount of material (3% to 10% of manure wet weight) making the application not practical in real-world conditions. Borgonovo et al. first published results on this specific commercial additive (SOP LAGOON), made of gypsum processed with proprietary technology, and found that the addition of the products to fresh liquid manure has a reduction potential of 21.5% of CH₄, 22.9% of CO₂, 100% of N₂O and 100% of NH₃ emissions on day 4, even at very low dosages. It should be mentioned that similar to other commercial additives, the exact manufacturing process of SOP Lagoon is unknown due to confidentiality.”); and Yang S., Xiao Y., Sun X., Ding J., Jiang Z., & Xu J. (2019) *Biochar improved rice yield and mitigated CH₄ and N₂O emissions from paddy field under controlled irrigation in the Taihu Lake Region of China*, ATMOS. ENVIRON. 200: 69–77, 69 (“These results suggest that 20 and 40 t ha⁻¹ biochar can be utilized under controlled irrigation not only for mitigation of CH₄ and N₂O emission but also to increase rice yield, soil fertility and irrigation water productivity. Therefore, the combination of biochar amendment and controlled irrigation might be a good option for mitigating greenhouse gases emission and realizing the sustainable utilization of soil and water resources of paddy fields in the Taihu Lake Region of China.”).

²⁴ Jackson, R. B., et al. (2021) *Atmospheric methane removal: a research agenda*, PHILOSOPHICAL TRANSACTIONS ROYAL SOCIETY A 379:1–17, 10 (“We recommend a Methane Removal Model Intercomparison Project to provide structure for a multimodel analysis. Similar to and inspired by previous analyses and intercomparisons for CO₂ (e.g. [28,103]), a full investigation for methane removal is needed to examine: (1) Scenarios of different timing and amounts of methane removal; (2) Comparisons of the climate impacts and Earth-system feedbacks of methane

removal in different atmospheric and climate scenarios (e.g. low- and high-emission); (3) Spatially explicit simulations of methane removal at prescribed locations and latitudes (requiring models to have an ‘emissions-driven’ methane capability); (4) Studies of how methane’s relatively short lifetime, in conjunction with climate feedbacks on natural methane emissions, influences metrics of cumulative methane removal; (5) Feedbacks with air quality, including tropospheric ozone (O₃) concentrations, through OH chemistry and/or secondary aerosol formation; (6) Interactions of methane removal with other mitigation and CDR approaches.”).

²⁵ Jackson R. B., *et al.* (2021) *Atmospheric methane removal: a research agenda*, PHIL. TRANS. R. SOC. A 379: 1–17, 10 (“We recommend a Methane Removal Model Intercomparison Project to provide structure for a multimodel analysis. Similar to and inspired by previous analyses and intercomparisons for CO₂ (e.g. [28,103]), a full investigation for methane removal is needed to examine: (1) Scenarios of different timing and amounts of methane removal; (2) Comparisons of the climate impacts and Earth-system feedbacks of methane removal in different atmospheric and climate scenarios (e.g. low- and high-emission); (3) Spatially explicit simulations of methane removal at prescribed locations and latitudes (requiring models to have an ‘emissions-driven’ methane capability); (4) Studies of how methane’s relatively short lifetime, in conjunction with climate feedbacks on natural methane emissions, influences metrics of cumulative methane removal; (5) Feedbacks with air quality, including tropospheric ozone (O₃) concentrations, through OH chemistry and/or secondary aerosol formation; (6) Interactions of methane removal with other mitigation and CDR approaches.”).

²⁶ Jackson, R. B., *et al.* (2021) *Atmospheric methane removal: a research agenda*, PHILOSOPHICAL TRANSACTIONS ROYAL SOCIETY A 379:1–17, 12 (“For many reasons, then, we believe a systematic research program for methane removal is needed today that includes experiments, technology development and modelling. The Methane Removal Model Intercomparison Project proposed here can quantify the global and local impacts of methane removal, and allow for comparisons with carbon dioxide removal, potentially informing policy decisions. Testing of various methane removal technologies and validation at scale will clarify which approaches are most effective, acknowledging the priority of emissions reductions for methane and carbon dioxide.”).