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Removal of non-CO_2 greenhouse gases by large-scale atmospheric solar photocatalysis

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A B S T R A C T

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 CO_2. Photocatalysis of methane oxidizes it to CO_2, effectively reducing its global warming potential (GWP) by at least 90%. Nitrous oxide can be reduced to nitrogen and oxygen by photocatalysis; meanwhile halocarbons can be mineralized by red-ox photocatalytic reactions to acid halides and CO_2. Photocatalysis avoids the need for capture and sequestration of these atmospheric components. Here review an unusual hybrid device combining photocatalysis with carbon-free electricity with no-intermittency based on the solar updraft chimney. Then we review experimental evidence regarding photocatalytic transformations of non-CO_2 GHGs. We propose to combine TiO_2-photocatalysis with solar chimney power plants (SCPPs) to cleanse the atmosphere of non-CO_2 GHGs. Worldwide installation of 50,000 SCPPs, each of capacity 200 MW, would generate a cumulative 34 PWh of renewable electricity by 2050, taking into account construction time. These SCPPs equipped with photocatalyst would process 1 atmospheric volume each 14–16 years, reducing or stopping the atmospheric growth rate of the non-CO_2 GHGs and progressively reducing their atmospheric concentrations. Removal of methane, as compared to other GHGs, has enhanced efficacy in reducing radiative forcing because it liberates more ◦OH radicals to accelerate the cleaning of the troposphere. The overall reduction in non-CO_2 GHG concentration would help to limit global temperature rise. By physically linking greenhouse gas removal to renewable electricity generation, the hybrid concept would avoid the moral hazard associated with most other climate engineering proposals.

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Abbreviations: Bioenergy with carbon sequestration and storage, BECCS; Carbon sequestration and storage, CCS; Carbon dioxide removal, CDR; Methane, CH_4; Chloro fluoro carbons, CFCs; Carbon dioxide, CO_2; Giant photocatalytic reactor, GCR; Greenhouse, GH; Greenhouse gas, GHG; Greenhouse gas removal, GHGR; Global warming potential, GWP; Hydro chloro fluoro carbons and hydro fluoro carbons, HFCs; Intergovernmental Panel on Climate Change, IPCC; Metal organic framework, MOF; Nitrous oxide, N_2O; Parts per billion, ppb; Parts per million, ppm; Photocatalytic reactor, PCR; Relative humidity, RH; Solar chimney power plant, SCPP; Solar radiation management, SRM; Ultraviolet, UV; Volatile organic compound, VOC

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1. Introduction

1.1. The need for hybrid renewable energy and greenhouse gas reduction technologies

Even if humans stop combusting fossil fuels and discharging CO_2 into the atmosphere, the average global temperature of the Earth will continue to increase for the rest of the century –for several reasons. Firstly, the long lifetime of CO_2 (estimated in the 100–300 year range [1]) means that the excess atmospheric stocks (515 Gt Carbon) would continue to drive radiative forcing and global warming for many decades [2]. Secondly, even if atmospheric concentrations were to decrease, CO_2 would outgas from the oceans and offset this decrease, because of the dynamic equilibrium between the CO_2 in the atmosphere and the (bi)carbonates HCO_3^-/CO_3^{2-} dissolved in the oceans [3].

Thirdly, there is the contribution of other GHGs, besides CO_2, which together account for about 34% of radiative forcing [4]. Even if all excess anthropogenic atmospheric CO_2 were removed, radiative forcing would only be reduced by half [5].

The insufficiency of mitigation measures based just on fossil fuel replacement has prompted research in carbon capture and sequestration (CCS), greenhouse gas removal (GHGR), and solar radiation management (also called solar reflection methods: SRM). But each of these approaches suffers drawbacks. CCS [6,7] may restrict release of CO_2 from stationary fossil-fuel power plants, but not from vehicles. It also carries significant water and energy penalties and a number of environmental risks associated with transport and storage of the CO_2 [7,8].

In order to stabilize the Earth surface temperature alternative processes are needed to decrease the level of atmospheric GHGs. Of the over 100 climate modeling simulations consistent with the Paris Agreement goals, as analyzed by the IPCC scientists, 87% involve net-negative emissions [9] by the end of the century: this can only be accomplished through the large-scale deployment of GHGR solutions.

According to the United Nations Environment Program [10], as shown in Fig. 1, the CO_2 emissions will have to be stopped by mid-century and by the second half of the century, GHGs will have to be removed from the atmosphere and among them non-CO_2 GHGs which have a significant contribution to the global anthropogenic radiative forcing (nearly 1/3rd).

Among GHGR technologies, those most frequently discussed target CO_2. Carbon dioxide removal (CDR) technologies include afforestation, reforestation, biochar [11,12], bioenergy with carbon capture and sequestration (BECCS) [13,14], direct air capture (removing the CO_2 from ambient air rather than from a smoke stack) [15], accelerated mineral weathering [16], ocean iron fertilization and several others [17]. Each incurs significant penalties with regard to land, energy and water usage [18]. BECCS offers the benefit of net energy production, but requires very large land usage which would impact on world food production [19].

SRM technologies include atmospheric aerosol injection using sulphur dioxide or other agents to induce negative radiative forcing by reflecting back to space incoming sunlight. Such interventions raise perplexing issues of international governance and security, prompting discussions about militarization, vulnerability to sabotage, and the risks of sudden termination causing catastrophic warming should the technology fail for any reason [20]. Over-reliance on SRM and CDR generally, and fear of resulting complacency as regards emissions reduction, has been referred to as *moral hazard* in debates about both these climate engineering measures.

The moral hazard can be avoided, however, if GHGR is physically linked to emissions reduction, thus making sure that the former does not neglect the latter. While BECCS achieves such linkage, another approach so far less discussed and evaluated is to hybridize Solar Chimney Power Plants (SCPPs) with Photocatalytic Reactors (PCRs) – see Fig. 2 and chapter 2. The idea of hybrid SCPP-PCR was introduced at the recent Oxford (UK) GHGR conference [21] and earlier at a SCPP conference in Bochum (DE) [22]. Like BECCS, SCPP-PCR would provide renewable electricity while removing GHGs; but

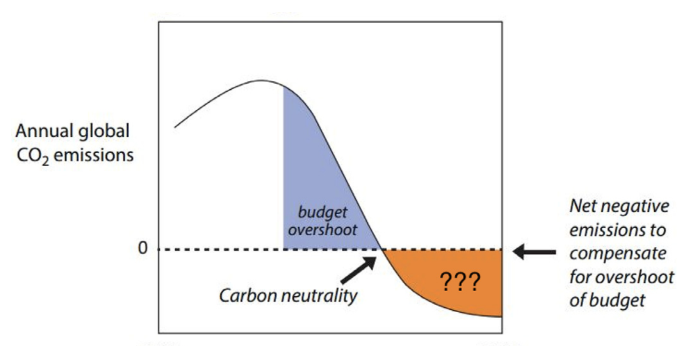

Fig. 1. CO_2 emissions pathway [10].