1

Introduction

1.1 Greenhouse Effect

The temperature in the Earth's atmosphere and at ground level is a result of a complex energy balance between incoming solar radiation energy and outgoing radiation energy from the Earth's surface and atmosphere. This balance varies naturally in daily and annual cycles. There are also variations with long-term cycles, such as the Milankovitch cycles, which are related to the Earth's orbital patterns (Milankovitch 1941). The heat balance is the basis for the temperatures that we have in the atmosphere and at ground level.

In general, the gases present in the atmosphere convert radiation energy into thermal energy – by *absorption*[1] *of electromagnetic radiation*, and vice versa – by *radiation*. The mechanism for absorption of radiation in a gas is that the gas molecules absorb the radiation energy by increasing its kinetic energy through molecular translation, rotation, and vibration, as well as electron translation and spin and nuclear spin. The increase in thermal energy of a gas translates into increased temperature. The longer the radiation travels through a gas, the more energy is converted. The radiation is at various wavelengths. The solar radiation is at rather low wavelengths (0.2–3 μm), either in the visible (0.4–0.8 μm) or in the near-visible (e.g. ultraviolet <0.4 μm) range. Radiation from the ground and from the atmospheric gases is at higher wavelengths (0.7–300 μm), which is known as infrared radiation.

The incoming solar radiation is about 342 W/m^2 (IPCC-WG1 2007). Some of this is reflected back into space by clouds, aerosols[2], and atmospheric gases, and some is reflected by the Earth's surface. About 240 W/m^2 is absorbed by the Earth's surface and atmospheric gases. In order to have a heat balance with constant temperature, the radiation from the Earth must be the same. Most of the outgoing radiation energy is at wavelengths in the range of 7–15 μm. Radiation from a surface is temperature dependent ($\propto T^4$), and a radiation of 240 W/m^2 would require an average surface temperature of $-19\,^\circ\text{C}$, which is much lower

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[1] Absorption of electromagnetic radiation is the process by which the energy of a photon is taken up by an atom. The photon is destroyed in the process. The term 'absorption' is also used within chemical engineering as uptake of a gas in a liquid or solid material, as described in Section 9.1. The term is also used in a number of other areas.

[2] Aerosol refers here to liquid droplets or solid particles in the atmosphere, such as sulfate aerosols from fossil fuel combustion or particles from volcano eruptions.

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than the actual average Earth surface temperature of +14 °C. Because some of the outgoing radiation is absorbed by clouds or gases in the atmosphere, the temperature at which radiation takes place is forced from −19 °C to +14 °C to maintain the average flux of 240 W/m^2. This absorption of radiation and the related increase in temperature is known as the *atmospheric greenhouse effect*. This is different from the effect observed in greenhouses, where the temperature increase is caused by suppression of convection.

A key issue is that the gases in the atmosphere have different properties with respect to absorption of radiation and radiation from the gases themselves. The absorption of radiation in a gas depends on the wavelength. Ozone (O_3) is a gas that absorbs ultraviolet radiation very well, whereas CO_2 absorbs at wavelengths around 3–5, and 12–20 μm. Water vapour absorbs at various wavelength ranges, including that of 7–15 μm. Visible light from the sun is absorbed by atmospheric gases only to a minor extent. The main bulk of infrared radiation, at wavelength 7–15 μm, is absorbed only to some extent. For some wavelength ranges, the absorption is about 100%.

The most important gases for the absorption of infrared radiation are water vapour (H_2O), carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), halocarbons (gases containing fluorine, chlorine, and bromine), and ozone (O_3).

Water vapour is the most important greenhouse gas in the atmosphere, accounting for about 60% of the natural greenhouse effect for clear skies. Human activities influence the atmospheric water vapour content to only a small extent; it depends much more on the temperature. The relation between temperature and water vapour content in the atmosphere is approximately a constant relative to humidity. The greenhouse effect of water vapour is much stronger in humid areas around the equator compared to that in polar areas where the air humidity is very low. Consequently, the importance of CO_2 as a greenhouse gas is more evident in polar regions, and changes in the concentration of CO_2 have a larger impact on the temperature in these regions.

The two most abundant gases in the atmosphere – nitrogen and oxygen – contribute almost nothing to the greenhouse effect. Homonuclear diatomic molecules such as N_2, O_2, and H_2 neither absorb nor emit infrared radiation.

The greenhouse effect was discovered by Joseph Fourier (1768–1830) in 1824. He was followed by John Tyndall (1820–1893), who did important work on the radiative properties of gases by verifying, through experiments, the absorption of radiation in gases and that emissions vary with wavelength and type of gas. In 1896, Svante Arrhenius (1859–1927) was the first to publish work on a quantitative investigation of the greenhouse effect, which he believed could explain the ice ages. At that time, the link between man-made emission of CO_2 and climate change was already established. Even though the calculations by Arrhenius were shown to be erroneous, he cleverly managed to collect information from a large number of sources and to make predictions not so very different from those recently made by the IPCC. Guy Stewart Callendar (1898–1964) made a very important contribution with a publication (Callendar 1938) presenting a comprehensive global temperature time series and a model, linking greenhouse gases and climate change (Fleming 2007). He found that a doubling of atmospheric CO_2

concentration resulted in an increase in the mean global temperature of 2 °C, with considerably more warming at the poles.

The effect of greenhouse gases in the atmosphere can be quantified on two different scales. One is the atmospheric lifetime, which describes how long it takes to restore the atmospheric system to equilibrium following a small increase in the concentration of the gas in the atmosphere. Individual molecules may interchange with the soil, oceans, and biological systems, but the mean lifetime refers to the net concentration change towards equilibrium by all sources and sinks. The other scale is the global warming potential (GWP), which is defined as the ratio of the time-integrated radiative forcing from a sudden release of 1 kg of a substance g relative to that of 1 kg of a reference gas, CO_2 (IPCC-WG1 2007):

$$\mathrm{GWP}(g) = \frac{\int_0^{t_x} r_g \cdot d_g(t)\mathrm{d}t}{\int_0^{t_x} r_{\mathrm{CO}_2} \cdot d_{\mathrm{CO}_2}(t)\mathrm{d}t} \tag{1.1}$$

r_g is the radiative forcing per unit mass increase in atmospheric abundance of component g, and $d_g(t)$ is the time-dependent abundance of g, and the corresponding quantities for the reference gas (CO_2) in the denominator. Radiative forcing is defined as the change in net irradiance at the tropopause. Net irradiance is the difference between the incoming radiation energy and the outgoing radiation energy in a given climate system and is measured in W/m^2. The GWP definition is time dependent (t_x), but, for any time horizon, the GWP of CO_2 is unity by definition.

In Table 1.1, a list of some selected greenhouse gases and their GWP and atmospheric mean lifetime is given. Water vapour is not included in the list even though it is an important greenhouse gas because the presence of water vapour in the atmosphere is mainly determined by the temperature. The short atmospheric lifetime of tropospheric ozone (hours to days) precludes a globally homogeneous distribution and is consequently not included in Table 1.1. Ozone concentrations, and associated radiative effects, are highest near their sources. CO_2 has an atmospheric lifetime that is difficult to specify precisely because CO_2 is exchanged with reservoirs having a wide range of turnover times: 5–200 years or even much longer than that.

1.2 Atmospheric CO_2

The concentrations of carbon dioxide, methane, and other greenhouse gases are currently increasing over time. The carbon dioxide concentration, measured as the mole fraction in dry air, on Mauna Loa, Hawaii, constitutes the longest record of direct measurements of CO_2 in the atmosphere. The average Mauna Loa CO_2 level for 2017 was 407 ppmvd (based on the monthly averages) compared to 316 ppmvd in 1959. The measurements were started by C. David Keeling of the Scripps Institution of Oceanography in March of 1958 at a facility of the National Oceanic and Atmospheric Administration (Keeling et al. 1976). NOAA started its own CO_2 measurements in May 1974, and they have run in parallel with those made by Scripps since then (Thoning et al. 1989).

Table 1.1 Global warming potential (GWP) – relative to CO_2 – as well as atmospheric concentration and lifetime of selected greenhouse gases.

Gas	Chemical formula	Recent tropospheric concentration	GWP (100 yr time horizon)	Atmospheric lifetime (yr)
Carbon dioxide	CO_2	≈386 ppm	1	100
Methane	CH_4	≈1800 ppb	25	12
Nitrous oxide	N_2O	≈320 ppb	298	114
Ozone	O_3	≈34 ppb	N/A	h/d
CFC-11	CCl_3F	≈240 ppt	4750	45
CFC-12	CCl_2F_2	≈536 ppt	10 900	100
CFC-113	CCl_2FFClF_2	≈76 ppt	6 130	85
HCFC-22	$CHClF_2$	≈200 ppt	1 810	12
HCFC-141b	CH_3CCl_2F	≈20 ppt	725	9
HCFC-142b	CH_3CClF_2	≈20 ppt	2 310	18
Halon 1211	$CBrCIF_2$	≈4 ppt	1 890	16
Halon 1301	$CBrCIF_3$	≈3 ppt	7 140	65
HFC-134a	CH_2FCF_3	≈52 ppt	1 430	14
Carbon tetrachloride	CCL_4	≈88 ppt	1 400	26
Methyl chloroform	CH_3CCl_3	≈10 ppt	146	5
Sulfur hexafluoride	SF_6	≈7 ppt	22 800	3200

Source: Data are based on CDIAC (2010).

For a much longer time frame, data from the Vostok ice core provide an insight into the variations in CO_2 levels for the past four glacial–interglacial cycles. In January 1998, the collaborative ice-drilling project between Russia, the United States, and France at the Russian Vostok station in East Antarctica yielded the deepest ice core ever recovered, reaching a depth of 3623 m (Petit et al. 1999). Data from this project are displayed in Figure 1.1. One conclusion from the authors was that present-day atmospheric burdens of CO_2 seem to have been unprecedented during the past 420 000 years. Note that the mean resolution for the CO_2 measurements was about 1500 years.

1.3 Natural Accumulations and Emissions of CO_2

There are many examples of natural emissions of CO_2 to the atmosphere. Forest fires and grass fires are natural phenomena, which release large amounts of CO_2 to the atmosphere. On the other hand, after such fires, new trees and grass will grow and eventually bind more or less the same amount of CO_2 as was released. One can say that as long as the total amount of biomass on a global basis is constant over a longer period of time, the combustion, decay, and growth of biomass is balanced with respect to CO_2. In this respect, the use of biomass for energy purposes can be regarded as *CO_2 neutral*. Consequently, if the amount of biomass is

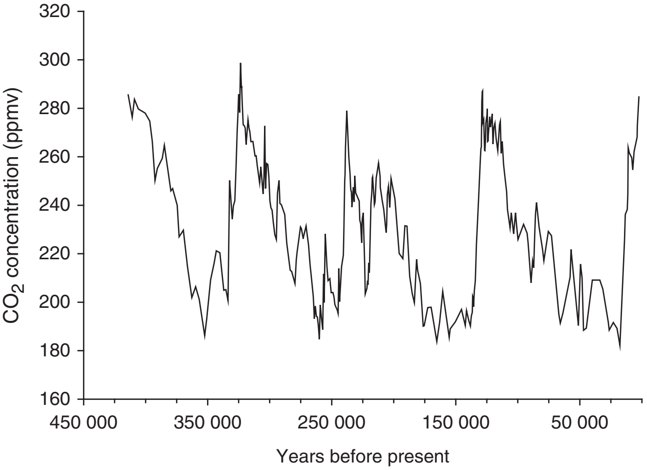

Figure 1.1 Vostok ice core data for 420 000 years. Atmospheric CO_2 concentrations from trapped gas bubbles. Source: Petit et al. (1999). Reproduced with permission of Springer Nature.

increasing through an expansion of the area covered by forests, this contributes to reducing the amount of CO_2 in the atmosphere. In reality, there is a significant ongoing reduction in biomass globally by man-made forest fires and wood cutting, which contributes to about 20% of the annual increase in greenhouse gas emissions.

There are also a number of examples of seepage of CO_2 from the ground. From many of the ground seepages, the flux is so small that it is hardly noticeable. Others have a high flux so that the ground-level concentration can cause death of plants through 'root anoxia' and even pose risks to human health and safety. At concentrations above about 2%, CO_2 has a strong effect on respiratory physiology, and at concentrations above 7–10%, it can cause unconsciousness and death. Exposure studies have not revealed any adverse health effect from chronic exposure to concentrations below 1% (Fleming et al. 1992).

One important aspect to note about CO_2 is that it has a higher molecular weight than air, meaning that it is denser than air. As a consequence, the release of CO_2 at a low point in the terrain, with little or no wind, can cause the concentration at that point to increase to a very high level, as the CO_2 will not be transported vertically by buoyancy.

Seepage of CO_2 from the ground has a relevance to CO_2 capture and storage because stored CO_2 may leak out to the atmosphere. The impact on human health from releases of CO_2 can be severe. Some examples are Lake Monoun, Lake Nyos, and Lake Kivu, which are the only three lakes in the world known to be saturated with CO_2 (Clarke 2001; IEA-GHG 2006). The former two are located quite close to each other in the Oku Volcanic Field in Cameroon, whereas the latter is in Rwanda. In these lakes, the content of CO_2 per volume of water increases with depth because the solubility of CO_2 in water depends on the pressure, as can be seen in Figure 4.17.

On 15 August 1984, *Lake Monoun* exploded in a limnic eruption probably caused by an earthquake, which resulted in the release of a large amount of CO_2. As a direct consequence of the release, 37 people were killed. A gas cloud came up from a crater in the eastern part of the lake at night. The 37 people who died were residents in a low-lying area close to the lake. Survivors reported that the whitish, smoke-like cloud smelled bitter and acidic. The vegetation was flattened around the eastern part of the lake, probably by a water wave caused by the gas cloud.

On 21 August 1986, *Lake Nyos* suddenly emitted a large cloud of CO_2 estimated as over 1 Mt, which travelled more than 10 km and suffocated 1700 people and 3500 livestock in the area near the lake. The lower levels of the deep lake had become saturated by CO_2 coming from a magma chamber beneath the region. The magma chamber is an abundant source of CO_2, which seeps up through the lake bed, charging the waters of Lake Nyos with an estimated 90 Mt of CO_2. It is thought that high rainfall just before the incident, and possibly a landslide, had displaced the CO_2-rich water at the bottom, releasing a massive bubble of CO_2 gas from the lake, in a natural phenomenon now referred to as 'lake overturn'. The heavy gas then sank to the ground and rolled in a cloud several tens of metres deep across the surrounding countryside.

Pipes have now been put in place in Lake Nyos and Lake Monoun to siphon water from the lower layers up to the surface and allow the CO_2 at the bottom of the lake to slowly bubble out. Events such as those in Lake Monoun and Lake Nyos can take place only in lakes that do not overturn annually, and where the water becomes stratified with very high concentrations of CO_2 at large depths. This can happen in deep tropical lakes.

In the Mammoth Mountain area in California, USA, CO_2 is leaking out of the ground at more than 1200 t/d (Farrar et al. 1995; Sorey et al. 1998). The concentration of CO_2 near the ground has been measured at over 50%. Three people are known to have died because of CO_2, and there is an area where the trees are dead.

There exist a few volcanoes with crater lakes. These lakes can be rather deep and still, with stratification and deep layers containing large amounts of CO_2, similar to Lake Nyos.

Geysers emit CO_2 to the atmosphere. Water charged with CO_2 rising from deep in the ground is released periodically in an explosive manner. *Hot springs* are similar to geysers but release CO_2-rich water at a continuous rate.

In 1979, at the *Dieng* volcano complex in Indonesia, a release estimated at about 200 kt of CO_2 took place in a rather short time before a major eruption. The CO_2 flowed from the volcano and down to a plain where 142 people were killed by suffocation.

There are a number of examples of CO_2 emissions from sedimentary basins. These emissions are characterised by being smaller and more stable over time compared to those previously given examples from volcanic areas. One example is the Southeast Basin in France where there are several small CO_2 fields in the ground leaking to the surface. The CO_2 is dissolved in the groundwater and comes out of the ground in springs as carbonated water. This has become the basis for a mineral water industry, such as Vichy and Perrier.

At some places, CO_2 is leaking from the seabed. Just outside the Aeolian Islands in the south of Italy, about 25 kt CO_2/yr is leaking over an area covering several square kilometres. Most of the CO_2 is being dissolved in the seawater.

A distinction should be made between natural emissions of CO_2 in volcanic areas and emissions in sedimentary areas. In volcanic areas, the emission of CO_2 is often characterised by a sudden release of CO_2, often caused by unstable seismic activities. High temperature and steam are often present, which builds up high pressure and severe emission with a very high concentration of CO_2. Emissions from sedimentary basins are characterised as more diffuse and definitely not sudden. Most sedimentary basins are located in tectonically stable regions with less or no seismic activity. They typically contain porous rocks or sandstone, which is gas permeable. In some areas, there are also impermeable rock layers that act as seals so that gas cannot go through and reach the surface. Oil and natural gas reservoirs in sedimentary basins have proved that such structures can hold gases locked in the ground for millions of years.

There are also natural accumulations of CO_2 in the ground (IPCC-CCS 2005). One example of such a sedimentary basin is the McElmo Dome in Colorado, USA. It contains about 1.6 Gt of CO_2 (98% purity) and is sealed by a 700 m-thick impermeable layer. Similar amounts of CO_2 are trapped in other fields in the United States: the St Johns Field in Arizona, the Bravo Dome in New Mexico, the Sheep Mountain in Colorado, the Jackson Dome, and the Pisgah Anticline in Mississippi. All fields mentioned above produce commercially traded CO_2, most of which is used to enhance oil production (refer to Section 2.1.2). The largest CO_2 accumulation known is the Natuna D Alpha field in Indonesia containing more than 9 Gt CO_2. In the Natuna field, there is also a substantial amount of natural gas, more than 700 Mt. Similar to the Natuna field, although smaller, is the La Barge field in Wyoming, USA. In general, there are many natural gas fields that contain a substantial amount of CO_2 and in some cases also some H_2S.

From some of these fields, there is a measurable leakage, whereas others appear to have no leakage. The mechanisms for these leakages are very well understood, so that one can tell with high probability which structures can hold CO_2 trapped for a long period of time.

1.4 Man-made Emissions of CO_2

Human activities result in emissions of mainly four greenhouse gases: carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and the halocarbons (a group of gases containing fluorine, chlorine, and bromine). These gases accumulate in the atmosphere, causing concentrations to increase with time. Significant increases in all of these gases have occurred in the industrial era.

The emission of CO_2 and other greenhouse gases does not only depend on technology but also depend on other mechanisms in society. The emission of CO_2 is closely linked to energy conversion, and one can depict the relation between the emission, the energy consumption, Gross Domestic Product (GDP), and population, as in Eq. (1.2), which is a slightly modified version of that presented originally by Kaya et al. (1989). This equation is a simplified way of expressing the relation between these parameters, but it provides a good qualitative illustration of how CO_2 emissions are related to these parameters. One obvious flaw in this equation is that the highest increase in population comes in areas where the GDP and energy consumption per capita is below the world average, and consequently, the equation overestimates contribution from the population factor.

$$\text{Emission of } CO_2 = \underset{(1)}{\frac{\text{Emission of } CO_2}{\text{Energy consumption}}} \cdot \underset{(2)}{\frac{\text{Energy consumption}}{\text{GDP}}} \cdot \underset{(3)}{\frac{\text{GDP}}{\text{Population}}} \cdot \underset{(4)}{\text{Population}} \tag{1.2}$$

(1) This ratio expresses the CO_2 formation related to energy consumption in society. The value of this ratio is closely related to the technology being used in terms of efficiency. The type of fuel (coal, oil, natural gas, and biomass) is also important for this ratio, which is further discussed in Section 4.2. The switch from coal to natural gas reduces the value, as do efficiency improvements. In some areas of the world, the dependence on coal is stronger than in others, where natural gas may be more accessible. The potential for reducing this ratio mainly depends on fuel availability and fuel selection and to some extent on how far efficiency is pushed when new power plants are built. There is still a huge potential for improving efficiency in power plants and other processing industries.

(2) This ratio is the *energy intensity*, which is calculated as units of energy per unit of GDP. The ratio says something about the structure of industry, transportation systems, and agriculture. An agricultural country would typically have low energy intensity, whereas, on the other hand, a country with a lot of metal production will typically have high energy intensity. In general, one can say that a high refinement ratio in the industry increases the energy intensity. One should here be aware that the international trade in products and raw materials that require a lot of energy (e.g. aluminium) is large. It may be that a country that is consuming a lot of energy-demanding products is importing most of them. In such a case, the energy intensity does not express the real energy intensity caused by the activities in that country. One example is cars, which are produced only in a few countries. Another example is production of heavy oil or bitumen, like in Venezuela and Canada, where much energy is spent on producing oil that is mainly exported.

(3) GDP of a country is defined as the market value of all final goods and services produced within a country in a given period of time. It is also considered the sum of value added at every stage of the production of all final goods and services produced within a country in a given period of time. One can consider the GDP as a measure of standard of living, which means consumption of goods, travel, heating, etc.

(4) According to Eq. (1.2), the population contributes proportionally to the emission of CO_2. The annual world population growth was about 83 million in 2017, which is about a 1.1% growth rate.

It is indeed a challenge to balance the population growth with technology improvements (term 1) and energy intensity reductions (term 2) with respect to CO_2 emissions.

1.5 Climate Change

Most people nowadays seem to accept that the observed change in global climate, especially during the past two decades, is man-made through emissions of greenhouse gases. Scientific knowledge of the global climate has developed greatly, not to fully understand it but to support a high probability for the link between change in global climate and man-made emissions of greenhouse gases (IPCC-WG1 2007). It is remarkable how the scientific community has moved over the past 20 years from strong disputes about climate science to the consensus observed today. Industrial corporations and most governments seem to accept the findings of the scientific community.

Is climate change good or bad? For some people, a warmer climate will definitely improve their life in many respects. Changes in the occurrence of animal and plant species may impact many people's comprehension of nature but would not necessarily be bad. The extinction of species and the appearance of new species is a continuous process in nature, and throughout history, climate change has been one cause for it. However, it is likely that climate change may result in a partial meltdown of the ice caps in Greenland and in Antarctica. The meltdown of thick ice caps was previously believed to be a very slow process, but recent knowledge indicates that the meltdown process is actually rather fast. Another negative effect is increased weather variability, with more storms and occurrences of heavy precipitation. In general, one can say that global warming will probably cause many problems for a large number of humans, resulting in the need for change with respect to where people can live and how to organise their society.

As mankind has always done in the past, he will somehow adapt to climate change. The ability and possibility to adapt will vary a lot depending on the location. Rich countries or countries with a lot of space will be able to adapt more easily to climate change than poor or densely populated countries. It is a paradox and an ethical dilemma that countries with the best ability to adapt to climate change are the ones emitting the most greenhouse gases. A very important decision we have to make is how we spend our resources between adapting to and reducing global warming.

In the future, we will most likely combine measures on how to reduce global warming and how to adapt to it. It is very difficult to know what measures should be emphasised. The longer we wait to introduce measures to reduce global warming, the more likely adaptation measures will become. It is also uncertain which measure will be most cost-effective, but today, it seems most people are in favour of reducing greenhouse gas emissions as a precautionary measure. Some countries and regions may only want to implement adaptation measures as they see the need for them.

1.6 Fossil Fuel Resources

Are we going to run out of fossil fuels soon? The answer is most likely no!

We are depleting the fossil fuel resources at an increasing rate. Nevertheless, there are still huge resources of coal and significant resources of oil and natural gas (Brandt and Farrell 2007; Chu and Goldemberg 2007; Freund and Kaarstad 2007). Both the amount of remaining resources and what is economic to produce have changed over time. It is hard to predict for how long there will be fossil fuel resources that are economic to produce. This mainly depends on fuel prices, cost of production, and environmental limitations, such as emissions of CO_2.

It should be noted that the technical challenge of producing fossil fuels is increasing. The resources that are easy and inexpensive to produce are the first to be exhausted, then comes the time of the more difficult ones. There is, however, no clear-cut transition between types of production, as one sees various kinds of ongoing fossil fuel production. Examples of resources from which it is difficult to produce are Canadian oil sand and Venezuelan heavy crude oil. The production of oil in these two examples requires a lot of expensive processing and use of energy before it can be fed to refineries. Other examples are oil shales, coal seams under the seabed, and natural gas hydrates. As the production on average becomes more difficult, there will be an increase in energy use and CO_2 emissions from the production of the fuels.

Production from gas shales, particularly in the United States, has changed the North American natural gas market with significant price drops. It is likely that unconventional gas will increase in importance, not just in the United States but also in other parts of the world.

If proponents of peak oil [3], such as Laherrère, Campbell, and Deffeyes (Swenson 2008), are correct, the predicted peak in oil production will take place before 2020 (Al-Husseini 2006). It is disputed whether this is likely. Even if this is the case, there are still so many fossil fuel resources left that we will probably produce transportation fuels by coal-to-liquid conversion.

There is one very important conclusion: the lack of fossil fuels *will not* cause a reduction of CO_2 emissions *soon enough* to avoid an unwanted high atmospheric concentration of CO_2. This implies that we cannot simply sit back and relax, waiting for the fossil fuels to be exhausted and let the problem with man-made climate change go away by itself.

1.7 Definition and Rationale of CO_2 Capture and Storage (CCS)

'*Carbon dioxide (CO_2) capture and storage*' *(CCS) or* '*carbon capture and sequestration*[4]' *is a family of methods for capturing and permanently isolating CO_2 that*

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[3] Peak oil is the point in time at which the maximum global oil production rate is reached, after which the rate of production starts to decline. Marion King Hubbert (1903–1989) first used the theory in 1956 to predict that US oil production would peak between 1965 and 1970. His model, the Hubbert peak theory, has since been used by some to predict the peak oil production globally.

[4] Sequestration is mainly used in North America. The term has different meanings in English, French, and Spanish and is also for many people a difficult word to relate to. Most people within the scientific community, including IPCC, have adopted 'capture and storage' IPCC-CCS (2005). IPCC Special Report on Carbon Dioxide Capture and Storage. Cambridge, United Kingdom, and New York, NY, USA, Cambridge University Press.

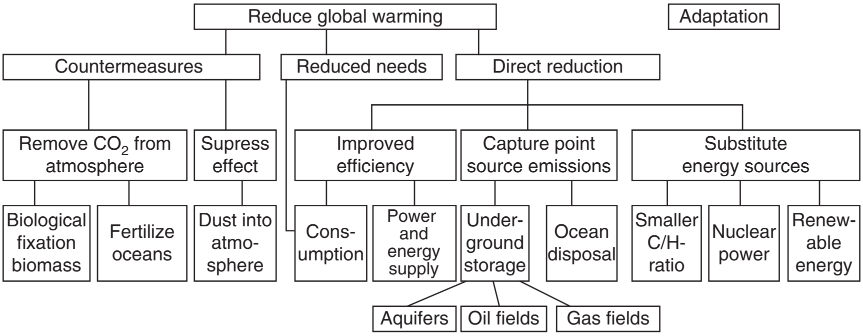

Figure 1.2 Options for reducing global warming. Source: Based on Lindeberg and Holloway (1998). https://www.osti.gov/etdeweb/biblio/20016103. Licensed under CC-BY 2.0.

otherwise would be emitted to the atmosphere and could contribute to global climate change.

If we want to reduce the greenhouse effect and global warming, we can implement *countermeasures*, reduce the *need for energy*, thereby reducing CO_2 emissions, or we can have a *direct reduction* (refer to Figure 1.2).

There are, in principle, four different ways of reducing CO_2 emissions:

(1) *Reduce the consumption of fossil fuels by*
  - increasing the efficiency of conversion processes
  - reducing the need for energy
  - the use of non-fossil energy sources such as hydropower, wind power, biomass, solar cells, and nuclear power.

(2) *Switch to fossil fuels with a lower carbon/hydrogen ratio*, by substituting coal and oil with natural gas.

(3) *Capture CO_2* from combustion plants and other industrial processes and store it in aquifers, in depleted or producing gas reservoirs, in oil reservoirs (enhanced oil recovery, EOR), or put it into the deep ocean.

(4) *Reduce deforestation* and thereby biologically fixate more CO_2 in biomass.

One could also include negative emission technologies by combining (1) and (3) to use bioenergy with CCS and possibly direct air capture (Bui et al. 2018).

Reducing the need for energy is a very difficult topic. The use of energy is very much related to the economic activity level and very important to things humans perceive as affecting their quality of life: the comfort of climatised homes, appliances, freedom to travel by car and to fly to faraway destinations by airplane, communication, and a rich supply of food and drink. These are highly esteemed values in the life of an increasing number of people around the world. Despite growing environmental consciousness among people, it is likely that these values will be very much contended.

Why capture CO_2? In the various processes where CO_2 is generated, it ends up *diluted* with a number of other gases. In power plants, the CO_2 concentration in the flue gas may be around 12–15 vol% for coal and 3–8 vol% for natural gas. In industrial processes, cement production gives a concentration of about 20 vol% of CO_2 and refineries 3–8 vol%. Most of the gas streams containing CO_2 are at or close to atmospheric pressure, which means that CO_2 is present at a rather low partial pressure[5]. It is commonly accepted that it requires too much energy to store the CO_2 with all its diluents away from the atmosphere. This means that CO_2 needs to be separated – or captured – from other gases and be brought up to a sufficient pressure for storage. The separation process involves mass transfer, which requires a certain volume and residence time, as well as energy use.

Where should the CO_2 be captured? In general, large point sources, preferably with a high CO_2 concentration, are the most economical. About 40% (42% in 2016) of the energy-related CO_2 emissions originate from the heat and power generation sector, which represents the largest CO_2 emitter followed by transport and industry (International Energy Agency 2018). Power generation is also important because it is the sector that has by far the highest number of the very large CO_2 emission point sources. Another area, which is of interest with respect to CO_2 capture, is that of plants producing synthetic fuels or chemicals from coal gasification. One should expect that the demand for transportation fuels will be larger than that which can be provided by conventional oil production and that unconventional oil resources have to be used. Gasification of coal, biomass, pet coke, and other heavy oils will likely be important in the future. Gasification processes are mostly well suited for CO_2 capture.

It is important to keep in mind that CCS is a *chain* consisting of capture, transport, and storage (Gielen and Podkanski 2004). The chain may be long geographically, with possibly several hundreds of kilometres between capture and storage. The chain would normally have a very limited buffer capacity, meaning that when CO_2 is captured, it has to be transported and stored within a short time frame (hours). Because of the large flow rates and volumes of CO_2 (refer to Example 2.1), it is not an easy task to have an intermediate storage that can remove the tight link between the capture and the storage. This means that the operation of the components in the chain is closely coupled. A larger number of sources (capture plants) and sinks (storage sites) can, to some extent, make the operation of the chain more flexible, in both the short term (hours) and the long term (years). In addition, one can think of intermediate storage of CO_2 in salt domes, as is being done with natural gas. Further, the storage has to be effective in terms of actually keeping the CO_2 away from the atmosphere for more than a thousand years.

1.8 Magnitude of CCS

Can CCS contribute to a significant reduction in greenhouse gas emissions? The International Energy Agency (IEA) projects the increase in world energy demand and the associated increase in CO_2 emissions by utilising different scenarios. Two of the scenarios are used in Example 1.1. The *New Policies Scenario* takes

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[5] Partial pressure relates to a specific gas in a gas mixture and the pressure that the gas would have if it alone occupied the volume (assuming ideal gas). According to Dalton's law, the sum of the partial pressure for the all the gases in a gas mixture equals the total pressure. Partial pressure equals the molar (or volumetric) fraction multiplied by the total pressure.

account of the broad policy commitments and plans that have been announced by countries around the world to tackle either environmental or energy-security concerns, even where the measures to implement these commitments have yet to be identified or announced. The *450 Scenario* includes actions necessary to have approximately a 50% chance of meeting a 2 °C increase in global average temperature from pre-industrial levels. To meet this goal, predictions indicate a need to limit the atmospheric CO_2 concentration to *450 ppmv*. To decrease the CO_2 emissions from the New Policies Scenario to the 450 Scenario, several measures would be necessary. This includes a focus on energy efficiency, an increase in renewable energy, nuclear energy, and CCS. Seventeen percentage of the decrease in CO_2 emissions would be covered by CCS by the year 2035 (IEA 2012). Example 1.1 gives an example on the number of CCS plants necessary to achieve this.

Example 1.1 ***CCS as part of the measures to get from the New Policies Scenario to the 450 Scenario.***

CCS under the '450 Scenario'

Assumptions:

New Policies Scenario does not include CCS.

17% (2.5 Gt CO_2) of the difference in CO_2 emissions from the New Policies Scenario to the 450 Scenario is covered by CCS by 2035 (IEA 2012).

CO_2 captured in a CCS plant: 2.5 Mt/yr.

Total number of CCS plants to build before 2035:

$$\frac{2.5 \times 10^3\ \text{Mt}}{2.5\ \text{Mt capt/plant}} = 1000\ \text{plants}$$

Annual CCS plants to build from 2016–2035:

$$\frac{1000\ \text{plant}}{20\ \text{years}} = 50\ \text{plants/yr}$$

The numbers in Example 1.1 point to the challenges for CCS to reach the goal of limiting the CO_2 concentration in the atmosphere to 450 ppmv. To build somewhere in the range of 1000 CCS plants before the year 2035 is a big economic, political, and practical challenge, but *not* unsurmountable.

1.9 Public Acceptance of CCS

Is CO_2 capture and storage a technology we will accept? If we develop CCS on a large scale, it will have an impact on society in general and on the way we live. Significant resources have to be invested to cover for operating expenses, which could be used in alternative ways in society. A very real concern – in particular, for onshore CO_2 storage – is whether the public will allow storage activities 'in their own backyard'.

Concerns and scepticism over CCS can be exemplified by the following questions and statements:

1. What is the point of CCS in a few rich countries, while other booming economies are increasing their emissions more than the reductions realistically achieved by CCS?
2. CCS will greatly increase the cost of power and of industrial production, so much of the industrial production will move to other countries with less stringent caps on CO_2 emissions and with even more greenhouse gas emissions as a consequence!
3. Climate change is already happening, and we are too late and not able to perform enough CCS to avoid a major change in the climate.
4. Will the CO_2 stored in the ground remain there or will it leak out into the atmosphere after a few years?
5. The CO_2 could leak out of the ground and kill people!
6. Who is responsible in the long term for CO_2 stored in the ground?
7. CCS is a methodology for the rich countries to continue their unsustainable way of life with an excessive use of energy!
8. In many countries, the challenge is to provide enough electricity, cars, and gasoline for the people. We cannot start CCS before we have developed our society into something closer to what they have in Europe and North America.
9. History shows that it will not be possible to make strong enough international agreements about CCS and other greenhouse gas emission limitations, so it is a waste of time and resources!
10. We should rather spend our money and engineering resources on renewable, non-fossil energy sources and technologies!
11. Some CCS scientists are suggesting use of CO_2 to increase the production of oil and thereby putting at least the same amount of CO_2 into the atmosphere as captured – why should we trust those people?
12. CCS requires the use of large amounts of chemicals, which will create the problem of handling toxic waste!
13. CCS requires the use of additional energy and will deplete fossil energy resources faster, resulting in less time to develop new energy sources.

The following statements could be made in favour of CCS:

a. The climate is changing and we should do what we can to reduce both the damage and the greenhouse gas emissions, which are a major cause of it!
b. In order to reduce greenhouse gas emissions significantly, CCS is the only realistic alternative to a substantial reduction in the use of fossil energy sources. Such a large reduction in the use of fossil energy sources could take us decades back in the development of our societies.
c. We have the knowledge, methodologies, and resources to perform large-scale CCS; what are we waiting for?
d. Underground storage of gases is something we know very well because we are today managing large-scale storage of natural gas.
e. We have experience with CO_2 capture – there are so many plants out there in the chemical industry!
f. We have experienced with CO_2 storage – there are many examples in the oil industry.

g. When storing CO_2 using our best knowledge, the possible leakage rate of CO_2 back to the atmosphere is so low that we can hardly measure it!
h. The rich countries should start CCS projects now to demonstrate to the world that this can actually be done, thereby setting the standard.
i. If we do not go for large-scale CCS, nuclear energy will be the alternative (Macfarlane and Miller 2007). There could be a lot of nuclear plants, most probably in many countries that do not have them today, and still no adequate long-term waste management, and possibly increased danger of nuclear war and terrorist actions with nuclear weapons and waste!
j. Large-scale CCS will cost us less than our military spending!
k. In order to cope with the challenge of man-made climate change, and because of its magnitude, there is no choice between CCS, renewable energy, nuclear energy, and energy conservation; we have to do them all!

1.10 Show-stoppers for CCS Deployment?

A number of studies have been conducted to find out what people in general (Curry et al. 2005; Itaoka et al. 2005; Uno et al. 2005; Huijts et al. 2007; Tokushige et al. 2007; Bachu 2008) and also what the experts (Gough 2008) think about the possibility and difficulties for the extensive use of CCS. Based on these reports, it seems that technology issues are *not* what people think will be stopping large-scale CCS from happening. The perceptions of environmental non-government organisations (NGOs) regarding CCS vary considerably. Most NGOs accept the necessity of CO_2 capture and storage in geologic formations, while only a small fraction does not (Wong-Parodi et al. 2008).

There are two main issues of concern: one is the *lack of long-term policy and national and international regulatory frameworks* and the other is the likely high and yet *uncertain cost*. Regulatory frameworks are necessary for commercial operation of CCS, and they are also necessary on an international level in order to create trust in CCS as a large-scale measure for reducing the emissions of greenhouse gases (Van Noorden 2010). One show-stopper could be that some countries take on the burden of CCS, while others do not, and that the latter benefit economically. A policy issue that needs to be addressed is the ownership of the pore space and the competition between storage rights and other mineral rights. Some people fear that CO_2 storage can reduce the economic value of underground structures where coal mining, oil production, or mining of minerals may take place in the future. Another policy issue is long-term (100, 500, and 1000 years) liability. It is not likely that private companies will have the longevity required for the time scale of safe storage of CO_2. In general, industry will not assume an indefinite and undefined risk. For this reason, the current thinking is that, after the active injection period, a closure period should follow where the operator demonstrates the safety of the storage, after which the state will assume long-term liability for the site. A possible complication here is when or if a CCS project is international in the sense that the CO_2 is captured and stored in different countries.

Public acceptance is an issue of major concern, and many people are worried about the storage of CO_2. The 'not in my backyard' (NIMBY) syndrome is growing globally, whereby the public in general accepts the technology and its benefits, but the local public does not want to accept the local economic and societal cost resulting from a CO_2 pipeline and storage. In this context, the need to resolve health, safety, and environmental (HSE) issues at an early stage is very important. An incident in the phase of installing and establishing CCS technologies can have a crucial impact on public acceptance and further development.

The risk of having a major leakage of CO_2 from a pipeline will most probably set a limit on the possibility of transporting CO_2 from a capture plant to a storage site. Sources and sinks of CO_2 do not in general match well (IPCC-CCS 2005), and it will be necessary to have a significant amount of CO_2 transported in pipelines. Assuming large-scale CCS with a larger number of capture plants, it is difficult to avoid transport of CO_2 through densely populated areas. A major leakage from a pipeline is potentially dangerous for humans. Therefore, it is of great importance to understand the human reaction to various extents of CO_2 exposure and, with this knowledge, generate the necessary safety measures. Issues could also arise from the transnational transport of CO_2 because of legal aspects (Leung et al. 2014).

Leakage of CO_2 from an underground storage site is also of concern. A major leakage from a storage site may irreversibly reduce the credibility of CCS. It is important and also difficult to inform society about the mechanisms of CO_2 storage and CO_2 flow underground and to deal with the risk of leakages, not least, mentally.

The whole process of CO_2 capture, transport, and storage combines not only existing and well-understood technologies in a new context but also new methods, unknown operating regimes, and quantities. This combination holds a lot of risk for both the separate process steps and the total technology. As for the capture part, these concerns are connected to the maturity of the novel technologies themselves and also with regard to the large scales of CO_2, which need to be captured and stored. Moreover, the assessment of these risks in a cost-efficient and systematic manner is very important. When it comes to transportation, the limited operating experience with CO_2 is a problem, as the standards for existing pipeline transportation cannot easily be transferred one-to-one. The aspect regarding generally consistent guidelines can also be applied to the storage locations. Moreover, monitoring and safety issues regarding the sites, wells, and the injection itself can also be seen as critical points (DNV 2010).

1.11 History of CCS

The technology for separating gases using chemical absorption was developed in the 1940s (Siddique 1990) and thereafter developed for separating CO_2 from gas mixtures. The purpose of separating CO_2 from other gases was, among other things, the carbonation of drinks. The technology was developed for separating CO_2 and other acid gases from natural gas (Kohl and Nielsen 1997). One of the first to suggest CCS was Marchetti (1977). He made reference to several methods for CO_2 capture from power plants and blast furnaces, and he proposed storing the CO_2 in the ocean. Marchetti worked for the International Institute for Applied Systems Analysis (IIASA) in Austria, where CCS was an issue being worked on during the 1970s. In 1980, Anthony Albanese and Meyer Steinberg published a paper with a very detailed discussion on capture technologies and energy use as well as storage (Albanese and Steinberg 1980). During the 1980s, Steinberg published a number of reports and papers dealing with CCS, and this is summarised in Steinberg (1992). Meyer Steinberg is the most obvious *father of CCS*, if one can use such a term, because of his contribution during the 1980s. Steinberg has published a large number of papers related to CCS and is also the co-author of a text book (Halmann and Steinberg 1999).

The Norwegians, Erik Lindeberg and Torleif Holt, did a lot of work on CCS in the late 1980s and were the initiators of CCS in Norway. A significant contribution was made by Wim Turkenburg, Kornelius Blok, and Chris Hendriks in the Netherlands, who, among other things, organised the *First International Conference on Carbon Dioxide Removal* (ICCDR) in Amsterdam in March 1992. ICCDR and the *Greenhouse Gas Mitigation Options Conference* were merged in 1997 to form the *Greenhouse Gas Control Technologies* (GHGT) conference series, which is now established as the main conference within CCS.

During the 1980s, work related to CCS took place at Oak Ridge National Laboratory (ORNL) and the Electric Power Research Institute (EPRI) (Steinberg 1992). There was also activity related to CO_2 capture at the Argonne National Laboratory from the early 1980s (Abraham et al. 1982).

It was not until close to 1990 that the volume of research effort in this field was increased to a significant level. The Japanese, and in particular, the RITE (Research Institute of Innovative Technology for the Earth), were already doing extensive work in the early 1990s. The main focus of research in Japan was on CO_2 fixation and utilisation.

In 1991, the IEA Greenhouse Gas R&D Programme (IEA GHG) was established. IEA GHG is an Implementing Agreement of the International Energy Agency. In January 2013, the IEA GHG consisted of 19 member countries, the European Commission, OPEC, and 21 multinational industrial sponsors. The IEA GHG is playing a very important role as a coordinator for R&D activities within CCS. The Intergovernmental Panel on Climate Change (IPCC) has been important for the development of the interest in CCS and in justifying the need for CCS, by pointing to the necessity both to reduce greenhouse gas emissions and to emphasise CCS as a mitigation option.

It is important to note that many CO_2 capture plants were built and operated with removal of CO_2 from natural gas and synthesis gas during the 1970s and 1980s. CO_2 was also captured from the flue gas in a number of plants that were built in this time period in order to provide CO_2 for EOR (refer to Section 2.1.2) as well as for other industrialised use (Pauley 1983; Barchas and Davis 1992; Sander and Mariz 1992; Suda et al. 1992). One can say that the technology for the capture of CO_2 from gas streams was already available at the time the idea of CCS emerged.