

D. J. Hofmann *, J. H. Butler, E. J. Dlugokencky, J. W. Elkins, K. Masarie, S. A. Montzka, and P. Tans
NOAA Climate Monitoring and Diagnostics Laboratory
Boulder, Colorado

1. INTRODUCTION

The perturbation to radiative climate forcing which has the largest magnitude and the least scientific uncertainty is the forcing related to changes in long-lived and well mixed greenhouse gases, in particular carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O) and the halocarbons (mainly CFCs). All of these gases have been monitored around the world since the 1970's mainly by NOAA's Climate Monitoring and Diagnostics Laboratory (CMDL), in Boulder, Colorado, and its forerunner, the Geophysical Monitoring for Climatic Change (GMCC) program. This paper discusses these observations for the past 25 years, a period when NOAA/CMDL's global network was adequate to form reliable global averages.

The observed mixing ratios and growth rates are converted to instantaneous direct climate forcing with standard IPCC-recommended expressions to obtain the total direct forcing by all the long-lived greenhouse gases. The interannual change in radiative forcing indicates a high degree of variability, providing an annual index of greenhouse gas radiative forcing. This index is a measure of the interannual changes in conditions that affect carbon dioxide emission and uptake, methane and nitrous oxide sources and sinks, and the decline in the atmospheric abundance of ozone-depleting chemicals.

2. MONITORING NETWORKS

One of the larger surface-based networks of greenhouse gas measurements is that operated by the Climate Monitoring and Diagnostics Laboratory (CMDL) of the U.S. National Oceanic and Atmospheric Administration (NOAA). In 1973, shortly after NOAA was formed, baseline climate observatories were initiated by the NOAA Geophysical Monitoring for Climatic Change (GMCC) program. These included Mauna Loa Observatory on the Mauna Loa volcano in Hawai'i, where Charles Keeling first began continuous measurements of carbon dioxide during the International Geophysical Year (IGY) period (1957-1958). Air samples from the South Pole Station, which could be returned to the U.S. and analyzed for carbon dioxide, also became available during the IGY. A complete NOAA climate observatory was also implemented at the South Pole

in the early 1970's. Shortly thereafter, similar observatories, capable of monitoring all the important climate forcing species, were implemented at Barrow, Alaska and in American Samoa.

In addition to Observatories, NOAA/CMDL also operates a global cooperative flask air sampling program, which began in 1967 and now numbers 56 global locations and three commercial ship routes (<http://www.cmdl.noaa.gov/ccgg/flask/> and Figure 1).

Air samples are collected in pairs weekly and are analyzed in the Boulder, Colorado Laboratory for CO₂, CH₄, carbon monoxide (CO), hydrogen (H₂), N₂O, and sulfur hexafluoride (SF₆). In addition to the mixing ratios of these species, the relative concentration of stable isotopes of CO₂ and CH₄ are determined at the University of Colorado (INSTAAR).

The halocarbon and nitrous oxide components of the atmospheric greenhouse species are determined both with continuous measurements at five global sites, including the Baseline Observatories mentioned previously and a site in Colorado, and by weekly air sampling at nine sites since 1979 (Figure 1). The major halocarbon greenhouse gases CFC-11 and CFC-12 are measured as is N₂O. The minor gases measured include CFC-113, CCl₄, CH₃CCl₃, COS, HCFC-22, HCFC-141b, HCFC-142b, HFC-134a, the brominated compounds halon-1211, and halon-1301, and SF₆. Although some of the halogens are declining substantially and others are declining more slowly, owing to Montreal Protocol regulations on gases that deplete the ozone layer, replacements such as the HCFCs and HFCs are increasing. It is expected that the radiative forcing from halocarbons will remain important at least until the middle of the 21st century.

3. THE OBSERVATIONS

Global trends in the monthly mean atmospheric mixing ratios of the five major long-lived greenhouse gases (CO₂, CH₄, N₂O, CFC-12, CFC-11) are shown in Figure 2 from about 1978 through 2003. The weekly station data are used to create a smoothed north-south latitude profile in sine latitude space from which a global average is calculated.

Seasonal variations are clear in the CO₂ and CH₄ data where best fit curves are drawn through the data in order to more clearly show the trend. The growth rate of CO₂ has averaged about 1.5 ppm per year over the past 25 years.

* Corresponding author address: David J. Hofmann,
R/CMDL, 325 Broadway, Boulder, CO 80305, e-mail:
David.J.Hofmann@noaa.gov

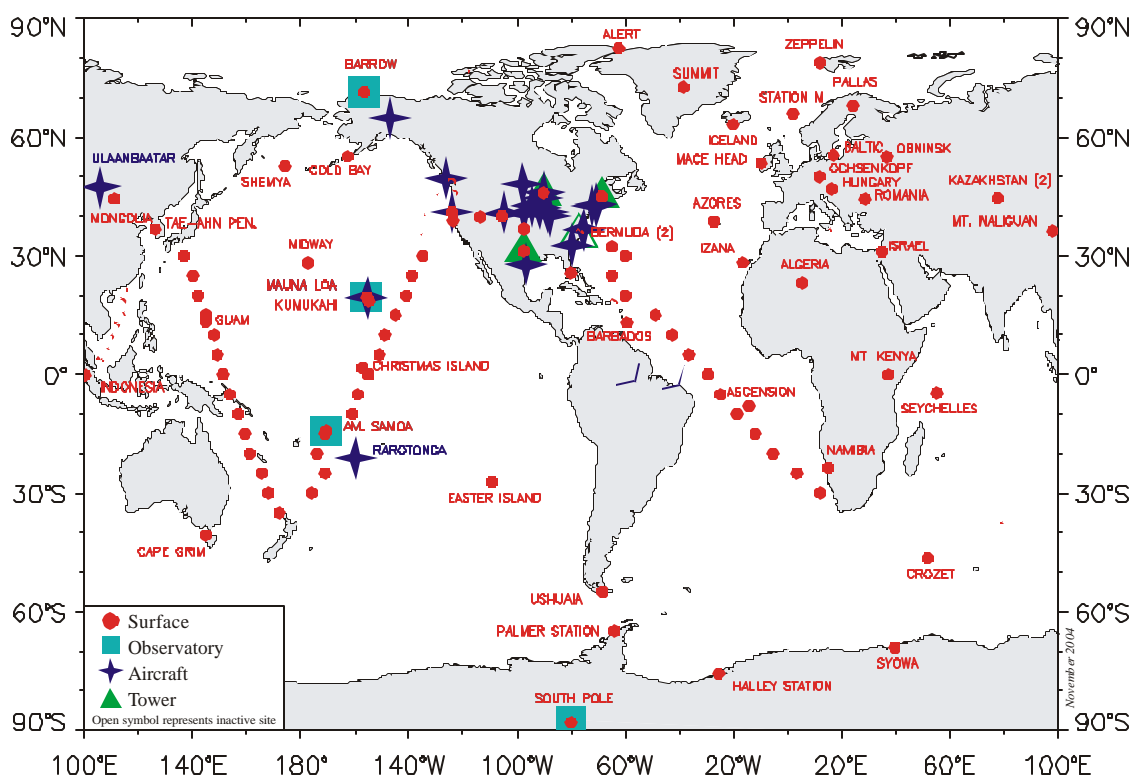


Figure 1. The NOAA/CMDL global cooperative air sampling network, including surface, ship, aircraft and tower sites.

The growth rate of methane has declined substantially since about 1992. The cause of this is likely related to several factors, including changes in emissions related to changes in the former Soviet Union and the short lifetime of methane (8-9 years) resulting in a pseudo-equilibrium between sources and sinks on this time scale (Dlugokencky et al., 1998, 2003).

Nitrous oxide continues to increase with a relatively uniform growth rate while the CFCs have ceased the increase observed before about 1992 and have either leveled off or are in decline (Montzka et al., 1999). The latter is a response to decreased emissions related to the Montreal Protocol on substances that deplete the ozone layer.

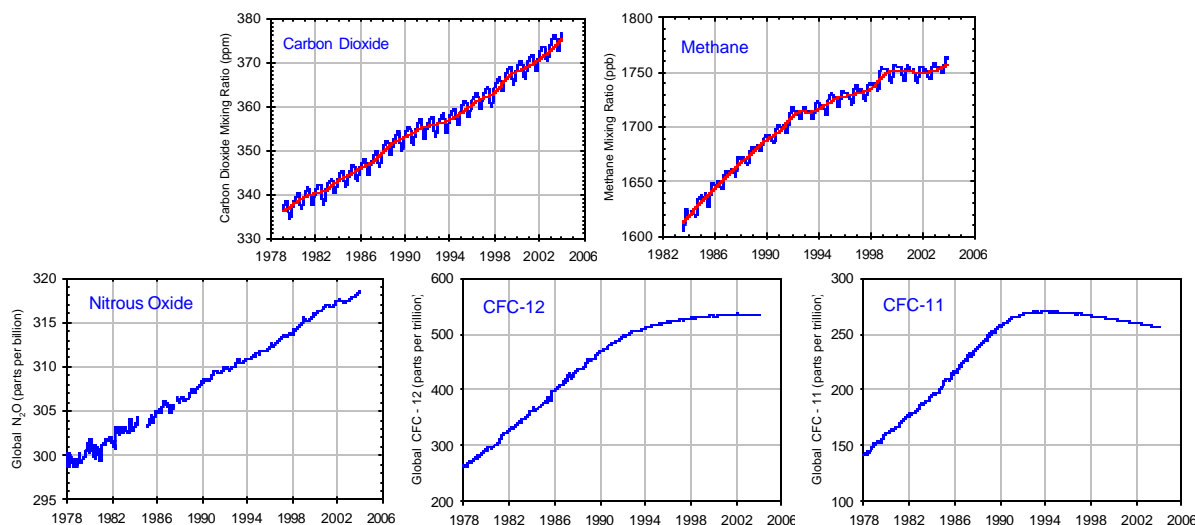


Figure 2. Global trends in major long-lived greenhouse gases through the year 2003. These five gases account for about 97% of the direct climate forcing by long-lived greenhouse gas increases since 1750. The remaining 3% is contributed by an assortment of 10 minor halogen gases, mainly HCFC-22, CFC-113 and CCl_4 .

4. RADIATIVE FORCING CALCULATIONS

To determine the total radiative forcing of the greenhouse gases, we have used IPCC (2001) recommended expressions to convert greenhouse gas changes, relative to 1750, to instantaneous radiative forcing. This allows for a year-by-year assessment of the change in the total direct radiative forcing by all the long-lived greenhouse gases. This is not a measure of the integrated climate forcing which can only be determined through an atmospheric radiative transfer model. In addition, only the direct forcing has been calculated. Model-dependent feedbacks, for example, due to water vapor and ozone depletion, have not been included. Contrary to climate models, these results are thus based mainly on measurements and have relatively high precision, for example, the estimated precision of the CO₂ measurement is 0.2 ppm (0.06%).

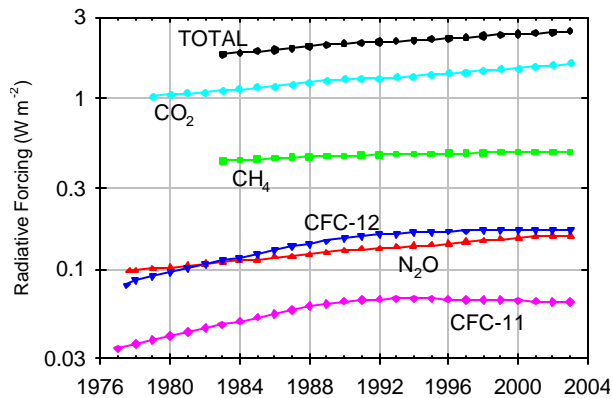


Figure 3. The direct radiative forcing by the major long-lived green-house gases based upon measured trace gas mixing ratios over time.

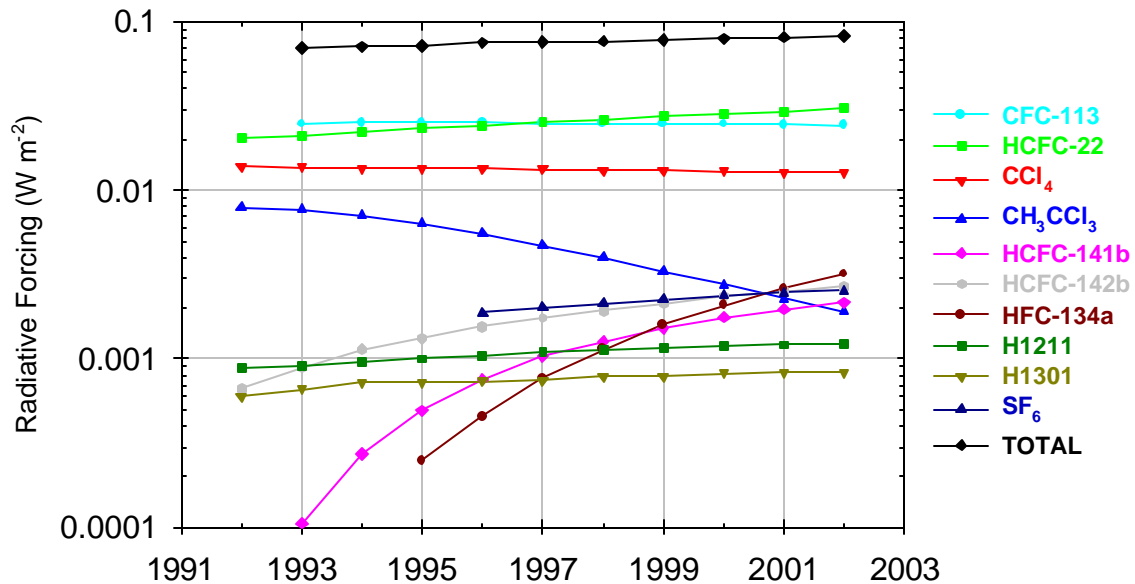


Figure 4. Direct radiative forcing due to ten minor gases, mostly measured in connection with the depletion of the ozone layer. The total forcing of all these gases is greater than that due to CFC-11 but smaller than that due to N₂O.

Table 1. Expressions for Calculating Radiative Forcing*

Trace Gas	Simplified Expression Radiative Forcing, (Wm ⁻²)	Const.
CO ₂	$A \ln(C/C_0)$	$A = 5.35$
CH ₄	$B (M^{1/2} - M_0^{1/2}) - [f(M, N_0) - f(M_0, N_0)]$	$B = 0.036$
N ₂ O	$D (N^{1/2} - N_0^{1/2}) - [f(M_0, N) - f(M_0, N_0)]$	$D = 0.12$
CFC-11	$E (X - X_0)$	$E = 0.25$
CFC-12	$F (X - X_0)$	$F = 0.32$

* IPCC (2001)

The subscript 0 denotes the preindustrial concentration

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

C is CO₂ in ppm, M is CH₄ in ppb

N is N₂O in ppb, X is CFC in ppb

4.1 The Major Gases

The expressions used for calculating the direct instantaneous radiative forcing for the major gases since 1750, from IPCC 2001, are given in Table 1.

Figure 3 shows the results from measurements beginning in 1977 and for the complete set of major gases, from 1983 to 2003.

4.2 The Minor Gases

Radiative forcing for a number of minor gases which are mainly monitored in connection with the recovery of the ozone layer, has also been determined. These include CFC-113, CCl_4 , CH_3CCl_3 , HCFC-22, HCFC-141b, HCFC-142b, HFC-134a, halon-1211, halon-1301 and SF_6 . The IPCC(2001)-recommended forcing constants were again used.

The results for the available data are shown in Figure 4. Most noticeable is the decline in forcing due to methyl chloroform (CH_3CCl_3), an industrial solvent which has been discontinued, and the rapid rise of some of the CFC replacements, HCFC-141b and HFC-134a, the latter used in automobile air conditioners. HCFC-22, used for large refrigeration applications, continues to increase slowly and appears to be driving the total increase of the minor gases.

4.3 Total Forcing by the Long-lived Gases

Figure 5 shows the total direct instantaneous forcing by all the long-lived greenhouse gases from 1983-2003. The CFCs and minor gases have been combined to form a "halogenated" group so that there are four classes. As expected, CO_2 dominates the total forcing with methane and the CFCs becoming smaller contributors to the total forcing over time.

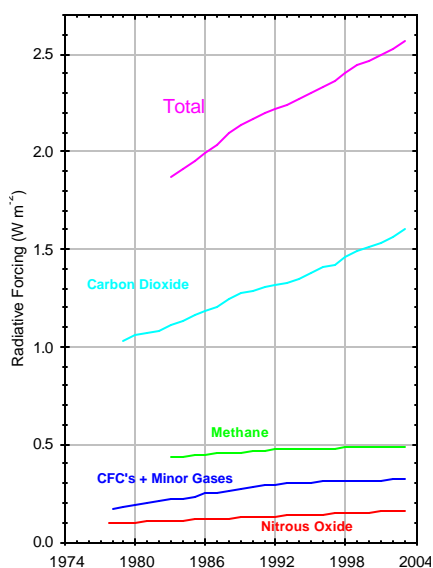


Figure 5. Instantaneous direct radiative forcing of four classes of long-lived greenhouse gases from the NOAA global network.

The increasing dominance of climate forcing by CO_2 can be seen in Figure 6 where the percentage of the total radiative forcing for the four classes of long-lived greenhouse gases is shown. Since 1979, the instantaneous radiative forcing by CO_2 has increased

from 59% to 62% of the total forcing by long-lived greenhouse gases.

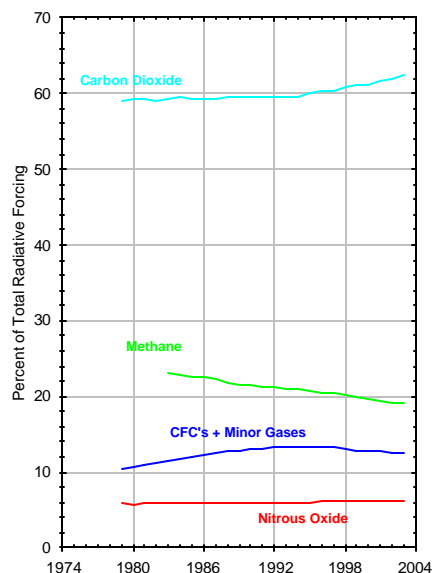


Figure 6. Percentage of the total instantaneous radiative forcing of four classes of long-lived greenhouse gases.

5. INTERANNUAL VARIABILITY IN RADIATIVE FORCING

Although the direct radiative forcing by long-lived greenhouse gases appears to increase relatively uniformly, there are interannual variations arising predominantly from CO_2 and CH_4 that are not well understood. Explaining this variance from year to year would provide an important step in improving climate projections.

The gases which cause the interannual variability can be determined from Figure 7 where the annual radiative forcing change is given for CO_2 , CH_4 , N_2O and the CFCs. Changes in CO_2 are the source of the largest magnitude interannual changes. The causes of these changes is a subject of current study.

6. CONCLUSIONS

Of the four major groups of long-lived greenhouse gases that contribute to radiative climate forcing, CO_2 and N_2O are the only ones that continue to increase at a regular rate. The contribution to radiative forcing by methane and CFCs has been declining in recent years with CO_2 now amounting to about 62% of the total direct radiative forcing.

The interannual variation in the total forcing is large and is the basis for an annual greenhouse gas index which is a measure of the interannual changes in CO_2 emissions and environmental conditions that affect CO_2 uptake, in CH_4 and N_2O sources and sinks, and in the decline in atmospheric levels of ozone-depleting chemicals in response to the Montreal Protocol.

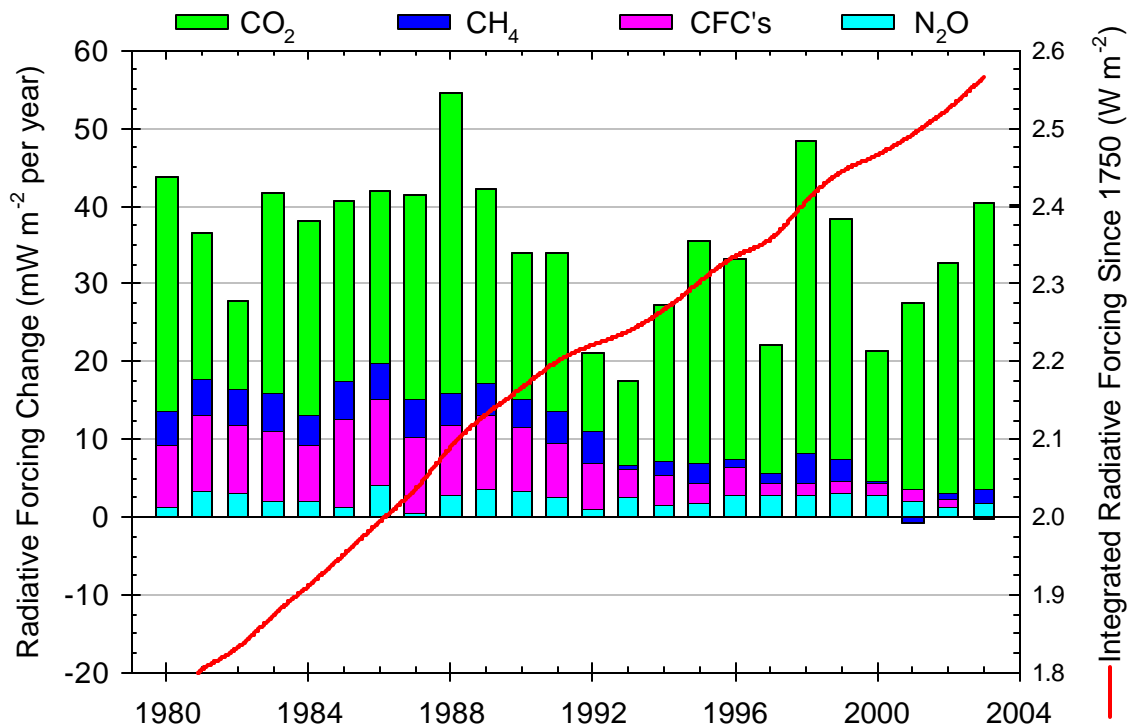


Figure 7. The interannual change in the radiative forcing due to changes in CO_2 , CH_4 , N_2O and the CFCs. The integrated total radiative forcing (red curve) is also shown. The growth rates of CH_4 and the CFCs were negative in 2001 and 2003, respectively.

References

- Dlugokencky, E. J., K. A. Masarie, P. M. Lang, and P. P. Tans, 1998: Continuing decline in the growth rate of the atmospheric methane burden, *Nature*, **393**, 447-450.
- Dlugokencky, E. J., S. Houweling, L. Bruhwiler, K. A. Masarie, P. M. Lang, J. B. Miller, and P. P. Tans, 2003: Atmospheric methane levels off: Temporary pause or a new steady-state?, *Geophys. Res. Lett.*, **19**: doi:10.1029/2003GL018126.
- Montzka, S. A., J. H. Butler, J. W. Elkins, T. M. Thompson, A. D. Clarke, and L. T. Lock, 1999: Present and future trends in the atmospheric burden of ozone-depleting halogens, *Nature*, **398**, 690-694.
- IPCC (2001) : *Climate Change 2001: The Scientific Basis*. Cambridge Univ. Press, Cambridge UK and New York, NY USA.