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now, however, Bitcoin has a big problem, and it is growing fast.

- 1. Chohan, U. The double spending problem and cryptocurrencies. 2017. https://doi.org/ 10.2139/ssrn.3090174.
- 2. Nakamoto, S. Bitcoin: a peer-to-peer electronic cash system. 2008. https://bitcoin. org/bitcoin.pdf.
- 3. Hileman, G., and Rauchs, M. Global cryptocurrency benchmarking study. 2017. https://doi.org/10.2139/ssrn.2965436
- 4. IEEE Spectrum. Why the biggest Bitcoin mines are in China. 2017. https://spectrum.ieee.org/computing/networks/why-thebiggest-bitcoin-mines-are-in-china.
- 5. Huang, Z., and Wong, J.I. The lives of Bitcoin miners digging for digital gold in Inner Mongolia. 2017. https://qz.com/1054805/ what-its-like-working-at-a-sprawlingbitcoin-mine-in-inner-mongolia/.
- 6. Tech in Asia. Cheap electricity made China the king of Bitcoin mining. The government's stepping in. 2017. https:// www.techinasia.com/inner-mongoliabitcoin-mine.
- 7. Hayes, A.A. Cost of production model for Bitcoin. 2015. http://www economicpolicyresearch.org/econ/2015/ NSSR_WP_052015.pdf.
- 8. Quartz. China's Bitmain dominates Bitcoin mining. Now it wants to cash in on artificial intelligence. 2017. https://gz.com/1053799/ chinas-bitmain-dominates-bitcoin-miningnow-it-wants-to-cash-in-on-artificialintelligence/.
- 9. Song, J. Just how profitable is Bitmain? 2017. https://medium.com/@jimmysong/justhow-profitable-is-bitmain-a9df82c761a.
- 10. Digiconomist. Bitcoin Energy Consumption Index. 2018. http:// bitcoinenergy consumption.com.
- 11. Wang, J. TSMC—the world's largest chip factory—is all about crypto all of a sudden. Bitmain is buying ~20k 16nm wafers a month. That's more than Nvidia. Twitter Post. 2018. https://twitter. com/jwangARK/status/ 954429531678543872.
- 12. Morgan Stanley. Bitcoin ASIC production substantiates electricity use; points to coming jump. 2018. https://ny.matrix.ms com/eqr/article/webapp/b974be48-effb-11e7-8cdb-6e28b48ebbd3.
- 13. Tahir, R., Huzaifa, M., Das, A., Ahmad, M., Gunter, C., Zaffar, F., Caesar, M., and Borisov, N. (2017). Mining on someone else's dime: mitigating covert mining operations in clouds and enterprises. Res Attacks Intrusions Def, 287-310.
- 14. Cointelegraph. Crypto farm with 6000 miners shut down in Russia for overdue electricity bill. 2018. https://cointelegraph.com/news/ crypto-farm-with-6000-miners-shut-downin-russia-for-overdue-electricity-bill.

- 15. International Energy Agency. World Energy Statistics 2017. 2017. https://www.iea.org/ publications/freepublications/publication/ KeyWorld2017.pdf.
- ¹Experience Center of PwC, Amsterdam, the Netherlands
- *Correspondence: alex.de.vries@pwc.com https://doi.org/10.1016/j.joule.2018.04.016

COMMENTARY

Research Opportunities for CO₂ Utilization and **Negative Emissions** at the Gigatonne Scale

Arun Majumdar^{1,*} and John Deutch²



Arun Majumdar is the Jay Precourt Professor in the Department of Mechanical Engineering and Co-director of the Precourt Institute for Energy at Stanford University. He served as the Founding Director (2009-2012) of the Advanced Research Projects Agency - Energy (ARPA-E) and the Acting Undersecretary for Energy (2011–2012) under Secretary Steven Chu in the US Department of Energy. He also served as Vice Chair of the Advisory Board (2014-2017) to Secretary Ernest Moniz. Arun Majumdar has published widely on the science and engineering of conversion, storage, and transport of energy, especially nanostructured materials and devices. He is a member of the US National Academy of Engineering and the American Academy of Arts and Science.



John Deutch is an emeritus Institute Professor at the Massachusetts Institute of Technology where he has been a member of the faculty since 1970. He has served as Chairman of the Department of Chemistry, Dean of Science, and Provost. In the Carter Administration, he served as Director of Energy Research (1977-1979), Acting Assistant Secretary for Energy Technology (1979), and Undersecretary (1979-1980) in the US Department of Energy. He has been a member of the President's Nuclear Safety Oversight Committee (1980-1981), the White House Science Council (1985-1989), the President's Committee of Advisors on Science and Technology (1997-2001), and the Secretary of the Energy Advisory Board (2008-2016).



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John Deutch has published widely on technical and policy aspects of energy and the environment and has been a member of the board of directors or of the technical advisory committees of several energy companies.

This commentary outlines a framework to reduce atmospheric CO2 concentration at the scale of gigatonne per year (GT scale) and identifies research opportunities in CO₂ utilization and negative emissions to achieve this goal. Given that the largest atmosphereland carbon flux occurs via photosynthesis (\sim 120 GtC/year), research focused on increasing the fraction of carbon stored on land via plant engineering and land management in agriculture and forestry deserves increased attention. Lowering the cost of carbon capture from dilute sources requires research on advanced sorbents and reactor systems with much improved performance. With the availability of low-cost renewable energy, research is needed on pathways to convert CO₂ into cost-competitive carbonbased chemicals and fuels. Finally, research on non-traditional enhanced oil recovery that produces a net positive in carbon stored in geological formations also deserves attention. For this GT-scale challenge, a systems approach is needed to understand the impact of various measures and explore "what if" scenarios.

There are two complementary approaches to avoid the adverse impacts of increasing global temperature due to anthropogenic emissions of greenhouse gases (GHG): mitigation, which refers to measures that reduce atmospheric CO₂ concentration by reducing the rate of emissions or increasing the rate of absorption of atmospheric CO2; and adaptation, related to measures that would help adjust to changed climate with as little disruption to our lives and well-being. Currently, there are \sim 3,200 GtCO₂ (\sim 870 GtC) in the atmosphere; ¹ the total global emissions rate is ~40 GtCO₂/ year. Therefore, any measure of consequence must have a scale of at least 1 GtCO₂/year (referred to as GT scale).

What measures can we adopt today, and where should we focus our research to have this scale of impact in the future? In June 2016, former US Secretary of Energy Ernest Moniz charged the Secretary of Energy's Advisory Board (SEAB) to establish a Task Force (TF) to undertake a study and identify such research opportunities.² The TF report³ is publicly available. In the TF's judgment, the scientific community is moving too slowly to assess the prospects for alternative GT-scale options despite significant recent attention.^{4,5} The pace of deploying GT-scale options is important as well. However, as Pacala and Socolow⁶ have emphasized, technology readiness only makes the transition to deployment if proper policy measures such as on GHG emissions are in place. This article is offered as a call to action for advancing technology readiness.

Looking at the global energy system, options to reduce emissions in the electricity sector⁷ are easier than those for the transportation and the industrial sectors. Hence, negative emissions and CO₂ utilization are worth assessing as counteractive measures. Furthermore, if the atmospheric CO₂ concentration rises above any dangerous threshold even with zero net emissions, negative emissions technologies could reduce the atmospheric concentration from increasing beyond that threshold.

Figure 1 provides a framework for CO₂ source, capture, conversion, storage, and/or utilization as an economically viable product. Undoubtedly, the number of pathways makes this a complex landscape since each pathway requires numerous questions about rates, locations, amounts, costs, infrastructures, chemical form, use, re-use, and fate of carbon, all of which need to be addressed systematically. Added to this complexity is the issue of scale: It is important to understand that GT scale is a demanding target.

First, the only industries on this scale today are steel, concrete, and agriculture, as well as coal, oil, and gas. If these industries were leveraged to achieve GT-scale carbon management, they would be part of the solution. Second, if an industrial activity cannot achieve 0.1 GT scale, it unlikely it will reach 1 GT scale. Hence, we need a roadmap, for example, using cement as a possible CO₂ sink and perhaps plastics⁸ in the future. Third, any process that captures, transports, and converts ~1 GtCO₂/year will require significant amounts of carbon-free energy,9 beyond what we estimate to decarbonize the electricity sector. Fourth, GT-scale carbon management will require massive investments (~\$100 billion) and thus present significant management, work force, supply chain, and financing challenges. Fifth, an endeavor at this scale will have consequences, intended and unintended, on our biosphere, which are difficult to predict a priori. Understanding and predicting the impact must be part of any research effort. Finally, it seems inevitable that to achieve GT scale, there will need to be a charge on CO₂. We do not dwell on this issue, since the policy landscape is still evolving. Rather, we ask the question: Where can research make the biggest difference to create new technoeconomic options or significantly expedite existing ones when scaled to ~ 1 GtCO₂/year? Here are some recommendations.

Systems Approach

The carbon fluxes between the natural systems (atmosphere, land, oceans) and commercial systems (electricity/ heat, transportation, industry) are highly coupled and must satisfy the laws of nature, with the commercial **Joule CellPress**

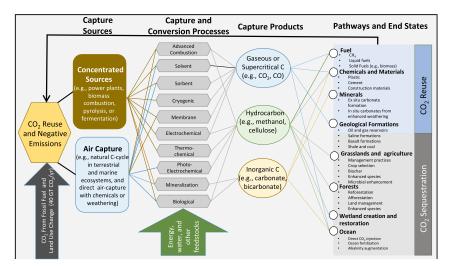


Figure 1. The Negative Emission and CO₂ Utilization Option Landscape

Options exist for the capture source, capture and conversion processes, capture product, sequestration repository, and the engineering approach to achieve negative emissions. Crosscutting research opportunities exist across the entire negative emissions landscape. Courtesy: Sally Benson, Stanford University. Note to reader: Not all pathways have equivalent climate change mitigation potential. Certain pathways place CO2 into the atmosphere, and so without negative emission, the result will be only partial decarbonization.

systems having to satisfy the imperatives of economics as well. Physics and economics both introduce feedbacks and non-linearity in the system, which could be potentially ignored at small scale, but cannot be ignored at the GT scale. This makes predictions of the overall system behavior very difficult. We need to intensify research that builds on and expands today's effort to create a constellation of systems models, beyond today's integrated assessment models of the global carbon balance, at different degrees of spatial and temporal resolutions. Such tools would guide holistic policies of carbon management, develop approaches to systematically study parametric sensitivity, and quantify uncertainties of different variables. These simulations would allow exploration of "what if" scenarios and thereby identify pathways that would produce GT-scale effects and identify performance and cost targets that would shape a research, development, and demonstration (RD&D) strategy for these technological pathways. There must also be an effort for global data collection and

analysis to validate such model predictions.

Harnessing the Natural Biological Cycle

The largest flux of carbon (~120 GtC/ year or 440 GtCO₂/year) between the atmosphere and land occurs via photosynthesis in plants (Figure 2). Roughly 2%-3% of this carbon remains stored on land for decades, while the rest is emitted back to the atmosphere. How might this natural biological carbon cycle be harnessed to absorb more carbon from the atmosphere, store more carbon on land, or use a combination of both to produce negative emissions? Could this be achieved with a positive co-benefit of increasing productivity of crops for food, bioenergy, feed, and fiber that the world will need, and thereby be of commercial value? Research using the modern tools of gene editing and manipulation holds promise to increase the photosynthetic efficiency¹⁰ and optimize crops for food, bioenergy, feed, and fiber, as well as trees for bioenergy, reforestation, and afforestation. But

this must be achieved with no marginal increase in resource inputs: fresh water, fertilizers, and pesticides. Research is also needed to identify approaches to reduce decomposition of soil organic carbon and N2O emission impact. Examples include creating roots that go deeper in the rhizosphere with higher lignin content. Such scientific progress should be complemented with no-till agriculture to stabilize soil carbon. Of course, the direct and indirect impacts of no-till agriculture on land use and productivity need to be carefully evaluated and documented.

Synthetic Transformation of CO₂

Carbon dioxide can be transformed into a variety of chemicals and fuels that have commercial value. But this requires significant carbon-free/neutral energy⁸ in the form of heat and/or electricity, which can often be the dominant cost. 11 Furthermore, these transformations need to follow one or a combination of chemical pathways-electrochemical, photochemical, biochemical, thermochemical—with sufficient efficiency and low infrastructure costs to produce market-competitive chemicals and fuels. Research is needed to reduce the cost of delivered carbon-free/ neutral exergy (electricity and hightemperature heat) with a target range below 3 cents/kWh. Fortunately, electricity generation from intermittent sources such as wind and solar are increasingly reaching this range, but it is important to achieve this without intermittency. Research on fundamentals of electro/photocatalysis is needed to identify catalysts for important redox reactions (e.g., CO₂ reduction and the O₂ evolution reaction) with low overpotentials and high reaction rates. It is also important to identify materials for thermochemical redox reactions at temperatures <1,000°C, making it compatible with today's chemical infrastructure, which is almost exclusively based on thermochemistry. Research opportunities to genetically manipulate **Joule** Cell^Press

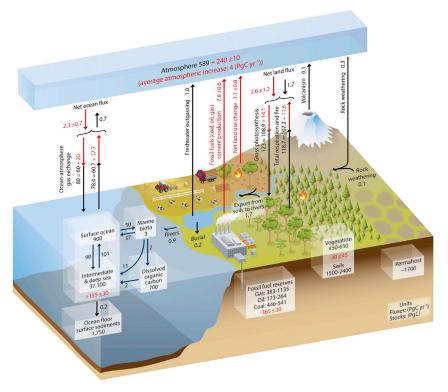


Figure 2. The Global Carbon Cycle

The numbers are in petagrams of carbon, which is the same as gigatonnes of carbon. Source: IPCC 5th Assessment Report, Working Group 1, Chapter 6 (2013). Note to Reader: The figure does not indicate which pathways have greatest potential for carbon sinks or possible leakage rates.

biological organisms that use non-photosynthetic biocatalysis for CO_2 fixation are certainly worth exploring. Finally, it is critical to create new systems designs and architectures for chemical reactors that utilize the science and are scalable to the GT scale while meeting market-competitive cost targets.

Technology pathways for synthetic transformation of CO_2 should be compared on a cost/effectiveness basis with technology pathways for negative emission technologies to remove CO_2 from the atmosphere by direct air capture, discussed in the next section.

CO₂ Sequestration in Geological Formations

Geological storage of CO₂ is an important piece of the puzzle for negative emissions since it has the potential to

store at GT scale. Over the past decade, several programs have been created to explore RD&D of CO2 storage in saline aquifers. By its very nature, this has no commercial value. On the other hand, the oil industry is using CO₂ for traditional enhanced oil recovery (EOR), which has commercial value for CO₂. However, because of the CO₂ purchase cost, the incentives in EOR are aligned to minimize CO2 use and maximize hydrocarbon recovery. If the incentives were changed, e.g., via a carbon charge, it would create a non-traditional EOR that valued the CO_2 storage (perhaps reaching GT scale) and simultaneously retained commercial value for the produced hydrocarbons? Could such nontraditional EOR lead to net reduction in CO_2 emissions? If the EOR sites and saline aquifers are co-located, would such formations offer a continuum of opportunities for CO2 storage

while also creating value out for CO₂? These questions have not been fully addressed via RD&D programs. Furthermore, while much research has focused on the fundamentals of CO2 mineralization in rocks, scalability to the GT scale remains undeveloped. It is important to focus research efforts on the fundamentals of pore-scale CO₂ displacements; co-optimizing CO2-EOR and CO2 storage in hydrocarbon reservoirs and underlying saline formations; understanding the geomechanical and geochemical effects of CO₂ transport and mineralization; addressing important questions on leakage; and developing the infrastructure to distribute CO2, as well as managing and sharing data obtained from pilot demonstrations.

CO₂ Capture and Separation

While some industrial processes such as oxy-combustion, fertilizer production, and chemical plants produce concentrated or pure streams of CO₂, most processes produce mixtures of CO₂ with other gases. Research is needed to reduce the cost of CO2 separation from gas mixtures, which can be highly energy intensive. Current CO₂ sorbents have either: (1) high rate constants and high binding enthalpy, and thereby high energy costs; or (2) low rate constants and low enthalpy, and thereby high capital costs. Research is needed to identify new, low-cost CO₂ sorbents that have sufficiently low binding enthalpy (<70 kJ/mol), and high binding rate constant (>12,000 $M^{-1} s^{-1}$). It is also worth identifying new, low-cost, noncorrosive, low-viscosity liquid solutions with lower heat capacity than water that selectively bind CO2 with the above characteristics. Furthermore, new materials and processes to separate miscible liquid mixtures are also needed. Finally, it is important the leverage the science of CO2 sorbents to design, build, and demonstrate reactors that can cost-effectively capture CO₂ at the GT scale.

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Direct Air Capture and Ocean Mineralization

The topic of direct air capture (DAC) has recently received attention from the scientific community. Almost a decade ago, 12 the cost estimate for capturing CO₂ from air using traditional sorbents such as monoethanol amine (MEA) was \sim \$600/tCO₂; a cost estimate, that remains highly uncertain. This underscores the need for research in high-performance sorbents that can dramatically reduce DAC costs. It is vitally important to conduct research to establish a practical lower bound of DAC.

The idea of CO₂ mineralization in oceans at the GT scale is attractive because the reaction CO_2 + H_2O \rightarrow H^+ + HCO_3 is exothermic and hence does not require any energy input. While the chemical transformation does not require energy input, there will be considerable energy cost in establishing the mineralization infrastructure. The bicarbonate anion (HCO₃⁻) can react with cations such as Na⁺ or Ca²⁺ to form stable bicarbonate or carbonate salts. However, the remaining proton (H+) reduces pH, which causes ocean acidification. Scalable approaches to form bicarbonate or carbonate materials would require large supplies of alkaline salts to neutralize the acidity. While alkaline rocks such as basalt and serpentine do exist, getting the alkaline materials to mix with water and form salts requires significant infrastructure and materials at scale.⁵ It is worth emphasizing that we must be very cautious in modifying the ecology of the oceans, since the complex interactions with our food chain and our environment are not completely understood.

The principal TF recommendation for the future is to encourage wide-ranging discussions with members of the technical community through organized workshops to better define carbon management options. It is essential that these discussions take place with a wide range of experts, not only with experience in one technology but also with broader aspects of managing and financing innovation. And given the magnitude of the challenge and the increasing rate of anthropogenic emissions, it is imperative that these discussions and the RD&D be undertaken with fierce urgency.

ACKNOWLEDGMENTS

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- 1. 1 ppm of atmospheric CO₂ corresponds to \sim 7.8 GtCO₂. The current CO₂ concentration of 410 ppm corresponds to ~3,200 GtCO₂.
- 2. Arun Majumdar was vice chair SEAB and chair of this task force; John Deutch was chair of SEAB and a member of the task force. Other members of this task force were: Sally Benson, Stanford University; Rafael Bras, Georgia Tech; Emily Carter, Princeton University; Don Ort, University of Illinois at Urbana-Champaign; Michael Ramage, formerly of Exxon-Mobil; Robert Socolow, Princeton University; Eric Toone, formerly of Duke University and currently at Breakthrough Energy Ventures; George Whitesides, Harvard University; Mark Wrighton, Washington University. The report was also reviewed by the following group of experts: Ken Caldeira, Carnegie Institute for Science; Michael Celia, Princeton University; Steven Koonin, New York University; Nathan Lewis, California Institute of Technology; Venkatesh Narayanamurti, Harvard University; Edward Rubin, Carnegie Melon University; Chris Sommerville, University of California, Berkeley; Ellen Stechel, Arizona State University. The report also received the benefit of collaboration with the following Department of Energy personnel: Adam Cohen, Doug Hollett, John Lytsinski, Harriet Kung, Ruben Sarkar, Sunita Satyapal, Eric Miller, Sharlene Weatherwax, and Ellen Williams.
- 3. The SEAB report is dated December 12, 2016, and the Department of Energy assessment of the report is dated January 18, 2017. Available at https://www.energy.gov/ seab/listings/seab-reports.
- 4. Fifth Assessment Report of the Intergovernmental Panel on Climate Change
- 5. National Research Council. (2015). Climate Intervention: Carbon Dioxide Removal and Reliable Sequestration, National Research

- Council Report (The National Academies
- 6. Pacala, S., and Socolow, R. (2004). Stabilization wedges: solving the climate problem for the next 50 years with current technologies. Science 305, 968-972.
- 7. The global implementation of carbon-free/ neutral electricity from wind, solar, nuclear, geothermal, biomass, and hydroelectric as well as those from low-carbon sources such as natural gas and coal with carbon capture would require serious combinations of policy measures and technological advancements.
- 8. If CO₂ was to be used as the source of all carbon in the global annual production of plastics (311 million tonnes [MT] per year in 2014), it would consume about 0.8 GtCO₂ per year. By 2030, the annual global plastic production is expected to rise to 700 MT, which would require roughly 490 MtC/year or about 1.8 GtCO₂/year.
- 9. The lower bound for the energy needed to achieve this is the energy released from combusting these chemical and fuels to form CO₂. A barrel of oil equivalent contains about 6.1 GJ or 1.7 MWh of energy. Hence, to convert CO₂ into a barrel of oil equivalent, the lower bound for the amount of carbonfree energy needed will be 1.7 MWh. In 2015, the US used 7.08 billion barrels of oil. If all the carbon in this came from CO₂, then the lower bound for the amount of carbon-free energy needed would be 12,000 TWh, which is about 41 Quads. As a comparison, the US uses roughly 100 Quads per year of primary energy. Also, the total electricity generation in the US in 2014 was 4093 TWh, of which 1340 TWh came from carbon-free sources (nuclear, wind, solar, hydroelectric).
- 10. It should be noted that among the most innovative and potentially high-impact strategies to improve photosynthesis in plants is to import carbon concentration mechanisms that operate in photosynthetic bacteria and algae. The opportunity is clear but the molecular technology to import these complex components is lacking, as is a full understanding of the biology of their
- 11. Note, there is an opportunity cost of using carbon-free/neutral energy to convert CO₂ into chemicals and fuels, since this energy could have been used to displace fossil fuels. A systems approach can be used to estimate the net \dot{CO}_2 impact of these two
- 12. Socolow, R. (2011). Direct Air Capture of CO₂ with Chemicals. APS Report (American Physical Society). https://www.aps.org/ policy/reports/assessments/upload/ dac2011.pdf.

¹Department of Mechanical Engineering & Precourt Institute for Energy, Stanford University, Stanford, CA 94305, USA

²Department of Chemistry, Massachusetts Institute of Energy, Cambridge, MA 02139, USA

*Correspondence: amajumdar@stanford.edu https://doi.org/10.1016/j.joule.2018.04.018