The Certainty of Uncertainty in Atmospheric CO_2 Removal: A Crisis-Response Abrupt Mitigation Scenario (CRAMS)

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Abstract

The gap between current emission trend and the expected 1.5 °C warming target forces the deployment of different carbon dioxide removal technologies (CDRs). Even though large discrepancies and uncertainties presents in studies investigating the CDR potentials, costs and side effects of bio-energy with carbon capture and storage (BECCS), direct air capture with CO_2 storage (DACCS) and enhanced weathering (EW), determining the appropriate actions to take in light of these uncertainties represents a core challenge of current research. Herein, under the proposed CRAMS, we estimated the CO_2 removal target to hold warming below 1.5 °C and re-calibrated the CO_2 removal potentials of CDRs against experimental data. The quantitative evaluation of both energetic and temporal cost provide certainties for CO_2 removal. Our findings suggested that the insufficiency and limitation of BECCS, DACCS and EW in combination call for techno-economic breakthroughs to these CDRs as our only way to reverse the temperature overshoot back to 1.5 °C in the future.

Introduction

Projections of future climate change are highly dependent on various different hypothetical scenarios and the conclusions are typically expressed in the form of confidence and certainty. However, such scientifically rigorous means of expression often fails to convey a definite message to non-experts, which has been found to cause confusion, suspicion and even inaction on climate issues for both governments and the public. Thus, how to identify actions that must be done out of the vast uncertainties in climate-related data is a core challenge of current research.

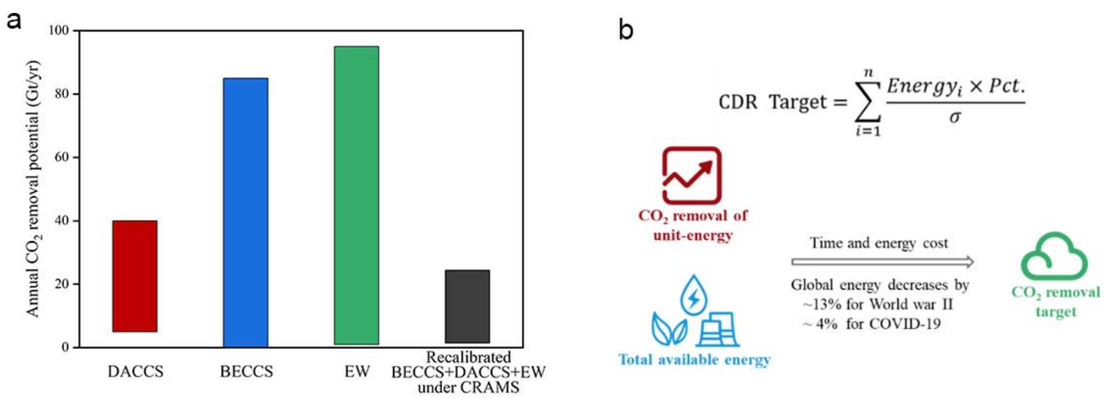

Figure 1 a) CO_2 removal potential reported in literatures and re-calibrated in this study; b) Scheme of the analysis framework for the certainty of uncertainty in atmospheric CO_2 Removal

Carbon Dioxide Removal (CDR) is widely recognized as one of the necessary strategies to achieve carbon neutrality and the Paris Agreement's temperature goals. Intensive research efforts have been devoted to several CDR technologies, including bio-energy with carbon capture and storage (BECCS), direct air capture with CO_2 storage (DACCS) and enhanced weathering (EW). However, significant uncertainties still exist for the carbon removal potential of these technologies. Various different studies suggest that by 2100, the annual carbon removal potential of BECCS, DACCS and EW will fall in a wide range from 0.1 to 85 Gt、 5 to 40 Gt and 1 to 95 Gt[1] (**Figure 1a**), respectively. If estimated with the lower end of these potentials, CDR would only be put into practice when global emissions are close to zero to offset residual unavoidable emissions from hard-to-abate sectors. In sharp contrast, the sum of the upper ends of these potentials is even much larger than current emission rate (*ca.* 40 Gt/yr[2]), which implies ostensibly that continuous emission as usual might be morally acceptable and large-scale mitigation might not even be needed. Such a potential misconception has drawn widespread concerns and criticisms given the increasingly risk with insufficient mitigation efforts. Clearly, identification of clear message in the vast uncertainties of CDR is urgent and important to cause meaningful actions against climate change issues.

Herein, we re-calibrated the CO_2 removal potentials of the three CDRs against experimental data reported in the literature. It was found to be insufficient in terms of energy and time cost when comparing with the required CO_2 removal target to hold warming below 1.5°C under the proposed crisis-response abrupt mitigation scenarios(CRAMS). Our results indicate that advanced CDR technologies with breakthroughs in CDR efficiency are urgently needed if we want preserve the possibility to reverse the temperature overshoot back to 1.5 °C in the future.

Results and Discussions

Estimated Mitigation potentials for the Three CDR Technologies

Experimentally re-calibrated mitigation potentials for the three CDR technologies were then evaluated. The amount of atmospheric CO_2 that can be removed upon consumption of each unit energy is defined as CDR coefficient (σ)[3].

BECCS:

In general, BECCS has a higher σ for electricity generation than for biofuel production since biofuel combustions in end use releases part of the CO_2 back into the air. In previous theoretical estimations, the CDR potential of BECCS is obtained based on 4 key parameters[4-7]: (1) the energy content and (2) carbon content for each type of biomass, (3) carbon-conversion efficiency and (4) CCS penalty.

The energy penalty and CO_2 emission in the biomass supply chain before transformation is often overlooked. Besides, the separate estimation of individual biomass feedstock ignored the mixture of various biomass feedstock in real word bioenergy supply, comprising agriculture residues, energy crops, forestry and organic waste at a global scale. There are large variations in energy and carbon contents, complexity and energy intensity of cultivation, processing and transport for different biomass resources[5, 8]. With relatively high moisture content, fresh woody biomass requires more energy for drying[9]. Even through energy crop are grown with low input requirement and high energetic value, its share in current biomass production with a minor fraction of 0.1%[8] may be attributed to the fear of competition with food production. The avoidance of long-distance transportation is also desirable to cut down the energy and CO_2 penalty for the use of organic waste in BECCS.

Notably, values from 330 g/kWh (92 g/MJ) to 4400 g/kWh (1220 g/MJ) have been previously reported for different types of biomass feedstock used in BECCS[10]. Separate estimation of individual feedstock ignored the mixture property in real word bioenergy supply, comprising agriculture residues, energy crops, forestry and organic waste at a global scale, which would not appropriately reflect the potential of BECCS. There are large variations in energy and carbon contents of biomass and energy intensity of cultivation, processing and transportation in realistic application. Thus, we concluded that these values are not suitable to analyze the overall CDR potential of BECCS. Besides, energy penalty and CO_2 emission in the biomass supply chain before conversion and the carbon-conversion efficiency may also be neglected sometimes[5].

To resolve these issues, we considered the influence of biomass supply chain by recalculating the energy and carbon penalty from the reported energy efficiency and carbon intensity of the bio-power production via various feedstock. As to carbon-conversion efficiency and CCS penalty, the data from the state-of-the-art technologies were also adapted. Analysis of a mix of bioenergy as the energy input is presented in this study. The deployment of BECCS consumes primary modern bioenergy as energy input for transformation and the source of CO_2 to be removed while final energy is also consumed as energy input for auxiliary operations.

Even through energy crop are grown with low input requirement and high energetic value, its share in current biomass production with a minor fraction of 0.1% may be attributed to the fear of competition with food production. The biomass feedstock composition of U.S, which contains nearly zero *ca.* 37% energy crop[11] as economically feasible predictions for 2040, is taken as an optimistic structure of modern bioenergy to calculate overall energy and carbon contents for future global BECCS. With all these considerations, we found that the lower and upper limit of σ for BECCS is 175 and 281 g/kWh (49 and 78 g/MJ, Table 1) considering only bioenergy, respectively. If only considering the final energy consumed, which shares 0.5~29% and 0.7~40% of total energy input, σ increases to 495 and 1675 g/kWh (138 and 465 g/MJ, Table 1), respectively.

	BECCS	DACCS	EW
Literature reported σ:	72-4400	2000-5035	364-3900
g/kWh (g/MJ)	(20-1222)	(556-1399)	(101-1083)
Re-calibrated σ:	175-281	505-805	0-19
g/kWh (g/MJ)	(49-78)	(140-224)	(0-5)

Table 1 Comparison of literature-reported and recalibrated mitigation coefficient σ.

DACCS:

There are generally two types of DAC systems based either on aqueous alkaline solutions with high temperature regeneration or on solid amine-based sorbents with low temperature regeneration. Values ranging from 2000 to 5035 g/kWh (556-1399 g/MJ) have been estimated theoretically for the upper limit of σ for DACCS. In sharp contrast, much lower values were obtained by optimistic linear scale-up of state-of-the-art engineering exercises, in which 505 g/kWh (140 g/MJ[12]) and 805 g/kWh (224 g/MJ[13]) were achieved by using CaO and amine as the sorbent, respectively. The use of thermodynamic minima in certain processes, such as capture, compression and thermal recycling, is the reason why previous theoretical analyses significantly overestimated the upper limit. We reasoned that a combination of both CaO and amine routes or either one of them may be adapted for DACCS in the future. Thus, 505~805 g/kWh (140-224 g/MJ, Table 1) appears to be a reasonable range for σ of DACCS.

The lower bound of our estimation is based on the $Ca(OH)_2/CaCO_3$ process with relatively cheap materials and high engineering readiness. The upper bound was achieved with the employment of expensive heat pumps and capturing 1 ton of CO_2 consumed *ca.* 7 kilograms of costly amine sorbent. As such, 805 g/kWh should be regarded as an optimistic estimation unless revolutionary techno-economic advancements occur in the future.

EW:

Enhanced weathering (EW) of (ultra)mafic rocks is widely considered as a promising option for carbon dioxide removal (CDR). For EW, reactive ultramafic and basaltic rocks are mined, ground and spread onto warm and humid areas in order to accelerate surficial weathering of silicates with dissolved atmospheric CO_2 in precipitation/irrigation via spontaneous acid-base neutralization. Fundamental physico-chemical characteristics for mineral weathering, such as 1) the content of reactive species for carbonation (R_{CO2}) associated with the rock composition and 2) weathering rate (W_r) associated with the rock composition and environmental parameters (temperature, precipitation/humidity and acidity), jointly determine the amount and the speed of CO_2 fixation, *i.e.* energy and time cost on CO_2 fixation.

To satisfy the requirements of environmental parameters, application to croplands has been actively suggested as an efficient route to enhance weathering rates and increase CO_2 drawdown[14-15]. However, the CDR potential of this route is also highly controversial. Accurately measuring its CDR potential remains unavailable due to sluggish weathering process. Previous models have estimated annual CDR potentials ranging from 1 to 95 Gt by 2100, with the maximum significantly exceeding the anthropogenic CO_2 emissions in 2021 (approximately 41 Gt[2]). This raises concerns that a misconception may arise, suggesting active mitigations of CO_2 emissions might not be necessary. We recently addressed this issue by partitioning the CDR potential of EW into two components, flow-through and non-flow-through processes, and developed an experimentally-calibrated model to reduce discrepancies between previous theoretical and experimental weathering rates.

While the shrinking core model (SCM) has been commonly adopted in previous theoretical and experimental studies, there still lacks a comprehensive assessment on the impacts of model parameters, such as rock particle size, size distribution, weathering rate and time length on the weathering kinetics and the resultant CDR potential[16]. We incorporated particle size distribution of rock powder into the surface reaction-controlled SCM, and conducts sensitivity analysis on EW's CDR potential quantitatively. Even fully powered by low-carbon energy in the optimistic case, the application of EW with olivine only achieves maximum CDR per unit of rock and energy consumption of 0.01 kg CO_2 per kg rock and 19 g per kWh at size of 8 and 22 μm respectively, indicating the limitations of EW. The derived optimal application parameters with olivine powers within 3.7–79 μm provide valuable insights into the practical real-world applications to achieve net CO_2 removal.

Our model estimates that the upper bound for the annual CDR potential of EW[17], without consideration of the uncertainties in overall soil carbon balance, should be revised from previously reported 1–95 Gt yr^{-1} by 2100 to 0.22 (±0.16) Gt yr^{-1}. This significant revision is mainly attributed to the observed sluggish weathering rates. Thus, the apparent observation from the analysis is that the pace of enhanced weathering is insufficient to match the competitiveness of other engineered CDR options. The development of ultra-enhanced weathering is crucial to significantly contribute to the substantial removal of CO_2 from the atmosphere.

From the perspective of σ based on total energy consumption, the priority order is as follows: DACCS > BECCS > EW. However, if only final energy is considered, BECCS becomes the most energy efficient way to remove atmospheric CO_2.

The Amount of Atmospheric CO_2 to Be Removed: A Crisis-Response Abrupt Mitigation Scenario (CRAMS)

How much atmospheric CO_2 will need to be removed globally depends on the actual CO_2 emission and mitigation pathways in the future. A realistic answer to such question is clearly crucial for evaluations of various CDR technologies. However, large uncertainty also exists in the literature data. For example, *ca.* 620 Gt and *ca.* 1180 Gt CO_2 need to be removed in SSP2' s 1.5 °C with low overshoot scenario and SSP5' s below 1.5 °C scenario, respectively, in IPCC AR6[18]. Therefore, a relatively definite estimation is urgently required. It's reasonable to expect that no abrupt changes in future global CO_2 emission scenarios before some kind of sociopolitical tipping points are reached (**Figure 2**). In retrospect, historic declinations of global CO_2 emission generally occurred in response to various major crises. Taking the past as a guide to the future, a crisis-response abrupt mitigation scenario (CRAMS) is developed below.

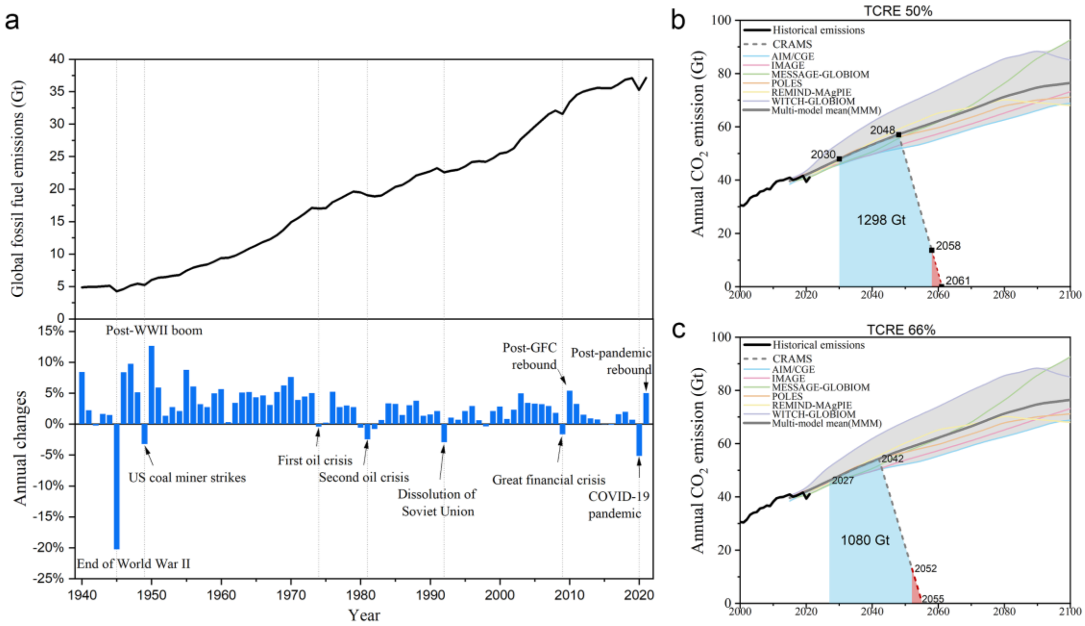

Figure 2. a) Historical global fossil CO_2 emissions and annual change[19], b, c) Emission pathways under CRAMS

CRAMS consists of a short-term business-as-usual emission phase followed by an abrupt mitigation phase. Ideally, the tipping point should emerge in the coming *ca.* 7 years before running out the remaining carbon budget for 1.5 °C (66% probability, transient climate response to cumulative carbon emissions, TCRE[20]). A further delay of 3~4 years leaves with half of the possibility. However, since it's unrealistic to expect drastic behavioral changes would occur, the tipping point is adapted here as the CO_2 emission peak when exhausting the 2°C carbon budget (66% probability, TCRE). We also deducted the extreme case with 50% of TCRE with more risk and less chance. The disastrous impacts of 2°C warming are assumed to be sufficient to force the entire world to take meaningful actions to remove the cumulative amount of CO_2 exceeding the threshold from air and reverse the CO_2 level back to the consensus target level of 1.5 °C. Global-wise mitigation at an annual reduction rate up to 7.6%, a very radical yet potentially affordable rate proposed by United Nations Environment Programme[21], is also adapted in the current analysis. When mitigation to the level that hard-to-abate residual emissions finally requires the deployment of CDR, CDR can work as a complement to reach zero emission. These unavoidable emissions in hard-to-abate sectors accounts for about 32% of the total energy emissions[22].

We reasoned that the amount of atmospheric CO_2 to be removed in reality should be higher than the amount given by CRAMS based on the following three reasons: 1) it should be noted that such a scenario is highly optimistic and temperature rise may very likely turn out to be larger than 2°C given the ongoing emission trend[3]; 2) a 7.6% annual emission reduction rate is rather optimistic compared with *ca.* a 3.3% annual reduction by EU from 1990 to 2020[23] and even the most ambitious national climate action plans in the real world; and 3) the remaining carbon budget with a 66% probability also forces earlier actions compared with that in lower percentile range.

In order to obtain the "crisis-response point", multi-model means (MMMs)[24-26] of various previous projection models were next deducted under current policies for short-term business-as-usual emission. MMMs shows that the peak of CRAMS and 2°C -exceeding year will arrive in *ca.* 2042 (66% probability, TCRE; 2048, 50% probability, TCRE; 2040~2043 by six projection models individually), while the time of CDR deployment turns out to be 2052 (66% probability, TCRE; 2058, 50% probability, TCRE).

If leaving the actual emission trajectory from the deployment of CDR out of account, mitigation to zero emission provides a direct estimation of the lower boundary for the amount of atmospheric CO_2 to be removed. In order to reverse back to the 1.5 °C target, c*a.* 1080 Gt (66% probability, TCRE; 1298 Gt, 50% probability) need to be drawdown. Notably, the delay of peaking results in more CO_2 to be removed.

It should be noted that AR6 finds that in the modest-mitigation SSP2-4.5 scenario the world is likely to exceed 2°C around 2052, with a range of 2037 to 2084[18]. For the high emissions SSP3-7.0 scenario, the world is likely to pass 2°C around 2046 (with a range of 2035-2062), while in the fossil-fuel-based SSP5-8.5 scenario it is 2041 (with a range of 2032 to 2053). However, regardless of the different years to pass 2°C, the amount of 1.5-to-2°C over emission should remain identical among various scenarios as long as a 66% probability of TCRE is adapted, although uncertainties regarding the carbon cycle, climate sensitivity, and the rate of change would influence the effect of cooling responding to CO_2 removal.

Projected Scale of Global Energy Available for the Three CDR Technologies

The global scale of specific energy available to be consumed for each CDR process is clearly a fundamental metric to determine the amount of CO_2 that can be removed from air by the three carbon-capture technologies. Statistical data of regional and global energy balance are typically categorized in a fixed framework consisting of various energy forms, including primary energy, secondary energy and final energy[27]. Notably, final energy, referring to those consumed by the end-user for energy services in different sectors, include both primary and secondary energies. The deployment of BECCS consumes both primary modern bioenergy and final energy as the source of CO_2 to be removed and the energy into for transformation and auxiliary operations. In contrast, DACCS and EW may consume a mixture of primary and secondary energy inputs that cannot be gauged solely by either global primary energy statistics or global secondary energy statistics. Thus, we reasoned that the CDR potentials of both DACCS and EW should be best evaluated based on the projections of total final consumption of energy (TFC) among all energy forms. It is worth noting that the total potential of these three CDR is limited by the overall availability of TFC.

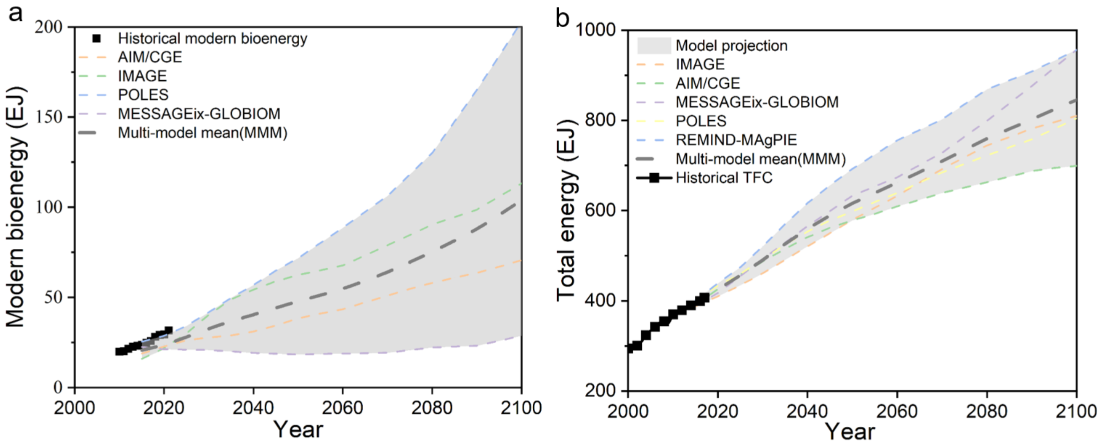

Figure 3. Projected modern bioenergy and total final energy consumption within 21 century

Subsequently, we calculated the Multi-Model Means (MMMs) of multiple previous projection models[26] for both global modern bioenergy and TFC (**Figure 3**), which reasonably comprise regional competitions and collaborations. The total global modern bioenergy and TFC were reported as 30.4 EJ (46% of total bioenergy) in 2017[28] and 416 EJ in 2018[27], respectively. Our MMMs models indicate that modern bioenergy and TFC will continuously expand to 57.3 EJ and 615 EJ, respectively, in 2050 as well as 129EJ and 845EJ, respectively, in 2100. The average annual growth rate is 2.4% for modern bioenergy and 1.3% for TFC which are comparable with the numbers reported in previous Current/Stated Policies Scenarios by International Energy Agency(IEA)[29] and International Renewable Energy Agency[30].

It should be noted that less energy consumptions have been envisioned in scenarios with more sustainable energy intensity in continuous economic growth. Thus, MMMs appears to be a good estimation for the upper limit of the amount of TFC available for CDR.

Energy and Time Costs for CO_2 Removal

Apart from the scale of global energy available, the time needed to fix certain amount of CO_2 also set limit to their feasibility of large-scale application. Therefore, a quantitative evaluation of both energetic and temporal cost for CO_2 removal can prioritize the three CDR technologies when planning on a multi-pronged mitigation portfolio. Considering the development of technical and engineering feasibility and public acceptance, we set different time schedule for the gradual upscaling of CDR from the starting year of deployment defined above.

According to our model, if all available of modern bioenergy is used for CDR, the annual CDR potential of BECCS reaches 5.2 to 8.3 Gt/yr in 2100 (**Figure 4a, c**). If starting from 2052 and achieving the energy-allowed maximum potential with only ten years, the upper bound of the cumulative CO_2 removal potential reaches 291 Gt in 2100 (**Figure 4b**). Early deployment and quick upscaling contribute to larger cumulative removal. Conversely, there is few difference in the average annual potential starting from different year. Compared with IPCC's modelled pathways limiting global warming to 1.5°C with limited or no overshoot, BECCS deployment is projected to range from 0–8 in 2050 and 0–16 $GtCO_2$ yr^{-1} in 2100, respectively[20]. IPCC's upper ends exceed the projected total available modern bioenergy indicates the risks and uncertainties of previous models. To put these numbers in a practical context, it has been estimated that a 40 Gt CO_2 yr^{-1} by BECCS requires 44.3 million square kilometers (Mkm^2), which is larger than the sum of land areas of Russia, European Union, United States and China[31]. In IPCC's AR6, among scenarios likely limiting warming to 2°C or lower, the cumulative contribution of BECCS reaches a median value of 328 Gt CO_2, even larger than the optimistic upper bound of our analysis, implying the significant reliance on BECCS to achieve the temperature goal.

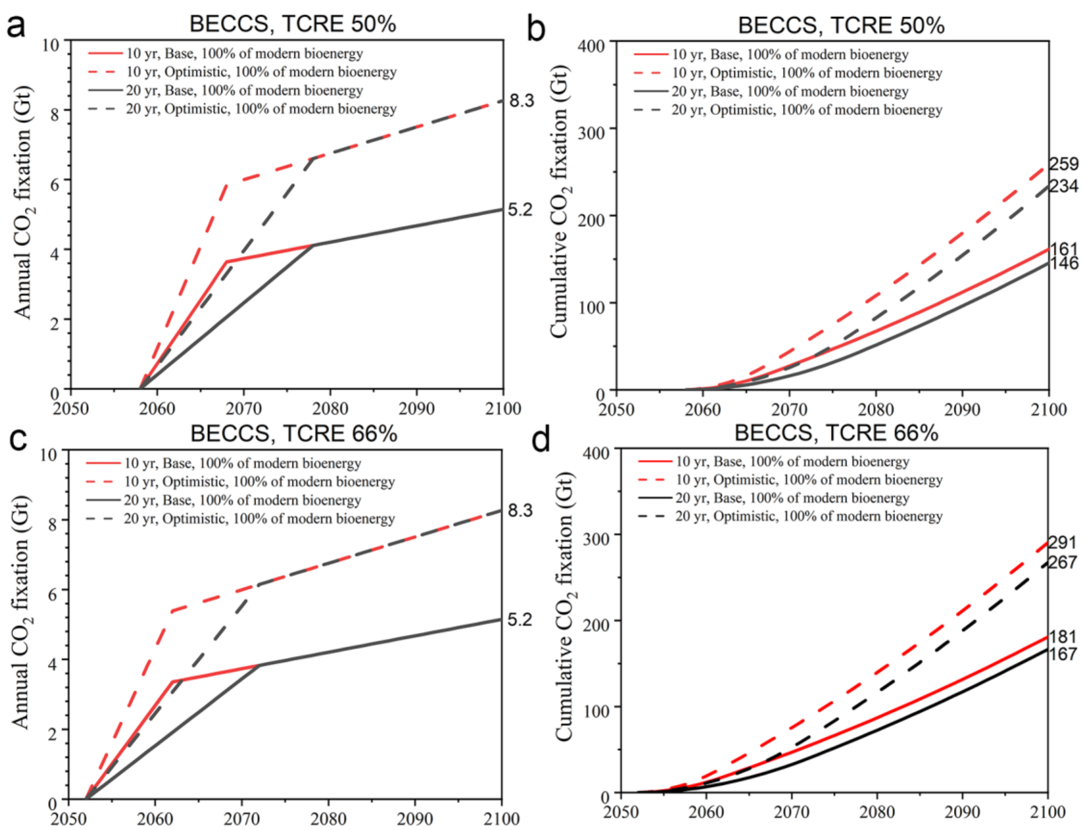

Figure 4 Annual and cumulative CO_2 fixation of BECCS limited by global modern bioenergy. a) Annual and b) cumulative CO_2 fixation with 50% of TCRE, c) annual and d) cumulative CO_2 fixation with 66% of TCRE

Our analysis shows that the deployment of BECCS only could not remove the overall beyond-budget emissions even with 100% of modern bioenergy input in the optimistic case (**Figure 4a, b**), which implies a long-lasting unpleasant environments with extreme warming. Limited scale of modern biomass renders BECCS alone incapable of the removal target.

Switching from BECCS to DACCS or EW, the available energy expands by one magnitude from modern bioenergy to TFC (**Figure 3**). However, it's impossible to use all TFC in atmospheric CO_2 removal. Notably, approximate 13%[19] and 4%[19] (4.2% from BP[32]) decline of annual global energy demand have come with profound sociopolitical and economic devastation in World War II and the outbreak of Covid-19 pandemics, respectively. Accordingly, we defined two levels of energetic cost in CDR, which is 13% and 4% TFC at parity of "world wartime costs" and "against pandemic costs", respectively.

Our analysis shows that the average annual CDR potential of DACCS range from 3.3 to 19.4 Gt/yr with energetic cost between "against pandemic costs" and "world wartime costs". The cumulative CDR potential expands to about four times larger than BECCS. However, even "world wartime costs" of 13% energy input only removes 835-951 Gt atmospheric CO_2 by 2100, which is still *ca.* 463~129 Gt less than the CO_2 removal target required by CRAMS (**Figure 5**).

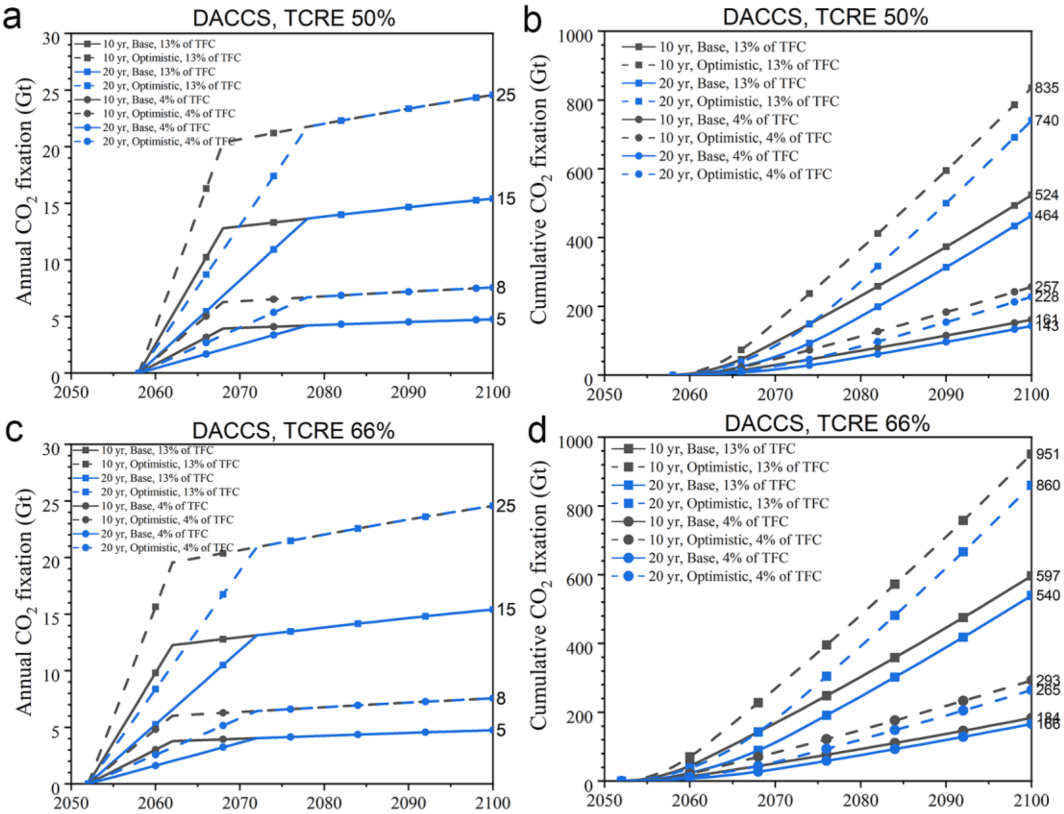

Figure 5 Annual and cumulative CO_2 fixation of BECCS limited by total final energy consumption. a) Annual and b) cumulative CO_2 fixation with 50% of TCRE, c) annual and d) cumulative CO_2 fixation with 66% of TCRE.

Finally, a combination of BECCS, DACCS and EW with our calibrated CDR potentials was analyzed. With 100% modern bioenergy input, the share of TFC consumed by BECCS was subtracted from the confined amount of TFC, the left of which is used to calculate the share of DACCS. However, the combination of three CDRs in the optimistic scenario remains below the removal target at the end of this century (**Figure 6**). Even though in the case of 66% percentile of TCRE, the overall removal of 1076 Gt is close to 1080 Gt removal target, an early emission peaking and fast upscaling of CDR is required. The overall potential is limited by the available energy and insufficient-enhanced weathering rate.

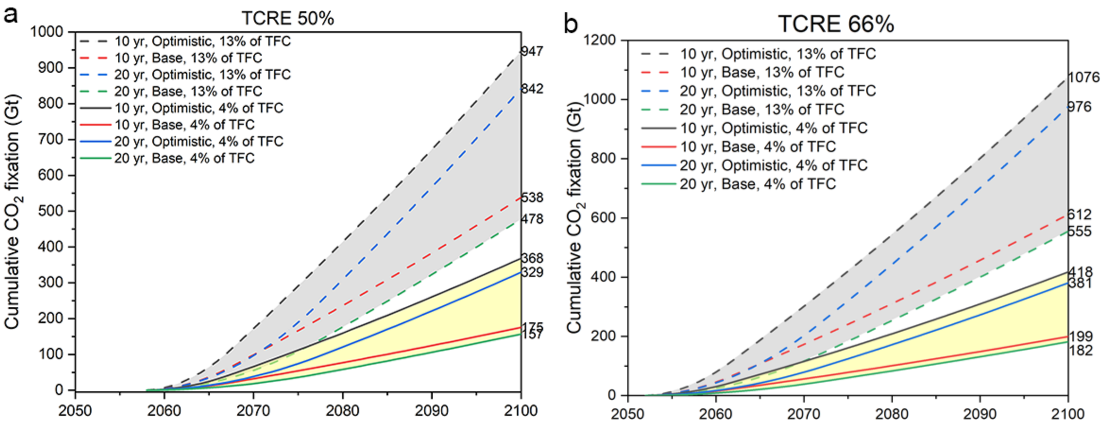

Figure 6 Cumulative total CO_2 fixation limited by global available energy. CO_2 fixation with a) 66% of TCRE and b) 50% of TCRE

Apart from BECCS and DACCS, most 1.5 ℃-consistent pathways in IPCC's report incorporate afforestation/reforestation (AR) as well, one of the only widely practiced CDR methods currently. Detailed estimation on the potential of this natural CDR strategy is out of the scope of this study. We took the reported potential from IPCC that cumulative 252 Gt CO_2 is projected for all net removal on managed land including AR from 2020 to 2100[18] alleviating the burden on engineering CDR strategies, although the magnitude of afforestation's albedo-warming effect is still debatable. By subtracting the contribution of AR from the CO_2 removal target, the remaining CO_2 to be removed, 828 Gt CO_2 (66% of TCRE; 1046 Gt CO_2, 50% of TCRE), still requires more than "world wartime costs" for the base technical levels and 10~11% of TFC even for the optimistic ones.

Combined with AR, less energy-intensive approach with 0.5-10 Gt CO_2/yr annual mitigation potential[1], we complete the net CO_2 emission trajectories with the same upscaling rate for AR (**Figure 7**). The year when net zero emission is achieved falls in 2056 to 2070, resulting in overall beyond-budget emission of 1113-1200 Gt (66% of TCRE; from 2062 to 2076, 1333-1429 Gt CO_2, 50% of TCRE). Corresponding cumulative removal till 2100 offset 887-50 Gt CO_2 (66% of TCRE; 625-26 Gt CO_2, 50% of TCRE), which implies the almost irreversible CO_2 levels in the atmosphere.

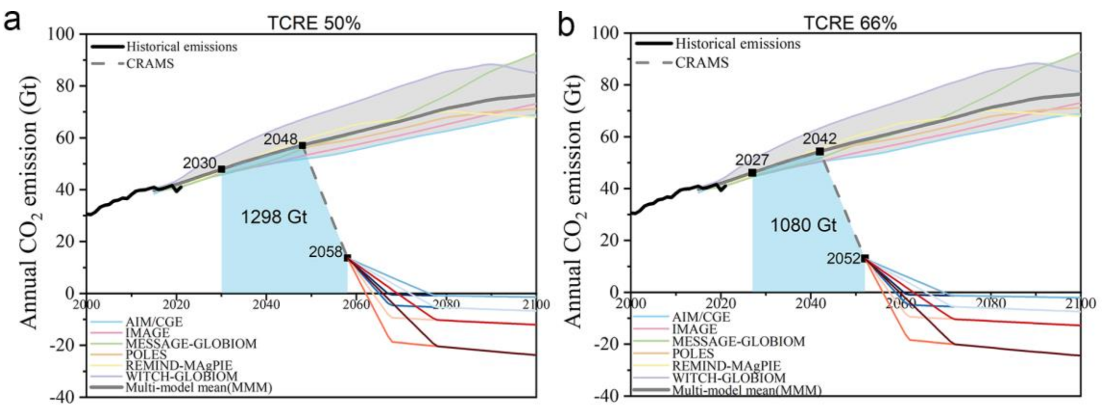

Figure 7 The projected net CO_2 emission trajectories with CDRs limited by global available energy.

Conclusion

Current CO_2 mitigation implementation lead to remarkable emission gaps to the Paris Agreement's temperature goals. Large-scale CDR strategies is indispensable when mitigation consensus is eventually reached at the exhaustion of carbon budget with emerging climate hazards. Under the proposed CRAMS, the balanced projections of CO_2 emission and mitigation pathways and global energy under current policies provide information and certainties for CO_2 removal target and available energy for the deployment of CDR strategies. The re-calibrated CDR potentials reveal the insufficiency and limitation of BECCS, DACCS and EW. Although still possessing many scientific and technical unknowns, techno-economic breakthroughs must be made to these CDRs as our only way to reverse the warming trend in the future.

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