

David Ussiri · Rattan Lal

# Soil Emission of Nitrous Oxide and its Mitigation

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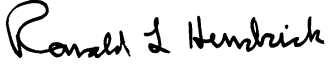
# Foreword

Humans are now the prime drivers of global change, which includes climate change. Atmospheric levels of heat-trapping greenhouse gases (GHGs) have increased, and the effects of human-induced climate change are now apparent. Global average temperature is rising, glaciers are melting worldwide, rainstorms are intensifying in many areas, and summer heat waves have become more intense while winters are less severe. Whether the governments and decision makers will act sufficiently fast to stem the global warming is uncertain because global warming presents unique challenges and the crucial research information is not readily available.

Nitrogen is essential for all life forms on Earth. Nature and its biodiversity can only exist because of the availability of reactive nitrogen in the system. Mankind has developed the technology to produce synthetic reactive nitrogen, which is used as a fertilizer to increase food production. Food production for about half of the current global human population of seven billion is being achieved because of the availability of synthetic nitrogenous fertilizers. Moreover, mankind also creates reactive nitrogen indirectly as a by-product from the transportation and energy sectors. Once reactive nitrogen molecule has been created, it is highly mobile and remains in the environment for a considerable time while contributing to several undesirable effects. Yet, anthropogenic activities have more than doubled the availability of reactive nitrogen in the biosphere, primarily through agricultural activities. Increasing nitrogen availability is leading to several unintended environmental consequences, including enhanced emissions of nitrous oxide (N<sub>2</sub>O). It is a long-lived radiatively active greenhouse gas (GHG) with an atmospheric lifetime of approximately 120 years, and heat trapping effects about 310 times more powerful than that of carbon dioxide (CO<sub>2</sub>) on molecular basis. Despite being a potent GHG, it also plays a significant role in the depletion of stratospheric ozone. The volume entitled *Soil Emission of Nitrous Oxide and Its Mitigation* by David Ussiri and Rattan Lal comprises a clear and concise analysis of the global budget of N<sub>2</sub>O and the factors controlling its emission. It also describes the anthropogenic sources of N<sub>2</sub>O with major emphasis on agricultural activities. The volume includes an extensive synthesis of mitigation techniques to reduce emissions from agricultural soils and diverse sources of nitrogenous fertilizers. Although significant research

information about  $\text{N}_2\text{O}$  emissions exist, this information has remained compartmentalized across a wide range of disciplines. This book fulfills the need for comprehensive multidisciplinary synthesis of the up-to-date scientific knowledge on  $\text{N}_2\text{O}$  and its pathways through nature. The volume is of interest to graduate students pursuing their career in soils and environmental sciences, academicians, policy makers and land managers. The comprehensive synthesis is timely and of relevance to an issue of global significance.

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# Foreword

The human population reached seven billion in 2011, and continues to grow unabated. The burden of feeding an ever-increasing population requires greater food production from a decreasing per capita land area, and dwindling water and nutrient resources. Meeting this challenge has resulted in greater applications of nitrogen-containing fertilizers to increase total food production. Yet, there are the consequences of this unparalleled release of anthropogenic nitrogen into the environment. One consequence is an increase in atmospheric concentration of nitrous oxide ( $\text{N}_2\text{O}$ ). Agriculture is the major source of  $\text{N}_2\text{O}$  emissions while the combustion of fossil fuels and industrial activities also have contributed to the increase. Carbon dioxide ( $\text{CO}_2$ ) is the major greenhouse gas (GHG) resulting in global climate change. Despite having a larger capacity to absorb long-wave radiation,  $\text{N}_2\text{O}$  – one of the potent GHG – has received far less attention than  $\text{CO}_2$ .

Nitrogen has been the subject of more intense research than most elements in the periodic table because of its ever increasing role in agriculture. The abiotic and biotic chemical processes that influence the efficacy of nitrogen-containing fertilizers, and the environmental impacts of nitrogen-transformation by-products have been studied by soil chemists, geochemists, agronomists, agricultural engineers, oceanographers, limnologists and climate researchers. A wealth of information exists on  $\text{N}_2\text{O}$  and its formation pathways in nature. Unfortunately, this information is scattered over a wide range of disciplines with different perspectives and priorities. Thus, there is a strong need to collate and synthesize this information and make it readily available in one reference for researchers,



academicians and land users. Comprehensive and up-to-date, *Soil Emission of Nitrous Oxide and Its Mitigation* by David Ussiri and Rattan Lal fills this gap admirably. The information presented in this book is crucial to any comprehensive plans for the mitigation of greenhouse gases, and developing any strategy of reducing anthropogenic global warming and global climate change.

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# Preface

Since the start of industrial age around 1750, significant increases have occurred in the atmospheric concentrations of several trace gases which have environmental impact at global and regional scales. Three of these gases namely carbon dioxide ( $\text{CO}_2$ ), methane ( $\text{CH}_4$ ), and nitrous oxide ( $\text{N}_2\text{O}$ ) – the so-called greenhouse gases (GHGs) – contribute to global warming. The  $\text{N}_2\text{O}$  also causes the depletion of stratospheric ozone layer. Emission of GHGs to the atmosphere is of primary concern worldwide because of their impact on radiative forcing and the climate change. In addition to three primary GHGs listed above, some halocarbons and aerosols present in trace concentrations also impact the radiative forcing. Atmospheric concentrations of these gases have been increasing rapidly since 1950s as a result of human activities. Molecules of these gases trap outgoing long wave radiation energy in the lower atmosphere, which raises the Earth's surface temperature. In its fourth assessment report, the Intergovernmental Panel on Climate Change (IPCC 2007) reported a linear trend in global surface temperature between 1906 and 2005 with a warming of  $0.74\text{ }^\circ\text{C}$  (range  $0.56\text{--}0.92\text{ }^\circ\text{C}$ ), with a more rapid warming trend over the past 50 years. This warming is primarily attributed to human-induced emission of these heat-trapping gases into the atmosphere.

Despite its low concentration in the atmosphere,  $\text{N}_2\text{O}$  is the fourth largest contributor to the greenhouse effect. Its contribution accounts for 7% of the current atmospheric radiative forcing. Therefore, this volume focuses on emission of  $\text{N}_2\text{O}$  and its mitigation strategies. It begins with an overview of the global N cycle, sources of N input to soil, the general soil–atmosphere exchange of  $\text{N}_2\text{O}$ , as well as key soil processes moderating  $\text{N}_2\text{O}$  fluxes. The volume integrates information from different disciplines and presents a holistic approach to  $\text{N}_2\text{O}$  formation in terrestrial and aquatic ecosystems and emission to the atmosphere, its effects on radiative forcing, and possible sinks and mitigation options. The volume is specifically prepared to provide academic and research information for undergraduate and graduate students, scientists, researchers, land managers and policy makers interested in understanding the environmental impacts of  $\text{N}_2\text{O}$  emission, sources, sinks of  $\text{N}_2\text{O}$ , role of different land use and land management on  $\text{N}_2\text{O}$  fluxes and

mitigation options. It places particular emphasis on the roles of anthropogenic activities on both direct and indirect increases in  $N_2O$  emissions.

The  $N_2O$  is produced in soils and sediments during N transformation by microbial processes of nitrification and denitrification as well as fossil fuel combustion and some industrial processes. Nitrification is the predominant process producing  $N_2O$  under warmer conditions. In contrast, denitrification is predominantly a process under wet and anaerobic conditions when ammonium ( $NH_4^+$ ) and nitrate ( $NO_3^-$ ) source is present in the soil and sediments. Historical data obtained from ice cores extending back 2,000 years indicate relatively stable  $N_2O$  concentration around  $270 \pm 7$  ppbv in pre-industrial revolution, followed by steady rise during the past 200 years to 319 ppbv in 2005. In the past 20 years, the atmospheric levels of  $N_2O$  have been increasing almost linearly at  $0.26\% \text{ year}^{-1}$ . This increase is generally attributed to the anthropogenic emissions. Human activities in the past century have accelerated release and removal of reactive N to the global atmosphere by as much as three to fivefold. Human activities that emit  $N_2O$  are those related to the transformation of fertilizer nitrogen into  $N_2O$  and its emission from agricultural soils, biomass and fossil fuel burning, animal husbandry, and industrial processes such as nylon manufacture and nitric acid production. In addition,  $N_2O$  is produced naturally from a wide range of biological sources in soils, water and sediments. Agricultural soils contribute about 65–70% of the total  $N_2O$  produced by the terrestrial ecosystems.

The  $N_2O$  is a long-lived radiatively active trace gas with atmospheric lifetime of 118 years. The only known main sink for atmospheric  $N_2O$  is stratospheric photo-reactions. Atmospheric  $N_2O$  is also one of the major sources of ozone depleting reactions in the stratosphere.

The fourth assessment report of the intergovernmental Panel on Climate Change (IPCC 2007) estimated  $N_2O$  emissions from both natural (i.e., oceans, tropical soils, wet forests, dry savannas, temperate soils-both forest and grasslands) and anthropogenic sources (i.e., fertilized agricultural soils, biomass burning, industrial processes, animal feeds and manures, sewage processing, landfills, atmospheric inorganic N deposition) at  $8.5\text{--}27.7 \text{ Tg } N_2O\text{-N } \text{year}^{-1}$  (Tg = teragram =  $10^{12} \text{ g}$  = million metric ton). Agriculture is the single largest anthropogenic source of  $N_2O$  fluxes contributing  $1.7\text{--}4.8 \text{ Tg } N_2O\text{-N } \text{year}^{-1}$ . This source is likely to be increased with the projected increase in use of nitrogenous fertilizers to meet the ever increasing demand for food of the growing and affluent world population. Natural sources of  $N_2O$  emission are estimated at  $11.0 \text{ Tg } N_2O\text{-N } \text{year}^{-1}$ . In addition to supply of N, temperature and moisture regimes are the principal controls of  $N_2O$  emission.

Collation, synthesis, and critical review of the available literature show that there are large uncertainties associated with the measured data on  $N_2O$  fluxes with the emphasis on spatial and temporal variability of measured fluxes. This uncertainty underscores the research needs to increase database by monitoring fluxes for long-term studies. In addition, there is a need for reliable flux data to improve model predictions, and for development of equipment for monitoring of fluxes for long-term plots under a wide range of land use and management systems. Also,

there is a strong need for robust models incorporating larger emission scenarios and more emission factors to improve the quality of N<sub>2</sub>O data. Yet, there is a strong need for development of nitrogenous fertilizers and the mode and method of their applications which enhance the nitrogen use efficiency while decreasing the losses of reactive nitrogen into the environment. While the Haber–Bosch process of manufacturing nitrogenous fertilizers have drastically enhanced crop yields and saved billions from starvation, the use efficiency of fertilizers must be improved while feeding the globe and mitigating global warming.

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# Chapter 1

## The Role of Nitrous Oxide on Climate Change

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**Abstract** Nitrous oxide is the third most important anthropogenic greenhouse gas in the atmosphere, contributing about 6% to the radiative forcing by long-lived greenhouse gases. In addition to its role as greenhouse gas, N<sub>2</sub>O also play significant role in ozone (O<sub>3</sub>) depletion. Its atmospheric abundance prior to industrialization was 270 parts per billion by volume (ppbv). Its current abundance is ~324 ppbv. Over past 10 years, the mean annual growth rate is ~0.8 ppbv year<sup>-1</sup>. It is emitted into the atmosphere from natural and anthropogenic sources. The anthropogenic activities have increased the concentration of long-lived greenhouse gases dramatically since industrialization, and altered the composition of global atmosphere, with great implications for current and future climate. Once emitted, the N<sub>2</sub>O is very stable in the stratosphere; the only known sink is the photo-oxidation tractions in the stratosphere. These reactions release oxides of N that contributes to ozone depletion. The magnitude of its current and projected future emissions leads to concerns about the timing of the recovery of ozone.

**Keywords** Greenhouse gases • Climate change • Radiative forcing • Global warming potential • Ozone depletion • Ozone depletion potential

## Abbreviations

UV	Ultraviolet GHGs greenhouse gases
IR	Infrared radiation
PFCs	Perfluorochlorocarbons
HFCs	Hydrofluorocarbons
GWP	Global warming potential
ODP	Ozone depleting potential
ODS	Ozone depleting substance
DU	Dobson unit
RF	Radiative forcing
IPCC	Intergovernmental Panel on Climate Change
FAR	First Assessment Report
RF	Radiative forcing
BP	Before present

### 1.1 Introduction

Nitrous oxide ( $\text{N}_2\text{O}$ ) is a colorless gas of slightly sweet odor and taste under ambient conditions, which was discovered by Joseph Priestly in 1772 from the reduction of nitric oxide (NO) with iron or iron sulfur mixtures. Its presence in the atmosphere has been known since 1939 (Adel 1939). However, its importance to global environment was only realized in the early 1970s when atmospheric scientists hypothesized that  $\text{N}_2\text{O}$  released into the atmosphere through denitrification of nitrates in soils and waters triggers reactions in the stratosphere that may lead to destruction of the ozone layer (Crutzen 1970, 1972, 1974). Ozone layer protects the earth from biologically harmful ultraviolet (UV) radiations from the Sun. Later on, the investigations of its radiative properties led to classification as an important greenhouse gas (GHG) which could modify the radiation energy balance of the earth—atmosphere system (Wang et al. 1976; Ramanathan et al. 1985). These two characteristics, in combination with observation of its increasing concentration in the atmosphere (Prinn et al. 1990; MacFarling Meure et al. 2006; Forster et al. 2007) and long atmospheric lifetime ( $114 \pm 10$  years; Forster et al. 2007; Wuebbles 2009; Prather and Hsu 2010) makes  $\text{N}_2\text{O}$  an important factor in global climate system and atmospheric chemistry of the ozone. In view of its potential to influence the global climate, it is necessary to gain knowledge about its global budget, source, sinks, and emission mitigations.

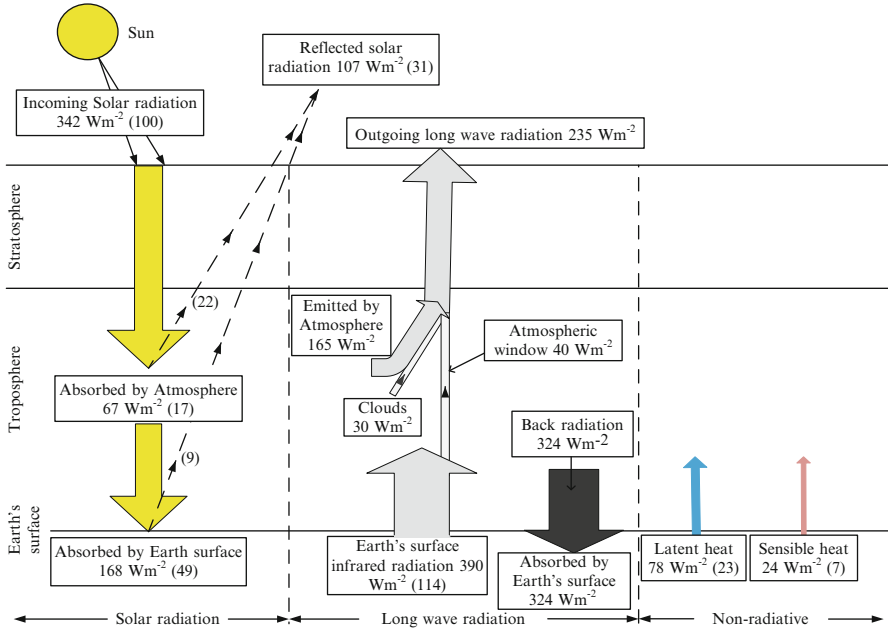
Measurements of entrapped  $\text{N}_2\text{O}$  in polar ice indicate that the global concentration of  $\text{N}_2\text{O}$  is higher now than at any time during the past 45,000 years (Leuenberger and Siegenthaler 1992; Schilt et al. 2010). After the last ice age,  $\text{N}_2\text{O}$  concentration increased and then remained at  $\sim 270$  parts per billion volume (ppbv =  $\text{nmol mol}^{-1}$ ) for about 10,000 years until the nineteenth century. Globally, the average mixing ratio has increased from  $\sim 270$  ppbv since pre-industrial

times in 1750s to current concentrations of 324 ppbv and continues to grow, currently at a rate of  $0.8 \pm 0.2 \text{ nmol mol}^{-1} \text{ year}^{-1}$  or  $0.25 \pm 0.05\% \text{ year}^{-1}$  and by 0.25% during 2000–2010 (WMO 2011).

## 1.2 Greenhouse Effect

The radiation from the sun is the source of Earth's energy that drives global climate. The climate system can be described as weather generating heat engine driven by the solar radiation energy input and thermal radiation output (Peixoto and Oort 1992). About 99.9% of the electromagnetic radiation emitted by the sun is in wavelengths of 0.15–4.0  $\mu\text{m}$  (i.e.,  $1 \mu\text{m} = 10^{-6} \text{ m}$ ). The balance between the solar energy that Earth receives from the Sun and that which it radiates out to the space is a major driver of the Earth's climate. Quantification of the amount of energy flow in and out of the Earth system and identification of the factors determining the balance between the incoming and outgoing energy helps in understanding the climate change. The Sun emits radiation over a spectrum of energies that exist in the form of waves; the radiation wavelength is the inverse of energy. On the high energy side of solar spectrum is UV and on the low energy side is infrared (IR) radiation. Nearly half of solar radiation is in the visible spectrum (i.e.,  $\sim 0.4\text{--}0.7 \mu\text{m}$ , short wave electromagnetic spectrum) and the other part is mostly in near infrared and UV electromagnetic spectrum. The amount of energy reaching the top of Earth's atmosphere during daytime is estimated at  $1,370 \text{ Watts (W) s}^{-1} \text{ m}^{-2}$ . Averaged over the entire planet, the Earth's atmosphere receives  $342 \text{ W m}^{-2}$ , predominantly as short wave electromagnetic radiation (Kiehl and Trenberth 1997; Le Treut et al. 2007; Fig. 1.1). About 31% (i.e.,  $107 \text{ W m}^{-2}$ ) of this solar energy is reflected back to space by clouds, aerosols, atmospheric gases, and the Earth's surface. The fraction of total energy reflected back to space can be referred to as *albedo*, and is influenced by surface conditions such as snow, ice, land versus sea, vegetation type, line of sight to the sun and other surface factors. It is also affected by atmospheric conditions, including cloud cover and wavelength of solar energy received (Pinty et al. 2008). Therefore, total albedo is highly variable from one place to another. The remaining energy ( $235 \text{ W m}^{-2}$ ) is mostly absorbed by the Earth's surface—land and ocean ( $168 \text{ W m}^{-2}$ ) and to a lesser extent by atmosphere ( $67 \text{ W m}^{-2}$ ). The energy absorbed by the vegetation layer drives the plant processes such as evapotranspiration, photosynthesis, and C assimilation, while the remaining fraction available in the underlying soils controls evaporation, snow melting, and other temperature-related processes (Sellers et al. 1997).

To maintain energy balance under stable climate, the Earth must radiate nearly the same amount of energy (about  $235 \text{ W m}^{-2}$ ) back to space. If the Earth was to emit  $235 \text{ W m}^{-2}$ , temperature at the Earth surface will be around  $-19^\circ\text{C}$  ( $255 \text{ K}$ ) based on the energy balance requirements, which is the temperature observed at the altitude of 5–6 km above the Earth's surface (Peixoto and Oort 1992). However, the



**Fig. 1.1** The flow of energy from the Sun to Earth and between the Earth's surface and the atmosphere. *Numbers in brackets* are percent of solar radiation (Redrawn with modification from Kiehl and Trenberth 1997; Baede et al. 2001; Le Treut et al. 2007)

Earth being colder than the Sun, is warmed and it radiates about  $390 \text{ W m}^{-2}$  (or about 114% of solar radiation from the Sun) as long waves (Fig. 1.1), primarily IR (i.e., radiant heat, Le Treut et al. 2007). The surface IR of  $390 \text{ W m}^{-2}$  corresponds to blackbody emission at  $14^\circ\text{C}$  on Earth's surface, decreasing with altitude, and reaching mean temperature of  $-58^\circ\text{C}$  at the top of the atmosphere (i.e., troposphere, the layer closest to the Earth) about 15 km above the Earth's surface. Much of this thermal radiation is absorbed in the atmosphere, and radiated back to the Earth ( $324 \text{ W m}^{-2}$ ), causing it to be warmer than as if the direct solar radiations were the only source of energy (Le Treut et al. 2007). As a result, the observed global temperature at the Earth surface is about  $14^\circ\text{C}$ ,  $33^\circ\text{C}$  higher than expected, which is attributed to absorption and re-emission of this long wave radiation in the form of IR by the atmosphere, as a result, radiant heat is trapped near the Earth's surface and keeps it warm. This process is referred to as the natural greenhouse effect. Two other mechanisms which transfers heat from Earth's surface to the atmosphere are sensible heat ( $24 \text{ W m}^{-2}$ ), and latent heat (i.e., water vapor ( $78 \text{ W m}^{-2}$ )) which eventually release its heat through condensation in the atmosphere (Kiehl and Trenberth 1997; Le Treut et al. 2007).

The outgoing long wave radiation leaving the atmosphere is estimated at  $235 \text{ W m}^{-2}$ , representing 74% of the incoming solar radiation (Fig. 1.1). Some of the radiation leaving the atmosphere originates from the Earth's surface and

transmitted relatively unimpeded through the atmosphere ( $40 \text{ W m}^{-2}$ ) in the areas where there are no cloud cover, and is present in part of the spectrum known as atmospheric window ( $8.0\text{--}12.0 \mu\text{m}$ ). The atmosphere emits about  $165 \text{ W m}^{-2}$ , and clouds about  $30 \text{ W m}^{-2}$ . Thus, very little radiation is transmitted directly to the space as if the atmosphere were transparent.

Presence of greenhouse gases (GHGs), also known as trace gases, those which account for less than 1% of the total volume of dry air in the atmosphere plays important role in Earth's energy budget by absorbing and re-emitting IR radiation emitted by Earth surface, preventing it from escaping to the space. The most important gases causing the greenhouse effect are water vapor ( $\text{H}_2\text{O}$ ), carbon dioxide ( $\text{CO}_2$ ), methane ( $\text{CH}_4$ ), nitrous oxide ( $\text{N}_2\text{O}$ ) and ozone ( $\text{O}_3$ ). As a result, most of the radiant heat flows back and forth between Earth's surface and atmosphere, and absorbed in the atmosphere to keep the Earth's surface warm (Le Treut et al. 2007). The exchange of energy between earth surface and atmosphere maintains global temperature near the surface at global average of  $14^\circ\text{C}$ , and decreases with increase in height above the earth surface. The GHGs occur naturally in the atmosphere and are responsible for natural greenhouse effect, which makes life possible, as we know it. The GHGs that are not naturally occurring include hydrofluorocarbons (HFCs), perfluorocarbons (PFCs; a class of organofluorine compounds that have all hydrogen (H) replaced with fluorine (F)), and sulfur hexafluoride ( $\text{SF}_6$ ) generated from a variety of industrial processes. Without this natural greenhouse effect, the life on Earth would not be possible (Le Treut et al. 2007). The most abundant constituents of the atmosphere- nitrogen (78.1% volume), oxygen (20.9% volume), and argon (0.93% volume) have minimal interaction with the incoming solar radiation and no interaction with IR radiation emitted by the Earth. On the other hand, clouds exert greenhouse effect; however, this effect is offset by their reflectivity such that clouds tend to have a cooling effect on climate (Le Treut et al. 2007).

Atmospheric concentrations of GHGs have changed naturally throughout the Earth's history. However, anthropogenic activities in post-industrial era have resulted in enhanced emission of four major GHGs namely  $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ , and  $\text{O}_3$ . In addition, new, manmade GHGs gases generally termed as high-global warming potential gases: HFCs, PFCs, and  $\text{SF}_6$  have been added to the atmosphere in the post-industrial era (Baede et al. 2001). These compounds are the most potent GHGs because they have very large heat-trapping capacity and some of them have extremely long atmospheric lifetimes (Table 1.1). Once emitted can remain in the atmosphere for centuries, making their accumulation almost irreversible. The atmospheric concentration of  $\text{CO}_2$  has increased due to fossil fuel use in transportation and power generation, cement manufacturing, deforestation and accelerated processes of organic matter decomposition. The  $\text{CH}_4$  has increased because of agricultural activities, natural gas distribution, and landfills. Wetlands also release  $\text{CH}_4$  naturally. The  $\text{N}_2\text{O}$  has increased as a result of agricultural soil management and N fertilizer use, livestock waste management, mobile and stationary fossil fuel, combustion, and industrial processes. Soils and oceans also emit  $\text{N}_2\text{O}$  naturally.

**Table 1.1** Global warming potentials of important anthropogenic greenhouse gases

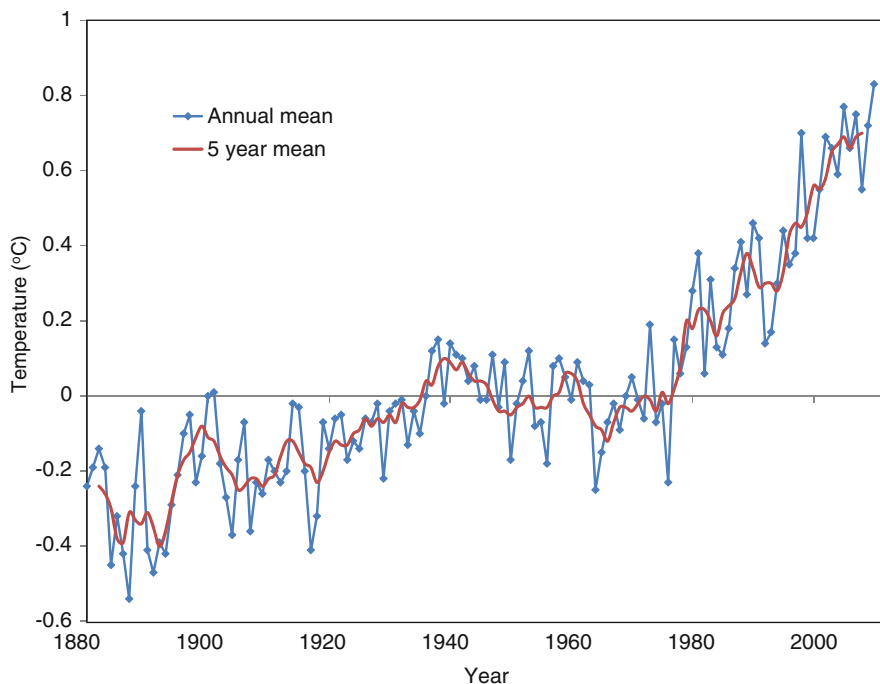
Greenhouse gas	Lifetime (years)	Radiative efficiency ( $\text{W m}^{-2} \text{ppb}^{-1}$ )	Time horizon (years)		
			20	100	500
Carbon dioxide ( $\text{CO}_2$ )	1.2	$1.4 \times 10^{-5}$	1	1	1
Methane ( $\text{CH}_4$ )	12	$3.7 \times 10^{-4}$	62	23	7
Nitrous oxide ( $\text{N}_2\text{O}$ )	114	$3.03 \times 10^{-3}$	275	298	153
Controlled by the Montreal Protocol					
CFC-11 ( $\text{CCl}_3\text{F}$ )	55	0.25	4,500	3,400	1,400
CFC-12 ( $\text{CCl}_2\text{F}_2$ )	116	0.32	7,100	7,100	4,100
CFC-13 ( $\text{CClF}_3$ )	640	0.25	10,800	14,400	16,400
Hydrofluorocarbons (HFCs)					
HFC-23 ( $\text{CHF}_3$ )	270	0.19	1,200	14,800	12,200
HFC-134a ( $\text{CH}_2\text{FCF}_3$ )	14	0.16	3,830	1430	435
HFC-152a ( $\text{CH}_3\text{CHF}_2$ )	1.4	0.09	237	124	38
Sulfur hexafluoride ( $\text{SF}_6$ )	3,200	0.52	16,300	22,800	32,600

(Source: Forster et al. 2007)

The HFCs are man-made chemicals developed as alternatives to ozone-depleting substances for industrial and consumer products, for example, HFC-134a, used in automobile air-conditioning and refrigeration. The PFCs are chemicals primarily produced from aluminum production and semi-conductor manufacture, while  $\text{SF}_6$  is gas used for insulation and protection of current interruption in electric power transmission and distribution equipment. Other GHGs include ozone ( $\text{O}_3$ ) continually produced and destroyed in the atmosphere by chemical reaction. Anthropogenic activities have increased ozone in the troposphere (i.e., the atmospheric layer closest to the Earth) through the release of gases such as CO, hydrocarbons, and NO which chemically reacts to produce ozone (Forster et al. 2007). Changes in atmospheric water vapor and  $\text{O}_3$  are climate feedbacks due to indirect effect of anthropogenic activities. Because GHGs absorb IR radiation, changes in their atmospheric concentration alters the energy balance of the climate system. Increase in atmospheric GHGs concentrations produces net increase in absorption of energy of the Earth, leading to warming of Earth's surface.

The characteristic absorption of  $\text{N}_2\text{O}$  in the IR radiation range of the atmospheric window of the Earth makes it a potent GHG. Since 1980s, a scientific consensus has emerged that human activities through increasing the concentration of GHGs in the atmosphere have intensified the natural greenhouse effect and set in motion a global warming trend (Fig. 1.2; IPCC 2001, 2007; USEPA 2007). For example,  $\text{CO}_2$  in the atmosphere has increased from about 280 ppm in pre-industrial era (1750) to the current 389 ppm (WMO 2011). Similarly, concentrations of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  have increased from 700 and 270 ppb in pre-industrial era to current levels of 1,808 and 324 ppbv, respectively (WMO 2011). This change represents an abundance of 139, 258, and 120 for  $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{N}_2\text{O}$ , respectively, relative to year 1750. The mean growth rate estimates of  $\text{N}_2\text{O}$  concentration in the atmosphere is at  $0.8 \text{ ppb year}^{-1}$  over the past 10 years (WMO 2011). Therefore, anthropogenic activities have





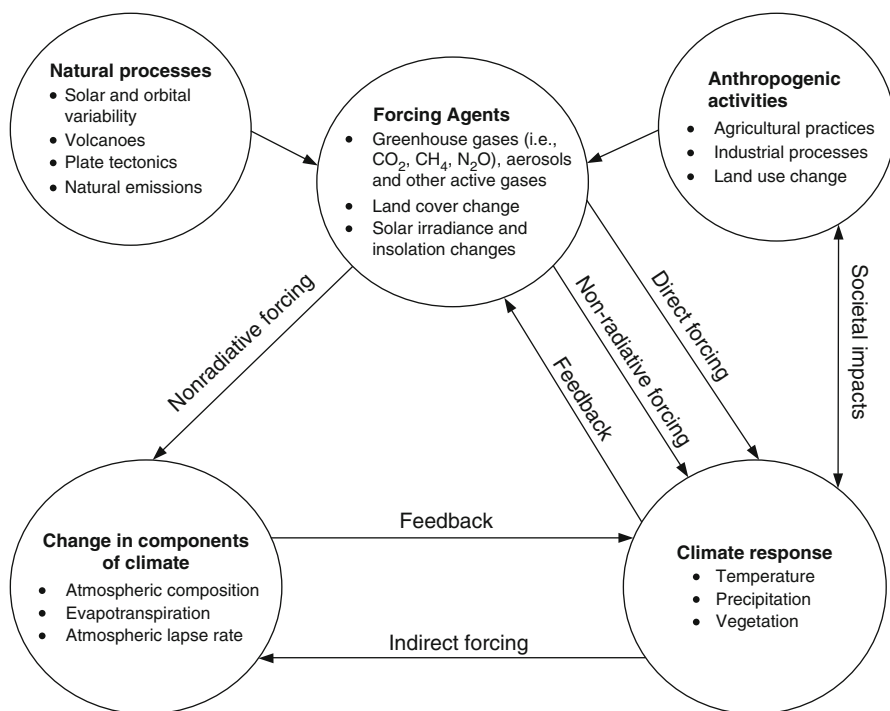
**Fig. 1.2** Global surface temperature from 1880 to 2010 based on the National Aeronautics and Space Administration Goddard Institute for Space Sciences (Data source: <http://data.giss.nasa.gov/gistemp/graphs>)

dramatically altered the chemical composition of the global atmosphere with great implications for current and future climate. Atmospheric theory predicts that changes in the concentration of trace gases will have dramatic consequences for the habitability of the earth, which may include (i) food insecurity, (ii) loss of biodiversity and ecosystems change (iii) destruction of the stratospheric ozone layer due to increase in  $\text{N}_2\text{O}$  and halogenated compounds, (iv) increase in the amount of tropospheric ozone due to increased emissions of  $\text{NO}_x$ , CO, and hydrocarbons. With the current trends, the earth is likely to warm by  $3\text{--}5^\circ\text{C}$  for the next century (Le Treut et al. 2007). This is as much as it has warmed since last ice age. Such a warming would have adverse impacts on ecosystems because ecosystems will not be able to adjust to such a rapid temperature changes.

Although the atmospheric concentrations of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  are much lower than that of  $\text{CO}_2$ , they each make a disproportionate contribution to atmospheric anthropogenic greenhouse effect in relation to their concentrations in the atmosphere. Methane contributes some 15%, and  $\text{N}_2\text{O}$  about 6% of the effect, making them the second and third most important GHGs after  $\text{CO}_2$ . This is because  $\text{CH}_4$  has a global warming potential 23 times and  $\text{N}_2\text{O}$  is 298 times that of  $\text{CO}_2$  on 100-year timescale (Table 1.1).

### 1.3 Climate Forcing

Human beings are now the prime drivers of global changes, which include climate change, reduced water quality, loss of diversity, and degraded ecosystem services – i.e., the services provided by nature for free, including fertile soils, clean air, pollination, and water purification (Rockström et al. 2009). The anthropogenic drivers include growing population, and changing per capita consumption patterns that affect food, feed, fiber production, land use change, and energy use. Climate change may be due to natural internal processes or external forcings, or persistent anthropogenic changes in the composition of the atmosphere or in land use (Fig. 1.3). Perturbations to energy balance of the earth system drives the climate change. Factors that affect climate change can be separated into forcings and feedbacks. The concept of climate forcing was introduced in the First Scientific Assessment Report (FAR) of the Intergovernmental Panel on Climate Change (IPCC 1990), and in the original framework and later text climate forcing and radiative forcing have been used synonymously. It is an energy imbalance (or radiative imbalance) imposed on the climate system either externally or by human activities. Examples of forcings include change in volcanic emissions,



**Fig. 1.3** Framework of radiative forcing, climate response and feedbacks of the climate change variables (Modified from NRC 2005; Liepert 2010)

land use modifications, changes in emissions of GHGs, aerosols and solar energy output (Fig. 1.3). The processes that amplify or dampen the initial forcing determine the relationship of radiative imbalance and climate response (Liepert 2010), and they are referred to as climate feedbacks. Climate feedback is the internal climate processes that amplifies or diminish the climate response to the forcing (NRC 2005). Positive feedback processes amplify response of the climate forcing. Examples of feedbacks include changes in atmospheric water vapor caused by increased warming associated with increase in GHGs concentration, which then amplify the warming through the greenhouse properties of water vapor. Warming causes ice to melt, revealing a darker land and water surfaces that absorbs more heat and causing more warming. Climate forcing can be radiative or nonradiative, and radiative forcing can be either direct or indirect. Direct radiative forcings influences the radiative budgets directly (e.g., added GHGs) which absorbs and emit IR radiations. Indirect radiative forcings causes radiative imbalances by first changing climate system components, which then lead to changes in radiative fluxes. One example is increase in evapotranspiration flux. Nonradiative forcings creates energy imbalance that have no direct effects on radiation (NRC 2005).

### ***1.3.1 Radiative Forcing***

Radiative forcing (RF) is a concept developed to provide a conceptual framework of understanding and quantifying Earths' energy budget modifications and their potential impacts on surface temperatures response (Hansen et al. 1997; Myhre et al. 2001). The effectiveness of climate forcings has practical importance because of the need to assess and compare the climate impacts of different changing atmospheric constituents. Strategies to reduce global warming can benefit from understanding of potential of different forcings in altering global temperature (Hansen et al. 2005). The RF quantifies the anthropogenic and natural influences on the climate system (Unger et al. 2010). The RF is a measure of how the energy balance of the Earth-Atmosphere system responds when factors that affect climate are altered. Factors affecting climate can be either natural or anthropogenic in origin. Natural factors include solar changes and volcanic eruption. In addition to cyclic changes in solar radiation that follow 10–11 year cycle, solar output has increased gradually in the industrial era, causing a small positive RF, warmer climate, and atmospheric abundance of stratospheric ozone (Forster et al. 2007). Volcanic eruptions can create a short-term (2–3 years) negative forcing through the temporary increase in sulfate aerosol in the stratosphere. Aerosol particles reflect and/or absorb solar and IR radiation in the atmosphere. Aerosols also change the cloud properties. Anthropogenic factors include increase in GHGs, increase in tropospheric ozone, decrease in stratospheric ozone, increase in aerosol particles in atmosphere, change in land cover and atmospheric contrails produced by aircrafts. The positive RF associated with increase in GHGs is the best understood of the anthropogenic RF, and they are positive because of absorption of IR

**Table 1.2** Simplified formulas for calculating radiative forcing due to carbon dioxide, methane, nitrous oxide, and halocarbons

Greenhouse gas	Formula for radiative forcing, RF ( $\text{W m}^{-2}$ )	Abundance of GHG in year 1750	Constant
Carbon dioxide ( $\text{CO}_2$ )	$RF = \alpha \ln\left(\frac{C}{C_0}\right)$	$C_0 = 278$ ppmv	$\alpha = 5.35$
Methane ( $\text{CH}_4$ )	$RF = \beta(\sqrt{M} - \sqrt{M_0}) - [f(M, N) - f(M_0, N_0)]$	$M_0 = 700$ ppbv	$\beta = 0.036$
Nitrous oxide ( $\text{N}_2\text{O}$ )	$RF = \epsilon(\sqrt{N} - \sqrt{N_0}) - [f(M, N) - f(M_0, N_0)]$	$N_0 = 270$ ppbv	$\epsilon = 0.12$
CFC-11	$RF = \lambda(X - X_0)$	$X_0 = 0$	$\lambda = 0.25$
CFC-12	$RF = \omega(X - X_0)$	$X_0 = 0$	$\omega = 0.32$

Modified from Ramaswamy et al. (2001)

$$f(M, N) = 0.47 \ln \left[ 1 + 2.01 \times 10^{-5} (MN)^{0.75} + 5.31 \times 10^{-15} M(MN)^{1.52} \right]$$

C is  $\text{CO}_2$  in ppm, M is  $\text{CH}_4$  in ppbv, N is  $\text{N}_2\text{O}$  in ppbv and X is CFC in ppbv

radiation. Changes in land cover, principally through forest clearing for cropland has modified the reflective properties of land. Anthropogenic activities have also modified the reflective properties of ice and snow. The overall anthropogenic impact has increased the reflection of solar radiation from the earth. The larger the forcing is, the larger the disruption of steady global surface temperature. It is defined as the change in incoming energy minus outgoing energy at the top of the atmosphere in response to a factor that changes energy balance with reference to unperturbed values (i.e., net energy flux difference at the top of troposphere caused by the imposed change in the pollutant relative to unperturbed initial state) (Ramaswamy et al. 2001; Forster et al. 2007). Mathematically, RF can be expressed as (Ramaswamy et al. 2001):

$$\Delta T_s = \lambda RF \quad (1.1)$$

where  $\lambda$  is the climate sensitivity parameter,  $\Delta T_s$  is the global mean temperature response,  $RF$  is radiative forcing. A number of radiative forcings of different origin and sign may exist together and to a first order, they can be added to yield a total forcing. Table 1.2 presents the simplified formulas for calculating RF for most common GHGs.

The year 1750 is a baseline for pre-industrial era benchmark for investigating changes in climate system associated with anthropogenic perturbations. The RF is quantified as a rate of energy change per unit area of a globe measured at the top of the atmosphere (troposphere). It is expressed in Watts per square meter ( $\text{W m}^{-2}$ ). It is the term used to denote externally imposed perturbation in the radiative energy budget of the earth's climate system. In an equilibrium climate state, the average net radiation at the top of the atmosphere is zero. A change in either solar or infrared radiation changes the net radiation. The corresponding imbalance is radiative forcing. When RF from a factor or a group of factors is positive, the energy of the earth-atmosphere system will increase, leading to a warming. On the other hand, negative forcing leads to decrease in energy and cooling of the system (Serreze 2010).

**Table 1.3** Concentrations and radiative forcing of the anthropogenic gases since industrialization to 2010

Component	2005	1750–2005 <sup>a</sup>	2010	1750–2010 <sup>b</sup>
	Concentration	Radiative forcing (W m <sup>-2</sup> )	Concentration	Radiative forcing (W m <sup>-2</sup> )
Carbon dioxide (CO <sub>2</sub> )	377.1 ppmv	1.66 ± 0.17	390 ppmv	1.791
Methane (CH <sub>4</sub> )	1,783 ppbv	0.48 ± 0.05	1,808 ppbv	0.504
Nitrous oxide (N <sub>2</sub> O)	318.1 ppbv	0.16 ± 0.02	324 ppbv	0.175
CFC <sub>12</sub> (CCl <sub>2</sub> F <sub>2</sub> )	541.7 pptv	0.173	530.1 pptv	0.170
CFC <sub>11</sub> (CCl <sub>3</sub> F)	250.9 pptv	0.063	240.2 pptv	0.060
15 minor long-lived halogenated gases <sup>c</sup>	–	0.092	–	0.105
Total	–	2.64	–	2.81

Notes: <sup>a</sup>Data from IPCC 2007<sup>b</sup>Data extracted from Butler (2011).<sup>c</sup>15 minor long-lived halogenated gases are: CFC-113 (CCl<sub>3</sub>FCClF<sub>2</sub>), CCl<sub>4</sub>, CH<sub>3</sub>CCl<sub>3</sub>, HCFC-22 (CHClF<sub>2</sub>), HCFC-141b (CH<sub>3</sub>CCl<sub>2</sub>F), HCFC-142b (CH<sub>3</sub>CClF<sub>2</sub>), HFC-134a (CH<sub>2</sub>FCF<sub>3</sub>), HFC-152a (CH<sub>3</sub>CHF<sub>3</sub>), HFC-23 (CHF<sub>3</sub>), HFC-143a (CH<sub>3</sub>CF<sub>3</sub>), HFC-125 (CHF<sub>2</sub>CF<sub>3</sub>), SF<sub>6</sub>, halon-1211 (CBrClF<sub>2</sub>), halon-1301 (CBrF<sub>3</sub>) and halon-2402 (CBrF<sub>2</sub>CBrF<sub>2</sub>)

Radiative forcing assesses and compares the anthropogenic and natural drivers of climate change. It is an important concept for prediction of surface temperature change, particularly for comparative studies of different forcings (Hansen et al. 1997). This concept originates from the studies of climatic response to changes in solar insolation and CO<sub>2</sub> using simple radiative convective models. It is applicable for assessment of climatic impact of long-lived GHGs (Ramaswamy et al. 2001). The importance of this concept is that it enables quantification and assessment of various factors that shift the energy balance relative to other factors. Radiative forcing is a simple measure for qualifying and ranking many different influences on climate change since it does not attempt to represent the overall climate response. It has advantage of being easier to calculate and to compare than estimates of the climate response, since climate response and sensitivity to external forcing are not easily quantifiable (Hansen et al. 2005; Colman and McAvaney 2011). The natural RF agents are solar changes and volcanoes. Anthropogenic activities contribute to RF through changing the amount of GHGs, aerosols and cloudiness. However, RF does not indicate how the climate will actually change due to particular forcing factor. Table 1.3 presents the RF of some of the common anthropogenic GHGs. The values reflect the forcing relative to start of industrial era in 1750s. The forcings of the greenhouse gases (e.g., CO<sub>2</sub>, N<sub>2</sub>O and CH<sub>4</sub>) are the best understood of those due to human activities and are all positive because each of these gases absorbs outgoing IR radiation in the atmosphere. Among the anthropogenic long-lived GHGs, increases in CO<sub>2</sub> concentrations have caused the largest forcing during post-industrial era (Table 1.3). Change in the mixing ratio of N<sub>2</sub>O from 270 ppbv in 1750 to 319 ppbv in 2005 results in RF of +0.16 ± 0.02 Wm<sup>-2</sup> (IPCC 2007;

Table 1.3). The RF of  $N_2O$  has increased by 11% since 1998. As CFC-12 levels slowly decline in response to regulation of ozone depleting gases (IPCC/TEAP 2005),  $N_2O$  with its current emission trends has taken over the third place in the ranking of the long-lived GHGs radiative forcings (Table 1.3). Increase in tropospheric ozone concentration also contributes to warming, while decrease in stratospheric ozone concentration contributes to cooling. Aerosol particles influence RF both through reflection and absorption of solar and IR in the atmosphere. Some aerosols can cause negative forcing, resulting into cooling, while others causes positive forcing contributing to warming. The overall effect of aerosols warming is negative (i.e., cooling), which is further intensified by changes they cause in cloud properties.

The RF is a useful tool for estimating the relative global impact of different climate change mechanisms (Ramaswamy et al. 2001); and is particularly useful in estimating the relative equilibrium of average temperature change due to different forcing agents. However, RF alone cannot assess the potential climate change associated with the emissions, since it does not take into account the lifetime of the forcing agents. With growing scientific insight, several attempts have been made since FAR to improve the RF concept by re-evaluating its definition (Forster et al. 2007) by including in it several other feedback processes. In addition, other simple metrics quantify the contribution of individual compounds to climate change. The Global Warming Potential (GWP) is the most established index that has been used in all past climate assessments (IPCC 1990, 2007; Ramaswamy et al. 2001).

### 1.3.2 Global Warming Potentials

Three characteristics of a gas determine its contribution to greenhouse effect, namely, its: (i) absorptivity for IR radiation, (ii) atmospheric lifetime (Table 1.1), and (iii) concentration in the atmosphere (CAST 2004). Gas absorbs IR radiation in specific energy bands influenced by its chemical properties. In addition, there is an overlap in the radiation-energy absorption bands for different GHGs. Therefore, the net effect of increasing the concentration of any single gas depends on inherent characteristics of the gas and its interaction with the energy absorption bands of other GHGs. The lifetime determines how long the emissions of GHGs into the atmosphere will contribute to global warming. The absorptivity and atmospheric lifetime characteristics of gas(es) is generally combined into a single index called Global Warming Potential (GWP). The concentrations of GHGs in the atmosphere play a significant role in influencing its GWP. If GHG is not present at a sufficiently high concentration, or does not have a potential to reach sufficiently high concentration, then it is not important even if its GWP is high. Only a few trace gases have the combination of the three characteristics to contribute significantly to global warming (Table 1.1; Forster et al. 2007).

When dealing with multiple GHGs, it is necessary to have a simple means of describing the relative abilities of emissions of each GHG to affect RF and