Geoengineering Responses to Climate Change

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Geoengineering Responses to Climate Change

Selected Entries from the Encyclopedia of Sustainability Science and Technology

Springer

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Chapter 1
Introduction

Timothy M. Lenton and Naomi E. Vaughan

This section focuses on ideas to deliberately remedy anthropogenic climate change, either by actively removing greenhouse gases from the atmosphere or by decreasing the amount of sunlight absorbed at the Earth's surface. The technologies discussed are commonly grouped under the term "geoengineering," defined in a 2009 report by the Royal Society as the "deliberate large-scale manipulation of the planetary environment to counteract anthropogenic climate change." Geoengineering methods can be contrasted with more conventional approaches to mitigating climate change that involve reducing the emissions of greenhouse gases, especially carbon dioxide (CO_2). However, there is some overlap as enhancing the sinks of greenhouse gases, for example, by afforestation, can be described as both geoengineering and mitigation. (The Intergovernmental Panel on Climate Change (Working Group III) states that "mitigation means implementing policies to reduce greenhouse gas emissions *and enhance sinks*" [our emphasis].)

Failure by the international community to make substantive progress in reducing CO_2 emissions, coupled with recent evidence of accelerating climate change, have brought urgency to the search for additional means of tackling climate change. This has fueled much recent debate about geoengineering and a flurry of mostly model-based research studies. There is widely expressed concern that undertaking, or even discussing, geoengineering poses the "moral hazard" of reducing efforts to tackle the root cause of climate change, namely, greenhouse gas emissions. However, few

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of those researching geoengineering advocate it as an alternative to reducing greenhouse gas emissions. Instead, current discussions are usually framed in terms of the possible use of geoengineering *in addition to* reducing greenhouse gas emissions in order to limit the magnitude of climate change, for example, to stay within the widely discussed policy "threshold" of limiting global warming to 2°C above preindustrial.

The technologies discussed herein can be subdivided into those involving carbon dioxide removal (CDR) from the atmosphere, and those involving reflecting sunlight, referred to by the Royal Society as "solar radiation management" (SRM) (although the word "management" implies a high level of understanding of the system in question that is probably not justified for the climate). The distinction can also be thought about in terms of wavelengths of radiation; short-wave geoengineering tries to reduce incoming sunlight, while long-wave geoengineering tries to reduce the return flux of heat to the surface from the increased blanket of heat-absorbing gases in the atmosphere. Sunlight reflection can never perfectly counterbalance an increased greenhouse effect because the two types of downwelling radiation have different spatial and seasonal patterns.

The section presents some of the more widely discussed geoengineering options, without being comprehensive. Here, we try to fill some of the gaps as well as introduce the entries herein.

Sunlight Reflection (SR)

Sunlight reflection can act rapidly to cool the climate if deployed on a sufficiently large scale. It could, in principle, be deployed to return the Earth to its preindustrial temperature, to hold it at some level of warming that has already occurred, or to lower future global warming. A popular framing is that sunlight reflection could "buy time" for decarbonising the economy and allowing greenhouse gas concentrations to stabilize and then come down. Alternatively, potent methods of sunlight reflection might be reserved for use "in emergency" should dangerous climate change become apparent, but it is as yet unproven that such a deployment after a threshold had been passed would prevent the change that was already underway.

Methods of sunlight reflection can be distinguished in terms of the altitude at which they are applied. They start outside the Earth's atmosphere with the idea of placing mirrors or sunshades in space to reduce the amount of sunlight reaching the Earth (D. J. Lunt "Sunshades for Solar Radiation Management"). To counteract a doubling of atmospheric carbon dioxide would require a roughly 2% reduction in the amount of sunlight reaching the top of the Earth's atmosphere. To achieve this, one proposal is to put tiny "flyers" (each about 0.3 m^2) between the Earth and the Sun at the first Lagrangian (L1) point. The roughly 5 million km^2 of sunshade required at this distance (around 1.5 million km) means the method will probably remain in the realm of science fiction for the foreseeable future. However, the model studies that have been conducted to examine the effects on the climate provide useful information on the effects of a uniform reduction in incoming sunlight, which might be achieved by other means.

Currently, the leading candidate for geoengineering a reduction in incoming sunlight is to inject tiny particles into the Earth's stratosphere which will scatter (or in some cases absorb) sunlight (B. Kravitz "Stratospheric Aerosols for Solar Radiation Management"). (The stratosphere is the thermally stratified layer of the Earth's atmosphere between about 10 and 50 km altitude, separated from the well-mixed troposphere below by the tropopause.)

A natural analogue here is a volcanic eruption, such as Mt. Pinatubo in 1991, which injected sulfate aerosol into the stratosphere and measurably cooled the climate (by around 0.5°C in the following year, followed by a tailing off). The stable thermal structure of the stratosphere means that aerosols stay aloft for much longer than in the troposphere below, and therefore, a much smaller loading of particles is required to have a given cooling effect. An estimated 1.5–5 MtS year^{-1} would need to be deliberately injected into the stratosphere to offset a doubling of CO_2, which is much less than the 50–100 MtS year^{-1} that human activity currently adds as pollution to the troposphere. The main unanswered questions surround how to inject sulfate aerosols into the stratosphere in a way that stops them from coagulating, because if the tiny particles combine to become larger particles, this can profoundly alter their radiative properties, at the extreme, turning a cooling effect into a warming effect. Other types of aerosol, notably soot, and engineered metal nanoparticles are also being discussed as possible candidates for stratospheric injection.

Moving down into the lower atmosphere, clouds are a major contributor to the reflectivity of the planet, and low-level clouds in particular have a net cooling effect at the surface. Hence, it has been proposed to cool the planet by making marine stratocumulus clouds more reflective (S. H. Salter "Solar Radiation Management, Cloud Albedo Enhancement"). This can be achieved by distributing the same amount of cloud water over more but smaller droplets, which requires a source of the tiny particles known as cloud condensation nuclei on which water condenses. Sea salt is the most obvious and ubiquitous candidate aerosol, and a means of spraying it from the ocean using wind-powered boats with Flettner rotors is described by Salter. An alternative is to enhance the source of cloud condensation nuclei from the biological production and air-sea exchange of the gas dimethyl sulfide. The methods may also suppress rainfall, increasing the lifetime of clouds and giving a further cooling effect. They should be most effective far from sources of human pollution (which also provide condensation nuclei), such as in the Southern Ocean. However, this means the cooling effects on the climate would inevitably be patchy, which in turn can cause unexpected climate changes far away, for example, in some model simulations, the Amazon gets drier.

Finally, a number of proposals have been made to enhance the reflectivity of the Earth's surface, focusing on deserts, grasslands, croplands, and human settlements. Here, the total area altered and the change in surface reflectivity (albedo) are the key determinants of the global cooling effect. As the land comprises only 29% of the Earth's surface, to achieve a significant global cooling effect would require major overcooling of parts of the land. Instead, these approaches are best thought of as means of achieving significant localized cooling. In the case of more reflective croplands, cooling effects would be largest just before the crop is harvested, so this could provide a means of, for example, cooling future European summers. However, there remain unanswered questions regarding the effects on crop yield. Reflective roof surfaces are now legislation in California and more widespread adoption of this approach could become an important adaptation strategy in tackling urban heat islands, even though its global cooling effect will be negligible.

Carbon Dioxide Removal

Current total CO_2 emissions from fossil fuel burning, land-use change, and cement production are rapidly approaching 10 billion tons of carbon per year ($\sim$10 PgC year^{-1}). Just to stop CO_2 concentration from rising in the atmosphere, the net anthropogenic source of CO_2 has to be reduced first by about 50% to match natural sinks and, then on, down to zero as the natural sinks decay. To help achieve this, carbon dioxide removal (CDR), and subsequent storage, is a clear complement to reducing CO_2 emissions. Doing both together could stabilize atmospheric CO_2 concentration sooner and at a lower level. Ultimately, CDR could be used to bring the concentration of atmospheric CO_2 down faster than natural sinks, to whatever is deemed a safe level. However, like reducing emissions, CDR will act relatively slowly to alter the rate and magnitude of climate change when compared to potent methods of sunlight reflection. To achieve significant global CO_2 removal, even with the most effective CDR methods, will require global deployment for decades. Furthermore, the Earth system actually works against deliberate CDR by always trying to maintain a balanced apportioning of CO_2 between the ocean, atmosphere, and land surfaces. Hence, if CO_2 is removed from the atmosphere, some leaks out from the ocean and/or land to partly compensate, meaning that the effect on atmospheric CO_2 concentration decays over time. (This is simply the opposite of the well-known land and ocean carbon sinks, which are generated by the addition of CO_2 to the atmosphere.)

Carbon dioxide removal covers a wide range of methods and pathways to storage. CO_2 can be removed from the air by photosynthesis (by plants, algae, or cyanobacteria) or by physical and chemical means, which are related to natural weathering reactions. The ease of removal varies with the pathway. The carbon may ultimately be stored as liquid CO_2 (in geological reservoirs or the deep ocean), in charge-balanced solution in seawater, as carbonate rocks, as charcoal, or as buried or standing biomass. The different forms of storage have differing stability in terms of thermodynamics, kinetics of reactions, and ease with which CO_2 might be returned to the atmosphere.

Photosynthesis is effectively solar-powered carbon capture for free (to us), although it is remarkably inefficient ($\sim$0.5% efficiency, compared to solar photovoltaic cells capable of ~20% efficiency), and biomass is the least stable form of carbon storage because it is a source of energy to other organisms. However, in the ocean, some of the carbon fixed in photosynthesis can sink to depths where it has a lifetime of up to 1,000 years. The amount exported to depth depends crucially on the supply of limiting nutrients to the surface ocean. Hence, several geoengineering proposals consider adding limiting nutrients, especially iron, to the surface ocean to stimulate biological productivity (P. W. Boyd "Ocean Fertilization for Sequestration of Carbon Dioxide from the Atmosphere"). Iron fertilization is unusual in being a form of geoengineering that can draw on a series of 12 experiments to investigate its biogeochemical consequences (though not geoengineering specifically). However, what those experiments have shown is that it is remarkably difficult to increase the sinking flux of carbon to the deep ocean. Model studies add that even with global iron fertilization maintained for a century, the potential impact on atmospheric CO_2 concentration is modest, lowering it by, at most, around 30 parts per million.

Land-based photosynthesis has the potential to fuel larger carbon dioxide removal fluxes, despite the smaller surface area of the land compared to the ocean and the need not to take land from natural ecosystems or to interfere with food production. The potential is greater because of the high productivity that can be achieved on land. The simplest method is afforestation, but it only works as a means of carbon dioxide removal if the conversion to forest is permanent and the carbon that is lost as trees decay is replaced by new trees. Still, afforestation is already underway at a global scale, with around 250 million hectares having been planted in recent decades, and this is creating a sink of circa 0.3 PgC $year^{-1}$, canceling roughly 3% of current total CO_2 emissions.

Carbon dioxide removal can also be achieved by converting waste biomass from farming and forestry into longer-lived forms for storage, although the long-term potential will depend more on the supply of deliberately cultivated biomass energy crops. Several conversion pathways are available. Biomass energy combustion coupled with capture and storage of the CO_2 given off (often referred to as BECS or BECCS) is more cost-effective than chemical methods of CO_2 air capture, although there are still energy penalties in capturing and compressing CO_2. Fermentation of biomass, for example, to produce liquid biofuels, yields a near-pure stream of CO_2 reducing the capture cost. Alternatively, pyrolysis of biomass (in the presence of little or no oxygen) produces charcoal, which can be returned to the soil as biochar (S. Shackley et al. "Biochar, tool for climate change mitigation and soil management"). Although the energy yield from pyrolysis is somewhat less than from combustion, biochar has a range of cobenefits, including improving soil water retention and fertility, which make it an attractive option. Energy remains in the biochar, but it is hard for organisms to break the material down, making much of it long-lived in soil.

Carbon dioxide can also be captured from the air by chemical means, using, for example, a strong alkali solution. When CO_2 has been captured into concentrated form in this way, or from combusting or fermenting biomass, it can be stored in liquid form. However, it is safer and more permanent to neutralize carbonic acid to form carbonate rocks or aqueous bicarbonates by mirroring natural rock weathering processes. Carbonate weathering brings CO_2 into solution as bicarbonate (although, ultimately, on a ~10,000-year timescale when carbonates are redeposited in the ocean, the CO_2 will be returned to the atmosphere). Silicate weathering followed by carbonate deposition is a permanent removal process for atmospheric CO_2. However, these reactions are generally rather slow, even when applied to a CO_2-rich gas stream. Heating to speed up the reactions is too costly; hence, better methods for accelerating carbonation are needed, and some work on using electrochemical energy to accelerate the reactions is underway.

Finally, CO_2 can be removed from the air by mining, crushing, and spreading on the land silicate minerals that weather rapidly such as serpentine or olivine (R. D. Schuiling "Carbon Dioxide Sequestration, Weathering Approaches to"). This represents a direct attempt to accelerate silicate weathering, which should be most effective in wet regions of the tropics under vegetation, because plants and their associated mycorrhizal fungi produce organic acids that accelerate dissolution. Olivine has relatively large deposits in the tropics, making it a promising candidate, although estimates are needed of the energy and CO_2 costs of mining, grinding, and distributing the rock on appropriate land. Weathering rates can be limited by factors other than substrate supply, and current estimates suggest this CDR method may be limited to at most around 1 PgC year^{-1}.

Broader Issues

The concept of geoengineering and the particular proposals, both CDR and SRM, provoke a plethora of social concerns ranging from specific questions of economical or political feasibility for each proposed method to broader ethical and philosophical debates about our relationship with nature.

Efforts are underway to establish a framework for governing geoengineering, particularly SRM. This poses a host of questions, relating both to research and possible implementation (S. Low et al. "Geoengineering Policy and Governance Issues"). Historical efforts at weather modification provide some past precedent (albeit at a smaller scale), but they offer little in the way of existing governance frameworks to draw on. Given the rapidly evolving nature of the field, flexibility and adaptability will be key requirements for whatever governance framework emerges.

The use of any geoengineering method as a response to climate change will ultimately be made by societies. Therefore, the public perception of, and engagement with, this group of emerging technologies is of critical importance in determining their future usefulness. Concepts of upstream engagement and responsible innovation can help incorporate a range of societal values into research at an early stage (and they are both, in different ways, built into two current UK Research Council–funded projects on geoengineering).

Early public engagement (e.g., “Experiment Earth” conducted by the UK Natural Environment Research Council) has already yielded unexpected responses to geoengineering. In particular, the “moral hazard” argument that geoengineering will suppress efforts to reduce greenhouse gas emissions has not been clearly borne out. Some members of the public responded to information on geoengineering with the opposite response; that if things are so bad that scientists are considering geoengineering, then efforts to reduce the root cause of climate change, namely, greenhouse gas emissions, must be strengthened.

Moving forward, the ethical and philosophical debate about geoengineering needs to distinguish SRM and CDR techniques. SRM only deals with the symptoms of climate change, notably rising temperatures. CDR, on the other hand, like conventional emissions reduction, tackles the root cause of climate change: rising greenhouse gas concentrations. Specific methods still raise specific concerns. But as research on geoengineering continues to escalate, we hope this section provides a stimulating introduction to the methods and the debates surrounding them.

Chapter 2
Sunshades for Solar Radiation Management

Daniel J. Lunt

Glossary

Geoengineering	The intentional large-scale manipulation of the environment.
Solar radiation management	Deliberate modification of the solar radiation budget, by either changing the amount of sunlight entering the Earth's atmosphere, or by changing the Earth's reflectivity, normally in an effort to counteract human-induced climate change.
Sunshade	A colloquial term for a giant reflector, or array of smaller reflectors, in orbit between the Earth and the Sun. Also known as a "space mirror."

Definition of the Subject and Its Importance

There is strong scientific consensus that the Earth's climate has been warming over the last century and that this warming is primarily due to human influences on the climate system (IPCC 2007). Attempts to curb human emissions of greenhouse gases have so far largely failed; as such, and in an attempt to avoid or delay potential dangerous climate change, several geoengineering schemes have been suggested for modifying Earth's climate directly. One such scheme is the construction of a sunshade in space, in

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orbit between the Earth and the Sun, with the aim of reducing the effective strength of the Sun's rays, and cooling the Earth's climate. However, it is now thought that such a sunshade, although highly effective in terms of cooling the planet, would have other, perhaps undesirable, impacts on Earth's climate, in particular on precipitation patterns. This highlights the importance of using a whole-Earth system approach when considering the potential impacts of any geoengineering scheme.

Introduction

One of the first references to a space-based sunshade was proposed in the context of modifying the climates of extraterrestrial planets, with the aim of making them habitable [1]. More recently, it has been widely discussed in the context of cooling our own planet, in an attempt to mitigate effects of human-induced global warming (e.g., [2–5]). The basic underlying concept is extremely simple – a reflective or deflecting substance or substances are placed between the Earth and the Sun in order to reduce the intensity of incoming solar radiation at the top of the Earth's atmosphere. The amount of reduction in solar radiation would be chosen to offset, or partially offset, surface warming induced by increases in greenhouse gases in the Earth's atmosphere.

The apparently alluring idea that a sunshade could perfectly offset greenhouse-gas-induced global warming is, perhaps unfortunately, an oversimplification. A sunshade positioned in space between the Earth and the Sun would reduce the effective strength of the Sun, resulting in an absolute change in the local solar forcing at the top of the Earth's atmosphere proportional to the local solar input, meaning that high latitudes and winter seasons receive less solar energy decrease than low latitudes and summer seasons (see Fig. 2.1). However, the radiative forcing due to increased greenhouse gases is more homogenous seasonally and latitudinally. As such, although the *global annual mean* greenhouse gas forcing could be exactly canceled by a solar shield, in such a "globally corrected" case the tropical and summer regions would be overcompensated, and high latitude and winter regions would be undercompensated. This would result in a surface temperature pattern of a globally corrected geoengineered world which, compared to modern, would be colder in the tropics than in the high latitudes. Along with this imperfect cancelation of temperature, other aspects of the Earth system would also remain unmitigated, such as changes to the hydrological cycle, as well as impacts resulting directly from the high CO_2, such as ocean acidification.

This entry focuses on the climatic effects (both desired and undesired) of placing sunshades in space. It also touches very briefly on some of the engineering considerations associated with the manufacture and launch of such a system. The substantial governance, political, and ethical considerations are not discussed here. Instead, the reader is referred to Virgoe [21], Blackstock and Long [7], and Corner and Pidgeon [8] for discussion of these more qualitative, but nonetheless important, issues.

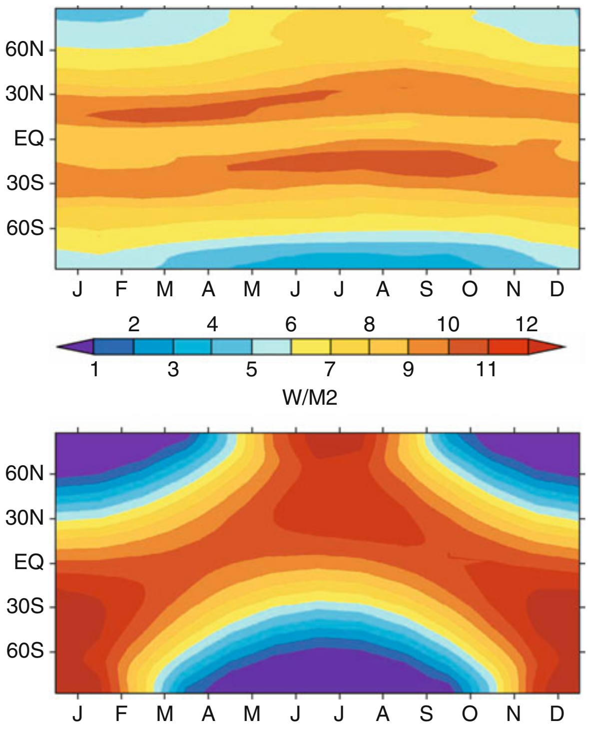

Fig. 2.1 Change in net long-wave radiative flux at the tropopause when CO_2 is quadrupled (*top panel*) with respect to the Control case and the reduction in incoming solar radiation (*bottom panel*) needed to compensate this forcing. Both values (W m^{-2}) are zonally averaged as a function of time of year. Change in solar radiation has a latitudinal and seasonal pattern markedly different from the radiative forcing of CO_2 (Adapted from [6], Fig. 2.1)

Modeling the Efficacy of Sunshades

Background

Because of the incredible complexity of the Earth system, and the expense and potential risks of carrying out large-scale field studies, probably the only way that the success and possible side effects of sunshade geoengineering can be assessed, at least initially, is through numerical climate modeling. Climate models, also known as “General Circulation Models,” or GCMs, consist of a numerical representation of our best understanding of the Earth system. In the atmosphere, they typically consist of a “dynamical core” which solves an approximation to the fundamental equations of motion of a perfect gas on a rotating sphere, a representation of radiative processes across a range of wavelengths from the solar to the infrared, and a set of “parameterizations,” which represent the large-scale effect of processes which occur at too small a scale to be resolved by the dynamical core, such as small-scale atmospheric waves, turbulence, processes associated with clouds, surface processes such as evaporation, and other aspects of the hydrological cycle. These atmospheric models can either have the temperatures of the ocean surface prescribed as boundary conditions, or can use a simple "slab" representation of the ocean in which the ocean interacts thermodynamically with the atmosphere (e.g., warming up if the atmosphere warms).

The last decade has also seen the development of coupled atmosphere-ocean models, which, as well as the atmosphere, also solve equations related to the fluid flow of the oceans. Given an initial condition (e.g., a static atmosphere and ocean, or the ocean–atmosphere state on a particular historical day since the advent of dense networks of observations) and a set of appropriate boundary conditions (such as the Earth's topography and bathymetry and land-surface characteristics, and atmospheric gas composition), a model will typically solve the equations in the dynamical core and associated parameterizations, and increment the state of the atmosphere forward in time (typically 15 min to 1 h, depending on the spatial resolution of the model – high-resolution models require short timesteps to maintain numerical stability). If the boundary conditions remain fixed over time, then the model will eventually (typically years to decades for an atmosphere-only model, centuries to millennia for an atmosphere-ocean model) reach a quasi-equilibrium, where dynamic weather systems are superimposed on an equilibrium circulation of the atmosphere-ocean system. In this case, the initial condition of the model becomes unimportant (unless there are multiple equilibrium states of the system for the given boundary conditions).

How can these models be applied to sunshades in space? In a typical numerical experimental design, three model simulations are carried out with three different sets of boundary conditions. Firstly, a "modern" (or "control" or "preindustrial") simulation is carried out, in which the boundary conditions are set as those of the modern era (or alternatively pre-industrialization values for atmospheric gas composition). Secondly a "perturbed" or "future" simulation is carried out, in which greenhouse gas concentrations are set at elevated concentrations (e.g., twice or four times preindustrial or modern values). Thirdly, a "geoengineered" simulation is carried out, in which the greenhouse gas concentrations are elevated and in addition the strength of the Sun's solar output (the "solar constant") is reduced. The reduction of solar constant acts as an approximation to the effect of a sunshade in space. The magnitude of the solar constant reduction is usually chosen so as to balance as closely as possible the global annual mean increase in surface air temperature caused by the elevated greenhouse gas concentrations. An alternative experimental design consists of a transient time-varying simulation, in which greenhouse gas concentrations are slowly increased in the "future" simulation, and a corresponding slow increase in the strength of the solar shield is applied in the "geoengineered" simulation.

Results

The first numerical model study of the impacts of geoengineering was carried out by Govindasamy and Caldeira [2]. Using an atmospheric GCM and a "slab" ocean model, they carried out three equilibrium simulations, as outlined above. They found a cooling in the tropics (20°S–20°N) in their geoengineered simulation compared to their control simulation, and a warming outside these regions. However, because of the relatively short length of the simulation (the climatologies were calculated over only 15 years of model time), only a small fraction of the temperature changes in their geoengineering simulation compared to their modern simulation were deemed statistically significant at a 95% confidence limit. Coupled with their use of a "slab" ocean model compared to a full atmosphere-ocean GCM, some of their other results (e.g., they observed an increase in sea ice in their geoengineered simulation compared to their control, and little change in the hydrological cycle) remained somewhat ambiguous. However, this paper was certainly pioneering, and inspired a series of subsequent studies which used a very similar methodology.

Govindasamy et al. [6] carried out a follow-on study, which increased the forcing to a fourfold increase in greenhouse gas concentration. This resulted in a larger signal to analyze, and, as well as confirming their earlier results regarding temperature changes, also resulted in a more physically realistic decrease in sea ice in their geoengineering simulation compared to their control. They also noted that this form of geoengineering, although doing a relatively good job of returning tropospheric temperatures back to modern levels, does little to counteract the cooling in the stratosphere associated with the warming in the troposphere. There was also a decrease in evaporative water flux from the ocean to the atmosphere in the tropics, associated with the cooler sea surface temperatures. However, they noted that all their results, and especially those associated with sea ice, should be regarded with some caution due to their use of a "slab" ocean model and lack of dynamics in the sea-ice scheme.

This issue was partially addressed by the study of Matthews and Caldeira [5]. They used a model with a representation of ocean dynamics, but due to computational constraints used a simplified representation of the atmosphere compared with Govindasamy and Caldeira [2]. They carried out a set of "transient" geoengineering simulations. Because their model was relatively computationally efficient, they were able to carry out long simulations and obtain statistically meaningful results. Their results largely agreed with those of previous studies regarding temperature change, but were very different in terms of precipitation. Matthews and Caldeira [5] found a widespread decrease in continental precipitation in a geoengineered world compared to modern, in particular over the rainforest regions of Amazonia, Central Africa, and South East Asia. They attributed this to decreased evapotranspiration in a high-CO_2 world, as plants used water more efficiently. Matthews and Caldeira [5] also addressed the question of the "safety" of sunshade geoengineering. In particular, several of their scenarios included a simulated catastrophic failure of the sunshade, such that the solar constant was instantaneously increased back to its normal value, with CO_2 levels still high. In this case, they found an extremely rapid warming, up to 20 times greater than the current anthropogenic warming. However, as with previous work, there remained some uncertainty in the validity of some of the results (in particular, the large reduction in precipitation in the geoengineered case) due to the lack of complexity in the atmospheric component of their model.

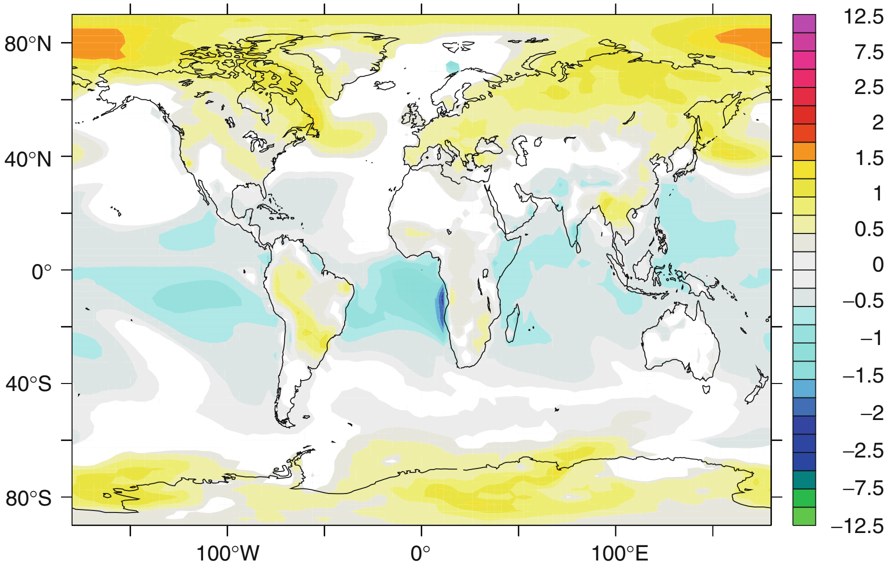

Fig. 2.2 Change in near-surface air temperature (°C) in a sunshade geoengineered world compared to modern, as predicted by a coupled atmosphere-ocean General Circulation Model. Regions where the difference is not statistically significant at a 95% confidence limit, as given by a Student *T* test, are masked out in white (Adapted from [4], Fig. 2.2)

The first study to use a fully coupled atmosphere-ocean climate model to investigate sunshade geoengineering was that of Lunt et al. [4]. They also carried out long simulations with averages calculated over 60 years. In general terms, their temperature estimates for the "sunshade world" were similar to previous, but with some new features, such as a maximum in cooling off the west coast of tropical Africa, associated with increased ocean upwelling in the geoengineered case, and maximum warming in the Arctic north of the Bering Strait, associated with a loss of sea ice (see Fig. 2.2). They also found new results in terms of precipitation, with a global mean decrease in precipitation predicted (in agreement with Matthews and Caldeira [5]), but instead of maximum reductions over continents, they found maximum reductions over the tropical oceans, in a regional pattern which correlated with that of temperature. The use of a fully coupled model also allowed them to investigate changes in El Nino Southern Oscillation (ENSO). They found a significant decrease in the amplitude of ENSO variability in sunshade world relative to modern, associated with a decrease in the tight coupling of atmosphere and ocean feedbacks due to the cooler tropics, but no significant change in ENSO frequency. They also found that the geoengineered world had a slightly more vigorous ocean circulation than the control, associated with a decrease in the intensity of the hydrological cycle, and decreased poleward moisture transport.

This study was followed up by Irvine et al. [3], who used the same model, but investigated a range of "strengths" of sunshade geoenginering, from 100% (which exactly balances the global annual mean temperature change associated with a greenhouse gas concentration increase), to 0% (no geoengineering). They also looked in more detail at the regional patterns of change following sunshade geoengineering. For example, they found that, relative to modern, the USA mainland region experiences a +8% increase in precipitation given 0% geoengineering, and an 11% decrease in precipitation given 100% geoengineering. According to their results, to achieve a precipitation rate close to that of modern, the USA would "choose" a sunshade of strength 40–50%. However, the east China region experiences precipitation back at modern levels with 100% geoengineering. The possible political implications are considerable. They also investigated the effects of sunshade geoengineering on croplands and densely populated regions, and found that in order to obtain modern average values of precipitation in cropland regions would require 75% geoengineering, whereas in urban areas it would require 85% geoengineering. Although this study highlighted the very regional impacts of geoengineering schemes, it should also be noted that no climate model is perfect, and in particular precipitation remains one of the variables which shows the largest inter-model spread. As such, the exact predictions of "ideal" geoengineering levels presented in Irvine et al. [3] should be regarded with some caution.

Several other studies have looked at the implications of sunshade geoeingineering on other aspects of the Earth system. Govindasamy et al. [9] looked at the impact of geoengineering on the biosphere. They used a coupled atmosphere-biosphere model with a slab ocean to carry out the classic three equilibrium geoengineering simulations. They found that the decrease in solar energy in the geoengineered world had very little effect on net primary productivity (NPP) by the biosphere, and instead that the CO_2 increase relative to modern resulted in almost a doubling of NPP. Their model did neglect some possibly important factors such as changes to nutrient cycles in the sunshade world. However, these are likely to be minor compared to the zeroth order result of increased biomass in the geoengineered world compared with modern.

Irvine et al. [10] addressed the possible mitigative potential of sunshade geoengineering for sea-level rise. This question was motivated by the "residual" Arctic warming seen in all of the sunshade geoengineering simulations. Would this residual warming be enough to significantly affect the Greenland ice sheet, and lead to sea-level rise? To address this question they used an offline ice sheet model, which, given fields of temperature and precipitation, calculates the surface mass balance (accumulation vs. melt) and flow of an ice sheet, to obtain an ice sheet geometry that is in equilibrium with the temperature and precipitation. They compared the ice sheets predicted to be in equilibrium with the "control," "future," and "geoengineered" climates from GCM simulations, and found that the "future" climate produced a Greenland ice sheet which was almost completely melted, with only small ice caps remaining on the high-altitude or high-accumulation regions in the east and south of Greenland, equivalent to a sea-level rise of 6.4 m. However, the "geoengineered" climate, despite the residual Arctic warming, was almost identical

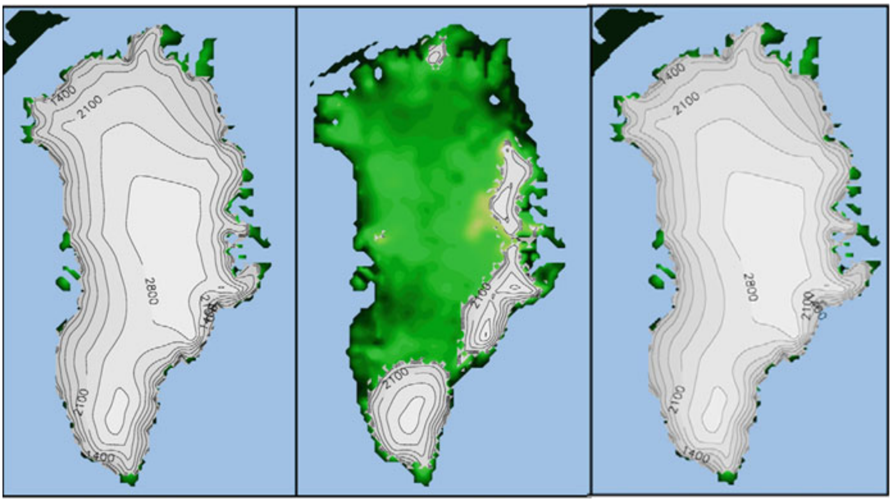

Fig. 2.3 Equilibrium ice sheets, as predicted by an ice sheet model. *Left*: ice sheet driven by modern climates (temperature and precipitation). *Middle*: ice sheet model driven by climate of a world with quadrupled atmospheric CO_2. *Right*: ice sheet model driven by climate of a world with quadrupled atmospheric CO_2 and a sunshade (Adapted from [10], Figs. 2.3 and 2.4)

to the "control" case, with essentially no sea-level rise (see Fig. 2.3). Irvine et al. [10] went further, simulating a range of strengths of sunshades, and found that a sunshade which reflected only 60% of the sunlight necessary to return global average temperatures back to modern, was sufficient to prevent any melting of the Greenland ice sheet; less than 60% and significant sea-level rise was predicted. Although this study highlighted the potential for sunshade geoengineering to mitigate sea-level rise, its conclusions should be viewed with some caution, as the ice sheet model used lacked some physical processes which could be important, for example, climate-albedo feedbacks, and lubrication of the bedrock by surface melting ice flowing down crevasses (e.g., [11]). It also contained simplified representations of other processes, such as surface mass balance (e.g., [12]) and flow regimes important for ice streams (e.g., [13]).

Many studies have qualitatively highlighted the fact that sunshade geoengineering, despite being predicted to bring temperatures close to those of modern, does nothing to mitigate the effects of increasing anthropogenic CO_2 leading to ocean acidification. Ocean acidification is the process in which a fraction of any increase in atmospheric CO_2 dissolves in the ocean, decreasing the ocean pH. This decrease in pH could have significant effects on ocean ecosystems; corals have been highlighted as being particularly susceptible to relatively small decreases in pH. Matthews et al. [20] noted that actually sunshade geoengineering would have some change on ocean pH compared to a modern world, due to the changes in ocean temperature which result from the sunshade's imperfect cancelation of the CO_2-induced warming, and due to changes in the terrestrial uptake of carbon. The cooler tropical temperatures in the tropics in the sunshade world result in increased solubility of CO_2, and therefore decreased pH. However, this is more than offset by the terrestrial carbon cycle change, where the cooler temperatures in the sunshade world result in increased terrestrial biomass compared with the unmitigated world, resulting in slightly lowered atmospheric CO_2, and therefore less dissolved ocean CO_2. So, a sunshade world might have a slightly less acidified ocean than an unmitigated future climate. However, the effect is relatively small, mitigating about 10% of the pH change by the year 2100 in their future scenario.

How does solar radiation management by sunshades compare to other geoengineering schemes? The Royal Society, in their report on geoengineering (2009), rate a number of geoengineering schemes according to their effectiveness, affordability, timeliness (how quickly the technology could be implemented and how quickly it would act once implemented), and safety (possible side effects). They rated sunshades as one of the most effective, but one of the least affordable and least timely, forms of geoengineering. In terms of safety it was ranked medium. A more quantitative assessment was carried out by [14]. They compared sunshade geoengineering with a range of surface albedo Solar radiation management schemes. Unsurprisingly, they found sunshades to be more effective than the other schemes (urban, crop, and desert albedo geoengineering) in terms of returning surface temperatures back to those of modern. And in spite of the previously noted changes to the hydrological cycle, they also found it to be more effective than the other schemes at returning precipitation back to modern levels.

Engineering Considerations

So, could a sunshade be feasibly built? Various engineering solutions have been suggested (e.g., [15, 16]), either fabricating a sunshade on Earth and transporting it to space, or building a sunshade out in space from materials available on asteroids. Sunshades have been suggested which consist of a single superstructure, or an array of millions of tiny reflectors or refractors. For those sunshades built on Earth, electromagnetic rails could launch the individual reflectors into space [15]. Estimates of cost vary, but a value in the region of several trillion dollars has been suggested [15]. Several authors have suggested that an obvious candidate for positioning a sunshade would be at or near the “first Lagrange point” (also known as the “L1 point”), which is an equilibrium point directly between the Earth and sun, about five times further from the Earth than the moon, at which the gravitational attraction of the Sun and Earth exactly cancel. An object placed at the L1 point is unstable, in that if it is displaced slightly, it will be accelerated further in the same direction, toward either the Earth or Sun. Such a displacement would occur due to the pressure on a sunshade exerted by the Sun’s radiation itself. Therefore in practice, a more suitable location for a sunshade would be slightly closer to the

Sun than the L1 point, allowing the radiation pressure to balance the offset in gravitational attraction. The ideal location (that which minimizes the mass of sunshade required for a given decrease in solar strength) depends on the density and reflective properties of the sunshades [16]. Others have suggested positioning a sunshade in a near-Earth orbit, or a ring of tiny dust particles, held in place by accompanying satellites [17]. However, these near-Earth solutions would potentially interfere with communications and other satellites.

Future Directions

So, where now for sunshade geoengineering? In terms of modeling, many of the predictions made so far have been carried out with models that can no longer be considered "state of the art." More recent models, such as those that will be used in the forthcoming IPCC fifth assessment report, run at higher resolution than older models, so they are able to resolve more regional features, and are expected to better represent the physics and dynamics of the atmosphere and ocean. Furthermore, they in general include more aspects of the system – they are true "Earth System" models, taking into account previously neglected processes such as atmospheric chemistry, aerosols, the terrestrial biosphere change, and ocean biogeochemistry. However, even these models are far from perfect, evident from the fact that even under identical scenarios, they make differing predictions. As such, it is important to consider predictions from a range of state-of-the art models. Such a task is currently being undertaken in the context of a recent project, "GeoMIP" (Geoengineering Modeling Intercomparison Project; [18]). This project defines two well-defined sunshade geoengineering experiments, which will all be carried out by a range of models (see Fig. 2.4). However, this multimodel approach still does not sample the full range of possible predictions. Each individual model has a large number of internal

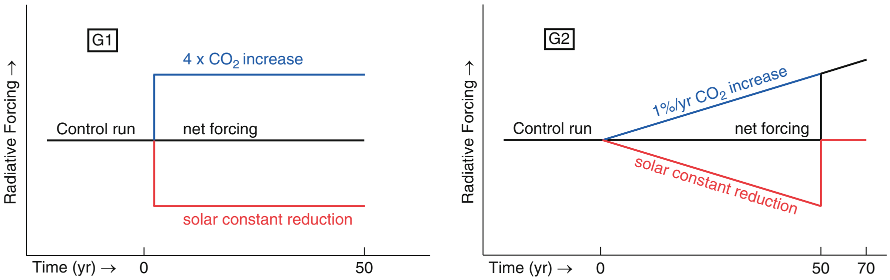

Fig. 2.4 Schematic of experiments"G1" and "G2" as defined in the GeoMIP project. G1: the experiment is started from a control run. The instantaneous quadrupling of CO_2 concentration from preindustrial levels is balanced by a reduction in the solar constant until year 2050. G2: the experiment is started from a control run. The positive radiative forcing of an increase in CO_2 concentration of 1% per year is balanced by a decrease in the solar constant until year 2050 (Adapted from [18], Figs. 2.1 and 2.2)

parameters which are not well constrained by observations, such as the relative humidity at which clouds start to form on the scale of a single model gridbox. As such, the very latest work is beginning to make predictions with an ensemble of models, which sample some of the model parameter space. This approach has been used for a number for years in future climate prediction (e.g., [19]), but work is just starting on applying this technique to geoengineering.

In terms of engineering solutions, it seems unlikely that a giant sunshade will be launched into space any time soon, due to the high costs associated with fabrication and/or launch, not to mention the possible legal, moral, and ethical questions. Other, cheaper, global geoengineering schemes are likely to be developed first, if at all, such as the injection of aerosols into the stratosphere. However, the concept of a sunshade in space does provide a neat idealized framework in which to study, with numerical models, possible geoengineering options. As such, the concept is likely to be investigated and discussed for some years to come.

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Chapter 3
Stratospheric Aerosols for Solar Radiation Management

Ben Kravitz

Glossary

Arctic oscillation	A pattern of sea-level pressures in the Arctic indicating large-scale circulation patterns. This serves as a proxy for the degree to which Arctic air penetrates to lower latitudes. During a positive mode of the Arctic Oscillation, which occurs both naturally and can be forced by large injections of stratospheric aerosols that enhance the polar jets, cold air gets trapped in the Arctic, resulting in warmer winter temperatures over the northern hemisphere continents.
Carbon dioxide removal (CDR)	Removing carbon dioxide from the atmosphere and sequestering it, either in geological formations or in the deep ocean, thereby decreasing the atmospheric concentration of anthropogenic greenhouse gases.
Pyrocumulus	A convective cloud that forms from a large amount of heating, and subsequent rising air, created by large fires.
Solar radiation management (SRM)	Reducing the amount of sunlight incident at the surface, thereby decreasing the globally averaged surface air temperature of the planet.

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Stratospheric aerosols	A layer of aerosols into the stratosphere which either scatter or absorb (or a combination of the two) a certain portion of sunlight that would, under normal circumstances, reach the surface.
Tipping point	The point at which global climate transitions from one stable state to another. Some extreme examples include ice ages, in which the climate rapidly cools, forming large ice sheets. The climate in this state is relatively stable, mainly due to an increase in planetary albedo, and is not easily returned to a different state (the present day climate, for example) without a large forcing of some kind.

Definition of the Subject

SRM in the context of this entry involves placing a large amount of aerosols in the stratosphere to reduce the amount of solar radiation reaching the surface, thereby cooling the surface and counteracting some of the warming from anthropogenic greenhouse gases. The way this is accomplished depends on the specific aerosol used, but the basic mechanism involves backscattering and absorbing certain amounts of solar radiation aloft. Since warming from greenhouse gases is due to longwave (thermal) emission, compensating for this warming by reduction of shortwave (solar) energy is inherently imperfect, meaning SRM will have climate effects that are different from the effects of climate change. This will likely manifest in the form of regional inequalities, in that, similarly to climate change, some regions will benefit from SRM, while some will be adversely affected, viewed both in the context of present climate and a climate with high CO_2 concentrations. These effects are highly dependent upon the means of SRM, including the type of aerosol to be used, the particle size and other microphysical concerns, and the methods by which the aerosol is placed in the stratosphere. SRM has never been performed, nor has deployment been tested, so the research up to this point has serious gaps. The amount of aerosols required is large enough that SRM would require a major engineering endeavor, although SRM is potentially cheap enough that it could be conducted unilaterally. Methods of governance must be in place before deployment is attempted, should deployment even be desired. Research in public policy, ethics, and economics, as well as many other disciplines, will be essential to the decision-making process. SRM is only a palliative treatment for climate change, and it is best viewed as part of a portfolio of responses, including mitigation, adaptation, and possibly CDR. At most, SRM is insurance against dangerous consequences that are directly due to increased surface air temperatures.

Introduction

Anthropogenic climate change has come to the forefront of scientific research as one of the most important problems facing society. The primary cause of climate change is the emission of greenhouse gases from fossil fuel combustion for production of energy, and the consequences have the potential to be severe [1]. Mitigation of emissions is the only viable permanent solution to the problem of climate change, but mitigation is likely to be difficult and expensive, and large-scale political will to abate emissions is currently lacking at a sufficient scale to prevent escalation of the problem.

As an alternative, there have been repeated proposals to deliberately alter the climate to counteract these changes, thereby avoiding some of the negative impacts. The proposed methods generally fall into two categories: SRM and CDR. Most of the solar radiation energy is contained within short wavelengths, to which the Earth's atmosphere is mostly transparent. This energy reaches the planet's surface and is absorbed, and the surface then radiates energy in longer wavelengths, which can be broadly interpreted as heat. Anthropogenic greenhouse gases, the predominant one being CO_2, absorb energy in certain bands of these longer wavelengths, thereby reducing the transmissivity of the atmosphere. Since the atmosphere retains this heat for a longer period of time, the atmosphere warms, radiating heat to the surface, and warming the surface. The purpose of SRM is to reduce this problem at the source by decreasing the amount of solar radiation that reaches the surface. Many methods for this have been proposed, but the one that has received the most attention and is likely to be the most feasible is placing a large amount of aerosols in the stratosphere, which will scatter or absorb the incoming solar radiation, preventing it from being converted to longwave energy. Conversely, the purpose of CDR is to remove CO_2 from the atmosphere and sequester it in long-term storage sites, generally in geologic formations or in the deep ocean. This has the potential to remove the cause of anthropogenic climate change and return the climate to a previous state.

This highlights that SRM and CDR are fundamentally different solutions to the climate change problem. Since climate change is caused by an increase in longwave energy, balancing this with a reduction in shortwave will necessarily result in imperfect compensation [2]. Conversely, CDR directly removes the source of the problem, so it will address all aspects of climate change, not just surface air temperature changes. SRM has the advantage that it will work quickly, reaching full effectiveness on the order of months to years [3]. CDR is likely to be slow and expensive, being effective at reducing climate change after several decades of sustained implementation [4].

Since SRM is only a palliative treatment and does little to reduce ambient concentrations of CO_2, if SRM were to cease, the climate would rebound to its pre-SRM state in a very short time [3, 5]. The ease of adaptation strongly depends on the length of time over which society can adapt, so the suffering felt during this rebound would be prominent. Therefore, if SRM were the only technology used to counteract climate change, due to the long atmospheric lifetime of CO_2 [6, 7], it would need to be done essentially forever.

One of the most commonly discussed methods of SRM is to mimic a large volcanic eruption by placing or creating large amounts of aerosols in the stratosphere. Large volcanic eruptions place SO_2 in the stratosphere which oxidizes to sulfate aerosols. These backscatter solar radiation, preventing it from reaching the surface, and the general effect is cooling for a short time after the eruption. This is primarily a radiative effect, independent of the way the aerosols were created, meaning to achieve surface cooling, aerosols could be placed in the atmosphere deliberately.

For SRM with aerosols to be feasible, the aerosols must be placed in the stratosphere. In the troposphere, the average aerosol lifetime is 1–2 weeks, meaning annual injection rates would need to be approximately two orders of magnitude larger than the desired equilibrium atmospheric loading of aerosol mass. However, the stratosphere is stable, resulting in atmospheric lifetimes of 1–3 years, depending upon the altitude and latitude of injection, as well as the aerosol size. Tropical injections would maximize this lifetime, as well as result in a more equal distribution of the aerosols throughout the global stratosphere, using the general atmospheric circulation to distribute the aerosols and transport them poleward.

Some past simulations have looked at optimizing cooling of the northern hemisphere continents, where the bulk of the world's population lives, instead of creating a worldwide aerosol layer, thus allowing for smaller injection amounts [8, 9]. However, doing so dramatically decreases the atmospheric lifetime of the aerosols, reducing their efficiency for SRM, as well as creating an interhemispheric imbalance of radiative forcing, which has profound implications for the hydrologic cycle, much more so than for tropical injections of aerosols.

The risks of SRM are potentially great, and at best, it is only a temporary measure, so some have questioned why research on SRM should even go forward [10]. The debate on SRM is exploring this question in great detail, but two answers have emerged prominently in the discussions. The first is to treat SRM as an insurance policy. If climate change were to reach dangerous levels, possibly even triggering climate tipping points, SRM gives society a means by which some negative impacts of climate change can be delayed, allowing time for a different, more permanent solution to be developed. Without having these emergency countermeasures available, the only choice for dealing with the impacts is adaptation, which is likely to be painful and costly [1, 11]. The second is pure scientific interest. SRM allows climate scientists to investigate scenarios that may not have previously occurred to them, allowing for new ways of thinking about the climate system and new understanding of physical processes. Regardless of whether SRM is actually implemented, this new knowledge is valuable to the scientific community.

History and Brief Literature Summary

The history of SRM is surprisingly lengthy, considering most of the attention to this topic has been concentrated in the last 5 years [12]. Arguably, the first mention of SRM, although not specifically in those terms, was by von Neumann in 1955 as an editorial opinion piece [13]. Although he understood mechanisms of climate change, including the ability of large volcanic eruptions to cool the climate and that warming would cause large-scale changes in ice sheet coverage, he made no mention of the connection between anthropogenic greenhouse gases and a warming planet. The first to make this connection in the context of SRM was Budyko [14, 15]. He understood that society's quality of life, obtained from combustion of fossil fuels, had the potential to cause massive changes in the climate system and that society may wish to deliberately cause compensatory cooling of the planet. Knowing that large volcanic eruptions cool the climate due to a layer of stratospheric sulfate aerosols, he did preliminary calculations of the amount of sulfate necessary to cause cooling to a certain level, also suggesting the amount of cooling could be controlled.

The study of SRM proceeded sporadically over the next three decades, with few papers and a small handful of reports. The National Academy of Sciences issued a report dealing with the practicality of inserting large amounts of aerosols into the stratosphere [16]. Some of the ideas they proposed include putting the material into a cargo airplane and flying it up to the stratosphere, firing it into the stratosphere using artillery, lofting it with weather balloons, or pumping it up a large tube, either through a tall tower or tethered to a stationary floating anchor (Fig. 3.1). Their rough estimates of costs suggested that SRM could be performed cheaply and, in some cases, with already existing technology.

This was followed by one of the first reviews of aerosol approaches to SRM by Dickinson, who provided preliminary estimates based on the eruption of Mount Pinatubo in 1991 of how much SRM would be required to cool the planet [17]. The next decade saw the first climate model simulations of SRM, performed by Govindasamy and Caldeira, in which they simulated a reduction in solar radiation to compensate for an increase in carbon dioxide concentrations [18].

Many of the reports early in the studies of SRM were completed at Lawrence Livermore National Laboratory under the guidance of Edward Teller [19–21]. These reports were often the source for many new ideas of how to perform SRM, including the use of black carbon aerosols, as well as some of the first estimates of costs. Most of the subsequent research over the next few years is summarized in a series of reports and articles [22–24] which tended to concentrate on the evaluation of the climate impacts of SRM.

SRM research received a large rejuvenation from a special issue of *Climatic Change* in 2006, most prominently due to the Nobel Prize winner Paul Crutzen [25]. His salient and timely calculations, showing SRM would likely require a small amount of effort when compared to mitigation, combined with worries about the implications of this conclusion, remain relevant and are often revisited by SRM researchers in all disciplines. He helped legitimize the study of SRM, resulting in an explosion of

Fig. 3.1 Various means of accomplishing SRM (Reproduced from Fig. 1 in [56]. Drawing by Brian West)

research and publications over the following years that continues to this day. This article was concurrent with simulations by Wigley, which confirmed that sustained injections of large amounts of stratospheric aerosols will cause surface cooling [5]. However, one of the most important findings of this study was that if SRM is stopped but CO_2 concentrations are not reduced, the climate will rapidly return to its pre-SRM state, making adaptation to the resulting change nearly impossible.

Another significant effect of SRM, as discussed by Trenberth and Dai the following year, is suppression of the global hydrologic cycle [26]. Although their findings were based on the evaluation of the effects of the eruption of Mount Pinatubo as an analogue, these findings were reinforced by Robock et al., who performed the first simulations of stratospheric aerosol SRM using a fully coupled atmosphere-ocean general circulation model [9]. This study was concurrent with a review paper by Rasch et al., comparing different modeling groups' simulations of SRM [27].

The first study of SRM with stratospheric sulfate aerosols to use a microphysical model found that aerosol microphysics plays a very important role in determining the particle size distribution and that nearly all past simulations of stratospheric sulfate SRM used an aerosol size that was much too small [9, 28, 29]. This suggests

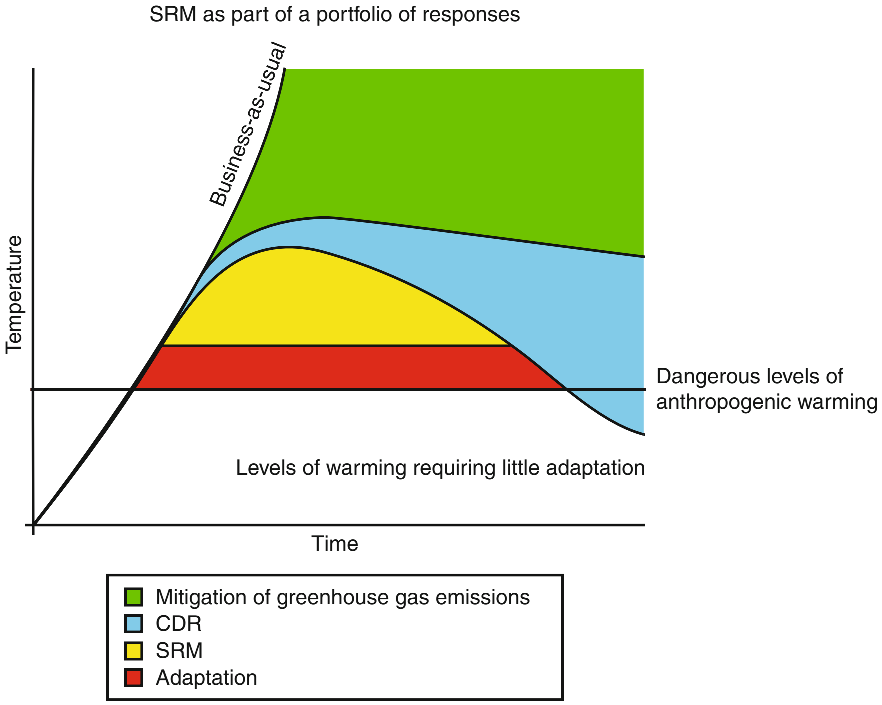

Fig. 3.2 SRM when viewed as part of a portfolio of responses to climate change, adapted from a personal communication with John Shepherd. Axes are left unlabeled deliberately to avoid assumptions about policy decisions or estimates of climate sensitivity

that SRM using injections of SO_2 would either require much more care and study or injections of much larger amounts [30].

The next stage of climate model research will largely be conducted under the framework of The Geoengineering Model Intercomparison Project (GeoMIP), which at the time of writing this entry is underway [31]. The need for this project, as well as a description, can be found in the last section.

Some attempts have been made to classify different SRM and CDR schemes and rate their effectiveness [32–35]. The findings generally show that SRM with stratospheric aerosols is likely to be cheap and highly effective, especially when compared to many other means. However, the uncertainties of this idea are quite large, and the adverse consequences are potentially severe, often more so than the other more expensive, less effective means.

Perhaps the most helpful view of SRM has arrived in recent years, in that it should be treated as part of a portfolio approach to climate change. Mitigation and possibly CDR are the only permanent solutions to anthropogenic climate change, but these are expensive and will take a long time to implement, during which time some dangerous levels of warming and their negative impacts could be felt. In this sense, SRM could be used to temporarily alleviate a small amount of warming to stay below the 2°C policy target [5] (Fig. 3.2).

Types of Aerosols

SRM with stratospheric aerosols implies that different kinds of aerosols could be used, each with their own array of impacts and consequences, some advantageous and some disadvantageous. It is well beyond the scope of this paper to discuss every possible aerosol, but two choices can be treated as bookends of a spectrum study. Sulfate aerosols backscatter sunlight almost perfectly, whereas black carbon aerosols are excellent absorbers, so the scattering properties of nearly any choice of natural aerosol actually used for deployment can, to first order, be seen as a linear combination of these two. Should an engineered, manufactured particle be deployed, perhaps the scattering properties could be tuned to be optimal, so for this case, properties other than extinction become of higher concern. In this sense, a great deal of information regarding the atmospheric physics impacts of SRM with stratospheric aerosols can be gained by study of only three kinds of aerosols: sulfate, black carbon, and engineered particles. Such a study must not only include the physical effects and climate impacts, but also practicality, i.e., how the aerosols can be created, placed in the stratosphere, and whether they will actually work at cooling the surface of the planet.

Sulfate

The most researched aerosol in the context of SRM is sulfate aerosols, mainly due to the well-studied analogue of large volcanic eruptions. The climate effects of large volcanic eruptions are well known [36]. The primary driver of these effects is the layer of stratospheric sulfate aerosols created by injection of SO_2 gas from the volcano. If the eruption is sufficiently large and at the proper time of the year [37], the aerosol layer will backscatter a sufficient amount of solar radiation to cause surface cooling, which primarily occurs over the northern hemisphere continents, since land has a lower heat capacity than the oceans. The summer of the year following the 1991 eruption of Mount Pinatubo, which was the largest volcanic eruption of the last 50 years, showed reduced globally averaged surface air temperatures by approximately 0.5°C [38].

Although volcanic eruptions are an inspiration for SRM, and many of the effects and impacts of large volcanic eruptions are due to a stratospheric aerosol layer, implying the effects would be similar, they are an imperfect analogue. Volcanic eruptions involve a pulsed injection of sulfate aerosol precursors. After the aerosols form, they begin to coagulate and fall out of the stratosphere, meaning the total stratospheric mass of aerosols decays from an early peak, until it reaches the levels before the eruption. To ensure a constant level of radiative forcing that would be required for SRM, a steady state is required, which necessitates smaller, regular injections, if not continuous injections.

Sustaining an aerosol layer would certainly have some differences from a pulse injection. As the aerosols stay in the stratosphere longer, they will grow from condensation of water onto the aerosols and coagulation of particles [39]. Therefore, the average particle size under SRM would likely be much larger than for volcanic eruptions, which reduces both the scattering efficiency of the aerosols and the atmospheric lifetime by increasing fall speed. Most simulations of stratospheric SRM using general circulation models are highly idealized in terms of the achieved sulfate aerosol forcing per unit mass, as they often neglect coagulation processes [28]. To achieve the radiative forcings that are often reported in the literature for a given stratospheric injection of SO_2 would likely require an order of magnitude more mass due to the microphysical effects [30]. Simulations have suggested that this significant hurdle could be overcome if pulsed injections were used instead of continuous injections [28]. Also, using aerosol precursors such as SO_2 could be bypassed entirely by directly condensing gaseous sulfuric acid into the atmosphere to achieve a monodisperse droplet size, although this idea requires further study and testing [30]. Moreover, any reduction of solar radiation will result in ocean memory of the cooling due to the ocean's thermal inertia, meaning surface cooling under SRM will necessarily be greater than cooling from a large volcanic eruption [40].

The negative impacts of SRM with sulfate aerosols have the potential to be quite severe. One of the most discussed aspects of the climate effects of SRM is changes to the hydrologic cycle. The eruption of Pinatubo dramatically suppressed the hydrologic cycle, leading to drought in many parts of the world, so SRM could reasonably be expected to have similar effects [26]. Some simulations of sulfate aerosol SRM have confirmed this expectation, showing a large reduction in the Indian/African/Asian monsoon system [9], although efforts to determine whether this effect is robust have not yet been conclusive [31, 41]. The impacts of SRM manifest on both a global and a regional scale, resulting in regional inequalities for any level of globally implemented SRM [2].

Another problem is that introduction of large amounts of aerosols into the stratosphere would damage the ozone layer through enhanced thermal decomposition from increased stratospheric temperatures and enhanced halogen-catalyzed depletion [42–44]. The eruptions of El Chichòn and Pinatubo both resulted in modest but noticeable depletion of the ozone layer [45, 46]. Simulations of SRM using models with sophisticated chemical components have confirmed this, showing a delay in the recovery of the Antarctic ozone hole by decades [47, 48].

Not all ancillary impacts of SRM would be negative. After the eruption of Pinatubo, due to the increased light scattering from the sulfate aerosols, the fraction of diffuse light increased significantly, promoting plant photosynthesis across most of the globe [49]. This showed that stratospheric aerosols could address climate change directly by increasing the biosphere carbon sink. The 2 years after Pinatubo showed an increase in carbon uptake by over 1 Pg a^{-1}, which resulted in a measurable drawdown of atmospheric CO_2 concentrations [50, 51]. The land carbon sink has also steadily increased from global dimming, giving further credence to this mechanism. However, the increase in the diffuse fraction of incident sunlight would negatively impact solar power generated from large thermal generation plants, with the amount of power scaling approximately linearly with the amount of available direct sunlight [52]. These two effects are simply due to the presence of scattering aerosols, meaning SRM would likely have similar effects.

An often cited concern of sulfate SRM is that when the aerosols eventually pass into the troposphere, they will fall as acid rain. However, current worldwide sulfur emissions are over 100 Tg [53], so any reasonable amount of SRM would be at least an order of magnitude less, meaning all but the most poorly buffered ecosystems would be negatively affected by the additional acid rain [54, 55].

Assuming SRM with sulfate aerosols was to be undertaken, the next natural question is how the aerosols can be placed in the stratosphere. The most commonly discussed means is through large injections of SO_2, mimicking volcanic eruptions and allowing the atmosphere to convert the SO_2 gas into sulfate aerosols. However, as discussed previously, microphysical issues would likely make this infeasible, prompting brainstorming of other methods [30].

The next stage of thought is transporting the aerosols or aerosol precursors to the stratosphere. Many means of this have been suggested in the past, including using airplanes, balloons, artillery, and space elevators [16, 56] (Fig. 3.1). Likely the most efficient means of achieving this, as well as the cheapest, is using planes to transport the material. Engineering considerations aside, this method is probably very cheap when compared to the cost of mitigation or climate change impacts, and cheap enough that it could be done unilaterally [56, 57]. The implications of this invite myriad legal, governance, ethical, and economic issues, but much more work on these topics is needed.

Black Carbon

Despite being repeatedly suggested throughout the history of SRM research [19, 21, 25, 40], formal study of deliberate large stratospheric injections of black carbon aerosols is distinctly lacking. At the time of the preparation of this entry, only one such study has been performed [58].

Black carbon SRM analogues are not nearly as closely related to SRM as is the case for sulfate aerosols. The only large injections into the stratosphere of black carbon aerosols are from pyrocumulus events, which occur from large fires, but the amount of stratospheric black carbon that results from these events is quite small and localized compared to the amount that would be needed for SRM [58, 59]. Measurements below the plumes of these large fires show surface cooling, lending credence to their consideration for SRM [60, 61].

Another analogue, albeit only in simulation form, is the climate effects of nuclear winter. The fires from nuclear war would cause pyroconvection, similar to pyrocumulus events but on a massive scale, lofting several megatons of soot into the stratosphere, which would reduce sunlight reaching the surface, causing cooling [62–67]. The amount of cooling that would result from such an injection would more than offset anthropogenic warming, causing enough temperature reduction to be catastrophic to agriculture [68]. Also similarly to sulfate aerosol SRM, these analogues involve one-time injections of aerosols, whereas SRM would involve continuous injections and a sustained radiative forcing.

Most of the negative impacts from stratospheric SRM with black carbon aerosols would be from the massive stratospheric heating due to absorption of sunlight by the aerosols. The heating would be much more than for sulfate aerosols, and the soot particles would also self-loft, potentially reaching the mesopause until the ambient air pressure can no longer suspend the particles [58, 69–71].

One of the most prominent side effects would be ozone depletion from accelerated thermal decomposition, much more than would be found for an equivalent mass of sulfate aerosols [42–44, 58, 71]. Additionally, the heating would allow more water vapor to enter the stratosphere, which would cause ozone destruction through chemical interactions [71]. Heterogeneous chemistry on the surface of the particles would also likely cause ozone depletion to some extent, but these effects are largely unknown.

The stratospheric heating would likely cause circulation changes by strengthening the polar vortices, forcing a positive mode of the Arctic Oscillation and potentially introducing an Arctic ozone hole [58]. Additionally, not only is black carbon hazardous to human health [72], but if black carbon falls on fresh snow, it would reduce the surface albedo, causing more absorption of sunlight by the surface, which is counterproductive to the purpose of SRM [73].

The effectiveness of black carbon SRM on reducing surface temperatures strongly depends on the particle size and altitude of injection, which has a direct impact on the feasibility of this choice of aerosol. The more efficiently a certain combination of SRM parameters cools the surface, the more efficiently the aerosols will heat the stratosphere, causing negative impacts. Smaller aerosols are more radiatively efficient and have a slow fall speed, resulting in more surface cooling per unit mass. If the aerosols are too large, they will actually have an effect in the longwave, resulting in warming [74].

The concerns with and mechanisms of deployment of SRM with black carbon aerosols are similar to those of sulfate aerosols. Soot is a by-product of incomplete combustion and not a gaseous aerosol precursor, so the means of creating the aerosols is different, but costs are of a similar order of magnitude. Transporting the aerosols to the stratosphere dominate the costs and feasibility of this method, but these costs are highly variable, dependent upon the way the aerosols are transported up to the stratosphere.

Engineered Particles

The drawbacks of aerosols with natural analogues, like sulfate and black carbon, have prompted the study of manufactured particles that are specifically engineered to optimize scattering of sunlight while avoiding some of the less desirable attributes. These engineered particles have been discussed briefly in the literature, mostly as thought experiments, as the feasibility of producing large amounts of these aerosols is uncertain.

One of the first mentions of this idea was by Edward Teller who wanted to optimize both the scattering properties and atmospheric lifetime by creating an efficient “self-lofting blue/UV chaff” or “resonant scatterers” [20, 21, 40]. Creating particles that photophoretically levitate is also possible, the advantage being that the atmospheric lifetime would be lengthened, so considerably fewer particles would be needed for a given amount of cooling [75, 76]. However, this has the serious problem of irreversibility of SRM unless a mechanism for removing the particles from the mesosphere could be designed.

One could certainly conceive of other desirable properties an aerosol might have as well as potential designs for it. The potential for research on engineered particles is large, as this idea is largely unexplored.

Future Directions

Although SRM with stratospheric aerosols has received a great deal of attention and study, this idea is still far from deployment-ready, notwithstanding the issue of whether deployment is necessary. However, assuming stratospheric aerosols is the method of choice for modifying the climate, the next stage of research must still resolve some serious fundamental questions.

Perhaps the most basic question that can be asked, which still has no satisfactorily all-encompassing answer, is: “What is SRM?” Implicit in this question is not just the method, but also the means. The scientific community, and more importantly, the policy community, has still not defined what SRM is supposed to do. Depending on what kind of aerosol is used, the aerosol size, the method of stratospheric delivery, and the location and timing of injection, as well as myriad other decidable parameters, the cooling effects and climate impacts of SRM can span a very large range and, to some extent, are tunable [9, 30, 37]. This invites a question often asked by Alan Robock, “Whose hand is on the thermostat?” [77, 78]. Moreover, this question suggests the most important variable to be adjusted by SRM is surface air temperature, but perhaps other climate variables could be tuned by adding more “thermostats.”

Assuming this decision could be made, SRM will not necessarily behave as predicted or intended. The radiative effects for given aerosol parameters can be determined quite well with various climate models at the disposal of the scientific community, but determining which radiative parameters are applicable to SRM is more uncertain. The recently highlighted issue of aerosol microphysics [28, 30] suggests other physical science issues with SRM could easily arise, dramatically changing the knowledge field. Equally as important are Donald Rumsfeld’s famous “unknown unknowns,” a phrase particularly applicable to a field with so much uncertainty. The only way to truly be sure of the effects of SRM is to conduct a test, but regarding tests involving actually placing aerosols in the stratosphere, the line between testing and deployment is quite blurred and the ability to observe the results of these tests is severely hampered by low signal-to-noise ratios [79]. Testing also has serious implications for issues of governance, which are well beyond the scope of this paper, although efforts are underway to investigate the issues involved (SRMGI, http://www.srmgi.org).

Despite the issues and inherent uncertainty with climate models, they are one of the most useful tools available to SRM research. Until recently, SRM studies with climate models were sporadic and uncoordinated, leading to modeling groups performing experiments that were insufficiently similar for comparison [27]. Even among modeling groups that did conduct similar experiments, some results were dissimilar enough to prevent robust conclusions on the expected climate effects from SRM from being reached [41]. The Geoengineering Model Intercomparison Project (GeoMIP, http://climate.envsci.rutgers.edu/GeoMIP/) provides a framework of coordinated experiments for all modeling groups wishing to participate, eventually allowing for robustness of results to be achieved [31]. In the project's current stages, the four proposed experiments are underway (Fig. 3.3) and, as of the time this entry was written, are being conducted by 16 different climate modeling groups.

Climate models used on a global scale have difficulty with accurate resolution of regional and local impacts, which will likely be some of the most important issues in the debate on SRM. Although assessment of these effects has begun [2], this direction will likely be one of the dominant avenues in the future of SRM research. Another important aspect in future research will likely be the increasing problem of ocean acidification, which SRM does little to prevent [80]. These reasons, and many others, have been suggested repeatedly as important investigations in the course of SRM research [77, 78].

If SRM were to be deployed, society's current ability to monitor the effectiveness of deployment has some prominent gaps. For such an endeavor, a global observation network is essential, and one of the most important parts of such a network would be satellites specifically geared to observe stratospheric aerosols and their climatic impacts. Although such instruments for directly observing aerosols have existed in the past, providing excellent observations of past volcanic eruptions [81], currently only one similar instrument exists [82]. Many of the current instruments in orbit are aging, and replacement plans are desperately needed to ensure uninterrupted observations. Before SRM deployment can even be considered, the observation network will require serious assessment and improvement on a large scale.

This entry has mostly focused on physical science aspects of SRM. Although such concerns are fundamental to understanding SRM, during the decision-making process of whether to undertake SRM, or even some forms of SRM research, physical science results should not and likely will not be the sole determining factor. Discussions will include myriad different values, necessitating research in many different fields, including (but certainly not limited to) law, governance, economics,

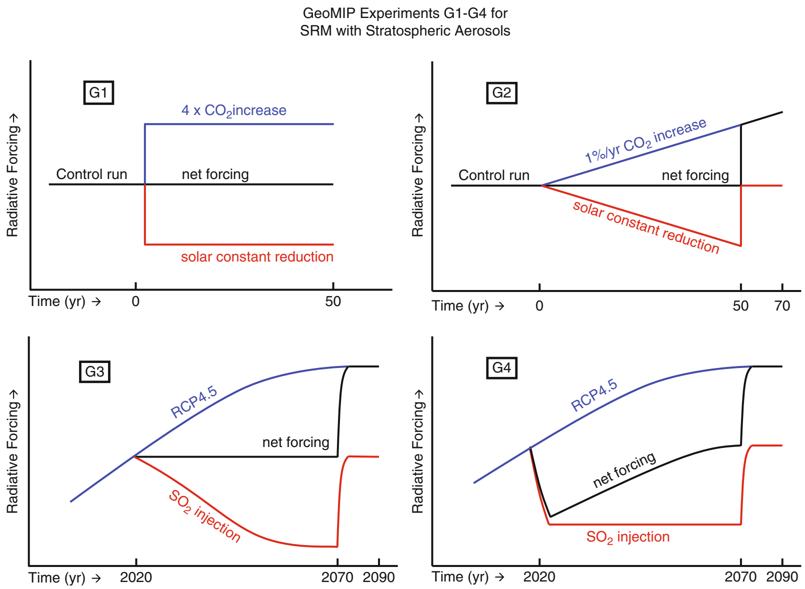

Fig. 3.3 Outlines of the four coordinated experiments (G1–G4) to be performed by the participants of GeoMIP. Panels are combined from Figs. 1–4 from [31]

ethics, and social policy. Research in these fields is essential and ongoing, but like physical science research of SRM, much more work is needed.

SRM has the potential to relieve some of the expected consequences of climate change, but not without numerous possible consequences, all of which must be evaluated. The decision to deploy SRM, as well as how long to deploy until mitigation has taken effect, must include all information possible to achieve the best outcome for society in the face of the daunting problem of climate change.

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Chapter 4
Solar Radiation Management, Cloud Albedo Enhancement

Stephen H. Salter

Glossary

Albedo	Whiteness or reflectivity. The ratio of outgoing to incoming radiation power.
Cloud condensation nuclei	The small fragment of material needed to start the growth of a cloud drop when the local relative humidity exceeds 100%.
MODIS	An acronym for moderate resolution imaging spectroradiometer. This is an instrument carried by two NASA satellites Aqua and Terra. The systems measure 36 spectral bands of radiation of the entire earth surface. Results are available free and are an invaluable aid to research into climate change.
Permafrost	Ground which remains frozen throughout the year. It can trap very large amounts of methane which would be released if it were to melt.
Positive feedback	A change in the value of a variable which leads to a further change in the same direction. It can often produce explosive effects and oscillations.
Relative humidity	The ratio of the amount of water vapor in a parcel of air to the maximum amount that it could hold if nucleating particles were present. Values can exceed 100% and even reach 300% if all nuclei are absent.

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Solar constant	The rate at which solar energy of all wavelengths crosses unit area at the top of the atmosphere. It is about 1,366 W/m^2 but is strictly not quite constant. There are cyclical variations with an amplitude of about 3.5% due the ellipticity of the earth's orbit around the sun, 11 year cycles and faster random variations of about 0.1%.
Twomey effect	The relationship of cloud albedo to cloud depth, liquid water content and concentration of cloud condensation nuclei elucidated by Sean Twomey. His work began following observation of increased albedo of ships tracks in satellite photographs.

Definition of the Subject

Cloud albedo enhancement is one of several possible methods of solar radiation management by which the rate of increase in world temperatures could be reduced or even reversed. It depends on a well-known phenomenon in atmospheric physics known as the Twomey effect. Twomey argued that the reflectivity of clouds is a function of the size distribution of the drops in the cloud top. In clean mid-ocean air masses, there is a shortage of the condensation nuclei necessary for initial drop formation in addition to high relative humidity. This means that the liquid water in a cloud has to be in relatively large drops. If extra nuclei could be artificially introduced, the same amount of liquid water would be shared among a larger number of smaller drops which would have a larger surface area to reflect a larger fraction of the incoming solar energy back out to space.

Introduction

At noon in the tropics, the top of the Earths' atmosphere receives about 1,350 W/m^2 from the sun. This is known as the solar constant but varies slightly with solar activity and orbital changes. Because of the high source temperature, most of the solar energy is spread over the short wavelengths – those to which our eyes have evolved. Some is reflected back to space, and some is absorbed by gases and particles in the atmosphere, thereby increasing their temperature. The amount reaching cloud level over 24h is about 440 W/m^{2}at the equator and about 340 W/m^2 globally. Some radiation is absorbed by clouds. Some passes through to reach the ground. Radiation from bodies at lower temperatures is at much longer wavelengths where atmospheric gases have different absorption properties.

Changes in the concentrations of atmospheric gases will affect the amount of heat retained, and the temperature then has to rise to restore the energy balance by increasing outward radiation. While much attention is given to carbon dioxide, many other gases are involved, some with stronger absorbing properties, particularly water vapor and methane. The maximum atmospheric water vapor concentration increases with temperature giving a positive feedback, but it can be rapidly returned to the sea as precipitation and condensation. A more abrupt and less reversible positive feedback could be triggered by the release of methane from melting permafrost.

It is widely thought that the change of atmospheric carbon dioxide and other greenhouse gases since pre-industrial times together with the resulting water vapor amplification has increased the amount of heat retained by the earth at a rate of about 1.6 W/m^2. This is only 0.0047 or one part in 212 of the 340 W/m^2 mean 24 h input. It is therefore possible that a very small increase in the reflectivity of the earth could reduce, cancel, or even reverse the anthropogenic increase and so provide time for the development of low-carbon energy sources and carbon sequestration.

Clouds cover a substantial fraction of the earth's surface and are good reflectors of solar energy. However, cloud drops cannot form without an initial small "seed" known as a condensation nucleus. In 1990, John Latham [1] proposed that the reflectivity of marine stratocumulus clouds could be increased by spraying submicron drops of sea water into the marine boundary layer. The drops would evaporate to leave salty residues. Air flows in the marine boundary layer are quite turbulent, and the residues would soon be well mixed through it. Salty particles are hydrophilic and so are ideal condensation nuclei. The most effective size range for nucleation is between 0.5 and 1 μm. Those nuclei which reach cloud level provide the seeds for new drops and so trigger the Twomey effect. Twomey [2] argued that the reflectivity of a cloud depends on its depth Z in meters, the liquid water content L in milliliters of water per cubic meter of air, and the drop concentration expressed in the number per cubic centimeter.

It may be convenient to rewrite the equations in Twomey's original paper with an intermediate term $K\ (Z, L, n) = 0.15\ Z\ L^{2/3} n^{1/3}$. Cloud albedo A is then

$$A(Z, L, n) = \frac{K(Z, L, n)}{K(Z, L, n) + 0.827}$$

Figure 4.1, adapted from Schwarz and Slingo [3], shows a plot of the relationship. For the typical range of conditions, a doubling of the number of nuclei will increase reflectivity by 0.056 from an initial value of 0.3–0.7. The vertical bars mark present condensation nuclei concentrations for clean mid-Pacific air and dirtier Atlantic air suggested by Bennartz [4] based on analysis of satellite observations.

It is the fractional increase in the number of nuclei in the effective diameter range that matters, not the amount of water sprayed. This means that the technique is far more effective in clean air and has diminishing returns as treatment progresses. If atmospheric observations are taken from short-range aircraft, there will be a bias towards higher numbers. Figure 4.2 shows worldwide drop concentration supplied by Wood [5] from the MODIS instrument on Aqua and Terra

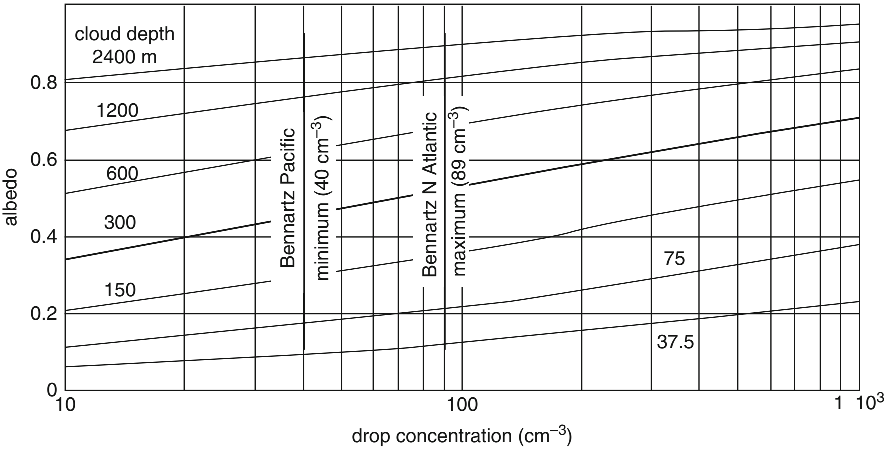


Fig. 4.1 Twomey's results for cloud albedo as a function of cloud depth, a typical liquid water content of 0.3 g/m^3 of air and drop concentration (Adapted from Schwarz and Slingo [3] with Bennartz [4] data for drop concentration)

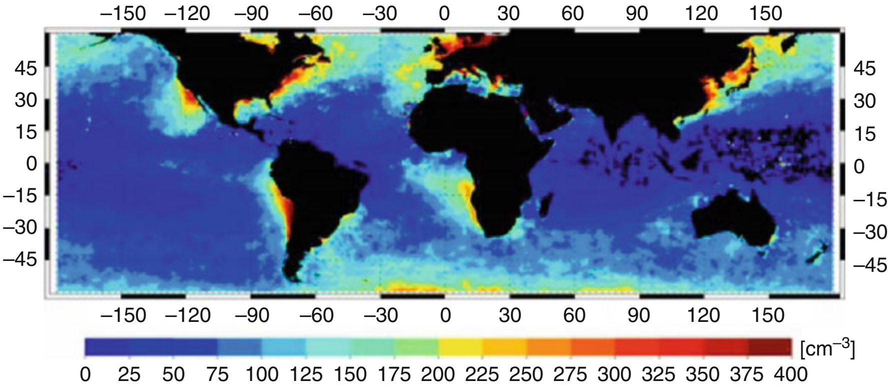
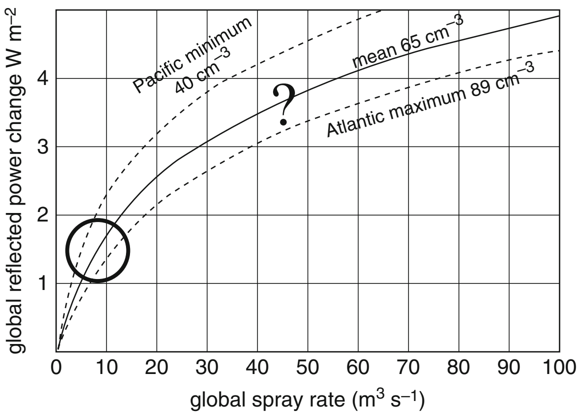


Fig. 4.2 Cloud droplet concentration for warm clouds as estimated by the MODIS instruments (Courtesy of NASA and Robert Wood of the University of Washington)

satellites [6]. Large areas of mid oceans have concentrations below 50 cm^{-3}. In some air masses, which have been over Antarctica for long periods or washed by recent rain, the concentration can be as low as 10 cm^{-3}. In contrast, values over land can be 1,000–5,000 cm^{-3}.

Latham found that, with the correct drop size, the amounts of spray needed to give a useful reduction of incoming power are surprisingly small. Figure 4.3 shows cooling for 340 W/m^2, a fairly typical selection of cloud depth liquid water content, the nuclei concentrations suggested by Bennartz and their mean. There is some uncertainty about lifetime of the condensation nuclei. This graph uses a half-life of



assumptions and sources	
boundary layer depth	1000 m
cloud depth	300 m
Schwartz and Slingo (1996)	
liquid water	0.3 ml m^{-3}
Schwartz and Slingo (1996)	
drop life	59 h
Smith et al. (1991)	
low, not high, cloud fraction	0.18
Charlson et al. (1987)	
initial albedo	0.495
24 h power in	340 Wm^{-2}

Fig. 4.3 Global cooling as a function of spray rate for the assumptions in the table to the right. The circle shows 1.6 W/m^2, often taken as a figure for anthropogenic change since pre-industrial times. The question mark might be the result of a doubling of carbon dioxide

59 h from Smith et al. [7] and is in good agreement with Hoppel et al. [8] which shows half-life as a function of drop diameter. If the assumptions on the right of the figure are reasonable, then the thermal damage since pre-industrial times could be offset by spraying about 10 m^3/s worldwide.

Several independent climate models [9, 10] support the effectiveness of Latham's proposal, and in some conditions, in clouds with no internal drizzle, the models predict an albedo change exceeding 0.4. In very rare circumstances with exceptionally clean air, it may be possible to produce clouds from a clear sky. However, the intention is to make existing clouds whiter.

Energy Requirements

The minimum energy requirement to produce a drop is the product of its surface area and its surface tension. For sea water, the surface tension is 0.078 N/m, a little higher than for fresh water, so making a 0.8 μm diameter drop needs at least 1.57×10^{-13} J. When a condensation nucleus reaches the cloud level, it will split an existing drop into two smaller ones with the same volume of liquid but with a reflecting area increased by the cube root of two, about 1.26. Figure 4.4 shows the range of drop sizes (in radius as used by scientists, not in diameter used by engineers) with much larger ones over mid ocean.

The solar energy that will be reflected by the two new drops will depend on the input power density, the time before the two drops coalesce and the effective reflection coefficient. The reflection coefficient of drops is an extremely complicated function, Mie [11] Laven [12], of wavelength, polarization, and angle of

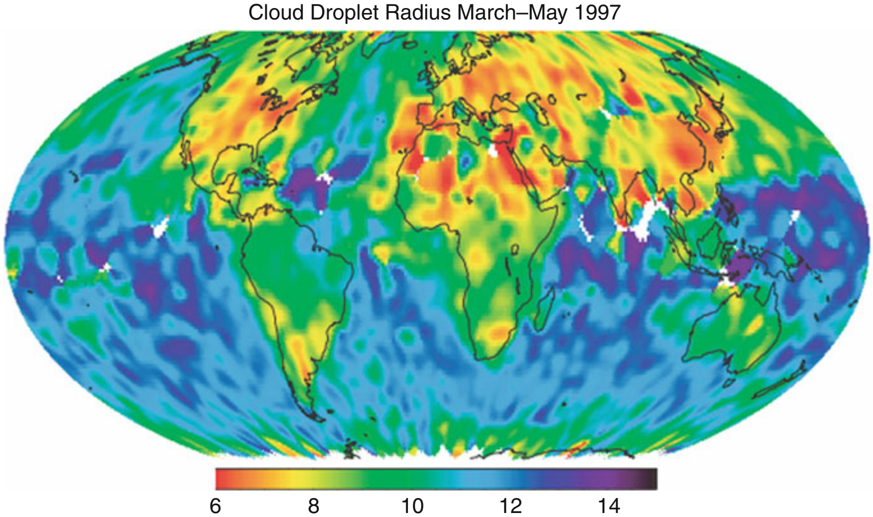


Fig. 4.4 Cloud droplet *radius* derived by Breon Tanre and Generoso (Courtesy of Science. From http://www.sciencemag.org/content/295/5556/834.full.pdf)

Fig. 4.5 The jar on the *left* contains 4 mm glass balls. The *grey scale* behind the jars has 10 bars going from roughly zero to nearly 100% whiteness depending on your printer and paper quality. This shows that the albedo of the 4 mm balls is about 0.6. The jar on the *right* contains 40 µm glass balls, 100 times smaller, and has an albedo of about 0.95

incidence but is of the order of 10%. Few people can believe the magnitude of the ratio of energies of reflected solar radiation to nucleus creation until they have done the calculations for themselves. Figure 4.5 demonstrates the effect of drop sizes on reflectivity using glass balls.

Treatment Sites

Good sites for spraying need incoming sunshine, low marine stratocumulus clouds, and clean air. The clean air requirement points to regions away from land masses and shipping routes. The sunshine requirement suggests the ability to migrate with the seasons so as to make use of the long hours of summer sunshine at high latitudes. The cloud requirement is less critical than the other variables. Cloud coverage may be inversely related to longer drop life. Spray produced under clear skies evaporates to leave a crystal of dry salt which would be much smaller than the typical 25 μm diameter liquid drop in a cloud but which would have an albedo well above a liquid drop and a lifetime which could be much longer. Furthermore, it is better to have a low dose over wide a area, and spray under clear skies would be more widely dispersed. It might be attractive to spray in air which we expect will later move to a cloudy region.

Mobility, energy, and long duty periods can be provided by unmanned sailing vessels. The chief engineering problem is the production of the correct diameter spray from a mechanism with a long service life, low energy requirements, and no need of supplies from land. A narrow range of diameters may reduce the probability of drop collision leading to coalescence which would shorten drop life and it may turn out to be useful to adjust diameter with remote commands. A number of techniques are being studied by Neukermans (2010, 2011, personal communication), Salter [13].

Environmental Impacts

Evaporation of water from the oceans is about 14 million tonnes a second of which about 12.8 million tonnes per second is precipitated back to the sea. There is no need to fear anything from the direct water output from spray vessels.

Breaking waves produce a wide range of drop sizes all over the oceans but especially at beaches. It is not easy to get a number for the salt transfer but Blanchard [13] explains the ways that an attempt could be made. Grini et al. [14] survey more recent estimates. These range from 1000 to 10,000 million tonnes a year or from 30 to 300 tonnes a second. Ten tonnes of sea water, a second needed to cancel 1.6 W/m^2, would transfer 350 kg of salt per second. What makes the salt from spray vessels so much more effective is the controlled size of the salt residues. Most of them would be returned to the sea and so would be very small compared with spray from beaches, much of which goes inland.

However, Victorian doctors knew very well that sea air was good for lung infections. A Polish doctor F Boshkowsky noticed that his patients who worked in the salt mines had no lung illness and enjoyed long lives despite hardworking conditions. In Eastern Europe, salt mine exposure is used for treating asthma, and in the West, there is a brisk trade in salt pipes. There is no need to worry about the effect of salt from spray vessels on human health.

The immediate effect of albedo control is a reduction of the sea surface temperature below the treated cloud. The reduction will be small compared with the normal changes from cloud to clear sky and even less compared with the change from day to night. But sea surface temperatures are a very strong driver for many other meteorological parameters. The laws of thermodynamics predict that although the effect will eventually diffuse worldwide, there can also be local deviations, and operators will have to build up a clear understanding of all the outcomes.

The greatest anxiety about side effects concerns changes to precipitation. Changes in either direction could be bad. During 2010 and 2011, there was severe drought in Russia and the most violent flooding in Pakistan, Queensland, Brazil, and Sri Lanka before anyone had attempted any form of intentional geoengineering. It may be that we have to choose the least bad outcome. But if we understand meteorology properly, we may be able to improve things in different places by drying out wet regions and getting more rain to dry ones. At least eight mechanisms are involved, and some, especially precipitation, are not well represented in present climate models. As so often in meteorology, factors can work in opposite directions, and we may be ignorant even of the polarity, let alone the magnitude, of the final outcome.

The mechanisms so far identified are:

1. The production of rain needs some large drops to fall through deep clouds fast enough to coalesce with other drops in their path so that they grow large enough to fall fast enough to reach the ground without evaporating in the dry air below a cloud. It is known from Albrecht [15] and Konwar et al. [16] that rain is less likely if there are too many small drops due to smoke and fine dust. This happens over land and could happen as a result of cloud albedo control but the depth of marine stratocumulus is mostly too thin to form rain and the effect might be to extend cloud lifetime.
2. However, if more small cloud drops means that there is less rainfall *over the sea*, it follows that there will be more water left in the air mass when it reaches land. The air mass will travel further inland before the original drop numbers are restored by coalescence. Rain inland is more valuable than rain at the coast.
3. Any reduction in sea temperature will tend to reduce evaporation and increase condensation from air back to the sea surface and this will reduce the water content of the atmosphere. The increase in evaporation rate increases sharply at sea surface temperatures around 28°C [17] and may have contributed to recent floods.
4. But cooling the sea means a larger difference between the temperatures of sea and land, more monsoon effect, with stronger winds bringing more water to land.
5. Wind is caused by differences in pressure which are a result of local temperature gradients. Wind causes turbulence in the marine boundary layer. The relative humidity is very high in the stagnant layer of air at distances of millimeters above the sea surface but falls to around 65% a few meters above. Turbulence has a much greater influence on evaporation rate than water temperature except at temperatures above about 27.5°C.

6. Wind makes waves steeper. Spilling breakers mix the thin top layer of water that has been chilled by evaporation and warm it with heat from below. Plunging breakers drive bubbles below the surface and throw spray above it to transfer more water vapor in the air. Evaporation rises steeply if wind speeds exceed 13 m/s.
7. If cloud albedo control at sea reduces temperatures on land, it will also reduce the evaporation rate so more of the precipitation will be retained as ground water.
8. Finally, we know that the regions with the most severe drought problems are dry because air which has been dried by being high in the atmosphere is subsiding and moving out to sea. This means what we do at sea cannot immediately affect the very driest land nearby.

A computer experiment by Jones, Hayward and Boucher [18] illustrates the possibilities. Their world climate model was set to allow the concentration of condensation nuclei to be increased in three small regions together representing 3.3% of the world area. Spray off California nearly doubled rainfall in the drought-stricken region of central Australia and caused a smaller increase in the Southern part of the Amazon basin. Spray off Peru increased rain in Brazil but reduced it in Argentina. But spray off Namibia produced a reduction of about 320 mm a year over most of the Amazon basin which could be partly offset by spray from the Pacific sites.

The spray was applied through the year rather than in any way related to the monsoon phase or wind direction and was confined to the small areas, which would probably not be realistic. But the important result of this work is that we can get changes in precipitation in either direction at distances far from the spray source. It would be no great surprise if the effects were strongly affected by the spray season relative to the monsoon cycle but this has not yet been established.

Other workers [10] have used treatment over much wider areas and have not observed similar effects. In particular, Bala [19] has found the net effect for the Indian subcontinent of widespread long-term cloud albedo control was slightly lower precipitation but a slightly greater run off from the Indian subcontinent because of slower evaporation of ground water in lower temperatures. Another result from [10] is that cloud albedo control carried out well away from the Arctic can reduce the rate of Arctic ice loss and so might prevent an irreversible tipping point. However, the better the climate modeler, the more caveats will accompany the predictions. Present models are not good at predicting precipitation.

It may be possible to use climate models to predict an everywhere-to-everywhere transfer function using pseudo random sequences of increases and reductions of the concentration numbers of condensation nuclei with different sequences in many regions. The sequences would be correlated with the computer model results of all the many meteorological parameters of interest. If small random changes are made to a signal consisting of 20 years of daily observations of temperature records, the scatter of the correlation is about 1% of the standard deviation of the variation of the original record, so very small signals can be detected. More importantly, the processes do not interfere with each other, so all can be done at the same time. By doing the correlations with variable time delays, we can gain information about the frequency response of the climate system.

Engineers know that an accurate control system needs a good way to measure the deviation from the desired state. The resulting error signal should then drive some form of correcting mechanism. Small errors need a high loop gain but require a high-frequency response with a low phase-shift if oscillations are to be avoided. Satellites, perhaps with their temperature-sensing analysis algorithms improved by true data collected from spray vessels, will be able to give fast and accurate measurements of worldwide sea temperatures. Particular attractions of cloud albedo control are that the introduction can be progressive and that the response is both fast and, initially at least, locally precise.

If we understand the flow patterns of winds and currents, we can aim immediate effects at specific regions to preserve Arctic sea ice and coral reefs or protect flood sensitive areas. Even if our understanding is not perfect, we can still apply neural network technology to guide deployment. Few people can explain how to ride a bicycle but many people can ride one. Some atmospheric physicists have said, perhaps not entirely seriously, that while saving humanity's existence on the planet might be regarded as a useful spin-off, the real value of cloud albedo control would be the gain in understanding of atmospheric aerosols.

It is reasonable to suppose that attempts to keep sea surface temperature at previous levels would be less damaging than unbridled increases which seem to be occurring now. Cloud albedo control is like adding steering, brakes and a reverse gear to an unfamiliar vehicle in which the foot of a criminal lunatic is pressing hard on the accelerator. One's first training in the use of a new machine tool should be finding the emergency stop button. It is comforting to know that if we command all vessels to stop spraying, the status quo ante will be restored in a few days. This offsets the frequent criticism of geoengineering by people hostile to it – that once you start using it you have to keep on doing it. This of course could also be an objection raised against electricity generation, sewage treatment, computing, money, and kidney dialysis machines. If you cut the strings of a parachute, you will quickly revert to your previous, rather high terminal velocity. It is a good idea not to cut the strings until you are on the ground being dragged along by a cross wind. In fact, we only have to keep using cloud albedo control until low carbon sources have been developed and excess atmospheric carbon dioxide or methane have sequestrated down to acceptable levels. The fact that the models say that the system quickly reverts gives comfort that we have done nothing irreversible.

Future Directions

Most people working in geoengineering do so with reluctance in the fear that action may be needed before low-carbon energy technologies can be deployed or atmospheric carbon dioxide removed on a sufficient scale. Even if politically impossible,

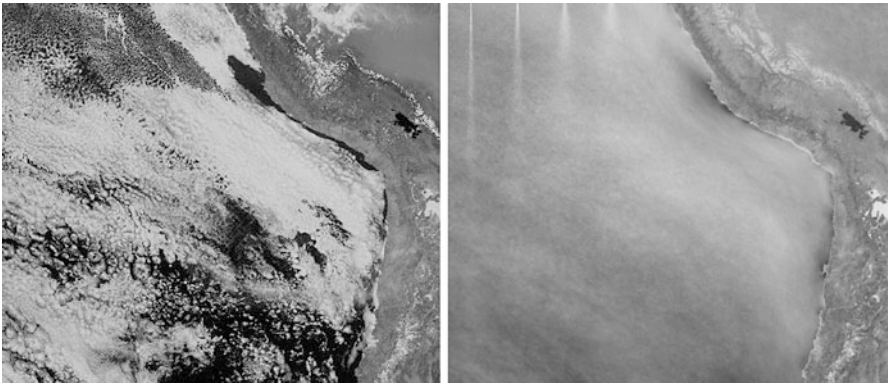


Fig. 4.6 Contrast enhancement by image superposition to demonstrate the effectiveness of small spray sources (Courtesy of NASA and Robert Wood and the VOCALS experiment)

reductions in emissions could take place today; our past emissions have still to take full effect. It is also possible that carbon dioxide may not be the only source of climate change with methane from permafrost increasing fast and possibly able to take the dominant role.

However, humanity is very ingenious at inventing reasons not to take unpleasant actions and ignoring reality. There are many organizations which have to protect their investments and which have perfected the skills to lobby politicians, some of whom were the same people who played a leading part in delaying the development of renewable energy. Despite achieving no reduction in atmospheric carbon dioxide, as shown by the continued acceleration of the Keeling curve, the carbon trading market has been very profitable, and people taking the profits have no wish to have competition from a parallel thermal trading market.

The near future will see activity on computer modeling of all the possible geoengineering techniques with about half the funding aimed at discrediting the ideas. Politicians try to follow public opinion rather than lead it, and until there has been much more suffering from climate extremes, there will be no political advantage in supporting the development of practical working hardware.

However, if spray hardware can be built, there are problems about producing experimental proof of successful operation because the change of cloud brightness needed is well below the contrast detection threshold of the human eye. A possible solution involves taking a large number of satellite photographs of cloud fields, shifting them to align the spray source, rotating them to align the local wind directions, and taking a pixel by pixel average of the full set. The cloud variations will be suppressed to converge towards a middle grey while the wakes downwind of the spray source emerge from the mist. Figure 4.6 (left) shows a single cloud image with four, mathematically generated 30-kg/s spray sources at four dispersion angles and contrast set by the Twomey equation. It would take a great deal of confidence to use the very faint vertical streak at the top left as proof. Figure 4.6 (right) shows the same wake patterns in 100 real superimposed images supplied from the VOCALS experiment off South America. The experiment should be done in a long cruise covering many sea areas and several months because the places where the system does not work will be just as interesting as those where it does.

Conclusions

By 2010, four independent climate models confirmed that the Twomey effect could be exploited to control world temperatures by the deliberate increase of cloud condensation nuclei produced from submicron sea spray.

One model shows that spray from small regions can affect precipitation in either direction at other, very distant regions, but this is not confirmed by studies of wider treatment areas.

With proper understanding of the results of pseudorandom increase and decrease of the concentration of cloud condensation nuclei in climate models, we may learn how to control fleets of spray vessels to obtain beneficial reduction of droughts, the prevention of floods, and increases in ice cover by the correct choices of place and season for spray operations.

The chief engineering problem for the cloud albedo system is the development of mobile, energy efficient spray sources of submicron monodisperse spray which can operate for long periods in any chosen set of mid-oceanic sites.

The effectiveness of cloud albedo control can be demonstrated with a small spray source by superimposing and aligning around 100 satellite images.

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Chapter 5
Ocean Fertilization for Sequestration of Carbon Dioxide from the Atmosphere

Philip W. Boyd

Glossary

Carbon sequestration	Is the removal of carbon by physical, chemical, and/or biological processes, and its long-term storage (i.e., decades to millennia) such that the carbon cannot return to the atmosphere as carbon dioxide.
Ocean fertilization	Is the purposeful modification of the chemical characteristics of the surface ocean, by the addition of plant macronutrients including phosphate and/or iron, or the deployment of equipment such as ocean pipes to enhance the supply of nutrient rich deep water to surface waters. This fertilization has the potential to enhance upper ocean productivity, some of which may eventually settle into the ocean's interior, thereby increasing carbon sequestration.
Global warming potential (GWP)	Provides a relative index between greenhouse gases, such as carbon dioxide or nitrous oxide, using their specific radiative properties to estimate the effect of anthropogenic emissions of each gas, over a specified time period relating to their atmospheric lifetime, on global climate.
Surface mixed layer	Refers to the less dense layer of seawater in the upper ocean (10–200 m thick) that overlies more dense (i.e., colder and/or saltier) waters. This mixed layer is persistently stirred by upper ocean processes such as turbulence and wind mixing.

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Free-drifting sediment traps	Are devices deployed, usually in the upper 1 km of the ocean, designed to intercept settling particles that are the conduit of carbon sinking into the deep ocean as part of the biological pump.

Definition of the Subject

The ocean is a major sink for both preindustrial and anthropogenic carbon dioxide. Both physically and biogeochemically driven pumps, termed the solubility and biological pump, respectively (Fig. 5.1) are responsible for the majority of carbon sequestration in the ocean's interior [1]. The solubility pump relies on ocean circulation – specifically the impact of cooling of the upper ocean at high latitudes both enhances the solubility of carbon dioxide and the density of the waters which sink to great depth (the so-called deepwater formation) and thereby sequester carbon in the form of dissolved inorganic carbon (Fig. 5.1). The biological pump is driven by the availability of preformed plant macronutrients such as nitrate or phosphate which are taken up by phytoplankton during photosynthetic carbon fixation. A small but significant proportion of this fixed carbon sinks into the ocean's interior in the form of settling particles, and in order to maintain equilibrium carbon dioxide from the atmosphere is transferred across the air–sea interface into the ocean (the so-called carbon drawdown) thereby decreasing atmospheric carbon dioxide (Fig. 5.1).

The strength of the biological pump therefore relies upon the global ocean inventory of plant macronutrients, and its impact on atmospheric carbon dioxide concentrations can only be altered under three scenarios [2]. These scenarios involve either a more efficient carbon fixation for each unit/mole of plant nutrient that is taken up, or additional carbon fixation fueled by either uptake of macronutrients in the open ocean that are currently not utilized, or by another source of macronutrients (such as from nitrogen fixation) that add to the preformed nutrient inventory of the upper ocean [2].

In this entry, the focus is only on the topic of iron fertilization and carbon sequestration as it has received considerable attention over 3 decades, whereas other forms of ocean fertilization including purposeful upwelling [3–5] and stimulation of nitrogen fixation [6–8] have received considerably less scrutiny, and only in the last few years. Iron plays a role in driving the uptake of ocean nutrients that are currently not utilized. In around one-fourth of the world ocean, residual nutrients in the surface ocean are present at significant concentrations (Fig. 5.2). These so-called HNLC (High Nitrate Low Chlorophyll) waters paradoxically have low stocks of microscopic plants called phytoplankton. Such cells are anemic in these low iron HNLC waters and cannot utilize fully the surface inventories of plant

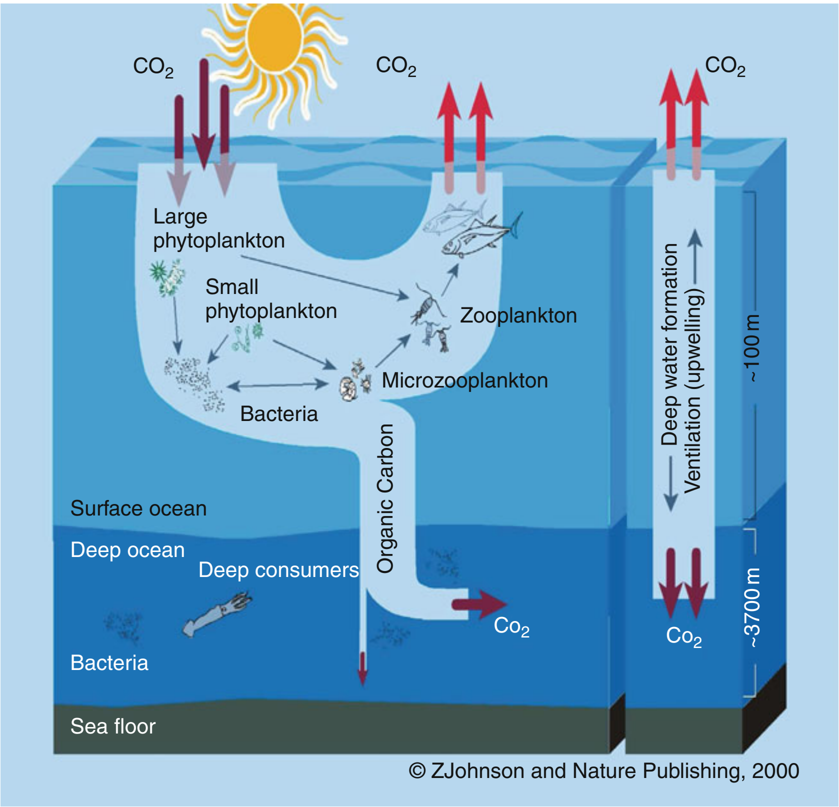

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Fig. 5.1 Cartoons of the biological (*left panel*) and solubility (*right panel*) pumps that drive carbon sequestration in the ocean (from Chisholm [65]). From [65]: "Together with the 'solubility pump', which is driven by chemical and physical processes, it maintains a sharp gradient of CO_2 between the atmosphere and the deep oceans where 38×10^{18} g of carbon is stored. Using sunlight for energy and dissolved inorganic macronutrients, phytoplankton convert CO_2 to organic carbon, which forms the base of the marine food web. As the carbon passes through consumers in surface waters, most of it is converted back to CO_2 and released into the atmosphere. But some finds its way to the deep ocean where it is remineralized back to CO_2 by bacteria. The net result is transport of CO_2 from the atmosphere to the deep ocean, where it stays, on average, for roughly 1,000 years. The food web's structure and the relative abundance of species influence how much CO_2 will be pumped to the deep ocean. This structure is dictated largely by the availability of inorganic macronutrients such as nitrogen, phosphorus, silicon and iron. Iron is the main limiting nutrient in the Southern Ocean, which is why the SOIREE experiment [37, 38] was conducted there (Figure modified from a graphic by Z. Johnson.)"

macronutrients such as nitrate. In summary, iron fertilization can potentially enhance the strength of the biological pump by driving down the inventory of plant macronutrients that are currently not taken up in surface waters, in up to a quarter of the World Ocean. By doing so, they can potentially enhance carbon sequestration and hence reduce atmospheric carbon dioxide concentrations.

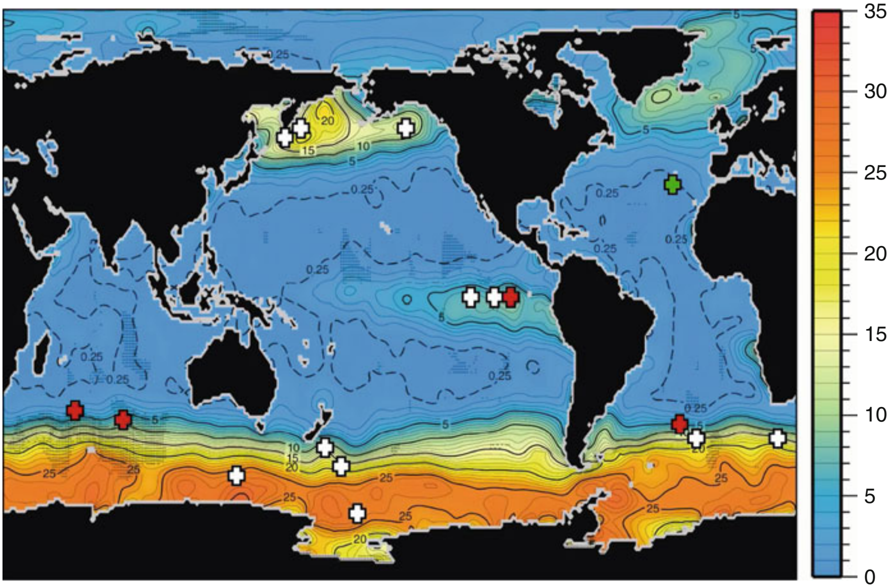


Fig. 5.2 Annual surface mixed-layer nitrate concentrations in units of μmol liter^{-1}, with approximate site locations of FeAXs (*white crosses*), FeNXs (*red crosses*), and a joint Fe and P enrichment study of the subtropical LNLC Atlantic Ocean (FeeP; *green cross*). FeAXs shown are SEEDS I and II (northwest Pacific; same site but symbols are offset), SERIES (northeast Pacific), IronEX I and II (equatorial Pacific; IronEX II is to the left), EisenEx and EIFEX (Atlantic polar waters; EIFEX is directly south of Africa), SOIREE (polar waters south of Australia), SOFEX-S (polar waters south of New Zealand), SOFEX-N (subpolar waters south of New Zealand), and SAGE (subpolar waters nearest to New Zealand). FeNX sites shown are the Galapagos Plume (equatorial Pacific), Antarctic Polar Front (polar Atlantic waters), and the Crozet and Kerguelen plateaus (Indian sector of Southern Ocean; Crozet is to the left of Kerguelen). (Reproduced from Boyd et al. [19].)

Introduction

The assembly of the history of atmospheric carbon dioxide concentrations in the geological past from core records such as Vostok [9] reveals that carbon dioxide is not constant over time but ranges between ca. 280 and 180 ppmv. The timing of these changes is clearly associated with the glacial–interglacial transitions (Fig. 5.3). This record of paleoclimate has led to much discussion regarding the underlying mechanism(s) behind such changes in atmospheric carbon dioxide. Candidate mechanisms for observed alterations in paleoclimate which were proposed included a shift in the latitude of winds in the Southern Ocean [10] (i.e., less communication between deep carbon dioxide–rich waters and the atmosphere) greater sea-ice extent [11] (i.e., reduced air–sea exchange due to capping of more of the polar ocean by sea ice) and iron supply to the ocean [12].

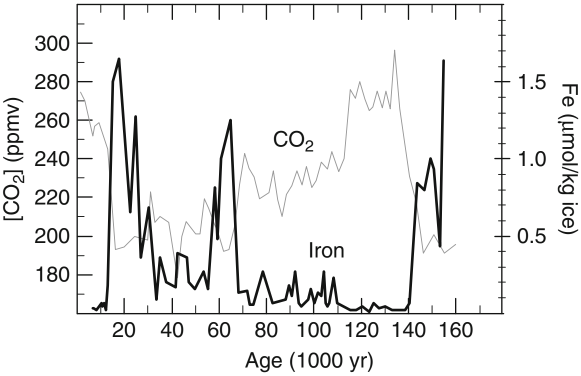


Fig. 5.3 Fe concentration versus mean CO$_2$ concentrations in air trapped in ice from the Antarctic Vostok ice core over the last 160 Ma (Reproduced from Martin [12].)

In 1990, John Martin [12] published the iron hypothesis which conflated trends in ice core records from Vostok (East Antarctica) with contemporary biological oceanographic studies. The former revealed a striking anti-correlation between atmospheric carbon dioxide and aerosol iron supply (Fig. 5.3), which Martin interpreted using evidence of phytoplankton in HNLC regions being stimulated rapidly by iron enrichment, leading to significant drawdown of the previously unutilized plant nutrients. Based on these combined observations, Martin hypothesized that increased dust supply during the windier and more arid glacial periods were responsible for a natural fertilization of the HNLC waters, in particular the Southern Ocean which is the greatest repository for unused upper ocean plant macronutrients in the World Ocean (Fig. 5.2). The consequent iron-mediated uptake of plant macronutrients enhanced the strength of the biological pump resulting in a gradual drawdown, over millennia, in atmospheric carbon dioxide of up to 80 ppmv (Fig. 5.3).

This iron hypothesis stimulated sustained scientific interest in the nature and magnitude of this potential causal mechanism in altering carbon dioxide in the geological past and hence as a drive of paleoclimate. However, Martin's hypothesis also aroused media interest in the application of ocean iron fertilization (OIF) as a "potential climate fix" for rising carbon dioxide concentrations in the Anthropocene [13].

Not long after the publication by Martin and his coworkers of their ship-based iron enrichment studies [14], and of his iron hypothesis [12], the attendant publicity led to ocean iron fertilization (OIF) making front-page headlines in the Washington Post [13]. This media attention raised awareness of OIF sufficiently to result in considerable interest in this approach as a climate fix by a range of commercial parties. These commercial groups adopted a range of approaches that are summarized elegantly as a timeline by Strong et al. [13]. They ranged from application for patents on OIF, to exploring the potential leasing of large tracts of the open ocean to conduct OIF trials. By the mid-1990s – just a few years after the first mesoscale (i.e., at least 10 km length scale) scientific iron enrichment experiments were conducted (Fig. 5.2; see next section) – the first trials of

OIF by commercial operators had taken place in the Gulf of Mexico [15] and another followed off the Hawaiian island chain [16]. A major distinction between these trials and the scientific studies into the biogeochemical impacts of OIF is that the latter were not reported in detail in the peer-reviewed literature [15], and in some cases they consisted only of a series of photographs [16].

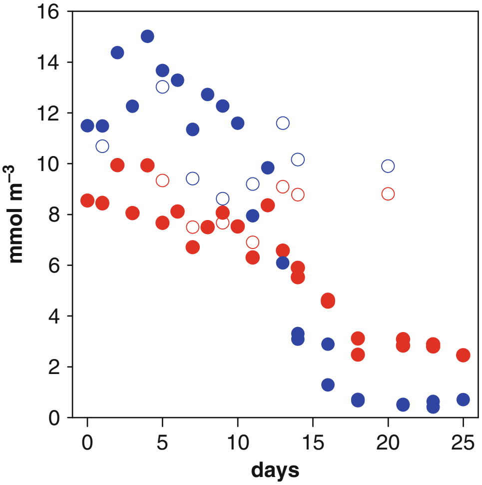
Since these preliminary OIF trials, the advocacy for OIF as a carbon sequestration tool has mainly been driven by selective use of the datasets from the scientific OIF experiments that were readily accessible as peer-reviewed literature in the public domain (see next section), with no further trials being conducted by commercial parties [13]. This potential carbon sequestration strategy is unique among other proposed geoengineering schemes in that there is a suite of scientific research studies into OIF, to examine both its paleoclimatic and contemporary ecological and biogeochemical influences, that commercial parties have been able to tap into and use for advocacy for this biogeoengineering scheme [17].

Over the period from 1993 to 2009 there have been 13 mesoscale iron enrichment studies [18, 19] and three large-scale investigations of regions of the oceans in which high iron supply occurs naturally [20–22]. Such studies were necessary to both overcome the artifacts/limitations of small volume shipboard OIF incubations (1–25 L) [14] and to provide a more realistic and holistic approach to study the ramifications of OIF on oceanic biogeochemistry and ecology [13]. These mesoscale studies were conducted at sites which straddled HNLC waters from polar to tropical (Fig. 5.2), and hence provide a comprehensive assessment of the range of ecological and biogeochemical responses of OIF that inform questions such as: Could sustained natural OIF over millennia be responsible for the observed 80–100 ppmv decreases in atmospheric carbon dioxide during the glacial–interglacial transitions [3, 19]?; How might changes in iron supply in the coming decades, mediated by a changing climate, alter ocean productivity and ecosystem dynamics [19]?

Several syntheses [18, 19] of the findings of these open ocean OIF experiments clearly reveal that iron supply results in increased phytoplankton stocks and growth rates, rates of net primary production, and concomitant uptake of more plant macronutrients such as nitrate (Fig. 5.4). These trends were also noted by investigators at all three sites of natural iron enrichment [20–22], and together confirmed the first tenet of Martin's iron hypothesis [12] – that iron supply to HNLC regions such as the Southern Ocean would increase phytoplankton growth rates, productivity, and nutrient uptake.

Another striking outcome of these scientific research studies was the pronounced drawdown in upper ocean carbon dioxide (conventionally expressed as the partial pressure of carbon dioxide – pCO_2) by up to 70 μmol during the iron-mediated phytoplankton bloom [18, 19]. The OIF experiments also revealed changes in the concentration of other climate-reactive gases during the resulting phytoplankton blooms, which could potentially either offset or enhance the effect of the drawdown of carbon dioxide. For example, increases in DMSP (dimethyl sulfoniopropionate), and subsequently in DMS (dimethyl sulfide) – a precursor of the sulfate aerosol

Fig. 5.4 Time series of observed decreases in the mixed-layer concentrations of silicate (*blue solid symbols*) and nitrate (*red solid symbols*) within the iron-labeled SERIES patch in the NE subarctic Pacific. Day 0 is when the iron was first added, and the open symbols denote the mixed-layer nutrient concentrations in the surrounding HNLC waters (Reproduced from Boyd et al. [43].)



hypothesized to enhance in increase cloud albedo by increasing cloud condensation nuclei [23] – were observed in experiments such as SOIREE in the Southern Ocean [24] but not in others such as SERIES in the NE Pacific [25] (see Fig. 5.2). Such an iron-mediated enhancement of cloud reflectance (i.e., albedo) could alter radiative forcing and along with concurrent carbon dioxide drawdown might enhance the ability of OIF to mitigate climate warming by both sequestering carbon and reducing incoming solar radiation (but only in the Southern Ocean where cloud condensation nuclei are undersatuarated [26]). Subsequent modeling studies have pointed to the complex interplay between surface mixed-layer dynamics, meteorological forcing, photochemistry, and microbial activity in determining whether DMS increases or remains unaltered following OIF [27]. The findings of this model [27] suggest that a wider extrapolation of the results of OIF at any particular site or region may be difficult.

Other climate-reactive gases that have been investigated and tracked during OIF scientific experiments include two gases with global warming potential (GWP, see Glossary) far in excess of carbon dioxide. Both N_2O and CH_4 production have been investigated in a small subset of scientific OIF studies which are summarized by Law [28]. As was the case for DMS, in some experiments there was a slight increase near the base of the surface mixed layer, thought to be associated with particle remineralization [29], whereas in other OIF studies there was no or a negligible change in the concentrations of these dissolved gases [19, 28, 30]. The SOFEX-South study (see Fig. 5.2) provided perhaps the best opportunity to study the impact of OIF on a wide range of gases, but despite this improved sampling coverage there were few significant changes in most of the biogenic gases sampled [26]. Given their high GWP, a relatively small increase in the concentration and subsequent efflux of either gas associated with the evolution of an iron-stimulated bloom, could readily offset the CO_2 mitigation effects that might result from OIF [28, 30].

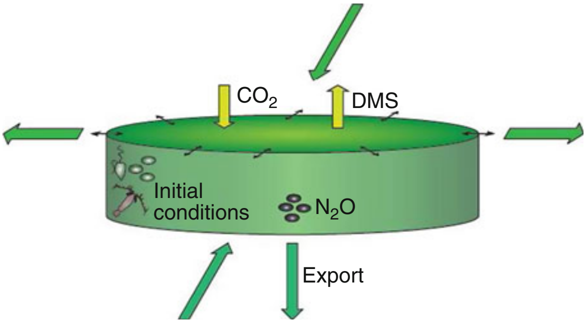


Fig. 5.5 A schematic of the interactions between the iron-enriched patch of upper ocean and the surrounding HNLC waters redrawn after Boyd and Law [31]. Solid *black arrows* denote diffusion of waters both to and from those at the edge of the patch. The solid *green arrows* denote the impact of strain – due to horizontal currents – which stretch the patch laterally. The effects of the physics can increase the areal extent of the initial patch from $< 100\ \text{km}^2$ to $> 1000\ \text{km}^2$ in several weeks [43]. CO_2, DMS, and export-labeled arrows represent the drawdown of atmospheric carbon dioxide into the iron-enriched waters; the potential efflux of elevated upper ocean concentrations of DMS; and the enhanced export of biogenic particles at the termination of the bloom (see Fig. 6 for more details on the timing), respectively. In some ocean iron fertilizations (OIFs) [34], the initial conditions – the biological composition of the iron-enriched waters (i.e., phytoplankton and zooplankton community structure) played a pivotal role in determining the bloom signature. As the bloom evolved in most OIFs there was shift from small phytoplankton to diatoms and in some studies the particles sinking to the base of the mixed layer later in the bloom were linked with increased nitrous oxide concentrations [28]

Thus, these largely unanticipated changes in other climate-reactive gases following OIF, and their potential impact on radiative forcing, must also be factored into any estimation of the net outcome of OIF on climate mitigation.

This suite of scientific mesoscale OIF experiments also revealed a range of other unanticipated side effects, that ranged from physical (dilution of the iron-enriched patch of water [31] (Fig. 5.5); enhanced upper ocean heating due to more light absorption by the phytoplankton bloom [32]), biological (production of a neurotoxin – domoic acid – by some of the blooms of pennate diatoms [33]), to ecological (importance of the initial conditions with respect to the zooplanktonic grazers to the outcome of the bloom [13, 34] (Fig. 5.5).

Two of the most striking conclusions to come out of the syntheses of the scientific OIF experiments [18, 19] were the all-pervasive influence of iron supply of ocean ecology and biogeochemistry, and that even mesoscale experiments of an initial areal extent of $100\ \text{km}^2$ were still prone to artifacts [31]. Together these outcomes were indicative of the complex interplay of physical, chemical, optical, biological, and ecological factors in determining the overall biogeochemical signature of OIF experiments. Such a signature has important ramifications for both carbon sequestration, and how additional iron-mediated processes, such as the production of climate-reactive gases, other than carbon dioxide, might either amplify or counteract the impact of such carbon sequestration on climate.

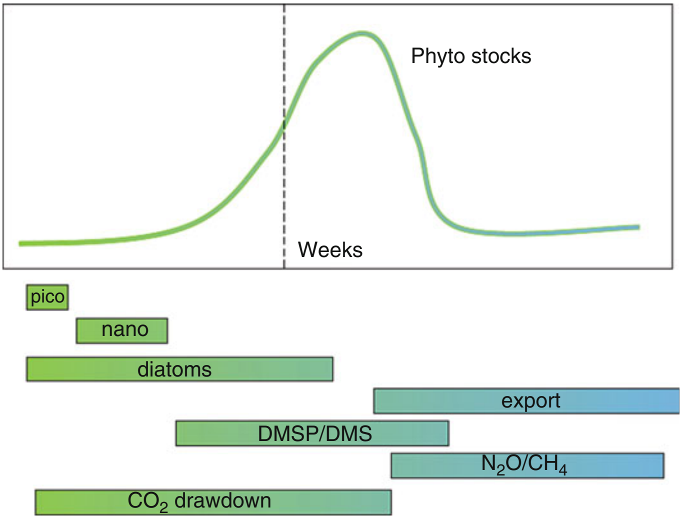


Fig. 5.6 A generic timeline of biogeochemical events overlaid on the pattern of bloom development, termination, and decline (based on the SERIES OIF [43]). The vertical dashed line denotes the bloom phase attained when most studies terminated, as the vessel had to depart the iron-enriched patch. Hence, only a few studies [41–43] sampled the biogeochemically important termination and decline phases of the iron-mediated blooms. Pico and Nano denote pico- (<2 μm) and nano- (2–20 μm) phytoplankton. The former dominate under low iron HNLC conditions, and are stimulated by iron supply, but are rapidly grazed down to low levels by microzooplankton. The nanophytoplankton also have a transient increase following iron supply, but are also grazed down, in some cases resulting in increased DMSP (dimethyl sulfoniopropionate) followed by elevated DMS (dimethyl sulfide) concentrations

The supply of iron resulted not only in an increase in the stocks of phytoplankton, but in a change of the dominant phytoplankton groups. In HNLC waters, the resident small phytoplankton cells were rapidly replaced by other larger-sized phytoplankton (Figs. 5.5, 5.6). Some of these larger cells – the prymnesiophytes – produce a precursor of the sulfate aerosol that is hypothesized to play a key role in the CLAW hypothesis [23]. CLAW proposes a link between the production of these phytoplankton compounds and enhanced cloudiness which in turn reduces incoming solar radiation – a potential natural form of SRM (Solar Radiation Management [26,35]). However, the prymnesiophytes may only dominate the iron-stimulated bloom for a few days, after which they are rapidly removed by grazing [36]. This grazing in turn helps transform the DMSP into DMS, the ultimate precursor of the sulfate aerosol. By this route, other climate-reactive gases are produced.

During the rise and fall of the stocks of prymnesiophytes, another group of larger phytoplankton – the diatoms – are increasing from very low initial stocks, and soon will come to dominate the phytoplankton bloom (Figs. 5.5 and 5.6), carbon fixation, and the drawdown of carbon dioxide – in some studies by as much as 80 μatm. Hence, different phytoplankton groups, with differing biogeochemical signatures, dominate these iron-stimulated blooms at different times (Fig. 5.6). Interpretation of these signatures is further confounded by three factors, the oceanographic conditions – the mixed-layer depth [18] (see Glossary) – which characterize the site for the experiment (Fig. 5.3), the seed stock of phytoplankton and zooplankton [34] (Fig. 5.5), and the role of upper ocean physics in exchanging the surrounding HNLC waters with those of the iron-fertilized patch [31] (Fig. 5.5).

Site-specific oceanographic conditions can influence the timing and the magnitude of the biological trends and the resultant biogeochemical signatures – outlined in Fig. 5.6 – which vary considerably between experiments. For example, the initial biological conditions – in particular the seed stock of phytoplankton and zooplankton – also appear to be key drivers of the outcome of scientific OIF experiments [19]. Although our interpretation of the role of seed stocks is restricted to just two of the thirteen OIF studies – SEEDS I and II in the NW Pacific [34] (Fig. 5.2), this comparison reveals marked differences in the characteristics of each iron-mediated bloom. Both SEEDS experiments took place at the same site, and season but in different years. SEEDS I was a diatom-dominated bloom that resulted in high phytoplankton stocks, whereas SEEDS II was characterized by a mixed community of many different phytoplankton groups and much lower phytoplankton stocks, relative to that recorded during the development of the SEEDS I bloom. Careful analysis of the bloom datasets revealed that unlike SEEDS I, in SEEDS II there were much higher initial stocks of zooplankton which probably kept the bloom in check and which provides the most compelling explanation for the different bloom signature [34]. This example, again points to the difficulty in generalizing on either the ecological or biogeochemical outcome of these iron-stimulated blooms and also provides insights into the complex array of factors contributing to the resultant bloom signature.

One of the most perplexing outcomes from this suite of scientific OIF studies was that the progression from shipboard experiments in 25 L incubation vessels to enriching 100 km^2 of HNLC ocean with iron did not overcome the issue of experimental artifacts [31]. It was evident from a comparison of the initial area of HNLC waters that were enriched with the areal extent at the end of the experiment that there was up to a tenfold increase in the bloom dimensions. Also, satellite imagery of the blooms – using ocean color (a proxy for phytoplankton stocks) revealed that the blooms had become elongated in one direction/plane and were often ellipsoidal [37, 38]. The analysis of Abraham et al. [38] revealed that the labeled waters interacted with the surrounding HNLC ocean via the strain and rotation of the patch and the dilution of the iron-enriched high chlorophyll patch with the surrounding low chlorophyll low iron waters (Fig. 5.5). Such dilution was reported to act like a chemostat [37] – an approach used in lab cultures of phytoplankton in which the culture is maintained under steady-state conditions by matching the phytoplankton growth rate with an equal rate of dilution with nutrient-enriched seawater. The so-called chemostat effect explains why some of these iron-stimulated blooms had a longevity not observed for natural blooms from the same ocean basins, for example, the Southern Ocean [39]. Perhaps the most significant effect of dilution of the OIF study by the surrounding HNLC waters was the delay of the bloom termination [40] (Fig. 5.6), and hence few studies were able to explore one of the key issues – how much of the iron-mediated increase in phytoplankton carbon sank out of the upper ocean and was sequestered within the ocean's interior.

This issue relates to the second tenet of the iron hypothesis of John Martin [12], and is central to assessing the contribution of the candidate mechanism of increased iron supply during the glacial–interglacial periods for the observed 80–100 ppmv decrease in atmospheric carbon dioxide over millennia. Despite its importance, few of the scientific OIF studies were able to measure whether any additional export or sequestration, over and above that occurring under ambient HNLC conditions, of carbon resulting from purposefully iron-stimulated blooms. Several reasons were responsible for this dearth of C export or sequestration measurements. First, most studies were of too short a duration to capture the transition from bloom development to termination and decline (Fig. 5.6) – as it is during this phase that the majority of export from the bloom will take place [19]. Second, because of the relatively slow phytoplankton growth rates in the cold waters of the Southern Ocean, the physical dilution of the iron-stimulated phytoplankton bloom was at a comparable rate in these waters [40]. This prolonged the development of the bloom and thus in most of these studies the bloom decline phase was never observed, for example, SOIREE, EisenEX, and SOFEX-South (see Fig. 5.2).

There are only two published scientific OIF experiments in which the duration of the experiment was sufficient to encompass the bloom termination and decline phase. In IronEX II, in tropical HNLC waters of the equatorial Pacific [41] (Fig. 5.2) the bloom evolution, termination, and decline took <20 days, and export was measured [42]. However, due to lack of space on the vessel, these export flux measurements were done opportunistically and hence are too sparse to construct a budget to resolve how much of the enhanced phytoplankton stocks from the bloom were exported to depth [42]. In the SERIES study in the NE subarctic Pacific (Fig. 5.2) water, temperatures were intermediate between tropical and polar HNLC waters and hence the length of the evolution, termination, and decline phases was within the duration of SERIES and enabled this study to conduct the most detailed assessment of the bloom decline to date [43].

The decline of the SERIES bloom occurred relatively rapidly – within 10 days of the bloom peak [43, 44] – and was evident both from several remotely sensed images of the bloom and from detailed vertical profiles from ships within the bloom. Together, these measurements provided an accurate inventory of the enhanced particulate organic carbon (POC) driven by the iron-stimulated diatom bloom. This inventory could then be compared with the export of POC that was intercepted by free-drifting sediment traps (see Glossary) directly below the surface mixed layer and deeper in the water column (i.e., below the depth of the permanent pycnocline below which the carbon will be sequestered on timescales of decades to centuries). Although, shallow sediment traps are widely viewed as imperfect tools to reliably estimate export flux [45], these traps had been carefully calibrated (using thorium), had fluorometers located on the upper part of the array (to ensure the traps always underlay the waters of the bloom), and such traps are thought to be most reliable at sampling export when settling phytoplankton from a bloom dominate this flux [46]. Again, such refinements to this sediment trap sampling approach illustrate the difficulties in making reliable estimates of export flux from such blooms.

During the SERIES OIF study, the comparison of the proportion of phytoplankton carbon fixed in the iron-mediated bloom that exited the upper ocean was around 18%, and around 6% of the phytoplankton carbon settled deeper than the depth of the permanent pycnocline – i.e., was sequestered on long timescales into the ocean's interior [43]. A coupled budget of C and Si revealed that much of the settling phytoplankton carbon was consumed by both zooplankton and in particular by heterotrophic bacteria. These fluxes of iron-mediated POC export and sequestration can be used to provide estimates of C flux per unit iron added – a valuable ratio required by biogeochemical modelers to assess the role of increased iron supply in the geological past [19, 47]. Significantly, the Fe:C molar ratios of 2×10^{-3} and 6×10^{-2} were considerably lower than those used by early models (i.e., 1×10^{-5} summarized by de Baar et al. [47]) – and based upon data from lab culture flasks of iron-replete diatoms – to estimate the role of iron in influencing atmospheric carbon dioxide concentrations in the geological past.

Since the SERIES study, two studies [21, 22] in regions of the Southern Ocean (Fig. 5.2) where natural iron supply each year stimulates phytoplankton blooms have provided additional estimates of carbon export per unit iron. The advantage of estimates from these natural iron supply laboratories is that their estimates of carbon export per unit iron are based on in situ iron supply c.f. a purposeful addition of an iron salt in OIF, as the latter are reported to overestimate the supply of iron as much of it is rapidly removed by transformations to non-accessible forms of iron for the biota [21]. The disadvantage of such natural laboratories is that it is difficult to assess the winter inventory of iron available to drive such natural blooms unless these waters are sampled in winter or early spring at the latest. Thus, it is possible to underestimate the winter iron supply and hence overestimate how much carbon export per unit iron supply occurs in each region.

There are wide disparities between the estimates of carbon export per unit iron supplied between the OIF study SERIES [43] and the CROZEX [22] and KEOPS [21] studies of natural laboratories. Even allowing for corrections to improve the reliability of each of these estimates (e.g., Chever et al. [48]) still results in a more than order of magnitude difference between them. Such a disparity between these estimates has major ramifications for assessing the role of increased iron supply in the geological past, with the estimate from KEOPS [21, 48] pointing to iron playing a major biogeochemical role in shaping paleoclimate, whereas the CROZEX [22] and SERIES [43] estimates, if broadly applicable suggest that increased iron supply played a much lesser role in altering the paleoclimate.

Thus, the present information from both a limited number of OIF's and natural laboratories of high iron supply provides too wide a range of estimates, and each with associated uncertainties, to resolve whether OIF sequestered sufficient carbon, on timescales of millennia, can play a pivotal role in the observed decreases in atmospheric carbon dioxide during the glacial terminations over the last 800,000 years [49].

Evaluation of OIF in Relation to Other Proposed Geoengineering Schemes

There have been only a few commercial trials [15, 16] to explore whether OIF is a potential tool in mitigating increasing emissions of anthropogenic carbon dioxide, and as stated none of these tools have been published in the peer-reviewed literature or is in any way comprehensive relative to the suite of scientific research OIF studies to address the role of increased iron supply in paleoclimate. Private sector interest in OIF for geoengineering has therefore based on scant data from any commercial trials, and thus has relied heavily on, and selectively used, datasets from the scientific OIF research studies. This presents problems in trying to do any initial assessment of how OIF compares with the pros and cons of other equally poorly resolved geoengineering schemes. Nevertheless, some attempts have been made at ranking OIF in relation to other schemes [17, 35, 50, 51]. Such ranking schemes have focused on criteria such as efficiency, cost, safety and side effects, mitigation speed, and emergency stop.

The estimates of the carbon exported per unit iron from scientific studies, to better quantify how much additional carbon was sequestered in the geological past due to enhanced iron supply over millennia, can be adopted cautiously to explore this criterion. The greater than an order of magnitude range in C exported per unit iron supplied will prevent refining the efficiency of OIF as a geoengineering tool but can be used to constrain the efficiency of OIF. Model estimates – summarized by Denman [52] range from 5 to 100 ppmv CO_2 (over decades to centuries), but are also subject to many unknowns, some of which – including the fate of the added iron (is it recycled or does it sink out) which several model outcomes are particularly sensitive to [53, 54]. Thus, it is presently difficult to relate the potential efficiency of OIF to other proposed geoengineering schemes. One issue that has also been overlooked is that even if enhanced iron supply was responsible for the majority of the decrease in atmospheric carbon dioxide, this probably took place over millennia in the Southern Ocean – and hence the rate of C sequestration would be 80 ppmv/5,000 years [55] (see later).

Following the banner headlines on OIF as a potential climate fix [13], it became further attractive to investors, etc., as it was reported to be a cheap climate fix – with estimates by one of the first commercial parties interest in OIF – Michael Markels Jr. publishing an estimate of 2$ US per ton of C sequestered [56]. Careful scrutiny of this estimate reveals it to have been based on erroneous and upper bounds for export efficiency [55, 57]. Again, some of the datasets derived from the OIF/paleoclimate scientific studies can be used to reevaluate the claims of 2 $ US per ton of C sequestered that were put forward by Markels. Boyd (2008) [55] rescaled these estimates by simply replacing the upper bound on C exported per unit iron used by Markel's (he had based his 2 $ per ton C estimate on the C:Fe ratio in a lab flask of iron-replete diatoms) with those measured during OIF experiments such as SERIES. The revised estimates increased to between 30 and 300 $ per ton of carbon sequestered when more realistic estimates from scientific research were applied.

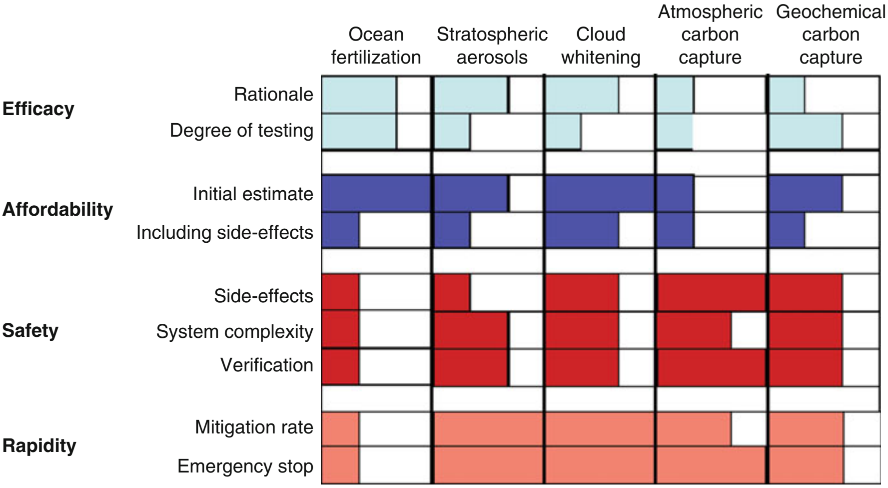


Fig. 5.7 Comparison of aspects of five geoengineering proposals. The schemes span both carbon storage and reductions of solar radiation, and have been prominent in both the popular and scientific media. The figure highlights schematically some facets of the four criteria: efficacy, cost, risk, and time. The assessment gives scores relative to other schemes. For each facet, more color denotes a higher ranking. Efficacy is assessed in the first line according to the provenance of a scheme, with those based on historical precedents rated higher than those derived from theory and/or models. The extent of testing is shown in the second line, with related observations from experiments or pilot studies scoring higher than model simulations, which in turn rank above a proposal with accompanying technical details. The full degree of efficacy is too uncertain at present to depict as a facet in this intercomparison and will need further research. Affordability is categorized as initial cost assessment from the designer of a scheme in the upper line and a more realistic cost assessment including additional costs that come with a scheme's risks in the lower line. Safety provides an assessment of risk, which is related to known side effects, with unknown side effects represented here by system complexity (biogeochemical complexity is larger than geochemical complexity, which is larger than physicochemical complexity) and the verification of both efficacy and side effects. Other important but very uncertain aspects of risk, such as geopolitical and economic changes, require further research. Relevant aspects of time include the rate of climate mitigation in the top line (higher rates are better), and the rapidity with which to halt any unanticipated deleterious effects, based on residence time of the agent of perturbation in the environment (shortest residence scores highest, reproduced from Boyd [17])

Such a range of costs suggests that OIF is not the cheap carbon fix that was originally claimed, indeed the upper bound for OIF C sequestration is comparable to some of the most expensive approaches [35] – "artificial tree" atmospheric carbon dioxide scrubbers – currently being discussed. It should also be noted that the $30–$300 per ton C sequestered estimates for OIF based on the rescaling by Boyd [55] did not include the additional costs associated with verification (Fig. 5.7) and implementation nor the hidden costs of any unknown side effects.

Insights into potential side effects of OIF have again been obtained from scientific research into the relationship between OIF and paleoclimate. These side effects include the concurrent production of other more potent greenhouse gases

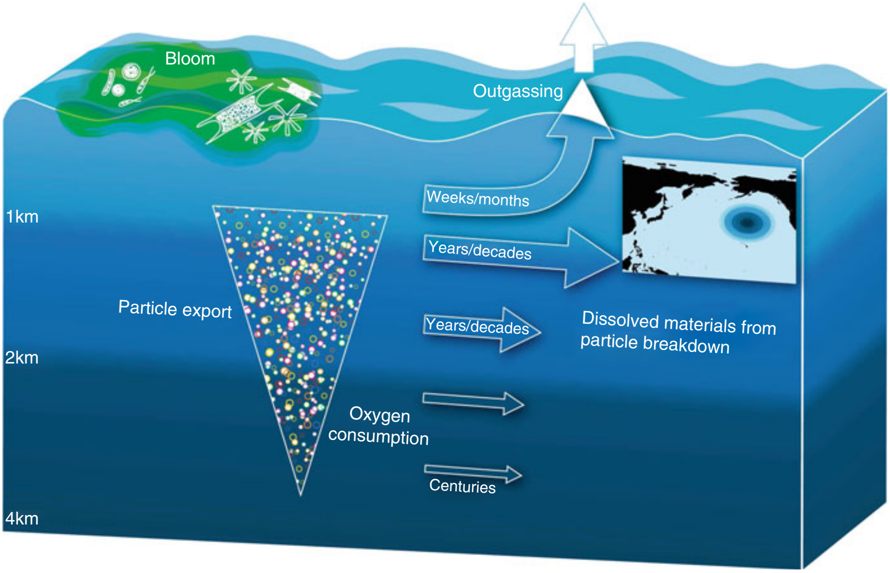


Fig. 5.8 Vertical (i.e., particle settling and remineralization) and horizontal (ocean circulation) transport processes over a range of timescales affect the fate of biologically fixed carbon in the ocean. The inset map of the NE Pacific denotes the likely initial size of commercial OIF as they would have to be large if they were to have any biogeochemical effect whatsoever [58]. The concentric rings represent the rapid spreading of the perturbed ocean waters due to ocean circulation, with major ramifications for detection and attribution issues (see main text)

(Fig. 5.5), and the release of neurotoxins by some pennate diatom species that have been reported to bloom [33], upon supply of iron. There are likely to be other side effects if OIF was to be implemented as a large scale – basin scale – geoengineering scheme, which would have to be the approach if it was to have any significant mitigation effects [58] (Fig. 5.8). Such findings are beyond the length scale of scientific OIF studies (10 km) and are derived from global and basins scale circulation models with embedded biogeochemistry [52–54]. One such side effect would be the potential removal (termed “nutrient robbing” [35]) of much of the northward supply of plant macronutrients from the Southern Ocean to the N hemisphere [53, 54]. The S Ocean is reported to supply around 75% of the macronutrients which drive the productivity of waters – and hence of fisheries – in the N hemisphere [59]. Another potential side effect that has until recently been overlooked is that of OIF resulting in ocean acidification of the ocean’s interior due to the purposeful increase in net carbon storage in the deep ocean [60]. In a recent ranking [17], due to the lack of detailed information, geoengineering schemes which had biological aspects such as OIF were ranked as more complex with more unknowns and hence less safe and with the likelihood of more side effects relative to schemes that were based on geochemical or physical–chemical foundations only.

If geoengineering schemes are to be applied, then they must be rapid and effective as it is thought that the time to mitigate climate change is short [61–63] – of the order of decades. However, based on the sustained OIF of the Southern Ocean over millennia in the geological past – it appears that at best OIF would have a mitigation rate of the order of 80 ppmv/millennia [55], which is several orders of magnitude too slow, relative to one of the stabilization wedges of 1 gT C proposed by Paccala and Soccolow [63] to mitigate rising atmospheric carbon dioxide concentrations on a timescale of decades.

A further important criterion, given the lack of pilot studies of trials of potential geoengineering schemes by the private sector, is what happens if some previously unknown and deleterious side effect(s) becomes apparent shortly after an ocean basin scale pilot study and/or scheme has commenced. How rapidly can such a scheme be shut down – what is the timescale for emergency stop? (Fig. 5.7). Again, due to the lack of information, on key issues such as what is the fate of the added iron – is it rapidly recycled or does it sink out – perhaps the best proxy to differentiate between a wide range of terrestrial, atmospheric, and oceanic reservoirs is the residence time in them [17]. This is relatively short for the atmosphere relative to the ocean and so suggests that oceanic schemes may well have longer emergency stop times – which would put them at a disadvantage when ranking with other schemes.

The detection of increased carbon sequestration and its attribution to purposeful OIF is not trivial [13, 58] (Fig. 5.8). Such detection and attribution issues are also pertinent to side effects such as the inadvertent production of other biogenic gases that are climate reactive [58]. There are also issues with increased anoxia in ocean waters [50] that could confound or mask detection and attribution of any decrease in dissolved oxygen due to OIF [13]. Detection of large-scale changes in ocean biogeochemical signatures such as in the concentrations of biogenic gases must overcome a wide range of hurdles, including the dynamic nature of the ocean, where large volumes of waters are rapidly transported both laterally and vertically [64]. Any such changes due to geoengineering must also be detected over and above other concurrent changes to the oceanic environment, such as by climate variability, and climate change [58]. Clearly, detection and attribution issues will help to frame the development of policy on geoengineering [50].

Conclusions and Future Directions

> *"The histories of the scientific and commercial interests in ocean iron fertilization (OIF) are intimately connected—co-evolving and transforming over time* [13]."

OIF is perhaps unique within the fledgling geoengineering debate, in that 13 relatively large-scale (10 km length scale) scientific research experiments have been conducted into better understanding the role of altering iron supply on paleoclimate, and how iron supply influences ocean ecosystems and in turn ocean biogeochemistry. The results from these experiments also have value for a wide range of disciplines surrounding geoengineering from policy formulation to implementation. For example, the results of these experiments point to a wide range of inextricably linked changes in ocean physics, chemistry, biology, and ecology that have both anticipated (increased productivity) and unforeseen (ocean acidification) consequences. The dynamic nature of the perturbed regions of ocean, and their interactions with surrounding waters, raises issues about implementing larger-scale schemes and in tracking such perturbed waters to both assess whether any additional carbon has been sequestered, and also if any chemical or biological side effects that might offset any additional carbon sequestration have occurred. Taken together, this research forms a body of rigorously conducted and communicated science that is indicative of the need to apply the precautionary principle when considering any future development of policy both into regulating future research into OIF and legislating any pilot bio-geoengineering schemes.

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Chapter 6

Biochar, Tool for Climate Change Mitigation and Soil Management

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Glossary

Biochar	The porous carbonaceous solid produced by thermochemical conversion of organic materials in an oxygen-depleted atmosphere which has physiochemical properties suitable for the safe and long-term storage of carbon in the environment and, potentially, soil improvement.
Black carbon	The continuum of solid combustion products ranging from slightly charred degradable biomass to highly condensed, refractory soot. All components of this continuum are high in carbon content, chemically heterogeneous, and dominated by aromatic structures.
Carbon (dioxide) equivalent	Common measure of global warming potential constructed by converting the emissions of the six greenhouse gases under the Kyoto Protocol of the UNFCCC into the equivalent radiative forcing units of CO_2. CO_2, N_2O, and CH_4 are the relevant Kyoto gases to be considered in evaluating biochar.
Carbon abatement (CA)	The net effect of changes in greenhouse gas fluxes that result from the production and application of biochar. This can include any or all of the following: carbon stored in biochar;

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	CO_2 equivalent emissions released during pyrolysis; offset CO_2 equivalent emissions arising from avoided fossil fuel combustion; offset CO_2 equivalent emissions from reduced chemical inputs to agriculture; change in nitrous oxide and/or methane flux through biochar addition to soils; change in carbon in soil organic matter due to biochar addition; and offset CO_2 equivalent emissions from changed operations in the field. Which of these components is included will be specified in the text.
Carbon credit	Any mechanism for allocating an economic value to a unit of carbon (dioxide) abatement. The most common units are EU Allowances (EUAs) (under the EU ETS), Emission Reduction Units (ERUs) (Joint Implementation, UNFCCC), Certified Emission Reductions (CERs) (Clean Development Mechanism, UNFCCC), and Verified Emission Reductions (VERs) (voluntary carbon market).
Carbon stability factor (CSF)	The proportion of the total carbon in freshly produced biochar which remains fixed as recalcitrant carbon over a defined time period (10, 100 years, etc., as defined). A CSF of 0.75 means that 75% of the carbon in the fresh biochar remains as recalcitrant carbon over the defined time horizon, and that 25% of the carbon has been converted into CO_2.
Charcoal	The solid product of natural fire and traditional biomass conversion under partially pyrolytic conditions without yielding bioenergy coproducts.
Mean residence time (MRT)	Inverse of decay rate, this is the average time for which carbon in new biochar remains present in a recalcitrant form.
Net primary productivity (NPP)	A measure of plant growth and the additional CO_2 fixed and stored into plant biomass over a period of, for example, 1 year; technically, it is calculated as the balance between net photosynthesis and plant (dark) respiration.
Pyrolysis-biochar system (PBS)	A combination of a specified pyrolysis technology, transport, distribution and storage infrastructure and application of biochar.
Recalcitrant carbon	Aromatic carbon which is resistant to chemical or biological oxidation and subsequent conversion to CO_2.
Terra preta	Literally “dark earth,” these are localized soils, intensively studied, whose dark color appears to result from historic and prolonged management with charcoal, probably for the enhancement of agricultural productivity in and around the Amazon Basin.

Definition of the Subject

Biochar is the solid remains of any organic material that has been heated to at least 350°C in a zero-oxygen or oxygen-limited environment, which is intended to be mixed with soils. If the solid remains are not suitable for addition to soils, or will be burned as a fuel or used as an aggregate in construction, it is defined as char not biochar. There is a very wide range of potential biochar feedstocks, e.g., wood waste, timber, agricultural residues and wastes (straws, bagasse, manure, husks, shells, fibers, etc.), leaves, food wastes, paper and sewage sludge, green waste, distiller's grain, and many others. Pyrolysis is usually the technology of choice for producing biochar, though biomass gasification also produces smaller char yields. Syngas and pyrolytic bio-liquids, which have a potential use as energy carriers, are produced alongside biochar.

The strongest evidence for the beneficial effects of char additions to soils arises from the *terra preta* soils of the northern Amazon, where dark, highly fertile soils with very high levels of both stable (char) carbon and organic carbon were established and remain today [1]. Char was also added historically to soils in parts of northern Europe (including Netherlands, NW Germany, and Belgium) [2]. Chars have been, and are currently being used as soil amendments in Japan and West Africa.

The contemporary interest in biochar started in the early part of the twenty-first century and arises from the bringing together of the potential benefits for soils and agriculture with the carbon storage or sequestration opportunity afforded by recalcitrant, stabilized aromatic carbon. Biochar production and deployment has the potential to do one or more of the following:

- Reduce atmospheric greenhouse gas concentrations through CO_2 removal and avoided greenhouse gas emissions (perhaps on a gigaton carbon abatement scale)
- Improve the structure, properties, and "health" of soils
- Increase crop productivity
- Provide energy (e.g., electricity from syngas, heat from syngas, power from liquid fuels)
- Safely dispose of certain waste materials with potentially useful recovered by-products
- Absorb pollutants and contaminants and reduce nitrate leaching to water courses
- Suppress soil emissions of nitrous oxide and methane

Biochar is one of only a few strategies for actually removing CO_2 from the atmosphere (in addition to reducing atmospheric emissions where the use of fossil fuels is substituted for). While this feature may not currently be a top priority, it will become increasingly important in the decades to come, as it is almost inevitable that atmospheric CO_2 concentrations are now set to exceed what is commonly regarded as a safe level (400–450 ppm). Biochar has consequently been termed ageo-engineering option, though it is a debatable point (See Box 6.1).

Introduction

Contemporary biochar research originates from several different sources: (a) research on *terra preta* soils from Amazonia dating back to the middle part of the twentieth century and earlier (e.g., the pioneering work of Sombroek) [3]; (b) research on the effects of charcoal on soils and plants, with initial contributions from the early- to mid-twentieth and more significant efforts in Japan in the 1970s and 1980s; (c) research on the properties and cycling of naturally occurring black carbon and charcoal; and (d) engineering RD&D on pyrolysis and gasification. The idea of the long-term storage of carbon in a stabilized form as found in charcoal (aromatic benzene-ring-type structures) was first proposed by Seifritz in 1993 [4], though his vision was storage in suitable land formations (such as valleys) rather than on agricultural land. This proposal was somewhat ahead of its time, and it was not until the first half-decade of the twenty-first century that the climate-change agenda provided a way of bringing the quite disparate areas of soil science, agronomy, environmental science, and engineering together under the banner of "biochar". Johannes Lehmann and Peter Read were important in making this conceptual linkage. A series of meetings took place in 2006 to 2008 which began to define and consolidate the emergent biochar community of researchers, practitioners, policy makers and entrepreneurs, including the first three meetings of the International Biochar Initiative (2007, Australia; 2008, UK; 2010, Brazil). In 2009, the first dedicated biochar book was published, edited by Lehmann and Joseph [5], and a series of national and regional meetings were held in 2009–2011, including in the USA, UK, Australia, China, Malaysia, and Brazil. Dedicated biochar research centers have now been established in the USA, Germany, New Zealand, and the UK, while existing departments, laboratories, or field stations in the disciplines of soil science, pyrolysis engineering and agronomy are increasingly turning their attention to biochar RD&D. Writing in 2010/2011, biochar has now become a distinct cross-disciplinary field of enquiry, a remarkable achievement given that the word was not even in circulation until the mid-2000s. Several comprehensive reviews of the biochar field were published in 2009 and 2010, and these can be read alongside the current chapter [2, 6, 7].

In this chapter, biochar is reviewed from the perspective of climate change, biomass and bioenergy resources, soils and agronomy. Biochar intersects all these issues and has to be evaluated against the dominant and emerging designs and options for solving problems and creating opportunities in those separate domains. As a multipurpose product and/or as an element of a multifunctional system, the different potential functions and purposes of biochar need to be dissected and analyzed. In section "What Is Biochar and How Can It Contribute To Carbon Mitigation?," biochar is defined, and the key arguments as to why it might be useful in carbon mitigation are presented. In section "Biochar Production," the main ways in which biochar can be produced are covered, briefly describing the key technological issues and challenges. Section "Properties of Biochar" covers some of the properties of biochar. Section "Carbon Mitigation Potential of Alternate Production

Technologies" provides an account of the energy and carbon balance of the pyrolysis process that is at the core of biochar as a carbon abatement strategy. Section "Evaluating Carbon Abatement from Biochar" extends this to an analysis of carbon abatement across the biomass-bioenergy-biochar lifecycle and addresses three crucial questions.

- How much potential carbon abatement might arise from biochar globally?
- How efficient is carbon abatement through biochar compared to alternative use of the same organic matter across the life-cycle of the system?
- How cost efficient is carbon abatement through biochar?

Section "What Are the Impacts of Biochar on Soil" is a detailed analysis of the impacts of biochar in soils. In section "Conclusion: Evaluating the Sustainability of Pyrolysis-Biochar Systems," some preliminary conclusions regarding biochar and its wider sustainability are presented. Finally, in section "Future Directions for Research, Development and Demonstration," some key research needs and future directions are considered.

What Is Biochar and How Can It Contribute to Carbon Mitigation?

We define biochar as the porous carbonaceous solid produced by the thermochemical conversion of organic materials in an oxygen-depleted atmosphere and which has physiochemical properties suitable for the safe and long-term storage of carbon in the environment and, potentially, soil improvement. This definition is deliberately flexible and refers to both the production of biochar and its application.

Combustion of organic matter will take place if there is too much oxygen present during thermal conversion, and the resulting solid will be ash which typically contains just a few percent of carbon by mass, in addition to compounds and elements (phosphorus, potassium, other metals, etc.). With a low-level of introduced oxygen and thermal treatment of the organic matter, gasification may take place, during which volatile organic compounds are generated and released as vapor. This energy-rich synthesis gas ("syngas") can be cleaned and used for electricity-generation.

Pyrolysis occurs where the organic matter is subject to heat in the absence of any introduced oxygen and yields about a third of the feedstock as char (by weight), while gasification produces up to 10% char by weight. Charcoal is a type of char that has been produced (intentionally or otherwise) from wood for millennia, and much of our knowledge of biochar derives from the study of charcoal. Charcoal has also been used in soil-management practices for millennia and has well-documented benefits. While these are best observed in tropical environments – most famously in the *terra preta* soils of the Amazon – they have also been observed in temperate and semi-tropical regions.

It would be a mistake to equate biochar and charcoal however, since biochar represents a much wider group of materials which are likely to have far more variable properties than charcoal. Biochar comprises stabilized plant material in which carbon is stored mainly in a chemically recalcitrant form which will not significantly degrade through microbial activity or chemical reaction in the environment. It is this recalcitrance which is of interest from a carbon mitigation perspective since the carbon is thereby unavailable to microorganisms and does not return to the atmosphere as carbon dioxide (CO_2). But how long does carbon remain fixed in biochar? The mean residence time (MRT, the inverse of the decay rate) is the average time for which carbon in new biochar remains present in a stabilized aromatic form. The MRT of charcoal and analogous material is in the order of millennia [8]. Biochar may therefore provide an effective long-term store of carbon in soil and thus offer a potential abatement option for anthropogenic carbon emissions [9].

Stabilization of Plant-Captured Carbon

Annually, plants draw down 15–20 times the amount of CO_2 emitted from fossil fuels (7.5 Gt C year^{-1}), up to 20% of the entire atmospheric pool. About half of this is returned immediately to the atmosphere through plant respiration, but about 60 Gt C year^{-1} [10] is invested in new plant growth (about 45% of plant biomass is carbon) and contributes to net primary productivity (NPP). Since plant biomass is relatively constant globally, the magnitude of new plant growth must be approximately matched by harvest, litterfall, exudation by roots, etc. The annual CO_2 release from decomposition of these products by natural pathways and human cycling of plant-derived materials and products roughly equals NPP. The annual return of carbon to the atmosphere from the decomposition of all prior cohorts of plant material is thus approximately equal to NPP.

Intercepting and stabilizing plant-biomass production reduces the return of carbon to the atmosphere, with a relative reduction in atmospheric CO_2 (see Fig. 6.1). This reduction can be quite immediate if the default rate of decomposition is months to years, as it is for the dominant portion of biomass returned to soil in managed (agricultural and forest) ecosystems. Controlled charring (pyrolysis) can convert up to half of the carbon in plant biomass into chemical forms that are recalcitrant and, in principle, managed soils have a capacity to store pyrolyzed biomass at a rate significant in terms of emissions of carbon from fossil fuel.

The conversion of carbon in plant biomass to charcoal during natural fire is only about 1–5%, but the high level of stability established for such material in soil, which is generally a highly active biological medium, leads to expectation for similar stability in deliberately produced biochar. Biochar, deployed as a “carbon-negative” technology at the scale of 1 Gt C year^{-1}, would be equivalent to expanding the natural cycle of fire-derived charcoal storage in geographic terms and increasing its global magnitude by a factor of 4–20 [11].

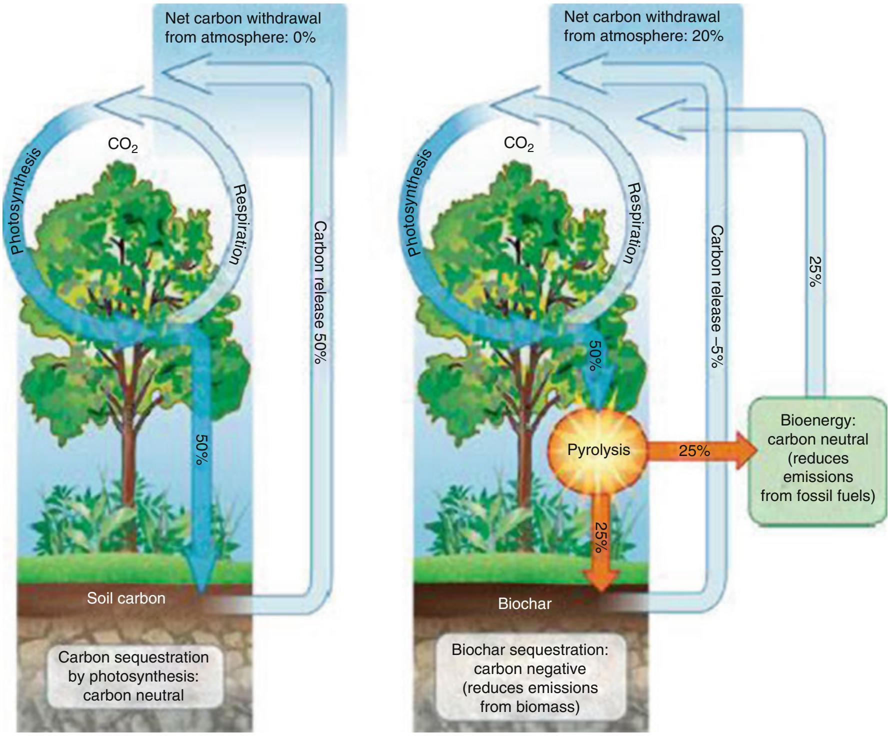


Fig. 6.1 Schematic illustrating the pyrolysis-biochar concept [9] (With permission from Nature Publishing)

Methane (CH_4) and nitrous oxide (N_2O) are the second- and third-largest contributors to radiative forcing of climate change after CO_2. Soil is a key source of both gases, which are emitted though natural microbial processes. The global significance of these processes has increased with agricultural expansion since higher CH_4 emission is associated with flooded soil conditions and with an enhanced nitrogen cycle (increased N_2O emissions) provided by the use of (synthetic) fertilizer, manures, and slurry. Since both CH_4 and N_2O are also associated in part with organic matter decomposition, stabilizing degradable organic matter could have a direct impact on soil-based emissions of these gases. Interventions in the nutrient or water balance of soil through changes in the dynamics of water in soil, or through the adsorption of nitrogen (as ammonium) may indirectly modify emissions of these gases from the soil.

To be a significant response to climate change, carbon abatement on a scale of millions of tons needs to occur, preferably hundreds of millions of tons. To intercept NPP and produce biochar at this scale presents a practical challenge, but still only involves a small fraction of total plant NPP (60 Gt C $year^{-1}$) of which 30% is already calculated to be “co-opted” by humans [12]. Increased efficiency of biomass recovery in managed ecosystems, diversion of biomass from current uses where it has a low value, and utilization of used biomass (organic waste) streams provide three ways in which it might be achieved. The fourth option is to sustainably harvest more biomass, which might be achieved by growing more productive plants, increasing the area of managed land, or adjusting harvesting regimes. Clearly, demonstrable effects on NPP arising from the deployment of biochar could be factored into such strategies.

Indirect CO_2 Equivalent Impacts

Biochar can improve the pore-size distribution of soils, resulting in an improved retention of plant-available nitrogen in the soil, increasing plant N uptake and fertilizer-use efficiency. This implies lower fertilizer requirement and lower eutrophication risks. If the accumulation of biochar results in beneficial change in pore-size distribution, it would result in a more permanent change than can be achieved through the effects of degradable and thus transient organic matter that can be used to condition soil.

The release of nitrogen by soil microbes from decomposition of crop residues in the low-growth winter period (in temperate regions) is a key source of nitrogen loss to water and the atmosphere. Increasing the use-efficiency of nitrogen in recoverable crop residues is therefore of relevance to eutrophication through leaching, and N_2O emissions.

Changing the abundance or physical position of aerobic space in soil (with respect to loci of microbial activity) has the potential to mitigate CH_4 emission. The emission of N_2O from soil could be suppressed by adsorption of nitrogen in the form of ammonium (NH_4^+). Emissions of both CH_4 and N_2O are notoriously variable both temporally and spatially, and are also sensitive to soil pH. However, these mechanisms may be relevant in the suppression of CH_4 and N_2O by biochar observed at certain locations, or under certain controlled conditions [13].

The Haber–Bosch process used to fix atmospheric nitrogen into solid soluble (ammonium) form is an energy intensive process that accounts for about 40% GHG emissions associated with arable agriculture. Typical use-efficiency for fertilizer nitrogen globally is 30–50% [14] attributable to leaching, immobilization and gaseous loss, and leaching. Technologies that improve the management of these processes through the soil can therefore offer an indirect gain in agricultural carbon-equivalent balance.

Liming of agricultural soil also transfers carbon from the geological pool to the atmosphere through production (calcining of limestone) and subsequent neutralization in the soil [15, 16]. The alkalinity typical of biochar can potentially substitute for the use of lime in the management of soil pH without emission of CO_2 [13].

Reliable and secure storage for annual and large cumulative amounts of biochar would have to be available, and while simple burial (for example, in landfill or disused mines) has been considered [4], these possibilities would be limited, costly and potentially dangerous. The broader land surface, and in particular soils that are already actively managed, may therefore provide the required capacity for a large and enduring strategy for storing carbon in biochar [17]. The key assumptions are that the estimates for stability of charcoal made so far are typical and accurate, that biochar from modern pyrolysis technologies using more diverse feedstock exhibits broadly similar levels of stability as natural charcoal, and that appropriate feedstocks can be provided sustainably and without adverse environmental or sociopolitical impacts.

Box 6.1: Is Biochar Geoengineering?

Geoengineering has been defined as the "deliberate large-scale manipulation of the planetary environment to counteract anthropogenic climate change" [18]. Biochar is regarded as a form of geoengineering by the Royal Society of London, along with afforestation and associated removal of wood for long-term applications and a range of physiochemical methods, including direct air capture, ocean fertilization, ocean alkalinity enhancement, etc. This is because, at a large enough scale, biochar could have a noticeable influence upon the global carbon cycle. Yet, there are reasons why using the moniker geoengineering for biochar is misleading. Firstly, biochar might be a small-scale intervention. Secondly, biochar is not solely concerned with moderating global warming, and there will be instances where its main function is for agronomic purposes and soil improvement, water retention, leachate reduction, or treating contaminated land. Evaluating such projects as geoengineering could be misleading and result in unhelpful comparisons with very different technologies. In many cases, better comparisons can be made between the use of agricultural and organic residues and wastes for composting, incineration, gasification, second generation fermentation, anaerobic digestion, and biochar production.

Biochar Production

Processes

There are several processes which can be used to produce biochar, pyrolysis being the most common. Pyrolysis is a thermochemical decomposition process in which organic material is converted into a carbon-rich solid and volatile matter by heating in the absence of oxygen [19]. The solid product, char or biochar, is generally of high carbon content and may contain around half the total carbon of the original organic matter. The volatiles can be partly condensed to give a liquid fraction leaving a mixture of so-called "non-condensable" gases. Each of the three product streams from pyrolysis – solid, liquid, and gas – can have properties and uses that provide value from the process. There are two main classes of pyrolysis process as well as a number of other more or less related technologies that may be considered for biochar production.

Fast pyrolysis is characterized by high heating rates and short vapor residence times. This generally requires a feedstock prepared as small particle sizes, and a reactor design that removes the vapors quickly from the presence of the hot solids, typically at around 500°C, leading to high yields of liquid products with low char yields. There are a number of established commercial processes (as well as many R&D examples) where the target products are liquids – bio-oils – although biochar from such processes has also been studied [20]. The area of bio-oil from fast pyrolysis has been extensively reviewed [21, 22].

Of more interest for biochar production is slow pyrolysis, which can be divided into traditional charcoal making and more modern processes. It is characterized by slower heating rates, relatively long solid and vapor residence times and usually a lower temperature than fast pyrolysis, around typically 400°C. The target product is generally the char, but this will always be accompanied by liquid and gas products, although these are not always recovered.

Traditional processes using pits, mounds, or kilns, generally involve some direct combustion of the biomass, usually wood, as heat source in the kiln, which reduces the char yield. Liquid and gas products are often not collected but escape as smoke. As well as particulates and carbon dioxide, other greenhouse gases such as methane may be emitted, as well as other hydrocarbons, and amines, leading to a net positive radiative forcing effect even if the biochar product is used for carbon storage [24]. Hence, traditional charcoal-making techniques are not generally compatible with the objectives of pyrolysis-biochar systems (PBS) for carbon abatement.

Industrial-scale charcoal-making processes, using large retorts operated in batch or continuous modes, achieve higher char yields and avoid most of the issues of emissions by allowing recovery of organic liquid products and recirculation of combustible gases to provide process heat, either internally or externally [26]. Other developments have led to slow pyrolysis technologies which are of most interest for biochar production. These are generally based on a horizontal tubular kiln design, where the biomass is moved at a controlled rate through the kiln and include agitated drum kilns, rotary kilns, and screw pyrolyzers [27], as well as some gravity-driven designs. In several cases, these have been adapted for biomass pyrolysis from original uses such as the coking of coal with production of "town gas" or the extraction of hydrocarbons from oil shale. Although some of these technologies have well-established commercial applications, production of biochar is not yet one of them, and only limited reviews are available [27, 28]. Other technologies that may be considered for biochar production include flash pyrolysis (cf. fast pyrolysis but shorter residence times), intermediate pyrolysis (cf. slow pyrolysis with improved heat transfer, allowing faster throughput), flash carbonization (partial combustion in pressurized reactor), gasification (partial combustion in a gas flow), and hydrothermal carbonization (aqueous process at high temperature and pressure with catalysis) [28]. Typical values and reported

Table 6.1 Scope of pyrolysis process control and yield ranges

Process	Slow pyrolysis	Intermediate pyrolysis	Fast pyrolysis
Temperature (in degrees centigrade)			
Range	250–750	320–500	400–750
Typical	350–400	350–450	450–550
Time			
Range	Min-days	1–15 min	ms-s
Typical	2–30 min	4 min	1–5 s
Yields (% oven dry weight)			
Char			
Range	2–60	19–73	0–50
Typical	25–35	30–40	10–25
Liquid			
Range	0–60	18–60	10–80
Typical	20–50	35–45	50–70
Gas			
Range	0–60	9–32	5–60
Typical	20–50	20–30	10–30

Based on review of over 30 literature sources [28]

ranges for key process variables and product yields of slow, intermediate, and fast pyrolysis processes are shown in Table 6.1 [28]).

Products

Composition of the three typical product streams from pyrolysis processes, solids, liquids, and gases, will vary with feedstock, process design, and conditions but can be generalized as follows. The solid product, char or biochar, has a varying carbon content, typically ranging from 60 to 90% [29]. In terms of proximate analysis, the char consists of four major components; fixed carbon, volatile carbon, ash and moisture. Energy contents of biochar range typically 20–30 MJ kg^{-1} (Higher Heating Value (HHV), [28]).

Liquid products from biomass pyrolysis are frequently termed bio-oil. However, this is a somewhat confusing term as the organic liquid product is generally hydrophilic, containing many oxygenated compounds, and is present sometimes as a single aqueous phase and sometimes phase-separated, together with water produced in the pyrolysis reaction or remaining from the feedstock [19]. Energy contents of bio-oils range typically from 15 to 30 MJ kg^{-1} (HHV, [28]), but figures quoted may be given after a degree of purification. The gas product is termed synthesis gas, shortened to syngas. It is typically a mixture of carbon dioxide (9–55% by volume), carbon monoxide (16–51%), hydrogen (2–43%), methane (4–11%), and small amounts of higher hydrocarbons [28]. Literature values for syngas energy content ranges are sparse, partly due to varying composition during processing and the presence of inert gas, with available values ranging from 8 to 15 MJ kg^{-1} (HHV, [28]).

Effect of Feedstock and Process Variables

The nature and preparation of pyrolysis feedstocks as well as the process conditions used influence both the composition and distribution of products. The main effects are caused by feedstock properties, the gas environment, and temperature control; and are summarized here for slow pyrolysis [26].

High lignin biomass will tend to give higher char yields, with other components leading to more liquid and gas products. Minerals present in the ash can have a catalytic effect by increasing char yields in some cases. Moisture can have a positive or negative influence on char yields depending on conditions. Larger particle size can increase char yields by restricting vapor disengagement and increasing the scope for secondary, char-forming reactions.

Factors affecting the gas environment that lead to a longer contact time between hot solids (feedstock or char) and primary vapor products lead to increased char yields resulting from secondary char-forming reactions at the hot surface. These factors include particle size, heating rate, and gas flow rate; increased pressure has a similar effect.

Temperature control is one of the most important operational variable, particularly peak temperature. Higher peak temperatures lead to lower char yields and higher liquid yields. For instance, a typical biomass pyrolysis might yield 38% biochar by weight on a dry basis at 350°C, but only 27% when heated at 550°C [30]. Heating rates have a smaller and inconsistent effect on slow pyrolysis. Increasing residence time at peak temperature will lead to lower char yield, but again this is a smaller effect than temperature itself.

Temperature also influences the composition and structure of the biochar formed. Higher temperatures and longer residence times lead to chars with higher concentrations of carbon and fixed carbon (determined by proximate analysis), as more volatile matter is driven off; pore structure and surface area also develop with more severe conditions. Figure 6.2 shows some of these temperature effects, including the effect on elemental composition of char, in a series of experiments with pyrolysed beech-wood [26]. Note that the complementary decrease in char (residue) yield and increase in fixed-carbon content at successively higher temperatures leads to a plateau in the fixed-carbon yield above about 400°C in this example. The relationship between fixed-carbon and soil-carbon stability is not yet clear [30].

Energy Balance

During pyrolysis, components of the biomass feedstock react by different pathways contributing to the complex products observed. Individual reactions may be endo- or exothermic and the combined process may also be endo- or exothermic depending on conditions of reaction [26]. Even when conditions favor exothermic reactions, initial heating to achieve onset temperature is required. Heat input may be provided by an external heat source, by partial oxidation of the feedstock, or by recycling and combustion of one of the product streams.

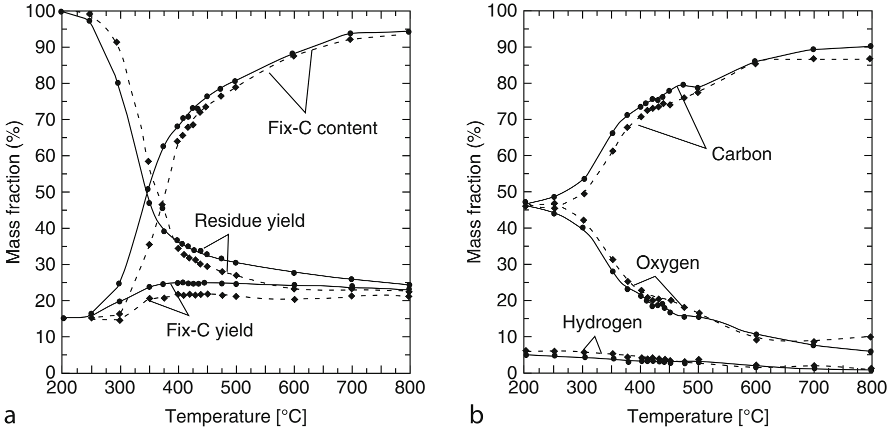


Fig. 6.2 Effects of temperature and heating rate on (**a**) char yield, and (**b**) carbon, hydrogen, and oxygen content of beech chars. *Solid and dashed lines* are 2°C and 10°C min^{-1} heating, respectively [26] (Reproduced with permission from the American Chemical Society)

In conceptual designs for pyrolysis-biochar systems, the syngas, bio-oil, or combined gas/vapor stream are the preferred energy sources for the process. Data for process energy is not generally available in the literature but estimates suggest a requirement in the order of 10% of the energy value of the dry feedstock. In most cases, the product distributions will leave an excess of energy in the gas and/or liquid streams which can be used for electrical generation or exported heat, but note that the higher the biochar yield obtained, the lower this excess energy will be, a significant point for the economics of the process.

Properties of Biochar

Cation Exchange Capacity

Biochar has the capacity to exchange cations (e.g., ammonium, NH_4^+) with soil solution, and thereby store crop nutrients. The extent of this cation exchange capacity (CEC) is effectively absent at very low pH and increases at higher pH [32]. Experimental results show that the CEC of fresh biochar is typically low but increases with time as the biochar ages in the presence of oxygen and water [33–35].

Specific Surface Area

Biochar has a very high specific surface area (SSA) of several hundred m^2 g^{-1} to a thousand m^2 g^{-1} (Fig. 6.3). The main parameters influencing SSA are pyrolysis

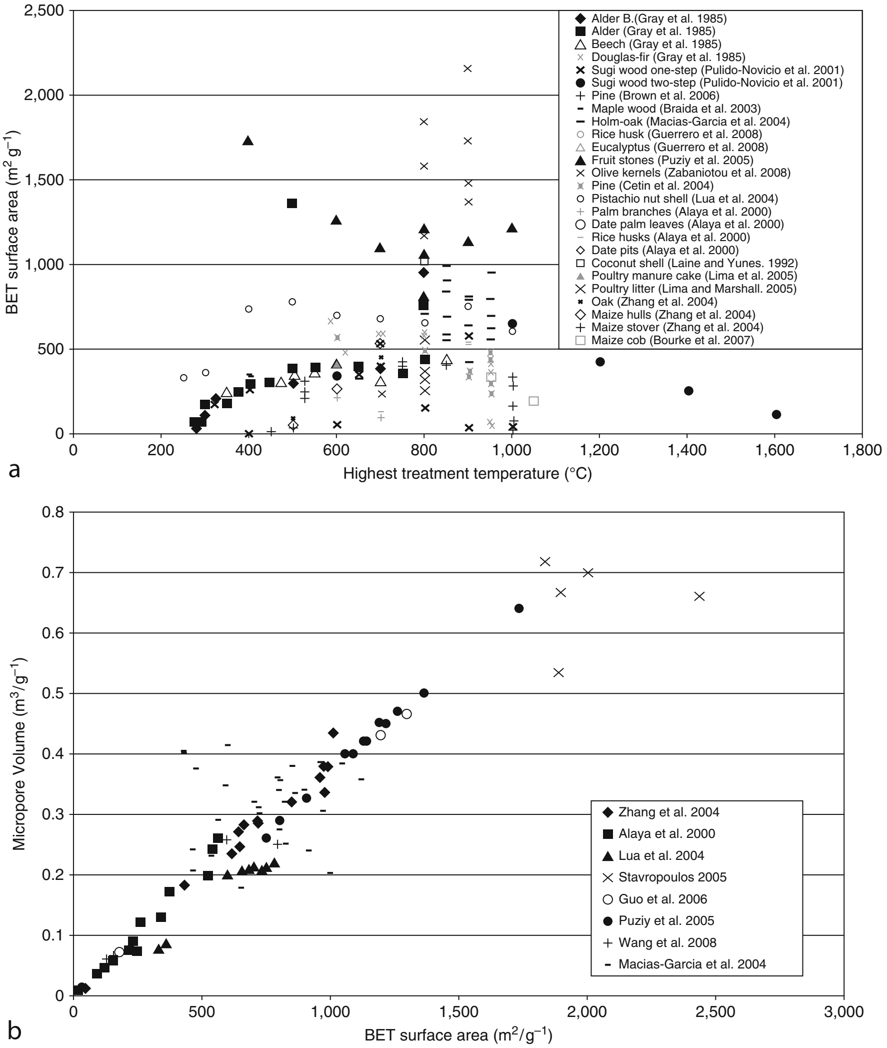


Fig. 6.3 Biochar surface area (**a**) plotted against treatment temperature and (**b**) its apparent relationship with micropore volume [36] (Reproduced with permission from Earthscan Ltd.)

temperature, heating rate, residence time, and presence of active reagents (e.g., steam, CO_2, O_2, etc.). Figure 6.3a shows that the total surface area of biochar from most feedstocks tends to increase with increasing pyrolysis temperature. This is mainly due to the development of micropores that are responsible for most of the surface area, see Fig. 6.3b.

At present, it is not clear whether the additional surface area presented by micropores plays as important role in soils as macropores and, therefore, whether it is beneficial to produce a biochar with very high SSA. It may be possible to produce biochar with high SSA in the macropore range. However, the physical structure of biochar tends to be defined by the starting material, so fine milling or compaction of the feedstock before pyrolysis is necessary to achieve a well-defined macroporous product [37].

Contaminants

There are two main potential sources of contamination in biochar: feedstock and the conversion process. Depending on the origin and nature of the pyrolysis feedstock, biochar may contain contaminants such as heavy metals (potential toxic elements, PTEs) and organic compounds. Some of these compounds will undergo changes in the conversion process and might be destroyed (or converted to benign compounds), while others will remain unchanged or give rise to potentially harmful substances. In addition to the contaminants introduced in the feedstock, some contaminants can also be formed in the conversion (pyrolysis) process. These include polycyclic aromatic hydrocarbons (PAH) and potentially, in some cases, dioxins. Additionally, the physical form of pyrolysis products may present a direct health risk, and increase or decrease the risk posed by elements, compounds, or crystalline material both in feedstock or formed during pyrolysis.

Heavy Metals

Heavy metals present in the feedstock (e.g., MSW, sewage sludge, treated wood, etc.) are most likely to remain and concentrate in the biochar (with lower char yields, resulting in higher concentration of PTEs) [38–42]. Therefore, careful selection and analysis of feedstock is necessary to avoid contamination of biochar with increased levels of heavy metals. Heavy metals are stable materials and therefore retained (conserved) during volatilization of associated organic molecules. The majority of metals will therefore be present as ash within biochar (together with nutrient elements such as phosphorus and potassium). It may thus be possible to manipulate contaminant loadings through selective removal of ash [43]. Alternatively, it has been shown that high-temperature pyrolysis can release heavy metals from the solid product, thereby yielding char with lower loading of these contaminants [44].

Polycyclic Aromatic Hydrocarbons (PAHs)

PAHs can be formed from any carbonaceous feedstock. The major chemical pathways for (PAHs) formation in the pyrolysis process are the high-temperature secondary and tertiary pyrolysis reactions (homogeneous and heterogeneous), as

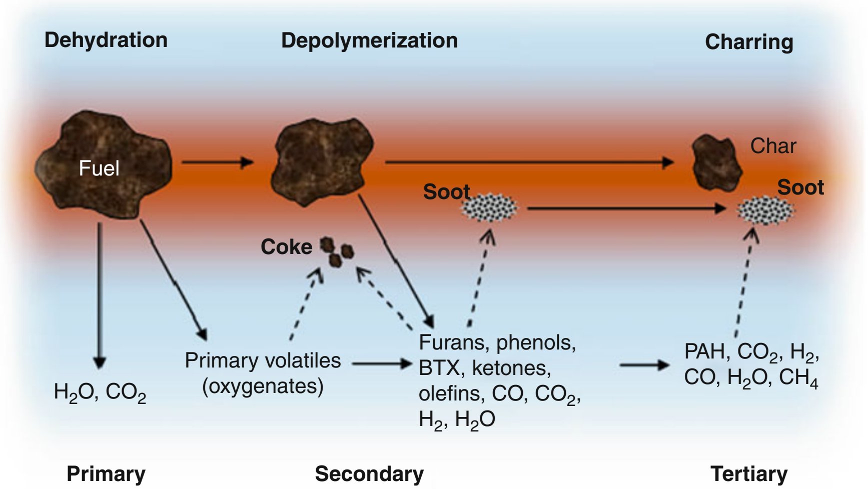


Fig. 6.4 Progress of fuel particle pyrolysis [51]

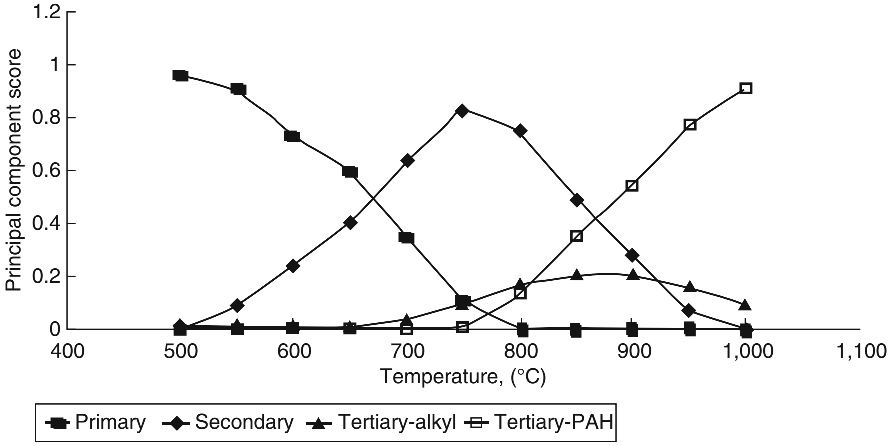


Fig. 6.5 The distribution of the four "tar" component classes as a function of temperature with 0.3 s gas-phase residence time [48] (Reproduced with permission from NREL)

shown in Figs. 6.4 and 6.5 [163]. The formation of these tertiary pyrolysis products increases with the pyrolysis severity (i.e., temperature and residence time) and becomes significant at temperatures around 750°C.

However, there exists also a second, less explored route for PAH formation. Evolution of PAHs from the solid substrate has been reported in the temperature range of 400–600°C [43, 45]. This pathway yields predominantly lower molecular-weight PAHs, although higher molecular-weight PAHs, such as benzo(a)pyrene, are also formed [45–47].

Table 6.2 Concentration of benzo(a)pyrene in biochar and UK soil [49, 50]

Benz[a]pyrene content (μg kg^{-1})				
Birch char	Pine char	Pine sleeper char	Urban soil (England)	Rural soil (England)
310	570	4,040	714	67

As the optimum temperature for biochar production is likely to lie in the region 450–550°C, in a well-controlled system (without hotspots), formation of PAH would proceed mainly by the evolution from solid substrates. PAH formation in the gas phase should be minimal due to the low pyrolysis temperature. Data on PAH content in pyrolysis char is scarce but indicate that the concentration and composition of PAHs in biochar are feedstock dependent [49]. Other data show that PAH concentrations in biochar produced from untreated biomass at temperatures of up to 600°C are considerably lower than those in urban soils in England, in the order of 10–100 mg kg^{-1} [50]. Biochar produced from chemically treated biomass is liable to contain considerably higher levels of PAHs than biochar from virgin feedstock due to the possibility of indigenous PAHs apparent in a study of biochar produced from railway sleepers previously treated with tar and creosote [49]. Available published data on the concentration of benzo(a)pyrene, one of the most toxic PAH compounds, is shown in Table 6.2.

Dioxins

Dioxins, unlike PAHs (which can be formed from any carbonaceous feedstock) require chlorine for their formation. Consequently, only biochar produced from feedstock containing significant amounts of chlorine (e.g. halogenated plastics) could be prone to generation of dioxins under certain conditions. The dioxin formation process is heavily dependent on the temperature history of the pyrolysis products, and relatively high temperatures (around 750°C) are required for the formation of dioxin precursors (chlorinated benzenes and phenols). Therefore, it is unlikely that biochar produced by pyrolysis at temperatures between 450–550°C would contain dioxins in significant amounts even when produced from feedstock containing chlorine, despite the fact that dioxins can be formed from their precursors in this temperature range. The only exception would be biochar produced from feedstock already contaminated by dioxins or dioxin precursors.

Stability

The stability of biochar is one of its key properties as it determines its potential for long-term storage of carbon. However, despite its importance, there is no recognized way of determining stability of biochar. It is very difficult to predict stability of biochar over timescales relevant to carbon sequestration, i.e., centuries to millennia. This difficulty stems from the diversity of processes (biological, chemical, and physical) responsible for biochar degradation in the environment and the wide range of properties biochar from different sources pose. It seems that simple correlation of long-term biochar stability with any particular physical or chemical property of biochar is difficult, and new methods, such as accelerated aging, are being investigated. Preliminary results have shown that high levels of stability can be achieved in a wide range of production conditions [30].

Carbon Mitigation Potential of Alternate Production Technologies

The overall effect of pyrolysis-biochar production on carbon abatement, prior to soil incorporation, can be described as the sum of two main factors: the carbon stored in biochar (related to CO_2 removed from the atmosphere) and the CO_2 emissions avoided through substitution of fossil fuels by combustion of pyrolysis products for energy. In calculating avoided emissions, a baseline case needs to be established for comparison, selection of which can have a large impact on the results. There are three elements for selecting a baseline:

1. *Carbon intensity of displaced fossil fuel energy*. Avoided CO_2 emissions are calculated relative to the average CO_2 equivalent emissions (including contributions of CH_4 and N_2O) from generation of grid electricity (or that from a specific facility). The carbon dioxide emission factor (CEF) of the grid varies over time with the mix of fuels used. In the UK, it has decreased over recent decades with the trend away from coal toward use of natural gas. Expressed as kg CO_2eq kWheq^{-1} electricity, CEF has fallen from 0.78 in 1990 to 0.55 in 2007 [52]. Given this trend, the UK government recommends using a CEF of 0.43 kgCO_2eq kWheq^{-1} for comparisons when considering renewable electricity development [53]. The actual CEF value, e.g., 0.5 kgCO_2eq kWheq^{-1} in 2008, could also be used. The most appropriate CEF would be employed in a specific case-study context. It will be less for an energy system that is less reliant upon coal and other fossil fuels.
2. *Carbon neutrality or otherwise of biomass system*. Combustion of biomass for energy generation releases c. 99% of plant carbon as CO_2; it is conventionally assumed that the same quantity of biomass is replanted (without incurring any additional greenhouse gas emissions), hence the same quantity of CO_2 is taken up by the growing plants and the system is “carbon-neutral”. Yet, if direct or indirect land-use change is entailed, carbon neutrality cannot be assumed: for instance, change from managed grassland (with c. 80 tC ha^{-1}) to energy-crop cultivation will involve immediate loss of CO_2 due to soil disturbance. Depending upon the energy crop, the equilibrium soil organic carbon content

may decrease (e.g., to 45 tC ha^{-1} in a wheat for bioethanol context) or possibly increase (e.g., by up to 20 tC ha^{-1} over 20 years in the case of Miscanthus) [54]. It is therefore more accurate to include considerations of land-use change and the feedstock carbon cycle explicitly. Below we calculate the net carbon abatement from PBS assuming biomass replacement (carbon neutrality) and no replacement.

3. *Carbon Stability Factor (CSF) of biochar*. The CSF is defined as the proportion of the total carbon in freshly produced biochar which remains fixed as recalcitrant carbon over a defined time period (see Glossary). As yet, little information exists on the actual CSF of specific biochar samples due to scientific uncertainties over biochar stability. A further uncertainty is the selection of the appropriate time period over which the stability is measured. Previous studies have used a range of values of the CSF from 0.68 over 100 years [55], 0.8 (time period undefined) [56], 1.0 (time period undefined) [57], 0.75 over 10 years [58], and 1.0 over 10 years [59]. As yet, there is no convention on the definition, measurement, and time horizon for reporting the CSF. To a certain extent, the selection of the time period is subjective and influenced by the decision-makers' preferences. In this review, we have adopted a time period of 100 years which is a compromise between the (multi-) millennial timescale of the climate system and the decadal (and frequently shorter) timescale of commercial and policy decision making. This also follows the convention of assessing Global Warming Potentials (GWP) to compare the radiative forcing of different GHGs over 100 years.

Equation 6.1 can be used to calculate the net carbon abatement arising from for combustion, PBS, or soil incorporation of biomass:

$$CO_{2na} = CO_{2av} + CO_{2fix} - CO_{2rel} \tag{6.1}$$

where:

CO_{2na} is net carbon eq. abatement
CO_{2av} is carbon eq. emissions avoided by replacement of fossil fuels
CO_{2fix} is carbon eq. fixed in the long-term (100 years)
CO_{2rel} is carbon eq. released by the biomass feedstock processing
(All expressed in $tCO_2eq.t^{-1}$ feedstock).

Meanwhile:

$$CO_{2fix(100)} = BM_{tot} \times BC_{yield} \times CO_{2tot} \times CSF \tag{6.2}$$

where:

$CO_{2fix(100)}$ is CO_2 eq. fixed over 100 years
BM_{tot} is biomass total dry weight
BC_{Yield} is biochar yield (ratio)

Table 6.3 Simple calculation of carbon stored and avoided CO_2 emissions arising from pyrolysis-biochar, combustion, and direct field incorporation for one oven dry ton of straw

Indicator	Combustion	Pyrolysis-biochar (with electricity generation)	Pyrolysis-biochar (no electricity generation)	Direct incorporation of straw into field[a]
Starting feedstock mass (t)	1	1	1	1
Carbon content at start (t)	0.45	0.45	0.45	0.45
Carbon content at end (stabilized) (t)[b]	0	0.25	0.25	0 year: 0.45 0.5 year: 0.27 1 year: 0.18 1.5 years: 0.11 2 years: 0.05
Expressed as CO_2 (t)	0	0.92	0.92	0 years: 1.65 0.5 year: 1.0 1 year: 0.66 1.5 years: 0.40 2 years: 0.18
Calorific value of straw: 13.5 GJ t^{-1}				
Efficiency of conversion	35%	15%	0%	0%
Delivered energy (GJ t^{-1})	4.725	2.025	0	0
Carbon emission factor: 0.5 kg CO_2 per kWh (2008 electricity mix)				
Delivered energy kWh	1,312.5	562.5	0	0
Avoided CO_2 emissions (t)	0.656	0.281	0	0
Total CO_2 abatement per ton feedstock (t) assuming carbon neutrality	0.656	1.2	0.92	
Total CO_2 abatement per ton feedstock assuming no biomass replacement (t)	−0.994	−0.449	−0.73	0 year: 0.18 0.5 year: −0.48 1 year: −0.81 1.5 years: −1.06 2 years: −1.28

[a] Assumes an exponential decay function with a decay constant of 1.0

[b] Assumes that 55% of the carbon in the feedstock is stabilized over 100 years

CO_{2tot} is total CO_2 eq. content of fresh biochar
CSF is carbon stability factor over 100 years
(All expressed in $tCO_2eq.t^{-1}$ feedstock).

Data for the example of 1 t of straw is given in Table 6.3. It can be seen that if combustion is used, 1.65 tCO_2 is released immediately, but there is an avoided emission of 0.66 tCO_2 arising from the substitution of fossil fuels (assuming a CEF of 0.5 $kgCO_2eq\ kWheq^{-1}$). The net CO_2 emission, assuming that there is no biomass replacement, is therefore c. 1 $tCO_2\ t^{-1}$ feedstock. If the same 1 t of straw is pyrolysed to produce biochar, the net CO_2 emission is lower at 0.45 $tCO_2\ t^{-1}$, assuming electricity generation from PBS is feasible. If no electricity cogeneration is possible, the net emission increases to 0.73 $tCO_2\ t^{-1}$, still lower than combustion (though obviously without the benefit of electricity generation). If full biomass replacement is assumed, biomass combustion and PBS both deliver net carbon abatement, though the biochar option more so.

If the alternative use of the straw is for incorporation into the soil however then the emission of CO_2 arising from decomposition would be slower. At day 1, 1.65 t of CO_2 remains in the biomass, and if we assume an exponential decay with a decay constant of 1.0, then after 4 years the vast majority of the straw has mineralized. Assuming that 5% of the straw biomass is stabilized as long-term soil carbon, then the direct incorporation abates more carbon from day 1 to 6 months. After that time, however, PBS with electricity generation and no biomass replacement abates more carbon, while after 1 year PBS without electricity generation and with no biomass replacement achieves higher carbon abatement than direct incorporation. The analysis is more complicated in the case of biomass replacement and is not attempted here. Clearly, this result is heavily dependent upon the decay function, and constant and slower rates of decomposition would give very different results under which direct incorporation would be more "competitive" in terms of carbon abatement relative to combustion and PBS.

In effect, pyrolysis has an associated "carbon debt" to pay-off due to release of CO_2 during pyrolytic conversion. The time period of this "carbon debt" is sensitive to the time horizon selected for measuring the CSF. If a shorter time horizon is chosen in measuring the CSF, then the carbon debt will appear to be smaller because the PBS CA will be larger, and vice versa. It is therefore important when evaluating options to be clear about what the PBS option is being compared to, what the CSF and time period is, and to use these numbers to calculate the carbon debt of PBS.

A simplified model was developed to calculate net carbon abatement for slow, intermediate and fast pyrolysis [28]. The data required for the model (Table 6.4) were gathered in a comprehensive literature review complemented by direct communication with relevant experts [36, 58, 60–64]. Model outputs are all expressed on a feedstock dry weight basis. A default value of 33% for electrical conversion efficiency of the main model output data is assumed but with no use of spare process heat (beyond drying the feedstock). The CEF used is 0.43 $kgCO_2eq$ $kWheq^{-1}$ and the results are shown in Table 6.5.

Results of the model show that fast pyrolysis may give the highest electrical energy product. This is due to the high liquid (oil) yields from the process, which can then be used for power generation. On the other hand, the electrical energy product is lowest for the slow pyrolysis process, where much of the energy value of the feedstock is stored in the biochar product. Abatement is greatest for slow pyrolysis, where most carbon is retained in biochar, and least for fast pyrolysis, where char yield is low. The values in Table 6.5 largely agree with those in Table 6.3, both in terms of energy product and net CO_2 abatement, with a 20% difference in calculated net carbon abatement for slow pyrolysis with associated energy generation.

Table 6.4 Data required for the pyrolysis process model

Model inputs	Slow pyrolysis	Intermediate pyrolysis	Fast pyrolysis
Process input			
Biomass			
Carbon content (%)	46	43	50
Energy value (MJ kg^{-1})	17*	16	19
Pyrolysis process data			
Gas yield (% input mass)	45	32	13
Liquid yield (% input mass)	15*	35	72
Char yield (% input mass)	40	34	15
Energy loss (% input)	6*	0*	3
Process energy (% input)	10*	10*	10
Primary process output			
Gas			
Energy value (MJ kg^{-1})	13.1	11.0	11.5
Carbon content (%)	37.4	30.0*	36.0*
Liquid			
Energy value (MJ kg^{-1})	0.0*	12.0	17.9
Carbon content (%)	0.0*	30.0*	46.5
Char			
Energy value (MJ kg^{-1})	25.0*	24.7	27.0
Carbon content (%)	72.3	70.0*	78.0*

* = Estimated. See text for sources of other figures

Table 6.5 Pyrolysis process model results (assumes carbon neutral bioenergy)

Model outputs, carbon stability factor 0.75	Slow pyrolysis	Intermediate pyrolysis	Fast pyrolysis
Energy product (kWh eq kg^{-1} oven dry feedstock)	0.38	0.56	1.18
Net CO_2 abatement (kg CO_2 eq kg^{-1} oven dry feedstock) Pyrolysis	0.96	0.88	0.80
Combustion	0.67	0.63	0.75

"Carbon-Negative" Energy?

It has sometimes been claimed that PBS is a "carbon-negative" energy system, this being an extrapolation from "carbon-neutral" bioenergy systems. Using Eq. 6.2, and assuming that BM_{tot} is 1 t, BC_{Yield} is 0.4, CO_{2tot} is 0.85 and CSF is 0.75, then the $CO_{2fix(100)}$ is 0.935 $tCO_2eq.t^{-1}$ (assuming biomass used is then replaced). From Table 6.4, this is associated with 380 kWh electricity generation, hence, it can be argued that PBS is not only carbon neutral, but in fact a carbon-negative system. This is technically correct but only under the assumption of biomass replacement, namely that the same quantity of carbon in the biomass is taken up as CO_2 through subsequent photosynthesis, and no other land-use emissions are entailed. Furthermore, since PBS is currently an inefficient way of generating electricity, the moniker "carbon-negative energy" can be misleading and generate confusion. If the prime intention is to generate electricity, it is likely far better in most cases to utilize a more efficient conversion technology than pyrolysis.

Evaluating Carbon Abatement from Biochar

In order to determine the importance of a carbon abatement strategy from PBS (or variants thereof, such as Gasification-Biochar Systems, GBS), a number of questions need to be addressed, taking a systems-wide view, including techniques such as resource assessment, land-use modeling, Life-cycle Assessment and Techno-Economic Modeling.

- How much potential carbon abatement might arise from PBS globally?
- How efficient is carbon abatement through PBS compared to alternative use of the same organic matter across the life-cycle of the system?
- How cost efficient is carbon abatement through PBS in economic terms?

What Is the Potential Carbon Abatement Level?

Carbon abatement from biochar is a function of the amount of biochar produced which, in turn, is a function of the amount of biomass or other organic matter that is available. The resource pyramid approach [65] can be used to distinguish between "theoretical available resources" (i.e., the total amount which exists), "realistic available resources" (which applies a first level of pragmatic judgment to limit the supply), and "viable available resources" (which applies a second level of pragmatic judgment to further limit supply, taking particular account of likely or possible other demands in the market place). Even so, resource availability scenarios are likely to be necessary to account for irreducible uncertainties in future supply and demand (such as scenarios reflecting lower supply, higher supply, and very high supply of feedstocks available for pyrolysis, or scenarios which reflect low, medium, and high levels of competition for any available biomass for uses other than PBS).

Lehmann et al. [17] estimate that current global *potential* production of biochar is 0.6 ± 0.1 gigatons (Gt) per year (10^9 t or PgC year^{-1}). Lenton [66] argues that the present potential for biochar production from agricultural wastes, forestry fellings, and from shifting agriculture ("slash and char") is somewhat higher than in [17], increasing the potential carbon abatement through biochar to 0.77–0.87 Gt C (PgC) year^{-1}. Lehmann et al. [17] estimate that by 2100, production of biochar could reach between 5.5 and 9.5 Gt year^{-1}, assuming that biomass is grown specifically for the purpose of PBS. There are very large uncertainties attached to these numbers however, arising from competition for land-use, competition for use of biofuels, agricultural residues, and organic wastes, and a huge divergence (of nearly 1,000%) in different expert estimates of the potential future global supply of biomass for bioenergy purposes (see Box 6.2). Roberts et al. [56] arrive at a much smaller value for global CA (0.65 Gt CO_2 year^{-1} or 0.18 Gt C year^{-1}) under the assumption that 50% of the 1.5 billion tons of currently unused crop residues globally is utilized for producing biochar. This might contribute 4% of the carbon reductions that are required globally by 2050 to limit climate change. However, after reviewing the literature on the potential production of biomass for bioenergy by 2100, Lenton [66] arrives at a carbon-abatement value from biochar that compares reasonably well with the larger Lehmann et al. value. He notes the potential constraint in carbon storage capacity in soil arising from biochar addition by about 2100, even assuming a high loading of 140 tC ha^{-1}.

Woolf et al. [68] have created and linked a global biomass feedstock availability model and a pyrolysis biochar production model, and calculate that between 1 and 1.8 Gt C year^{-1} is feasible by about 2050 from the biochar mitigation option. The range is explained by the use of three different scenarios of biomass supply; in each scenario, sustainability constraints have been met such that land upon which food is grown is not used for growing biomass for biochar production (i.e., only agri-residues and clean wastes are utilized) and indirect land-use change is not induced, incurring carbon-debt problems. Woolf et al. found that carbon abatement from use of biomass for biochar production was, on average, c. 20% higher than where the same biomass is used for bioelectricity generation through combustion. The comparison depends however on the fossil fuel offset. Where coal is offset, then biomass combustion and pyrolysis-biochar have similar carbon abatement. The higher bioelectricity generation from use of combustion avoids more carbon emissions due to the high carbon intensity of coal (compared, say, to gas, nuclear, renewables, etc.). Pyrolysis-biochar performs better in terms of carbon mitigation, where offsetting is against a fuel with a lower carbon intensity because the energy penalty of pyrolysis is therefore less important (from a carbon point of view) (a point also made by [69]). A further issue explored by Woolf et al. is the potential role of soil fertility in influencing the carbon mitigation potential of biochar; namely, in areas of low soil fertility, biochar is anticipated to have greater agronomic benefit, increasing yields with the use of fewer inputs (and their associated greenhouse gas emissions). This means that, according to this analysis and in terms of carbon mitigation, biochar is most effective, where it is incorporated into poorer quality soils in regions where less carbon-intensive fuels are being displaced.

The IEA's Energy Technology Perspectives project has suggested that a reduction of 13 Gt C year^{-1} is needed in 2050 relative to the "do-nothing" business-as-usual scenario [75]. If biochar could contribute 1 Gt C year^{-1} to "filling" this overall 13 Gt C year^{-1}, then its contribution would be roughly similar to the potential role of nuclear power expansion or enhanced power-generation efficiency and fuel switching.

Box 6.2: Estimates of the Global Potential of Biofuels

A review by the OECD identifies four potential sources of bioenergy: additional land brought into production; crop residues; forest residues; and other organic waste (plant and animal) [70]. The OECD report suggests that, globally, 0.44 Gha is the upper limit on the land area that could be made available for dedicated bioenergy crop production by 2050. Models of land availability tend to underestimate the land that is already in use (by 10–20%), while overestimating the amount of land that could be brought into production. Limited water availability and competition for food and fiber production are frequently overlooked. The OECD estimate on new land available for bioenergy cultivation compares with the average of 0.59 Gha calculated from 11 studies reviewed [71].

The OECD report estimates that the total bioenergy available from the 0.44 Gha of new land is 100 EJ year^{-1}. The potential for marginal and degraded land is put at 29–39 EJ year^{-1}. As for crop residues, only 25–33% of residues are available for extraction because of competing uses and the need to return some to soil for nutrient replacement. Using yields from IIASA [72], bioenergy from crop residues is estimated at 35 EJ year^{-1} in 2050. The IIASA study estimates bioenergy from forestry residues to be 91 EJ year^{-1}, while other organic residues and wastes are expensive to collect, hence the potential is limited to 10 EJ year^{-1} by 2050 [73]. All in all, the OECD estimates that the primary energy available for heat, electricity, and motive power that could technically be made available globally is 245 EJ year^{-1}, which is at the lower end of the range reported by the IPCC in its Fourth Assessment Report (125–760 EJ year^{-1}) [74].

The Carbon Abatement Efficiency of Pyrolysis-Biochar Systems

Carbon abatement efficiency (CAE) is defined as the net carbon abatement delivered for a given function unit (e.g., processing of a unit of feedstock, delivery of a kWh of electricity or heat, utilization of a given area of land, etc.). It is a way of comparing abatement efficiency between alternative uses of the same feedstock, land, or per unit of delivered energy. This is important in deciding how to use limited resources. The CAE is calculated from a life-cycle assessment (LCA) of the full PBS chain. An example of an LCA of a biochar system, including the impacts of biochar in the soil, is illustrated in Fig. 6.6.

A number of LCA studies of PBS have been conducted for a range of different feedstocks, technologies, and agricultural contexts. The results are summarized in Table 6.6.

Table 6.6 shows that some studies present significantly higher net carbon-abatement values than others. For instance, Gaunt et al. [59] present values for switchgrass that are several times larger than those of Roberts et al. [56]. Their

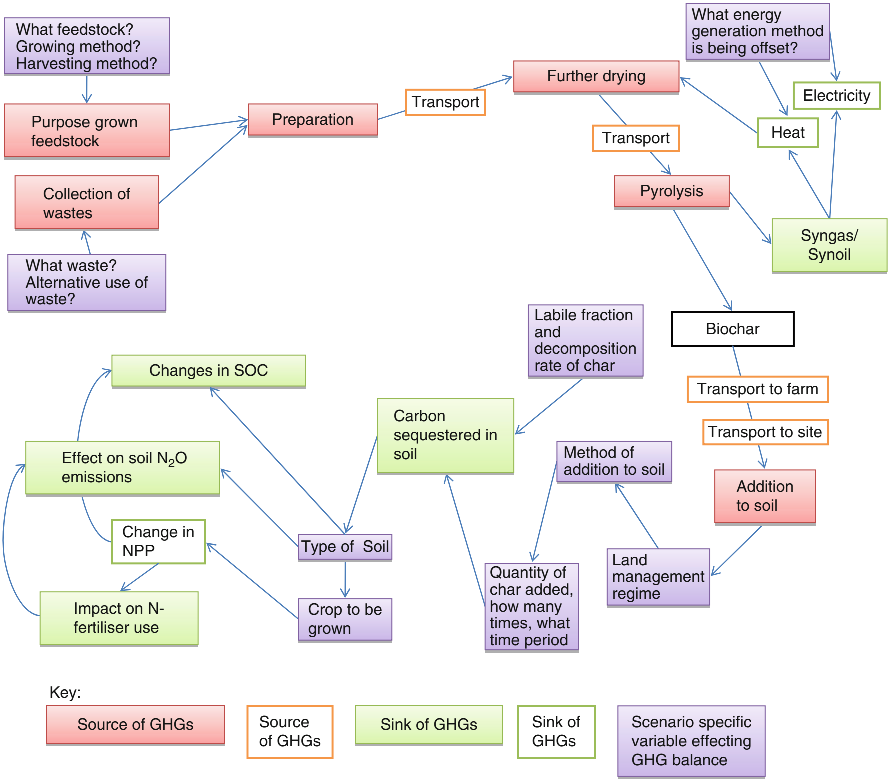


Fig. 6.6 A life-cycle system model for pyrolysis-biochar systems [55]

respective values are closer for corn stover (30% different). At first glance, the net carbon abatement for Miscanthus from Gaunt [59] and Hammond et al. [55] appear to agree, but not when the results are compared with inclusion of the indirect effects of biochar in soil in the former study (in which case, the Gaunt et al. study has 60–100% greater net carbon abatement than the Hammond et al. study). While some results do appear to converge, the overall impression is of a wide range of different assumptions leading to a high degree of uncertainty. At present, it is not known which assumptions are most appropriate and in what context. For instance, the assumed CSF of studies in Table 6.6 varied from 0.68 to 1.0 (for a range of [not always specified] timescales). This difference alone can account for a 50% variation in net CA.

Different assumptions about useable or delivered energy from pyrolysis are also important contributors to the uncertainty. Some studies assume a much higher energy conversion efficiency than others, e.g., Gaunt and Lehmann assume a value that is substantially higher than conventional biomass combustion, and even gasification, suggesting that useable heat is also being utilized. Roberts et al. also assume effective use of heat from pyrolysis, hence use a high overall energy

Table 6.6 Comparison of different life-cycle assessment studies of pyrolysis-biochar systems

Variable	Description	Gaunt and Lehmann [59]	Gaunt and Cowie [58]	McCarl et al. [76]	Roberts et al. [56]	Hammond et al. (2011) [55]	Ibarrola [57]
Feedstocks		Switchgrass, miscanthus, forage corn, wheat straw, corn straw	Green waste, cattle manure, wheat straw	Maize stover	Corn straw, yard waste, switchgrass	Wheat, barley, and oilseed rape straw, UK and Canadian forestry residues, short-rotation forestry, short-rotation coppice, miscanthus, sawmill residues, arboricultural arisings	Green and garden waste, sewage sludge, food waste, anaerobic digestate, construction and demolition wood waste
National context and applicability		USA	Undefined	USA (mid-West)	USA	UK	UK
Energy ratio	Energy outputs divided by energy inputs	Forage corn: 1.3 All others: 4.3 to 5.9	Not specified	Not specified	Corn stover: 2.8 Switchgrass: 3.1	Not specified	Not specified
Overall energy efficiency	Delivered energy (heat or power) divided by energy in feedstock + fossil fuel energy to operate process	38%	Green: 20% Cattle man.: 15% Wheat straw.: 25%		29% Heat	6–15% Electricity	20% Electricity
CO_{2av}	CO_2 avoided through fossil fuel substitution (% of total)	19–25%	10–18%	Not specified	26–40%	10–25%	20–30%
BC_{Yield}	Fresh biochar yield (as proportion of feedstock) (% DM)	8.5–8.7% (fast pyrolysis)	35–42%	35%	29%	33.5%	35%
CSF and time period	Carbon stability factor	1.0 over 10 years	0.75 over 10 years	1.0 time period not defined	0.8 (loss over small number of years then stable)	0.68 over 100 years	0.68 over 100 years

(continued)

Table 6.6 (continued)

Variable	Description	Gaunt and Lehmann [59]	Gaunt and Cowie [58]	McCarl et al. [76]	Roberts et al. [56]	Hammond et al. (2011) [55]	Ibarrola [57]
CO_{2tot}	Carbon content of fresh biochar			0.75	0.63–0.68	0.75	C&I wood: 0.72 Green: 0.50 Sewage sludge: 0.38 Food: 0.62
Proportion of the CA that is stabilized C in char (% of total)		58–63%	41–45%	Not specified	54–66%	40–50%	45–55%
Proportion of the CA that is indirect effects of char in soil (% of total)		17–19%	40–48%	Not specified	2–10%	25–40%	15–25%
Assumed size of pyrolysis unit (tons per annum)			16,000	70,000	10 t h^{-1} c. 50,000 tpa	Small: 2,000 Medium: 20,000 Large; 100,000	
CO_2na ($tCO_2eq.t^{-1}$ oven dry)	Net carbon-equivalent abatement	Gaunt and Lehmann [59]	Gaunt and Cowie [58]	McCarl et al. [76]	Roberts et al. [56]	Hammond et al. [55]	Ibarrola [57]
	Corn stover			0.8–1.1	0.793–0.864		
	Switchgrass	1.307	1.16/1.82		0.442		
	Green waste/yard waste				0.885		0.855
	Miscanthus	1.328				1.14	
	Wheat straw		0.99/1.65			0.84	
	Cattle manure		1.085/1.745				
	Sawmill residues					1.17	
	Forestry residues					1.24	
	Short-rotation coppice					1.13	
	Sewage sludge						0.774
	Food waste						0.965
	Anaerobic digestate						0.785
	Construction and demolition waste						1.048

Notes	1	2	3	4	5	6
Reference	Gaunt et al. [59]	Gaunt et al. [58]	McCarl et al. [76]	Roberts et al. [56]	Hammond et al. [55]	Ibarrola [57]

1. The CEF assumes 50% coal and gas, hence it is the average of the two values in Table 6.3 in Gaunt and Lehmann [59] converted from per hectare to per ton basis using the yield figures provided in the paper. The yield for corn stover is not given, so the CO_2na cannot be calculated.
2. The CEF assumes 50% coal and gas, hence it is the average of the two values in Table 18.2a ([58]). The first value shown does not include the application of biochar to crops or indirect effects of biochar upon soil incorporation. The value of these effects can be inferred to be from 0.2 (low), 0.66 (medium) and 1.14 (high) $tCO_2eq.t^{-1}$. The second value shown uses the medium values of the indirect effects and adds this quantity to the pyrolysis + fossil fuel offsets value. If the lower estimate of the indirect effects is used instead, the overall effect is reduced by c. 0.4 $tCO_2eq.t^{-1}$. In calculating the proportional contribution to total CA from the avoided fossil fuel emissions, stabilized carbon in char and the indirect impacts of biochar in soils, the medium value of the indirect effects have been assumed.
3. Lower value for switchgrass refers to slow pyrolysis; the higher value refers to fast pyrolysis.
4. For corn stover, the lower value is early stover and the higher value is late stover. In the case of the switchgrass, this does not take account of indirect land-use change (ILUC) arising from use of bioenergy crops. If ILUC is included, then the net carbon abatement is -0.036 $tCO_2eq.t^{-1}$ (i.e., an emission).
5. The value for wheat straw is the same as for barley and oilseed rape straw. Where large quantities of forestry residues are imported into the UK from Canada, the CO_2na is 1.08 $tCO_2eq.t^{-1}$. The value for short rotation coppice (SRC) is similar to the values for small round wood (1.15 $tCO_2eq.t^{-1}$).
6. Results for green waste, sewage sludge, and food waste are relative to incorporation of material in landfill sites with UK-levels of methane recovery. Values are also provided for incorporation of material into fast pyrolysis (10% char yield).

efficiency. Hammond, on the other hand, uses a more conservative value for net energy efficiency, which is substantially lower than straight combustion. How energy is treated in the LCA is important in making comparisons with CA from straight combustion or gasification since it is frequently assumed that heat is not readily used from such technologies where the principal purpose is electricity generation. Comparing net CA from PBS with delivered power and heat with biomass combustion with only power generation is probably not a fair comparison to make.

Some element of “biochar proponent optimism” has likely entered into the existing studies, and a more critical approach will be needed for the future. In summary, there is a moderate to high level of uncertainty attached to all existing values, and one should not pay too much attention to the precise numbers as they are very likely to change in the future as more understanding and experience is gained. The lack of reliable engineering data on the slow pyrolysis process at a commercial scale is one of the critical uncertainties. Most of the existing studies have used data from a single technology (BEST Energies, now Pacific Pyrolysis Pty. Ltd.), yet the results from this process have not been published in the peer-reviewed literature. This creates a potential weakness in the current argument in favor of biochar that needs to be addressed by acquisition of much better engineering data on slow pyrolysis.

Energy-Output to Energy-Input Ratios

The energy-output to -input ratio (also known as the energy yield) is the quantity of delivered energy (i.e., useable power and/or heat) divided by the quantity of energy required to produce that energy. Bioenergy systems typically utilize as fuels widely distributed biomass resources which require energy – frequently supplied by energy dense fossil fuels – to cut, prepare, transport, store, process, and ignite feedstocks. Fossil fuel–derived energy is also required to manufacture, transport, and erect the equipment and infrastructure that is needed for the bioenergy system to function. In calculating the energy ratio, the energy content of the biomass itself is not included as an energy input since this is treated as “free energy,” having been derived from the sun’s energy through plant photosynthesis. The biochar energy system can be depicted as a set of inputs and outputs as in Fig. 6.7.

The energy ration in Fig. 6.7 is calculated as:

$$\text{Energy ratio} = \frac{\text{net energy output}}{\text{energy inputs}} = \frac{\text{E5}}{\text{E2} + \text{E3} + \text{E4}}$$

The use of highly dense energy sources (fossil fuels) to enable the utilization of very distributed bioenergy sources has to be carefully assessed to avoid the problems that have beset the production of bioethanol from maize in the USA. In

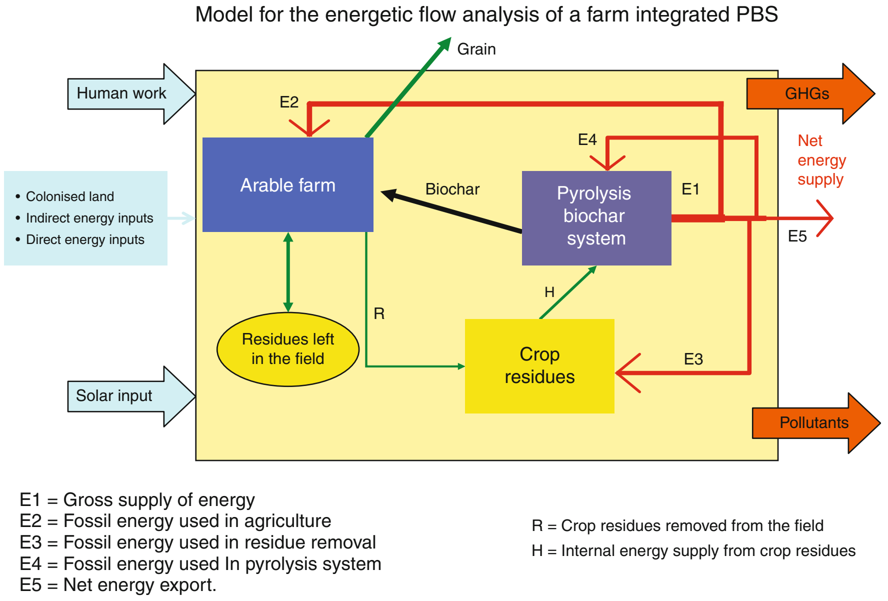


Fig. 6.7 Pyrolysis-Biochar as an Energetic System (Jason Cook after Giampietro and Mayumi [77])

that case, the energy output/energy input ratio (at between 0.7 and 2.2 MJ MJ^{-1}) is generally too low for the system as a whole to make energetic sense: what happens in effect is that the energy content of fossil fuels is being released to produce bioethanol with a similar energy content. The energy output/energy input ratio needs to be 2.0 or more for a bio-energy system to make energetic sense [77].

Several estimates of the energy-output/energy-input ratio of PBS are available, though due to the lack of reliable data on the pyrolysis process itself, especially at commercial scale, such estimates remain tentative. Gaunt et al. [59] provided a range of values that range from 2.3 to 7, depending upon the feedstock. However, Gaunt et al. use the gross energy output in calculating the energy yield, whereas the convention is to utilize the net energy output. The recalculated energy yield ranges in [59] from 1.3 (forage corn), 4.3 (switchgrass), 4.6 (miscanthus), to 5.9 (wheat straw and corn stover). The highest energy yields are associated with the use of crop residues (wheat straw and corn stover) since the energy inputs are lower for these feedstocks than for dedicated bioenergy crops (switchgrass and miscanthus). The study [59] assumes a biochar yield of c. 8.5–9%, with 38% of feedstock energy available as delivered energy. If a more modest net energy efficiency of 15% is assumed, however, then the energy yield is reduced to 1.1 (switchgrass), 1.2 (miscanthus), 0.1 (forage corn), and 1.7 (wheat straw and corn stover). With a more conservative, and some would argue, more realistic assessment of the net energy efficiency, the energy yield falls below the critical value of 2 and is unlikely to make energetic sense.

Roberts et al. [56] provide values for the energy ratios of 2.8 for corn stover and 3.1 for switchgrass; however, as with Gaunt et al., this study makes highly optimistic assumptions regarding the net energy efficiency, using a value of 37%, requiring productive use of the heat from syngas combustion. Reliable and economic markets for heat from power plants are notoriously difficult to create, and much analysis avoids inclusion of heat in calculations of avoided fossil fuel emissions for this reason. It is therefore optimistic to assume effective markets for heat from pyrolysis plants, therefore. If the more conservative assumption is made that only electricity generation from pyrolysis at 15% efficiency will find an economically viable market, then the energy yields from Roberts et al. can be recalculated downward as follows: late stover from 5.5 to 1.65, early stover from 3 to 0.63, switchgrass from 5.5 to 1.65, and yard waste from 9.5 to 2.4. The energy yields turn out to be highly sensitive to the efficiency of the conversion process to delivered energy.

Assuming that Giampietro and Mayumi are correct in identifying 2 as a critical value for the energy yield for biofuels, below which the basic energetics of bioenergy systems cease to make sense, then it is apparent that pyrolysis-biochar systems need to be operating at net energy efficiencies of at least 20–30%, depending upon the individual feedstock and technology assumptions. Anything which increases the use of fossil fuels in the PBS (*ceteris paribus*) will also pose a challenge to the system energetics. Roberts et al. [56] report, for example, that an increase in transportation distance from the baseline (15 km) to 200 km reduces the net energy by 15%, while at 1,000 km, the net energy decreases by 79%. An 80% reduction implies an energy yield of 1 or below, but even a 15% reduction in net energy could bring the energy yield below 2.

More work on accurate calculation of energy yields is therefore urgently required. The most promising scenarios will be where forestry residues and other organic wastes are being utilized, i.e., where fossil fuel inputs to the provision of the feedstock are minimized (in the case of many wastes because some form of treatment is required in any case) and where long transportation distances are not required. The carbon-equivalent production emission for sawmill residues in the UK is 4 $\text{kgCO}_2\text{odt}^{-1}$, while in the case of forestry residues, there is a negative emission of c. 50 $\text{kgCO}_2\text{odt}^{-1}$ as a consequence of avoided methane emissions from wood that otherwise decomposes [55]. UK arable straw entails a higher production emission of c. 200 $\text{kgCO}_2\text{odt}^{-1}$ (partly because c. 15% of the fossil fuels required for the arable crop production are allocated to the straw on economic grounds) [55]. This is actually greater than the production emissions of SRC and Miscanthus in UK conditions, where chemical fertilizers are not used (c. 20–40 $\text{kgCO}_2\text{odt}^{-1}$). Utilization of sewage sludge in SRC and Miscanthus results in higher N_2O emissions, though the sludge has to be disposed of in the baseline case so could arguably be ignored in LCA calculations.

It could be argued that if a key purpose of PBS is carbon abatement, critical values arising from energetic analysis are not necessarily relevant. This point may be valid, where the production of energy from the biomass is an ancillary benefit of a PBS development. For instance, the main purpose of a project may be the more effective disposal of an organic waste stream, and energy production a fortunate by-product and bonus: the waste would otherwise need to have been managed in some fashion. Yet, where the biomass has an alternative use as a fuel in co-firing, anaerobic digestion (AD), fermentation, dedicated biomass combustion or gasification, and where a market for such biomass and for bioenergy exists (e.g., whether with or without the aid of incentives), then an energetic analysis is appropriate to use. This is because a lower energy yield has to be compensated by increased energy production (or reduced demand) from some other part of the system.

Key Findings from Existing LCA Studies

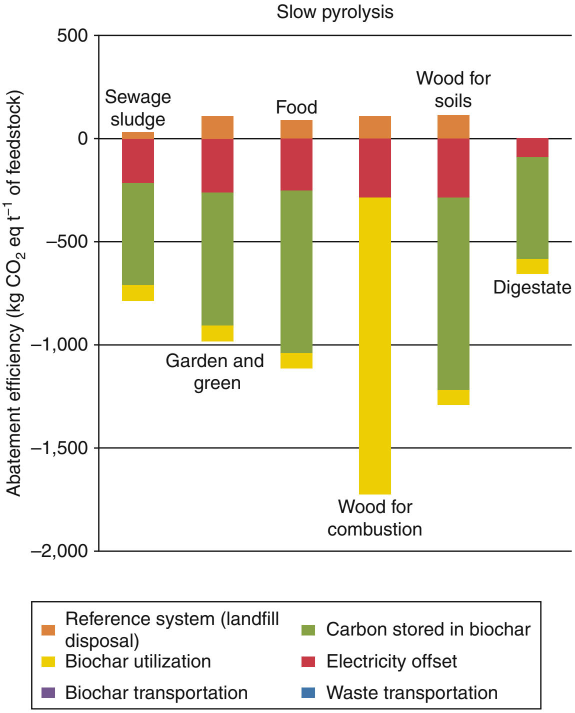
Feedstock Suitability

Hammond et al. [55] found that systems which utilize woody residue feedstocks tend to have the highest CAE, closely followed by purpose-grown woody feedstocks. Small-scale straw-based systems have a 15–30% lower CAE than wood residues, partly because of assumed scale-factors; the rest of the difference is explained by higher inputs for straw-based systems versus wood residues.

Roberts et al. [56] examined the impact of land-use change arising from the conversion of cropland from annual crops to perennial switchgrass (direct change) and the subsequent need to convert land to cropland to replace lost agricultural land (indirect change). They included two estimates of the size of these direct and indirect land-use changes (886 and 406.8 $kgCO_2\ t^{-1}$ dry switchgrass). If the larger land emission value is used, then the overall CA of the PBS is negative (i.e., a positive emission of 36 $kgCO_2\ t^{-1}$), but is positive if the lower land-use-change value is used (442 $kgCO_2\ t^{-1}$). Roberts et al. comment that PBS could conceivably *increase* net radiative forcing from GHG emissions if direct and indirect land-use-change emissions are associated with energy-crop establishment.

Ibarrola [57] found that biochar production from pyrolysis of wood waste (construction and demolition, plus commercial and industrial), garden and green waste, and food waste have greater CAE than sewage sludge or AD digestate (Fig. 6.8). This is because of the higher calorific value of the former, and the higher stabilized carbon content of their biochar product. Gaunt and Cowie [58] present a similar figure for CA of green waste compared to conventional landfill with CH_4 recovery (1.0–1.2 t $CO_2eq.t^{-1}$ oven dry feedstock). Joseph et al. [79] have recently presented more results for a range of nonvirgin biomass feedstocks in Australia, including poultry litter, paper sludge and green waste: the CA is between 1.4 and just over 2.0 t $CO_2eq.t^{-1}$ oven dry feedstock, the somewhat higher values being explained by the assumed waste-management baseline (which recovers less CH_4 emissions than is typical for many European countries).

Fig. 6.8 CA of PBS using different nonvirgin biomass feedstocks



Life-Cycle Stage Contributions to Carbon Abatement

Hammond et al. found that the largest contribution to CA is from stabilized carbon in biochar, accounting for approximately 40–50% of total CA. The next largest contribution is from the indirect impacts of biochar in the soil, all of which are currently uncertain: lower crop fertilizer requirement, lower soil N_2O emissions and increased SOC. These account for 25–40% of CA (the proportion changing with the size of other CA categories). The third major CA category in Hammond's et al. study is fossil fuel offsets from renewable electricity generation at 10–25% of total CA. Similar information is presented in Fig. 6.9 in which the life-cycle stages of the UK lower resource supply scenario are broken-down to illustrate CO_2 emitting and abating stages.

In the Roberts et al. study, the proportion from stabilized carbon in the biochar is larger at 54–66%. The proportion from avoided fossil fuel emissions is also larger at between 26% and 40% depending on feedstock.

The main difference between Hammond et al. and Roberts et al. is that the former study assumes a higher value for the indirect soil impacts of biochar, principally due to the assumed accumulation of soil organic carbon as a consequence of biochar addition. As a consequence, the proportional contribution of stabilized carbon and avoided fossil fuel emissions is lower than in Roberts

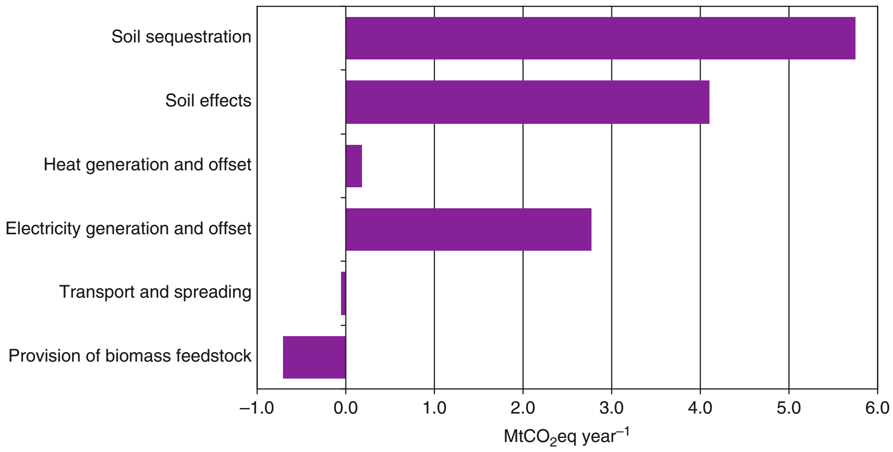


Fig. 6.9 CO_2eq abatement by life-cycle stage for lower biomass supply scenario (UK conditions)

et al., which makes more conservative assumptions about the indirect impacts of biochar in soil. Of the studies reported in Table 6.6, the indirect impacts of biochar in soils upon net carbon abatement tend to be smaller than assumed in Hammond et al., although Gaunt and Downie assume even higher values. Ibarrola's study presents a similar contribution breakdown as Roberts: the largest contribution to CA for both fast and slow pyrolysis in the case of wood, food, and green wastes is carbon stabilized in biochar (45–55%, excluding use of digestate case). The second largest contribution comes from offset GHG emissions from fossil fuel emissions (20–30%). Where the assumed indirect impacts are lower, then the contribution of stabilized carbon and avoided fossil fuel emissions are proportionally higher. The studies in Table 6.6 indicate that transport emissions are a relatively minor contribution to overall lifecycle emissions (several percent for biomass and biochar movement of c. 20 km each).

CO_2 Equivalent Emissions Per Unit Delivered Energy

Hammond et al. presents a carbon abatement for PBS of 1,500–2,000 kg CO_2eq MWh^{-1} (1.5–2.0 kg kWh^{-1}) for large systems, compared to a UK CEF of 0.56 $kgCO_2kWh^{-1}$ in 2006 [53]. For comparison, modern bioenergy systems (combustion with grate or fluidized bed, gasification) produce *emissions* from between 0.03 and 0.07 kg CO_2 kWh^{-1} [80], or from 0.05 to 0.30 kg CO_2 kWh^{-1} according to the Environment Agency [81]. While PBS appears to offer far better CA MWh^{-1} than conventional bioelectricity, this is a somewhat misleading finding. Much of the CA from PBS results from stabilized carbon in the biochar and from indirect soil effects (rather than from offset fossil fuel emissions), while the

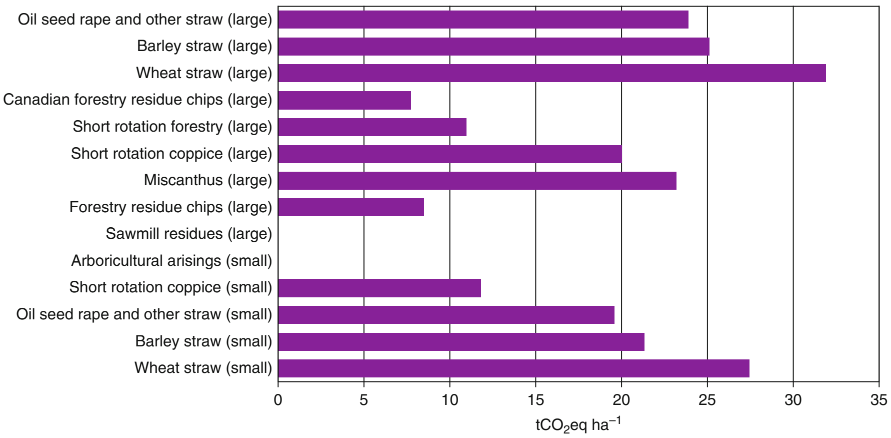


Fig. 6.10 Annual CO_2eq abatement per hectare for UK conditions. No value is given for sawmill residues or arboricultural arisings since the plants are not grown specifically for pyrolyzable residues

denominator – electricity generation per unit biomass – is lower than for conventional bioelectricity due to lower efficiency. Thus, the CA per unit electricity is high, but electrical efficiency is low.

CO_2 Equivalent Emissions Per Hectare

On an area basis in the UK, Hammond et al. found that CA ranges from seven to nearly 30 $tCO_2eq\ ha^{-1}\ year^{-1}$, depending on PBS feedstock (Fig. 6.9). Waste feedstocks such as arboricultural arisings are the most efficient in terms of land use as they do not require any additional land use change and do not interfere with crop-production systems; moreover, they are commonly disposed of as wastes and hence PBS incurs less emissions through additional transport, handling, and storage stages. Such numbers compare favorably with conventional bioenergy in the UK, which abate between 1 and 7 $tCO_2eq\ ha^{-1}\ year^{-1}$; the most productive biofuel system in the world – bioethanol from sugar cane in Brazil – abates c. 16 $tCO_2eq\ ha^{-1}\ year^{-1}$, so biochar performs well under this metric (Fig. 6.10).

Delivered Energy Generation from Pyrolysis-Biochar Systems Versus Combustion

Roberts et al. compared use of biochar for soil with use of biochar to replace coal and found that biochar to soil resulted in 29% more carbon abatement (627 vs 864 $kgCO_2eq\ t^{-1}$). A more realistic comparison is between PBS and direct biomass combustion and, in this case, Roberts et al. found that carbon abatement was actually less for PBS than for combustion (987 vs 864 kgCO_2eq t^{-1}) (using a CEF for natural gas). On the other hand, approximately half of the CA from PBS involves removal of CO_2 from atmosphere, compared to biomass combustion, where all the carbon abatement arises from avoided fossil fuel emissions.

In Hammond's et al. study, PBS performs somewhat better compared to combustion than in Roberts et al., partly reflecting somewhat higher CA per ton feedstock in the former than in the latter. The comparison depends upon the relative energy efficiencies of the two processes and upon the CEF used to calculate avoided fossil fuel emissions. For example, at an efficiency of 15% and with a CSF of 0.68, PBS appear to offer greater CA than combustion at 33% efficiency (using an average UK grid CEF) even without inclusion of indirect soil effects, but cannot compete with combustion (or gasification) at an efficiency over 40%. In systems co-firing biomass with coal, efficiency can be significantly higher in new plants (up to 42%). Hence, either the indirect benefits of biochar upon net GHG emission fluxes would need to be on the scale proposed in Hammond or else the efficiency of the PBS would need to increase (or some combination of the two). Where biomass co-firing is combined with CO_2 capture and geological storage (Bioenegy CCS, or BECCs), PBS is not able to compete in terms of carbon abatement (because the CCS process captures c. 90% of the carbon in the feedstock compared to c. 50% of feedstock carbon conserved during pyrolysis).

If grid average of 80 kg CO_2eq MWh^{-1} is attained by 2030 (which is required in the UK context if the government's carbon-reduction targets of an 80% reduction by 2050 relative to 1990 levels are to be met) and assuming biomass is still available as a resource, biomass combustion offers almost no carbon abatement benefit [82]. PBS meanwhile still offers CA benefits, i.e., it has net negative CO_2eq MWh^{-1} emissions. Yet, as noted above, at current net energy efficiencies, PBS is unlikely to be the technology of choice for generating electricity so it may have a rather limited role.

From the results of Roberts et al. and Hammond et al., it is evident that PBS is not necessarily more efficient in terms of CA than other bioenergy options: it will depend on the detailed analysis of each individual case. It is clear in general that conversion of the energy in feedstock into useful delivered energy (heat and power) will have to be reasonably efficient (20–30%) for PBS to compete on any scale with direct combustion. In the longer-term perspective, however, PBS may become more valuable (compared to combustion technology) due to its ability to actually remove carbon from the atmosphere. Exactly when this benefit would be realized is highly uncertain and context-dependent (e.g., reliant upon the outcome of other highly uncertain processes).

Sensitivity Analysis

Hammond et al. found that the following variables were all important in influencing the overall net carbon abatement: total handling losses, char yield, total electrical efficiency, use of heat, soil organic carbon accumulation, the allocation of GHG emissions to the production of the feedstock, fraction of labile carbon and – most importantly – the Mean Residence Time (MRT). If the MRT is below 500 years, there is a reduction in the CAE which begins to look concerning, although this partly depends on the time horizon over which an analysis is undertaken. Hammond found low sensitivity to the following variables: distance travelled (biomass and biochar) (up to 200 km), reduced nitrogen fertilizer application (0–50%), and reduced nitrous oxide emissions (0–100%).

In their LCA, Roberts et al. identified broadly similar sensitivities to Hammond et al. The results were sensitive to: emissions entailed in feedstock collection, stability of the carbon, biochar yield, and syngas yield. There was a lack of sensitivity to N_2O emission suppression and to distance travelled (except where this was 500 km +). Needless to say, the results of any such LCA are sensitive to the assumed CEF of the fossil fuel which is offset.

There are large uncertainties associated with the LCA work presented here. Biomass production systems vary in space and with time, making a calculation using a single number problematic. For the nonvirgin waste feedstocks, considerable uncertainties occur with respect to the management of individual landfill sites (e.g., the biodegradable fraction, oxidation factors, CH_4 recovery, etc.), making comparison of PBS to existing options difficult.

How Cost-Effective Are Pyrolysis-Biochar Systems?

Gaunt and Lehmann [59] calculated that the cost of reducing a ton of CO_2 in the PBS they examined was between \$9 and \$16 compared to utilizing the same char as a fuel. Since they did not undertake a full economic costing, this figure does not allow comparison with other marginal abatement carbon costs (MACCs).

McCarl et al. [76] undertook a full economic costing and found that the net present value of the PBS examined (70 ktpa corn stover, mid-west USA conditions) was $-\$70\ t^{-1}$ feedstock for slow pyrolysis and $-\$45\ t^{-1}$ for fast pyrolysis, i.e., it is a loss-making venture under these assumptions. This assumed a carbon value of \$4 $tCO_2{}^{-1}$ abatement and an agronomic value of $\$33t^{-1}$ biochar or $\$11.5\ t^{-1}$ feedstock. The biochar production cost (i.e., ignoring revenues from biochar as a form of carbon storage or arising from its agronomic value) is therefore approximately $\$85\ t^{-1}$ feedstock or c. $\$240\ t^{-1}$ biochar. The "energy penalty" cost of utilizing char as a soil amendment rather than as a fuel is \$40 $tCO_2{}^{-1}$, considerably higher than Gaunt and Lehmann's estimate of \$15 $tCO_2{}^{-1}$, though similar to other estimates (e.g., Lehmann [9]).

Roberts et al. [56] present data on net present value of their USA-based PBS (c. 50 ktpa) which appears to indicate some positive NPVs. This is as a consequence of a very high carbon-price assumption (\$80 $tCO_2{}^{-1}$). Even their "low" carbon price (\$20 $tCO_2{}^{-1}$) is actually higher than the 2008–2010 EU ETS market value of \$10–20 $tCO_2{}^{-1}$. If we remove the carbon revenue from the calculation along with the small benefit in increased fertilizer efficiency (but retaining the value of the

P and K nutrients in the char), the NPVs are all negative. The production costs, expressed per ton of biochar, are: \$155 t^{-1} for late stover, \$124–142 t^{-1} for switchgrass, and \$13 t^{-1} for yard waste. These costs are lower than McCarl et al. [76], but not much so (40% or so lower). The exception is for yard waste in which case the cost of production is much lower due to the revenue gained through tipping fees and the other avoided costs of organic waste management.

Shackley et al. [158] present economic data on the situation in the UK for a range of feedstocks, with three plant sizes (small: 2 ktpa; medium: 16 ktpa; large: 184 ktpa). They provide a range of values of the costs of biochar production from \$-220 to \$580 t^{-1}. The negative values arise from waste feedstocks, where there is a revenue stream from tipping fees which can otherwise be large in the UK context. The production costs are typically lower for the large-scale pyrolysis units due to lower capital, operational, and maintenance costs per unit production. The costs also vary depending upon the assumed storage option. For the virgin feedstocks, production costs are closer to McCarl et al. than to the Roberts et al. estimate above.

Brown et al. [161] compared the internal rate of return (IRR) for slow and fast pyrolysis assuming an increasing carbon price to 2030, use of gas co-product for heating and bio-oil refined to gasoline (in the case of fast pyrolysis only). The projected IRR for slow pyrolysis is negative out to 2030 for feedstock costing \$83 per ton. A zero cost feedstock would deliver an IRR of between 8% and 17%, but this is not considered sufficiently profitable for investment in a new technology. Brown et al. [161] found that the profitability of both slow and fast pyrolysis depended primarily upon the value of the energy product(s). Yoder et al. [162] develop a product transformation curve using optimisation and econometric analysis of existing data for quantifying the trade-of in pyrolysis between bio-oil and biochar production.

As for the LCA, costs are typically context-specific, and there are some niche applications where the NPV will be more evidently favorable, e.g., where a type of biochar has a high agronomic or soil-related value. A further example is where the biochar is a “waste” product from an economically viable energy project (Box 6.3).

To summarize, the economic viability of biochar production and application are currently highly uncertain. Feedstocks – especially clean ones – are frequently expensive in developed countries and increasingly in demand by other users such as for Anaerobic Digestion, composting, combustion, gasification, and so on. Technology costs associated with pyrolysis are especially hard to predict at the present time, and most estimates in the literature are based upon one or a few designs. At present, the incentives structure in most countries is focused upon renewable electricity generation, and there is no mechanism for rewarding stabilized carbon abatement in the soil. If carbon abatement is the primary policy driver, inclusion of stabilized carbon in biochar and its indirect impacts on soil GHG fluxes would need to be given some value alongside renewable electricity generation. One problem with carbon-based land crediting is that it raises the issue of how to establish a baseline, and many current land-owners and occupiers, e.g., farmers, are reluctant to begin to establish inventories of carbon-equivalent fluxes over their land (such as might be required to establish a baseline against which a biochar project could be assessed). Some countries, e.g., in Europe, have previously expressed their skepticism at including land-based carbon crediting as a major carbon-abatement strategy within the UNFCCC. Scientific uncertainties and technical challenges surrounding monitoring, verification, accounting, and reporting (MVAR) in relation to biochar additions will mean that developing a robust methodology for inclusion of biochar in carbon markets (voluntary or through the Clean Development Mechanism) will be challenging until scientific knowledge improves.

Where biochar is potentially more economically viable is where it is able to treat wastes that incur high tipping fees to landfills. In those situations, PBS may be a cheaper disposal route than landfill or incineration. Whether the char produced can be used in agricultural soils remains uncertain due to the risk of contamination. At present, there is a lack of a clear risk assessment and regulation pathway for such substances. A further situation where biochar might already be economically viable is the case of gasification char – which is a waste product of a financially solvent energy generation technology. If this char can be shown to be beneficial to soils and to avoid introducing contamination, then it could be financially viable to distribute such material to agricultural systems.

Box 6.3: Gasification of Rice Husks: Case Study from Cambodia

Rice husks are gasified in an Ankur gasifier to produce syngas that is fed into an engine that powers a rice mill. The system is economically viable (due to mill being off-grid, hence otherwise having to rely upon expensive diesel fuel for power generation). Carbonized rice husks (CRHs) are the waste product, which accumulate, and can become an environmental problem. The CRH yield is c. 30%, and the carbon content of the char is c. 35%. Therefore, for each ton of rice husk, 300 kg of CRHs are produced containing 105 kgC or 385 kgCO_2. The labile C content is <1% and the CSF is 0.92 hence the stabilized carbon content is c. 350 kg$CO_2$$t^{-1}$ biochar. The cost of application to agricultural fields in Cambodia is low – estimated at \$1t^{-1}, while the agronomic value (based upon unpublished research) appears to be c. \$2 to 8 t^{-1}. Since the CRHs are (currently) free, the value of the biochar is c. \$1 to 7 t^{-1}; this is an important potential source of additional income in a subsistence farming system such as Cambodia.

If the rice husks were otherwise disposed of in irrigated paddy, there is also the value of avoided methane emissions from gasification. For every 1 t of applied rice husks, 40 kg of carbon is converted into 53 kg of methane, equivalent to 1,219 kg CO_2.

If the stabilized carbon can have a value, say at \$5 t^{-1} CO_2, then the additional value of the CRHs is \$1.6 t^{-1} biochar. Adding to the agronomic value, the overall value is between \$3 and 8 t^{-1} biochar. And if the CA value of the avoided CH_4 emission is included, this would further rise to about \$9–14 t^{-1} biochar. Clearly, if the CRHs do indeed have a demonstrable and predictable agronomic value, demand for its use may increase, and the producer may begin to sell the CRHs rather than give it away free as a waste product.

What Are the Impacts of Biochar on Soil?

In this section, we turn to the question of the impacts of biochar on soil. A summary of the published literature has been presented previously [6, 7]. Much of the evidence comes from the study of charcoal in the natural environment: this provides the only source of relevant direct evidence for long-term stability of biochar, having been used historically by humans or as a result of deposition after periodic fires in many natural ecosystems. Given the similar formation and chemical characteristics, charcoal in the natural environment provides a powerful tool to investigate the long-term stability of biochar. However, the short-term impacts of biochar may not be well represented in studies of old charcoal. This is in part because the feedstock can be quite different, and partly because the more complex composition of biochar is overlooked. Also, any labile components associated with the charcoal will have been mineralized prior to sampling.

The more temporary beneficial impacts of biochar may be chemical and result from leachable ash and modification of soil pH, promotion of short-term microbial activity, including the effect of small labile fractions. Physical benefits may arise from modification of soil bulk density, water holding capacity, and promotion of soil aggregation (possibly in combination with soil biological effects). These effects may be temporary or long term. Thermal properties may change as well [83].

Other effects relate to the provision of cation exchange capacity (CEC) and specific surface area (SSA), biological associations (with micro-organisms, fungi, and with plant roots), and bio-physical benefits (mediating the connection of micro-organisms and microbial substrate, promotion of mesofaunal activity, including earthworms). The potential for detrimental effects on the soil would depend on the source of the biochar applied, and the rate and timing of its application. Negative impacts could include leaching of nutrients, addition of toxic elements (metals), or the introduction of organic contaminants. Where biochar has a high affinity for nitrogen, there may be negative short-term effects on crop nutrient supply, i.e., potentially reducing nitrogen availability to the plant in the period after application [84].

Scientific research of biochar is a relatively new topic, and therefore generality in site-specific observations is not yet apparent, while extrapolation from individual observations is not yet possible. At this point, a convergence in methodologies has not emerged and until recently, there have been no strategic research programs to provide a systematic evaluation. The nature of PBS also demands coordination and consolidation of research efforts with pyrolysis engineering in order to produce selected biochar that expresses particular and possibly multiple beneficial functions in soil.

Key Functions of Biochar

Provision of Labile Organic Carbon

Rapid utilization of labile substrates in soil can build a store of nutrients in soil microbial biomass, which may become available for plant acquisition and growth over time. The potential benefits of labile carbon in soil can create a constraint to crop growth if substrate nitrogen is low, and if at the time of addition inorganic (i.e., available) nitrogen in the wider soil is limited. This is because nitrogen as well as carbon is required to build new biomass, and microbes will out-compete roots.

Nitrogen is progressively volatilized during pyrolysis so the ratio of carbon to nitrogen in biochar is generally much higher than in the feedstock. However, if biochar is entirely stable, it will not present the readily accessible carbon substrate necessary to create microbial demand for external nitrogen. Whether significant nitrogen immobilization of soil nitrogen occurs should therefore depend on the size of the biochar addition, the size of the labile fraction, and whether the ratio of nitrogen to carbon of the labile fraction reflects that of the bulk biochar sample.

Storage of Stable Carbon

The stable portion of biochar is the fraction for which, in the future, a carbon credit might be claimed and, for the purpose of climate-change mitigation, may be the component that remains in the soil for at least 100 years [7]. There is, as yet, no robust methodology for establishing the MRT of a specific biochar product: this is one of the key scientific uncertainties and policy needs. The sum of stable and labile carbon should not reflect the total carbon content of biochar as fractions of intermediate stability are also likely to be present.

Supply of Plant Available Nutrients

Aside from nitrogen, most potential nutrients in pyrolysis feedstock are largely conserved during pyrolysis (as also are potentially toxic elements). Progressive elimination of carbon, oxygen, and hydrogen during pyrolysis therefore increases the total concentration of minerals in the char residue, and in potentially extractable forms as ash. Biochar ash content increases in inverse proportion to retained carbon feedstock, analogous to that which arises during combustion but distributed within a complex physical matrix.

Solubilization of ash may result in minerals becoming available to plants on addition to soil, although since phosphorus (as phosphate) is rapidly complexed with minerals in soil, this may depend on scavenging directly from char by roots or symbiotic mycorrhizal fungi. In general, introduction of readily available crop nutrients can promote mineralization of organic matter, especially in marginal environments.

However, porosity, and more specifically pore connectivity, may control the release of soluble nutrients from char, making release progressive rather than instantaneous as may be the case in the solubilization of combustion ash. This process could be associated with the mineralization of condensed tars and oils that appear to block biochar pores [85].

Modification of Soil pH

The typically alkaline nature of biochar may increase microbial activity in acidic soils by increasing pH and with this, another potential source of "priming" for the decomposition of pre-existing organic matter, although modification of soil pH may also increase plant productivity and thus the amount of carbon substrate added to the soil through roots and residues. Mass for mass, the value of char in pH modification may be up to one third that of agricultural lime [86] and at experimental rates can increase soil pH by 1 unit.

Modification of Soil Physical Characteristics

Depending on the distribution of particle size in the soil, the rate and nature of biochar applied and the time since application, soil pore–size distribution and water holding capacity may be affected. Porosity in char may occur at a range of scales, which affects the proportion of water that can be retained, and equally the accessibility of held water and solutes to plants which can exert sufficient tension to extract the contents of macropores (0.1–30 μm diam) that may not drain naturally. Structurally sound pores of this dimension are abundant in fresh wood-derived charcoal [87], and the connectivity of the relatively small number of larger pores has been investigated in three dimensions by tomography [88].

However, the fresh particle size of powdery charcoal created from grass feedstocks appears to be less than 50 μm [89], and weathered charcoal, while generally found in larger fragments, also resides in this size range [90]. In clayey soils, particles may be less than 5 μm [91]. During weathering, and particularly for char from woody substrate, the position of char fragments within the soil mineral matrix is likely to alter over time. The effect that this has on total porosity, accessible pores, and accessible surface areas has not been explicitly examined.

Nonetheless, while initially macroporous, it is established that the great majority of total porosity in wood-derived charcoal may reside in micropores of nanometer size [92]. Oils and tars could represent less stable components of biochar around which microbial activity could promote micro-aggregation, relevant to water infiltration and resistance to water erosion. An apparent role for charcoal in aggregation has been observed in field soils [90], although short-term incubation with activated charcoal did not cause aggregation under controlled conditions [93].

Cation Exchange Capacity and Sorption

Progressive abiotic and biotic surface oxidation of charcoal results in surface proliferation of carboxyl groups and an increasing ability to sorb cations [33, 34], explaining high cation exchange in archaeological soils [35]. Negative charge provides the possibility for reversible storage of available nitrogen (ammonium, NH_4^+) relevant to soil-based N_2O emissions and nitrate leaching. A mechanism based on the dehydration of phosphate and charcoal has also been described for the adsorption of phosphorus [94], which may explain the apparent impact of biochar on crop phosphorus uptake, possibly aided by arbuscular mycorrhizal fungi [95].

Charcoal has the capacity to sorb polar compounds, including many environmental contaminants [96], particularly PAHs for which it may be the dominant sink in soils and sediments [97]. The significance of biochar addition in removing contaminants from the environment depends on its capacity to fulfill this function relative to charcoal, the affinity (security and reversibility) of stabilization, and the ultimate fate of both char and contaminants [164].

Microbial Activity

The possibility that biochar catalyses breakdown of organic matter by providing microbial habitat alone is improbable since sustainable microbial proliferation depends on a renewable source of accessible carbon substrate as well as nutrients. Provided that the majority of biochar carbon is highly stable, after an initial flush of mineralization, microbes that inhabit biochar pores will depend primarily on the indirect effects of biochar to obtain an enhanced supply of substrate. This could either be through the capture and retention of soluble organic matter otherwise lost to deeper horizons or watercourses, or through a change in loci of plant root activity. Increased plant productivity however should be reflected in increased exudation of carbon through roots, and the deposition of carbon from residues of above-ground growth. The complication that this adds to interpretation of field data has been highlighted by Major et al. [98].

Many plants can form symbiotic associations with mycorrhizal fungi, whose filamentous hyphae provide an extension to plant roots which can enhance acquisition of both nutrients and water, at the expense of some host plant carbon. Although potentially limited by inoculum, soil-chemical conditions and the abundance of exploitable nutrients are more likely to limit mycorrhizal proliferation in most circumstances. Reported promotion of mycorrhizal activity by biochar [99, 100] could reflect utilization of reversible stores of water and soluble nutrients, or exhaustible “mining” of nutrients embedded in ash. Warnock et al. [101] proposed a range of possible mechanisms.

Limitations of Existing Research Base

Until recently, there have been no directed research programs to strategically evaluate biochar for its function in soil. Much of the current understanding of the function of biochar rests on published data for charcoal, often in the context of natural systems and wildfire. For reasons highlighted in sections “Biochar Production” and “Properties of Biochar,” the function of biochar in soil is strongly influenced by feedstock and formation conditions, and charcoal may only provide an insight into some general principles of biochar function in soil. In addition to the problem of extrapolating from studies of charcoal, pilot- and commercial-scale pyrolysis may produce biochar that differs from the products of bench-scale pyrolysis systems operating under ostensibly the same conditions. There is no existing research to evaluate char from gasification, which is likely to provide a function between that of biochar and ash from straight combustion.

Categorization of Current Literature

- *Sediment or soil*: Early evidence for the stability of biochar arose from sediment studies, where charcoal is preserved under anoxic conditions in which decomposition proceeds inherently slowly [102, 103].
- *Static or dynamic*: Static studies provide snapshot comparisons for a response variable at locations where a relevant soil (or other) variable differs, e.g., presence or absence of vegetation burning history [104, 105]. In a dynamic experiment, a “treatment” is imposed, and change in response variables over a time period determined, or ideally its trajectory observed through intermediate measurement. Chronosequence studies are a variant of the latter that enable long-term dynamics to be studied using samples deemed comparable, aside from the point in history at which the (ideally singular) change or intervention occurred [106–108].
- *Biochar or charcoal*: Natural fire contains an anoxic zone, where biomass pyrolysis prevails over combustion. Natural fire yields low charcoal conversion rates in the range 0.1–5% [109]. Wildfire is typically brief and the peak temperature variable. With rapid heating rates, the conversion may be partial, superficial, or progressive and affected by vegetation moisture content. Wildfire and charcoal are significant considerations in the global carbon cycle, and now an established topic for research.
- Much work on the dynamics of charcoal has been undertaken in this context, and laboratory studies have aimed to mimic wildfire carbonization by exposing biomass to similarly brief, variable, but generally low (ca. 350°C) temperature

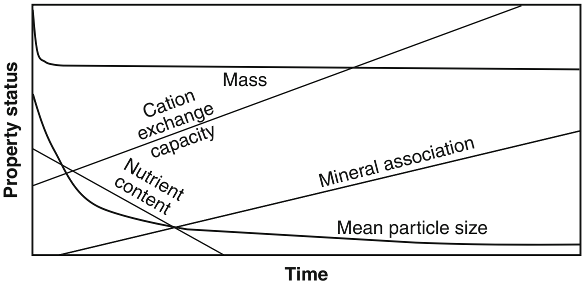


Fig. 6.11 Schematic to illustrate the challenge of unraveling multiple functions of biochar whose possible trajectories strongly differ

and semi-oxic conditions, i.e., limited or partial restriction of air flow. Experiments with char produced with complete exclusion of oxygen are much more limited and recent. Published research using char from commercial pyrolysis reactors rather than material produced in a laboratory-scale batch process are very scarce. Only eight studies categorized in [7] used pyrolysis char.

- *Short term or long term*: The various functions of biochar (Fig. 6.11) may be manifest over different timescales and, crucially, the trajectory of these functions appears to be nonlinear and may not be entirely independent of other functions or the wider system. The average duration of the dynamic studies identified in one literature review [7], including those undertaken in field plots (but excluding long-term chronosequences), was 11 months. Although the chronosequence approach has been employed to observe the development or demise of functions that change slowly over time (rather than emerge or decline rapidly at the start), few attempts have been made to short cut such change, other than by imposing favorable laboratory-imposed conditions.
- *Gasification or fast or slow pyrolysis*: After oven-drying, plant biomass usually contains about 45% carbon by mass, and a few per cent mineral ash. Ash is broadly conserved, but the proportion of carbon that is retained is specific to the process. Combustion leaves trace amounts of carbon, gasification less than 10%, and pyrolysis typically 30–40%. Ash includes key mineral nutrients such as phosphorus and potassium, other metals and a range of micronutrients concentrated by loss of total feedstock mass in the conversion process [110]. The nutrient value of the products differs markedly on a carbon-mass basis, but in terms of their value to crops will depend not only on rate of char application but on the physical accessibility of nutrients in the char to leaching, plant roots, and mycorrhizal fungi. The production process and feedstock mineral content will also modify the pH of the char by-products, which tend to be moderately to highly alkaline.
- *Feedstock*: Scanning electron microscopy of fresh charcoal and charcoal aged in the natural environment reveals a cellular structure resembling that of the woody feedstock from which it was derived, e.g., lignified cell walls of dead xylem cells. The structure of char from grass and nonwoody plant material is rarely

reported and, similarly, nor is the structure of char produced from digested or composted materials.

- *Tropical or temperate*: Under otherwise equal conditions (moisture, nutrient, and substrate availability), biological activity increases with temperature. Consequently, soils in the tropics tend to be depleted in organic matter and associated biological activity relative to those from temperate regions. In addition, soils that are very old have usually been subjected to extensive weathering and leaching and display low-inherent fertility and are often acidic. Although all functions of biochar may be expressed at all locations, some will be more conspicuous in such soil. In field studies and controlled experiments, half of the research effort has been undertaken in regions with above 20°C mean annual air temperature, and only one fifth in temperate zones below 10°C.
- *Laboratory or field*: Laboratory conditions enable variables and functional attributes to be isolated or controlled, and the impact of climatic variation to be removed. Permutations of different factors are possible since the space and resource requirement may be relatively small and good replication is possible. The interactions between functional attributes of soil and biochar with the wider environment, such as fluctuations in rainfall and evaporation impacting leaching, soil structure, and microbial community composition, can only be assessed in the field. However, the rate at which processes proceed in the field is dictated by the ambient climate and cannot be manipulated. Spatial heterogeneity demands intensive sampling which can constrain experimental design.
- *Soil or soil with plants*: Plants provide a sink for soil nutrients, exert suction on soil pores, and secrete compounds and enzymes that mobilize nutrients and modify soil surfaces. The microbial activity that concentrates around plant roots may "prime" processes that would not otherwise occur, for example, the co-mineralization of recalcitrant biochar and labile glucose [111]. However, the complex soil environment does not comprise such discrete components and plant-derived substrates are separated by the soil mineral matrix.
- *Empirical (descriptive) or mechanistic (predictive)*: Empirical studies identify statistical relationships between two or more test variables; mechanistic studies seek to understand the reason for such relationships. Mechanistic approaches should offer greater prospect for prediction of effects at other locations, being based on a fundamental understanding of the underlying process. Although technically more robust, mechanistic understanding may take time to acquire and still not be accurate; both approaches require considerable validation especially where multiple variables or processes are involved (Fig. 6.11).

Extrapolation from Studies of Environmental Charcoal

Given the basic similarity in formation and the relatively abundant literature, it is necessary to view biochar in the context of the existing understanding of charcoal, especially in terms of long-term stability. To date, however, studies that compare char produced under a range of conditions, e.g., varying levels of oxygen exclusion, in terms of agronomy, mineralization, or other parameters relevant to biochar deployment, are lacking. Until these studies have been completed, the congruence in the properties of these materials remains uncertain.

Does charcoal in soil constitute soil organic matter? In the discrimination of more and less recalcitrant forms of organic matter in soil generally, the ratio of oxygen to carbon broadly decreases with age with progressive removal of oxygen through biological or chemical "oxidation." Charcoal has a characteristically low O:C ratio, while graphitic black carbon (the most stable form derived in combustion) is essentially elemental carbon [112]. By comparison, charcoal and biochar are merely highly depleted in oxygen and hydrogen, containing groups that are strictly organic (most particularly aromatic forms), and part of the soil organic carbon pool.

Can charcoal be distinguished from other soil organic matter? Charcoal is particularly abundant in aromatic carbon that occupies a distinct position in the nuclear magnetic resonance spectrum for carbon (^{13}C NMR), displays a minor depletion in the abundance of the scarce carbon isotope (^{13}C) relative to other compounds, and is characterized by certain biomarkers (so far, benzene polycarboxylic acid and levoglucosan have been most extensively evaluated). Charcoal is partially resistant to some chemical oxidants typically used to quantify total soil carbon (potassium dichromate), and both chemical and photo-oxidation have therefore been used to quantify it. However, none of these signals have proven entirely exclusive, and the procedures for measurement are complex or slow. Because the functionally relevant level of stability is itself ill-defined or context specific, the analytical separation of charcoal and "ordinary" soil organic matter has yet to be perfected.

What is the historic significance of charcoal in the global carbon cycle? Models describing soil carbon in the agronomic or global change context consider a near-inert soil-carbon fraction to correctly simulate response to altered climate or organic matter inputs, which being site-specific generally reflect, at least in part, contrasting abundance of charcoal in regions where wildfire is more or less frequent. The rate, extent, and completeness of conversion of biomass to charcoal in wildfire is highly variable. Nonetheless, making assumptions about such factors based on available evidence leads to estimates for a mean residence exceeding 1,000 years [113]. Charcoal is thought to account for 1–20% of organic carbon in soils, and at least 150 GtC of the global soil pool comprises charcoal or its more condensed or graphitic relatives, soot and elemental black carbon [11]. This implies that up to 150 Mt of carbon has annually cycled through the biosphere in the formation and degradation of charcoal-derived carbon.

Are the impacts of biochar carbon analogous to those of soil organic matter? Some properties of biochar, and specifically its impacts on physical soil properties, are also associated with bulk soil organic matter. It is not, however, safe to assume that the mechanisms by which these are provided are identical, or that the impacts are equivalent in magnitude, direction, or duration – and thus that biochar can provide a direct substitute for higher levels of other organic matter in soil. This is of practical importance since biochar is carbonaceous and thus not readily distinguished from organic matter using current techniques. This is analogous to the challenge of discriminating chalk and limestone from organic carbon in soil.

Can impacts be predicted from ex situ properties? The ex situ characteristics of organic matter and biochar are unlikely to be additive with respect to a soil's properties. For example, certain soil minerals (clays) have a high cation exchange capacity; although organic matter has higher specific exchange capacity mass for mass, binding between organic and mineral fractions shields exchange sites at the molecular-scale and reduces the sum effect. At the moment, it is not completely clear whether interactions between char and mineral particles will occur significantly at this physical scale or primarily as discrete, disparate particles. Cation exchange capacity of biochar also appears to evolve over time, and it is likely that many other properties have a trajectory which is currently ill-defined.

Evidence to Address Key Questions around PBS

Biochar and Contaminants

In terms of human health and the food chain, the irreversibility of biochar addition is a key consideration. Existing soil amendments contain immobile components, albeit in less visible form and biochar inherits the potential risk posed by the feedstock that might otherwise be directly applied (see section "Properties of Biochar"). However, the class of these compounds known to be formed in the charring process itself (polycyclic aromatic hydrocarbons, PAHs) is process dependent. Without extensive evaluation of pyrolysis char, it is difficult to assess the risk posed by PAHs in PBS specifically as most data available relates to charcoal.

Charcoal is generally produced at lower temperatures that might favor PAH formation, but vapors may combust rather than condense and could thus be eliminated. Levels of extractable PAHs in charcoal are variable, but reported concentrations [114–117] generally fall between those reported for urban and rural soil on a mass basis (see Table 6.2). These compounds are persistent but ultimately degradable in soil [50].

The effect of association with chars on rate of degradation of PAHs, and the balance between rates of accumulation and release has not been systematically addressed. Concentrations of PAHs in soils subjected to natural fire suggest, however, that degradation is in excess of sorption. The capacity of both activated and nonactivated charcoal, typically as charcoal from or mimicking natural fire to adsorb PAHs and other organic contaminants, has been relatively well assessed [118, 119].

Since metals are broadly conserved in pyrolysis, the total metal content of biochar will be determined largely by the feedstock content and the yield of char. The higher the carbon content of the char, the lower will be the mass concentrations of metals. On a biochar mass basis, the metal concentrations in products from gasification where char yield is small (a few to 10%) are likely to be up to tenfold higher than in slow pyrolysis. Data on the availability of metals from charcoal or biochar in soil is lacking. However, the potential for pyrolytic char to remediate land contaminated by metal cations has been demonstrated [120].

Stability of Biochar Carbon

About 60% of the literature evaluated in [7] related to the stability of charcoal or to the quantification of char in soil (equally divided). Stability has been addressed both in real-time observation, where sensitivity can be enhanced through isotope tracers [121, 122], or extrapolation from measurements of soils from systems routinely experiencing natural fire [8, 106, 107]. In three cases, the effects of biological activity enhanced by substrate addition have been investigated [108, 111, 122]. Only in one case has pyrolysis char been evaluated [123] and, most often, the feedstock has been wood-derived.

Inference from measurements on soils in systems subject to natural fire suggests millennial stability, and in extrapolation from controlled incubations (elevated temperature and optimal moisture), the general acceleration of mineralization (decomposition) that occurs in such systems has been noted and accounted for [124].

Newly formed char appears to contain a small biologically labile fraction (see below), alkaline pH (mean pH = 8.5; ten studies), and nutrients available in ash from partial combustion embedded in the residual matrix. Only in one published study is this labile fraction deliberately extracted prior to evaluation [125]. Allowing for these characteristics that may lead to nonlinear carbon loss, other deficiencies in laboratory studies (simulation of natural char formation), and effects of induced changes in soil chemistry, the assumption of centennial to millennial stability does not appear unreasonable [122].

Experiments using newly formed charcoal have generally used particles <2 mm diameter (with no minimum particle size), but it has been noted that the physical fate of charcoal is predominantly in fine fractions, broadly <50 μm [90], and that its physical diminution appears relatively rapid, presumably through physical weathering and abrasion. There appears to be substantial evidence for intimate mineral–char interactions which, it has been hypothesized, might guard against degradation; however, discrete char particles have been found to persist within free organic fractions over a period of decades [126].

Three studies found no evidence for a role of tillage in the mineralization of wood-derived charcoal [122, 126, 127], and the single study that has explicitly examined the stability of pyrolytic char from wood and cereal straw suggested a slow and predominantly abiotic degradation which has been convincingly demonstrated for wood charcoal in a climosequence, which confirmed the sensitivity of absolute rates of degradation to temperature [33].

Oxidative measures are one of the key methods used to quantify char in soil, however they are used with the objective of retaining all charcoal in order to identify the wide continuum of char on soil (ranging from black carbon to soot), rather than identifying more or less stable subcomponents. However, the potential to develop artificial aging techniques (that simulate enzymatic oxidative degradation in soils) to rapidly compare and evaluate biochar stability (relative to charcoal) appears to have been rather overlooked and will be useful in seeking greater certainty on this critical matter [113, 112].

Labile Biochar Fractions

Incubation of soil with manufactured or un-aged char typically results in higher CO_2 evolution than from the same soil without char. However, the degree of excess declines over time in a strongly nonlinear fashion in the short term [121, 123, 128–130]. This suggests a "priming" of decomposition of carbon, either of that already in the soil, or more likely of carbon in the added char. Priming of existing soil carbon could be a consequence of the modification of the soil-chemical environment (see below), while loss of carbon from biochar can result from the mineralization of a labile char fraction. These patterns also suggest that priming is complex and that extrapolation of stability using short-term decay rates will be unreliable; such rates are not consistent with the age of charcoal found in archaeological soils such as the *terra preta*. The parameters that govern the balance between labile and stable components are not yet fully understood.

Priming of Soil Carbon or Biochar Loss

The potential for biochar, in the form of synthetic charcoal, to cause or accelerate the decomposition of preexisting soil organic matter (priming) has been reported in medium-term study of litter layers in the boreal zone [131]. The loss of litter carbon was measured over a period of 10 years, but almost all the loss occurred prior to the first annual sampling. The litter into which charcoal was introduced was likely to be acidic and nutrient constrained, in which case the decomposition response is expected.

In Canada, and with the benefit of a carbon isotope trace, information on priming in tilled arable soil was obtained on a 65-year timeframe. This indicated that the mean residence time for particulate organic matter in soil increased by a factor of 2.5 at sites where charcoal derived from historic natural fire was present [126]. Another study, also with a carbon isotope trace, suggested slower and less complete utilization (high stabilization) of organic material added to soils from a tropical environment containing aged charcoal [132].

A single laboratory study [111] showed an approximate doubling of charcoal degradation rates (charcoal priming) with the addition of glucose to soil, a compound often used as a simple analogue for the labile carbon exuded into soil by living plant roots. The initial rates of loss were still low (0.5% over 60 days, which is a smaller proportion of charcoal carbon that might reside in a labile charcoal component) – especially for higher temperature char created from wood and for an experiment conducted under optimal conditions in a sand matrix.

Quantitative extrapolation of such laboratory studies to the priming that might be likely to occur in the field, particularly with contrasting levels of microbial and plant root activity, is difficult.

It has been noted that if priming of soil organic matter is a permanent function of charcoal, the amount of non-charcoal carbon present under equilibrium field conditions must be lower than in charcoal-free soils [133]. Available data do not support this, and the Amazonian *terra preta* are enriched in organic matter relative to the surrounding soils, as well as containing large amounts of aged charcoal. Due to climatic influences, the Amazonian soils are rather low in organic matter naturally. The likelihood of an analogous accumulation in temperate soils amended with biochar is not certain, given higher background soil-carbon mineralization rates. Other studies have also suggested stabilization of soil organic carbon pools [134] and suppression of CO_2 production in biochar-amended soils [135].

Biochar and Soil Nutrient Dynamics

Reported increases in crop yield with charcoal addition have precipitated a number of plot-scale field trials to evaluate impacts on soil fertility, mainly through crop grain or biomass yield, usually with some measure of nutrient uptake. Reviewing 19 relevant articles in the literature [7], none of the reported studies have been undertaken in temperate zones. As such, caution should be adopted in directly transferring knowledge gained from tropical environments to temperate regions. In the tropical environment, the impacts have generally been positive, though most often in combination with fertilizer nitrogen. Less than one third of these studies have used char application rates of less than 15 tC ha^{-1}, however, and only three used pyrolytic char.

Verheijen et al. [2] undertook a meta-analysis of the effects of biochar addition to soil on crop production using nine studies (all of which used replicates to measure variance), involving 86 separate "treatments". The results are reproduced in Fig. 6.12.

The sample means indicate a small, but positive, effect on crop productivity with a grand mean of c. 10%. While there is some apparent trend of increased biochar additions, resulting in higher yields, this is not statistically significant at the $P = 0.05$ level as can be seen from the overlapping error bars at the 95% confidence interval. Biochar additions at rates of 10, 25, 50, and 100 t ha^{-1} led to statistically significant increases in crop yields compared to a control with no addition, though other studies using 40 and 65 t ha^{-1} did not show any statistically significant yield increase. Figure 6.12 illustrates that there is a wide variance in the response to biochar addition, e.g., at the 5.5, 11, and 135.2 t ha^{-1} application rates. Verheijen et al. speculate that the reasons for this are variability in the biochar, crop, and soil types. They also note that the means for each application rate are positive, and that no single biochar application rate had a statistically significant negative effect on crop productivity (though individual experiments have demonstrated negative effects, e.g., [84], [159]). On the other hand, the studies they examined do not cover a wide-range of latitudes and are heavily skewed toward (sub-) tropical conditions.

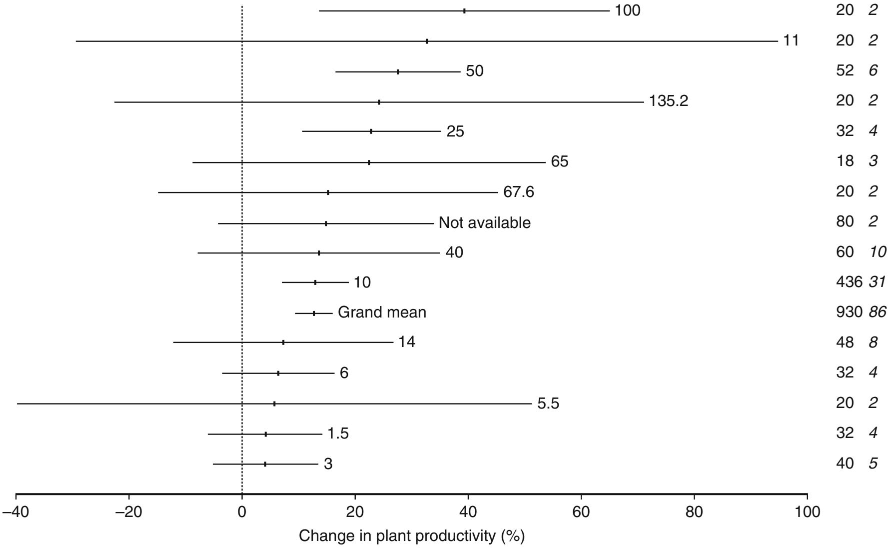


Fig. 6.12 The percentage change in crop productivity upon application of biochar at different rates from a range of feedstocks along with varying fertilizer co-amendments. *Points* represent mean, and *bars* represent 95% confidence intervals. Numbers next to bars denote biochar application rates (t ha^{-1}). Numbers in the two columns on the right show number of total "replicates" upon which the statistical analysis is based (*bold*) and the number of "experimental treatments" which have been grouped for each analysis (*italics*) (Reproduced with permission from Verheijen et al. [2])

Substituting chemical fertilizer for the nutrients added into the soil as biochar has not resulted in the same increase of crop productivity as provided by biochar addition. In two cases where the effect of pH modification was controlled for by liming, the effect of char was still superior [7]. This suggests that char might impact crop growth through its impacts on soil physical properties and/or on mediation of nutrient exchange between soil and plant.

In classic studies of *terra preta* fourth-season maize yields were much higher in plots amended with char and fertilizer than the non-fertilized and non-char amended control [136], and similar results have been shown in experiments with maize carried out in Columbian savannah soil [137]. However, regarding the *terra preta* studies, it is strictly incorrect to say that the effect of the treatments was to increase yield, since the yields for all treatments displayed post-clearance decline, and the control yields were ultimately very low.

Limited evidence under tropical soil conditions suggest that the addition of fresh charcoal can reduce nitrogen leaching loss. Soils with higher and long-established charcoal content had enhanced nutrient status but leached extra added nitrogen [1]. There is a relatively large amount of consistent evidence for the partial surface oxidation of char by chemical and biological processes in soil and proliferation of carboxyl groups [138]. It appears that this is reflected in the cation exchange capacity of aged charcoal and charcoal-rich soils, but not shown in new char.

Plot-scale experiments indicate that the uptake of other nutrients may be enhanced by charcoal, in particular phosphorus as ash in charcoal may be more available than phosphate in the soil.

Since biochar has a high carbon-to-nitrogen ratio, it is likely that rapid mineralization of a labile carbon fraction could – by immobilization – contribute to a draw on soil mineral nitrogen, in addition to an effect of ammonium sorption, and potentially reduce crop nitrogen supply. Evidence for this effect is relatively abundant and consistent in the literature, but the effect depends on the status of indigenous soil mineral nitrogen, and these studies have been undertaken exclusively in the tropics.

Immobilization tends to enhance soil nitrogen supply to the crop in the longer term since microbial proliferation builds a reservoir of mineralizable nitrogen. In field studies of charcoal or biochar extending beyond a single season, this effect may be observed, where second- or subsequent-season (but not first-season) are elevated relative to non-amended controls.

Biochar and Emission of Nitrous Oxide and Methane from Soil

A single peer-reviewed study reports suppression of nitrous oxide emission from soil from charcoal [125]; however, in the light of the importance of N_2O emissions to total agricultural greenhouse gas emissions and emerging evidence reported in recent studies (e.g., [13, 139, 140, 141]) the effect warrants further attention. Results from planted fields in Columbia showing a large positive effect are unpublished [142] as are three studies showing conflicting outcomes in laboratory studies using soils from Australia [143, 144] and New Zealand [139]. Laboratory studies used high rates of application [125, 135], single soils [145], or single types of charcoal. Results from a laboratory study using biochar made from poultry manure and *Eucalyptus* wood showed N_2O emissions were reduced over a long-term (5-months with three wet-dry cycles) period [140].

Nitrous oxide is emitted mainly by specific groups of bacteria, which under anaerobic conditions reduce nitrate rather than oxygen (nitrate to N_2O via nitrite and nitric oxide). Emission of N_2O at low rates may also occur under aerobic conditions from the activity of chemotrophic bacteria converting mineralized organic nitrogen (ammonium) to nitrate. Higher soil organic matter increases nitrification, but the application of nitrogen fertilizer has a greater immediate impact on soil nitrate concentrations and, hence, N_2O emission.

Proposed mechanisms for biochar suppression of N_2O revolve around modification of soil-water dynamics, e.g., drawing soil solution and dissolved nitrate into inaccessible pores (small pores saturating first) and maintaining aerobicity in inhabited soil-pore space; increase of soil pH which under anaerobic conditions favors completion of nitrate reduction to N_2 from N_2O; or the adsorption of ammonium and its protection from nitrification and denitrification [125, 145, 146]. Singh et al. [140] also propose the importance of time-induced changes (“aging”) on biochar properties which were attributed to the reduction in ammonium leaching and N_2O emissions they observed.

The effect of water addition cannot be completely evaluated under constant conditions, but Yanai et al. [125] found suppression was reversed when water-filled pore space was increased from partial to near-complete saturation. In the same study, the addition of combustion ash to the soil, separately from charcoal, did not suppress emission. However, [140] and [147] have demonstrated reduced N_2O emissions after a sequence of wet-dry cycles. Therefore, there is the important influence of time which must be taken into consideration. Simultaneous monitoring of N_2 is also required to confirm N_2O reduction. It has also been shown that nitrate has also been reported to accumulate where N_2O is suppressed [147].

Mobility of Char

Biochar and charcoal fines have a low bulk density of approximately 300 kg m^{-3} against a typical soil bulk density of 1,300 kg m^{-3}. Particles may be very fine in size and, in addition, surfaces may be hydrophobic [148]. Collectively, these characteristics indicate a higher potential for lateral transfer in water than for other soil components [149], and applied at a high rate in tropical environments subjected to frequent intense rainfall, erosion of charcoal off-site has been measured in proportions up to 25% in 2 years [150], and most of the 66% loss calculated by Nguyen et al. [106] in 30 years after surface deposition was attributed to erosion.

Negligible longer-term losses [106] and low rates of movement apparent for natural charcoal in a temperate environment [126] suggests a rapid decline in hydrophobicity, physical breakdown, and development of association with mineral particles [87, 151, 152]. The “anchoring” of particles within the soil matrix at depth may be critical in limiting erosion. The apparent combustion of fire-derived char in dry regions, where material remains at the surface between fires [153, 154], emphasizes the role of incorporation into deeper soil in ensuring longevity in the natural environment, and that in more biologically active soils, this must therefore occur. However, measured rates of transport into subsoil appear to be slow [150].

Meanwhile, studies of the global cycle of “black carbon” have established the existence of significant flux from land to ocean at a macroscale [102, 155]. Little literature has addressed the process of transport of char through the environment, although it has been noted that PAH is high in organic matter dissolved in alkali extracts after natural fire [156].

Char, Soil-Water Dynamics, and Irrigation

In large quantities, wood-derived charcoal modifies soil physical properties. It has a low-inherent bulk density of 0.3–0.5 t m^{-3}, which is one third to one fifth that of typical NW European arable soil. Depending on particle-size distribution of the char relative to that of the soil and the extent to which added char may locate within existing pores, higher experimental rates of application could directly reduce soil bulk density and increase soil volume. This affects water-holding capacity and water-filled pore space, but declining hydrophobicity and the effects of weathering on particle size will determine the duration of this effect. In the experimental context, water-holding capacity is measurably increased by adding fresh charcoal and must be considered in the design of laboratory soil incubations [132]. Studies of amended soils can be adjusted for either equal gravimetric water content, or to equal tension (depending on the hypothesis). Water storage could be of critical value, yet the factors that determine the efficacy of char in this context have not been clarified.

Published evidence [88] for the effect of biochar on pore-size distribution, however, is remarkably scarce. Some assessments have been made, and the problem appears to be in the level of replication required to demonstrate significant affects using methods best used in comparison of different soils. One study [157] has reported water-holding capacity of soils amended at low, medium, and high rates with pyrolytic char; one study has focused solely on pore-size characteristics of charcoal and pyrolytic char [88]; and one has measured the impact of charcoal residues on water-holding capacity at old kiln sites [83].

Summary

The evidence for the function of biochar in soil is based largely on evidence from studies of charcoal, and predominantly in the tropical environment. Triangulation of existing knowledge with systematic studies of biochar produced using technology and feedstocks relevant to viable temperate systems is needed, combined with techniques to rapidly assess long-term stability, and potential soil agronomic benefits.

Conclusion: Evaluating the Sustainability of Pyrolysis-Biochar Systems

A *sustainable biochar system* can be defined as one which: (a) produces and deploys biochar safely and without emitting non-CO_2 greenhouse gases; (b) reduces net radiative forcing; (c) does not increase inequality in access to and use of resources unduly, and (d) provides an adequate return on investment. Condition (a) is important to ensure that PBS technologies and practices do not pose undue risks to human health and safety and the environment (e.g., through inhalation of dust, biochar particles turning waterways or surrounding vegetation "black," adding N_2O, CH_4, or black carbon soot particles to atmosphere increasing net greenhouse forcing, etc.). Condition (b) is important to ensure that the net result of a PBS is indeed to reduce net radiative forcing relative to a baseline case. This is primarily due to reduction in atmospheric concentration of greenhouse gases through removal and avoided emissions. Condition (b) also takes account of direct and indirect land-use changes

(I/LUC), which can result in one-off emission of hundreds of tons of carbon per hectare in the case of tropical and peatland rainforest. Clearing of Brazilian wooded *cerrado* incurs a carbon loss of c. 45 tC ha^{-1}, US grassland c. 30 tC ha^{-1}, and abandoned US crop land zero or only a few tons per hectare [160]. Clearly, there is no point in converting land that incurs a large loss of carbon to biomass production for energy if the main purpose (or a large part of the rationale) of a project is to abate carbon through biochar production.

Condition (c) is relevant because an increase in demand for biomass will have knock-on impacts upon other users or potential users of that biomass, or upon other biomass, demand for which increases due to substitution effects. This also relates to LUC and ILUC, which frequently encounters equity and justice problems and questions. Condition (d) refers to economic viability since in market economies investment will only follow favorable rates of return. Defining an "adequate rate of return" is fraught with difficulties and depends upon subjective considerations such as the discount rate selected.

Systems which meet all of the above criteria do not exist at demonstration or commercial scale at the current time, and do not include traditional charcoal production. This is not surprising, of course, because biochar has only been proposed as a carbon abatement and agronomic improvement technology since the early- to mid-2000s. The further development and eventual deployment of biochar will be driven by one or more of the following policy and economic drivers.

(a) *Agronomic value of the biochar*: This value could be quite large on depleted soils, but any value has to be offset against the costs of production, delivery, and application to soil. Where the biochar is a by-product of electricity (or heat) generation, it may have a zero production cost. Examples of zero-cost biochar are few and far between and unlikely to be the norm. The cost of biochar production will, in many cases, be hard to recoup from the value of the agronomic gain alone.

(b) *Carbon storage value of the biochar*: If biochar can be ascribed a carbon storage value, this could greatly accelerate its successful take-off. The difficulty in ascribing a carbon value to biochar relates to the scientific uncertainty over long-term stability and interactions with soil organic carbon, lack of agreement on (and difficulty relating to) inclusion of land-based carbon budgets in trading arrangements, and technical uncertainties around Monitoring, Verification, Accounting, and Reporting (MVAR).

(c) *Waste management*: If pyrolysis-biochar systems are a cost-effective way of dealing with certain organic waste fractions (relative to other disposal/management options), this could drive the deployment of PBS through the value of the avoided tipping-fees to waste-management companies. The difficulty of this route is that waste biochar products are likely to pose more environmental and health and safety risks and will need to be carefully assessed and controlled. Such regulation is likely to drive-up the costs of biochar from wastes and to increase the regulatory barriers and hurdles, reducing interest from the industry. Furthermore, tipping fees are related to land-fill costs and taxation, which are subject to political change.

(d) *Renewable energy incentives*: To make pyrolysis-biochar add-up, it is likely that incentives will be required for the generation of renewable electricity and/or heat. Where the incentives are for electricity generation, however, other thermal conversion technologies such as combustion and gasification (with limited char production) will frequently have a competitive edge. Such incentives and subsidies are also vulnerable to political change.

(e) *Agri-environmental policies*: Another potential route to biochar deployment is through appropriate agri-environmental policies. If biochar addition can be demonstrated to have multiple benefits to agricultural land, agricultural support policies could be used to promote its adoption, for example, through inclusion in measures that are eligible for area-based and environmental stewardship payments.

One way forward in the short- to medium-term is to promote the use of biochar in particular niche applications, where its potential can be demonstrated and from there broader applications identified. This is broadly consistent with the innovation studies literature on socio-technical transitions: new technologies nearly always begin as niche applications that, if useful, find a wider role (3). The identity of the niche application will vary, depending on local, national, and regional context; policy; and socioeconomic drivers and incentives. In Europe, it may be sustainable biochar production from particular organic waste streams (paper or sewage sludge, for example), while in Asia, it may be carbonized rice husks from a gasifier as described in Box 2. In North America and Scandinavia, woody wastes from the timber industry may be ideal feedstocks, while in Africa, agricultural residues that would otherwise be burnt may offer-up opportunities.

Future Directions for Research, Development, and Demonstration

Biochar is a complex, multifunctional material that requires improved mechanistic knowledge and understanding – of its production, properties, impacts, interactions, costs, and benefits. Without this mechanistic process understanding, it is difficult if not impossible to predict and assess accurately the benefits of biochar for either greenhouse gas abatement or for addition to the soil even with evidence from individual field trials. We have identified the key research needs according to three broad headings below.

Pilot Production Research Facilities for Biochar and "Engineered" Biochar

What is the need? A strategic approach to producing, testing, and comparing biochar samples from different technologies, under specific reproducible conditions, would improve the evidence base. Facilities are needed to serve the research community, focusing current and future effort away from charcoal and toward biochar, produced from designated feedstocks under highly specified process conditions.

Why? The engineering and technological challenges are intimately related to the engineered biochar concept – namely engineering biochar with specific and controlled properties, designed for particular purposes (e.g., carbon abatement, short- and long-term agronomic gain, waste-management and pollution control, energy production, etc.) and contexts of application (soil types, agricultural systems, other land-uses, etc.).

When? The need is urgent if biochar is to have a role in tackling climate change in the next several decades and necessary for any future soil application of biochar.

Resource implications: The resources required are reasonably large, but not large compared to much technology-development since biochar can be produced at small-scales using relatively straightforward equipment.

How well advanced is existing research? There is currently limited activity or capacity anywhere in the world for technological evaluation of biochar production.

Ability to address key questions: Once equipment is in place, it should be relatively straightforward to address the key questions which arise, though some issues will inevitably take time to answer.

Examples of key questions to address:

Recipes for producing engineered biochar with specific properties and functions – acquiring the technological know-how to produce biochar with defined properties based upon a process understanding of production conditions.

Better understanding of the carbon and energy balance of alternative biochar production technologies – as yet there is little consistent and high-quality data on pilot- and commercial-scale pyrolysis (especially slow and intermediate) without which any evaluation of biochar is impaired.

Better understanding of the superlabile, labile, and stabilized components of biochar – better knowledge of what influences the Carbon Stability Factor for biochar is required, along with the effects in soil and field of labile versus stable carbon.

The Predictability and Certainty of the Impacts of Biochar

What is the need? If biochar is to be a commercial proposition, it will be necessary for reliable predictive knowledge of its impacts in particular soil and agronomic contexts to be well established (just as is the case for chemical fertilizers or pesticides). There is also need for a practical and scalable method by which the stable component of biochar can be established experimentally through acceleration of initial degradation as well as for examining the change that occurs soon after incorporation in soil.

Why? Only if the user is confident of positive and cost-effective benefits of biochar, when applied at particular rates, will a biochar market emerge. If the purpose is solely carbon storage, then the key issue will be long-term stability of the biochar.

When? For the purposes of carbon storage, urgent knowledge of long-term stability will be necessary. Predictive knowledge of soil and plant impacts is necessary for development of a market for biochar addition to land.

Resource implications: the resources required are moderate, but progress is being held back by lack of samples and equipment.

How well advanced is existing research? Evidence for the stability of charcoal in agriculture and the wider environment has been inferred in detailed studies in the USA and Australia. Work on the definition and stability of biochar is ongoing by established research groups. Work on soil and plant effects is well established internationally (especially in the USA, Australia, Brazil and Japan). However, this research is still in its infancy with no systematic effort: the opportunity exists for a focused and deterministic approach.

Ability to address key questions: In principle, it should be possible to address the stability question relatively quickly. Addressing the soil and plant effects will be more complex and time-consuming because of variability and complexity.

Examples of key questions to address:

These questions/topics can be addressed/answered fairly quickly.

Short- and long-term effects – separating out the long-term and short-term effects of biochar by comparing the functions of fresh and artificially aged material

Available nutrients and contaminants – Agreement on a methodology to quantify "available" nutrients and contaminants in a biochar matrix.

Deployment equipment and appliances – Develop modifications of existing agricultural equipments and implements so as to develop effective and efficient ways of storing and deploying biochar in realistic farm-based scenarios.

Biochar in grassland systems – The potential to reduce methane emissions from cattle, biochar as a slurry additive for odor control, etc.

Methodologies for evaluating the migration of biochar by movement through the soil profile, wind-erosion, water-erosion, etc. – necessary for regulation and carbon accounting.

Low-cost monitoring of biochar – Investigation of field spectroscopy and remote sensing for the low-cost monitoring of biochar added to soil.

These questions/topics are more difficult, complex, and/or simply time-consuming, and will take longer to address or answer

Nitrous oxide suppression – Examination of the mechanism by which biochar, at least under certain conditions, can suppress nitrous oxide emission from soil.

Field experimentation and trials strategy – Field experiments and trials that encompass diverse rotations and systems (arable, horticulture, and grassland) and including feedstocks derived from (inter alia) agricultural residues.

The value of biochar-based soil management – Compare the likely value of biochar-based soil management against the return that has been established for active use of other organic resources in management of soil.

Biochar for the control of diffuse pollution – More research on the ability of biochar to reduce leaching from land (e.g., using buffer ditch experiments) or from other sources (e.g., waterways, road surfaces).

Wider Biochar Sustainability Issues

What is the need? What are the wider impacts of biochar as a system, potentially deployed at different scales and in different spatiotemporal and socioeconomic contexts? How can the biochar system be made sustainable?

Why? Pyrolysis biochar systems (PBS), or variants thereof, only make sense if they meet minimum sustainability requirements (standards) and avoid incurring adverse environmental, social, or economic impacts. Sustainability appraisal methods can be utilized to ensure that biochar at a system level “adds-up”

When? It is important to understand system level impacts as these help direct more basic research and development by identifying key sensitivities. It is necessary to evaluate system-level effects prior to real-deployment to understand knock-on effects and potential problems.

Resource implications: The resources required are small compared to the technological and basic scientific research and development required because equipment and experimental costs are typically lower.

Ability to address key questions: Rapid progress can be made once resource is available, though accuracy and precision is dependent upon the availability of new data from technological and natural scientific R&D.

Examples of key questions to address:

Better Life-Cycle Assessments of pyrolysis-biochar systems – improved data across the whole PBS supply chain (from feedstock to field) and alternative biochar-producing systems.

Better techno-economic cost modeling – using more accurate data and with an improved representation of the key processes and stages, including production, distribution, storage, and deployment.

Better comparative analyses of biochar versus *other resource-use options* – need for explicit and transparent comparisons using best-available data of the most effective way of using and managing limited biomass resources for, e.g., bioenergy generation, carbon/greenhouse gas abatement, sustainable soils and waste management, composting and sustainable agri-food systems.

Assessment of land-use implications of biochar deployment – how biochar might influence the competitive advantage of different crops and the knock-on impacts on land-use decisions, supply, and demand.

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Chapter 7
Carbon Dioxide Sequestration, Weathering Approaches to

R.D. Schuiling

Glossary

CO_2 capture	Any process whereby CO_2 is sustainably removed from the atmosphere for a long period.
Dunite	Common rock type consisting for more than 90% of the mineral olivine.
Enhanced weathering	Any process whereby weathering reactions are sped up, like crushing the rock into fine particles and spreading these in suitable climates.
Laterite	Iron-rich tropical soil, which is the insoluble residue left after the weathering of the rock.
Mineral carbonation	The reaction of CO_2 with minerals (particularly Mg or Ca silicates), leading to the formation of solid and stable carbonates. This carbonation is preceded by the transformation of CO_2 gas to bicarbonate solutions.
Olivine	A silicate which is a mixed crystal of Mg_2SiO_4 and Fe_2SiO_4. It is the fastest weathering common silicate.
Weathering	The process whereby rocks are decomposed by reaction with water and acid (usually carbonic acid).

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Definition of the Subject

The aim of enhanced weathering is to capture CO_2 by the carbonation of silicates, or by dissolution of these silicates during which the greenhouse gas CO_2 is converted to bicarbonate in solution. Research in this field is still focused on increasing the rate of reaction, but the required additional technologies add considerably to the cost of the process. In this entry, the focus is on the optimization of the weathering conditions, by selecting the most reactive abundantly available minerals, grinding them, and spreading the grains over land. Thereafter nature takes its course. Since its formulation in the late 1990s, more and more people realize that this simple and natural approach may well turn out to be one of the most promising and environmentally friendliest ways to counteract climate change and ocean acidification.

Introduction

CO_2 is a greenhouse gas. The rising concentration of CO_2 in the atmosphere by the burning of fossil fuels is considered by many to be the main cause of climate change. Most efforts on carbon sequestration so far have focused on the physical removal of CO_2, by locking it up as a supercritical fluid in available spaces in the subsoil, like abandoned oil and gas fields or aquifers. Sequestration by mineral carbonation* has long been neglected, as it was deemed to be too slow or too costly. The main arguments for a slow rate of reaction stem from an extrapolation of abiotic experiments in the laboratory. Observations from the “real world” only now begin to show that weathering rates are often 1–2 orders of magnitude faster than rates determined in the laboratory. Arguments based on the apparent high-cost aspect are due to the fact that almost all researchers assume that mineral carbonation must be coupled to a technology to speed up the reaction. This may involve thermal treatment of the mineral, a chemical or mechanical pretreatment or a technology based on subjecting the mineral to high pressures and temperatures in autoclaves. Most of these approaches are successful to some extent, but their high cost makes such mineral carbonation technologies unattractive [1], and some produce large amounts of CO_2. If there is no objection to the reaction taking a few months or a few years to run to completion, then there is no need for the use of expensive equipment, which must meet minimum throughput rates for financial reasons. Given these conditions, chemical weathering in favorable environments becomes an attractive proposition. It can be applied on a world scale, it is a low-cost operation per ton of CO_2 captured, it is sustainable, it provides employment opportunities in developing countries, and unpleasant environmental surprises are unlikely, as the same process has already operated over the entire geological history.

This entry, therefore, will focus on the enhancement of the process of chemical weathering, by which a mineral is converted to a bicarbonate solution. These bicarbonate solutions are transported by rivers to the oceans, where they will be captured as carbonate rocks, which are the ultimate sink for CO_2. During the precipitation of solid carbonates, half of the CO_2 is released again, but this process takes on average several hundreds to thousands of years, so the bicarbonate step is the relevant step for climate change mitigation.

Materials

Most materials to be used for carbonation are naturally occurring rocks or minerals, although residues from industrial processes are also considered. Industrial residues have the advantage of being readily available and sometimes need to be removed or treated anyhow. However, their total volume is too small to make more than a small dent in the CO_2 problem, but they may provide an easily accessible and cost-effective part of the solution. Some of the materials in this category are (coal) fuel ashes, oil shale ashes, metallurgical slags, including slags from the production of elementary phosphorus, red mud from the alumina industry, and wastes from the demolition of concrete buildings [2, 3]. It was shown that wastes from the demolition of concrete are rapidly carbonated in soils, and their carbonation compensates for decarbonation during manufacture.

Among the naturally occurring materials for carbonation, olivine and olivine rocks, called dunites, take a prominent place because olivine weathers fast and is abundantly available (see Fig. 7.1). Other candidate materials include basalts and basaltic tuffs, and possibly anorthosites. A mineral that is also frequently mentioned is wollastonite ($CaSiO_3$). Wollastonite does react quickly with CO_2 and water, but wollastonite deposits are rare and small. It is evident that priority will be given to material that is already crushed or milled, as this saves mining and milling costs. Many ore deposits of chromite, nickel, magnesite, peridot (a semiprecious variety of olivine), or even diamond have olivine-rich host rocks. Billions of tons of such rocks can be found in crushed form as mine tailings. Later on, in the section on rate of weathering, some spectacular evidence on the rate of weathering of powdered olivine or serpentine (the hydration product of olivine) on mine dumps will be presented.

Modes of Application

a. In dedicated industrial installations
b. In situ injection in suitable rock types
c. Ex situ after crushing and spreading suitable rock types

Fig. 7.1 Distribution of dunites in the world. *One dot* in a country may represent several dunites

a. Industrial installations. These require an input of fairly pure CO_2 to reach a maximum efficiency. This means that they can be dependent on point sources of pure CO_2, or on more dilute sources of CO_2 from which the CO_2 must first be captured and purified. Industrial treatments, involving thermal or chemical activations, or large autoclaves through which the mineral powders must pass as fast as possible have significant energy requirements. It is doubtful that large-scale application of thermal or chemical treatment of olivine, for instance, can be carried out at the required scale of 25 billion tons of olivine annually, let alone in a cost-effective way.

b. In situ injection in suitable rock types. The major distinction between this type of injection and the most commonly proposed CCS (Carbon Capture and Storage) is that injection in flood basalts or olivine rocks aims at converting the injected CO_2 gas into carbonate rock by reaction with the host rock. This reaction produces some heat, which may further speed up the reaction. In oil and gas fields, or in saline aquifers, the CO_2 remains in the reservoir as a gas (or more correctly as a supercritical fluid), with the inherent risk of leakage, or even explosive escape. The main conditions for a reservoir rock in which the CO_2 will be injected and converted to solid carbonates are reactivity and, what can be called, "selective" permeability. Thick sections composed of many superimposed volcanic flows with a horizontal extension of sometimes more than 1 million km^2, so-called flood basalts, may be an option. A number of features of lava flows, including flows that are separated from the overlying flow by a rubble zone, which can have high permeability, often finding a high concentration of bubbles near the top of the flow where these bubbles contribute to the accessibility of the rock for rock/fluid interaction; and some lava flows vertical structure with more or less hexagonal columnar jointing that originates from shrinkage during cooling, which permits easy access for fluids throughout the basalt flow. The combination of these properties makes piles of basalt flows almost ideal targets for injection, although their reactivity is considerably lower than that of olivine-rich rock types. Flood basalts can occupy enormous volumes, in several cases in excess of 1 million km^3. Well-known examples are the Deccan traps in India, the Siberian traps, the Karroo basalts, and the flood basalts of the Parana basin, each covering more than 1 million km^2. The Columbia River flood basalts and the Deccan traps are presently under investigation on how they can be used for carbon capture [4]. Logistics of CO_2 transport and its associated cost, as well as the rate at which it can be injected, and the reactivity of the rock will probably limit their use as a CO_2 trap to locations close to voluminous point sources of CO_2. They potentially constitute a safer storage than aquifers or abandoned oil and gas fields because after reaction with the rock, the CO_2 is no longer stored as a gas but as a solid carbonate.

The situation with direct injection into olivine-rich rocks is more or less the reverse of injection into basalt piles. Permeability is often much smaller, as most dunites (dunite is the rock type that consists for more than 90% of olivine) are massive rocks. They may contain joints, but these are often sealed with a thin veneer of serpentine or magnesite ($MgCO_3$). On the other hand, their reactivity is

Fig. 7.2 Multiple generations of carbonate cement around dunite pieces in alluvial terraces in Oman [5]

considerably better. Spectacular examples (Fig. 7.2) of carbonation of olivine-rich rocks in Oman were presented by [5]. These authors propose that annually up to one billion tons of CO_2 can be injected into these rocks in Oman. Transport of such volumes of CO_2 from industries and coal-fired power plants in the industrialized world, as well as purifying it and bringing it to a remote site, poses, however, a huge logistical problem. Even if the logistical problems are not insurmountable in terms of cost, there remains the problem that the rocks must be intensely fracced for injection. Kelemen and Matter suggest that the considerable heat of reaction from the hydration [5] and carbonation [6] may help to crack the rocks further. Although from the reactivity point of view direct injection of CO_2 + water into dunites is an attractive proposition, the likely high costs may limit its use to locations where a coal-fired power plant is situated at short distance from a dunite massif like near Orhaneli/Turkey [7], or where, like in Oman, the CO_2 produced by the installations on oil and gas fields can be injected directly into the dunites nearby.

c. Ex situ after crushing and spreading suitable rock types. This option is better known as enhanced weathering [8]. This is probably the most promising solution for the sustainable capture of CO_2 on the scale of billions of tons of CO_2 annually. It is only for this option that a more complete description of the proposed process will be given, as well as some quantitative estimates of its costs and energy requirements. Furthermore, the possible collateral benefits of the method will be considered.

Weathering

It is estimated that the Earth produces around 0.5 billion tons of CO_2 each year, mainly from volcanic sources [9]. Other important sources of CO_2 are carbonate rocks that decompose after having been transported to great depth and subjected to high temperatures in subduction zones [10], or limestones and dolomites in the contact aureole of intrusive rocks, or marbles or siliceous dolomites undergoing high-grade regional metamorphism. If there were no feedback mechanism or mechanisms by which this CO_2 is constantly removed from the atmosphere and sustainably stored in rocks, the Earth would now have an atmosphere similar to Venus, where the CO_2 pressure of the atmosphere is 80 bars, and the surface temperature of the planet is 450°C, due to the excessive greenhouse effect. The major feedback mechanism is the weathering of silicate minerals, in particular Ca and Mg silicates, which is only possible thanks to the presence of liquid water on Earth, whereas it is absent on Venus. A smaller feedback mechanism is the storage of CO_2 as organic carbon (oil and natural gas, coal, and organic carbon dispersed in sediments). Chemical weathering can be described as the neutralization of an acid by rocks. H_2CO_3 is the dominant acid during weathering. It is converted to bicarbonate in solution. The rock is partly dissolved. A residue of clay minerals can form as the solid end product. Weathering has been a life-saving process for our planet. Not only has the interaction of CO_2 and water with rocks saved us from a greenhouse problem as found on Venus, but it has provided us also with fertile soils, without which it would have been impossible for any evolved life forms to develop, and it has released the mineral nutrients from the rocks which are a basic requirement for plant life.

Some typical weathering reactions are as follows, weathering of olivine:

$$Mg_2SiO_4 + 4CO_2 + 4H_2O \rightarrow 2Mg^{2+} + 4HCO_3^{-} + H_4SiO_4 \tag{7.1}$$

and weathering of anorthite:

$$CaSi_2Al_2O_8 + CO_2 + 2H_2O \rightarrow CaCO_3 + Al_2Si_2O_5(OH)_4 \tag{7.2}$$

The first reaction, the weathering of olivine, describes an intermediate step in the weathering process. After this dissolution step, the Mg-bicarbonate solution is transported to the sea, where ultimately it will precipitate in the form of carbonate sediments (limestones and dolomites). That process, however, takes on average hundreds, if not thousands of years, so the first step, the formation of Mg- or Ca-bicarbonate waters is the relevant step for CO_2 capture on a time scale of a few tens of years. The second reaction (Eq. 7.2) demonstrates the formation of a carbonate and a clay mineral by the weathering of anhydrous silicates.

Many nongeologists think that the CO_2 that is dissolved in the oceans, present as CO_2 in the air, or contained in biomass plays the major role in the CO_2 balance of the Earth. In fact, these reservoirs are only short-lived and minor transitory storage rooms. The major part of the geochemical CO_2 cycle is represented by emission of CO_2 from the Earth, its transformation by weathering into a bicarbonate solution, followed by its transport to the oceans and deposition as carbonate rocks. These carbonate sediments form the ultimate sink on a human time scale. Geologically speaking, even these solid carbonate sinks are not eternal, but are recycled as well,

Table 7.1 Distribution of carbon on Earth. Modified after [11]

	Amount of carbon ($\times 10^{15}$ kg)	Relative amount (%)
Limestone ($CaCO_3$)	35,000	46.6
Dolomite	25,000	33.3
Sedimentary carbon	15,000	20
Recoverable fossil fuels	4	0.005
Oceanic CO_2	42	0.056
Atmospheric CO_2	3	0.004
Biomass	0.056	0.0007
Anthropogenic emission	0.03/year	
Input from Earth's interior	0.002/year	

albeit with time scales of hundreds of millions of years. The relations can be demonstrated with the following table (Table 7.1) [11].

The process of chemical weathering has always provided the main mechanism to keep the CO_2 levels of the atmosphere within reasonable bounds. Of course, there have been fluctuations in the course of geological history because the available surface area where reactive rocks were exposed to the atmosphere varied; during or shortly after major orogenic periods, or after large-scale volcanic activity, the area of fresh rocks available to weathering was larger than in periods where the landscape was more mature, and where many easily weatherable rocks were covered by a thick weathering crust, which virtually stopped their interaction with CO_2 and water. At the same time, it is likely that the annual emissions of CO_2 from the Earth were also subject to large variations caused by the intensity of volcanism, related to the rate of plate spreading.

To a certain extent, such variations are counteracted by the CO_2 concentration of the atmosphere itself, which acts as its own negative feedback. The higher the CO_2 pressure, the more acidic the water, and the faster the weathering which removes that same CO_2 from the atmosphere.

There is geological evidence that factors like mountain building or massive volcanism, through their effects on rates of weathering and emission of CO_2, have influenced the CO_2 pressures of the atmosphere in the past and may have led to periods of glaciation [12–14]. Since the industrial revolution, atmospheric CO_2 levels have risen rapidly (from 260 to 392 ppm), largely due to mankind burning in a few 100 years the fossil fuels that took hundreds of millions of years to form. If weathering is going to be used as a tool to counteract this rise in CO_2, caused by mankind annually emitting more than ten times as much CO_2 than the Earth normally emits, then the weathering process must also be made more than ten times more effective than normal to reach a new balance. The process as discussed here is to select widely available rock types that weather easily, mine them and increase their reactive surface area by crushing them, and spread the crushed rocks over land or along beaches in areas with the most suitable climate for weathering.

In subsequent sections, only olivine and olivine rocks will be discussed. Other rock types can also be used, but they are less effective or less available, although locally, there may be good reasons to use other rock types, such as basalts, anorthosites, or nepheline syenites. In the following weathering reactions, the formula of olivine will be simplified to Mg_2SiO_4, although olivine normally is a mixed crystal of Mg_2SiO_4 and Fe_2SiO_4, with the Mg-endmember usually dominant. One finds a number of ways in which the weathering reaction of olivine is described. As this affects the amount of CO_2 that can be captured for a given amount of olivine, these different weathering reactions will be briefly discussed, and the major reaction that takes place in nature will be identified. A common way is the reaction in which during weathering two new solid minerals are formed, namely, magnesite ($MgCO_3$) and serpentine ($Mg_3Si_2O_5(OH)_4$).

Weathering Reactions of Olivine

$$2Mg_2SiO_4 + CO_2 + 2H_2O \rightarrow MgCO_3 + Mg_3Si_2O_5(OH)_4 \tag{7.3}$$

In this reaction, 2 olivine moles capture 1 CO_2 mole.

A second reaction is

$$Mg_2SiO_4 + 2CO_2 + xH_2O \rightarrow 2MgCO_3 + SiO_2\ xH_2O \tag{7.4}$$

In this reaction, 1 olivine mole captures 2 CO_2 moles.

The reaction that occurs normally in places with sufficient rainfall

$$Mg_2SiO_4 + 4CO_2 + 4H_2O \rightarrow Mg^{2+} + 4HCO_3{}^- + H_4SiO_4 \tag{7.5}$$

In seawater, it is convenient to split the reaction into

$$Mg_2SiO_4 + 4H_2O \rightarrow 2Mg_2{}^+ + 4OH^- + H_4SiO_4 \tag{7.6a}$$

followed by

$$4OH^- + 4CO_2 \rightarrow 4HCO_3{}^- \tag{7.6b}$$

In reactions (7.5) and (7.6a and 7.6b), each olivine mole captures 4 moles of CO_2. Evidence will be presented that reaction (7.5) is the common weathering reaction. It has been mentioned already before that the resulting Mg-bicarbonate waters will ultimately form carbonate rocks in the oceans. During the precipitation of those carbonates, half of the captured CO_2 returns to the atmosphere, but as this process may take thousands of years, we will neglect it as far as it affects climate change in the short run.

In order to find out what is the normal reaction mechanism for olivine weathering, a suite of springwaters issuing from olivine rocks in Turkey were

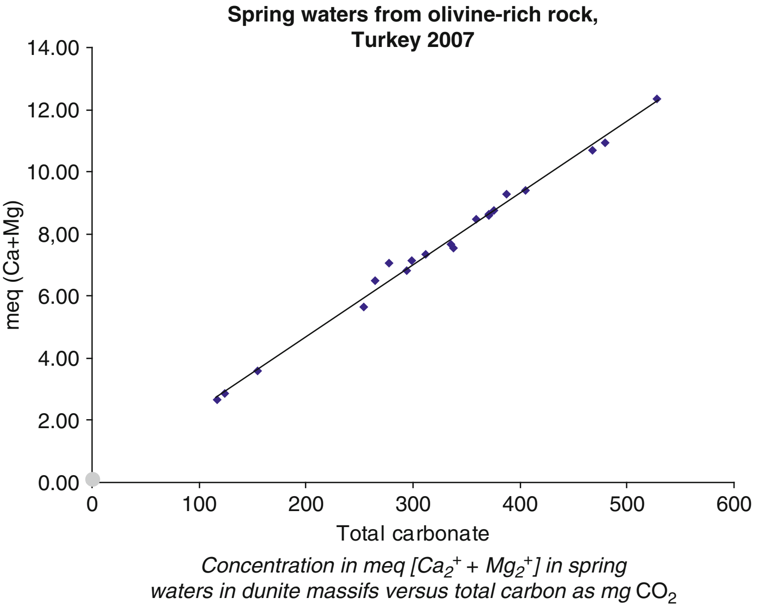

Fig. 7.3 Relationship between calcium + magnesium extracted from the rock and amount of CO_2 captured as bicarbonate in springwaters issuing from olivine rocks in Turkey

collected [15]. One can distinguish the following steps during the formation of such springwaters:

1. Rainwater falls on the ground.
2. It infiltrates the soil. The CO_2 concentration in soil atmospheres is usually around a 100 times larger than in the open air, because litter in the soil decays and soil fauna respires, both contributing to a high concentration of CO_2 in the soil [16, 17].
3. After equilibration with this CO_2-rich soil atmosphere, the water infiltrates the underlying rock and reacts with it (the weathering step).
4. Finally, the water emerges again as a spring at some lower point.

Olivine-rich rocks contain generally, next to the dominant mineral olivine, some other silicates, including calcium silicates. From Fig. 7.3, it is evident that there is a close relation between the (Mg + Ca) content of the water, expressed in milliequivalents, and the amount of CO_2 that has been converted to bicarbonate. This close relation has been confirmed in a number of cases all over the world, and it can be concluded that the reaction leading to bicarbonate in the solution is the major pathway in which CO_2 is removed from the air, taken up as bicarbonate in the water and transported to the oceans.

Even when the problem is considered on the scale of the whole Earth, the picture remains essentially the same. All rivers together transport annually 3.6×10^{16} kg of water to the oceans. The weighted average of their concentration of Ca is 11 ppm, of

Mg 2.6 ppm, and of HCO_3 42.4 ppm. Expressed again in milliequivalents, this means that Ca + Mg make up 0.77 meq and bicarbonate 0.7 meq, so, despite all complications, there is still a satisfactory agreement. If we calculate from these figures the amount of CO_2 that is annually brought to the sea by rivers, this comes out as 1.1 billion tons of CO_2, the same order of magnitude as the annual emission of CO_2 by the Earth, which is obviously larger, as emissions of CO_2 from the seafloor are only partly taken into account. Some of this CO_2 returns to land, takes part in weathering reactions, and returns as bicarbonate to the oceans.

Enhanced Weathering: What Does It Involve?

Having established that the principal weathering reaction is reaction (7.5), the formation of magnesium bicarbonate solutions, it can now be calculated from the stoichiometry of this reaction how much olivine is required each year to sequester all the emitted CO_2 by enhanced weathering, under the assumption that energy savings, switching to green energy and change of lifestyle, will not lead fast enough to a lowering of the world's CO_2 emissions, and other technologies for CO_2 capture will remain negligible. In this scenario, the required volume turns out to be approximately 7 km^3 of olivine rock. This is, of course, large because it must match the carbon contained in all the oil, gas, and coal that is burnt. It is, however, within the range of modern large-scale mining. This volume of olivine is equivalent to 10 km^3 of oil. It is worthwhile to come to grips with the scale of this mitigation effort (Fig. 7.4). The largest mine in the world, the copper mine at Bingham, Utah, USA, has an excavated volume of 25 km^3. Olivine mining at this scale, however, means that olivine moves up from a modest commodity to third place in the mining industry, after construction materials and coal. One should, obviously, not open only one single, huge olivine mine but spread the mining operations over 30–50 open-pit olivine mines. In order to profit from the most suitable climatic conditions for weathering, these mines should be strategically spread along the tropical zone. A larger number of olivine mines will be able to service wider areas without requiring large transport. Each of the selected sites for olivine mining must produce in the order of 500 million tons of olivine each year to reach the required 7 km^3 per year. It may well turn out that it is more favorable both economically and environmentally to increase the number of olivine mines and decrease their size. At the same time, this will spread the economical benefits of olivine mining over a larger number of countries and further limit the required transport distances. This volume of olivine rock must be crushed and milled to grains of around 100 μm in diameter. If 7 km^3 is spread over an area of 10 million km^2, it will occupy a layer of 0.7-mm thickness. In the section on rate of weathering, it will be shown that grains of olivine of 100 μm will weather in approximately 5 years in tropical soils. It will, therefore, be cheaper to spread a layer of 3.5-mm thickness each year over an area of 2 million km^2, shift to the next area in the following year, and come back to the first after 5 years.

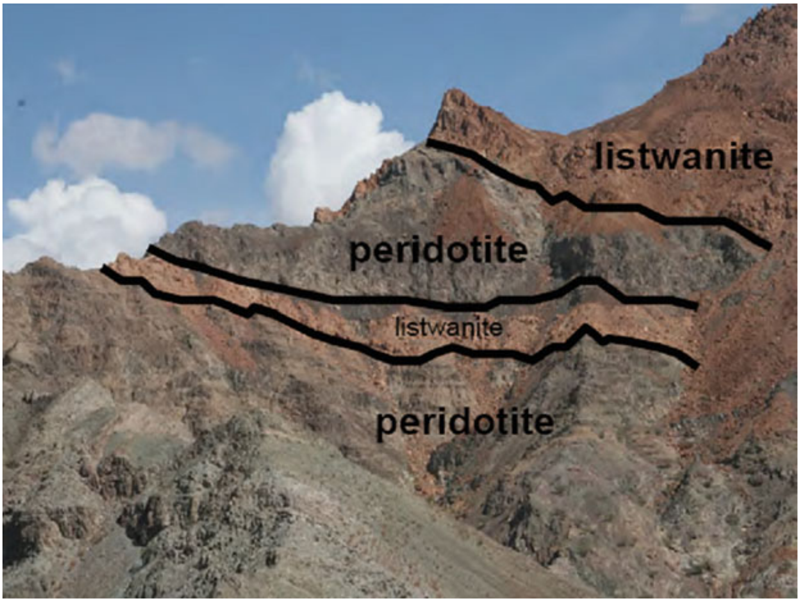

Fig. 7.4 It is a sobering thought that these huge bands of listwanite (completely carbonated dunites consisting of magnesite + quartz) in Oman, containing 500 million tons of CO_2, are equivalent to only 1 week of the world's anthropogenic CO_2 emissions [4]

In a few instances outside the tropical zone, conditions may be so favorable that large olivine exploitations can be located there. For example, a possible site for an olivine mine outside the tropical zone might be Oman, which has the largest ophiolite zone in the world and a long coastline along the Indian Ocean. Deeply weathered sections of dunite, dating from earlier more humid periods, can be found, but in its present desert climate, the olivine does not weather, but forms huge talus deposits on the slopes of the mountains, at a short distance from the Indian Ocean. These scree deposits have no overburden which must first be removed, like in most tropical dunite deposits. In Oman, the loose pieces, after some additional crushing, can be loaded into ships that have direct access to the Indian Ocean.

Rate of Weathering of Olivine

Much of the debate on the potential of enhanced weathering using olivine grains to counteract climate change centers on the problem of the rate of weathering. The rate of weathering is enhanced in hot humid conditions, hence why this idea is focused on the application of crushed olivine in the wet tropical regions.

There are a number of reasons to focus on the wet tropics for large-scale applications:

- Weathering is faster under hot, humid, climatic conditions.
- Tropical soils are usually very poor and can benefit from the addition of mineral nutrients.

This does not mean that the olivine option is useless in temperate climates, it is just that the rate of weathering will be slower.

In abiotic laboratory experiments, it was found that the surface of olivine grains retreats at a few tenths of a micron per year [18]. This is described by the shrinking-sphere concept. Such low rates would make it difficult to use enhanced weathering to mitigate the greenhouse effect. Fortunately, there is observational evidence on rates of weathering of olivine in the real world (see below), which shows that the rates are more than tenfold, and probably 100-fold larger, than those found in the laboratory. Qualitative information on fast rates of weathering is obtained from volcanic terrains with rocks containing olivine. When volcanism started in the Eifel/Germany, synchronous Rhine sediments downstream in the Netherlands immediately started to contain a wealth of volcanic minerals, but *no olivine*, despite the fact that these volcanic rocks contain plenty of that mineral. Contrary to the other minerals of volcanic origin, olivine has not survived the short trip from Bonn to the Dutch border. Similar observations are reported from many other volcanic terrains in the world. Although suggestive of fast weathering, this evidence is difficult to quantify.

In contrast, the rate of weathering of dunite massifs in the tropical zone can be quantified, or at least a minimum rate of weathering can be firmly established. The first example is the dunite massif of Conakry/Guinea. This dunite occupies the entire peninsula on which Conakry, the capital of Guinea, is situated. It has an approximate length of 50 km and an average width of 5 km. Over its entire surface, it is covered by a thick lateritic weathering crust, which is very clearly visible as a purplish red area on satellite pictures (see Fig. 7.5). This lateritic crust, which is the iron-rich insoluble red residue of the dunite after deep tropical weathering, contains virtually no silica, magnesium or calcium oxides which were completely leached out during the weathering process [19]. These components make up around 90% of the original dunite. This means that 1 m of laterite is equivalent to 10 m of dunite, or even more if the remaining components of the laterite were not completely immobile but have also been leached to some extent. The same author presents evidence that iron has in fact been fairly mobile and was partially leached out as well, which means that 1 m of laterite is equivalent to more than 10 m of dunite. The weathering crust has a thickness between 30 and 100 m. The age of the dunite (that is to say the time at which this dunite intrusion formed) has been determined as 195 million years. From these data, it is simple to calculate the minimum rate of weathering as follows: 50 m of laterite is equivalent to 500 m of dunite, 500 m (= 500 million microns) divided by 195 million years is 2.6 μm/year. This is already ten times faster than deduced from laboratory experiments, but the real rate of weathering must have been considerably faster. The rock is an *intrusion*. That means it was emplaced between rocks at some depth and covered by other rocks, which had to be removed first by erosion before the dunite became exposed

Fig. 7.5 The dunite massif of Conakry, Guinea. The length of the dunite massif (in *purplish red*) is approximately 50 km

and could start its weathering process. If the dunite intrusion has taken place at 2 km depth, it would take 100 million years before the dunite massif was entirely laid bare by erosion at an estimated erosion rate of the order of 1–2 cm/1,000 years [20]. This is the average erosion rate for all continents. This correction alone more than doubles the calculated rate of weathering. That is not the only positive correction that must be made. In more recent times, the weathering process, under such a thick weathering crust, has virtually come to a standstill, as the thick laterite crust effectively shields the underlying rock from further interaction with the atmosphere. This shortens again the time span over which weathering was active, and thereby increases the rate of weathering.

A further positive correction concerns the difference between weathering of a solid rock as opposed to loose grains. A rock is attacked by weathering from above along a two-dimensional front, whereas loose olivine grains in soil are attacked from all sides. It seems certain that olivine grains in tropical soils dissolve at least at a rate of 10 μm/year, but most likely even faster. Even when their surface retreats only by 10 μm/year, a grain of 100 μm will disappear in 5 years. A similar calculation can be made for the dunite body at Jacupiranga, Brazil [21]. Here, the rock has an age of 130 million years, and it is covered by a weathering crust of >40 m (this is where the drill hole stopped, but at 40 m, the drill was still in lateritic weathering crust). The minimum rate of weathering turns out to be >3.1 μm/year, but the same positive corrections have to be applied as in the Conakry case.

Fig. 7.6 Efflorescence of nesquehonite on the serpentinite tailing heap at Clinton Creek (Photograph by S.A. Wilson)

From a global balance of weathering and erosion, similar minimum rates of weathering emerge. The average rate of erosion of the continents is 1–2 cm in a 1,000 years [20]. As olivine grains from the interior of the continents do not make it to the oceans, this means that olivine rocks dissolve (= weather) at least at the same speed, which is 10–20 μm/year. The most dramatic evidence for fast weathering of crushed magnesium silicate rocks comes from observations of weathering rates of mine dumps of such rocks [22]. By measuring the amount of a suite of newly formed Mg carbonates, it was shown that the mine tailings of two abandoned asbestos mines in British Columbia weather extremely fast. In this case, it does not involve fresh olivine, but its hydration product serpentine ($Mg_3Si_2O_5(OH)_4$) that weathers and produces carbonates. This carbonation proceeds as follows:

$$Mg_3Si_2O_5(OH)_4 + 3CO_2 + 2H_2O \rightarrow 3MgCO_3 + 2H_4SiO_4 \qquad (7.7)$$

At low temperatures, magnesite seldom forms, but in its place, hydrated magnesium carbonates, like nesquehonite $Mg(HCO_3)(OH)\cdot 2H_2O$, are found instead (Fig. 7.6).

In order to make sure that these carbonates have indeed newly formed, ^{14}C analyses were performed on these carbonates which gave an age of about 0, showing that the carbon in these minerals really represents the sequestration of present-day atmospheric carbon [23]. In one of the cases, the mine dump, occupying a surface area of 0.5 km^2, had captured 82,000 t of CO_2 between 1978 and 2004, more than 50 times the maximum ever recorded for natural weathering under the most favorable conditions. The real rate of weathering is even higher because the authors have only taken the solid products into account, whereas the waters that percolate through the mine dumps carry an additional load of dissolved weathering products. These waters become quite alkaline, and their high silica

Fig. 7.7 The olivine reactor (artist's impression)

content leads to small diatom blooms in a pool at the foot of the tailings dump and in at least one of the mine pits [23–25].

One may wonder why there is such a large discrepancy between laboratory experiments, showing low rates of weathering, and the real world, where weathering rates are 100 times larger. The answer is relatively simple. Higher plants live in symbiosis with mycorrhizal fungi in and around their root system. These fungi secrete low molecular organic acids like acetic acid, malic acid and oxalic acid that rapidly attack mineral grains in the soil [26]. This liberates mineral nutrients that are subsequently taken up by the higher plants. In turn, the higher plants "reward" the fungi by providing them sugars. Lichens act in a similar way by secreting oxalic acid that "eats" the underlying rock [27]. In the laboratory, mycorrhizal fungi and lichens are absent, and this is the reason why the abiotic reaction rates that were found in the laboratory are much lower than weathering rates in nature.

This all refers to weathering rates of olivine on land. Much less information is available for olivine on beaches, in the surf zone, shallow seas, and tidal flats. To understand what happens to olivine upon weathering we must distinguish between the chemical reaction of olivine with seawater and mechanical impacts during grain transport. To measure quantitatively the chemical weathering rate of olivine in seawater, one could use a so-called olivine reactor (see Fig. 7.7), a concrete box 50 m long and 4 m wide, constructed perpendicular to the coast and filled with olivine sand. To obtain even more information from this experiment, the reactor can be partitioned lengthwise, and the two compartments can be filled with olivine sand of

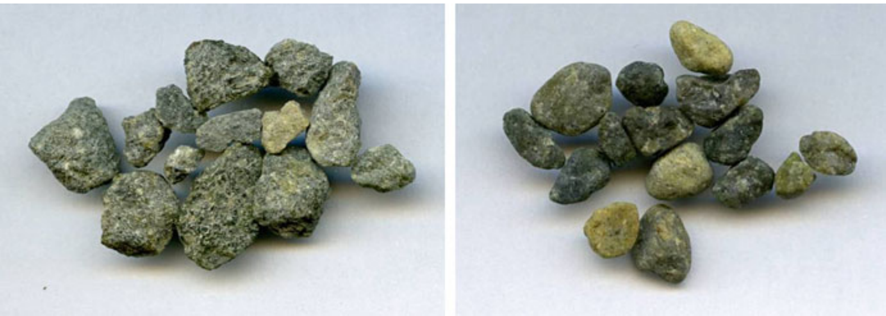

Fig. 7.8 Simulation of surf action. *Left*: Olivine grit (2–5 mm) at start. *Right*: Olivine grit, rounded and polished after 3 days shaking

different grain sizes. On its coastal side, there is a low inlet, permitting seawater to enter the box during high tide. The seawater will drain from the other side toward the sea during ebb, after having passed through 50 m of olivine sand. By analyzing the water before and after it has passed the olivine reactor, one can calculate how much olivine reacts during each tidal cycle, how much CO_2 it captures, and if and when the reaction starts to be hampered by reaction products attached to the grains. Once the CO_2 that is transformed into bicarbonate arrives in the seawater, it will take time before it is transformed into carbonates in the form of shells and corals, but no figures are available for this final uptake. Upon the formation of carbonate, half of the captured CO_2 is liberated again, but that may take centuries by when CO_2-neutral energy production may have been well established. It is reported that olivine dissolves somewhat faster in saline water than in fresh water [28].

The mechanical action, the grinding down of olivine grains, by waves and currents largely determines the rate of weathering of olivine on beaches and in shallow seas with strong bottom currents. There is a paper in which the rate of weathering of olivine grains on beaches is calculated [29], but that is based on theoretical modeling and overlooks the mechanical consequences of the surf, where grains are wearing down by the constant rubbing and bumping against each other. Olivine beaches are practically restricted to places where cliffs of olivine-rich rocks overlook the beach. Like on any other beach sand, the olivine grains are rounded, which means that they have lost little slivers of olivine due to bumping and scraping against each other. This has the advantage that one does not need to mill the olivine at high cost to very small grain sizes because the sea takes the place of the ball mill. These little slivers will quickly disappear by chemical weathering. In a recent experiment, this surf action was reproduced [30]. Grains of olivine were rotated in conical flasks. Within 24 h, the crushed olivine grains that were originally angular, with a rough surface, had transformed into rounded and polished grains (Fig. 7.8). The clear water at the start had become an opaque white suspension of very tiny olivine slivers, half of which had a grain size of less than 5 µm. The system reacts fast, the pH shoots up to 9.4, and a clay-type magnesium mineral is newly formed. The surf is clearly the world's largest, most efficient and cheapest ball mill. The experiments also showed that a mixture of different grain sizes of olivine wears down more quickly than single grain sizes. There are strong currents along the bottom of the Southern North Sea, the Channel, and the Irish Sea. If one would cover this sea bottom with a layer of 1 cm of crushed olivine, this would compensate 5% of the global CO_2 emissions, more than the combined CO_2 emissions of the adjacent countries, England, France, The Netherlands, Belgium, and Ireland, which together produce about 4% of the world's emission.

For tidal flats, a very different picture emerges. Tidal flats are inhabited by huge populations of lugworms (*Arenicola maritima*). The upper few centimeters of the mud pass on average three times each year through the guts of the lugworm. In its digestive system, weathering is 700–1,000 times faster than outside [31]. In experiments in which lugworms were fed with small fragments of basaltic rock, these pieces came out as small heaplets of clay [32]. As basalt weathers much slower than dunite, it can be expected that spreading of olivine grains over tidal flats is a very effective application of enhanced weathering/sequestration of CO_2.

Costs

Except in cases where the required olivine-rich rock is already available in crushed form on mine dumps, the application of enhanced weathering implies mining and milling of solid dunite rock from large open-pit mines. In 2002, two Swedish mining engineers calculated the cost of mining and milling of bulk rocks in large open-pit mines [33]. This turns out to be 6 Euros/t. After passing the primary and secondary crushers at the mine site, the granulated material must then be transported from the mine to the points of use. If we restrict the transport distance to a maximum of 300 km around the mine, this adds on average 6 Euros/t for transport. From the stoichiometry of reaction (7.5), one can calculate that 1 t of olivine will sequester 1.25 t of CO_2. This brings the price for the sequestration of CO_2 to slightly under 10 Euros/t, making it by far the cheapest method to sustainably capture CO_2. In comparison, CCS, the most commonly discussed technology for the sequestration of CO_2, costs between 70 and 100 Euros/t of CO_2 [34]. Even though the costs are modest, compared to other options that are considered, the cost to sequester all man-made CO_2 emissions by enhanced weathering of olivine still comes to around 250 billion US$/year worldwide.

The energy costs, expressed in terms of the CO_2 expenditure for mining, milling, and transport, have been calculated in an LCA (life cycle analysis) [35]. The total CO_2 requirement for the operation as a whole turns out to be 4% of the CO_2 that will later be captured by the extracted olivine. In order to bring the olivine grains from the mine to the point of use, several transport systems can be considered, including road, railroad, river, or sea. Making maximum use of the opportunities provided by local conditions will keep costs lower.

Mitigating Environmental and Social Costs

(*This section is mainly based on a personal communication by Mrs. Gwendolyn Wellmann, community development specialist, Port Elizabeth, S.A.*)

There are, however, also environmental costs (and benefits) to the olivine option. Every mining operation has a negative impact on the natural and social environment, even if the mining is done for the purpose of solving a worldwide environmental problem. Fortunately, there are ways to overcome the environmental damage, or at least diminish it. All dunite bodies in the tropical zone are covered by a thick laterite (laterite is the typical red soil of tropical countries, an iron-rich residue that is left after the weathering of rocks in tropical climates). Dunites contain on average 0.2–0.3% nickel, which is not very soluble during weathering. As most of the major components (SiO_2, MgO, CaO) are leached out during weathering, the Ni content of the residue rises accordingly and may reach 2% to sometimes over 3% Ni, making these nickel laterites a rich nickel ore [36]. In a number of countries (e.g., Brazil, Cuba, Philippines, Indonesia, New Caledonia, Madagascar, Malawi), these Ni laterites are mined, or mining options are being studied. By continuing the mining below the laterite crust and also mining the underlying dunite, one can avoid the impact of clearing a new mining site; the infrastructure for mining is already in place at such locations, and there is a local population that depends on mining for their living. The removal of the overburden consisting of Ni-laterite ore is an additional financial bonus for the mining of dunite below. This way, one can save the environment, help the people, and at the same time make olivine mining cheaper.

Even though mining brings with it many positives to an area, including employment opportunities, infrastructure development, and local economic development; and even if it is an olivine mine that will benefit the earth, mining also brings with it many negatives, such as an influx of job seekers, the increase of HIV/AIDS, violence, and discontent. In terms of mitigating the negative impact on the social fabric in a mining area, what is most important is the attitude of the mining company. The majority of mining companies adhere to the International Council on Mining and Metals' *Sustainable Development Principles* [37], with its underlying message that the overall success of any mining venture in a populated area is to consult, consult, consult with the local people. This requires the company from the onset, which might mean as early as the time of exploration activities, to meet with the local people; gain their views; understand their concerns; seek first to understand what makes the local community tick, before expecting them to understand your needs; and to devise long-term mitigating actionable plans for the rehabilitation of the area.

Should an olivine mine be established in a poor, developing country, it is vital that as the mining company starts to invest in the community, whether through infrastructure, local economic development, or alternative livelihood projects, it also invests in the development and promotion of a functional local government. This is important because the management and maintenance of these projects will ultimately be the responsibility of the local government. A weak, nearly nonexistent local government will lack the correct policies, procedures, structures, and systems to ensure sustainability of any project invested in by the local mine. The result of this is that there is a tendency for both the affected communities and the government to rely on the mine to assume local government roles. This can be problematic as implementing local governance is not the mine's expertise or responsibility, and as has been demonstrated numerous times in many countries – when the community feels the lack of service delivery – they will express their discontent by attacking the mine. One of the ways of investing in local governance is to expand the skills base of local government officials through training. The skills that are most frequently lacking in these areas are those needed for planning, budgeting, project implementation, and public consultation. Once olivine mining is a certified means of obtaining carbon credits, it is recommended that it is supervised by a special international body.

Applications of the Olivine Option

It should be emphasized that the presence of liquid water is essential. This rules out all countries with a desert climate except along their coasts, where olivine grains will disintegrate in the surf and be spread by wind and currents. The rate of reaction is increased by a high temperature. There are a number of arguments to focus on the wet tropics for *large-scale* applications:

- Weathering is fastest under these climatic conditions.
- Tropical soils are usually very poor, and can benefit from the addition of mineral nutrients.

For all mines, not just the ones in the wet tropics, the following arguments hold:

- Large mines profit from the economy of scale.
- Olivine rocks sometimes contain marginal chromite, nickel, or magnesite deposits, and the related rock kimberlite is the normal host rock for diamonds. If the host rock itself is also put to good use, and brings a profit in the form of carbon credits, instead of being dumped as a mining waste, this may make such marginal deposits economical.
- Once the olivine option is accepted as a legitimate means of carbon capture, the olivine mines can sell carbon credits.

There may be additional novel or niche applications suitable to certain regions, such as using olivine sand instead of quartz sand to strew into Astroturf playing fields, using olivine gravel on tennis fields, spreading olivine grains on lawns to reduce the growth of moss, using olivine as the top layer on footpaths or bicycle paths, adding a layer of olivine as roof covering, using olivine instead of quartz in sandblasting, using olivine grit on icy roads, adding olivine powder to digesters to improve the quality of the biogas by transferring the CO_2 part of the biogas to bicarbonate in the liquid, and using olivine sand in sound barriers along main roads.

These are all fairly minor applications, although taken together they are not negligible.

A larger application that can be used in the tropics as well as in temperate climates is to cover beaches that are subject to erosion or tidal flats by olivine sand. Olivine is considerably heavier than quartz and will not be eroded and transported away from the beach as easily as quartz sands. The tides will alternately wet the beach and drain the pore water. During such a tidal cycle, the olivine will react with the seawater, thus adding alkalinity to the sea, which makes it possible to store more CO_2 as bicarbonate in the seawater without acidifying it. The olivine grains bump into each other or scratch each other when they are moved by the surf. They become rounded, and the tiny slivers that have come off the grains will very rapidly weather [38]. If marine constructions (dams or artificial reefs) are built with olivine blocks and sand, this will conceivably lead to well-cemented structures after some time. Seawater is saturated with $CaCO_3$. If the water between the olivine pieces is only slowly replaced, it can react with the olivine for some time. This raises the pH of the interstitial seawater, causing a shift in the carbonate equilibria. This leads to a supersaturation of calcite which precipitates as a cement between the olivine. This process is similar to the formation of beach rocks. Filling the solution holes left by underground solution mining of salt with olivine powder serves two, possibly three purposes. After the cavity is filled with olivine, it is no longer necessary to keep the brine-filled hole permanently pressurized to prevent it from collapsing because the olivine will support the cavity. By injecting CO_2 into the mixture of olivine and brine, one captures CO_2 as bicarbonate. The heat of reaction of carbonatation + hydratation may heat the water sufficiently to use it for warm thermal medicinal baths, or swimming pools.

There are possible ideas for large-scale or economical applications of olivine. One such novel idea is the application of olivine in biodigesters. Biodigesters are installations where agricultural waste, urban sewage sludge, and organic waste stream from industry are anaerobically digested; during digestion, the organic material is turned into biogas. The biogas that is produced consists for around 2/3 of methane and 1/3 of CO_2. By adding olivine powder to the digester, part of the CO_2 gas will be converted to bicarbonate in solution, so the quality of the biogas will improve. As yet unpublished experiments at the Wageningen Agricultural University have shown that this is indeed the case, but there were two unexpected additional positive results. Most of the smell disappears. This can be explained by the fact that the FeO that is liberated when the iron part of the olivine dissolves reacts immediately with H_2S and forms solid iron sulfides. A second effect was even more unexpected. The methane increased not only in relative amount but also in absolute amount. In nature, serpentinization of olivine under exclusion of free oxygen is often accompanied by methane emissions. These are due to the following reaction:

$$6Fe_2SiO_4 + CO_2 + 14H_2O \rightarrow CH_4 + 6H_4SiO_4 + 4Fe_3O_4 \quad (7.8)$$

Sometimes this leads to methane flames, like in the Turkish Yanartasi (the rock that always burns), where a peridotitic rock is being serpentinized at some depth and in Los Fuegos Eternos (the eternal fires) in the Zambales ophiolite complex on the main island of the Philippines, Luzon. The increase in methane production in the biodigesters is probably due to the same reaction.

Collateral Benefits

It was already pointed out in the report entitled "Geoengineering the climate" of the commission on geoengineering to the Royal Society that the consequences of enhanced weathering could be benign in principle [39]. One of the most obvious advantages of enhanced weathering is that it has the potential to mitigate ocean acidification, the decrease of pH in response to rising atmospheric CO_2 concentrations [40]. The ongoing lowering of the pH endangers the growth of coral reefs and likely affects marine life in general. A number of other collateral benefits of the olivine option can be mentioned.

Improvement of Soil Productivity

Tropical soils, except in areas with recent volcanism, are generally very poor, as they lack important mineral nutrients like magnesium, calcium, potassium, and phosphate. A limited amount of these nutrients is recycled by the standing forest, but once this is cut, the trees are removed and the soil is used for agricultural crops, it loses its productivity very fast. Spreading crushed dunite rock will solve the magnesium problem, and the calcium problem as well to some degree. Kimberlites, the host rock of diamonds, are also olivine-rich, but in addition they contain a potassium mineral (phlogopite) as well as often some phosphate. In order to provide a more balanced mixture of major nutrients, one should spread the crushed kimberlite of which hundreds of millions of tons are lying on the mine dumps of diamond mines. At the request of the author, seepage waters from kimberlite tailings in India and South Africa were analyzed. It was found that the waters that seep through these kimberlite tailings have a high pH and sequester large amounts of CO_2 as bicarbonate. Moreover, they can contain significant concentrations of potassium (data from the de Beers mine tailings in South Africa and the Panna diamond mine in India, Schuiling, unpublished). Again, one should avoid as much as possible mining wastes that contain chrysotile, although this white asbestos is considerably less dangerous than crocidolite (blue asbestos) or amosite (brown asbestos), which are not associated with dunites or peridotites. Recent research by Arcadis has shown that chrysotile weathers quite fast, and thereby loses its fibrous character [41].

Mindful of the beneficial effects of volcanic rock on the fertility of soils, it may also be a good idea to spread mixtures of crushed dunite and rock meal, like volcanic tuffs in areas with poor soils. Although the rock meal weathers considerably slower than dunite powder, and thus contributes much less to CO_2 sequestration, the combination is likely to increase productivity of the soil for a number of years, by which it can contribute to world food production. In view of the scarcity of potash fertilizer and its high price, it is recommended [42] to use powdered nepheline syenites to provide potassium to poor tropical soils, as the nepheline $(Na,K)AlSiO_4$ weathers considerably faster than potash feldspar. Acid sulfate soils form another problem that can be solved by spreading olivine. The Mekong delta, as well as a number of soils in estuaries along the East coast of Australia, suffers from high acidity, once the soil is plowed in preparation for agriculture. This exposes the pyrite in these soils to the oxygen in the air. The pyrite oxidizes and produces sulfuric acid, which reduces their rice productivity (Mekong) or sugarcane production (Australia) considerably [43]. If crushed olivine is spread on these acid sulfate soils, its weathering will neutralize the acid and thus increase their productivity. If the neutralization reaction is simplified to

$$2FeS_2 + 2Mg_2SiO_4 + 7.5O_2 \rightarrow 4MgSO_4 + Fe_2O_3 + 2SiO_2 \tag{7.9}$$

this would mean that 1 t of soil with 1% of pyrite would require close to 12 kg of olivine for complete neutralization.

Production of Biofuels from Siliceous Algae

In a very different manner, olivine can help to produce biofuel from algae instead of from land-based crops which use up land that is thereby lost for world food production and cost large amounts of scarce irrigation water [44, 45]. Diatoms (siliceous algae) grow very fast and are a proven raw material for the production of biodiesel, as they have a lipid content around 50%. Their growth, unfortunately, is often limited by the availability of silica, which they need to build their silica skeletons. During weathering, olivine releases large amounts of silica in solution. If one constructs a lagoon, preferably where the coastal waters carry large loads of urban or agricultural waste, diatoms can be grown in the following manner. The lagoon should be along a coastal stretch where the beach can be covered by olivine sand. A dam must be constructed around the lagoon to separate it from the open sea, except for one or a few U tubes through this dam. These U tubes permit the exchange of water between the lagoon and the open sea. The opening on the lagoon side of each tube should be covered by a perforated metallic plate, which acts as a support for a plankton net. During high tide, the water will flow into the lagoon and wet the olivine sand on the beach. During low tide, there will be an outflowing current of the water, including the part that formed the pore water between the olivine grains on the beach that is drained during ebb. This outflow will also carry the diatoms, but these are retained on the plankton net, where they can be harvested to be used as the raw material for biodiesel production. It is expected that in such diatom farms almost pure diatom cultures will form, as they have a competitive edge over organisms that do not use silica.

The growth of diatoms might also be stimulated for quite a different purpose. More and more often, coastal waters are threatened by poisonous dinoflagellate blooms ("red tides"), causing massive fish kills, and threatening human health. The main reason is believed to be the supply of untreated urban waters that contain high levels of nutrients. If one could stimulate the growth of fast-growing and nonpoisonous diatoms to consume the contained nutrients in these coastal waters, this may reduce or eliminate the dinoflagellate blooms. Besides, diatoms are an excellent fish food. The stimulation can be done by applying the same method, covering the beaches with olivine sand which will provide the silica that is necessary for the growth of the diatoms.

Geopolitical Implications of the Olivine Option

Although many scientists realize that CCS is a very costly operation [34], and is unlikely to solve the CO_2 problem at the required volumes, it is still the most widely discussed and preferred option, particularly among Western politicians and those that may benefit from the uptake of the technology. This can make for a powerful lobby. Many scientists urgently call for a reduction of CO_2 emissions, by increasing efficiency, changing our lifestyles, and switching to sustainable energy (wind, water, solar power, biomass, geothermal energy, or even nuclear energy). In the long run, mankind will be obliged to make the change to renewable energy anyhow, as our reserves of oil and gas are dwindling. Although there are still vast reserves of coal, these too are not endless. It is evident that the world should reduce its carbon footprint as quickly as possible. Some are concerned that the possibility of CO_2 sequestration may provide an excuse not to change current lifestyles, sometimes called the "moral hazard" argument. Trying to change people's attitudes and lifestyles is necessary, but it may be a slow process, whereas the danger of climate change requires immediate action. The two sides have a different time frame, and both are needed to avoid the imminent climate change caused by rapidly rising CO_2 levels in the atmosphere.

The picture is complicated by the fact that the largest industrializing nations (such as China, India, Brazil, South Africa, and Indonesia) all possess vast coal reserves and want to lift the standard of living of their people as quickly as possible. One requirement to reach that goal is the unlimited access to cheap energy. Reduction of emissions is, therefore, not necessarily a top priority. Enhanced weathering ideas may be a way to break this deadlock by providing a means of compensating their emissions by using olivine. All five industrializing nations mentioned also possess vast reserves of olivine rocks. They can exploit these with their own workforce for 10% of the cost of CCS for the same amount of CO_2 sequestered, and it will give them large employment opportunities. Moreover, if they produce more olivine than required to meet their own agreed quota, they can sell surplus carbon credits to the other countries for 15 Euros/t of CO_2, and still make a profit. This will permit them to continue to use their cheap energy based on coal, while still doing their part to counteract climate change.

Future Directions

A number of niche applications of the concept of enhanced weathering of olivine have already been set into motion, like the inclusion of olivine in roof coverings, the addition of olivine to fertilizers or potting soil, or the covering of (bicycle) paths with olivine sand. Several other niche applications are under development. A breakthrough, though, depends on the certification of the olivine option for carbon credits, and the start of large-scale olivine mining. In the global climate debate, enhanced weathering can play a major role to reconcile the views and aims of the West and those of the emerging economies (China, India, Brazil, South Africa), if it is accepted that these countries may continue to use their vast coal reserves as a source of cheap energy, on condition that they compensate their CO_2 emissions by enhanced weathering of olivine.

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Chapter 8
Geoengineering Policy and Governance Issues

Sean Low, Nigel Moore, Zhewen Chen, Keith McManamen, and Jason J. Blackstock

Glossary

Carbon geoengineering	A variant of geoengineering also known as Carbon Dioxide Removal (CDR) concepts aimed at capturing carbon dioxide directly from the atmosphere by either enhancing existing natural sinks or by using chemical engineering technologies.
Geoengineering	Also known as "climate engineering" refers to the deliberate and technological manipulation of the climate system to forestall the worst effects of global warming.
Governance	The management of political issues and physical systems that relies not only on (traditional) government at state level, but upon a wider range of actors at the international (international organizations, minilateral clubs), substate (provincial and municipal government), and nonstate (industry, civil society, knowledge networks) levels with such management often involving coalitions across multiple levels and actor types.

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Solar geoengineering	A variant of geoengineering also known as Solar Radiation Management (SRM) concepts aimed at enhancing reflecting incoming sunlight back into space, preventing absorption by Earth's atmosphere, surface or oceans, and thereby reducing global temperatures.
Weather modification	An antecedent to climatic geoengineering focused on the manipulation of local atmospheric conditions to induce short-term, bounded changes in weather. Such methods are different from geoengineering methods in that they are smaller in scale and intent of atmospheric modification.

Definition of the Subject

Geoengineering – the deliberate and technological manipulation of the climate system to forestall the worst effects of global warming (*also referred to as climate engineering*) – has recently emerged as a novel and controversial issue in climate governance. It is sometimes proposed as an insurance policy, should either (a) primary efforts to develop sustainable energy and societal systems prove unable to quickly enough overcome the inertia of current ones, or (b) uncertainty in the climate system lead to unexpectedly large damage to societies and ecosystems [1]. This entry explores the current attempts and future ideas for governing emerging geoengineering research programs and technologies in ways that effectively manage their climatic and societal impacts.

Introduction

The international and national discourses surrounding geoengineering are currently in an early formative stage, bringing together scientific, ethical, legal, and political dimensions, whose details and interlinkages have been at best only preliminarily mapped to date. Given the potential accessibility and proposed utility of various geoengineering techniques, the discussion of geoengineering is expected to expand and evolve significantly as new actors begin to engage with the issue.

As a result, the core challenge of designing governance frameworks for geoengineering technologies, particularly at the international level, stems from uncertainty about the future. On the scientific side, research and deployment is confronted by large constraints in forecasting impacts and in running meaningful field tests. Political contentions include the potentially unequal distribution of technology ownership; risk in field-testing and deployment; the unclear effects of the debate's politicization by a range of states, researchers, private developers of technology, and civil societies; the potential introduction of moral hazard into climate change mitigations efforts; and complex intersections with a number of global governance issues and policy communities. Underlying each of these political contentions are ethical perspectives on how humanity should interface with the natural world.

Hence, the crux of governance is not only to provide guidelines and structure for managing near-term actors and issues, such as scientists and research agendas. It must also set a framework for managing new related issues and interconnections as they evolve. In this context, caution regarding "locking in" governance mechanisms designed to manage only near-term issues is needed, as narrow frameworks might hinder capacity to address future possibilities and issues. Even independent of lock in, any regulation enacted in the near future could clearly have a formative impact on the future governance of geoengineering. However, with the current lack of an institutional forum for discussion, national legal frameworks, or even coherent rules and framings among the actors now driving the debate, projecting what that impact might be is exceedingly difficult. For these reasons, recent initiatives – such as the Solar Radiation Management Governance Initiative (SRM-GI) and Asilomar Conference on Climate Intervention Technologies [2, 3] – aiming to promote the development of responsible governance for emerging geoengineering research and technologies have focused first on expanding the conversation to include greater diversity in the perspectives involved.

The following sections will provide a general overview of the various components of the geoengineering governance debate. For more detail on any of the issues raised, readers are encouraged to examine the references herein in detail. The section on "History of Weather Modification and Governance Frameworks" chronicles historic environmental modification activities as limited antecedents with potential influence on how geoengineering governance may evolve. The section on "Geoengineering Technologies and Governance Challenges" outlines important distinctions between different geoengineering technologies and stages of research, and highlights how these relate to a range of technical and sociopolitical issues that may require governance. The section on "Current Governance Landscape" then provides an overview of the currently emerging governance and policy landscapes, including identifying key actors, institutions, and issues that have shaped and are currently shaping the governance landscape thus far. Finally, as the current landscape remains far from comprehensively developed, the last section on "Future Directions" overviews existing proposals for the future governance and regulation of geoengineering research and technologies.

Given the nascent state of national and international discourses on geoengineering, this entry necessarily focuses as much on speculative future proposals for governance as on existing governance or policy. This balance highlights the uncertainty – and potential for influencing future developments – that exists within the discourse.

History of Weather Modification and Governance Frameworks

While the current concept of geoengineering is relatively modern, it can be situated within a long tradition of human attempts to influence their environmental surroundings. Throughout history, mankind has imagined and fantasized about weather control; such stories have been found in myths, religious texts, traditional and cultural practices, and science fiction. Granted, the nature of weather control aligns only partially with geoengineering. Yet, its historical treatment has implications for looking at geoengineering from the point of view of antecedent governance efforts.

In practice, the attempt to manipulate atmospheric conditions had its roots in the Enlightenment vision of deconstructing natural processes for the betterment of human society. However, it was not until the nineteenth century that scientific research programs materialized in this capacity. The early weather engineers were rainmakers who proposed various physical, chemical, and electrical approaches to manipulate precipitation.

Prominent figures included James Pollard Espy (1785–1860), a US government appointed meteorologist, who actively promoted the idea of commercial rainmaking by cutting and burning forest woods. Edward Powers and Daniel Ruggles were former US generals who theorized that cloud condensation by explosive agent could lead to rainfall [4]. Other prominent figures of this “concussive” stream included Robert St George Dyrenforth (1844–1910), a lawyer from Washington D.C. who received government funding to lead such experiments, J. B. Atwater, who filed a weather patent to disrupt tornadoes, and Laurice Leroy Brown, who filed a patent application for an “automatic transporter and exploder for explosives aiding rain-fall” [4]. Instead of engaging in serious science, rainmakers of the era often sought to defraud desperate farmers. A famous example was Charles Hatfield, who evaporated chemicals to “coax(ed) rain from the sky” [5]. Projects of the age were backed by poor theory, speculative knowledge, and insufficient financial support; and regulatory frameworks were largely nonexistent.

Whereas agriculture interests were the primary driver for nineteenth-century weather controlling schemes, research into weather control during the two World Wars was supported mostly by military patronage as well as some corporate financial support. Aerial fogs and vapors impeded forward visibility of troops and bombing raids, and the escalation of World War II called for immediate scientific attention. In 1940, Britain created the Petroleum Warfare Department (PWD) and charged it with the task of developing a reliable method of clearing fogs called the Fog Investigation and Dispersal Operation (FIDO), the project brought together scientists, engineers, industrialists, and policy makers under one umbrella. With FIDO, the British and Allied air forces were able to assume normal military activities as the Germans were bound to the ground. Despite its success as a wartime invention, FIDO ultimately proved to be too costly to be commercially viable [4]. However, this complex of interests carried over to the Cold War, with even greater strategic interests of weaponizing weather control techniques.

The scientific foundation began in the General Electric (GE) Research Laboratory, where in 1946, Vincent Schaefer and Irving Langmuir discovered a weather-control technique that used dry ice and silver iodide to "supercool" clouds [4]. Langmuir envisioned that the technology would have the potential to deflect hurricanes, generate large-scale precipitation, and clear the sky for aviation services. The confluence of military interests and GE's concerns over liability threats prompted the transfer of research to a newly established classified cloud-seeding program called Project Cirrus, a collaboration between GE, the US Air Force, the US Army Signal Corps, and the Office Naval Research. Cirrus resulted in more than 250 trials and experiments during 1947–1952, but was canceled due to tort liability lawsuits [4]. Project Stormfury, successor to Cirrus and operational from 1962 to 1983, was another collaboration between the Weather Bureau and the military in exploring cloud-seeding experimentation.

During the Vietnam War, Operation Popeye, a clandestine field trial of cloud seeding was conducted along the Ho Chi Minh Trail to stonewall traffic. With support from the US administration, a much larger scale program known as Operation Motorpool, covering areas over Laos, Cambodia, North and South Vietnam, ran until July 1972 [6]. Similarly, French forces also attempted artificial rainmaking to impede the movement of Vietnamese troops; not to mention the frequent practice of cloud seeding by Moscow in the 1950s and the 1960s.

The fact that many of the research and trials remained in the domain of military operations shielded them from public criticism and regulatory oversight. Therefore, despite scant court cases on the tort liability of cloud-seeding experiments, no statutory laws were erected to regulate weather modification during the interwar period. However, when a story about the operations in Vietnam broke in the Washington Post in March 1971, it became a public relations disaster that led to pressure from members of the Congress on the Nixon administration to cease the operations. The efforts to ban environmental warfare were led by Senator Claiborne Pell in the Senate and Representative Donald M. Fraser in the House, leading to the passing of numerous bills in 1975 that sought to prevent the weaponization of weather modification [4].

The Soviet Union was quick to respond to the "Watergate of weather warfare." First, it invited the Nixon administration to sign the "Joint Statement Concerning Future Discussion on the Dangers of Environmental Warfare" at the Moscow Summit in 1974 [7]. It then presented the United Nations with a proposal to establish an international convention to outlaw weather modification as a weapon of war. This diplomatic initiative caused the Ford administration to commit the USA to the negotiation and eventual signing of the Convention on the Prohibition of Military or Any Other Hostile Use of environmental Modification Techniques (ENMOD), which banned all militarized environmental modifications with "widespread, long-lasting or severe effects as the means of destruction, damage or injury to any other State Party." ENMOD went into effect in October 1978 with 70 state members. However, the treaty was viewed as deeply flawed: It contained vague texts, established a high threshold for violation, and did not prohibit research and development in the field, which made the treaty almost unusable [4]. Nonetheless, the ENMOD treaty has still been discussed as potentially applicable to certain geoengineering techniques due to the potential for detrimental transboundary impacts [8–11].

Despite limited scientific evidence of success over the past half-century, the turn of the century still witnessed governments in drought-prone countries resort to cloud-seeding techniques to enhance precipitation. During the 2008 Summer Olympics, Beijing deployed over 1,000 rain dispersal rockets to prevent rain during the opening and closing ceremonies [12]. Over the summer months of 2010, the Abu Dhabi government commissioned an $11 M project that used ionizers to generate storms [13].

The boundary between what constitutes weather modification (*a single nation attempting to modify their own weather*) versus geoengineering (*sufficiently large modification of the atmosphere to have significant transboundary impacts*) currently remains ill defined – and, for the moment at least, uncontested. However, the potential for both evolving weather modification and geoengineering experiments to encroach on this boundary points to the importance for effective governance of understanding the above history. In the long tradition of weather modification, human civilization has largely bypassed the ethical, social, and legal dimensions of the issues, rendering much of its governance and legislative aspects unresolved to the present day. Yet, the nature and magnitude of weather modification efforts to date are child's play compared to the potential for some geoengineering technologies to generate impacts over much larger areas and timescales. Thus, the historical incapacity to regulate atmospheric interference causes concerns that geoengineering research will demonstrate a repeat of the same evolutionary pattern.

Geoengineering Technologies and Governance Challenges

Distinguishing Carbon and Solar Geoengineering

Geoengineering is a blanket term that encompasses a suite of very different techniques for intervening in the climate system [14]. In designing effective governance arrangements for different geoengineering technologies and research, it is therefore crucial to take account of the differences between them. Moreover, as these technological constructs will themselves evolve with time, governance frameworks need to be adaptable to addressing their possible evolution as research progresses in climate science and geoengineering. What follows is a basic description of prominent geoengineering methods that have been envisaged to date, including identification of the various technical and social uncertainties surrounding each.

The broadest differentiation of prominent geoengineering technologies is between *carbon* and *solar* geoengineering, which tackle the climate challenge from very different directions. Through removing excess carbon from the atmosphere, carbon geoengineering (*also called carbon dioxide removal or CDR techniques*) aims to treat the *cause* of climate change by removing carbon dioxide already in the atmosphere. Solar geoengineering (*also known as solar radiation management or SRM techniques*), on the other hand, aims to reflect incoming solar radiation before it is absorbed by Earth's atmosphere, surface or oceans. This strategy aims to offset the energy imbalance created in the Earth's climate by greenhouse gasses trapping outgoing infrared radiation, thereby avoiding or reducing the anticipated "global warming" from that imbalance.

The actual technologies being proposed to implement either carbon dioxide removal or solar radiation deflection vary significantly. Carbon geoengineering, for example, can be broken down into two subcategories: engineered and ecosystem. The prominent techniques in these categories differ in ways that would suggest the requirement of very different arrangements for their effective governance. The most studied engineered technique is *direct air capture*, which would utilize machines (the size of large buildings) that draw in ambient air, chemically remove carbon dioxide from it, and store that carbon somewhere for a very long time [15]. On the other hand, the ecosystem carbon geoengineering technique that has garnered the most attention from scientific and environmental governance communities is ocean fertilization, which is premised on the enhancement of a process of CO_2 removal that already exists in nature. With ocean fertilization, nutrients (of which iron is most often discussed) that limit the growth of organisms such as algae and phytoplankton are added to the oceans to stimulate the growth of these species. These organisms require carbon to live and grow which they fix from the atmosphere, and when they die they sink and eventually sequester the carbon in deep ocean sediments [16].

Solar geoengineering tackles the climate challenge from a different direction, focusing instead on attempting to enhance the albedo of the planet. Notable options for achieving this include launching mirrors into space to reflect sunlight before it reaches the atmosphere, brightening clouds by spraying a mist of cloud condensation nuclei (fine particles) into the lower atmosphere, and injecting reflective aerosols such as sulfates [17] – or possibly synthetic variants made of nanoparticles [18] – into the stratosphere. Space mirrors remain presently unfeasible at scale, while the most prominent cloud brightening technique would use globally distributed oceangoing vessels to remove sea salt from the water below them and spray it into the air above [19]. Stratospheric aerosols are perhaps the most plausible solar geoengineering method thus far proposed, in part because they mimic a natural process that is known to cool the planet significantly – large volcanic eruptions which send sulfate aerosols high into the stratosphere for a period of about a year. Delivery methods for distributing aerosols in the stratosphere include balloons, guns fired from ground level, and airplanes [1].

At this early stage of technical research and development, it is important that governance frameworks be developed with recognition that the technicalities of hypothesized geoengineering methods will evolve significantly. To date, most concepts remain largely theoretical, with the technologies for deployment not yet having been developed or demonstrated. Over the coming decades there will assuredly be changes to these proposals. Some may be scrapped and forgotten, and new ideas will emerge. As a result, the early arrangements for governance would best be made flexible to these future developments, whatever they happen to be.

Nonetheless, governance assessments of geoengineering have emphasized that uncertainty about future technologies should not be an excuse for complete inaction today, as there is a great deal of information that can be teased out of prior geoengineering research – and relevant precedents for other evolving technologies – that can guide near-term governance efforts [20]. For one, it has become apparent that some techniques could be extremely inexpensive and yet have the ability to significantly impact climate on a global scale [21]. Take, for example, stratospheric aerosol injection, which may be able to cool the planet by multiple degrees centigrade over a timescale of a few years at a cost of a few billion dollars per year [21]. The technologies necessary to develop this capability are relatively simplistic by modern standards, and more than likely could be developed by a small group of actors [22, 23]. On top of this, deployment has the potential to be carried out clandestinely given the relatively small amount of aerosol loading required and deployment of this kind would impact the globe, with potentially severe side effects for some human and natural systems. Clearly, with this type of scenario, there is a near-term need for suitably tailored governance.

Despite the negative connotations of geoengineering in the previous example, the large consequences that will likely be faced if climate change is under addressed by insufficient mitigation efforts suggests that geoengineering research may be a prudent investment. If this is the case, there may be a need for near-term governance frameworks for geoengineering research to foster open and collaborative research projects aimed at uncovering whether or not there are techniques that might be worthy of deployment should dangerous climate change appear unavoidable [22].

Research Stages

Research into geoengineering, some of which is already ongoing in the absence of such a governance framework, will follow a predictable set of stages beginning with basic theory and modeling [1]. Understanding stages and categories of research, along with challenges and impacts they may pose, could be a starting point for establishing governance by pointing out which types of activities deserve governance attention, which do not, and therefore at what point in the future a legitimate system of research governance must be put in place . See Table 8.1 for further detail on different categories of research and the potential governance issues they raise.

The first two stages of research do not involve the release of substances into the environment – they involve strictly lab-based activities. Thus, in the eyes of many, governance of these stages is not warranted – any restriction on these activities would not directly protect the environment or citizens from harm, and likely would be very difficult to enforce because geoengineering research at these stages appears (and could even be disguised as) very similar to other kinds of research such as climate

Table 8.1 Geoengineering research stages

Stage	Description	Challenges	Status	Direct environmental impact
1. Theory and modeling	Publications and computational models studying the anticipated climatic impacts of geoengineering techniques	International cooperation, research funding, and development of more comprehensive models	Studies began more than 20 years ago and continue for both carbon and solar technologies	None
2. Technology development	Design and laboratory development of geoengineering deployment technologies	Emergence of governance issues when technologies are patented or classified. Who has access to and control over new technologies?	Many carbon geoengineering technologies are currently under development	None
3. Subscale field testing	Feasibility testing of geoengineering deployment technologies at levels posing “demonstrably negligible” environmental and transboundary risks	Evaluating risks and modeling uncertainties related to the environmental impacts of field testing	Limited recent tests of atmospheric aerosol injection and ocean iron fertilization have taken place	Small, regional, temporary (negligible)
Difficult to define threshold				
4. Climatic impact testing	Testing the global climate impacts of geoengineering deployment, nominally at scales below actual deployment, but with notable transboundary environmental impacts	Environmental and governance challenges of experiments spread unevenly at local, national, and regional levels. Definition of large scale	A few, limited proposals for this kind of testing to begin soon	Potentially global and large (nonnegligible)

science, meteorology, volcanology, etc. However, other perspectives hold that even these early stages of research will have important path-dependent impact of the types of technologies that evolve and are eventually used, and thus need to be incorporated into broader governance frameworks – even if only through broader discussions of the implications of certain research directions. Moreover, issues of technology ownership and transparency of research activities are directly raised at this stage – both of which are at the core of most evolving geoengineering governance proposals.

The two latter stages of research involve an intentional release of substances into the environment of some kind. Small-scale field studies are defined as having a negligible environmental impact and for this reason there is justification of some reticence regarding strict regulation as these activities would not have transboundary effects, nor would their effects be climatically relevant. Examples might include testing the effectiveness of a nozzle for spraying sulfate particles. To many within the field of geoengineering science, a key governance question is where to define the threshold between negligible and nonnegligible, as this ought to be the point where governance restrictions kick in. Unfortunately, as long as the threshold between negligible and nonnegligible remains difficult to define, this stance toward governance may be problematic to implement.

An alternative viewpoint worthy of mention sees research stages as irrelevant. Proponents of the "slippery-slope" notion hold the view that early stages will increase dramatically the likelihood of eventual deployment and thus are nonnegligible and should themselves be subject to regulation [11]. This argument raises an important set of questions: Is geoengineering research unique? Ought it to be treated like chemical weapons and other kinds of research that have obviously dangerous consequences that make their restriction critical even at early theoretical stages? Can and should geoengineering researchers be trusted? Where in all of this do the widely held value of scientific freedom, and the commitment to the scientific search for truth fit? These are difficult questions that can foster conflict between alternative viewpoints rooted in different ethical perspectives.

Issues

Sociopolitical Linkages

Geoengineering requires a highly nuanced governance approach to accommodate the various impacts of research, development, and possible deployment. Unfortunately, thinking of geoengineering in and of itself is not necessarily helpful for governance – in fact, this perspective could be damaging to the design of a successful governance regime because geoengineering is tied inexorably to many other issues. Prudence dictates that these linkages should be accounted for in discussions and deliberations of geoengineering governance challenges.

The climate regime is the most obvious and fundamental decision-making structure with which geoengineering governance must be intimately tied. Further potential need for geoengineering can be seen as proportional to the lag in progress toward mitigation, and related to limited progress toward (or capacity for) adaptation. With their successes and failures, as well as the progression of geoengineering research from theory to practice, the potential future role of geoengineering will become clearer. For this reason it appears important to meaningfully link decision-making structures that dictate responses to climate change – whether they be mitigation, adaptation, or geoengineering based.

Another importantly linked issue is that of the economic and social development of least developed nations. The largest impacts of both climate change and geoengineering are likely to be felt by the poorest communities of the world, leading to an important question of how their needs should be addressed [24]. Another tightly linked issue area is that of international security, as both climate change and geoengineering research or attempts at deployment could have destabilizing geopolitical effects [25]. Such linked issues – also currently including agriculture and food security – are likely to only grow as geoengineering becomes a more well-defined proposition. Air pollution could also be a future candidate because the injection of sulfates into the stratosphere is one of the prominent techniques proposed. The line between stratospheric and tropospheric sulfate could blur in the future – in fact some people have already begun to examine the dichotomy between costly decreases of sulfur emissions over the oceans from ships which do little to improve human health, yet increase warming through reducing cooling tropospheric sulfate aerosols in ways that may accelerate the danger to societies and ecosystems from climate change [26]. This suggests there may be legitimate reasons to coordinate air pollution and geoengineering governance regimes in some respects as well.

There are additional questions regarding post-deployment governance that while far off, must be agreed upon well in advance of deployment and almost assuredly before any large-scale field-testing is underway. The tough governance questions raised by the prospect of having winners and losers – some nations that benefit more from a geoengineering intervention than others, to the point where some areas could face serious negative side effects while others reap tangible benefits from climate change avoidance – are prime examples of this.

Ethics

Those who decide whether or how to utilize geoengineering in response to climate change will wield a tremendous amount of power. The expansive normative and ethical implications of such decisions are the core reasons why geoengineering is such a controversial topic, even in its current formative stage. Though seemingly unwieldy, such far-reaching questions can be teased into a number of more precise ones for understanding the implications for governance of the ethical issues entangled in the prospect of geoengineering our climate.

One example is the question of informed consent. Geoengineering resembles the concept of an experiment being performed on the entire global population [27]. Much like experimental medicine, it would be performed in order to avoid a larger problem; however, there is potential for severe negative side effects that could be very damaging. In medical scenarios, informed consent from the patient is required to carry out treatment. However, in the example of geoengineering, this is impossible: One cannot procure informed consent from everybody on the planet. Implementing geoengineering and large-scale tests of geoengineering methods necessitates a decision-making structure that is remarkably inclusive. Unfortunately, the degree of inclusivity required to make such an experiment ethically acceptable will always be a highly controversial question in and of itself.

There is also the question of liability if something goes wrong. In a scenario where unintended negative side effects are felt by one group of stakeholders, there will certainly be finger-pointing. A successful governance regime will have well-defined courses of action to deal with liability and compensation issues in place well in advance of any large-scale testing [11]. Actors currently engaged in geoengineering discussions today should pay close attention to their personal responsibilities and actions, and how they may be relevant to future liabilities. Scientists in the early stages of research who are pushing for field-testing, as well as those who speak out proposing a ban on all research, should acknowledge the influence that such stances can already have in dictating the future of geoengineering research and development.

These examples are a few of the many ethical issues that arise when geoengineering (both research and potential deployment) is considered. They tend to stem from a small group of high-level concerns. One is the issue of intent. Though humans have been changing our environment for centuries, the scale and intent of this incarnation is different, [28] and intentionality in most moral (and legal) constructs brings with it added responsibility and liability for wrongdoing or negligence. Another issue for many who are skeptical of geoengineering's acceptability is that it is a technologically based fix to a problem caused in large part by the use of other powerful technologies. People with this viewpoint see climate change as a problem requiring behavioral changes – not technical solutions – to ultimately solve. Finally, there is the high-level question of who decides [1]. This brings up issues of informed consent, global participation in the decision-making process, the importance of collaboration, the primacy of the interests of the most vulnerable among us, and many others.

Current Governance Landscape

As the subject of climate intervention technology has gained attention within the scientific community, public debates on the topic have grown in their legitimacy and significance. As a result, the constellation of actors involved has also grown to encompass an array of groups and individuals within the public and private sector, government and NGOs, foundations, private philanthropists, and firms [29]. In spite of the diversity of interests at play, most actors to date have presented arguments somewhere in between the extreme poles of either aiming to have geoengineering implemented in some fashion, or aiming to establish a full moratorium on deployment of the technology.

The recent expansion of attention to geoengineering was nucleated by a landmark 2006 article by Nobel Laureate Paul Crutzen [17]. Within a few years, the emerging attention prompted the UK Royal Society and the US National Research Council to explore geoengineering, and issue reports calling for geoengineering research, and a joint statement by the G8 + 5 nations included a call for an international meeting on geoengineering [30–32]. Policy statements calling for research into climate engineering have also emerged from the American Meteorological Society, the American Geophysical Union, and the UK Institution of Mechanical Engineers [33–35]. Each of these documents emphasized the primacy of reducing GHG emissions, but recommended developing conventions for the scientific community and the blueprints for a formal geoengineering governance framework. The Royal Society concluded:

> Little research has yet been done on most of the geoengineering methods considered, and there have been no major directed programmes of research on the subject. The principal research and development requirements in the short term are for much improved modelling studies and small/medium scale experiments (e.g. laboratory experiments and field trials). Investment in the development of improved Earth system and climate models is needed to enable better assessment of the impacts of geoengineering methods on climate and weather patterns. . .as well as broader impacts on environmental processes [30].

Prompted by the increasing scientific attention to these issues, committees of the UK Parliament and US Congress released their own reports on geoengineering [36]. Both bodies explored related issues in a series of science and technology hearings on the subject, and reached conclusions endorsing increased research into emerging geoengineering technologies. As an important indicator of concern for the international dimensions of geoengineering research and technologies, the UK Parliament and US Congress organized their hearings jointly, allowing testimony, documents, and reports from each other's hearings to be presented in their own. This is the first, and thus far only, time in history that such an arrangement between the UK and US legislative bodies has been used.

Government funding for geoengineering research is currently scarce in the US, where scientists rely primarily on private philanthropy and redirected federal research grants. Some researchers have argued that a delay in establishing a federal program will make it progressively harder for the US government to guide these efforts in the public interest as the dialogue continues to move rapidly forward [37]. Elsewhere, public funding has been gradually forthcoming. Both the UK and the European Union have recently provided preliminary grants to a few moderate research projects [38–40]. Small, government-funded projects using computer models are also underway in Germany, [41] and Russia has already conducted at least one geoengineering field test [41]. Nevertheless, a document from the UK Parliamentary Office of Science and Technology conceded:

> There is currently very little public funding specifically earmarked for geo-engineering. Despite a US Department of Energy White Paper (Unpublished) that in 2001 recommended a $64 M, five-year programme, less than $1M of public money is currently directly funding geoengineering research in the USA. In the UK, the Engineering and Physical Sciences Research Council (EPSRC) has proposed a £3M "Ideas Factory" commencing in 2010. To date, therefore, most research has been either funded using existing climate science grants or has been unfunded, performed in researchers' spare time [42].

The same report posited that an international research program of $100M would significantly increase the scientific and engineering knowledge, as well as provide greater understanding of the risks associated with altering climate system. (Also in March 2009, the UK's Royal Society proposed that a $200 M international fund be established for research into geoengineering [43].)

Much of the present funding for geoengineering research comes from the private sector. Leading organizations such as Environmental Defense Fund, Novim Group, and Climate Response Fund rely on the philanthropy of private donors. Even billionaires Bill Gates and Richard Branson have also brought money to bear on geoengineering research. (Since 2007, Gates has put at least $4.5 M into the Fund for Innovative Climate and Energy Research [44, 45] and Branson offered a $25 M cash prize rather than a research grant, which went unendowed [46]). To date, despite significant concerns that vested fossil fuel interests may back geoengineering technology development, there remains no evidence that corporations and industries standing to benefit from continued GHG emissions are investing publically in geoengineering research [47, 48].

A group of corporate actors did emerge in the mid-00s with the goal of generating carbon credits to sell from using ocean-fertilization techniques. Ocean fertilization company Planktos' plan to sequester carbon dioxide through a release of iron filings into the Pacific Ocean was blocked by a petition to the EPA by environmental groups invoking the Marine Protection, Research, and Sanctuaries Act (the "Ocean Dumping Act"). Though Planktos attempted to flout the regulation by using a vessel flying under a different national flag, a lack of investors forced the company to abandon the project [49].

As a consequence of such activities, the International Maritime Organization (IMO) became engaged in the governance of iron fertilization projects. (In July 2007, EPA dispatched a memo to the IMO revealing that Planktos intended to proceed with its planned project without the permit required under the Marine Protection, Research, and Sanctuaries Act (the "Ocean Dumping Act"). The document informed IMO that Planktos, opting not to fly a U.S. flag, would be able to avoid U.S. regulations. EPA with no power to regulate advised the member states of the London Convention to carefully evaluate Planktos' plans.) Following an IMO statement issued in 2007 and a report on iron fertilization drafted in 2008, a resolution was made in 2008 at the London Protocol and Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter that "ocean fertilization activities, other than legitimate scientific research... should be considered as contrary to the aims of the Convention and Protocol and do not currently qualify for any exemption from the definition of dumping" [50–52].

To assess the research and governance challenges of geoengineering, the Climate Response Fund (CRF) organized a conference of nearly 200 experts in various scientific and policy disciplines, which gathered at historically resonant Asilomar in 2010 (the site of a prior 1970s conference on genetic recombination). Created in 2009, CRF was established in order to fund geoengineering research projects and work with national and international partners to communicate information about geoengineering research (*referred to as climate intervention research*) to interested groups and the general public. Asilomar conferees set out to develop a set of voluntary guidelines, or best practices, for the least harmful and lowest risk conducting of research and field-testing [53]. The meeting brought together social and natural scientists to deliberate about geoengineering governance, in a tentative first step toward international dialogue between scientists and nonscientists on principles for future research [54, 55].

The Royal Society also maintains a strong leadership role in the continuing climate engineering discourse. Building from its 2009 report, and in partnership with the Academy of Sciences for the Developing World and Environmental Defense Fund, the Royal Society launched the SRM Governance Initiative (SRM-GI) in March 2010. It aims to develop regulatory frameworks and best practices for the research and possible deployment of SRM technology [2]. SRM-GI remains as an advisory body for international organizations and national governments engaging in the debate, bringing together scientific and policy experts to provide guidance for the conduct of research, and incorporating input from a number of civil society stakeholders.

Opponents to geoengineering have also put forth initiatives to try and prohibit or otherwise hinder its advance. The Action Group on Erosion, Technology and Concentration (ETC Group), a Canadian NGO, has led the organization of the "Hands Off Mother Earth campaign" for a moratorium on real-world geoengineering experiments or deployment (The ETC group turned down it's invitation to the Asilomar Conference on Climate Intervention Technologies and attempted to mobilize opposition against the summit. ETC Group, "Open Letter to the Climate Response Fund and the Scientific Organizing Committee," http://www.etcgroup.org/en/node/5080) [56]. This campaign lobbied the Convention on Biodiversity at its 10th Conference of Parties in 2010, contributing to a decision establishing qualifications on the scope of field tests and deployment [57]. The nonbinding resolution stated that:

> No climate-related geo-engineering activities that may affect biodiversity take place, until there is an adequate scientific basis on which to justify such activities and appropriate consideration of the associated risks for the environment and biodiversity and associated social, economic and cultural impacts, with the exception of small scale scientific research studies that would be conducted in a controlled setting in accordance with Article 3 of the Convention, and only if they are justified by the need to gather specific scientific data and are subject to a thorough prior assessment of the potential impacts on the environment [58].

Meanwhile, the Intergovernmental Panel on Climate Change in 2011 held scoping meetings for the inclusion of geoengineering in its 2013–2014 Fifth Assessment Report (AR5).

Future Directions

The attempts of a nascent landscape of governance actors to assess and regulate geoengineering (see the section on "Geoengineering Technologies and Governance Challenges") faces a complex of ethical, social, and political concerns and the diversity of potential future technological options (see the "Introduction" section). This section outlines a variety of proposed and potential governance options, as well as their capacity and adaptability to confront current and future issues. Some of these proposals – such as the SRM Governance Initiative (SRM-GI) fostering broader international dialogue and decisions at the CBD and London Convention, both discussed above – are already in play, although their longevity and impacts on governance remain uncertain. Most options remain proposals as yet unexplored in depth. Although there are many ways to categorize the range of proposals, most assessments have tended to characterize governance mechanisms primarily by the level and locale of authority [9–11, 59]. Accordingly, discussion of proposed governance frameworks here are divided into the categories of: (1) Nonstate, (2) National and Minilateral, and (3) International, or treaty-based varieties.

Nonstate Frameworks

These tend to emphasize nonbinding guidelines and other forms of soft law, and possess a degree of autonomy from government regulation. They are also intended to engage a transnational range of nonstate actors: researchers and private sector developers of technology, with varying degrees of public participation and feedback. Proposals range in regulatory strength and organizational complexity from an absence of regulation, to informal and ad hoc peer regulation, to professional or institutional codes of conduct. For the lattermost option, it has been suggested that research standards could be cohered by a network of research institutes or an international geoengineering research organization [59].

Variations of self-regulating standards, albeit with broad societal input, have traction among many early proponents of further research, and have also received support from more reserved actors that nonetheless see the need for further exploration of the technology and its attendant issues and impacts. A number of the leading scientific voices driving the development of geoengineering technologies and debate were key participants in the Royal Society's SRM-GI and Asilomar II (David Keith and John Shepherd are two examples pointed out by Banerjee [9] p20). The Royal Society's 2009 report recommends that it should itself partner with international scientific bodies to derive best practices and transparency mechanisms for research [30]. Meanwhile, Asilomar II was itself modeled on the first Asilomar meeting's normative thrust toward self-regulation [53].

Among social scientists, Victor argues that bottom-up norms and assessments formed by the collaborations of international science organizations and research groups actively engaged in the debate represent a more flexible and evolutionary governance mechanism than existing international treaties. Such an option might be "an active geoengineering research programme, possibly including trial deployments, that is highly transparent and engages a wide range of countries that might have (or seek) geoengineering capabilities." Victor adds that "[s]imilar approaches have been followed in other international scientific collaborations that have had potentially hazardous side effects, such as the European Organization for Nuclear Research (CERN) and the Human Genome Project" [59].

The benefits of such codes of conduct center around their relevance to the immediate activities of the geoengineering research community. Most notably, such codes of conduct enable responsible exploration of scientific questions that require significant research in order to guide the development of responsible governance frameworks. Moreover, as they would be driven by a more homogeneous range of actors and could potentially remain free of geopolitical motives, self-derived and imposed codes of conduct for research would likely be much easier to negotiate and swifter to implement through transnational scientific institutions. Finally, soft guidelines may have great flexibility to adapt to evolutions in the debate than rigorously negotiated international agreements [59].

There are, however, drawbacks to this approach. Depending on the extent and severity of the code, a voluntary system raises the important issue of noncompliance, particularly by actors operating outside of publically peer-reviewed scientific literature – such as private actors or clandestine government programs. Moreover, while many researchers support some form of public input, it is uncertain what effect the inclusion of more oppositional civil society groups might have on a code of principles, or on the subsequent desire of researchers and technology developers to be restricted by them. Nonetheless, if research codes forego public participation or government oversight, there may be a backlash that questions the legitimacy of the process. Codes of conduct are also most amenable to the current phase of laboratory research, and potentially – though ambiguously – in small-scale field-testing, when impacts and controversies are limited and more easily defined. However, they will almost certainly prove less effective or relevant to large-scale testing or deployment, as the entry of governmental agendas into governance may render self-regulation obsolete. Finally, there is the basic question of whether some of the actors most in favor of geoengineering should be allowed to formalize the first principles on its regulation.

National and Minilateral Frameworks

The potential for national regulatory frameworks has received the least attention in both academia and policy prescription. Hypothetically, the accepted legal mandate and enforcement capacities (within its own borders) of developed states makes governmental regulation a strong candidate for creating legally binding strictures at all stages of technology development. Besides national-level legislation or regulation, governmental legal frameworks could take a variety of forms. Within federal structures, subnational levels of government (municipal or provincial, with networks thereof) could enact legislation at a level below the state, and might feasibly network across national borders. There is also the possibility for states to cooperate on geoengineering development and regulation on a minilateral basis – a club-based approach that eschews the complications of universal participation in international frameworks.

Few of these options have been discussed in detail, likely because governments have thus far been hesitant to take strong positions on an issue whose controversies – and resulting political blowback – cannot be accurately predicted [47]. SRM-GI and Asilomar II have both noted the need for governments to scope their positions on geoengineering, and the eventual need for state-led oversight mechanisms [60]. Although no governments have formulated a clear position, the initial scoping efforts by the USA and the UK demonstrate the possibility that domestic legal frameworks will be developed. (See, for example, [61]. For example of hearing, see [62] or [63]). Besides the creation of novel legislation, Hester notes that existing national environmental and air pollution laws and agencies, such as the Clean Air Act and the Environmental Protection Agency in the USA, might be reoriented to regulate the emissions of materials upon which geoengineering initiatives depend (e.g., sulfate aerosols) [49].

A government-focused array of prescriptions could capitalize on the ability of developed states with strong bodies of environmental law and regulatory capacities to create and enforce geoengineering legislation. Such an approach would also uphold the principle of sovereignty, potentially avoiding the problems inherent in determining the extent and form of collective action. Initial actions taken by states also could serve as testing grounds for regulatory measures, societal debate, and building blocks for the negotiation of a more comprehensive framework at the international level. Even if the state proves hesitant to regulate geoengineering, action could also begin at the subnational levels in federal structures.

However, the efficacy of state regulation also relies on the strength and coherence of its own structures. While states of the OECD could legitimately enact strong rulings, it is difficult to forecast how many capacity-deficient states would be able to follow suit. Federalism may be a double-edged sword, as conflicts of interest between the national and subnational levels of government may emerge. Moreover, there are many issues buried in the logic of unilateralism, especially in the development of novel and risky technologies. If states move actively to develop and deploy geoengineering technologies without multilateral consultation, a geoengineering "race" could begin. This could in turn entrench technological know-how, with first-movers maintaining a lead on latecomers. It is possible that an incentive to act early on technology development and to maintain a technological advantage would emerge, and that contestations over intellectual property rights and ownership would follow. Finally, the uncertainty of the transboundary impacts of large-scale field-testing or deployment makes governance options that rely on the good intent of sovereign states, at the least, a controversial prospect [22, 64, 65]. (It is worth, however, noting the arguments of Horton [69], that the dangers of unilateralism are overstated, and that there are rational incentives for states to collaborate on geoengineering development and regulation.)

International Frameworks

Existing analyses of and proposals for international governance of geoengineering have tended to focus on ways existing international environmental agreements (IEAs) and organizations (IOs) might be "co-opted" to address the core governance issues. These explorations have focused particularly on IEAs and IOs whose mandates either regulate materials that are components to forms of geoengineering, or impact geoengineering more broadly.

A number of academic studies have mapped out the potential intersections of geoengineering regulation with the mandates and the governing capacities of numerous IEAs [9, 11, 20, 47, 59, 67]. There is a general consensus that while no single IEA has a direct mandate for geoengineering or could govern it in light of all its stages of development and interdependent issues. On the other hand, many studies name treaties or organizations that could assert a regulatory jurisdiction over some aspect of the technology, or some stage of the research to deployment process. For example, stipulations of the Convention on Long-Range Transboundary Air Pollution and the International Marine Organization on sulfur emissions limits might be used to regulate solar engineering testing or deployment [10, 11]. The Montreal Protocol might be called upon to fulfill a similar function should sulfate be proven to be an ozone-depleting substance. (Tilmes et al. [71] demonstrated with laboratory research that sulphates deplete the ozone layer, but corroborations are ongoing and the item has not made it onto the agenda of the Montreal Protocol.) For carbon engineering, the Law of the Sea convention might similarly be used to regulate ocean fertilization, as the London Convention and Protocol recently did [10, 11].

IOs and regimes that might address geoengineering more generally range from the UNFCCC (*as the default locus of any debate about geoengineering as a supplement to the climate issue*), to the Convention on the Prohibition of Military or Any Other Hostile Use of Environmental Modification Techniques (ENMOD), a treaty of the Cold War era that prohibits environmental modification for military or hostile purposes. (Lin [66] notes both, with a primary focus on the UNFCCC. Banerjee [8, 9] is in favor of rebooting ENMOD's mandate to ban geoengineering as a reconceptualized form of environmental modification. MacCracken [70], on the other hand, advocates modifying ENMOD to permit certain forms and amounts of geoengineering. A host of other IOs have been cited, some quite obscure: the Outer Space Treaty, the Antarctic Treaty System, etc. See [10, 11, 20].) The Convention on Biological Diversity's 2010 decision on geoengineering field-testing was a seminal event. Although of uncertain legitimacy, efficacy, or longevity, the CBD's actions may set a precedent for other IOs to make decisions on geoengineering, with varying intents and governing capacities.

Although little discussed, a new regime might be able to govern geoengineering as a holistic issue. As a blank slate, a novel mechanism could be specifically tailored to the issues and stages unique to geoengineering. However, the international arena already suffers from a glut of IOs and IEAs [68], and the creation of a new body might create jurisdictional overlaps and competition for visibility with existing ones. Moreover, the creation and implementation of a new regime with broad participation and legitimacy is a gradual process in and of itself. Combined with existing disagreements over the myriad and evolving facets of geoengineering, as well as the near absence of governmental positionings, a drawn-out time lag for creation may make it difficult for a new multilateral body to be a source of strong regulation in the near future.

The benefits of using co-opted regimes stem from leveraging frameworks with strong capacities and legitimacy, where governance – even if it targets only one facet of the geoengineering issue – might be more quickly enacted. On the other hand, a patchwork of co-opted regimes may reveal regulatory gaps that cannot be filled, or overlaps and conflicts that cannot be mediated. A multiplicity of governance forum with no key institutional home could also create an incentive for proponents or opponents of geoengineering to forum shop at the body most amenable to their interests, creating the potential for conflicting bodies of international law. Finally, existing institutions contain established mandates, organizational structures, and political logics that inevitably calcify over time. As such, they may have neither the desire nor flexibility to adjust to an issue as novel and complex as geoengineering [20].

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