USE OF FULL OXY-FUEL COMBUSTION AND ACCELERATED CARBONATION CURING FOR CARBON DIOXIDE FOR THE REDUCTION OF CARBON DIOXIDE EMISSIONS IN CEMENT MANUFACTURING

GLOBAL GOVERANCE OF GEOENGINEERING METHODS: THE MISSING PUZZLE PIECE

A Thesis Prospectus
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Bachelor of Science in Chemical Engineering

By
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On my honor as a University student, I have neither given nor received unauthorized aid on this assignment as defined by the Honor Guidelines for Thesis-Related Assignments.

Signed Camille Cooper Date: 11/2/2020

Approved: Date:
Catherine D. Baritaud, Department of Engineering and Society

Twelve years. That is how long many activists and scientists say the world has before the impending climate catastrophe hits (Wagner and Samaras, 2019). Twelve years before the Earth’s temperature reaches levels far above preindustrial levels, causing irreversible damage. Only twelve years to do something to stop this from happening. Already across the United States, environmental disasters and extreme weather events occur at higher frequency and severity (Branch and Plumer, 2020). The Intergovernmental Panel on Climate Change (IPCC) met in 2015 to create the Paris Agreement. Chief among the promises was a resolution that member countries would do what they could to limit the temperature increase to 1.5 degrees Celsius. Greenhouse gas (GHG) emissions would have to drop by about 50% by 2030 and reach net zero by approximately 2050 to achieve this goal (Wagner and Samaras, 2019, p. 1). Carbon dioxide (CO_2) is the most prevalent GHG and the most discussed in research and policy to stop climate change. According the United States Environmental Protection Agency (n.d., p. 1), 81% of U.S. GHG emissions in 2018 were CO_2. Traditional methods of climate change mitigation generally primarily call for carbon emission reductions. Examples of this are better fuel efficiency in cars, carpooling, and switching to renewable energy sources, among many others. On top of the challenges that come from changing norms on a societal or industrial level, cars are on the road for an average of twelve years, and power plants from the 1950s are still generating electricity (Wagner and Samaras, 2019). Therefore, most changes implemented now through traditional methods could take decades to create results, and the world might not have that long.

This global urgency and cultural lag have legitimized other faster and more dramatic climate change prevention methods commonly called geoengineering or climate engineering. Geoengineering is the intentional manipulation of Earth’s climate in order to fight the impacts of global warming. Within geoengineering, there are two categories (1) albedo modification

(AM), also known as solar radiation modification, and (2) carbon dioxide removal (CDR). Geoengineering techniques are highly controversial and have been for decades. AM is more divisive among experts and policymakers than CDR, and for that reason, less technology and research has been developed for AM. The technical portion of this prospectus will discuss a CDR design proposal for a cement manufacturing process that adds full oxy-fuel combustion and accelerated carbonation curing to make the process a means of carbon capture and storage. The sociotechnical portion will discuss the global governance of geoengineering methods as a means to promote safe research and innovation in the fight against global warming through the lens of Actor-Network Theory. These two topics are tightly coupled.

USE OF FULL OXY-FUEL COMBUSTION AND ACCELERATED CARBONATION CURING FOR THE REDUCTION OF CO_2 EMISSIONS IN CEMENT MANUFACTURING

Concrete is the most widely used human-made material in existence. In terms of human consumption, it is second only to water (Rodgers, 2018). Unfortunately, cement, the principal component of concrete, leads to massive CO_2 emissions; in fact, approximately 2.2 billion tons of CO_2 per year are emitted by cement manufacturing, making up 8% of all global carbon emissions (Rodgers, 2018). This amount represents the second-largest single industrial process emitter after iron and steel production (Harvey, 2018). The cement industry is a promising opportunity for industrial sustainability due to its scope, already discussed, and worth. Market projections suggest that the cement market value will grow from $312.5B in 2018 to $682.3B by 2025 (Grand View Research, 2018). To meet the standards outlined in the 2015 Paris Agreement, it will require that global carbon emissions associated with cement production drop 16% by 2030 (Rodgers, 2018).

This prospectus will propose an alternative to the clinker-making process and a production method for using concrete as a carbon capture and storage (CCS) technology. In terms of cement produced, the de-carbonation of limestone in the kiln produces about 525 kg CO_2 per ton of clinker, and the fuel combustion produces about 335 kg CO_2 per ton of cement (Bosoaga et al., 2009). Over the last few decades, many advances have been made regarding kiln and feedstock design to make the process more environmentally-friendly. However, only recent studies have explored the possibility of CCS and cement's role in this environmentally-sustainable technology. The resulting designs of these studies face adverse market conditions. An economically-viable design will be necessary to garner the support needed to make a substantial shift toward sustainable cement production.

BACKROUND ON ORDINARY PORTLAND CEMENT MANUFACTURING

Ordinary Portland Cement (OPC) is the industry standard cement. The following description of the process commonly used to manufacture OPC comes from "How cement is made" (n.d.). OPC is produced when limestone, or another source of calcium carbonate, is crushed and mixed with other ingredients, such as clay, shale, or slag, and subsequently heated in a cylindrical, rotary kiln. The kiln reaches temperatures as high as 2,700 degrees Fahrenheit. The solid ingredients have a chance to thoroughly mix and incorporate as they move from the top of the kiln downward, exposed to even heating. The product of this kiln firing are small and circular pellets, called clinkers. The following equation describes this reaction:

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$$

These hot molten pellets are sent through multiple coolers until they reach a cold enough temperature to be handled by humans. The clinkers are then ground and combined with small amounts of gypsum and limestone. After this mixing process, water can be added to the mixture to form concrete. This process is outlined below in Figure 1:

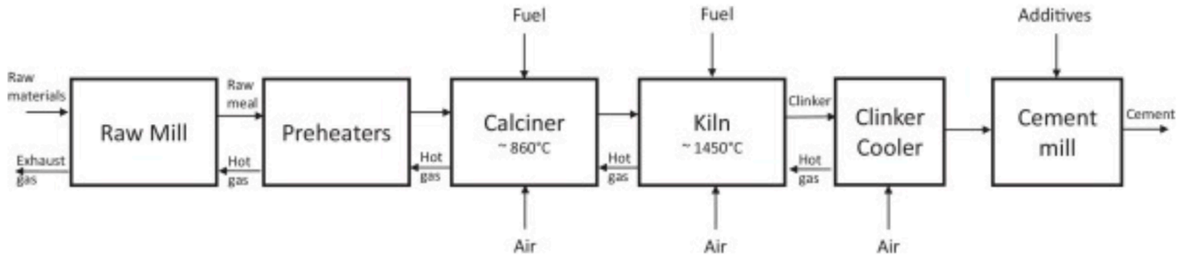

Figure 1. OPC manufacturing process: A simplified flow diagram for OPC concrete production. (Carrasco-Maldonado et al., 2016).

OBTAINING CO_2: THE ROLE OF OXY-FUEL COMBUSTION

Oxy-fuel combustion is a CCS technology. The following paragraph describes the oxy-fuel combustion process as it is described in “Oxy-fuel combustion systems” (n.d.). The process begins with an air separator, which produces a pure O_2 feed to the combustion process. The oxygen is then fed, in conjunction with a hydrocarbon fuel, to the boiler which facilitates combustion. CO_2 and H_2O are the main products of this process and are taken off as flue gas. The stream then splits into two, one which recycles part of the flue gas back into the boiler, and one which leads to cooling, compressing, and dehydrating the stream to produce pure CO_2. Figure 2 shows how this process works in cement manufacturing.

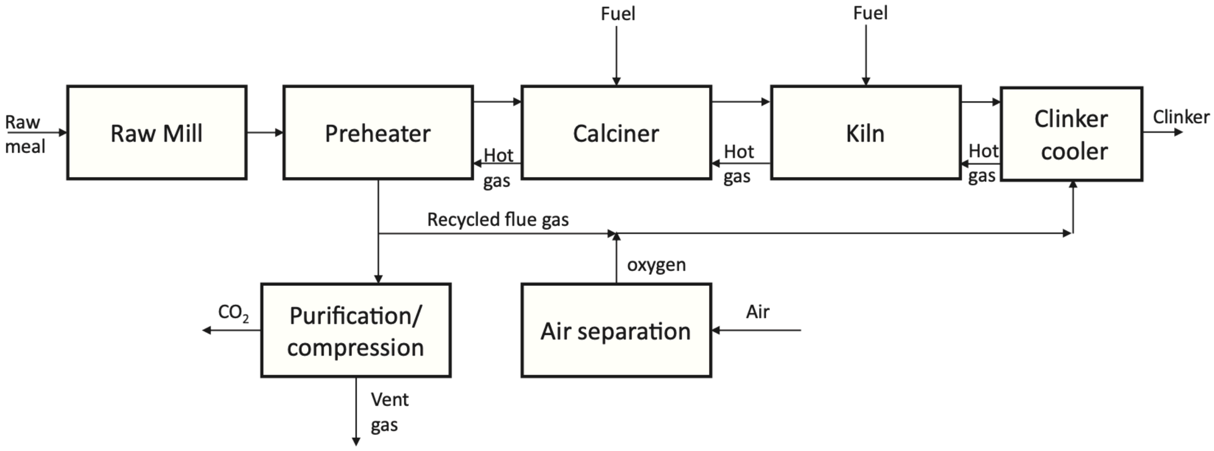

Figure 2. Full oxy-fuel configuration layout: Modifications that must be done to the OPC process as seen in Figure 1 to achieve the full oxy-fuel configuration. (Carrasco-Maldonado et al., 2016).

Part of what sets oxy-fuel combustion apart from other CCS technologies is that the process utilizes a pure O_2 stream and recycles flue gas back into the combustion process. This is as opposed to feeding air into the process, which presents inert nitrogen to dilute the O_2 in the boiler. Using flue gas to dilute oxygen instead allows for higher conversion to the products. This process is also one of the most viable and efficient CCS technologies available today.

CARBON STORAGE: IMPLEMENTING ACCELERATED CARBONATION CURING

Carbonation naturally occurs over time when cement paste and atmospheric CO_2 meet. This reaction is dictated by the following equation:

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$$

However, this process can be accelerated for precast concrete to significantly increase carbon storage capacity, minimizing the cement-making process' overall carbon footprint (Zhang and Shao, 2016). The following description of the accelerated carbonation process comes from Zhang and Shao (2016). Accelerated carbonation occurs when CO_2 gas diffuses into the porous concrete. The solvation of gaseous CO_2 occurs before it is hydrated to H_2CO_3. Once H_2CO_3 forms it is then ionized. The newly formed H^+ ions cause the pH of the concrete to decrease. Finally, the nucleation of the $CaCO_3$ occurs, and C-S-H gel forms. Instead of hydrating, CO_2 could instead react with silicate phases, dicalcium silicate and tricalcium silicate, to dissolve exothermically and form stable calcium carbonates. The $CaCO_3$ then precipitates out of the solution. Figure 3 shows the proposed carbonation curing procedure for wet-mix cement.

Figure 3. Process of precast carbonation curing: Carbonation curing procedure for wet-mix concrete. (Adapted by Cooper from Zhang and Shao, 2016).

BARRIERS TO IMPLEMENTATION

Barriers to implementing a CCS process in concrete production are mostly in the oxy-fuel combustion component. High combustion temperatures and costs associated with using oxy-fuel combustion - more fuel, energy to increase temperature - are issues associated with our design that should be considered. The introduction of a pure O_2 stream to the kiln, instead of an air stream, also increases the partial pressure of CO_2 in the kiln, and changes the reaction from a gradual to a threshold reaction, threatening re-carbonation of lime to limestone, and making the reaction much fickler (Zeman, 2009).

THE TEAM'S PLAN

The team consists of chemical engineers Camille Cooper, David Reed, Sarah Gill, and Nirasha Abeysekera. The team plans to use AspenPlus to simulate the process. This will come together in a fashion similar to the process flow diagram in Figure 4. This process flow diagram will be built upon to properly model the entire process including the oxy-fuel combustion, CCS, and various coolers beyond the kiln. For this diagram, the rotary kiln is represented by the combination of a reactor and cyclone solid-gas separator. The team will focus on adding oxy-fuel combustion to the kiln, and creating a carbon capture stream off the kiln.

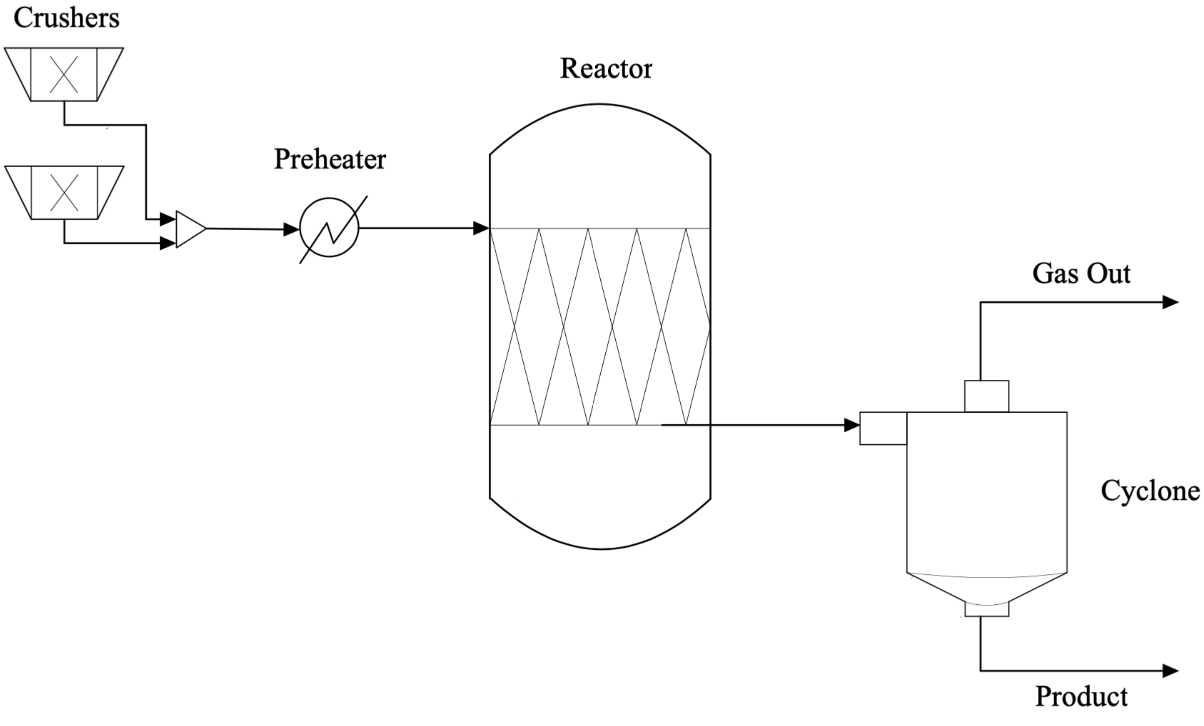

Figure 4. Proposed process flow diagram: The layout of unit processes that will be used for the AspenPlus simulation (Cooper, 2020).

The process begins with the formation of OPC. Then, oxy-fuel combustion will be used in the kiln to provide pure CO_2 for carbon capture. This specific kiln set-up is known as a

Reduced Emission Oxygen (REO) kiln, according to Zeman (2009), whose work will be used to guide the team in the use of oxy-fuel combustion in the kiln. In addition, a study on oxy-fuel combustion by Ditaranto and Bakken (2019), which utilizes a scale of 3000 tons of clinker production per year, and a kiln of 60 m long with an inner diameter of 3.76 m, which can be revised and scaled up in later calculations by the team, will be used. CO_2 from the REO kiln will be taken off as a stream fed to the concrete, with excess CO_2 from contact with the concrete led out of the plant and to an alternative method of CCS, such as geologic formation storage.

In terms of process variables for this technical project, the team hopes to experiment with cement composition including limestone, fly ash, etc., clinker quantity which depends on kiln size, and CO_2 exposure time, as well as kiln temperature and kiln fuel, while monitoring the percent recovery of CO_2 in the recycle stream and percent of CO_2 absorbed by the concrete. The team will assume that the factory where the cement is manufactured generates precast concrete slabs so that CO_2 exposure can be manipulated in the designed plant.

This project will be worked on over the course of two semesters in CHE 4438/4476 under the advising of professor Eric Anderson of Department of the Chemical Engineering. The team will meet once a week at minimum to discuss advances made and establish new goals for the following week of research, modeling, and analysis. Every three weeks there will be a review and presentation to professor Anderson. The team has also recently discovered companies, for example, Central Concrete and Solidia Technologies, that are producing carbonated concrete using similar, though not fully identical, methods. The team plans to reach out to a representative from these companies to see if they can establish a partnership for the Spring 2021 semester. The end result will be a phase I design package and decision. The team hopes to show that 20% carbon reduction is feasible and economical.

GLOBAL GOVERANCE OF GEOENGINEERING METHODS: THE MISSING PUZZLE PIECE

Humanity is quickly approaching a dangerous slope into a world-wide apocalyptic environmental disaster, at least from the perspective of human adaptability. Traditional climate change mitigation has been slow to adoption and slow to produce results. Many experts fear that, when global leaders at last recognize the danger at hand, they will have no choice but to implement engineering projects whose risks have not been thoroughly researched and could bring worse consequences than the problem they fix (Anderson, 2017). In fact, calling climate change and global warming *emergencies* or *crises* has been criticized as these framings could be abused to rationalize setting aside democratic decision making for unilateral, radical attempts at fixes (Gupta et al., 2020).

Currently, geoengineering, especially albedo modification (AM), is considered highly risky, so risky that some believe even its research is unethical (Gertner, 2017). Admittedly, geoengineering has only been explored in small field experiments; therefore, its methods are underdeveloped, and its risks and effectiveness are both inadequately understood (Reynolds and Parson, 2020). This should change as the abilities and limitation of the technology need to be understood before the technology must be implemented in political desperation (Blackstock, 2012). Moving forward, regulation and oversight is necessary as the few geoengineering experiments in the works are funded by philanthropists without public accountability or checks and balances (Anderson, 2017).

Actor-Network Theory (ANT) was proposed in the 1980s and is used to investigate the relationships surrounding a technological advancement. The *network* is composed of a heterogeneous incorporation of textual, conceptual, social, and technical *actors* ("Actor Network Theory," 2004). These *actors* can be human and non-human artefacts that have the capacity to act on other actors and or be acted upon themselves (“Actor Network Theory,” 2004). In this case of geoengineering, ANT will be used to investigate how global governance agencies play a critical role in the safe development and implementation of geoengineering techniques.

PROS AND CONS OF ALBEDO MODIFICATION

The Intergovernmental Panel on Climate Change (IPCC) has conceded that solar geoengineering could limit warming to below 1.5 degrees Celsius (Reynolds and Parson, 2020). This is an impressive feat for a single technology, which fuels the fears of many of its opponents that reliance, or even any extent of use, of AM will dissuade many from exerting effort to lower GHG emissions, committing to sustainable practices, and other more acceptable and established ways of climate change mitigation (Robock et al., 2008). In fact, AM is well positioned to be practiced concurrently with mitigation, adaption, and CDR not to replace these methods (Reynolds and Parson, 2020; Anderson, 2017). That being said, a real risk comes from the fact that once begun AM must be sustained else a rapid reversal of any temperature decrease will occur (Reynolds and Parson, 2020). Future, large-scale deployment of AM without “competent, prudent, and legitimate international control could trigger international destabilization or conflict” (Reynolds and Parson, 2020, p. 1). This is due to the unilateral nature of AM, for only $1 billion each year, any party with access to high altitude aircraft can implement AM (Gertner, 2017). However, the further danger in this arises when geopolitical calculations factor this decision not technical assessments (Blackstock, 2012). Gunderson et al. (2018, p. 1) states that AM upholds powerful economic interests and protects “an inherently ecologically harmful social formation.” It is an answer to a lazy, capitalistic society that does not want to implement major changes to their life and treats nature like “passive resources” for human satisfaction. The fact is that climate change affects socioeconomic and geographic demographics differently. Branch and

Plumer (2020) point out that investors are more likely to invest based on where the crisis is most obvious and who is getting effected; more urgency with funding research would occur if Washington D. C. was hit not Oregon. In the end, politics cannot be avoided, and climate negotiators and political leaders need to develop strategies to fill the governance gap (Blackstock, 2012).

CURRENT STATUS OF GEOENGINEERING

Global governance is linked to responsible innovation (Jinnah et al., 2018). However, state actors have yet to become involved in the debate over geoengineering (Reynolds and Parson, 2020). Field trials are already underway in countries like the United States, so the state parties do not have much more time to act, if they wish to do so proactively (Jinnah et al., 2019). Some argue that scale-up of geoengineering experiments require state oversight (Reynolds and Parson, 2020). A field trial was canceled because of lack of rules on how to proceed (Anderson, 2017). Another highly publicized field trial, the Stratospheric Particle Injection for Climate Change field trial, was cancelled over a patenting problem that arose from corporate ownership taking precedence over global public interest (Blackstock, 2012). The U.N. tried to create the first global governance regulation to greenlight investigatory research into geoengineering, but the U.S. and Saudi Arabia blocked the efforts (Watts, 2019). State oversight, regulation, and enforcement has been proven to be successful in environmental protection before. The 1987 Montreal Protocol is a great example of international governmental cooperation. Today, 99% of manufactured ozone-depleting substances have been phased out, chlorine and bromine stratospheric levels are decreasing, and scientists are reporting the ozone layer healing (Anderson, 2017).

Anticipatory governance should be employed in order to provide structure to research that is already underway as a flexible decision framework. State regulation can be enabling not just restrictive. Figure 5 categorizes four rationale behind state governance of AM, specifically solar geoengineering (SGE). The spectrum on which each rationale group is placed implies a relationship between the freedom associated with the given rationale for governing and the threat of a future with and without SGE. Each of these rationales is useful; however, *govern to exercise oversight* is the most needed in this situation. However, further research may prove this incorrect. The state actors need to carefully discuss each of these rationales to decide what is best for their public. The International Academic Working Group on Climate Engineering Governance stated in their final consensus report that establishing legitimate deliberation bodies, leveraging existing institutions, and making research transparent and accountable is the way society must move forward with geoengineering (Gupta et al., 2020).

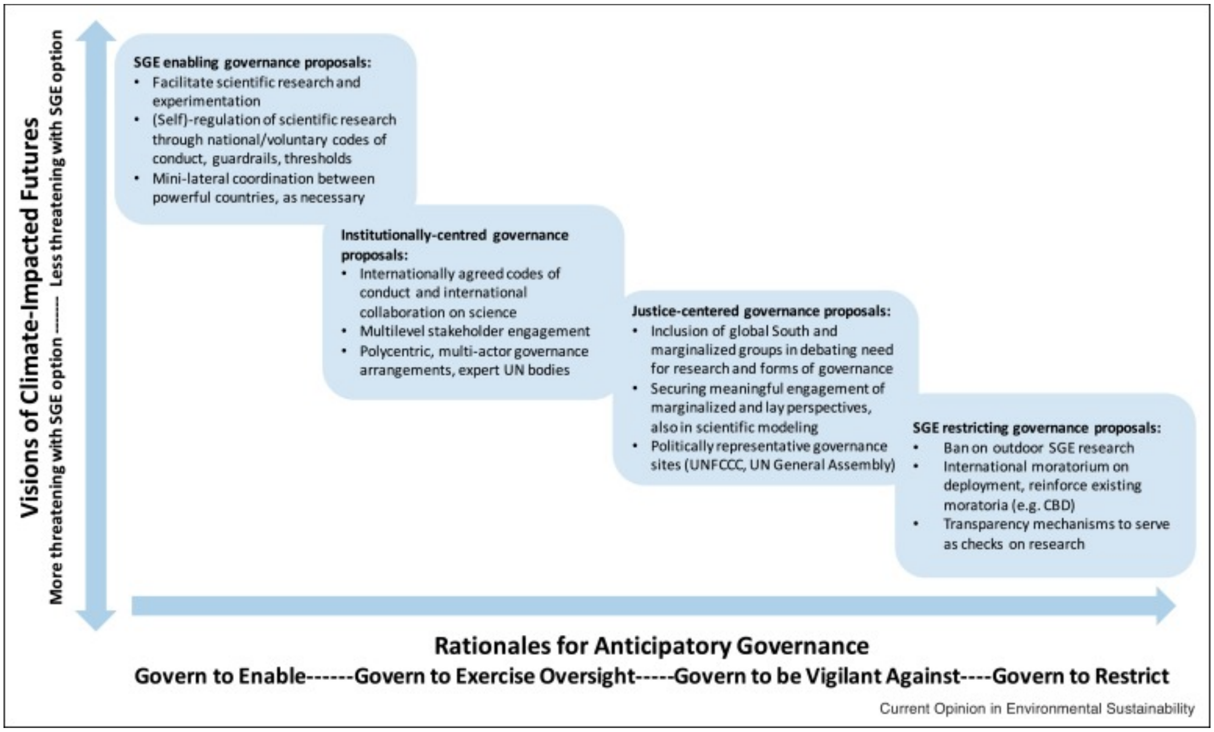

Figure 5. Anticipatory governance of solar geoengineering: Why govern? Four roles that anticipatory governance can assume in the regulation of albedo modification techniques. (Gupta et al., 2020).

ACTOR NETWORK THEORY AND GEOENGINEERING: THE FUTURE

The wider actor network of AM consists of scientists, investors, regulators, politicians, protestors, and major carbon emitting nations. AM policy, as opposed to the technology itself, also matters highly to the energy and agriculture industries and the public (Blackstock, 2012). Currently, these parties do not effectively communicate with each other nor does there exist an established and recognized platform for discussion. There are a handful of conferences and initiatives launched by researchers that target geoengineering, such as the Solar Radiation Management Governance Initiative launched by the Royal Society in London and the Asilomar Conference in California in 2010. These parties attempted to craft recommendations for geoengineering policy. More than researchers need to act. Once AM reaches the implementation stage, all the aforementioned parties must be acting in harmony; however, at the current research stage the task at hand can be expedited by focusing on researchers, investors, and regulatory agencies. Regulatory agencies include state and non-state parties and are grouped together for these purposes as the distinction between the two is not significant at the current stage of geoengineering innovation. Figure 6 shows the proposed actor network for the continued and improved research of AM and other geoengineering endeavors. The figure shows that all parties need to be in mutual communication with the others in order for AM to be successful. The primary difference of this proposed network from that which currently exists is the involvement of regulatory agencies, who should take inspiration from the Montreal Protocol in establishing new regulation and norms for geoengineering across the world.

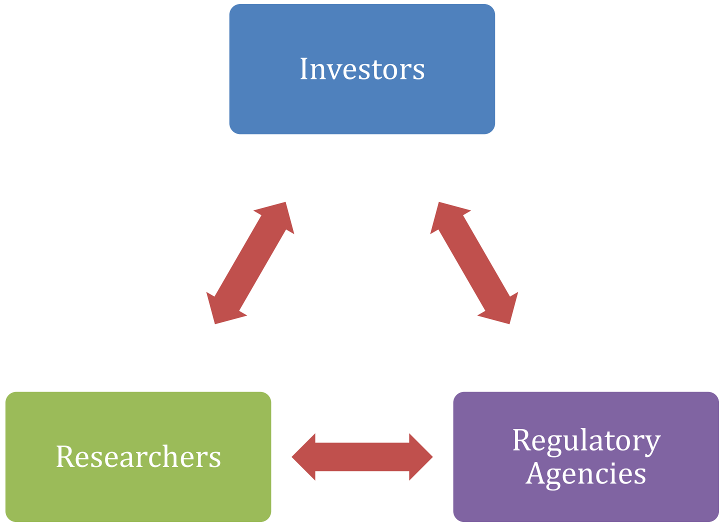

Figure 6. Proposed actor network: The actor network for expedited research of albedo modification. The three parties that need to communicate effectively for successful research. (Cooper, 2020).

Geoengineering researchers need to create guidelines that can become a practical regulatory framework, which will allow for research to proceed more effectively. These guidelines need to be communicated and accepted by fellow researchers, regulatory agencies, and investors. Researchers rely on investors to fund the research. The investors depend on the researchers to use the money effectively. A registry of ongoing and planned research could aid in this communication (Blackstock, 2012). Investors are able to establish norms of international cooperation before those norms are set by international regulatory groups, and if they create incentives on collaboration on research that will responsibly oversee and monitor the controversial projects (Blackstock, 2012). Regulatory agencies will be necessary to ensure that the research is being conducted ethically and that all parties are held accountable. Geoengineering innovation can only continue if regulatory parties begin to work cohesively with researchers and investors. They are the missing puzzle piece necessary for the smooth course of innovation of geoengineering.

THE PLAN

This sociotechnical research paper will be written during the Spring of 2021 during STS 4600. Chemical engineer Camille Cooper will undergo this part of the thesis independently. The next step after turning in this prospectus is presenting an oral presentation to her STS 4500 class on November 19th. The final thesis will be turned in the first week of April. In between these dates are deadlines for an executive summary, abstract, and more.

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