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Below zero†

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The current climate debate focuses on how to reach net zero latest by 2050. Most transformation pathways rely on negative emissions to compensate "hard-to-avoid" emissions, for example in aviation, industry or livestock farming. However, even a constant global heating at 1.5 °C may trigger climate tipping points, such as the loss of cryosphere, permafrost or ecosystems. It therefore becomes necessary to achieve "below zero" with large-scale negative emissions, reducing atmospheric CO₂ concentration and climate forcing. This paper argues for a systemic view and shows with a comparison of past, current and future carbon stocks and flows that storing the minimally necessary removals will already be challenging. Consequently, continued fossil emissions shall be avoided completely, as their compensation increases removals and binds societal resources. For delivering the required scale and speed of negative emissions, scalable technical solutions will have to developed, as bio-based solutions are limited though essential for reverting land use impacts and safeguarding biodiversity. In this context, it is important to investigate the potential of a circular carbon economy, storing carbon in safe and reliable material cycles.

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Environmental significance

Negative emissions are foreseen at large scale to achieve the intermediary target of net zero emissions. Even though politically endorsed and dominating the debate, the net zero narrative disregards the required reduction of atmospheric carbon to achieve long-term climate stability and the cumulative storage capacity for negative emissions. From an Earth system perspective, compensating "hard-to-avoid" emissions cannot be sustained indefinitely and distracts from returning to a safe climate regime as it binds materials, energy and societal resources. Furthermore, it reveals the limited, though important, potential of biobased solutions, necessitating to design and investigate scalable and reliable technical carbon storage.

Introduction

Earth is experiencing rapid loss of ice and permafrost,^{1,2} increase in weather and ocean extremes,^{3,4} declining biological productivity^{5,6} and many more severe consequences already now at only 1.19 °C global heating.⁷ The international, political consensus is to limit global heating to well below 2 °C and preferably 1.5 °C,⁸ which means a further substantial increase compared to today. Though still attainable in principle,⁹ sluggish climate action requires ever faster and more ambitious strategies.¹⁰ While it is most urgent to limit peak heating to prevent severe short term damages, it is insufficient to avoid climate tipping with high confidence.^{11,12}

During the past one million years, atmospheric CO₂ concentration had been between 180 ppm in ice ages and 280 ppm in warm periods.¹³ Anthropogenic CO₂ emissions are accumulating in the atmosphere and upper oceans, leading to an increase in atmospheric CO₂ concentration, the main driver

to minimize cumulative emissions. However, constant global heating at 1.5 °C may still exceed vital limits for other climate impacts—such as sea level rise, ocean acidification or decline in biological productivity¹²—and trigger a tipping cascade, inducing runaway heating with disastrous consequences. Consequently, it is necessary to actively remove CO₂ from the system in order to reduce the induced heating and halt or even revert the loss of cryosphere, forests and other essential Earth systems. An atmospheric CO₂ concentration of 350 ppm has been proposed as a safe level for long term climate stability. Reaching 350 ppm—or any other long term climate target—inevitably requires below zero emissions at a massive scale. 21,22

for global heating.14 It is rising faster than ever and currently

crossing 417 ppm.15 For limiting peak heating, it is imperative

The current debate on climate action centres around reaching net zero emissions globally in about 2050. In this narrative, which can be summarized as "Do your best, remove the rest", 23 "hard-to-avoid" emissions can be continuously compensated by negative emissions. 24 What is considered "hard-to-avoid" is currently discussed in a socio-economic perspective: either substitution with emission free alternatives is considered "too costly" (e. g. hydrogen reduced steel, 25 synthetic fuels and chemicals 26) or shifting and reducing consumption, often related to affluence, 27,28 "too inconvenient" (e. g. shift to

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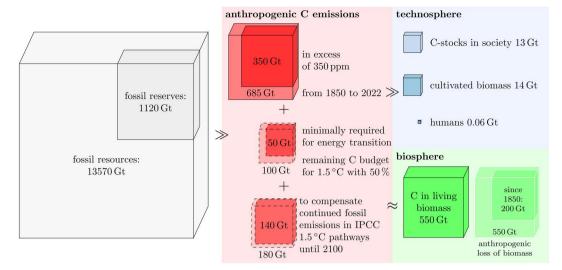


Fig. 1 Comparison of anthropogenic C emissions (red) with C stocks in fossil fuels (grey, more than one order of magnitude larger), technosphere (blue, one order of magnitude smaller) and biosphere (green, the same order of magnitude). Solid black lines denote current, dashed lines future and white lines past C stocks

predominantly vegan diet29 or alignment of energy demand with solar supply³⁰). Compensation is assumed as possible between all kinds of greenhouse gas emissions and across different locations and time scales.31 It leads to delaying actions for avoiding emissions, which has been termed mitigation deterrence.32 It further gives rise to concerns such as possibly negative effects on biodiversity, infringement of indigenous rights or "climate-colonialism", 33-35 for a minority of rich individuals, companies and countries compensates lifestyle-dependent emissions on foreign land.27

In transition pathways aiming at limiting peak heating to 1.5 °C considered by IPCC, for example, negative C emissions have to start this decade and increase to approximately -3 Gt/ a in 2050.36 This is necessary to compensate the remaining fossil emissions of equal magnitude³⁶ (i. e. 27% of current fossil emissions²⁴). After this important milestone is reached, fossil emissions decrease only slightly, while negative emissions increase to about -5 Gt/a in 2100 (Fig. S1 and Section S2†). Global temperature correlates almost linearly with increasing cumulative emissions14,37 and non-linearly (i. e. with a hysteresis) with decreasing ones.38-40 Until 2100, negative C emissions cumulate between -220 Gt and -260 Gt in IPCC pathways (Fig. S2†). Yet, only 1/3 (-70 Gt to -90 Gt) reduce climate forcing and are thus truly negative emissions, while the rest (-140 Gt to −180 Gt) is compensating continued fossil emissions (Fig. 1, S1 and S2, and Table S1†). As a consequence, these projected negative emissions will have little effect on global temperature reduction despite tremendous efforts (260 Gt is as much C as had been emitted over the past 30 years).

Regardless of with or without compensation, emissions need to reduce to (net) zero soon to limit peak heating. For stabilizing the climate in the long term, "cleaning-up" the atmosphere and returning to 350 ppm inevitably requires below zero emissions at a large scale. The question is, if and how much hard-to-avoid emissions society can and wants to afford, which need to be

continuously compensated in addition. Avoiding emissions completely will remove the underlying cause for climate change and necessitates faster actions.32,41 Yet, fossil fuels cannot be switched off immediately, as the replacing renewable energy system first needs to be built. Installing the necessary infrastructure requires energy in addition to (reduced) societal demand. In the beginning of the transition, it can only come from the fossil energy system. 9,21,30 A minimum of 50 Gt of C has to be emitted to achieve the energy transition.9 When exhausting the remaining carbon budget for 1.5 °C with 50% confidence, this increases to 100 Gt of C (Fig. 1 and Section S1†). Together with the 350 Gt C already in excess in the atmosphere and upper oceans, at least 400 Gt to 450 Gt has to be removed and stored safely as below zero emissions to reach 350 ppm. For comparison, this is about as much pure carbon (C) than the mass of all concrete in use in society today42,43 (Fig. 2). The required scale of negative emissions is thereby one order of magnitude larger than C currently contained in or managed by the technosphere, in the same order of magnitude than C contained in living biomass and two orders of magnitude smaller than fossil fuel resources (Fig. 1). This is—simply put a gargantuan task ahead. Hard-to-avoid emissions in IPCC pathways³⁶ necessitate to increase negative emissions by 40% within this century and more thereafter.

Negative emission routes

Different technical and nature based negative emission technologies (NET) are being discussed in literature (Fig. 2).47,48 Most of them remove CO₂ by reverting the mode of release (i. e. biomass growth and direct air capture), while some propose new routes, such as enhanced weathering49,50 or ocean fertilisation.51 Considering that fossil energy use created the climate crisis unintendedly, the potential risks and side effects of new geo-engineering experiments are high.48 Consequently and in

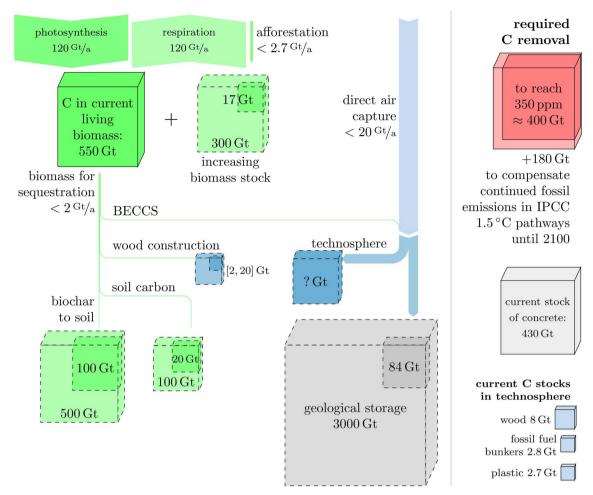


Fig. 2 Comparison of carbon stocks (cubes) and flows (Sankey) for different negative emissions routes. All stocks and flows are representing C mass content (except concrete). For reaching 350 ppm, CO₂ currently in excess and minimally required emissions during the transition (red cube) have to be removed. Continued fossil emissions have to be removed in addition (light red cube). This is comparable to the mass of current concrete stocks,⁴³ while two orders of magnitude above current C stocks in society: wood, fossil fuels bunkers and plastics (right). Current living biomass processes around 120 Gt/a of C (photosynthesis and respiration)⁴⁴ and may provide a limited flow of C for sequestration. This flow can be stored either in wood construction,⁴⁵ through bioenergy with carbon capture and storage (BECCS) in geological or technical stores, as biochar in soil or by increasing soil carbon sequestration (SCS). Biochar and SCS are limited by the maximum C content of soil.⁴⁶ Afforestation can increase C in living biomass. Direct air capture (DAC) is limited by the sustainable potential of renewable energy²¹ and by far the largest potential NET flux. C from DAC and BECCS can be transported and stored in geological storage or incorporated in products and cycled within the technosphere.

precaution, reverting anthropogenic emissions shall be preferred through their mode of release, *i. e.* technical and biological NETs for fossil and land use emissions. In the following, the possible scales for such NETs are put in perspective with past, current and future carbon stocks and flows (Fig. 2 and S3 and Table S2†) applying an Earth systems perspective irrespective of social and economic aspects. Negative emission potentials are counted as the actual removal capacity, disregarding indirect substitution effects *e. g.* resulting from replacing fossil energy through bioenergy with carbon capture and storage (BECCS) or concrete with wood as a construction material.

Increase C stocks in living biomass

Restoring and increasing C stocks in the biosphere, e. g. through afforestation, 52 can, if implemented properly,

safeguard biodiversity and restore ecosystem integrity in addition. The C-flux for afforestation is limited by the available land and the growth dynamics of forests and may range between 0.12 Gt/a and 2.7 Gt/a. 48,52-56 Total storage potential is limited due to saturation of C-uptake in mature forests after approximately one century.56 Estimates for the cumulative storage potential vary widely in the literature between 17 Gt and 300 Gt.53,56 For comparison, land use change has emitted about 200 Gt of C since 1850,57 which is within the range for afforestation potential (Fig. 2). Even though difficult to achieve, reverting land use change to pre-industrial levels can at best remove half the C necessary to reach 350 ppm. It would be insufficient to deliver the negative emissions necessary in IPCC 1.5 °C pathways alone (260 Gt of C until 2100). Today, the biosphere contains 550 Gt of C in living biomass, the majority in forests ecosystems. 44,58 Bar-On et al. 58 estimate that living biomass halved since humanity's

existence, *i. e.* living biomass has been reduced by 550 Gt. The even more hypothetical case of restoring biomass to pre-human level could just be sufficient to reach 350 ppm.

Biomass for sequestration

In addition to increasing biomass stocks, the biosphere can also provide a C-flux for permanent sequestration through wood for construction, 45 BECCS, 53,59 soil carbon sequestration (SCS) or biochar to soils.46 Sustainable wood production, useable for wood construction, BECCS and biochar, is limited to a C-flux of 0.6 Gt/a. 60,61 Agricultural residues (currently 2.46 Gt/a (ref. 62)) and dedicated biomass production on marginal land may increase the C-flux for sequestration.53 However, human appropriation of net primary production is already substantial⁶³ and causing severe pressure on planetary boundaries. 29,64,65 Estimates for biomass sequestration C-flux in literature range between 0.14 Gt/a (ref. 48) to 3.3 Gt/a (ref. 54) (up to 11.6 Gt/a, 53 which seems unrealistic in comparison to total current C harvest of 8 Gt/a (ref. 62)). Considering decline in biological productivity, loss of fertile land and increased desertification⁵ and a sequestration efficiency of about 50%,53 C-flux of biomass sequestration is unlikely to exceed <2 Gt/a. Throughout Earth's history, the size of this biological "leak" has been four orders of magnitude smaller. During the Carbon period (350 to 250 million years before now), net surplus produced by forests transferred 12 300 Gt of C to coal deposits.66-68 The average Cflux to coal had been 0.000123 Gt/a over this entire period. Similarly, oil and gas deposits had formed during the Jura period (190 to 175 million years ago) and stored 550 Gt in oil and 1 820 Gt in gas. 66-68 The average C-flux into these deposits had been 0.000158 Gt/a. This comparison suggests that unperturbed natural ecosystems may have a long term potential to remove atmospheric C of around 0.00016 Gt/a only, making it necessary to investigate in detail if 2 Gt/a could even be sustained.

The cumulative storage potential of biomass sequestration varies for each route. Wood construction may increase C stock by 2 Gt to 20 Gt this century, depending on construction demand and wood content. This may roughly double the wood stock in society (currently 8 Gt (ref. 42)). Carbon captured with BECCS has to be stored in technical or geological storage (see below), whereas SCS and biochar increase the C stock in soil. Currently, soils are estimated to hold between 1 500 Gt and 2 400 Gt of C (excluding living biomass) and permafrost may contain 1 700 Gt in addition (Fig. S3†). Biochar may add 100 Gt to 500 Gt (ref. 53 and 56) and SCS 20 Gt to 100 Gt (ref. 56) to soils (Fig. 2). Biochar may contribute a significant storage potential, still impacts of application at scale on soil productivity, biodiversity, C release and resistance to extreme events—like wildfires—remain to be investigated.

Direct air capture

Capturing CO_2 directly from the ambient air is energy intensive, but in contrast to bio-based solutions nearly independent of land availability.⁶⁹ When powering direct air capture (DAC) with solar PV from the already built environment,²¹ additional land

conversion is negligible.70 There are different DAC technologies available, some already at pilot scale and others in development.71-76 When powered with renewable energy, DAC has negligible C emissions stemming from the production of the materials contained in the infrastructure, 77,78 which can also be avoided by decarbonising the supply chains. Resources required for the plant and sorbent are considered uncritical.78,79 For these reasons, DAC are foreseen as a major part of future energy systems, 21,69,80-82 yet it faces the challenge of upscaling from pilot scale to a global industry. 56,83 Furthermore, DAC may be constrained by economic and social limitations, 48,54 however, they can be overcome in principle. 47,84,85 C-flux from DAC is ultimately limited by the availability of excess solar energy on the already sealed surface of the built environment. At constant or decreasing energy demand from society, solar PV could power a C-flux from DAC of <20 Gt/a (Section S4†),21 one order of magnitude larger than for biomass sequestration or afforestation. Yielding this potential will depend on the mobilisation of resources building the required solar infrastructure, the subsequent handling (e. g. transport) as well as the energy and resource requirements for technical and geological storage.

Geological storage

The principal challenge of BECCS and DACCS is the long-term safe storage of technically captured CO2.86 Geological storage is the injection of CO2 into geological formations, which may hold CO2 over centuries and millennia.86,87 Under certain conditions, CO2 reacts with the surrounding rock to form carbonates.88 Current geological storage projects are, however, often associated with oil and gas production.89 One example is the Sleipner project in Norway.90 This offshore gas field has a high content of CO2 in the gas, which is separated and injected back into the gas field to recover more gas ("enhanced oil/gas recovery"). In the first 20 years of operation, the Sleipner project stored 4.4 Mt of C. During the same time (1996-2016), oil and gas had been extracted worth 48 Mt of C emissions,91 paradoxically with the help of the separated and injected CO₂. Consequently, the project avoided 9% emissions. Other projects without enhanced oil/gas recovery are currently under development (e. g. Climeworks in Iceland92). At the scale of injections at Sleipner, about 10⁴ similar sized storage operations would be necessary to store the minimally required 400 Gt of C before the end of this century, highlighting the challenge of upscaling

The Global CCS Institute estimates the global potential for C storage in saline formations to 84 Gt, ⁹³ while IPCC estimate the total geological potential to be in the range of 500 Gt to 3 000 Gt. ⁷⁵ Even if the cumulative storage potential may be sufficient, it may be difficult to find enough suitable and safe geological formations for permanent CO₂ storage in time. Additionally, leakage has to be stored again, increasing the required C-flux. ⁹⁴

Technical storage

Geological storage is an end-of-pipe solution, as such a burden (or "cost") to society. In contrast, incorporating C in products

and cycling C in the technosphere can create value. A "circular carbon" economy may make excess C the main constituent of the socio-economic metabolism. Currently, the technosphere holds about 13 Gt of C, mostly in wood and paper products (8 Gt (ref. 43)), plastics (2.7 Gt (ref. 43)) and fossil fuel bunkers (2.8 Gt (ref. 67) Fig. S3†). In contrast to today, C would need to be cycled within the technosphere, preventing leakage to the atmosphere. It would also need to increase C stocks in the technosphere by more than one order of magnitude, for example by incorporating it in long lived products, such as buildings and infrastructure. This, however, has to go much beyond current efforts of wood construction (see above) and "CO2 binding concrete". Concrete takes up about 10% of CO₂ emissions previously released in cement production during the service life of buildings. This may be increased to <30% during recycling, when exposing crushed concrete under increased pressure and concentration of CO2.95-97 If all current concrete (430 Gt (ref. 43)) would take up 30% of CO2 emissions from their production (about 30 kg C t⁻¹ concrete⁹⁸), it could remove about 4 Gt of C. Consequently, CO₂ binding concrete may at best contribute <1% to below zero. Using captured C in synthetic fuels or short lived products (e. g. carbonated drinks, single use plastics) has a storage potential proportional to the stocks of these products. For example, bunkering synfuels to the equivalent of one year's consumption of today's fossil fuels, would only hold 13 Gt of C out of the atmosphere. Consequently, storage of C in short lived products and synfuels may only make a minor contribution to C storage, while greatly increasing C circulation from and to the atmosphere and its associated energy demand.

In contrast, it would be necessary that C becomes the main constituent of any bulk material were we to store a significant fraction of removed C in the technosphere. Research is necessary for finding practical means to convert atmospheric ${\rm CO_2}$ into synthetic polymers, graphite, graphene, diamonds or other C containing materials and keeping them out of the atmosphere for centuries at low energetic costs.

Conclusions

Below zero emissions are inevitable to reduce atmospheric CO₂ concentration and stabilize the climate. A minimum of 400 Gt of C has to be removed and stored permanently and safely. This is as much pure C as all the concrete in society or almost as much as contained in currently living biomass. Negative emissions for compensating continued fossil emissions have to be stored in addition. Already for the minimally required negative emissions, finding practical solutions at scale is a challenge. Consequently, the notion of "hard-to-avoid" emissions has to be rethought, finding ways to avoid them by substitution with expensive but emission-free technology as well as shifting and reducing consumption. By looking beyond net zero and applying a systems perspective, our strategy has to change: compensation of continued fossil emissions is no longer viable, in contrast, it distracts from the major task of returning to safe climate conditions. It conveys a false hope, leads to stranded investments, binds materials, requires energy and generates continuous need for storing C. These resources are more

urgently needed for building the replacing renewable energy infrastructure and removing excess CO₂ from the atmosphere to stabilize the climate in the long run.²¹ Restoring the biosphere has co-benefits of safeguarding biodiversity along with storing C. As the biosphere's stock and flow capacities are limited, it is relevant to design and investigate a leading role of direct air capture, which has the potential to remove C one order of magnitude faster than bio-based NETs (Fig. 2). Safe, reliable and scalable storage possibilities at low energy costs have to be developed, *e. g.* as circular carbon materials in the technosphere. While the remaining resource flows have to drastically decrease to return to the safe operating space for humanity,⁹⁹ C-fluxes out of the atmosphere into stocks in the technosphere will have to increase: a huge potential market that will have to grow fast.

Author contributions

Conceptualization, H. D.; method, H. D.; validation, H. D.; formal analysis, H. D.; investigation, H. D.; visualization, H. D.; writing-original draft preparation, H. D.; writing-review and editing, H. D.

Conflicts of interest

The author declares no known competing interests, which could have appeared to influence the work reported in this paper.

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