The Carbon Problem

The "carbon problem" refers to the ongoing increase in atmospheric concentrations of the greenhouse gas carbon dioxide (CO₂) observed over the last two centuries. This increase is being driven almost entirely by anthropogenic emissions, with most of the emissions associated with combustion of fossil fuels. If humankind decides, at some point, to reduce significantly the anthropogenic emission rate, new or different technologies will almost certainly play a central role. In this book, we are interested in one specific technology: carbon capture and storage (CCS), wherein the CO₂ produced from the use of fossil fuels is captured at large stationary sources like power plants and is stored somewhere other than the atmosphere. We are specifically interested in the storage part of this operation, and even more specifically, in geological storage, where the captured CO₂ is injected into appropriate geological formations deep underground. Proper analysis of the operations and possible consequences of this kind of injection requires careful mathematical and computational models to predict the system behavior. We focus on such models in this book.

1.1 BACKGROUND

The concentration of CO_2 in the atmosphere is naturally dynamic. Figure 1.1 shows the so-called Keeling curve, named after Charles Keeling, who initiated a program of ongoing measurements of atmospheric CO_2 in the 1950s. These data show annual cycles of variability superimposed on a monotonic increase over the half century of measurements. Atmospheric concentration of CO_2 in the late 1950s was around 315 parts per million (ppm), while today's concentration has grown to about 390 ppm. To put these numbers into historical context, consider the data shown in Figure 1.2. There the Keeling data, measured at Mauna Loa, Hawaii, are combined with data from ice cores to show atmospheric concentration of CO_2 over the last 1000 years. These data show a stable atmospheric concentration of about 280 ppm, which is the concentration to which the atmosphere stabilized at the end of the last ice age. The increase above 280 ppm began with the industrial revolution and has accelerated continuously to the present day. The range of values shown in Figure 1.2, between

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Figure 1.1 Atmospheric carbon dioxide as a function of time, measured at Mauna Loa (modified from wikipedia.org/wiki/Keeling_curve).



Figure 1.2 Atmospheric carbon dioxide as a function of time, over the past 1000 years. Also shown is the range of CO_2 concentrations measured over the past 650,000 years (modified from http://www.britannica. com/eb/art-69345/Carbon-dioxide-concentrations-in-Earths-atmosphere-plotted-over-the-past).

a low of about 170 ppm and a high of about 300 ppm, indicates the maximum and minimum values of atmospheric CO_2 concentration seen in ice core data over the last 650,000 years. In those ice core records, clear 100,000-year cycles of glacial–interglacial periods can be seen, with corresponding maximum and minimum values of atmospheric CO_2 . From these data, we conclude that the current concentration is about 100 ppm above the "natural" equilibrium associated with the current interglacial period. We also conclude that the current value of 390 ppm is larger, by about



30%, than the highest value seen in at least the last 650,000 years. As such, we humans are collectively performing an interesting global-scale experiment to see how the earth system will respond to significant increases of an important greenhouse gas. The consensus expectation is that these increases will lead to dangerous climate change unless they are reduced or reversed.

In order to understand the problem, it is helpful to identify the specific sources of anthropogenic CO₂ associated with combustion of fossil fuels. Figure 1.3 shows global estimates for the different sources of CO₂ emissions, indicating the fraction of total fossil fuel-related emissions coming from each major sector. The dominant source of CO₂ emissions is electricity generation, accounting for approximately 40% of emissions, followed by transportation at slightly more than 20%. A recent estimate for total annual anthropogenic emissions (for calendar year 2008) is between 8 and 9 gigatonnes (Gt) of carbon (where 1 Gt C = 1×10^{15} g of carbon = 1×10^{9} metric tones of carbon). The molecular weight of carbon is 12, while the weight of a CO₂ molecule is 44; therefore, the conversion factor from carbon to CO₂ is 3.67, which means that the global annual anthropogenic emission rate measured in mass of CO₂ is about 30 Gt CO₂/year. In the remainder of this section we will use 8 Gt C/year as the estimated current emission rate.

Given this profile of emissions, it seems logical that any successful strategy for carbon mitigation will involve decarbonization of electricity generation coupled with associated strategies that may include the use of decarbonized electricity in the remaining sectors. For example, one can consider electrification of the transportation sector and modified designs for both residential and commercial buildings to take advantage of carbon-free electricity. Overall, development of effective solutions to the carbon problem constitutes a grand challenge for the early 21st century.

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1.2 STABILIZATION WEDGES AND TECHNICAL SOLUTIONS

To understand the overall size of the problem and the scales of effort needed to solve it, specific units of measure have been introduced. One that is particularly useful is the so-called stabilization wedge. In their seminal paper from 2004, Pacala and Socolow introduced the concept of stabilization wedges. A stabilization wedge is a unit of measure corresponding to avoidance of emission of 25 Gt C over the next 50 years. The concept arises when a typical "business-as-usual" scenario for future emissions is compared to a so-called "flat path," where emissions are held constant at their current rate for the next 50 years. With business as usual corresponding to doubling of the current emission rate over the next 50 years, the flat path implies that instead of increasing emission rates to 16Gt C/year from the current 8Gt C/ year, the rate would instead be held constant at 8 Gt C/year. The concept is shown in Figure 1.4, which is modified from the original figure of Pacala and Socolow (2004), which used the then-current emission rate of 7 Gt/year. We see that the difference of 8 Gt C/year after 50 years can be broken into 8 "slices," where each slice, or "wedge," corresponds to emissions avoidance that increases linearly over 50 years to a value of 1 Gt C/year after 50 years. This "slice," or "wedge," is indicated on Figure 1.4. Among many other things, Pacala and Socolow (2004) considered a variety of existing technologies and estimated the effort needed to implement those technologies in order to achieve one wedge. A partial list of available technologies, and the associated effort to achieve one wedge worth of emissions avoidance, is given in Table 1.1 (this has also been updated from the original 2004 publication).



Figure 1.4 Stabilization wedges, modified from the original presented in Pacala and Socolow (2004), which used a "current" emission rate of 7 Gt C/year.

Technology	Effort required to achieve one wedge
Automobile efficiency	Increase fuel efficiency from 30 mpg to 60 mpg for 2 billion cars (currently there are fewer than 1 billion cars worldwide).
Nuclear power	Add twice the currently installed nuclear capacity.
Solar power	Add 350 times the currently installed solar power capacity.
Wind power	Add 15 times the currently installed capacity of wind- generated electricity.
Carbon capture and storage	Install carbon capture and storage at 800 large-scale coal-fired power plants.

 Table 1.1
 Examples of Technologies and Associated Efforts to Achieve One Wedge (Updated from Pacala and Socolow 2004)

Notable technologies include nuclear power, which requires addition of twice the current global installed capacity to achieve one wedge; solar photovoltaics, which require about 350 times the global installed capacity (as of 2008) to achieve one wedge; automobile efficiency, which requires 2 billion cars to increase fuel efficiency from 30 miles per gallon (mpg) to 60 mpg (there are currently about 650 million cars worldwide); and installation of a technology called CCS at about 800 large-scale coal-fired power plants. Based on these calculations, Pacala and Socolow (2004) made three important points. The first is that several current technologies exist that can produce at least one wedge of emissions avoidance. The second is that each technology requires an enormous effort to reach one wedge. And the third is that none of the technologies is capable of producing the entire eight wedges needed to achieve the flat path for future emissions. As such, a portfolio of technologies must be used if the carbon problem is to be solved.

When considering energy portfolios of the countries that are the largest emitters, coal plays a central role. Abundant and cheap coal can be found in both the United States and China, which means that coal-fired electricity is likely to be a significant part of the energy portfolio in each country for at least the next several decades. This makes decarbonization of the power grid a problem of use of coal without the CO_2 emissions. The only currently available technology to achieve this outcome is CCS. To see the potential impact of CCS technology, we can consider that there are currently about 2100 large coal-fired power plants worldwide. Furthermore, the current rate of construction of new plants in China alone is widely estimated at between one and two plants per week. Both of these numbers imply that a full-scale implementation of CCS coupled with coal has the possibility to contribute several wedges to the overall solution. And, as the Massachusetts Institute of Technology (MIT 2007) report on the future of coal concluded in 2007, "carbon capture and sequestration (CCS) is the critical enabling technology that would reduce CO₂ emissions significantly while also allowing coal to meet the world's pressing energy needs."

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1.3 CCS

As a possible large-scale mitigation strategy for the atmospheric carbon problem, CCS has emerged as a serious option. The concept of CCS is simple: capture the CO₂ produced when the chemical energy in fossil fuels is converted to electrical energy, and sequester the captured carbon somewhere other than the atmosphere. The most likely location for large-scale sequestration is in deep geological formations, where the CO₂ would be injected into formations that are (1) sufficiently permeable to accept large quantities of CO2 and (2) overlain by very low-permeability formations that will keep the injected buoyant CO2 in place. Deep sedimentary basins are the likely target for large-scale CO₂ injections, using some combination of depleted oil and gas reservoirs (with or without enhanced oil recovery), unminable coal seams, and deep saline aquifers (see Figure 1.5). The recent Intergovernmental Panel on Climate Change (IPCC)'s special report on CCS provides many details about the overall concept and the technological approaches for its possible implementation. The report presents data showing that overall global storage capacity, in terms of total amounts of CO₂ that can be stored underground, appears to be more than adequate, with the largest capacity associated with deep saline aquifers. Figure 1.6, taken from the IPCC report, identifies promising areas for CO_2 storage across the globe.

The concept of CCS involves capture of the CO_2 prior to release to the atmosphere and storage (or sequestration) away from the atmosphere. In principle, technology exists to achieve both of these steps. Capture can be achieved through separation of the CO_2 from the dilute gaseous waste stream of traditional power plants. Or it can be achieved through the use of different designs to process the fuel and produce the electricity, for example, splitting the hydrogen and carbon in the



Figure 1.5 Different kinds of geological formations suitable for geological storage of CO₂ (image adapted from IPCC 2005).



Data quality and availability vary among regions. **Figure 1.6** Prospective areas for geological storage of CO₂ (from IPCC 2005 report).

fossil fuel hydrocarbon early in the process and using the hydrogen as the energy carrier while producing an essentially pure stream of CO_2 . This latter concept is the basis of the so-called integrated gasification combined cycle (IGCC) designs. The limiting factor in carbon capture at this time is cost, with 80–90% of the cost of CCS being associated with the capture process. While estimates still vary significantly, if a traditional coal-fired power plant were replaced by a CCS power plant, the added cost to consumers would be about 5 cents (U.S.)/kWh. Recognizing that implementation of large-scale CCS operations on many power plants will take several decades to implement, the annual cost increases would be a small fraction of 1 cent/kWh.

Once the CO_2 has been captured, it must be kept out of the atmosphere. In geological storage, the captured CO_2 is compressed and injected into deep geological formations. Almost all strategies considered to date involve injection at sufficient depth so that both the pressure and temperature exceed the critical point of CO_2 : 7.4 mega-Pascal (MPa) and 31.1°Celsius. Supercritical CO_2 is significantly denser than gaseous CO_2 . Therefore, the same amount of mass occupies a much smaller volume and has a concomitantly weaker buoyant drive. For typical geothermal gradients, CO_2 is supercritical at depths below about 800 m.

One of the main advantages of CCS is that all of the needed technology already exists. Therefore, large-scale implementation is not limited by the need for new technological developments, although better capture technologies would improve efficiencies and lower cost, and new technologies may be needed for longer-term monitoring of the injection system. In addition, the oil industry and the waste disposal industry have extensive experience with injection of both gases and liquids

BOX 1.1 Existing CO₂ Injection Operations

Carbon dioxide has been injected into the subsurface for more than 25 years as a means of enhancing oil recovery (EOR). Because the objective of EOR operations is to maximize profit rather than reduce carbon emissions, the CO_2 is usually not taken from industrial sources, but rather from natural CO_2 reservoirs because it is currently a cheaper and more reliable option.

Commercial carbon injection for the purpose of emissions reduction requires economic incentives that are just beginning to be developed. An early mover was the Norwegian government, which instituted a tax for carbon emissions from the offshore petroleum industry. One target of this tax is CO_2 that is separated from natural gas before it is sold to the market. Consequently, in the mid-1990s, the Norwegian oil company Statoil found it economically favorable to capture and inject the CO_2 associated with producing the Sleipner gas field rather than vent that CO_2 to the atmosphere. The captured CO_2 was injected into the shallower Utsira formation, as shown in the schematic in Figure 1.A. This storage operation came online in 1996, and at the time of writing represents 14 years of operational experience with an injection rate of about 1 million tons per year through a single well. The Sleipner storage project has been closely monitored with a series of seismic measurements, which have provided large amounts of data on CO_2 migration within the formation.

A second commercial-scale ongoing storage operation is being conducted by BP and their partners at their In Salah gas field in Algeria. Since 2004, close to one million tons have been injected annually into a water-filled portion of the formation from which gas is produced.

It is worth noting that both the Sleipner and In Salah operations are situated in geographically challenging locations: Sleipner is in the middle of the North Sea, while In Salah is in the Sahara Desert. Still, the technological challenges associated with injec-



Figure 1.A Schematic of the Sleipner injection (courtesy of Statoil ASA).

tion have been overcome. The major challenge is therefore not in the technical implementation, but in our understanding of the subsurface system and our ability to predict the fate of injected CO_2 , especially if CCS is implemented at a scale that is significant (at least one wedge) relative to solving the carbon problem.

into deep subsurface formations. This includes injection of significant quantities of liquid hazardous wastes in the United States and so-called "acid gas" (a mixture of CO_2 and hydrogen sulfide $[H_2S]$) in Canada. However, there are other issues that have slowed the implementation of large-scale CCS projects. These include the lack of a regulatory framework within which large-scale injection operations would be permitted, the lack of an economic system where the cost of emitting CO_2 to the atmosphere is internalized within an individual operator such as a power plant, and the lack of international agreements that would lead to effective global solutions.

When both regulations and economic credit systems are considered, a common question focuses on the fate of the injected CO_2 , and more generally, the overall effects of large-scale injection on the subsurface system. If a significant amount of the injected CO_2 leaks out of the formation into which it is injected, then the operation may be viewed as a failure from an environmental, regulatory, and economic–credit perspective. A broader analysis of the overall system shows that leakage of either the injected CO_2 , or of brine that is displaced by the injected CO_2 , is the major issue associated with geological storage of CO_2 . Therefore, a comprehensive analysis of the overall subsurface system is required, including the movement and ultimate fate of the CO_2 and other subsurface fluids affected by the injection operation.

The complete physical description of the subsurface system involves the physics of multiphase fluid flow in porous media, phase behavior of the fluids involved, heat and energy transport, geomechanics, and strongly nonlinear geochemical reactions. When all of these processes are included, the problem can become very complex, and different kinds of numerical and computational challenges arise. Engineering judgments about when certain simplifying assumptions can be made, their implications, and their limitations, lead to many interesting questions about underlying physics, the nature of mathematical (and computational) modeling, and the overall engineering and scientific approaches that are most appropriate for a given set of problems. In order to make these kinds of decisions about physics and mathematics, an understanding of the subsurface environment is necessary. We now present an introduction to the fascinating environment of the earth's subsurface.

1.4 THE SUBSURFACE SYSTEM

To understand geological storage and develop mathematical models to describe the movement of fluids in the subsurface, including injected CO_2 , we need to have some understanding of the subsurface environment. If we were to start at the land surface

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and begin to move downward, we would first encounter soil, which is familiar to everyone. Soil is composed of different kinds of solid particles, collectively forming the solid part of the soil. These solid particles are not bound together, but rather are loose, and are referred to as *unconsolidated*. Between the solid particles are spaces filled with one or more fluids; for soils, these fluids are usually air and water. The space filled with fluids is referred to as the *pore space* or the *void space*. Our interest is primarily in the movement of fluids within the pore space.

As we continue further downward in the shallow soil zone, the mineral composition of the solid particles will typically change, with less organic material at greater depths. At some point, we will begin to notice that more of the pore space is filled with water, and eventually, we will reach a zone where all of the pores are filled with water. This is referred to as the *saturated zone*, while the zone above it, in which water and air coexist, is the unsaturated zone. As we proceed deeper, we may encounter layers of different kinds of materials, indicative of the particular geological history of the area. In areas of sediment deposition, these layers may be a few meters to tens of meters to perhaps hundreds of meters thick. In coastal areas like the coastal plain of New Jersey, USA, we can see clear layered structures with the layering alternating between layers of fine-grained, clay-rich materials and layers of coarser-grained sand deposits. Figure 1.7 shows a cross section of the New Jersey coastal plain, extending from Philadelphia, PA to Atlantic City, NJ. Two comments are of interest at this point. First, sandy materials tend to be much more permeable than clays. This is because the pores in coarse-grained sands are much larger than those in clays, and the permeability of the material depends strongly on the pore sizes. The higher permeability means that water can flow much more easily through the sand formations than through the clays. Water is often extracted from these kinds of sandy formations through pumping wells, thereby using so-called groundwater



Figure 1.7 Sedimentary structure shown for a cross section of the New Jersey coastal plain (modified from Epstein 1987).

as a water supply. Groundwater supplies several billion people worldwide with water and is a vital resource. The second point to be made is that in Figure 1.7, the formations appear to have a significant slope toward the Atlantic Ocean. It is important to realize that this is a distortion of the true situation, because the scale on the vertical axis has been expanded, or exaggerated, by a factor of about 50 relative to the horizontal scale. If these were plotted with equal scales in the horizontal and vertical, the formations would be seen more properly as very thin, almost flat structures. It is a good idea to remember this vertical exaggeration and to interpret various images accordingly. It is also important to remember that the actual structures do indeed have much greater horizontal extent than vertical, and they commonly have slopes less than 1%.

Formations that contain freshwater and have permeability high enough to yield water in significant quantities for human use are referred to as *aquifers*. The lower permeability layers, like the clay layers in Figure 1.7, are often referred to as *confining layers* or *aquitards*. Aquitards do not allow water extraction in significant quantities, but they have enough permeability to allow for slow leakage of water across the formation, thereby providing hydraulic connection between aquifers immediately above and below the confining layer.

As we move deeper into the subsurface, we will often see similar layered structures. At depth, the rock tends to be *consolidated* rather than unconsolidated, but patterns of relatively permeable formations alternating with layers of very low permeability are often seen. A good example is the Alberta Basin, whose general vertical structure is characterized by alternating permeable (aquifer) and relatively impermeable (aquitard) formations. In deeper zones, the low-permeability layers are often referred to as *caprock formations*. Also, within the deeper formations, we encounter elevated pressures and temperatures, and the water tends to have high levels of dissolved solids, usually well above the level of salt found in seawater. Salt concentrations in the deeper formations across the Alberta Basin range from less than 50,000 ppm to more than 300,000 ppm. This compares to seawater at about 35,000 ppm. Water with high salt content is referred to as brine and is not considered suitable for either human consumption or agricultural use without significant desalination. Of course, the shallow zones continue to be filled with freshwater, and groundwater is an important resource in the Province of Alberta.

Thick sedimentary sequences like the Alberta Basin often contain oil and gas reservoirs. In Alberta, these producing reservoirs typically correspond to formations that would be called "aquifers," based on their flow properties. Existence of a mature oil and natural gas industry in Alberta has advantages for CO_2 storage operations because the properties of the subsurface, such as porosity and permeability, have been studied and characterized in detail, especially around the oil and gas fields. Furthermore, the associated infrastructure for oil and gas extraction operations might be used profitably for CO_2 injection projects. However, the development of the oil and gas fields has resulted in hundreds of thousands of wells having been drilled in the Alberta Basin. Many of these wells perforate tight confining units or seals (i.e., caprock formations), which have held hydrocarbons within underlying reservoirs over geological time scales. Without well perforations, these caprock



Figure 1.8 Density of wells (number per area) drilled across the world (from IPCC 2005).

formations are unlikely to allow CO_2 to leak should the CO_2 be injected into the underlying formation. However, the wells that have been drilled through the caprock formations might compromise the seal integrity because they provide potential leakage pathways for a buoyant fluid like CO_2 . Leakage along wells is especially of concern in regions with a rich history of oil and gas exploration and production. This is apparent in Figure 1.8, which shows spatial densities of oil and gas wells worldwide, with North America in particular showing high densities of wells extending over significant distances.

The general approach for geological storage of CO_2 is to identify formations in the deep subsurface that have sufficiently high permeability to allow reasonable quantities of CO₂ to be injected. These are the formations corresponding to "aquifers" or "productive hydrocarbon reservoirs." The injection formation should be overlain by a low-permeability aquitard, or caprock formation, that has sufficiently low permeability to keep the CO₂ from migrating upward out of the injection formation. If the injection is into a deep brine-filled aquifer, then the CO_2 will displace the brine out of the pore space, so that CO_2 will now occupy a significant fraction of the pore space around the injection well. As the injection proceeds, the region occupied by CO₂ expands, and more brine is pushed outward away from the well. Because the CO_2 is significantly less dense than the resident brine, the injected CO_2 tends to migrate upward by buoyancy as it spreads away from the well, forming a characteristic displacement pattern within the formation. Of course, if wells have perforated the caprock, then some CO₂ and/or brine might leak along one or more of the wellbores, forming the more complex pattern of flow and leakage illustrated schematically in Figure 1.9. In addition to mass movement involving CO2 and brine, the imposed pressure increase at the injection well also propagates into the formation, serving to drive the flow of both the CO₂ and the brine. How far this pressure pulse extends is also of interest, because it provides a way to define the



Figure 1.9 Schematic of CO_2 injection, migration, and interaction with existing oil and gas wells (from Gasda, S.E., S. Bachu, and M.A. Celia. 2004. Spatial characterization of the location of potentially leaky wells penetrating a deep saline aquifer in a mature sedimentary basin. *Environmental Geology*, 46, 707–720).

Area of Review that is a natural concept to include in regulatory structures. For all of these aspects of the CO_2 storage problem—movement of the injected CO_2 , movement of the displaced brine, possible leakage of one or both of the fluids into and through a complex vertical sequence of formations, and propagation of pressure increases into the formations—we will want to develop appropriate mathematical models and the concomitant methods to solve those mathematics. This book is meant to provide the basic tools to develop, implement, and solve these kinds of models.

1.5 THE APPROACH TAKEN IN THIS BOOK

With the exception of this first chapter, the focus of this book is on mathematical descriptions of fluid movement in the subsurface with a strong focus on CO_2 injection into deep saline aquifers. We have tried to write the book to cover the basics of the processes involved, beginning with a simple description of the experiments

of Henry Darcy some 150 years ago, and explaining how those experiments form the basis for much of our current analysis. The mathematical derivations become increasingly complex as the book progresses, but we have tried to write the text and the associated equations so that the material is accessible to anyone with an understanding of basic calculus and a willingness to work through the derivations. Note that a brief review of notation and basic theorems is given in the Appendix. In Chapter 2, we develop the mathematical models appropriate for systems where only one fluid exists in the pore spaces of the soil or rock. We refer to these systems as single-fluid or single-phase porous media. We use Darcy's equation along with a statement of mass balance and information about fluid and solid compressibility to derive the equations that govern the system. Those resulting equations that govern the movement of single-phase flows are usually partial differential equations, but for most systems, they are linear equations that can be solved readily with either analytical solution techniques or numerical methods. Chapter 2 allows us to introduce much of the notation and terminology that is used throughout the book.

In Chapter 3, we extend the analysis begun in Chapter 2 to include multiple fluid phases in the pore spaces. This leads to more complex governing equations which are typically nonlinear and coupled. We develop a series of specific cases that allow for some simplifications under certain circumstances, and demonstrate how these simplifications allow for more convenient solutions. When appropriate, the CO_2 -brine system is used as the example of two-phase flow in porous media.

The remaining Chapters 4–6 focus entirely on the CO_2 injection and subsequent migration in deep saline aquifers. Those chapters present a specific modeling approach based on considerations of length and time scales. We develop models that are noticeably simpler than the full multiphase models reviewed in Chapter 3, but still capture the essential behavior of the system at the relevant scales of interest. These models can provide substantial insights into system behavior. They also allow tools to be developed that can simulate large-scale injection, migration, and leakage of both CO_2 and brine, while also accommodating interphase mass exchange and subsequent mass transport of CO_2 as a dissolved component in the aqueous (brine) phase. These models represent all of these processes at the appropriate scales and are sufficiently simple to allow for very efficient solution methods. As such, the methods presented herein can allow realistic CO_2 problems to be analyzed directly, including problems with complex patterns of flow and leakage.

We have included an appendix, meant to provide helpful background and reviews. It includes notation and an overall review of vector and matrix operations as well as a few important results from calculus including Leibnitz' theorem and Gauss' theorem.

1.6 FURTHER READING

At the end of each chapter, we will provide suggestions for further reading on the topic. For Chapter 1 we suggest the following additional readings.

Emissions and Rise of Atmospheric CO₂ Concentrations

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