

# Annual Report 2021



High Meadows  
Environmental  
Institute

Carbon  
Mitigation  
Initiative







Matteo Detto in Harvard Forest (Massachusetts), taking canopy leaf samples using a bucket truck (Photo by Hannes de Deurwaerder)



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# Introduction



Peat from the Pine Barrens, New Jersey with American Tree Moss  
(Photo by Linta Reji and Xinning Zhang)





The Carbon Mitigation Initiative (CMI) at Princeton University is the longest running partnership with industry in the university's history. Sponsored by bp and independently operated and administered by the High Meadows Environmental Institute (HMEI), CMI funds 15 principal investigators (PIs) and over 50 researchers who are tasked with finding compelling and sustainable solutions to the carbon and climate problems.

For 22 years, CMI has been leading the way in developing cutting-edge research in engineering, ecology, earth sciences, and environmental policy. This research has led to new insights into the impacts of climate change, developments in environmental engineering principles, and to salient policy recommendations and analyses that continue to impact the scientific and policy-making spheres, both domestically and internationally.

In this report, readers can find the latest information about CMI's key activities and research initiatives. Each of the fourteen highlights, written by CMI PIs and their teams, features a summary of the latest, on-going research conducted by their groups. The Highlights section is followed by a complete list of this year's CMI publications.



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## Ongoing Initiatives

Despite the COVID pandemic of the last two years, CMI researchers have maintained unabated research progress. One research initiative, which continues to gain traction globally and from which new projects were borne, The *Net-Zero America* project, has had outsized impact on policymakers and scientists in the US and abroad. Evolving from *Net-Zero America*, the REPEAT project allows policymakers and the public to view the impacts of proposed climate and energy policies before they are voted into law.

Another focus of CMI research in 2021 was carbon capture and storage (CCS), an important component in the transition to net-zero. Indeed, most models that show the economy achieving net-zero by 2050 rely heavily on this technology. CCS can be deployed by using hubs that carry CO<sub>2</sub> from various capture sites via pipeline networks to a centralized injection site. One 2021 CMI highlight posits that CCS will be limited by large-scale geologic limitations on the rate at which CO<sub>2</sub> can be injected. Another group of CMI researchers built a computer simulation tool to predict how geologic conditions can impact larger-scale geological carbon storage. Addressing the disconnect between CCS ambitions and constraints is crucial to successful CCS investment and policy decisions.

Other initiatives in the CMI research realm described in more detail in the following section include:

- Determining the impact of aerosol particles on global radiative forcing
- Consequences of hydrogen leakage on atmospheric methane
- Impacts of wetlands on methane emissions
- Predicting biodiversity responses to climate change
- Carbon capture through mineral-carbon interactions in water
- Using mathematical models to predict future climate
- Impacts of climate and fire on the fate of Amazonian forests
- Predicting reduced oxygen levels in the world's oceans
- Understanding the frequency of tropical cyclones
- Using calcium compounds for carbon capture



## Annual Meeting

For the second consecutive year, the CMI Annual Meeting was held virtually in April 2021. Despite the lack of in-person interaction, the meeting allowed over 100 participants to come together from across the globe. The virtual program included several deep dives that explored a variety of energy and land-based climate solutions.

Speaking to over 100 attendees, Stephen Pacala, the Frederick D. Petrie Professor in Ecology and Evolutionary Biology and the Director of CMI opened the meeting stating, “There seems to be a recognition happening all at once in different parts of the world, in companies, board rooms, halls of government and the academy, that the transformation of our energy system to net-zero emissions of all greenhouse gases is feasible, economic and necessary.”



*Stephen Pacala (left), the Director of the Carbon Mitigation Initiative (CMI), and Kelly Goddard (right), former Vice President for Carbon Ambition at bp and the bp executive sponsor of CMI, opened the 2021 CMI Annual Meeting held virtually on April 20-21.*

During her welcoming remarks, Kelly Goddard, then Vice President for Carbon Ambition at bp and CMI’s bp executive sponsor remarked, “In early 2020, bp announced its ambition to become a net-zero company by 2050 or sooner, and to help the world get to net-zero. bp is now over one year on from announcing this ambition and we’re pleased that we have made progress against what we have planned. Societal expectations and calls for action continue to increase, reinforcing what we have set out and the need for collaboration to support action.”

“We continue to see our long-term collaboration with Princeton’s CMI as an important science and technology partnership and we value the research being done by CMI to help the understanding of challenges and opportunities in the energy transition. Over the next two days, we will hear about the relevance of this research to bp and to broader stakeholders including policymakers,” said Goddard.



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The meeting's first deep dive included speakers from the Biden Administration, non-governmental organizations, and bp who discussed the role of carbon capture utilization and sequestration (CCUS) in net-zero energy systems in the U.S., Australia, China, and Europe.

In another deep dive, CMI leadership team members and principal investigators, Jonathan Levine and Amilcare Porporato, described a newly launched CMI initiative aimed at determining how land-based climate solutions can be deployed globally to maximize carbon storage on land, while at the same time maintaining global biodiversity and food security.

"If we are going to actually achieve net-zero at a global level, country level or even a company level, land-based climate solutions must be deployed at massive spatial scales to have impact," said Levine.

In addition, Pacala announced the following recipients of two awards named in honor of Robert H. Socolow, Emeritus Professor of Mechanical and Aeronautical Engineering at Princeton and CMI Co-director from 2000 to 2019.

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## Best Paper Awards 2020

Since 2010, the CMI Best Paper Award for Postdoctoral Fellows has been presented annually to one or two CMI-affiliated postdoctoral research associate(s) or research scholar(s) selected for their contribution to an important CMI paper. In late 2019, CMI created a similar award honoring a CMI-affiliated doctoral student for their contributions to an important CMI paper.

Former Postdoctoral Researcher Erin Mayfield, now an assistant professor at Dartmouth College, received the Robert H. Socolow Best Paper Award for Postdoctoral Fellows for her work on the *Net-Zero America* report. The second Robert H. Socolow Best Paper Award for Doctoral Students was given to Ching Ho Justin Ng, who received his Ph.D. in Atmospheric and Oceanic Sciences from Princeton in 2019, for his paper "Large-scale environmental controls on the seasonal statistics of rapidly intensifying North Atlantic tropical cyclones," published in *Climate Dynamics*.



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Erin Mayfield (left), a postdoctoral research associate in the High Meadows Environmental Institute received the Robert H. Socolow Best Paper award for the Postdoctoral Fellows. Justin Ng, who received his Ph.D. in Atmospheric and Oceanic Sciences from Princeton in 2019, was awarded the Robert H. Socolow Best Paper Award for Doctoral Students.



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## Collaborations

In addition to the external collaborations undertaken through the Princeton-led Net-Zero America project and the National Academies of Sciences, Engineering and Medicine Committee on Accelerating Decarbonization of the U.S. Energy System, CMI continued its engagement with three excellent research programs bp has long supported: the Center for the Environment at Harvard University; the Center for International Environment and Resource Policy at Tufts University; and the Thermal Engineering Department and the Tsinghua-bp Clean Energy Research and Educational Center at Tsinghua University.

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## New Collaborative Initiative

In 2021, CMI established a new initiative, *Land-Based Climate Solutions: Variable Responses to Economic Incentives*, in collaboration with University of California Santa Barbara and the Environmental Defense Fund. The project seeks to understand how economic, institutional, political, and cultural differences between countries are likely to affect the outcome of policy incentives encouraging private landowners to internalize climate impacts in their land-use decision making. The econometric analysis is expected to be completed in 2024.



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## Honors and Appointments

In 2021, CMI scholars were awarded with honors and appointments. Steve Pacala was appointed member of the President’s Council of Advisors on Science and Technology (PCAST); Gabriel Vecchi was appointed Director of the High Meadows Environmental Institute; Laure Resplandy received an National Science Foundation Career Award to study the formation and future of Pacific and Indian Ocean dead zones; and Jesse Jenkins was awarded the Undergraduate and Graduate Engineering Council Award for Excellence in Teaching.



# Research – At a Glance

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## Toward Accelerating the Deployment of CO<sub>2</sub> Capture and Storage Hubs

**PRINCIPAL INVESTIGATOR: ERIC LARSON**

Carbon dioxide (CO<sub>2</sub>) capture and storage, including at bioenergy conversion facilities, will be crucial if the U.S. is going to reach net-zero emissions by 2050. The Energy Systems Analysis group launched new research in 2021 to explore with high spatial and temporal resolution how these features of the future energy landscape might evolve most expeditiously. The research attempts to understand the potential performance, benefits, costs, and challenges of deploying regional “hubs,” which are clusters of CO<sub>2</sub> capture sites linked via pipeline networks to storage injection sites. A deep understanding of such hubs will help inform public and private decision-making to accelerate their deployment. An initial regional focus is the Louisiana Gulf Coast.

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## Bridging the Gap Between CCS Ambition and Reality

**PRINCIPAL INVESTIGATOR: CHRIS GREIG**

Most integrated assessment and other macro-scale energy system models find that widespread carbon capture and storage (CCS) at very large scales is crucial to achieve ambitious CO<sub>2</sub> reduction goals. However, these models assume that abundant low-cost geological storage is available to meet all needs. This research presents the contrasting view that storage capacity uncertainty could seriously hamper the pace and scale of CCS deployment, especially in developing Asian economies. This storage capacity uncertainty leads to “chicken-or-egg” challenges that deter investment. The implications for emissions reduction goals and the role that CCS should play in a net-zero future warrant more attention.

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## REPEAT Project Provides Real-time Look at Evolving U.S. Climate Policy

**PRINCIPAL INVESTIGATOR: JESSE JENKINS**

The REPEAT Project is led by Jesse Jenkins of the Princeton ZERO Lab. It provides a detailed, “real-time” evaluation of the United States’ evolving energy and climate policies and the country’s progress on the road to net-zero greenhouse gas emissions. The Project uses a novel suite of geospatially-granular planning, modeling, and visualization tools coupled with macro-scale optimization models of the United States energy system. The goal is to publish regular, timely, and independent environmental and economic evaluation of federal energy and climate policies as they are proposed and enacted. (See [repeatproject.org](https://repeatproject.org).)

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# Nucleation and Growth of Nano-Aerosol Particles

**PRINCIPAL INVESTIGATOR: IAN BOURG**

The Bourg group is working to better understand the initial stages of nucleation and growth of secondary organic aerosol particles. Atomistic-level simulations reveal a strong affinity of organic molecules for the water-air interface and a previously unknown transition between two highly distinct structures during the initial stages of nano-aerosol particle growth. These particles, which spontaneously form in the atmosphere through clustering of water, natural or anthropogenic semi-volatile organics, and ions, are key unknowns in extant climate model predictions. Since the formation of these particles can be enhanced by land-use changes, their impact on climate must be considered in carbon mitigation strategies that modify carbon and water dynamics at the land-atmosphere interface.

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# Soil Uptake and Methane Feedback of Atmospheric Hydrogen

**PRINCIPAL INVESTIGATOR: AMILCARE PORPORATO**

Hydrogen ( $H_2$ ) plays a crucial role in global energy scenarios aimed at achieving net-zero. Because it is not a greenhouse gas,  $H_2$  has been touted as an alternative to fossil fuels in certain energy sectors. But the environmental consequences of perturbing the global hydrogen cycle are still largely unknown. Specifically, there are concerns around hydrogen interference with the methane ( $CH_4$ ) atmospheric sink by the hydroxyl radical (OH). To sharpen future  $H_2$  projections, the Porporato group has been quantitatively addressing the major sink of tropospheric  $H_2$ , namely the soil uptake by bacteria, and the methane feedback of  $H_2$  fugitive emissions. This research informs bp's aims of developing the  $H_2$  economy in a manner that minimizes adverse climate impacts.

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# The CMI Wetland Project: Understanding the Biogeochemical Controls on Wetland Methane Emissions for Improved Climate Prediction and Methane Mitigation

**PRINCIPAL INVESTIGATOR: XINNING ZHANG**

Methane ( $CH_4$ ) is the second most important anthropogenic climate forcer after carbon dioxide. Determining the importance and mechanisms of different anthropogenic and natural methane sources and sinks across temporal and spatial scales remains a fundamental challenge for the scientific community. Wetlands are dominant but highly variable sources of methane and are predicted to play a critical role in carbon-climate feedbacks. Methane emissions from these areas are shaped by a complex and poorly understood interplay of microbial, hydrological, and plant-associated processes that vary in time and space.

The CMI Wetland Project aims to identify the biological and chemical mechanisms that promote methane emissions from wetlands. The goals are to improve predictions of carbon-climate feedbacks and strategies of methane mitigation. A better understanding of the factors responsible for the greatest methane emissions from wetlands is crucial to bp's actions aimed at targeting this powerful greenhouse gas and thus a vital step towards a low-emissions future.



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# Modeling Large-Scale CO<sub>2</sub> Injection in Highly Reactive Rocks

**PRINCIPAL INVESTIGATOR: MICHAEL CELIA**

When carbon dioxide (CO<sub>2</sub>) is injected into highly reactive rocks like basalts, the injected CO<sub>2</sub> will react quickly to form new carbonate rock. This is the most stable form of geological carbon storage. The Celia group built a computer simulation tool to study this process across a range of spatial scales. Small-scale injections show fast reaction rates, on the order of months to years that are consistent with results from small-scale field experiments. However, at the large spatial scales associated with practical industrial-scale injections, large-scale mass transfer limitations lead to much longer time scales for the reactions to proceed, on the order of a century or more. The newly developed computational tool allows these and other issues to be investigated efficiently. Progress in modeling and reliably assessing the potential of carbon sequestration in deep geological formations can help accelerate bp's efforts in decarbonizing heavy industry, while seeking new energy solutions.

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# Improving Forecasts of How Biodiversity Responds to Climate Change

**PRINCIPAL INVESTIGATOR: JONATHAN LEVINE**

To accurately assess the biodiversity benefits of slowing climate change through land-based climate solutions, the Levine group is challenging key mathematical assumptions in the leading biogeographic modeling tools for forecasting biodiversity response to climate and developing solutions critical for their accurate implementation. Reliably gauging the impact of mitigation initiatives on biodiversity is vital to current bp efforts towards a sustainable energy world.

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# The Efficiency of Enhanced Weathering

**PRINCIPAL INVESTIGATOR: AMILCARE PORPORATO**

Enhanced weathering (EW) is a negative-emission technology that holds the potential of alleviating the acidification of soils and natural waters. It is a carbon capture process designed to enhance and accelerate the chemical weathering of natural minerals that, when dissolved, remove carbon dioxide (CO<sub>2</sub>) from the atmosphere and store it in natural waters. However, because the precise characterization of EW efficiency is still little understood, Porporato's research group has been working on quantifying the Alkalinization Carbon Capture Efficiency (ACCE) of any mineral dissolution in various natural waters. The findings provide an important step forward in the identification of suitable environmental conditions for EW applications, better quantification of EW carbon sequestration potential, and important context for bp's plans for natural climate solutions.

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# Pacala Group

**PRINCIPAL INVESTIGATOR: STEPHEN PACALA**

The Pacala group's CMI research in the last year has embraced a large number of topics, including the final report of the Princeton *Net-Zero America* project. The group also produced or co-produced a series of papers, including: (1) a paper on possible failure modes of President Biden's U.S. decarbonization agenda; (2) a paper on the need for separate national targets for CO<sub>2</sub> and methane emissions; (3) two papers (one in *Science*) on how fire and other disturbances work against land-based climate solutions; and (4) multiple papers (one in *Science*) that improve the capacity of climate models to represent the carbon cycle in tropical forests and other ecosystems. Although not directly supported by CMI, Stephen Pacala chaired the effort of the National Academies of Science, Engineering, and Medicine that produced a peer-reviewed policy manual for a U.S. transition to a net-zero economy, which extensively used the *Net-Zero America* report and was a primary reason that CMI initiated the *Net-Zero America* effort in the first place.

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## Future Fires Compromise Amazon Forest Resilience to Climate Change

**PRINCIPAL INVESTIGATOR: ELENA SHEVLIAKOVA**

Assessments of alternative mitigation strategies to limit the impact of global change increasingly rely on simulations of Earth System Models (ESMs). In the tropics, a major biodiversity refuge and a net sink for anthropogenic carbon dioxide (CO<sub>2</sub>) emissions, ESMs consistently project that forests will thrive through the century due to CO<sub>2</sub> fertilization. In contrast, ecological models warn about a potential catastrophic forest loss under future drying conditions. A team of CMI researchers from the Pacala group and NOAA-GFDL used a state-of-the-art ESM to assess the impact of global change on tropical forest dynamics under alternative emission scenarios. Their ESM accounts, for the first time, for complex ecological mechanisms and large-scale biophysical forcing of vegetation dynamics. This research has broad consequences for the monitoring and management of tropical forests and opens new avenues for the design and implementation of carbon mitigation strategies. Of particular relevance to bp's natural climate solutions initiatives, this work informs the stability of carbon mitigated through avoided deforestation in tropical regions.



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# Diverging Fate of Oceanic Oxygen Minimum Zone and Its Core Under Global Warming

**PRINCIPAL INVESTIGATOR: LAURE RESPLANDY**

Global warming and anthropogenic activities are contributing to a loss of oxygen in the world's oceans. However, it is still unknown if this systematic deoxygenation will expand oxygen minimum zones (OMZs). These are areas where low oxygen levels threaten marine life and perturb the carbon and nitrogen cycles, potentially acting as an amplifying feedback on climate change. The Resplandy group uses the latest generation of climate model projections to evaluate how OMZs will evolve in the future, a key step to anticipate impacts on ecosystems, ecosystem services (e.g., fisheries), and greenhouse gas emissions. This work increases our understanding of an important oceanic impact of climate change that had many in the scientific community profoundly worried. It shows that, while serious, expanding oceanic dead zones do not represent the kind of tipping point that might significantly alter bp's plans for the energy transition.

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## Understanding Tropical Cyclone Frequency

**PRINCIPAL INVESTIGATOR: GABRIEL VECCHI**

The objective of this research from the Vecchi group is to better understand all aspects and variations in the statistics of tropical cyclone (TC) activity and other climate impacts over the past few centuries, as well as the coming one. An equally salient objective is to better understand the likely range of equilibrium and transient climate sensitivity, such as how much warming to expect from a doubling of atmospheric CO<sub>2</sub>. Key tools in these studies are climate and atmospheric models. These, along with analyses of the observed record, help researchers to distinguish whether observed multi-decadal to centennial changes in TC activity have been driven by large-scale factors such as ocean temperature changes, greenhouse gases, volcanic eruptions, or the El Niño, as opposed to random atmospheric fluctuations.

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## Calcium (Ca)-Based Solid Sorbents for Low Temperature CO<sub>2</sub> Capture

**PRINCIPAL INVESTIGATOR: CLAIRE WHITE**

White and her group are developing novel calcium (Ca)-based solid sorbents that are capable of selectively capturing carbon dioxide (CO<sub>2</sub>) from a mixed gas stream, or air, at ambient temperature. By understanding the solution chemistry during synthesis, the researchers have obtained phase pure Ca-based layered double hydroxides and demonstrated that these sorbents can selectively adsorb CO<sub>2</sub>. Ongoing efforts are focused on engineering energy-efficient regeneration methods and quantifying life cycle environmental and economic aspects. This project aligns with bp's goal of developing solutions to decarbonize the cement production process, thereby helping cities and corporations to decarbonize.

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# Toward Accelerating the Deployment of CO<sub>2</sub> Capture and Storage Hubs

PRINCIPAL INVESTIGATOR: ERIC LARSON

## At a Glance

Carbon dioxide (CO<sub>2</sub>) capture and storage, including at bioenergy conversion facilities, will be crucial if the U.S. is going to reach net-zero emissions by 2050. The Energy Systems Analysis group launched new research in 2021 to explore with high spatial and temporal resolution how these features of the future energy landscape might evolve most expeditiously. The research attempts to understand the potential performance, benefits, costs, and challenges of deploying regional “hubs,” which are clusters of CO<sub>2</sub> capture sites linked via pipeline networks to storage injection sites. A deep understanding of such hubs will help inform public and private decision-making to accelerate their deployment. An initial regional focus is the Louisiana Gulf Coast.

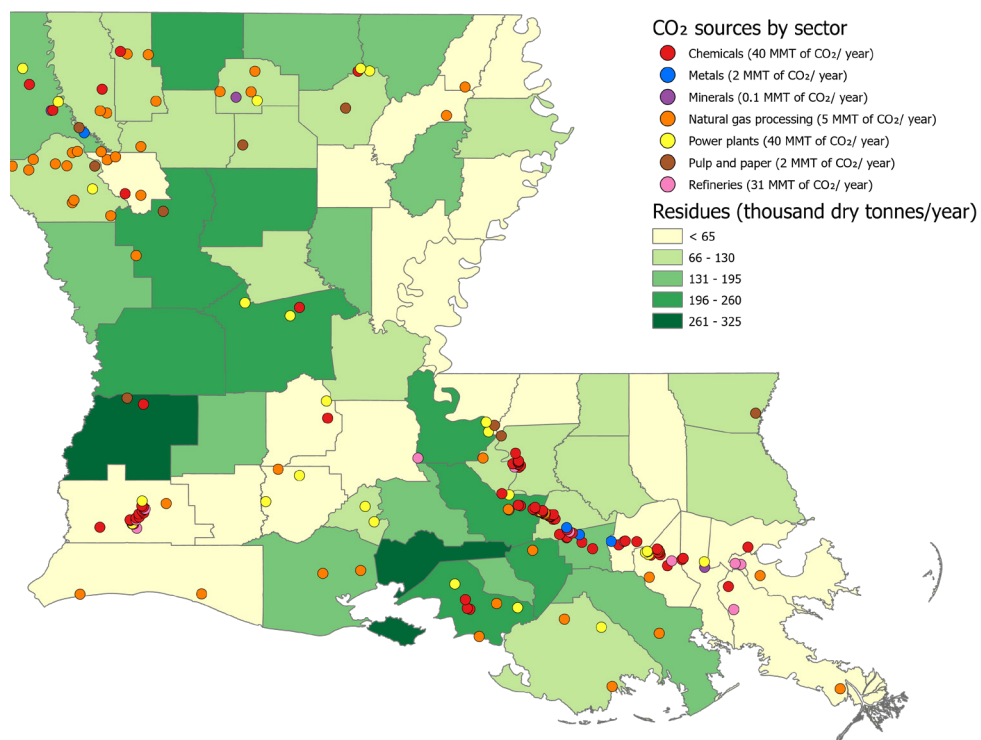
## Research Highlight

Last year, Princeton’s *Net-Zero America* (NZA) study (Larson et al., 2021) highlighted the key role that both CO<sub>2</sub> capture and storage (CCS) and biomass conversion to clean energy carriers with CO<sub>2</sub> capture and storage (BECCS) will need to play for the country to reach net-zero emissions by 2050. A deep-dive research effort was launched in 2021 to explore with high spatial and temporal resolution how these features of a future energy landscape might be evolved most expeditiously. The organizing focus is regional “hubs,” which are clusters of CO<sub>2</sub> capture sites linked via pipeline networks to storage injection sites. CCS hubs have been proposed for launching and upscaling a CCS industry (Oil and Gas Climate Initiative, 2022; Abramson et al., 2022; Meckel et al., 2021), including to help mitigate employment losses in the energy transition as fossil fuel industries shrink (Energy Futures Initiative, 2021). However, there has been relatively little detailed public-facing analysis to better understand bottlenecks and roadblocks that development of CCS hubs might encounter and how these might be overcome to accelerate deployment. There has been even less analysis of the challenges for developing CCS hubs that include BECCS facilities. The research is adopting a case-study approach and focusing initially on the Louisiana Gulf Coast region.



Louisiana is an attractive initial case study region. The state has mandated greenhouse gas emission reduction goals, including net-zero by 2050 (Office of the Governor of Louisiana, 2022), and its Climate Action Plan (Louisiana Climate Action Plan, 2022). These plans call for incentivizing industrial CCS hubs. Two-thirds of Louisiana's 200+ million tonnes per year of emissions are of industrial origin (the U.S. average is 17% [Dismukes, 2021]), and many of the emitting facilities are clustered in the southern part of the state (Figure 1.1). This part of the state is where the subsurface geology is among the most prospective anywhere in the country for long-term underground CO<sub>2</sub> storage (Roberts-Ashby et al., 2014). These are favorable conditions for establishing CCS hubs, and some already-published studies (Global CCS Institute, 2020; Dismukes et al., 2019) provide a useful starting point for the Energy Systems Analysis group's work.

**Figure 1.1.** Louisiana industrial CO<sub>2</sub> emitters and potential biomass residue availability. Individual facilities with CO<sub>2</sub> emissions of 25,000 tonnes per year or more in 2019 are shown (U.S. EPA, 2022). A total of 180 such facilities collectively emitted 121 million metric ton (MMT) in 2019. The distribution of potential future residue-biomass energy feedstocks by parish (county) is also shown. The degree of shading reflects the U.S. DOE (2016) estimate of potential availability of residues of agricultural and forestry operations having roadside costs of \$100/tonne or less. Total residue availability is 7 MMT/y (dry weight).



Additionally, Louisiana has significant agricultural and forestry activities that could source biomass feedstocks for future BECCS facilities. There are an estimated seven million tonnes per year of residue biomass potentially available in Louisiana for energy use with no land-use change from today (Figure 1.1). Currently there is also a net annual addition of tree biomass on Louisiana's extensive forested lands (Office of Forestry, 2020). In 2018 (most recent data) this was about seven million tonnes (U.S. Forest Service, 2020). To put these figures in perspective, the carbon in seven million tonnes of biomass is equivalent to about 13 million tonnes carbon dioxide (tCO<sub>2</sub>).

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The research is at an early stage. It includes (1) data collection to characterize and map existing CO<sub>2</sub> emission sources, biomass resources, and potential CO<sub>2</sub> storage reservoirs; (2) establishment of a biogenic carbon accounting framework, especially for forest-derived biomass; (3) techno-economic assessment of CO<sub>2</sub> capture retrofits for a variety of existing process technologies and plant configurations, as well as of future BECCS power, hydrogen, or liquid fuels production; and (4) development and application of methodologies for cost- and risk-optimized design of integrated CCS hubs.

In addition to support from CMI, ExxonMobil, Deloitte, and Weyerhaeuser are all interested in engaging with and co-funding the effort, and these relationships are now being formalized. Methods and modeling tools developed in this initial two-year effort will facilitate future CCS hub analyses for other regions of interest across the U.S., including the Ohio River Valley, the Upper Midwest, and California's Central Valley, among others.

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# Bridging the Gap Between CCS Ambition and Reality

PRINCIPAL INVESTIGATOR: CHRIS GREIG

## At a Glance

Most integrated assessment and other macro-scale energy system models find that widespread carbon capture and storage (CCS) at very large scales is crucial to achieve ambitious CO<sub>2</sub> reduction goals. However, these models assume that abundant low-cost geological storage is available to meet all needs. This research presents the contrasting view that storage capacity uncertainty could seriously hamper the pace and scale of CCS deployment, especially in developing Asian economies. This storage capacity uncertainty leads to “chicken-or-egg” challenges that deter investment. The implications for emissions reduction goals and the role that CCS should play in a net-zero future warrant more attention.

## Research Highlight

Integrated assessment and other macro-scale energy systems optimization models (IAMs) for exploring decarbonization pathways are influential in shaping international, national, and subnational energy and climate policy. Most such models indicate very large-scale deployment of CCS in all major emitting nations. For example, in IAM scenarios which limit warming to 1.5 °C, median CCS deployment reaches 11Gt CO<sub>2</sub>/y by 2050 and around 13-23Gt CO<sub>2</sub>/y of CCS by 2100 (Huppmann et al., 2018). At a national scale, Princeton’s *Net-Zero America* study featured at least 1 Gt/y of CCS across all but one scenario that expressly precluded CO<sub>2</sub> storage (Larson et al., 2021). Notwithstanding these modeling insights, there remains disagreement among academics, policymakers, and other stakeholders about the need for CCS.

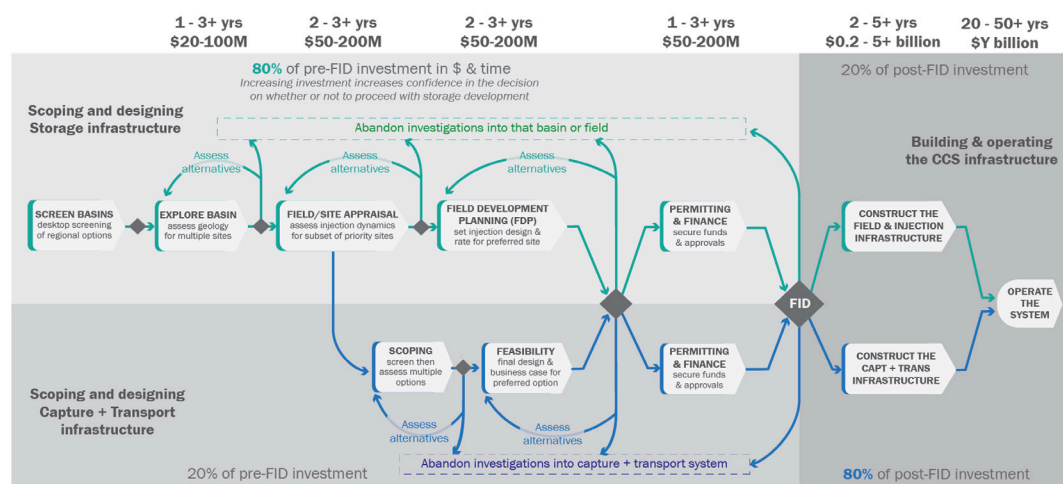
In 2021, the researchers reviewed a range of decarbonization scenarios and assigned the value of CCS in time-bound net-zero pathways to three levels: *threshold* value (without CCS, net-zero outcomes are likely to be implausible at a global scale); *commercial* value (where minimizing cost is a central goal of a net-zero transition); and *option* value (maintaining a credible CCS option mitigates the risk of other, for example, renewable heavy pathways, from faltering) (Greig and Uden, 2021). This value of CCS is driven by its versatility — providing firm low-carbon power, industrial decarbonization, low-carbon hydrogen production, and generating negative emissions via engineered removals.

The research is also examining the ongoing failure to deliver on these modeled scenarios and increasing calls to improve the feasibility of modeled pathways. CCS is just one example where real-world deployment has fallen short of the ambition in modeled scenarios.

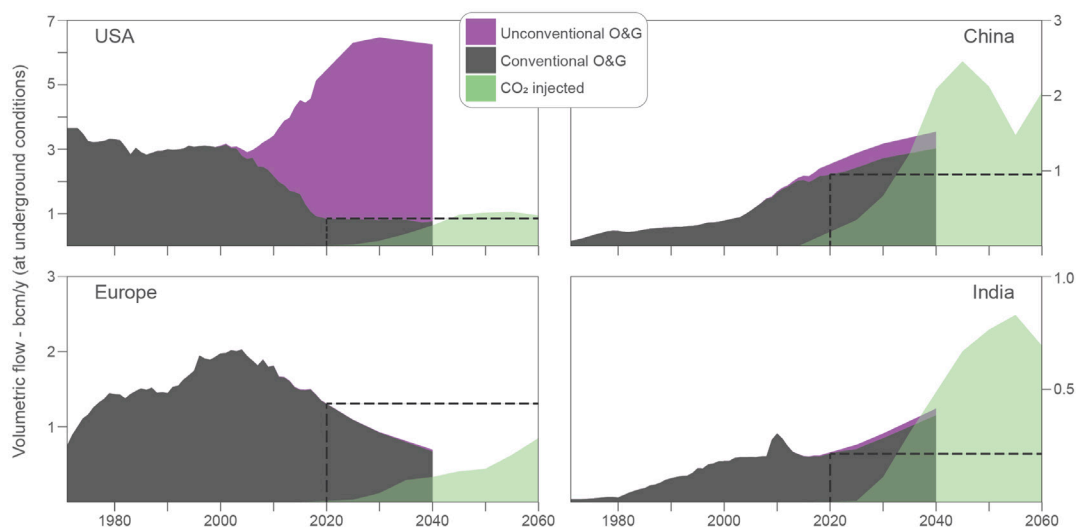
In 2021, with Postdoctoral Researcher Joe Lane, the research challenged the prevailing supposition behind most IAMs, namely, that CCS capacity can be expanded rapidly to circa gigatonne-per-year levels by mid-century, in most major emitting regions. This supposition assumes that CO<sub>2</sub> storage capacity is ubiquitous, especially in deep saline formations. This modeling perspective derives from discounted static estimates of subsurface spore volume. Investments in CCS, on the other hand, require confidence in accessible, dynamic (injection rate-based) capacity estimates. The latter is much more uncertain and heterogeneous within and across regions. Reducing these uncertainties requires serious site-based investment in characterization, exploration, and appraisal.

The researchers posited a conundrum for CCS (Lane et al., 2021) that creates a gap between ambitions and reality: despite the crucial value offered by CCS, its rate of expansion is constrained by a lack of investment confidence due to chicken-or-egg type challenges that are founded in uncertainties around storage capacity coupled with the threat of technology substitutes. The chicken-or-egg investment challenge is reflected in Figure 2.1. Failure to address this conundrum presents a risk to CCS deployment levels implied in net-zero targets, especially in fast-growing Asian economies with limited oil and gas production history. Figure 2.2 highlights the gap between historical oil and gas production and one International Energy Agency (IEA) scenario (IEA, 2017) for CCS in India and China.

**Figure 2.1.** Stylized representation of the CCS investment decision and asset development sequence illustrating the potential for chicken-or-egg challenges that might deter investment and constrain the rate of expansion (Lane et al., 2021).



**Figure 2.2.** Comparison of historical oil and gas production data to critique the relative prospects of meeting the regional CCS targets implied in the 2 °C scenario of the IEA's 2017 Energy Technology Perspectives (IEA, 2017; Lane et al., 2021).



The researchers are also working with collaborators at Imperial College London to evaluate the impact of regional CCS constraints on 1.5 °C-compatible scenarios using the TIAM-Grantham IAM, a version of ETSAP-TIAM (Loulou and Labriet, 2008). This research considers the effects of constraining annual CO<sub>2</sub> storage rates to the maximum historical annual oil and gas extraction rates in each region. The research suggests a need to rethink the allocation of CCS as a mitigation option across electricity, fuels, industrial, and negative emissions, with a significant shift in the optimal resource mix to 2050 (Grant et al., submitted 2021).

Looking forward, opportunities for further research include:

1. methods to develop plausible regional assessments of dynamic CO<sub>2</sub> storage capacity; and
2. methods to inform the execution feasibility of modeled CCS scenarios based on:
  - reverse-engineering the asset delivery sequence across CO<sub>2</sub> storage, CO<sub>2</sub> pipelines and CO<sub>2</sub> capture facilities to meet those targets; and then
  - reverse engineering the investment decision sequence implied by this asset delivery sequence.

Such research could help identify policy and critical infrastructure investments necessary to bridge the gap between CCS ambition and reality.



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# REPEAT Project Provides Real-time Look at Evolving U.S. Climate Policy

PRINCIPAL INVESTIGATOR: JESSE JENKINS

## At a Glance

The REPEAT Project is led by Jesse Jenkins of the Princeton ZERO Lab. It provides a detailed, “real-time” evaluation of the United States’ evolving energy and climate policies and the country’s progress on the road to net-zero greenhouse gas emissions. The Project uses a novel suite of geospatially-granular planning, modeling, and visualization tools coupled with macro-scale optimization models of the United States energy system. The goal is to publish regular, timely, and independent environmental and economic evaluation of federal energy and climate policies as they are proposed and enacted (See [repeatproject.org](https://repeatproject.org)).

## Research Highlight

The Biden Administration took office in January 2021 with a promise to pursue a “whole of government” approach to tackle climate change and cut emissions of greenhouse gases at least 50% below peak levels by 2030 and to net-zero by 2050.

Recognizing that the present moment constitutes the most significant opportunity for federal clean energy and climate policy in a decade, the Princeton Zero-Carbon Energy Systems Research and Optimization Laboratory (ZERO Lab), led by Jesse Jenkins (Department of Mechanical and Aerospace Engineering and the Andlinger Center for Energy and Environment), launched a new project to provide regular, timely, and independent environmental and economic evaluation of federal energy and climate policies as they are proposed and enacted.

The REPEAT (Rapid Energy Policy Evaluation and Analysis Toolkit) Project is a unique, public-facing research effort that provides a detailed, “real-time” look at the United States’ evolving energy and climate policies and the country’s progress on the road to net-zero greenhouse gas emissions.

The Princeton ZERO Lab leads the REPEAT Project in partnership with Evolved Energy Research and Erin Mayfield of Dartmouth College (formerly a CMI postdoctoral research associate). The Project developed a suite of geospatially-granular planning, modeling, and visualization tools coupled

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with macro-scale optimization models of the United States energy system. The REPEAT team employs these novel tools to rapidly evaluate policy and regulatory proposals at state, county, and sometimes finer resolutions. These tools also provide politically salient analyses of the impacts of the nation's changing energy infrastructure on air quality and public health, energy sector employment, household and business energy expenditures, and more. Results and publications are intended to provide independent, timely, and credible information and analysis for broad educational purposes, including as a resource for stakeholders, decision-makers, and the media.

The REPEAT toolkit reflects further development and refinement of the models and methods used in the landmark Princeton *Net-Zero America* study, which “set an entirely new standard” in energy transition modeling by offering an “unprecedented degree of clarity and granularity” in its results, according to John Holdren, former Science Advisor to President Obama and Director of the White House Office of Science and Technology Policy. The influential report was widely covered in major media, and the spatially explicit and granular results and associated data and maps have proven to be highly relevant to a wide range of stakeholders and decision makers. The impact of the *Net-Zero America* project demonstrated an appetite for more politically salient outputs from energy systems models. It also inspired and motivated the REPEAT Project, which has successfully refined, updated, and automated these detailed methods to enable evaluation of federal policies at an unprecedented pace and level of detail.

Through modeling and analysis of the Infrastructure Investment and Jobs Act (a.k.a. the Bipartisan Infrastructure Bill) and the Build Back Better Act published over the last six months, REPEAT Project has established itself as a critical resource that has helped shape policy negotiations, reporting, and public understanding of these major Congressional bills. Living up to its name, REPEAT Project published several timely analyses of each bill as they evolved, frequently publishing a new analysis within days or weeks of major milestones in progression of each piece of legislation through the Senate and House of Representatives. The Project's website at [repeatproject.org](https://repeatproject.org) also provides a detailed data portal that allows anyone to explore a wide range of quantitative outcomes under each version of the policy and to compare outcomes under each policy to benchmark scenarios or other policies. This tool puts all the quantitative results from the Project at the disposal of decision makers, media, stakeholder groups,



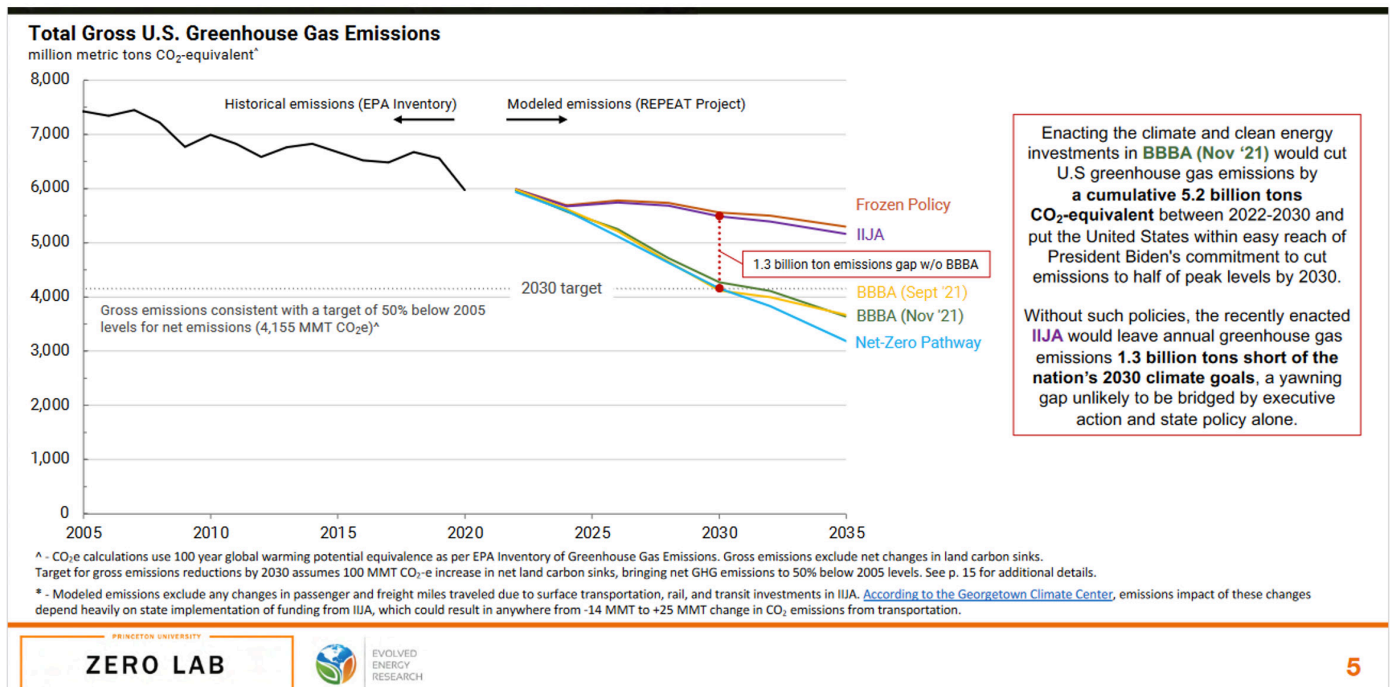
and the public.

The REPEAT Project website was accessed by over 5,000 unique visitors between October 2021 and March 2022, and the Project's reports were downloaded over 1,600 times during this period. The Project's analysis has also featured in 22 news stories to date, ranging from *The New York Times* and *Washington Post* to *Nature*, *Axios*, and *The New Yorker*.

With the Build Back Better Act stalled in the Senate, REPEAT Project is standing by to assess any Senate legislative vehicle that may emerge in the remainder of the 117th Congress. In addition, the Project will assess proposed regulations and other notable executive actions to assemble a more accurate picture of the impact of regulatory policy absent of or in conjunction with further legislation.

Finally, the ZERO Lab and REPEAT Project collaborators continue to develop novel methods for robust evaluation of energy infrastructure deployment and options. The aim is to manage trade-offs and maximize benefits across impacts on social equity, labor, air quality, land use, and other politically salient outcomes to provide improved decision support as the United States charts a course to a net-zero emissions future.

**Figure 3.1.**  
An analysis of various potential policies' impacts on US greenhouse gas emissions by 2035: Frozen Policies – policies as of January 2021; Infrastructure Investment and Jobs Act (IIJA), enacted November 2021; Build Back Better Act, passed by the US House in November 2021 but is currently stalled in the Senate; and Net-Zero Pathway, a cost-optimized pathway to reduce economy-wide U.S. greenhouse gas emissions 50% below 2005 levels by 2030 and to net-zero by 2050. (credit: Zero Lab Report Summary).



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# Nucleation and Growth of Nano-Aerosol Particles

PRINCIPAL INVESTIGATOR: IAN BOURG

## At a Glance

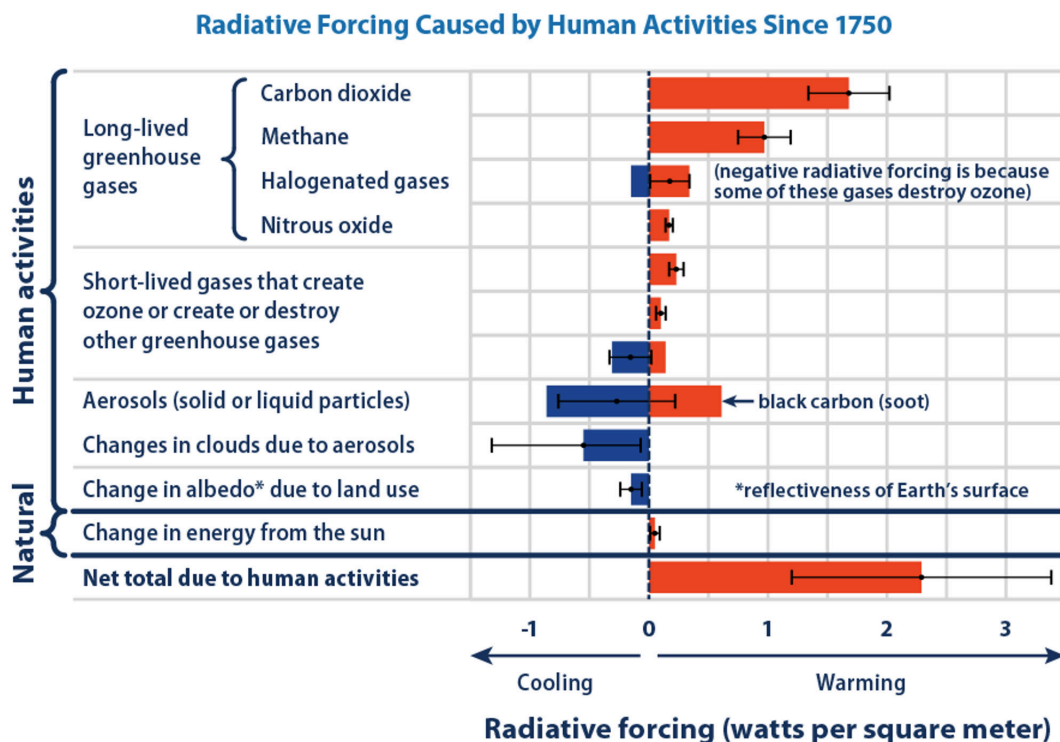
The Bourg group is working to better understand the initial stages of nucleation and growth of secondary organic aerosol particles. Atomistic-level simulations reveal a strong affinity of organic molecules for the water-air interface and a previously unknown transition between two highly distinct structures during the initial stages of nano-aerosol particle growth. These particles, which spontaneously form in the atmosphere through clustering of water, natural or anthropogenic semi-volatile organics, and ions, are key unknowns in extant climate model predictions. Since the formation of these particles can be enhanced by land-use changes, their impact on climate must be considered in carbon mitigation strategies that modify carbon and water dynamics at the land-atmosphere interface.

## Research Highlight

Aerosol particles are the second largest anthropogenic contributors to global radiative forcing, on par with methane. They are the largest source of uncertainty in current estimates of this forcing and one of the largest sources of uncertainty in climate model predictions (Figure 4.1). A key cause of this uncertainty is our limited ability to predict the nucleation and growth of secondary organic aerosol particles. In short, how do water vapor molecules, semi-volatile organic molecules, and ions present in the atmosphere spontaneously cluster into nano-aerosol particles, and how do these nano-droplets eventually grow into cloud droplets?

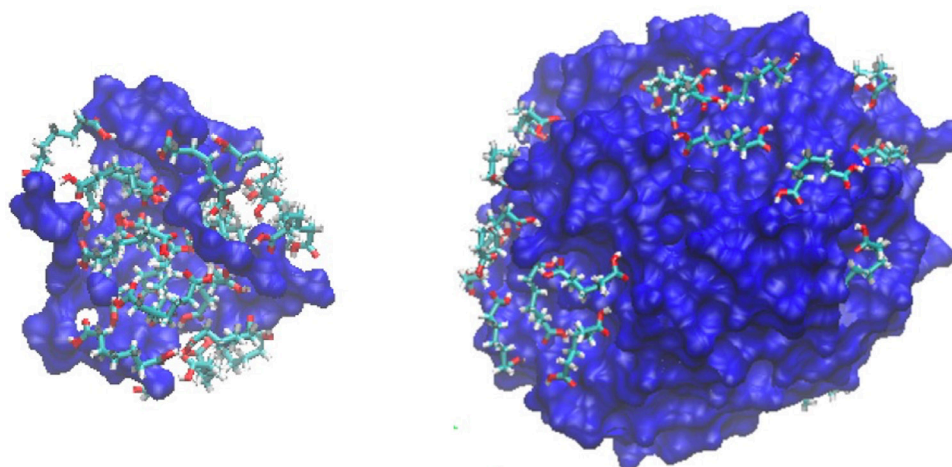
As a new effort within CMI, the Bourg group has been using all-atom molecular dynamics simulations of nano-aerosol particles carried out on U.S. Department of Energy supercomputers to re-examine fundamental theories of the microphysics of these particles. In 2021, this research was focused on two major tasks: (1) examining the properties of nano-scale droplets containing water, a natural semi-volatile organic compound (pimelic acid), and ions (NaCl) as a function of droplet size and composition (Li and Bourg, in preparation); and (2) examining the affinity of a wide range of organic molecules (a suite of 80 different natural and anthropogenic compounds) at the water-air interface (Lemay et al., in preparation).

**Figure 4.1.** Magnitude of different anthropogenic contributions to global radiative forcing according to the Intergovernmental Panel on Climate Change (IPCC, 2013). The large uncertainties associated with aerosols and their impact on clouds (8<sup>th</sup> and 9<sup>th</sup> rows) contribute most of the uncertainty in the net total due to human activities.



Preliminary results show that natural and anthropogenic semi-volatile organic compounds, which play key roles in the formation of nano-aerosol droplets, accumulate strongly at the water-air interfaces in droplets larger than ~2 nm (approximately 500 water molecules). This finding contradicts extant models of aerosol chemistry used in air quality and climate modeling, where organic molecules are assumed to be predominantly dissolved within the aqueous core of aerosol droplets. These results also reveal that nano-aerosols smaller than ~2 nm form a highly distinct structure with a water-coated organic core, versus an organic-coated water core for larger droplets (Figure 4.2). This unexpected transition may hold a key to predicting the initial stages of nano-aerosol droplet growth and to reducing uncertainties associated with these particles in air quality and climate models.

**Figure 4.2.** Molecular dynamics simulation snapshots showing representative structures of droplets formed by water and a natural semi-volatile organic compound (pimelic acid). Water is represented as a dark blue surface, while organic molecules are displayed using a “stick” representation in light blue. Simulations predict an unexpected transition, with increasing droplet size, from an organic core coated by water (left panel) to a water core coated by organic matter (right panel).





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Li, X., and I.C. Bourg, 2022. Structure, dynamics, and energetics of liquid water in nano-aerosol droplets. Proceedings of the National Academy of Sciences, in preparation.

# Soil Uptake and Methane Feedback of Atmospheric Hydrogen

PRINCIPAL INVESTIGATOR: AMILCARE PORPORATO

## At a Glance

Hydrogen ( $H_2$ ) plays a crucial role in global energy scenarios aimed at achieving net-zero. Because it is not a greenhouse gas,  $H_2$  has been touted as an alternative to fossil fuels in certain energy sectors. But the environmental consequences of perturbing the global hydrogen cycle are still largely unknown. Specifically, there are concerns around hydrogen interference with the methane ( $CH_4$ ) atmospheric sink by the hydroxyl radical (OH). To sharpen future  $H_2$  projections, the Porporato group has been quantitatively addressing the major sink of tropospheric  $H_2$ , namely the soil uptake by bacteria, and the methane feedback of  $H_2$  fugitive emissions. This research informs bp's aims of developing the  $H_2$  economy in a manner that minimizes adverse climate impacts.

## Research Highlight

Hydrogen will play a crucial role in the decarbonization of energy systems and may provide a cost-effective option to replace fossil fuels in applications where emission reductions are difficult, such as in heavy transport. However, large scale  $H_2$  production with its consequent fugitive emissions may increase the  $H_2$  atmospheric concentration, which currently hovers around 530 ppb (Novelli et al., 1999). The environmental implications of this are not yet clear.

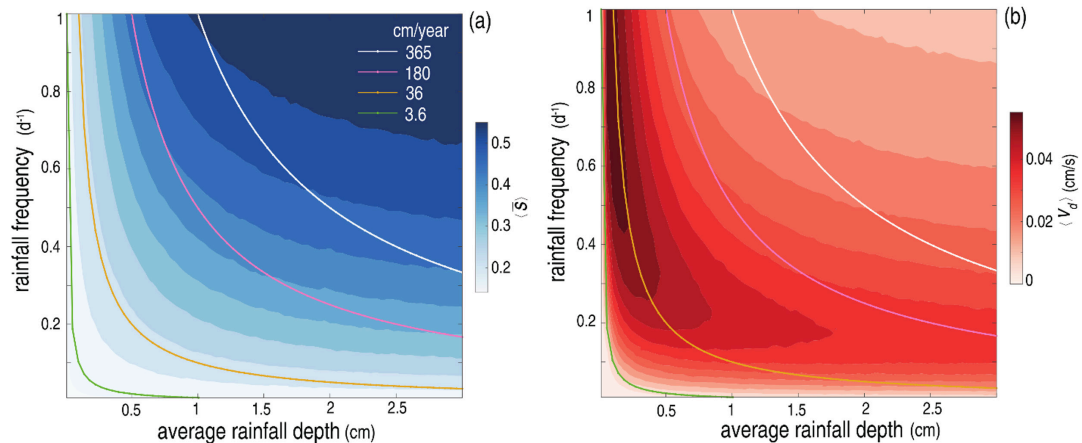
Hydrogen has an indirect global warming effect due to its interactions with other greenhouse gases (GHG) in the atmosphere, such as, for example, ozone, water vapor, and methane. Indirect radiative forcing of  $H_2$  is expected to be small compared to that of fossil fuels. However, recent global climate models (Paulot et al., 2021) have raised concerns about some possible consequences of an increasing concentration of hydrogen on tropospheric methane ( $CH_4$ ), the second most important GHG.

To improve our understanding of the global hydrogen cycle and the consequences of its possible perturbation, Porporato's research group has refined the modeling of the major sink of atmospheric  $H_2$ , namely the uptake by soil bacteria, and quantitatively addressed the methane feedback of  $H_2$  fugitive emissions.

H<sub>2</sub> uptake by soil bacteria currently accounts for nearly 80% of tropospheric removal (Ehhalt and Roher, 2009) and is widespread in all ecosystems worldwide, including extreme environments (Ji et al., 2017). The main abiotic driver of the uptake is soil moisture, which influences both the biotic (the bacteria metabolism) and the abiotic (H<sub>2</sub> diffusion through the soil) processes that govern the H<sub>2</sub> uptake.

Bertagni et al. (2021) have improved the mechanistic representation of H<sub>2</sub> uptake as a function of soil moisture and highlighted the influence of rainfall-driven moisture variability on the H<sub>2</sub> biotic consumption (Figure 5.1). Results show that limitations to the uptake are ecosystem dependent. H<sub>2</sub> diffusion through the soil generally limits the H<sub>2</sub> uptake in humid temperate and tropical regions, while biotic limitations tend to occur in arid or cold regions.

**Figure 5.1a-b.** Average soil moisture (a) and H<sub>2</sub> uptake rate (b) as a function of the average rainfall depth and frequency. Note the strongly nonlinear relationship.



An increase in global average temperature is expected to slightly favor the uptake on a global scale, while shifts in rainfall regimes can be important drivers of H<sub>2</sub> uptake changes at the local scale (Figure 5.1). Results also suggest that, although there is a greater potential of H<sub>2</sub> consumption by soil bacteria, worldwide-spread diffusive limitations will likely impede the complete offset of additional H<sub>2</sub> emissions.

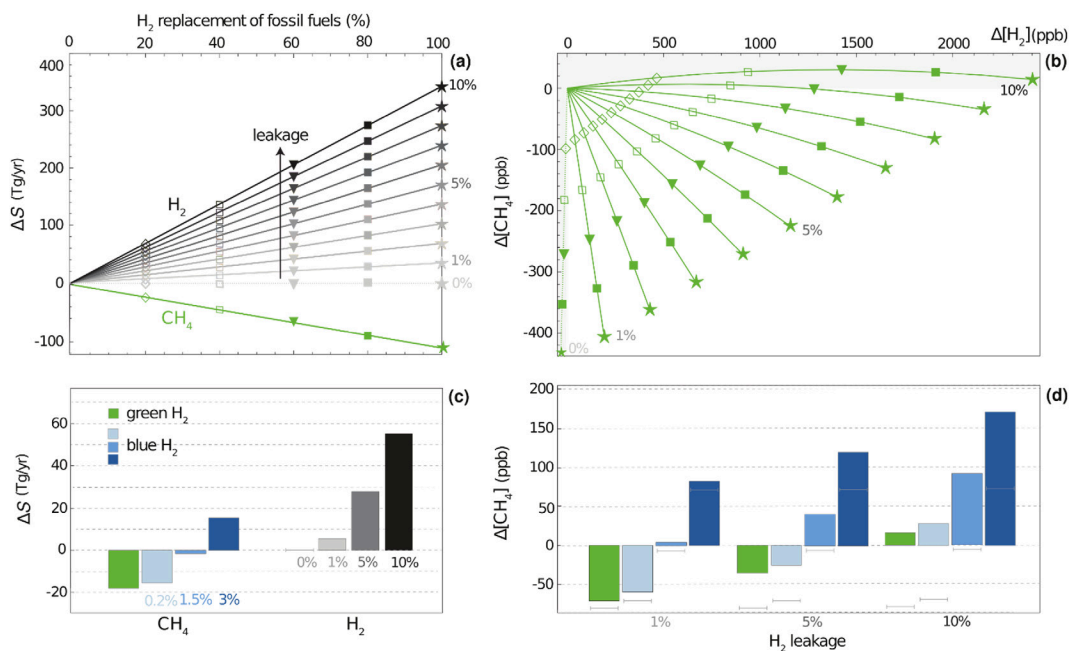
It is crucial to accurately evaluate the feedback on the methane burden because H<sub>2</sub> fugitive emissions are likely to increase the H<sub>2</sub> atmospheric concentration. Porporato's group has been addressing this problem using a box model for the coupled atmospheric system CH<sub>4</sub> - H<sub>2</sub> - OH (Bertagni et al., in preparation). The tropospheric budgets of H<sub>2</sub> and CH<sub>4</sub> are, in fact, deeply interconnected. Methane is a primary precursor of hydrogen, and the two gases share the sink by the hydroxyl radical OH. Furthermore, H<sub>2</sub> and CH<sub>4</sub> are linked at the



industrial level because most of current and near-term future  $H_2$  production comes from steam methane reforming.

Model results show that  $H_2$  emission pulses cause a small, transient growth of tropospheric  $CH_4$  that slowly decays in a few decades. Moreover, the replacement of fossil-fuel energy with renewable or low-carbon hydrogen can have very different consequences for tropospheric  $CH_4$ . This depends on the  $H_2$  production method and the amount of  $H_2$  lost to the atmosphere (Figure 5.2). For renewable  $H_2$ , tropospheric  $CH_4$  would decrease due to the fossil-fuel displacement only if the rate of  $H_2$  leakages is kept below a critical rate. This critical rate is around 8%, but with an uncertainty interval of between 5 and 11% related to how OH consumption is partitioned among the tropospheric gases and how much of atmospheric  $H_2$  is consumed by soil bacteria. For blue  $H_2$ , not only would the  $CH_4$  emissions increase, but the combination of  $CH_4$  and  $H_2$  leakages may have undesired consequences for the tropospheric burden of  $CH_4$  (Figure 5.2).

**Figure 5.2a-d.** Scenarios of a hydrogen-based economy.  $\Delta S$  is the source variation (panels a and c).  $\Delta[CH_4]$  and  $\Delta[H_2]$  are the consequent variation in tropospheric concentrations (panels b and d). (a-b) Changes in  $H_2$  and  $CH_4$  sources (a) and tropospheric concentrations (b) as a function of the  $H_2$  leakage rate and the green  $H_2$  replacement of fossil fuels energy (%). (c-d) Changes in  $H_2$  and  $CH_4$  sources (c) and  $CH_4$  concentration (d) for a 15% fossil-fuel replacement with green or blue  $H_2$  with different  $CH_4$  and  $H_2$  leakage rates.



These preliminary results call for more precise estimates of future  $H_2$  leakage rates and additional analyses with high resolution three-dimensional atmospheric chemistry models.

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# The CMI Wetland Project: Understanding the Biogeochemical Controls on Wetland Methane Emissions for Improved Climate Prediction and Methane Mitigation

PRINCIPAL INVESTIGATOR: XINNING ZHANG

## At a Glance

Methane (CH<sub>4</sub>) is the second most important anthropogenic climate forcer after carbon dioxide. Determining the importance and mechanisms of different anthropogenic and natural methane sources and sinks across temporal and spatial scales remains a fundamental challenge for the scientific community. Wetlands are dominant but highly variable sources of methane and are predicted to play a critical role in carbon-climate feedbacks. Methane emissions from these areas are shaped by a complex and poorly understood interplay of microbial, hydrological, and plant-associated processes that vary in time and space.

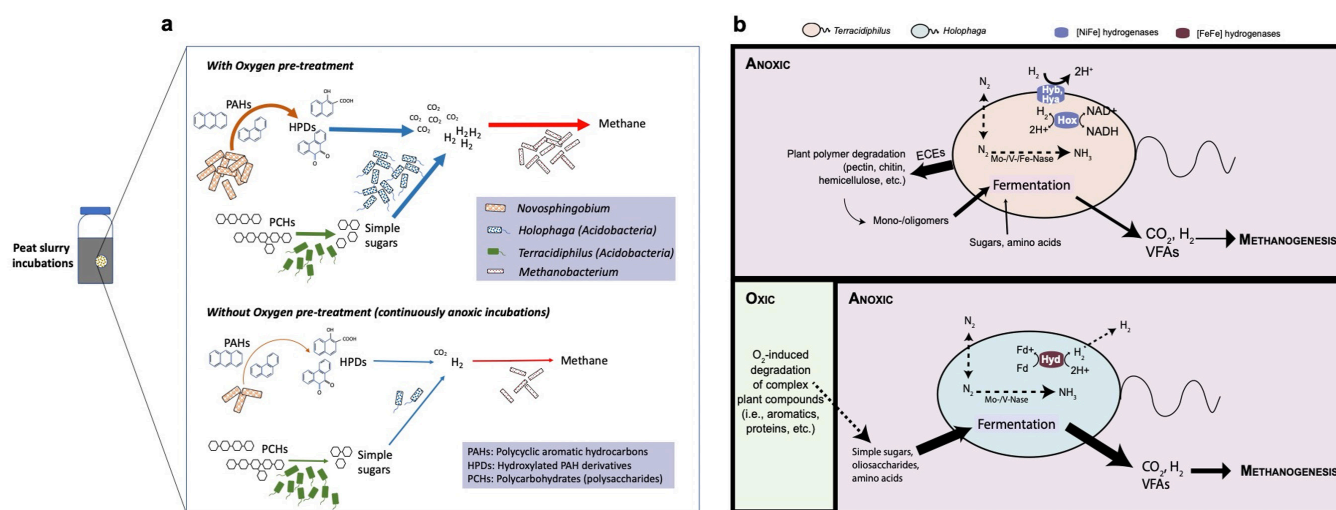
The CMI Wetland Project aims to identify the biological and chemical mechanisms that promote methane emissions from wetlands. The goals are to improve predictions of carbon-climate feedbacks and strategies of methane mitigation. A better understanding of the factors responsible for the greatest methane emissions from wetlands is crucial to bp's actions aimed at targeting this powerful greenhouse gas and thus a vital step towards a low-emissions future.

## Research Highlight

Atmospheric CH<sub>4</sub> has risen to levels roughly 150% above preindustrial concentrations due to human activities. These levels continue to rise despite a short period of stabilization between 1999 and 2006. Wetlands are geographically and biogeochemically diverse environments that together constitute the largest and most variable sources of methane to the atmosphere. CMI Wetland Project researchers are investigating the microbial, chemical, and hydrological pathways that regulate methane emissions from diverse wetland soils that vary in biogeochemical composition and hydrologic environment.

Ongoing research builds on prior CMI discoveries that transient oxygenation associated with hydrological variability unlocks a microbial "latch" on wetland carbon flow that ultimately makes mineral-poor, peaty wetlands drastically

more methanogenic (Figure 6.1a, Wilmoth et al., 2021). The researchers have pieced together fragments of genetic information from peat microbiomes to recreate microbial genomes. This has allowed the researchers to show that transient oxygenation selects for different keystone microorganisms at multiple steps of the microbial food chain underlying peat carbon conversion into methane (Figure 6.1b, Reji et al., submitted).



**Figure 6.1a-b.** (a) Transient oxygen exposure triggers a shift in microbial community succession during microbial degradation of complex aromatic peat carbon that promotes methane formation. (b) Genome reconstructions indicate functionally distinct microbial organisms with varying metabolic adaptations that are enriched under anoxic versus oxygen-oscillated conditions (Reji et al., submitted).

To better constrain the effects of hydrologically driven oxygen variability on methane emissions from a greater diversity of wetlands, current work examines wetlands along a fresh to saltwater continuum, including organic-rich peat, mineral-soil marsh, and saltmarsh sediments. Preliminary results indicate that oxygen-rich to poor transitions accelerate methane emissions from peat and marsh soils, but not from saltmarsh sediments. Geochemical variables such as pH, mineral composition, and organic carbon content are significantly different between the peat and marsh soils. This suggests that distinct microbial mechanisms underlie the observed methane emission patterns. Researchers are in the process of disentangling these biological and geochemical mechanisms. They have also started to examine the efficacy of chemical amendments like biochar in reducing wetland methane emissions.

The CMI Wetland Project has identified the influence of environmental conditions (e.g., O<sub>2</sub>, soil saturation, water table, salinity) and soil molecular form on microbial biodiversity as keys to better constrain and mitigate wetland methane emissions. The researchers urge the adoption of strategies to limit greenhouse gas emissions from natural and constructed

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wetlands, as part of land-based climate solution initiatives in freshwater wetlands (e.g., Wilmoth et al., 2021; Calabrese et al., 2021). Ongoing collaborations with the Bourg, Stone, and Porporato groups (Yang et al., 2021) address how soil mineralogy and biophysics can be manipulated to support soils-based carbon mitigation efforts.

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# Modeling Large-Scale CO<sub>2</sub> Injection in Highly Reactive Rocks

PRINCIPAL INVESTIGATOR: MICHAEL CELIA

## At a Glance

When carbon dioxide (CO<sub>2</sub>) is injected into highly reactive rocks like basalts, the injected CO<sub>2</sub> will react quickly to form new carbonate rock. This is the most stable form of geological carbon storage. The Celia group built a computer simulation tool to study this process across a range of spatial scales. Small-scale injections show fast reaction rates, on the order of months to years that are consistent with results from small-scale field experiments. However, at the large spatial scales associated with practical industrial-scale injections, large-scale mass transfer limitations lead to much longer time scales for the reactions to proceed, on the order of a century or more. The newly developed computational tool allows these and other issues to be investigated efficiently. Progress in modeling and reliably assessing the potential of carbon sequestration in deep geological formations can help accelerate bp's efforts in decarbonizing heavy industry, while seeking new energy solutions.

## Research Highlight

In carbon capture and storage (CCS), anthropogenic CO<sub>2</sub> is injected into deep geologic reservoirs. The injected CO<sub>2</sub> needs to remain underground for hundreds to thousands of years. The most secure form of underground carbon storage, called “mineral trapping,” occurs when the injected CO<sub>2</sub> reacts with minerals in the subsurface to form carbonate rock that will remain buried underground for a very long time.

A few recent small-scale field tests have shown this kind of trapping to be highly effective. For example, as part of a project called CarbFix, a group in Iceland injected CO<sub>2</sub> into basalt formations. They discovered that the injected CO<sub>2</sub> had transformed to solid rock over a fairly short time (Matter et al., 2016). In that experiment, the injected CO<sub>2</sub> was first mixed with water and the resulting aqueous solution was injected. In another small-scale experiment, a group in the Pacific Northwest of the United States (McGrail et al., 2017) injected separate-phase CO<sub>2</sub> into a basalt formation and also reported significant reactions. These examples demonstrate the great potential mineral trapping holds for the permanent storage of CO<sub>2</sub>.

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Several issues need to be addressed before this method can be adopted in practice. One is the issue of scale: Do the same fast reaction rates apply to more realistic large-scale injections, or are other rate-limiting processes involved at larger scales? Another issue is the characterization of the rock mineralogy and reaction kinetics: How much uncertainty is there in the definition of these reactions? And will the precipitation of new rock mass in the subsurface clog the pore space and render the injection system ineffective?

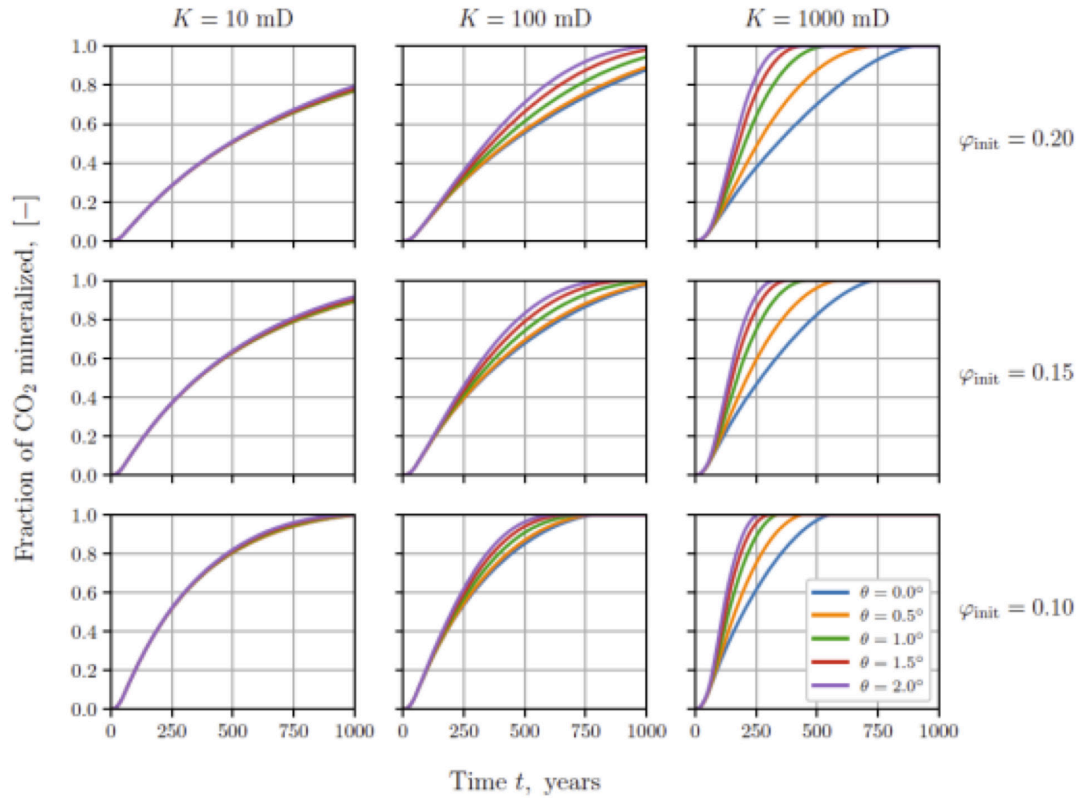
To answer these and other questions, Ph.D. student Tom Postma built a new computer simulation tool that combines earlier work in the Celia group on modeling large-scale two-phase fluid flow in the subsurface (Nordbotten and Celia, 2006, 2012; Gasda et al., 2011) with a flexible geochemistry solver that accommodates a broad range of mineral reactions. The code maintains the efficiency of the original formulations from Nordbotten and Celia while enhancing mass tracking and including different kinds of geochemical reactions. Details of the methodology have been published recently in Postma et al. (2021, 2022).

One key finding is that scale matters. The researchers ran simulations with realistic injection rates (in the order of one million tonnes of CO<sub>2</sub> per year) and varied parameters like porosity, permeability, dip angle, and reactive surface area. Small-scale injections show that large fractions of the injected CO<sub>2</sub> react on a time scale of months to years. Large-scale injections, by contrast, show that reactions are limited by the much slower mass transfer between regions containing separate-phase CO<sub>2</sub> and those containing only brine. An example of these results is shown in Figure 7.1, where the amount of CO<sub>2</sub> stored as carbonate rock is shown as a function of time. Even the fastest reaction cases show century time scales for the reactions to proceed.

Another key finding is that reductions of porosity can take place, with the largest changes found in the vicinity of the injection wells. While this could pose some risk of excessive permeability reduction, early results suggest that the porosity reduction, which is not more than a few percent (that is, reduction of porosity from, say, 15% to 13%), may not be enough to cause pore-space clogging. The actual effect on permeability is especially difficult to assess in basalts, but a first, simple estimate is a reduction of about a factor of 2. This reduction tends to be offset by the reduced resistance to flow associated with CO<sub>2</sub>.

The most sensitive parameter in these simulations is also the most uncertain: reactive surface area. The researchers have taken a range of values from the literature and included them in a wide range of simulations. The researchers are also in the process of investigating different mineralogies, such as basalts from different parts of the world, to determine what characteristics are most effective for mineral trapping. Upcoming publications will report on these findings.

**Figure 7.1.** Fraction of the total amount of CO<sub>2</sub> trapped as carbonate minerals over time, for injection into reservoirs of varying porosity, permeability, and dip angle.



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# Improving Forecasts of How Biodiversity Responds to Climate Change

PRINCIPAL INVESTIGATOR: JONATHAN LEVINE

## At a Glance

To accurately assess the biodiversity benefits of slowing climate change through land-based climate solutions, the Levine group is challenging key mathematical assumptions in the leading biogeographic modeling tools for forecasting biodiversity response to climate and developing solutions critical for their accurate implementation. Reliably gauging the impact of mitigation initiatives on biodiversity is vital to current bp efforts towards a sustainable energy world.

## Research Highlight

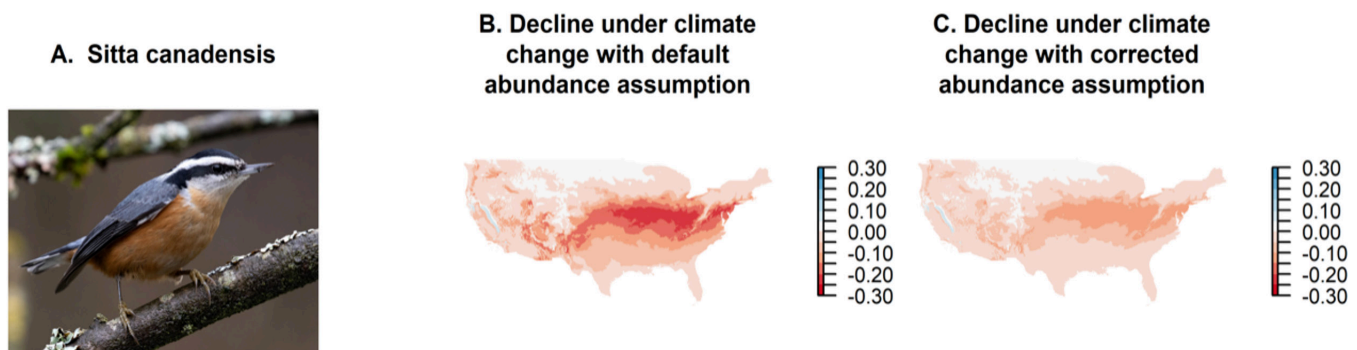
Land-based climate solutions have great potential to slow climate change and will thereby benefit global biodiversity. Countering this benefit, however, are the biodiversity impacts of the land use change associated with some of these solutions, including for example, afforestation or bioenergy with carbon capture. Weighing the costs and the benefits of land-based climate solutions for global biodiversity therefore requires accurate predictions of how climate change impacts biodiversity.

“Species distribution models” are ecologists’ best tools for forecasting biodiversity responses to climate change. They underlie nearly all research quantitatively predicting global biodiversity under a future climate. These models take known occurrences of a given species today, align those occurrences with current climate and other environmental variables, and then explore how the species’ distributional range will shift with changing climate variables.

Researchers can predict how the diversity of species at each geographic location changes with future climate when this approach is repeated for many taxa. As powerful as these approaches are, research in the Levine group has shown that key mathematical assumptions in these models are undermining our ability to draw meaningful conclusions, leading to erroneous estimates of biodiversity responses to climate change.

The Levine group has found that species distribution models accurately represent the relative suitability of different points in space for a given bird species. However, these same models struggle to accurately predict the species' true presences and absences when compared to predictions based on a detailed survey of hundreds of bird species with known presences and absences. This is because in the absence of outside information on species' abundance across the landscape, most implementations of these models simply assume a default value for species abundance.

The Levine group's research, however, shows that this default assumption greatly inflates the response of bird species' ranges to climate change, and ultimately, the overall biodiversity response to climate. Finally, their work shows that species distribution models can more accurately reflect real-world observations by using an alternative metric of biodiversity change, one that characterizes multiplicative rather than additive changes and proves insensitive to assumptions about species abundances. Adopting this approach will allow investigators to more accurately assess the benefits of slowing climate change for biodiversity, information critical to assessing the benefits of emissions reductions and land-based climate solutions against potential costs.



**Figure 8.1A-C.** Response of *Sitta canadensis* to climate change across its United States range with the default assumption for species abundance in species distribution models versus the correct abundance assumption based on known presences and absences.

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# The Efficiency of Enhanced Weathering

PRINCIPAL INVESTIGATOR: AMILCARE PORPORATO

## At a Glance

Enhanced weathering (EW) is a negative-emission technology that holds the potential of alleviating the acidification of soils and natural waters. It is a carbon capture process designed to enhance and accelerate the chemical weathering of natural minerals that, when dissolved, remove carbon dioxide (CO<sub>2</sub>) from the atmosphere and store it in natural waters. However, because the precise characterization of EW efficiency is still little understood, Porporato's research group has been working on quantifying the Alkalinization Carbon Capture Efficiency (ACCE) of any mineral dissolution in various natural waters. The findings provide an important step forward in the identification of suitable environmental conditions for EW applications, better quantification of EW carbon sequestration potential, and important context for bp's plans for natural climate solutions.

## Research Highlight

Enhanced weathering (EW) has been gaining attention as a promising geoengineering technology with large potential for CO<sub>2</sub> removal and limited technological requirements (Beerling et al., 2020). EW consists of spreading finely ground alkaline minerals in environments where the mineral dissolution might be favored, such as, for example, in the acidic soils of croplands and forests. In these environments, the minerals would also counteract soil acidification and promote biomass growth with the addition of important biological macronutrients. Some of the mineral dissolution products would be transported along the hydrologic cycle to surface freshwaters and, eventually, the ocean, mitigating ocean acidification and stably sequestering atmospheric CO<sub>2</sub> for geological timescales (Renforth and Henderson, 2017).

The transfer of CO<sub>2</sub> from the atmosphere to water is triggered by alkalization, or an increase in water alkalinity, which is caused by the mineral dissolution. However, whether CO<sub>2</sub> transfer occurs depends on the water's chemical conditions in which the minerals dissolve. Although this has long been recognized (Hartmann et al., 2013), the conditions that discriminate between efficient and inefficient carbon capture is currently little understood and lacks an objective quantification.

Using principles of aqueous chemistry, Porporato's group has derived an analytical factor that quantifies the increase in the Dissolved Inorganic Carbon (DIC) in the water solution in response to a small variation in water alkalinity (Alk). The factor, referred to as Alkalinization Carbon Capture Efficiency (ACCE), enables an exact definition of the alkalinization carbon capture efficiency of any mineral. This, in turn, is indicated as ACCEM, which quantifies the amount of  $\text{CO}_2$  captured per molecule of mineral dissolved (Bertagni and Porporato, submitted). Figure 9.1 shows that ACCE and ACCEM strongly depend on the water pH.

**Figure 9.1.** Alkalinization Carbon Capture Efficiency (ACCE) as a function of the water pH for freshwater and seawater in equilibrium with the atmosphere. ACCEM is the ACCE of a specific mineral.

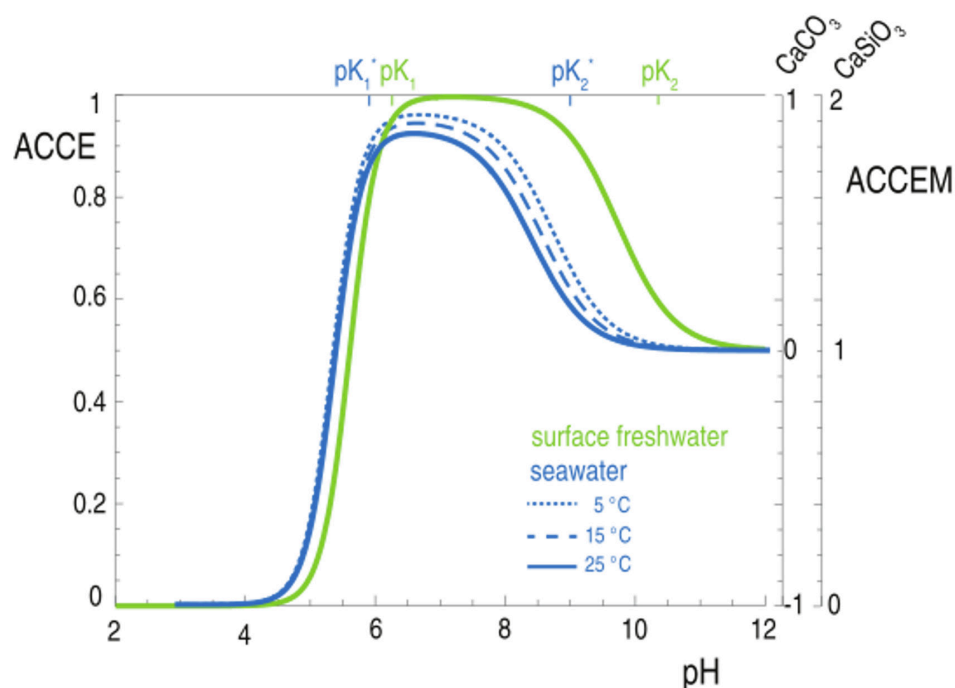
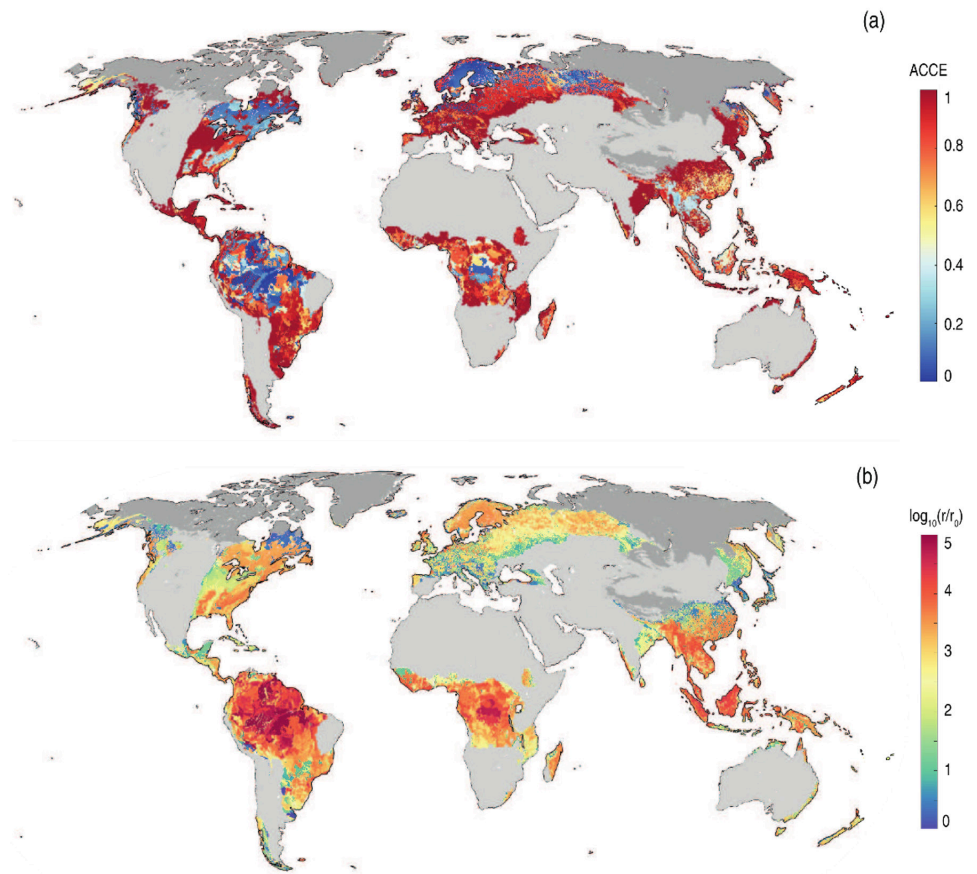


Figure 9.2a reports ACCE evaluation for the world's topsoils that are sufficiently wet and warm for EW applications. ACCE is maximized in mildly acidic or alkaline areas (e.g., Central Europe, Eastern Asia) and is minimized in very acidic soils (e.g., Scandinavia, central Amazon). Mineral dissolution rates are higher in very acidic soils (Figure 9.2b), resulting in an important trade-off between carbon capture efficiency and enhanced chemical dissolution. This means that, in acidic soils, either the EW application promotes a large alkalinity perturbation that raises the soil water pH to values that are favorable to carbonate formation, or the cations released by the mineral dissolution would need to reach another water solution before transferring any atmospheric  $\text{CO}_2$  to the water.



**Figure 9.2a-b.** Map of ACCE (a) and mineral dissolution rates (b) in global topsoils (0-30 cm). Dissolution rates account for temperature, moisture and water acidity and are normalized to the minimum. Dark and light grey regions are respectively too cold or too arid for EW applications. The carbon capture efficiency is lower where the dissolution rates are higher (acidic soils).



There is, therefore, a need for further research on the timescales of the mineral cation transport during the hydrological cycle and on the fraction of cations that can be lost to ecosystem uptake or soil adsorption.

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# Pacala Group

PRINCIPAL INVESTIGATOR: STEPHEN PACALA

## At a Glance

The Pacala group's CMI research in the last year has embraced a large number of topics, including the final report of the Princeton *Net-Zero America* project. The group also produced or co-produced a series of papers, including: (1) a paper on possible failure modes of President Biden's U.S. decarbonization agenda; (2) a paper on the need for separate national targets for CO<sub>2</sub> and methane emissions; (3) two papers (one in *Science*) on how fire and other disturbances work against land-based climate solutions; and (4) multiple papers (one in *Science*) that improve the capacity of climate models to represent the carbon cycle in tropical forests and other ecosystems. Although not directly supported by CMI, Stephen Pacala chaired the effort of the National Academies of Science, Engineering, and Medicine that produced a peer-reviewed policy manual for a U.S. transition to a net-zero economy, which extensively used the *Net-Zero America* report and was a primary reason that CMI initiated the *Net-Zero America* effort in the first place.

## Research Highlight

The Pacala group's work over the past two years illustrates how curiosity-driven research by CMI continues to produce applied dividends. Over its history, CMI has contributed to NOAA's Earth System Model (ESM) that predicts climate. The Pacala group developed the fundamental equations that govern the terrestrial carbon and hydrologic cycles in the model, which were implemented by the team led by Elena Shevliakova at NOAA's Geophysical Fluid Dynamics Laboratory, some of whose work was also supported by CMI. That model and other ESMs that use these equations spontaneously predict the coexistence of plant species. This is interesting because both biodiversity and climate are imperiled. These problems might be addressed within the same modeling system, including threats to biodiversity from climate change.

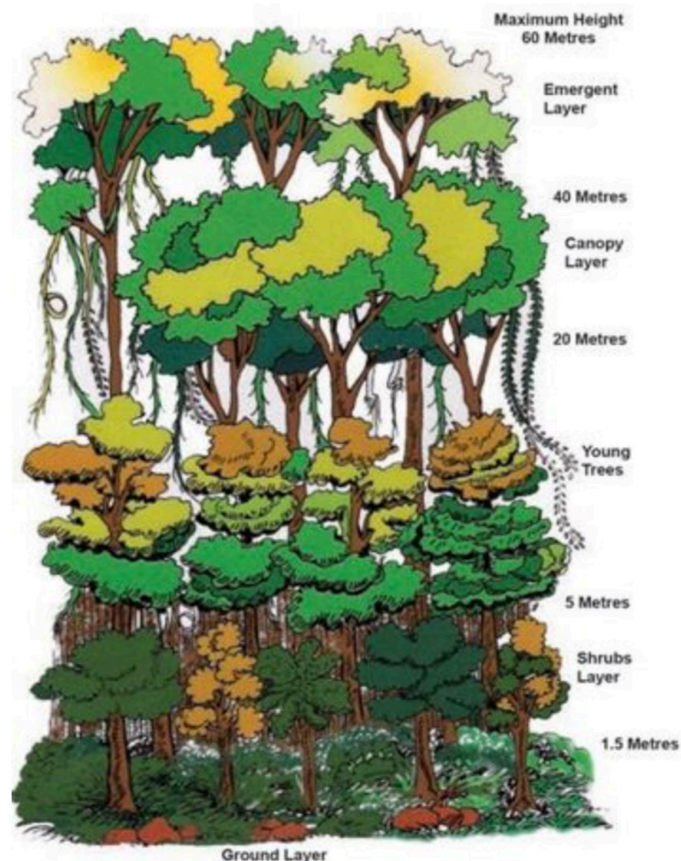
Matteo Detto, Jonathan Levine and Stephen Pacala developed mathematical methods to examine this question in a simplified version of the land model, which retains its critical features. This work was published in a paper in *Ecological Monographs* this year. One of the analyses examined the so-called shade tolerance tradeoff, which is thought to underpin successional diversity in forests. Tree species are arrayed along an axis that separates those that grow quickly in

full sunlight but die quickly in shade from those that grow slowly in the sun but survive in the shade. The researchers showed that this tradeoff can maintain the coexistence of a theoretically infinite number of species.

## Relevance to bp

Some scientists argue that forest carbon storage would be maximized by offsets projects that promote rapidly growing species, so that carbon is stored quickly. Others advocate trees capable of attaining very large size, which maximizes the total amount of carbon that will ultimately be stored. Giant, rapidly growing, but shade intolerant trees, like members of the tropical genus *Ceiba*, offer both attributes – rapid growth and large total carbon storage. The analysis shows that stands composed of both these giants and slow growing shade tolerant species will store more carbon than either type in pure stands. This is because the shade tolerant species form an extensive carbon-storing subcanopy beneath the giant species (Figure 10.1). More surprisingly, when a multispecies forest is co-managed for biodiversity and carbon storage, the shade tolerant species allow the forest to store carbon after a disturbance that destroys the canopy trees, because their high low-light survival means that many are waiting in the understory to rapidly restore the canopy. Thus, the biodiverse forest will both store more carbon and increase resilience by restoring lost carbon faster after disturbance.

**Figure 10.1.**  
Diagram of a  
multilayered tropical  
forest with giant  
shade intolerant  
species on top and  
adult shade tolerant  
species and young  
trees beneath.  
([logicalclass.com/  
contant/view/1040](http://logicalclass.com/contant/view/1040))



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# Future Fires Compromise Amazon Forest Resilience to Climate Change

PRINCIPAL INVESTIGATOR: ELENA SHEVLIAKOVA

## At a Glance

Assessments of alternative mitigation strategies to limit the impact of global change increasingly rely on simulations of Earth System Models (ESMs). In the tropics, a major biodiversity refuge and a net sink for anthropogenic carbon dioxide (CO<sub>2</sub>) emissions, ESMs consistently project that forests will thrive through the century due to CO<sub>2</sub> fertilization. In contrast, ecological models warn about a potential catastrophic forest loss under future drying conditions. A team of CMI researchers from the Pacala group and NOAA-GFDL used a state-of-the-art ESM to assess the impact of global change on tropical forest dynamics under alternative emission scenarios. Their ESM accounts, for the first time, for complex ecological mechanisms and large-scale biophysical forcing of vegetation dynamics. This research has broad consequences for the monitoring and management of tropical forests and opens new avenues for the design and implementation of carbon mitigation strategies. Of particular relevance to bp's natural climate solutions initiatives, this work informs the stability of carbon mitigated through avoided deforestation in tropical regions.

Land Model LM4.1 development and testing were guided by extensive analyses of global databases and of more than 30 years of continued study and monitoring at Barro Colorado Island (BCI), Panama.



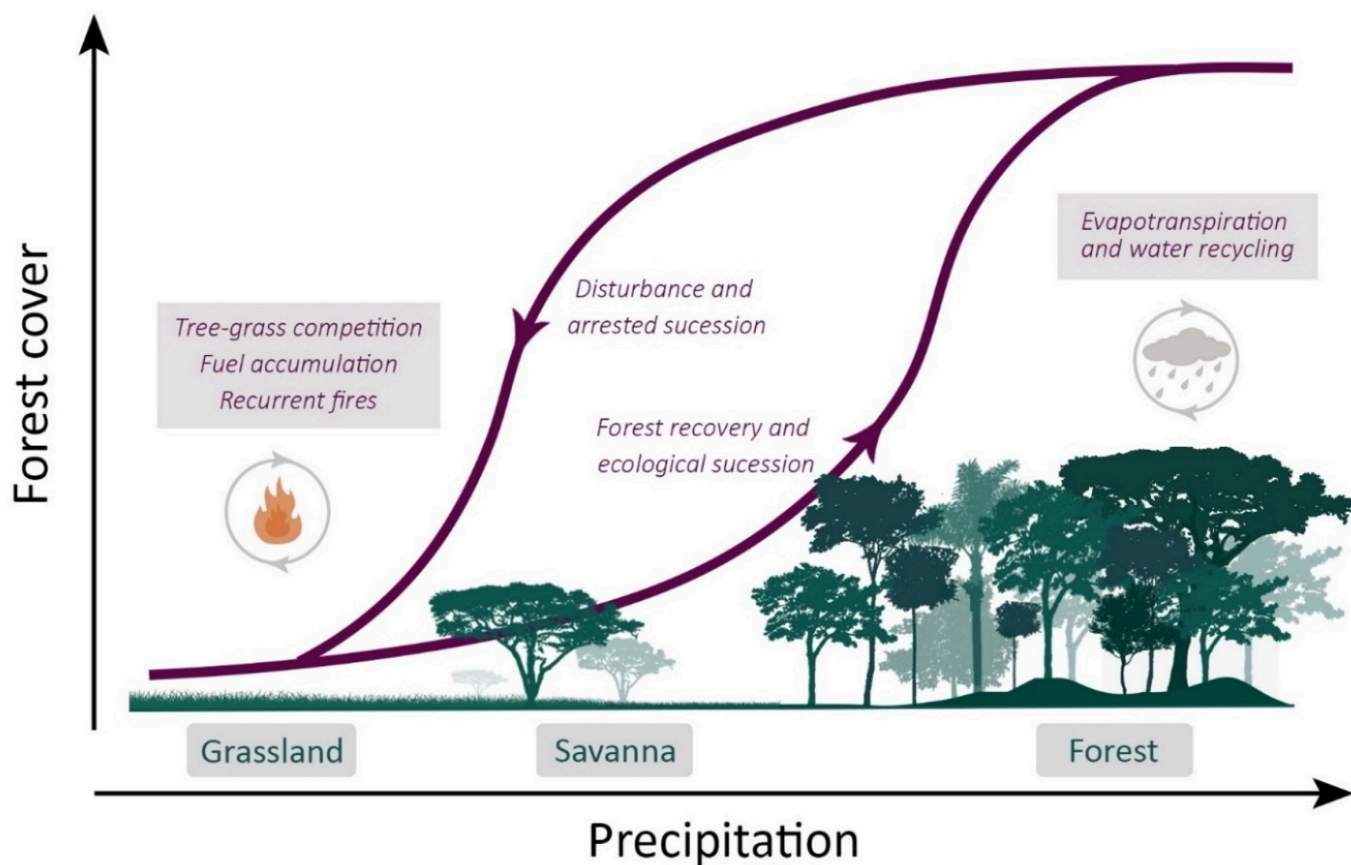
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## Research Highlight

The scientific consensus about the future of tropical forests remains unsettled. One body of literature, rooted in field observations and ecological theory, predicts that the Amazon rainforest will cross a tipping point in a few decades. As climate change brings more frequent extremes such as droughts and fires, increases in tree mortality and arrested recovery may trigger a rapid transition to grass-dominated savanna ecosystems. On the other hand, modeling studies based on Earth System Models (ESMs) come to the opposite conclusion, and consistently predict that tropical forests will thrive through the century.

To reconcile these opposite views, researchers in the Pacala group working in collaboration with climate modelers from NOAA's Geophysical Fluid Dynamics Laboratory (GFDL), have developed an ESM with realistic fire, height-structured competition and subgrid-scale heterogeneity. Such an approach enables the model to simulate the successional mosaic in ecological models of savannas, coupled to large-scale, climate-vegetation interactions. Simulation experiments with GFDL-ESM4 project that, under the extreme emission scenario SSP5-8.5, Amazon forests may begin to convert to savanna before mid-century as a consequence of increased forest fires. Projected fires resemble contemporary responses to dry conditions associated with El Niño Southern Oscillation and the Atlantic Multidecadal Oscillation, exacerbated by an overall decline in precipitation. Following the initial disturbance, grassland dominance promotes recurrent fires and competitive exclusion, which lead to an arrested successional state that prevents forest recovery and impairs the tropical carbon sink.

The effort of CMI and NOAA-GFDL researchers provides a unique tool to understanding the interaction of forest management with the dynamics of disturbance and successional recovery in forest ecosystems. Climate-induced tree mortality due to fires emerges as a potential key driver of forest damage during this century, prompting the inclusion of fire disturbances and height-structured competition in other ESMs. The team continues working to improve the ability of ESMs to assess the impact of meteorological extremes on global forests and the potential onset of abrupt transitions in land ecosystems.



**Figure 11.1.**

Emergence of alternative states and hysteresis in the structure of tropical vegetation along a gradient of water availability. The schematic highlights key mechanisms implemented in the dynamic land model LM4.1 embedded in ESM4.1. Low precipitation regimes favor the dominance of grasslands and savannas where seasonal fuel accumulation promotes recurrent fires that keep a state of arrested succession. At the other extreme, high precipitation regimes converge toward a high tree cover state where the closed tree canopy inhibits grasses, reduces evaporative water loss and increases transpiration to enhance moisture recycling at regional scales. Fire and humidity feedback mechanisms reinforce the resilience of each state and result in their coexistence at intermediate precipitation levels, where the dominant formation becomes contingent to past conditions. After a string of wet years, trees may be able to displace grasses, form a closed canopy and reach a new alternative equilibrium. As conditions become drier, a closed forest canopy resiliently keeps humidity and prevents its own collapse until disturbances like fires prompt an abrupt transition to the low cover state.

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# Diverging Fate of Oceanic Oxygen Minimum Zone and Its Core Under Global Warming

PRINCIPAL INVESTIGATOR: LAURE RESPLANDY

## At a Glance

Global warming and anthropogenic activities are contributing to a loss of oxygen in the world's oceans. However, it is still unknown if this systematic deoxygenation will expand oxygen minimum zones (OMZs). These are areas where low oxygen levels threaten marine life and perturb the carbon and nitrogen cycles, potentially acting as an amplifying feedback on climate change. The Resplandy group uses the latest generation of climate model projections to evaluate how OMZs will evolve in the future, a key step to anticipate impacts on ecosystems, ecosystem services (e.g., fisheries), and greenhouse gas emissions. This work increases our understanding of an important oceanic impact of climate change that had many in the scientific community profoundly worried. It shows that, while serious, expanding oceanic dead zones do not represent the kind of tipping point that might significantly alter bp's plans for the energy transition.

## Research Highlight

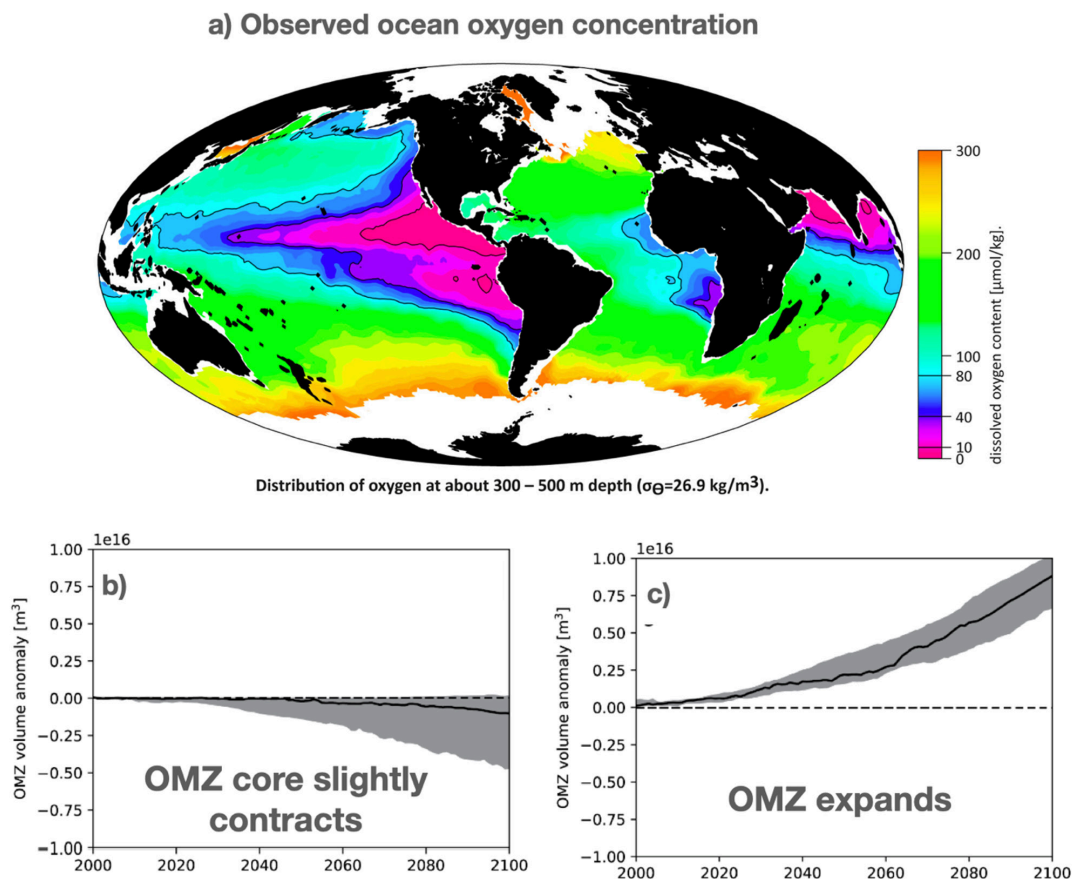
The ocean has lost dissolved oxygen ( $O_2$ ) in response to global warming in the past 50 years (Schmidtko et al., 2017). A serious threat of this systematic ocean deoxygenation is the expansion of tropical oxygen minimum zones (OMZs) located in the subsurface ocean (Figure 12.1a), such as the already vast OMZ that is currently expanding over the tropical Pacific Ocean. Uncertainties in the spatial and temporal evolution of the OMZ severely restrict our ability to anticipate its ecological, climatic, and societal impacts (Resplandy 2018; Busecke et al., 2019). Low oxygenated OMZ waters (typically  $O_2 < 120 \mu M$ ) delimit the optimum habitat of valuable commercial fishes, such as sardines and billfishes (Bertrand et al., 2011), while the core of the OMZ, where  $O_2$  concentrations are the lowest (typically  $O_2 < 20 \mu M$ ), will determine the rate of production of nitrous oxide, a potent greenhouse gas (Bianchi et al., 2018).

In this project, the Resplandy group leverages the latest generation of Earth System Models (CMIP6 ESMs) to examine the future evolution of the tropical Pacific OMZ and its core under the warming scenario SSP8-5.8 ("business as usual").



For the first time, the researchers found a consistent response across models, which was lacking in prior work. ESMs project a robust expansion of low oxygenated waters (Figure 12.1c), but a slight contraction of the volume of the OMZ core waters (Figure 12.1b). While thermal changes lead to a relatively homogeneous oxygen loss (warming waters reduce oxygen solubility), the team found that non-thermal changes dictate the switch from core contraction to broader OMZ expansion. The OMZ expansion can be attributed to a slowing down of the tropical ocean circulation that supplies oxygen to the OMZ, while the core contraction is associated with both a change in ocean turbulent mixing and a decline in biological production and respiration (Busecke et al., 2022).

**Figure 12.1a-c.**  
(a) Global map of oxygen distribution at 300-500m water depth in the world ocean (Source: GEOMAR).  
(b-c) Projected evolution of the tropical Pacific OMZ core ( $O_2 < 20 \mu M$ ) and OMZ ( $O_2 < 120 \mu M$ ) in the newest Earth system model generation CMIP6 under the warming scenario SSP8-5.5 (multi-model median in solid, first, and third interquartile in shading).



If emissions are not curtailed, the expansion of the OMZ will yield a contraction of the habitat of numerous valuable marine species, confining them closer to the surface where oxygen levels are suitable for their metabolism, and making them more prone to overfishing. The slight contraction of the core, however, would likely have a limited impact on the production of nitrous oxide.



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# Understanding Tropical Cyclone Frequency

PRINCIPAL INVESTIGATOR: GABRIEL VECCHI

## At a Glance

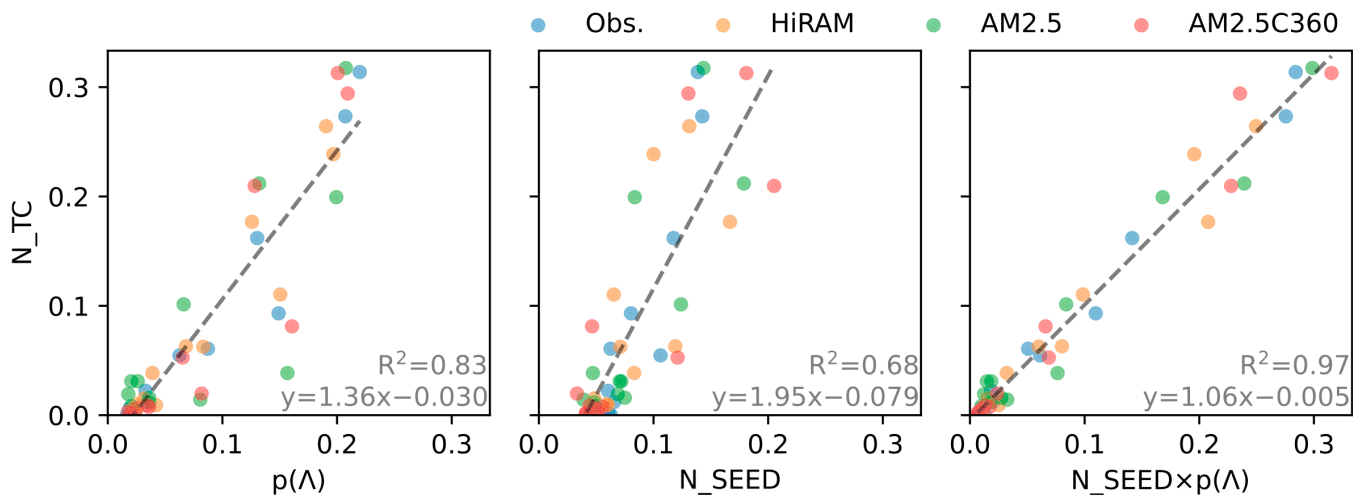
The objective of this research from the Vecchi group is to better understand all aspects and variations in the statistics of tropical cyclone (TC) activity and other climate impacts over the past few centuries, as well as the coming one. An equally salient objective is to better understand the likely range of equilibrium and transient climate sensitivity, such as how much warming to expect from a doubling of atmospheric CO<sub>2</sub>. Key tools in these studies are climate and atmospheric models. These, along with analyses of the observed record, help researchers to distinguish whether observed multi-decadal to centennial changes in TC activity have been driven by large-scale factors such as ocean temperature changes, greenhouse gases, volcanic eruptions, or the El Niño, as opposed to random atmospheric fluctuations.

## Research Highlight

Understanding tropical cyclone frequency remains a challenge for the tropical cyclones community (Knutson et al., 2021; Sobel et al., 2021). The Vecchi group has worked in recent years to build a consistent and physically grounded framework to understand the mechanisms controlling tropical cyclone frequency (Vecchi et al., 2019; Hsieh et al., 2020). Through this effort they have developed a new paradigm, which considers the change in the number of pre-tropical cyclone vortices (or “TC seeds”). The paradigm also looks at the impact of large-scale environmental conditions on the probability of cyclones arising from these seeds. Once one accounts for the distinct climate dependence of seeds and genesis probability, one can accurately predict the sensitivity of TC frequency to a wide range of global forcing. Prior to this work, the literature on TC frequency had focused primarily or solely on the role of climate on genesis probability.

The seed-probability framework for understanding TC genesis was originally developed to understand the response of TC frequency to climatic changes and to accurately capture the observed and modeled annual cycle (Yang et al., 2021; see Figure 13.1). The North Atlantic hurricane season is very narrow, with the vast majority of hurricanes occurring between August and October. However, the extremely sharp transition between the inactive and active periods during the year cannot alone be explained by the environmental

conditions that favor tropical cyclone genesis (“genesis probability” in the left panel of Figure 13.1) or the frequency of pre-TC “seeds” (center panel). However, the combined effect of variations in genesis probability and seeds yields a sharp annual cycle that accurately explains hurricane climatology across both climate models and observations (right panel). The ability of this theoretical framework to explain the annual cycle of hurricanes provides an observational test of the hypothesis, developed in climate simulations, that the combined role of seeds and genesis probability in the climate impacts hurricane frequency.



**Figure 13.1.**  
TC seeds and genesis probability together explain the annual cycle of North Atlantic (NA) hurricane frequency in observations and climate models.

Scatter plots of monthly climatology of NA TC frequency versus TC genesis probability index (left panel), frequency of vortex seeds (center panel), and the combination of both (right panel) from observations and model historical simulations from HiRAM, AM2.5, and AM2.5C360. All the quantities are normalized by their annual total. Dashed lines show the linear regression, for which the equation and the variance explained are shown on the bottom right of each panel. The sharp annual cycle of Atlantic hurricane frequency is due to the combined effect of both seeds and genesis probability (Yang et al., 2021).

A related study explores the impact of TC seeds on genesis across a broad suite of climates and climate model configurations and develops a theory to link seed frequency to large-scale environmental factors (Hsieh et al., 2022, submitted). This work has shown that inter-model spread in TC genesis sensitivity to changing climate is largely driven by differences in the response of pre-TC synoptic disturbances. The climatological changes in these disturbances can be understood in terms of changes to large-scale aspects of the atmosphere (such as the amount of energy converging in the atmosphere and the rate at which air ascends). The researchers are now working to connect observed and modeled changes in TC frequency to large-scale climatic parameters using first principles (e.g., conservation of energy, mass and momentum), in an effort to build a theoretical constraint on tropical cyclone frequency.

The Vecchi team also explored impacts of uncertainties in ocean temperature reconstructions and changes in hurricane monitoring as a way to assess past changes in hurricane activity. They found that improvements in ocean temperature

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estimates over the 20<sup>th</sup> century allow for a reproduction of multi-decadal historical tropical cyclone frequency changes with high-resolution atmospheric models. This has enhanced confidence in the models' ability to reproduce future changes in TC frequency (Chan et al. 2021). The researchers have also extended a methodology to account for the impact of past changes in hurricane monitoring on tropical cyclone frequency estimates. This has allowed the researchers to build a homogenized record of major (Category 3-5, the most destructive storms) hurricanes in the Atlantic from 1851-2020. This new homogenized record suggests that expected century-scale increases in major hurricane frequency have been masked by a combination of multi-decadal climate variability and late-20<sup>th</sup> century aerosol forcing. The Vecchi team is currently developing high-resolution climate model simulations to test this hypothesis.

In addition to developing a new understanding of the climate-tropical cyclone connection, the team have worked to review the state of the literature and highlight key open challenges. This work has appeared in two review articles, one on tropical cyclones and global warming (Knutson et al., 2021) and the other on tropical cyclone frequency more generally (Sobel et al., 2021).

## Relevance to bp

bp has long been interested in tropical cyclone risk because of the vulnerability of its coastal and offshore infrastructure, and because increases in the severity or frequency of tropical cyclones is an important driver of public opinion in support of the energy transition. The most interesting conclusion of the highlighted studies is that the 20<sup>th</sup> century increase in the frequency of major hurricanes would now be higher if not for past and ongoing anthropogenic aerosol emissions (primarily from coal combustion), which will continue to decrease during the energy transition. In the near term, we should thus expect more rapid increases in the frequency of major hurricanes than would be expected because of elevated greenhouse gases alone.

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# Calcium (Ca)-Based Solid Sorbents for Low Temperature CO<sub>2</sub> Capture

PRINCIPAL INVESTIGATOR: CLAIRE WHITE

## At a Glance

White and her group are developing novel calcium (Ca)-based solid sorbents that are capable of selectively capturing carbon dioxide (CO<sub>2</sub>) from a mixed gas stream, or air, at ambient temperature. By understanding the solution chemistry during synthesis, the researchers have obtained phase pure Ca-based layered double hydroxides and demonstrated that these sorbents can selectively adsorb CO<sub>2</sub>. Ongoing efforts are focused on engineering energy-efficient regeneration methods and quantifying life cycle environmental and economic aspects. This project aligns with bp's goal of developing solutions to decarbonize the cement production process, thereby helping cities and corporations to decarbonize.

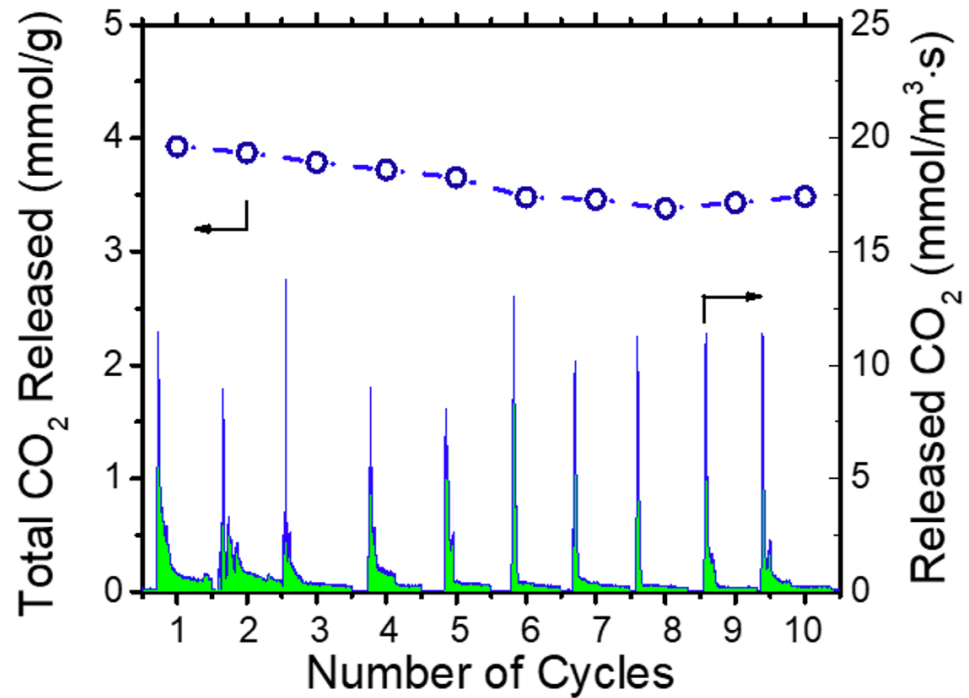
## Research Highlight

To reach net-zero by 2050, it is imperative that capturing CO<sub>2</sub> both directly from the air and at point sources become routine approaches. However, current capture technologies are comparatively expensive and have yet to be widely implemented. High costs are associated with the capture material and with the energy required for the release of CO<sub>2</sub> after capture, a process called regeneration. Solid sorbents are ideal for CO<sub>2</sub> capture, but at present there is a lack of inexpensive sorbents for low temperature capture that selectively adsorb CO<sub>2</sub> from a mixed gas stream (or air) and that require lower energy for their regeneration.

White and her group have recently begun research on layered double hydroxides (LDHs). These are a class of materials that have been assessed for intermediate temperature capture using calcined LDHs, yet low temperature capture using Ca-based LDHs remains unexplored. The White group successfully synthesized phase pure CaAl- and CaFe-LDHs for low temperature CO<sub>2</sub> capture by uncovering the complex speciation chemistry that occurs during solution-based synthesis. They have also manufactured Ca-based LDH gas filters consisting of the LDH phase deposited on carbon substrates using an electrodeposition synthesis approach.

Ongoing research is focused on determining the mechanisms and the strength of gas molecule binding (gas-solid interactions) using first principles calculations validated against experimental data. Other research explores various methods for material regeneration (i.e., release of CO<sub>2</sub> after capture), and quantifying capture capacity and cyclability.

**Figure 14.1.**  
CO<sub>2</sub> capture  
cyclability of CaAl-  
layered double  
hydroxide supported  
on carbon substrate.



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