Title

Appendix D: CO2 Equivalency Factors: An Appendix to the Report, “A Lifecycle Emissions Model (LEM): Lifecycle Emissions From Transportation Fuels, Motor Vehicles, Transportation Modes, Electricity Use, Heating and Cooking Fuels, and Materials”

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APPENDIX D: CO_2-EQUIVALENCY FACTORS

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Note: Most (although not all) of the material in the sections "Brief Overview..." and "Major Emission Sources..." is from the mid 1990s, and does not reflect the most recent findings of the Intergovernmental Panel on Climate Change (IPCC). The reader is referred to IPCC (2001) for more current and comprehensive information. The material in the other sections does reflect the most recent information from the IPCC and other sources.

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BRIEF OVERVIEW OF GREENHOUSE GASES AND CLIMATE CHANGE

Human activity is altering the atmospheric concentrations and distributions of greenhouse gases (GHGs) and aerosols. Although to some extent such changes have occurred for centuries, at no time in human history have they been as great as they are at present. These changes in gas and aerosol concentrations have the potential to affect earth's climate by altering the reflection or absorption of incoming solar radiation and the emission or absorption of terrestrial radiation.

In general, the energy that enters the earth / atmosphere system from the sun is balanced by the energy that leaves the system. However, it is possible for the level of energy that is retained in different components of the earth / atmosphere system to change over time. The Intergovernmental Panel on Climate Change (IPCC) has defined the term *radiative forcing* to refer to a change in the energy that is available to the earth / atmosphere system (IPCC, 1996).

Some gases have a direct effect on climate by reducing the efficiency with which energy in the atmosphere is lost to space. These gases absorb outgoing terrestrial radiation and re-radiate energy in all directions, and this tends to warm the atmosphere and the surface of the earth. This effect has come to be known as the enhanced greenhouse effect, because it is an enhancement of the effect that has occurred naturally for millions of years. Without the greenhouse effect, the temperature on the surface of the earth would be much colder, and most species of life that currently proliferate would be unable to survive. Gases that provide this direct effect on climate include water vapor, CO_2, O_3, CH4, and N_2O.

In addition to these gases, some other gases influence climate indirectly by altering the concentrations of the direct GHGs through chemical reactions. In some cases, the relevant chemical reactions proceed very quickly (such as for the reactions that produce tropospheric O_3), while in other cases it may take many months for the final effects to be realized (such as for the destruction of stratospheric O_3 by CFCs).

Atmospheric aerosols cause other types of climatic effects. Aerosols are tiny particles suspended in the atmosphere, derived primarily from the combustion of fossil fuels and biomass, and from volcanic eruptions. They have very complex effects on climate because they can either reflect or absorb incoming solar radiation, depending on their specific characteristics, and also because they exert an indirect effect through their role in cloud formation. There is considerable uncertainty at present about the net climatic effects of different aerosol species.

Table D-1 presents the gases that are currently believed to influence climate, along with the nature of their influence. With regard to Table D-1, it is important to note that water vapor (H_2O) is actually the main climate-controlling gas. However, H_2O emissions from fossil-fuel use worldwide are a tiny fraction of global evaporation (about 0.0013%). Thus, anthropogenic emissions of H_2O are not very significant, and probably can be ignored. (I ignore them here.) It is also important to note that while CFCs have a very strong effect on radiative forcing, they are being phased-out under the Montreal Protocol. Replacements for CFCs, such as HCFCs, tend to have lower, although still significant, net positive radiative forcing (IPCC, 1996).

MAJOR EMISSION SOURCES OF GHGS AND AEROSOLS

Human activities have become important sources of GHG emissions to the atmosphere relatively recently. In past centuries, the human influence on atmospheric concentrations of GHGs was primarily limited to the effects of land conversion, livestock cultivation, and biomass burning. Today, however, motor vehicle use, industrial processes, and many other minor sources are present. Concern about these emission sources has been the primary motivation for the Kyoto Protocol to limit the growth in GHG emissions.

Table D-2 shows some basic figures related to the pre-industrial and present concentrations of a few GHGs, along with an estimation of the current rate of change. In the following sections, the major sources of the main GHGs are discussed.

Carbon dioxide

Carbon dioxide (CO_2) emissions have increased from about 280 ppmv in pre-industrial times to approximately 360 ppmv at present. Prior to the recent increase in CO_2 concentration levels, over the last 1000 years the concentration of CO_2 hovered around 280 ppmv, changing only by about ±10 ppmv. The major sources of CO_2 emissions include fossil fuel combustion, land-use conversions, and cement production (IPCC, 1996).

CO_2 concentrations over the past forty years have been well-characterized, primarily by data collected on the summit of Mauna Loa in Hawaii. Over the past 1000 years, CO_2 concentrations are reasonably well-characterized from ice core data. However, understanding the ultimate fate of the CO_2 that is released into the atmosphere is complicated by the wide range of different processes that act as sinks for CO_2, and the different time-scales under which these processes operate. Unlike some GHGs, whose removal is dominated by a single mechanism, CO_2 is removed by processes that act as quickly as a few years and as slowly as a century or more. The IPCC notes that 40-60% of the CO_2 that is released into the atmosphere is removed within 30 years, but that it is difficult to make exact predictions for the future because of the uncertain rates of deforestation and regrowth in the tropics and mid-latitudes, and because the mechanisms for CO_2 fertilization (the increase in crop growth due to higher levels of atmospheric CO_2) remain poorly quantified at the ecosystem level (IPCC, 1996) .

Methane

Atmospheric methane (CH_4) concentrations have increased since the early 19th century from a pre-industrial level of about 700 ppbv to the current level of 1721 ppbv, which is the highest level in at least 160,000 years (IPCC, 1996. CH_4 concentrations grew rapidly during the 1970's, due to an annual rate of increase of about 20 ppbv/yr., but the rate of increase declined through the 1980's to about 9 ppbv/yr. in 1991 (IPCC, 1996) . In 1992-1993, anomalously low CH_4 growth rates were observed, with virtually no net increases in some areas, but by 1994 the rate of growth rebounded to about 8 ppbv/yr. (IPCC, 1996) . The cause of the 1992-1993 anomaly is not yet fully understood, but may be due to a combination of the eruption of Mt. Pinatubo (which enhanced troposphere-stratosphere exchange through stratospheric heating), decreased emissions from fossil fuels in the Northern Hemisphere, and decreased biomass burning in the tropics (IPCC, 1996) . Thus, atmospheric concentrations of CH_4 are still increasing at a substantial rate, but not as rapidly as has been observed over the previous few decades.

Most CH_4 is known to come from the anaerobic fermentation of organic matter in rice paddies and swamps, from the fermentation of mammalian organic excrement, and from enteric fermentation in ruminants (Bolle, et al., 1986; Chamberlain, et al., 1982; Mooney, et al., 1987; Ramanathan, et al., 1985; Wahlen, et al., 1989; Watson, et al., 1990) . The primary pathway for CH_4 removal is through reaction with the hydroxyl radical ($OH^\bullet$), via the reaction: $CH_4 + OH^\bullet \text{-->} CH_3^\bullet + H_2O$. There is a positive feedback loop for accumulations of CH_4 in the atmosphere as additions of CH_4 to the troposphere can deplete OH levels, thereby reducing the rate of CH_4 removal (IPCC, 1996) . Increases in carbon monoxide levels can also deplete OH (via $2CO + 2OH^\bullet \text{-->} 2CO_2 + H_2$), and this can also decrease the rate of CH_4 removal (Chamberlain, et al., 1982; Stauffer, et al., 1985) . The use of fossil fuels accounts for roughly 20% of yearly global CH_4 emissions, and anthropogenic activities are in total responsible for 60-80% of current CH_4 emissions (IPCC, 1996) .

According to the Intergovernmental Panel on Climate Change (Shine, et al., 1990) , CH_4 could contribute more to global warming than any other non-CO_2 greenhouse gas -- about 15% of the total warming over the next century. Molecule per molecule, CH_4 has about 21 times the radiative forcing of CO_2, although it also has a much shorter lifetime (IPCC, 1996) . When CH_4 is destroyed by the OH radical, it forms CO_2 and H_2O, both of which are greenhouse gases. Also, an increase in the concentration of CH_4 may result in increases in O_3, which is a greenhouse gas as well (Ramanathan, 1988) .

Nitrous oxide

N_2O is a potent greenhouse gas that is formed in complex ways in combustion settings, and in soils and water where nitrogen is present. It may be the least well-understood greenhouse gas. The concentration of tropospheric nitrous oxide (N_2O) has been increasing at about 0.2%-0.3% / year over the last 40 years, from a preindustrial concentration of about 275 ppbv to a 1994 level of 312 ppbv (IPCC, 1996; Watson, et al., 1990) . The primary natural sources of N_2O are soils, with annual emissions of 3.7 to 7.6 Tg N per year, and oceans, with annual emissions of 1.4 to 2.6 Tg N per year (Kramlich and Linak, 1994) . The main anthropogenic sources of N_2O are from agriculture and a variety of industrial processes, such as adipic and nitric acid production. Total anthropogenic emissions are 3 to 8 Tg N per year (IPCC, 1996) . The combustion of fossil fuels appears to contribute only a minor amount to the global N_2O budget (and less than previously thought, as explained below), although uncertainties remain. N_2O is very stable in the troposphere, and as a result it has a long atmospheric lifetime of about 120 years (IPCC, 1996) . The main sink for N_2O is photolysis in the stratosphere, a process that accounts for about 90% of N_2O destruction (Kramlich and Linak, 1994; Watson, et al., 1990) .

N_2O has a per molecule radiative forcing over 200 times that of CO_2 (Watson, et al., 1990) . N_2O also is involved stratospheric O_3 chemistry. However, global emissions of N_2O emissions are small compared global emissions of CH_4 and carbon dioxide, and hence the contribution of N_2O to future warming is expected to be relatively small (Shine, et al., 1990) .

Ozone

Ozone (O_3) is an effective greenhouse gas both in the stratosphere and the troposphere (IPCC, 1992) . The concentration of O_3 near the earth's surface appears to be increasing, but there is no clear trend in the upper troposphere, where O_3 is most effective as a greenhouse gas (Johnson, et al., 1992; Watson, et al., 1990) . In the mid-to high-latitudes of the Northern Hemisphere an increase has been observed in the last two decades, particularly in the summer (Bolle, et al., 1986) , due at least in part to emissions of O_3 precursors from airplanes, industry, and vehicles.

An important source of tropospheric O_3 in the lower atmosphere in urban areas is the complex series of photochemical reactions involving reactive organic compounds (ROG), nitrogen oxides (NO_x), carbon monoxide (CO), hydroxyl radicals, and other compounds. (See the National Research Council (1991) for an excellent review of the science of tropospheric ozone pollution.) The major ozone precursors -- ROG, NO_x, and CO -- are emitted from highway vehicles and other fossil-fuel combustion sources.

Emissions of ozone precursors are relatively well characterized in the U.S. and many other countries, because concern over urban smog has led to the regulation and careful study of these pollutants. However, the atmospheric chemistry and activity of O_3 is complex, and much remains to be learned about the production and destruction of O_3 (all of the trace greenhouse gases considered here, and others, are involved); the movement of O_3 vertically and latitudinally within the troposphere, and between the stratosphere and the troposphere; the relationship between changes in the vertical and latitudinal distribution of O_3 and the surface temperature (O_3, unlike the other trace gases, is not uniformly mixed in the troposphere); and the feedback effect on temperature of increasing temperature, changes in precipitation and concentrations of other trace gases.

CFCs, HCFCs, and HFCs

Chlorofluorocarbon compounds (CFCs) are important environmentally in two ways: they are strong infrared absorbers, and they destroy O_3 in the stratosphere. A CFC molecule has several thousand times the direct radiative forcing of a CO_2 molecule, and can persist for centuries before being photodissociated in the stratosphere (Watson et al., 1990). However, CFCs also have indirect effects on climate, and these tend to cause global cooling. The chlorine released by the decomposition of CFCs destroys odd oxygen (O or O_3), and thereby depletes O_3 levels throughout the stratosphere. Because O_3 is a radiatively active greenhouse gas, the reduction in O_3 levels in the lower stratosphere (especially near the tropopause) reduces the radiative forcing of the surface-troposphere systems (IPCC, 1996) . As Ramaswamy et al. (1992) note, this cooling may counterbalance the direct warming effect of CFCs. However, even if the indirect and direct effects of CFCs on climate roughly cancel each other as a global average, the cooling effects of CFCs have strong latitudinal gradients, and further increases in CFCs can therefore affect climate at the regional level (Wuebbles, 1994) .

The concentration of CFCs increased from a pre-industrial level of 0 to nearly 1 ppb in 1992 (IPCC, 1996) . The concentration of CFC-12, the most abundant of the CFCs, was increasing at about 6%/year during the early 1980's (WMO, 1985) , but due to phase-out efforts required by the Montreal Protocol on Substances that Deplete the Ozone Layer, and subsequent amendments thereto, the rate of increase began to drop in the early 1990's, and by 1995 was about zero (IPCC, 1996). Still, all of the CFCs are long lived (e.g., over 100 years for CFC-12), and so will persist for quite some time even if net emissions reach zero (IPCC, 1996).

CFCs are used mainly as refrigerants, blowing agents for foam packaging and materials, and solvents (EIA, 1997). The EIA (1997) estimates 1995 emissions of 52,000 metric tons CFC-12, 36,000 metric tons CFC-11, and 22,000 metric tons other CFCs. Automobile air conditioning systems are a major source of CFC-12.

In response to the Montreal Protocol, CFCs have been replaced in most end uses by compounds with little or no potential to deplete stratospheric ozone. Most of the replacements are hydrochlorofluorocarbons (HCFCs) or hydrofluorocarbons (HFCs). HCFCs are much less stable than CFCs, and hence less likely to migrate to the stratosphere where they can destroy ozone. However, they do still deplete stratospheric ozone, and consequently are controlled by the Copenhagen Amendments to the Montreal Protocol and are scheduled to be phased out by 2020 (HCFC-22) and 2030 (all other HCFCs) (EIA, 1997).

HFCs do not contain chlorine or bromine, and hence do not deplete stratospheric ozone. In mobile air conditioners, a hydrofluorocarbon, HFC-134a (tetrafluoroethane, CH_2FCF_3), has replaced[1] CFC-12. HFC-134a causes less direct radiative forcing than does CFC-12 (it has a similar infrared absorption band, but a lifetime of only about 15 years, compared to the 102 year lifetime of CFC-12), but because it does not deplete stratospheric O_3, it does not indirectly cause global cooling the way CFC-12 does. Consequently, the global warming potential of HFC 134a, while less than that of CFC-12, is still significant.

Aerosols and climate

Aerosols are small particles and droplets in the atmosphere. They may comprise dust particles, or of other particles and droplets with a wide variety of different chemical compositions. They can be produced through both natural processes, such as dust storms and volcanic activity, and anthropogenic processes, such as fossil fuel and biomass burning. Aerosols affect the radiative balance of the earth directly, by scattering and absorbing radiation, and indirectly, by modifying the amount, lifetime, and optical properties of clouds. Anthropogenically produced aerosols are found in the lower troposphere (below about 2 km), where they rapidly undergo chemical and physical changes, particularly through interactions with clouds. Aerosols are even more regionally concentrated than is tropospheric O_3, which in turn varies in concentration much more than the globally well-mixed greenhouse gases such as CH_4. The main removal mechanism for tropospheric aerosols is through precipitation, and as a result such aerosols have atmospheric lifetimes of only a few days. In contrast, some naturally occurring aerosols, particularly those produced through volcanic activity, can reach the stratosphere where they can persist for months or years (IPCC, 1996).

Considerable uncertainty surrounds many aspects of aerosol behavior, particularly with regard to indirect effects on cloud properties, and as a result the net climatic effect of aerosols is unclear. The direct effects of aerosols, in scattering and absorbing radiation, depend importantly on the details of atmospheric aerosol chemistry, aerosol spatial distribution, and the microphysical properties of the aerosol particles themselves (IPCC, 1996; Jacobson, 2001). Such properties as aerosol scattering efficiency and upscatter fraction depend in turn on particle size, composition, and relative humidity (Pilinis et al., 1995) . Due to the inhomogeneous spatial distribution of aerosols, and the importance of the details of this distribution to their climatic effects, estimates of globally averaged radiative forcing estimates are especially uncertain.

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[1] The November 1992 revisions to the Montreal Protocol required all CFCs to be phased out by January 1, 1996. However, some German automakers converted virtually all new production to HFC-134a in 1992-93 (Chemical and Engineering News, 1994; Reichelt, 1993) , and by 1995 U. S. and Japanese automakers had followed suit (EIA, 1997; Walsh, 1993). At the same time, though, automakers have been looking for long-term, climate-friendly alternatives to HFC-134a (Reichelt, 1993) .

Some progress has recently been made in understanding the behavior of tropospheric aerosols. Estimates by the IPCC (2001), Harvey (2000), and Jacobson (2001, 2002, 2003) are presented later in this appendix. Recent work suggests a positive direct radiative forcing of the black carbon component of aerosols that is more than had been estimated previously. Jacobson (2001) estimates that black carbon (soot) from fossil-fuel and biomass combustion has a direct forcing of 0.55 W/m^2, assuming that the black carbon ends up as mainly as coated particle core aggregated with other aerosol components. He concludes that "the warming effect from black carbon may nearly balance the net cooling effect of other anthropogenic aerosol constituents" (p. 695). More recently, Jacobson (2002) has estimated that overall radiative forcing due to fossil-fuel black carbon and organic matter (in a "with versus without" analysis) is 0.50 W/m^2. However, Chang's (2002) three-dimensional aerosol-climate model shows that black-carbon emissions cause increases in surface temperature in some regions but decreases in others, with no easily discernible mean global tendency.

The complex and incompletely understood effects of aerosols on climate suggests that estimates of direct forcing effects from anthropogenic sulfate aerosols, such as are reviewed here, are uncertain by more than a factor of two, and that estimates of the direct forcing effects of anthropogenic soot aerosols are uncertain by at least a factor of three (IPCC, 1996; Nemesure et al., 1995) .

The indirect effects of aerosols on cloud properties are even less well understood. Certain types and sizes of aerosol particles are well known to serve as cloud condensation nuclei, and it is believed that aerosol particles can modify cloud droplet size distributions, and thus the optical and radiative properties of clouds (Hobbs, 1993; IPCC, 1996) . The evidence collected to date suggests that the indirect effects of aerosols results in a negative radiative forcing, but the number of processes involved in determining the ultimate distribution of cloud droplet sizes makes it very difficult to directly relate aerosol emissions to the radiative properties of clouds. Recent studies have confirmed that the indirect effects of aerosols have caused a substantial negative forcing since pre-industrial times, and despite the remaining uncertainties the IPCC (1996) suggests a negative radiative forcing in the range of 0 to -1.5 Wm^{-2} and uses an average value of -0.8 Wm^{-2} for aerosol indirect effects in some scenario analyses. The results from eight published studies listed in Harvey (2000) range from - 0.55 W/m^2 to – 4.8 W/m^2, although most of the studies fall between – 0.6 and – 1.5, with an average of – 0.91 W/m^2 ignoring the two presumably anomalously highest results. (For purposes of comparison, note that the eruption of Mt. Pinatubo produced a global mean radiative forcing effect of -3 to -4 Wm^{-2} for two to three years [IPCC, 1996].)

The inclusion of aerosols in climate models has been a recent focus of attention, and a study by the Hadley Centre for Climate Prediction and Research indicates that including aerosols along with greenhouse gases provides a better fit of model prediction to reality (Mitchell, et al., 1995).More recent work by Jacobson (2002) confirms this. The study concludes that the omission of sulphate aerosols from previous models may help to explain why model predictions of warming have typically been larger than warming actually observed, and particularly why warming has been substantially lower than predicted over the northern mid latitudes (Mitchell, et al., 1995). Other research suggests that aerosols may be offsetting a substantial portion of the global mean response to greenhouse gas induced forcing, but that representing the spatial and seasonal distribution of aerosols is important to understanding climatic responses at more detailed levels than the global and annual mean (Cox, et al., 1995) .

ESTIMATING CO_2 EQUIVALENCY FACTORS (CEFS): BACKGROUND

Introduction

In order to estimate the combined impact of emissions of all of the different greenhouse gases, mass emissions of the non-CO_2 greenhouse gases are converted into the mass amount of CO_2 emissions that would have the same impact on some measure of interest – say, global climate or global economy. The first CO_2-equivalency factors (CEFs), developed around 1990, equated emissions solely on the basis of global warming (in terms of integrated radiative forcing), and hence were called “global warming potential” factors, or GWPs. More recently, researchers have estimated equivalency factors that equate emissions on an economic basis, in terms of the present dollar value of the impacts of climate change or the costs of controlling greenhouse-gas emissions. Table D-3 summarizes published estimates of GWPs and indices based on economic impacts (“damage-cost” indices) or control costs (“cost-effectiveness” indices).

Economic damage-cost indices (EDIs) potentially are an advancement over GWPs because they incorporate the present value of the economic damages of future global warming, whereas the GWPs reflect only the future radiative forcing. However, most of the EDIs developed thus far do have some shortcomings: those of Reilly (1992) are based on now-outdated estimates of climate effects; those of Hammit et al. (1996) do not include the indirect effects of CH_4 or halocarbons on H_2O or O_3, or the effects of CO_2 fertilization; and those of Tol (1999) include CO_2 fertilization but not indirect effects. Because of this, and because some of the issues involved in the calculation of GWPs also are involved in the calculation of EDIs (GWPs in essence are partial EDIs), I will discuss the calculation of GWPs as well as the calculation of EDIs in some detail.

After I discuss GWPs, EDIs, cost-effectiveness indices, and the discount rate, I present the methods and parameter values in our own estimation of CEFs. To estimate CEFs for gases with significant direct radiative forcing effects, I develop a method that is a hybrid of the methods for GWPs and EDIs. To estimate CEFs for gases that affect radiative forcing mainly indirectly, I use different methods that for the most part are original with us.

Gases with significant direct radiative forcing effects, considered in this analysis:

- CO_2 (carbon dioxide)
- CH_4 (methane)
- N_2O (nitrous oxide)
- chlorinated or fluorinated compounds (detailed below)
- O_3 (ozone)
- CO (minor direct radiative forcing effect)
- aerosols (several different kinds of aerosols; discussed below)

Gases that affect radiative forcing indirectly, considered in this analysis:

- CO (carbon monoxide)
- H_2 (hydrogen)
- NMOCs (non-methane organic compounds)
- SO_2 (sulfur dioxide)
- NO_X (nitrogen oxides)
- aerosols (several different kinds; discussed below)

Table D-1 summarizes the pollutants, climate effects, and estimated CEFs in this analysis. Table D-3 shows some recent estimates by others of GWPs and EDIs. In the following sections I review the construction of GWPs, EDIs, and cost-effectiveness indices before presenting the development of our own CEFs.

The calculation of GWPS.

To calculate a GWP, one needs to know, for both CO_2 and non-CO_2 gases, the relationship between radiative forcing and atmospheric concentration and the relationship between an increase in yearly emissions and the increase in the equilibrium atmospheric concentration. One also must consider interactions between gases (for example, CO and CH_4), and the ultimate fate of the gases (for example, CH_4 ends up being oxidized to CO_2 and H_2O by the OH^- radical). Finally, one must pick a period of time to do the analysis, because inasmuch as one is equating radiative forcing integrated over a period of time, the equation will depend on the length of time chosen. This choice is important.

Sophisticated estimates of GWPs are available in several papers (Derwent et al. 2001; Johnson and Derwent, 1996; IPCC, 1996; Lashof and Ahuja, 1990; Martin and Michaelis, 1992; Rodhe, 1990; Shine, et al., 1990; Wilson, 1990) . All papers use the same basic method, paralleling that used to develop ozone depletion potentials for CFCs. Generally, analysts distinguish between the “direct” warming effect of a greenhouse gas, which is due directly to the radiative forcing of the gas, and the “indirect” warming effect, which is due to the effect of the gas on the concentration of *other* radiatively active trace gases. The basic formula (IPCC, 2001; Lashof and Ahuja, 1990; Shine, et al., 1990) for calculating *direct* GWPs for radiatively active trace gases (CH_4, N_2O, O_3, and CFCs) is:

$$GWP_{i,T} = \frac{\int_0^{TH} a_i(t) \cdot X_i(t)/MW_i \, dt}{\int_0^{TH} X_{CO2}(t)/MW_{CO2} \, dt} \qquad \text{eq. D.1}$$

where:

$GWP_{i,T}$ = the global warming potential for gas i, relative to CO_2 (on a mass basis), over a period of T years[2]

$a_i(t)$ = the relative radiative forcing of gas i as a function of time, in units of: (W per m^2 per ppm-increase for gas i)/(W per m^2 per ppm-increase for CO_2) (discussed below)

$X_i(t)$ = a function expressing the amount of gas i that remains in the atmosphere over time (discussed below)

MW_i = molecular mass of gas i (g/mol)

MW_{CO2} = the molecular mass of CO_2 (44.01 g/mole)

$X_{CO2}(t)$ = a function expressing the amount of CO_2 that remains in the atmosphere over time over time

t = time (years)

TH = time horizon; the period of time for which the GWP is calculated (years)

It is typical to express X(t) as an exponential decay function:

$$X(t) = e^{\left(\frac{-t}{L_i}\right)} \qquad \text{eq. D.2}$$

where:

t = defined as above

L_i = the residence time or lifetime of gas I (years) (this also is called the "e-folding time" for the gas, because after t = L years the concentration of the gas will be reduced to 1/e, or 37%)

The relative radiative forcing term can be expressed as a constant, based on the areal radiative forcing and the concentration of each gas:

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[2] To convert the GWPs or EDIs from a mass basis to a mole basis, multiply by Mi/44, where Mi is the molecular mass of gas i (e.g., 16 for CH_4), and 44 is the molecular mass of CO_2.

$$a_i(t) = \frac{\frac{F^*_i}{C_i}}{\frac{F^*_{CO2}}{C_{CO2}}} \qquad \text{eq. D.3}$$

where:

F^*_i = the radiative forcing of gas i associated with some concentration increment N_i (watts / m^2)

C_i = the concentration increment of gas i resulting in the radiative forcing F_i (ppm)

Making the substitutions for X(t) (eq. D.2) and a(t) (eq. D.3) into eq. D.1 and taking and evaluating the integral (eq. D.1) from time t = 0 to t = TH results in the following expression for a GWP:

$$GWP_{i,T} = \frac{\frac{F^*_i}{C_i}}{\frac{F^*_{CO2}}{C_{CO2}}} \cdot \frac{MW_{CO2}}{MW_i} \cdot \frac{L_i}{L_{CO2}(t)} \cdot \frac{1-e^{\left(\frac{-T}{L_i}\right)}}{1-e^{\left(\frac{-T}{L_{CO2}}\right)}} \qquad \text{eq. D.4}$$

where:

the lifetime of CO_2 as a function of time $L_{CO2}(t)$ is discussed later.

With eq. D.4, and data on the lifetime and radiative forcing parameters, one can calculate the GWPs used by the IPCC in its third assessment report (IPCC, 2001) (see Table D-3).

To obtain a total GWP, which accounts for indirect warming effects as well as direct effects, one either can include in eq. D.4 the indirect warming effects (e.g., one can include in the radiative forcing term, a_i, the indirect as well as the direct forcing, as in Lashof and Ahuja [1990] , or else one calculate indirect effects separately and add them to the direct effects. In the case of CH_4, the IPCC (2001) simply multiplies the calculated direct-effect GWP by a factor that accounts for the indirect effects.

The GWPs for N_2O and HFC-134a are relatively simple to estimate, because they do not appear to have significant indirect effects on climate. The recent IPCC estimates are shown in Table D-3. The GWPs for CH_4 and CFCs are more complicated because of their indirect effects on ozone and water vapor.

Issues with GWPs. Not surprisingly, there is disagreement about every key parameter in the calculation of total GWPs, including: the effective lifetime, the relative radiative forcing, the effect of changing concentrations of all gases on their residence times and radiative forcing, the indirect global warming effect of chemical reactions among gases, and the selection of the appropriate time period for the analysis.

But there also are more fundamental disagreements about whether GWPs as presently constructed are suitable for policy analysis (Reilly et al., 2003; Smith and Wigley, 2000a, 2000b; Lashof, 2000; O'Neill, 2000). For example, Smith and Wigley (2000a, 2000b) argue that GWPs have three structural flaws that render them unsuitable for comparing different GHG emissions-reductions policies: i) GWPs compare radiative forcing rather than the impacts of climate change, and it is the latter that we care about; ii) GWPs arbitrarily ignore radiative forcing and hence climate-change impacts beyond the chosen time horizon; and iii) GWPs are based on a one-time emissions of one unit of a gas given a constant concentration of all gases, whereas any real policy will result in different emissions trajectories over time. Smith and Wigley (2000a, 2000b) develop a measure of CO_2 equivalence based on temperature change or sea-level rise (rather than radiative forcing) and on a more realistic emissions and concentration scenario, and find that these more realistic equivalency factors can differ substantially from GWPs.

Manne and Richels (2001) add economic considerations to the debate, and argue that an ideal index "would be the outcome of an analysis that minimizes the discounted present value of damages and mitigation costs" (p. 675). Similarly, Reilly et al. (2003) argue that CEFs should be based on the *dollar costs* of the impacts of climate change:

> Still another area of criticism of the current GWP formulation is its lack of an economic basis...In proposed economic-based indices [EDIs – my note], the comparisons go beyond just the effect on radiative forcing to consider the actual implications of these gases for climate change and, in most cases, the economic damages estimated to result from it. An economic discount rate is used to evaluate the fact that damages from different substances are spread over different time periods because of varying lifetimes of the gases. Discounting and tracking damages as they vary over time with rising atmospheric concentrations of gases allows these approaches to take into account the timing of climate benefits resulting from control strategies, something current GWPs cannot do. The economic formulation provides, therefore, a better rationale for comparing short-lived gases...with longer lived gases. (p. 38).

However, Reilly et al. (2003) go on to note that there is a great deal of uncertainty in estimating EDIs, and that as a result one cannot unambiguously recommend present EDIs over present GWPs.

Note on the time period assumed for the GWP. GWPs depend greatly on the length of time considered. For most gases, the GWP is much smaller if one considers a 500-year period rather than a 10-year period, because most greenhouse gases have a greater radiative forcing per mole but a shorter residence time than has CO_2. If, in the calculation of the GWP for a particular greenhouse gas, one considers a period of time less than or equal to the life of the greenhouse gas, the GWP will be relatively high, because over the whole period of analysis the greenhouse gas will be in the atmosphere and causing a greater forcing than will CO_2. However, over a much longer period, the greenhouse gas will have been gone from the atmosphere for most of the time (or may have oxidized to CO_2 and water), and so its average warming effect over the period will be less than over the shorter period. The importance of the time period can be seen in Table D-3, which shows the IPCC's GWPs for 20, 100, and 500 year periods.

There are two ways to approach the question of the right time period for an analysis using GWPs: 1) use a time cutoff, with no “discounting” of warming in future years (i.e., simply integrate the GWP formula over a specified number of years), or 2) use a non-zero discount rate, with effectively no time-cutoff (integrate to several thousand years). DeLuchi (1991) argues that it might be inappropriate to use a discount rate for this problem, because very small and thus relatively meaningless changes in the discount rate have significant effects on GWP values. He suspects that most policy makers can grasp more readily the concept of a time cutoff, and its effect on the GWP, than the significance of small changes in the discount rate. However, as discussed below, there is evidence that people discount the long-term future with a rate that declines over time. Furthermore, the use of an EDI instead of a GWP requires the use of a discount rate rather than a time cutoff. The CEFs developed in this report use both a non-zero discount rate and a time-horizon cutoff.

In any case, most analysts who use GWPs use the values for a 100-year time horizon, with no discount rate. The case for a relatively long time horizon is obvious: climate change is a very long-term phenomenon[3] which will affect many generations to come, and it seems unreasonable to use a time horizon that excludes significant long-term effects.[4]

An economic damage index as an alternative to the GWP index

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[3] For example, the deep ocean, which stores CO_2, turns over on a time scale of centuries.

[4] An argument that has been advanced in support of using a very-short-term horizon is that we should be concerned with the *rate* of warming as well as with the ultimate increase in equilibrium surface temperature, and that the short-term horizon represents the effect of different gases on the rate of global warming (Mitchell, et al., 1990; Shine, et al., 1990; Wilson, 1990). The implicit claim is that the greater the 20-year warming factor for a particular greenhouse gas, the greater the rate of warming due to that gas. Now, it certainly is true that the higher the GWP the higher the implied *average* rate of warming. But this is true for any time horizon, not just short ones. Moreover, the actual warming trajectory over time due to emissions of a greenhouse gas may or may not follow the straight-line average. We ought to be interested in the actual warming trajectory, which cannot be deduced or inferred from a calculated GWP. Moreover, the overall global rate of warming depends on the rate of emission of all gases, and the GWPs consider neither rate nor all gases simultaneously -- they tell us the relative radiative forcing of a one-time emission of a unit of one gas. To construct a measure that captures the bad effects of a “fast” warming, one must actually estimate the warming trajectories due to different emission scenarios, and formally relate some measure of the undesirable effects of global warming to the functional form of a warming trajectory. See Lashof (2000), O’Neill (2000), and Smith and Wigley (2000a, 2000b) for discussion of these and related issues.

Shortly after the original GWPs were developed, Reilly (1992) and Schmalensee (1993) derived equivalency factors based on the marginal damage of greenhouse-gas emissions. These “economic damage indices,” or EDIs, recently have been developed further by Hammitt et al. (1996), Kandlikar (1996), Tol (1999), Gottinger (1999), and Reilly et al. (2001). (Manne and Richels [2001] and Gottinger [1999] discuss a related economic index, based on cost-effective control costs rather than damage costs; see the discussion below.)

In addition to accounting for the relative lifetimes and radiative forcing potencies of different gases, the EDI goes a step further and compares greenhouse gases with respect to their potential economic welfare damages. The EDI's focus is thus on the *effects* rather than the magnitude of climate change, and as a result it is able to account impacts such as stratospheric ozone depletion from CFCs or CO_2 fertilization of crops. Gottinger (1999) shows that GWPs can be derived from EDIs by assuming that: i) damages are a linear function of temperature, ii) the climate lag time due to the response of the ocean is zero, and iii) the discount rate is zero.

The EDI, as defined by Hammitt et al. (1996), is the reduction in emissions of a reference gas (CO_2) that would be required to offset the incremental damage that would otherwise result from increased emissions of a particular greenhouse gas. It can also be defined as the partial derivative of the present value of economic welfare loss with respect to the emissions of a particular greenhouse gas, relative to the partial derivative of welfare loss with respect to CO_2 emissions. Hammitt et al. (1996) use the following equation for an EDI:

$$EDI_i = \frac{\frac{\partial}{\partial e_i} W\left[C(t)\right]}{\frac{\partial}{\partial e_o} W\left[C(t)\right]} = \frac{\int_0^\infty \Delta C_i(t) \cdot \lambda_i(t) dt}{\int_0^\infty \Delta C_o(t) \cdot \lambda_o(t) dt} \qquad \text{eq. D.5}$$

where:

EDI_i = the economic damage index for gas i
e_i = emissions of gas i
e_o = emissions of reference gas, CO_2
W[C(t)] = the economic welfare loss due to the time path of GHG concentrations, C(t)
$\Delta C_i(t)$ = the change in the concentration of gas i
$\Delta C_o(t)$ = the change in the concentration of the reference gas, CO_2
$\lambda_i(t)$ = the marginal social cost of an additional unit concentration of gas i
$\lambda_o(t)$ = the marginal social cost of an additional unit concentration of the reference gas, CO_2

Hammitt et al. (1996) base the calculation of EDI values on a simple damage function that relates economic damages to the magnitude of temperature change over time:

$$W[\Delta T(t)] = \int_0^\infty \left(\frac{1}{1+r}\right)^t \cdot a \cdot GDP(t) \cdot D[\Delta T(t)]\, dt \qquad \text{eq. D.6}$$

where:

W[ΔT(t)] = economic damages from a change in average global temperature
r = the discount rate
α = a scaling constant
GDP(t) = gross world product
D[ΔT(t)] = economic damage function, related to magnitude of temperature change
ΔT(t) = the increase in global annual-mean surface temperature from its 1990 value

In addition to simple damage functions, where damages are a linear, quadratic, or cubic function of ?T, Hammitt et al. also investigate a more complex, "hockey-stick" damage function that can be varied from a quadratic function to a highly convex function by varying a parameter χ from 0 to 1. The authors choose a χ value of 0.1, resulting in a highly convex function that might represent the possibility of catastrophic damages with high levels of ?T. The authors note that more complicated and disaggregated damage functions, perhaps related to the rate of climate change as well as the ultimate magnitude, could readily be substituted for the simple forms that they used. They also note that additional damages (or benefits), including damages unrelated to climate change, could be included in the EDI, but they stop short of including non-climatic effects.

Hammitt et al. (1996) assess various scenarios with and without incremental greenhouse gas emissions, and use the Integrated Science Assessment Model to determine resulting global average temperature changes. The use of a climatic model to determine the temperature changes resulting from different scenarios of greenhouse gas emissions represents an advancement over previous attempts to develop economic indices for greenhouse gases. For example, Reilly (Reilly, 1992) has developed an economic damage index (which he calls a Trace-Gas-Index) that relies on instantaneous radiative forcing estimates. These estimates are in turn based on the assumption of a constant-composition atmosphere. The use of a climate model allows Hammitt et al. to compute EDI values without accepting this potentially problematic assumption.

Both Reilly's (1992) and Hammitt et al.'s (1996) economic damage indices are reported in Table D-3. Reilly's three estimates are shown, based on different assumptions of the form of the damage function. Several EDI estimates are reported from Hammitt et al. (1996), reflecting different assumptions of climate sensitivity, discount rate, emission timing and magnitude, and damage function exponent. It is worth noting that the highest and lowest EDI values for each gas that are obtained in 81 combinations of the above parameters are increasingly disparate in the case of relatively short lived CH_4, producing a range of values from about 3 to 50 (Hammitt, et al., 1996) , without accounting for the indirect effects of CH_4 on climate. For longer lived N_2O and CFC-12, the variation is more modest, with the low value being on the order of 70% to 75% of the high value.

Recently, Tol (1999) has estimated the marginal damage cost of CO_2, CH_4, and N_2O emissions. The ratio of the marginal damage cost of CH_4 or N_2O to the marginal damage cost of CO_2 is in essence the EDI of Hammit et al. (1996). The damage ratios from Tol's (1999) model are 348 for N_2O and 14 for CH_4, very close to the "middle case" EDI's of Hammit et al (1996).

Finally, Reilly et al. (2001) derive a trace gas index from the "optimal dynamic control problem that balances the cost of controlling each of the greenhouse gases with the damages associated with climate change" (p. 3). Their work advances previous similar work in several ways: they calculate the trace gas index for the economically optimal emissions path, rather than for a reference emissions scenario; they consider additional gases (PFCs and SF_6); they consider a scenario in which the discount rate declines over time to zero (see the discussion of the discount rate, below); and they formulate the optimal control problem within a general equilibrium rather than partial equilibrium cost-benefit framework. However, it appears that there characterization of damages is simpler than that in, say, Hammit et al. (1996): Reilly et al. (2001) assume that damages are linear with radiative forcing, such that damages from a doubling or radiative forcing are 1% of world GDP.

Table D-3 shows two sets of estimates from Reilly et al. (2001). Their EDI for N_2O is somewhat lower than the IPCC GWP, but their EDI for CH_4 ranges from much lower to much higher than the IPCC GWP for CH_4.

Cost effectiveness economic CO_2-equivalency indices

A variant on the damage-cost EDI presented above is a "cost-effectiveness" index (CEI) (Gottinger, 1999; Manne and Richels, 2001). In this formulation, CEI_i is the ratio of the marginal costs of abatement for gas *i* relative to the marginal cost for CO_2 when one minimizes the *total* abatement cost (over all gases) given an exogenously determined temperature or temperature-trajectory constraint. A CEI, then, requires estimates of abatement-cost functions (the cost of reducing emissions of each gas as a function of the amount reduced) rather than damage-cost functions. Because emissions abatement costs are easier to estimate than are climate-change damage costs, CEIs are less uncertain than are damage-cost EDIs. (See also Reilly et al. [2002] for a discussion of GHG emission abatement costs.) Moreover, if we agree on a limiting temperature (warming) trajectory or ceiling, and are certain we are going to meet it, then society does maximize its welfare by minimizing the total abatement costs. In this case the CEI tells us the true social cost of one gas relative to another. However, if emissions are not constrained by an agreed-upon temperature cap, then the true social cost of each gas (and hence the appropriate CEF) is based on the damages from the emitted gases, not on the cost of abating emissions.

Manne and Richels (2001) use computable general equilibrium model called MERGE to determine CEIs for CH_4 and N_2O. The model has reduced-form descriptions of the energy sector, the economy, emissions, concentrations, and temperature change. Manne and Richels (2001) assume a temperature constraint, or ceiling, and then use MERGE to identify an economically efficient strategy for staying with the prescribed ceiling. They consider temperature-increase ceilings of 2^o and 3^o over 200 years, and, in a separate scenario, an additional constraint that limits the decadal temperature change to 10% of the total allowable increase of 2^o and 3^o. Given these assumed ceilings, MERGE is used to calculate the price / ton of CH_4, N_2O, and CO_2, where the price expresses willingness to pay to emit an additional ton of the gas. The CEI for gas *i* is based on the price of gas *i* relative to the price of CO_2 as determined by their contribution to the ceiling.

Manne and Richels (2001) find that in the case of a temperature-increase ceiling of 3^o, the CEI for N_2O is about 650 (more than twice the official IPCC GWP), and the CEI for CH_4 ranges from near zero today to about 15 100 years from now (less than the official IPCC GWP). The CEI for CH_4 is near zero today because present-day emissions of CH_4 contribute much less to reaching the temperature ceiling than do present-day emissions of CO_2: present-day emissions of CH_4 will last for only the next decade or so, whereas present-day emissions of CO_2 will last for the next century or so, and the temperature ceiling does not begin to become binding (compared with a no-controls reference case) until several decades from now. However, as the temperature ceiling is approached, it becomes more valuable to reduce emissions of a short-lived gas compared with a long-lived gas.

The Manne and Richels (2001) CEIs are quite a bit different from the EDIs estimated thus far (compare the CEI values discussed above with EDI values in Table D-3). Because CEIs are based on abatement costs, whereas EDIs are based on damage costs, the implication is that the Manne and Richel (2001) estimates of relative marginal abatement costs are quite a bit different from estimates of relative marginal damage costs. This in itself is an interesting finding, one which highlights the considerable uncertainty in estimating any kind of economic CEF.

The discount rate

Background and theory. EDIs, CEIs, and our own CEFs (discussed below) all require the specification of the discount rate. The discount rate is the rate at which some future event (such as a monetary payment) is valued compared with the same event happening today. An annual discount rate of *r*% tells us that an event happening *t* years from now is worth $1/(1+r)^t$ as much as the same event happening today.[5] (Note that if the discount rate is negative ($r < 0$), then the discount factor above is greater than 1.0, and the future event is worth more than the present event.)

There are three basic reasons why the present value of an event may depend on when the event occurs:

i) Individuals' tastes (valuation functions) or circumstances (e.g., wealth) may depend on time (e.g., if one expects to like something more in the future than today, then $1/(1+r)^t > 1.0$ and $r < 0$).

ii) The future is uncertain: there always is some risk that one will not be able to enjoy future consumption (for example, one might die first). All else equal, we prefer certain consumption today to uncertain consumption tomorrow (in which case $1/(1+r)^t < 1.0$ and $r > 0$).

iii) Present investments produce future income, which means that future events are worth less than present events by the amount of income that would have been produced by the present event. This rate of return is the same as or a component of the discount rate.

In the case of estimating CEFs, the discount rate reflects how we value the impacts of climate change as a function of the time when they occur. A positive rate discounts climate-change impacts in the distant future compared with climate change impacts in the near future. The higher the rate, the more the distant future is discounted. On the time scale of climate change – decades or even centuries – the effect of discounting can be dramatic. The following table illustrates this by showing the present value of an impact occurring Y years in the future given a discount rate of *r%*:

r Y -->	*1*	*10*	*20*	*50*	*100*
1.0	0.99	0.91	0.82	0.61	0.37
2.0	0.98	0.82	0.67	0.37	0.14
5.0	0.95	0.61	0.38	0.09	0.01
10.0	0.91	0.39	0.15	0.01	0.00

In most analyses of public investments, discounts rates are 5% or higher. The table above shows that with rates of 5% or higher, events that occur more than 50 years from now are discounted to close to zero present value. Now, many of the actions that we take today will affect climate for more than a century. Thus, if we apply standard discount rates to these actions, we will be quite literally completely discounting the

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[5] The discount factor $1/(1+r)^T$ also may be estimated as e^{-rt}. The exponential formulation is the continuous compounding case of the other formulation -- that is, as the time step and hence the discount rate go to zero, e^{-rt} gives the same discount factor as $1/(1+r)^t$. For relatively low annual discount rates (less than 5%), the two formulations give similar results out to a couple hundred years.

long-term effects of our actions. Many analysts and lay persons feel that this is wrong. (For example, the IPCC [2001a, p. 467] cites a survey of 1700 economists that found that they believe that lower discount rates should be applied to problems with long time horizons.) As a consequence, analysts have put forward arguments for using very low discount rates in the evaluation of the impacts of climate change.

More than a decade ago, Cline (1992) provided a comprehensive analysis of the appropriate discount rate to use in the context of global warming, and settled on a rate of a bit under 2%. Shortly thereafter, Cropper et al. (1994) surveyed adults in Washington, D. C. about their preferences regarding saving lives now versus in the future, and found that the implicit discount rate declined over time: 16.8% for a time horizon of five years, 7.4% for a time horizon of 25 years, and 4.8% for a time horizon of 50 years. This notion of a declining discount rate over time has been extended to the case of global warming, and even developed into a formal proposition. For example, in the survey of 1700 economists, cited above (IPCC, 2001a), economists also distinguished between the immediate future and the far distant future, step by step. The resultant discount rate implied by the analysis dropped progressively from 4% for the near term (5 years) to 0% for the far distant future (beyond 300 years). The IPCC (2001a) offers further discussion of this phenomenon. In recognition of this phenomenon, Reilly et al. (2001) use a declining discount rate in some runs of the model they use to calculate EDIs.

In 1998, Weitzman (1998) formalized the proposition that the far-distant future should be discounted at its lowest possible rate. The essence of his argument (a proof, actually) is presented in the following eight steps:

o) Define the following:

-- the *instantaneous discount rate* r at any time *t*.

-- the *instantaneous discount factor* (DF) corresponding to time *t*, which tells us the value today of a dollar in year t: $DF = e^{-rt}$

-- a *discount rate trajectory* (DRT), in which the instantaneous discount rate *r* changes over time, following some trajectory: DRT = r(t).

-- the *integrated trajectory discount factor* (ITDF), which is the discount factor corresponding to the area under DRT up to time T:

$$\mathrm{ITDF} = 1 / T \int^{T} e^{-r(t)t}\, dt$$

i) We acknowledge that we don't know what the discount rate *r* will be in the future: it may be higher than the rate today, it may be the same, or it may be a lot lower. It will be lower if the rate of return to capital declines, because of stagnating productivity, or if the social rate of time preference, for whatever reason, is very low.

ii) This uncertainty about future discount rates can be represented by positing various discount rate trajectories over time. Let trajectory *i* be represented as DRTi.

Each discount rate trajectory DRTi implies an integrated trajectory discount factor (ITDFi) up to some far-distant time T.

iii) Among the various trajectories posited there is one in which the discount rate R approaches some lower limit L as time approaches infinity. Define the quantities related to this trajectory to be: the lower-limit discount rate trajectory DRT_L, the lower-limit instantaneous discount rate r_L, the instantaneous discount factor at time *t*, DF_L, and the integrated trajectory discount factor for the lower-limit trajectory up to time T, ITDF_L.

iv) We can assign a probability Pi to each trajectory DRTi and corresponding integrated trajectory discount factor ITDFi. We can then estimate an overall probability-weighted discount factor (ITDFpw), over all the trajectories:

$$\text{ITDFpw} = ?\ _{\text{i}}\text{Pi} \cdot \text{ITDFi}$$

Note that the probability weights are applied to the integrated trajectory discount factors up to time T, not to the instantaneous discount factor at time T, because the instantaneous discount factor at time T does not account for the time-path of the discount rate from now to time T (whereas ITDF reflects the cumulative impacts of the discount rate trajectory over time).

v) Because $\text{DF} = e^{-rt}$, the lower the value of the discount rate *r*, the higher the value of the discount factor DF. As *r* approaches zero, DF approaches 1.0, regardless of *t*. Conversely, as *t* gets large, DF approaches 0.0 for all but small values of *r*. From this, we can infer that the integrated trajectory discount factor for trajectories in which *r* approaches some lower limit -- ITDF_L -- will be larger (and probably *much* larger) than all other ITDFi.

vi) If ITDF_L >> all other ITDFi, and if the probability of ITDF_L is not particularly small, then $?\ _{\text{i}}\text{Pi} \cdot \text{ITDFi}$ will be close to ITDF_L. Hence:

$$\text{ITDFpw} \sim \text{ITDF}_\text{L}$$

In words, the discount factor for the trajectory in which the discount rate approaches the lower limit will be so much greater than the discount factor for the other trajectories that the probability-weighted result will be close to the result for the single trajectory in which the discount rate approaches the lower limit.

To this point I have shown that the probability-weighted overall discount-rate trajectory will be similar to the one in which a lower-limit discount rate is approached. This probably is enough to make the point about discounting the far-distant future at the lowest possible rate, but we may go a few steps further.

vii) For each trajectory i there is an instantaneous discount rate r_I at time *t* which produces a discount factor DFi equal to ITDFi up to time T. If from above:

$DF = e^{-rt}$

then $r_I = -\ln(DFi)/t = -\ln(ITDFi)/T$

Now, we have defined r_L to be the instantaneous discount rate corresponding to the integrated-trajectory discount factor for the case in which the discount rate approaches a lower limit L (DF_L). Next, let r_{PW} be the instantaneous far-future discount rate corresponding to the probability-weighted integrated trajectory discount factor (ITDFpw). Thus

$r_L = -\ln(ITDF_L)/T$
$r_{PW} = -\ln(ITDFpw)/T$

viii). If from vi) ITDFpw ~ $ITDF_L$, then:

$r_{PW} \sim r_L$

That is, the instantaneous far-future discount rate (Rpw) corresponding to the probability-weighted integrated trajectory discount factor (ITDFpw) will be relatively low -- indeed, close to R_L -- because ITDFpw will be close to the discount factor corresponding to the trajectory in which the discount rate approaches a lower limit.

To put it more intuitively, it is possible that in the future people will use a zero or near-zero discount rate, and this possibility carries great weight today (relative to any other possibility) because if the discount rate is zero the future carries great weight (because the discount factor is close to 1). In other scenarios the future carries very little weight (the discount factor is small) and hence do not matter much today. Thus, we ought to “bias” our discount rate to account for the possibility that the future might carry great weight. We can do this by positing a discount rate trajectory in which the discount rate declines over time, towards some relatively low limit. I do this in a later section of this report.

CO_2 EQUIVALENCY FACTORS (CEFS): METHODS AND PARAMETER VALUES FOR GASES WITH DIRECT RADIATIVE FORCING EFFECTS

The general method

In this section, I develop a hybrid EDI/GWP for gases with a significant direct radiative-forcing: CO_2 CH_4, N_2O, O_3, CFC-12, HFC-134a, SF_6, CF_4, C_2F_6, and six different kinds or components of aerosols: black carbon (BC), organic matter (OM), sulfate, nitrate, secondary organic aerosols (SOA; formed from emissions of NMOCs), and dust[6]. (I also estimate the minor direct radiative forcing effect of CO.) In the subsequent major section I use different methods to estimate CEFs for the "indirect" gases CO, H_2, NMOCs, NO_X, and SO_X.

The hybrid method is meant to address some of the shortcomings in GWPs. As discussed above, the IPCC-estimated GWPs incorporate simplifications that can lead to differences between the relative integrated radiative forcing (meant to be a proxy for temperature change) estimated by a GWP and the same quantity estimated by a sophisticated model:

i) GWPs assume a constant e-folding lifetime for all gases, whereas in reality the e-folding time of some gases is a function of the concentration of the gas, which changes over time.

ii) GWPs assume a constant radiative forcing per unit (e.g., Wm^{-2} $ppbv^{-1}$) for all gases, whereas in reality the radiative forcing per unit of some gases is a function of the concentration, which changes over time.

iiii) Assuming that the relative integrated radiative forcing estimated by a GWP is meant to be a proxy for relative integrated temperature change, then another simplification is that GWPs ignore the time lag between an initial change in radiative forcing and the reaching of the final equilibrium atmospheric temperature due to the thermal inertia of the oceans.

This method avoids the simplifications of i) and ii). I do not address simplification iii) (i.e., I *do* assume that the equilibrium temperature change is simultaneous with a change in radiative forcing).

Apart from these technical simplifications, GWPs also are based on certain conventions that one might not wish to accept:

iv) GWPs are based on relative integrated radiative forcing (a proxy for temperature change), whereas one might be interested instead in the impacts (such as economic damages) resulting from temperature change (and impacts might not be linearly related to temperature change)

v) GWPs integrate radiative forcing from the present day to some fixed future time T (e.g., 20, 100, or 500 years from now), in effect giving a weight of 1.0 to every year between now and T and a weight of 0.0 to every year beyond T, whereas one might prefer to apply weights that decline continuously (and nearly indefinitely) starting from 1.0 today.

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[6] I chose these six because they are all of the types of aerosols for which estimates of radiative forcing are available today. (Ideally, I would further disaggregate dust, OM, and SOA to more specific compounds, but at present there are no estimates of radiative forcing for these more specific compounds.) Most of the mass of total global aerosol emission is one of these six kinds.

Our CEFs are based, crudely, on the impacts of radiative forcing rather than on radiative forcing itself, and apply a non-zero discount rate over a long period of time.

For CO_2 CH_4, N_2O, O_3, aerosols, CFC-12, HFC-134a, SF_6, CF_4, and C_2F_6, I estimate CEFs as a function of the rate of decay of a unit emission of a gas (which in turn is a function of the concentration), the unit radiative forcing of the gas remaining in the atmosphere (also a function of the concentration), the relationships between temperature and radiative forcing and between damages and temperature, and a present-value or discounting factor (which varies over time). Our CEF formula, unlike the GWP formula, is too complex to be integrated easily, and hence is evaluated year-by-year over 1000 years, with the discounted yearly impacts summed. (Presumably, integratability is one reason the IPCC chose a simple GWP formula.) Formally:

$$CEF_{i,T} = \frac{MW_{CO2} \cdot \sum_0^{TH} X_i(C,t) \cdot D_i\left\{T_i\left[\left(F_i(C), I_i(C)\right)\right]\right\} \cdot DF\left[r(t)\right]}{MW_i \cdot \sum_0^{TH} X_{CO2}(t) \cdot D_{CO2}\left\{T_{CO2}\left[\left(F_{CO2}(C), I_{CO2}(C)\right)\right]\right\} DF\left[r(t)\right])} \qquad \text{eq. D.7}$$

where:

$CEF_{i,T}$ = the CO_2-equivalency factor for gas i over a period of T years (expressing the total discounted impact of a one-gram year-zero emission of gas i relative to the total discounted impact of a one-gram year-zero emission of CO_2)

$X_i(C,t)$ = the time- and concentration-dependent decay an emission of a unit of gas i, represented as the fraction of a unit emission of gas i that remains in the atmosphere over time

$D_i\{T_i[F_i(C), I_i(C)]\}$ = damages associated with gas i due to changes in temperature

$T_i[F_i(C), I_i(C)]$ = changes in temperature associated with gas i due to changes in direct and indirect radiative forcing from gas i

$F_i(C)$ = the direct radiative forcing of gas i in the atmosphere, as a function of concentration, in units of: W per m^2 per ppmv-increase for gas i (except in the case of aerosols, where the units for aerosols and CO_2 are W per gram increase of gas i)

$I_i(C)$ = the indirect forcing of gas i in the atmosphere (due to the effects of gas i on the concentration of other directly radiatively active gases) as a function of concentration, in units of: W per m^2 per ppmv-increase for gas i

$DF[r(t)]$ = the discount factor as a function of the discount rate

$r(t)$ = the discount rate as a function of time

MW_i = molecular mass of gas i (g / mol) (not used in the case of aerosols, because the forcing term is calculated directly in mass terms)

MW_{CO2} = the molecular mass of CO_2 (44.01 g / mole) (not used in calculating CEFs for aerosols, because in the case of aerosols the forcing term is calculated directly in mass units)
t = time (years)
TH = time horizon; the period of time over which the CEF is calculated (years)

In the following sections I discuss each of the terms in the general CEF formulation.

Note that I estimate CEF parameters for an incremental change in emissions or concentration, rather than for "average" conditions. Consider for example the parameter radiative forcing. The radiative forcing of some gases is a nonlinear function of concentration. This means that for any given concentration, the radiative forcing of an incremental change in concentration, in $Wm^{-2}ppmv^{-1}$, is different from the average forcing at that concentration (where the average is equal to the total Wm^{-2} divided by the total concentration). Similarly, the lifetime of some gases is a nonlinear function of concentration, and as a result, the effective lifetime of an incremental change in concentration will be different from the average lifetime of the entire atmospheric burden.

Underlying emissions and concentrations scenarios

For some gases, some of the parameters in our CEF formula, such as the exponential decay of a unit emission of a gas and the unit radiative forcing of a gas, depend on the concentration of the gas. (Generally, as the concentration of a gas increases, it's decay is slowed and its radiative forcing is reduced). Since our CEF expression sums the year-by-year impacts of a gas over a long period of time, and the concentration of the gas and hence the parameters related to concentration will change over time, it is appropriate to first specify emissions and concentration scenarios, and then estimate concentration-dependent parameters in accordance with the projected concentrations.

Because the LEM can perform analyses for any target year between 1970 and 2050, we also must include historical data on actual concentrations back to 1970, so that the model will appropriately back-cast for scenarios that start as early as 1970. And because we wish to be able to calculate CEFs on the basis of time horizons of a 1000 years or more, we must project concentrations for at least 1000 years into the future. In the following I discuss my assumptions for three time periods: 1970 to 2000, 2000 to 2100, and beyond 2100.

1970 to 2000. Appendix II of the IPCC (2001) reports the concentration of CO_2, CH_4, and N_2O in 1970, 1980, and 1990. Table 1 of the IPCC (2001) reports the concentration in 1998. I use these values here.

2000 to 2100. The IPCC (2001, Appendix II) provides several scenarios of emissions and related concentrations for each major GHG, from the year 2000 to the year 2010. For the purpose here of estimating parameters dependent on the concentration, I have chosen as my starting point the "A1B" scenario, in which emissions and concentrations are roughly in the middle of the range of all the IPCC scenarios[7]. This scenario estimates the following changes in concentrations of GHGs over time:

Year	*CO_2 (ppmv)*	*CH_4 (ppbv)*	*N_2O (ppbv)*	*O_3 (DU)*	*CFC-12 (pptv)*	*SO_4 (TgS)*	*OC (Tg)*	*BC (Tg)*
2000	367	1760	316	34.0	535	0.52	1.52	0.26
2010	388	1871	324	35.8	527	0.66	1.70	0.29
2020	418	2026	331	37.8	486	0.76	1.75	0.30
2030	447	2202	338	39.3	441	0.69	1.86	0.32
2040	483	2337	344	39.7	400	0.52	1.94	0.33
2050	522	2400	350	39.8	362	0.48	2.01	0.34
2060	563	2386	356	39.6	328	0.35	2.06	0.35
2070	601	2301	360	39.1	298	0.27	2.11	0.36
2080	639	2191	365	38.5	270	0.23	2.22	0.38
2090	674	2078	368	38.0	245	0.22	2.43	0.42
2100	703	1974	372	37.5	222	0.21	2.67	0.46

(OC = organic carbon, which is not necessarily the same as OM [organic matter aerosol], the quantity considered here; DU = Dobson Units. The CO_2 concentrations are the reference case from the Bern-CC model.) I adopt these projections for the years 2000 to 2100.

Note that the IPCC A1B scenario shows CH_4 emissions and concentration rising through 20250, then declining. By contrast, recent detailed model projections of emissions by Webster et al. (2002) show CH_4 emissions rising continuously through the year 2100, although at a declining rate after 2050. (Webster et al.'s [2002] projections of N_2O and CO_2 are broadly consistent with the IPCC A1B scenario.)

Beyond 2100. For projections beyond the year 2100 I use my judgment. I assume that CH_4 emissions continue to decline for a while but then rise once more, before beginning a final decline.

My assumptions are as follows:

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[7] The "A1" family of scenarios describes a future world of rapid economic growth, population declining after mid-21st century, and rapid develop of new and efficient technologies (IPCC, 2001, p. 63). In the "A2" scenarios the world is more heterogeneous; in the "B1" scenarios the economy tends towards information and service technologies, and in the "B2" scenarios sustainability is emphasized. Within the A1 family, the "A1B" scenario assumes a "balance" of fossil and non-fossil energy sources.

Year	*CO2 (ppmv)*	*CH4 (ppbv)*	*N2O (ppbv)*
1970	325	1420	295
1980	337	1570	301
1990	352	1700	308
1998	365	1745	314
2000	367	1760	316
2010	388	1871	324
2020	418	2026	331
2030	447	2202	338
2040	483	2337	344
2050	522	2400	350
2060	563	2386	356
2070	601	2301	360
2080	639	2191	365
2090	674	2078	368
2100	703	1974	372
2110	731	1,895	376
2120	757	1,857	379
2130	778	1,857	383
2140	793	1,876	387
2150	805	1,894	391
2160	813	1,913	395
2170	820	1,933	399
2180	824	1,952	403
2190	826	1,971	407
2200	826	1,991	411
2300	776	1,971	403
2400	722	1,951	395
2500	664	1,932	387
2600	611	1,913	379
2700	562	1,894	371
2800	517	1,856	364
2900	476	1,819	357

3000	438	1,782	350
3100	403	1,747	343
3200	370	1,712	336
100000	320	1,575	330

Because the period-to-period differences are relatively minor (e.g., the difference between the CH_4 concentration in 2170 and 2180 is quite small), I do not bother to interpolate values for years that fall between periods (e.g., for 2173, which falls between 2170 and 2180), but rather use the value corresponding to the lower of the two bounding periods (e.g., 1,933 for CH_4 in 2170 in this case).

Decay of emissions in the atmosphere (function X(t))

I follow the IPCC, and assume that the decay of a gas is represented by a simple exponential function (basically eq. D.2 above):

$$X_i(C,t) = e^{\left(\frac{-t}{L_i(C)}\right)} \qquad \text{eq. D.8}$$

where:

$X_i(C,t)$ = the fraction of a unit emission of gas i that remains in the atmosphere over time

t = time

$L_i(C)$ = the e-folding time of the gas (the number of years after which the concentration of the gas has decayed exponentially to 1/e, or 37%, of its initial value; in some cases, this is a function of the concentration C)

The task here is to estimate L, the e-folding time of each gas. For some gases, the parameter L technically is a function of the concentration of the gas *C*. And in the case of CO_2, the e-folding time also is a function of the time-horizon considered.

In the following, I first review data and estimates pertinent to L(C), then summarize my assumptions. There are extended discussions for particulates and CO_2.

Gases except aerosols and CO_2. IPCC (2001, Table 4.1a) shows the following estimated atmospheric lifetimes (see also Jain et al., 2000, for similar values from a World Meteorological Organization study):

CH_4 (years)	*N_2O (years)*	*O_3 (days)*	*CFC-12 (years)*	*HFC-134a (years)*	*CO (days)*
8.4 / 12	120 / 114	3-20	100	13.8	30-90

The IPCC (2001) notes that these lifetimes are also the e-folding times. In the case of CH_4 and N_2O, the number before the slash is the average lifetime of the entire atmospheric burden given approximately year-2000 concentrations, and the number after the slash (which the IPCC calls the "perturbation" lifetime) is the effective lifetime of an infinitesimal increment to the current concentration, accounting for the affect of the increment on the lifetime of the original atmospheric burden. In the case of CH_4, an incremental increase competes for the OH radical (which is the main sink for CH_4) and hence reduces the oxidation rate and increases the lifetime of all CH_4. In the case of N_2O, an incremental increase eventually results in an increase UV radiation, which increases photolytic destruction and hence reduces the lifetime of all N_2O.

The perturbation lifetime (which we will designate PT, following the IPCC) is appropriate for our analysis because as mentioned above we wish to estimate CEFs for incremental changes in emissions and concentration, and the perturbation lifetime is the effective lifetime of an incremental change in concentration, as opposed to the average lifetime of the entire burden. However, in the case of CH_4 and N_2O, the perturbation or incremental lifetime itself is a function of the concentration of the gas. That is, for these gases, both the average and the incremental lifetimes change as the concentration changes (although the ratio between the average and the incremental lifetime is a constant in the IPCC formulation).

The IPCC (2001) provides data and formulae that allow us to estimate the incremental or perturbation lifetime as a function of concentration (or burden). The IPCC (2001, Table 4.5 p. 254) represents the perturbation lifetime as a function of the atmospheric burden lifetime and a sensitivity parameter that describes the sensitivity of the atmospheric burden lifetime to changes in concentration (or burden) (see also Joos et al. 2001). Thus:

$$L_i(C) \equiv PT_B = \frac{LT_B}{1-S} \qquad \text{eq. D.9}$$

$$S = \frac{\ln\left(\frac{LT_B}{LT_{B*}}\right)}{\ln\left(\frac{B}{B*}\right)} \qquad \text{eq. D.10}$$

where:

$L_i(C)$ = the e-folding time of the gas (the number of years after which the concentration of the gas has decayed exponentially to 1/e, or 37%, of its initial value; in some cases, this is a function of the concentration C) (from eq. D.8)

PT_B = the perturbation lifetime at the concentration (burden) B; the effective lifetime of an incremental change to the concentration (burden), where the increment is assigned its effects on the lifetime of the original entire atmospheric burden (years)[8]

LT_B = the average lifetime or turnover time of the atmospheric burden B (years)
S = lifetime / concentration sensitivity parameter (0.28 for CH_4, -0.05 for N_2O)
LT_{B*} = the average lifetime or turnover time of a reference atmospheric burden (concentration) B* (8.4 years for CH_4, 120 years for N_2O)
B = the atmospheric burden (or concentration) (Tg or ppm)
B* = a reference atmospheric burden or concentration (1745 ppb for CH_4, 330 ppb for N_2O)

Because the IPCC provides estimates of the sensitivity parameter S and the lifetime at a reference burden LT_{B*}, we want an expression for the perturbation lifetime PT as a function of S and LT_{B*}. First we solve eq. D.10 for LT_B:

$$S \cdot \ln\left(\frac{B}{B*}\right) = \ln\left(\frac{LT_B}{LT_{B*}}\right)$$

$$\ln\left(\left(\frac{B}{B*}\right)^S\right) = \ln\left(\frac{LT_B}{LT_{B*}}\right)$$

$$\left(\frac{B}{B*}\right)^S = \left(\frac{LT_B}{LT_{B*}}\right)$$

$$LT_B = LT_{B*} \cdot \left(\frac{B}{B*}\right)^S \qquad \text{eq. D.11}$$

Substituting eq. D.11 into eq. D.9 results in:

$$PT_B = \frac{LT_{B*} \cdot \left(\frac{B}{B*}\right)^S}{1-S} \qquad \text{eq. D.12}$$

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[8] The perturbation lifetime can be u understood by considering the following “budget-balance” equation (IPCC, 2001, Table 4.5, p. 254):

$B / LT_B = B^* / LT_{B*} + ?B / PT$

where ?B = B-B*.

This shows that any changes in the lifetime of the original burden B* are assigned to the increment ?B (because the original burden B* is given its original lifetime LT_{B*}).

I use eq. D.12 to estimate the e-folding time of CH_4 and N_2O for a given target emissions year, where the values of B*, S, and LT_{B*} are given above, and the concentration *B* at the target emissions year is looked up from the table showing concentration scenarios, above[9].

Particulates (aerosols). The lifetime of aerosols generally is rather short compared to the residence times of greenhouse gases, on the order of a week, but it depends strongly on particle size. Both larger and smaller particles persist in the atmosphere for shorter periods than do medium-sized particles, with the highest residence times exhibited by particles with an aerodynamic radius of about 0.3 µm. Larger particles are removed more rapidly due to their larger settling velocities, and smaller particles rapidly become transformed to larger particles through Brownian motion and coagulation (Jaenicke, 1993). Data in Jaenicke (1993) and Pilinis et al. (1995) show that the most aerosol particles are in the range of radius size from 0.001 to 10 µm, with an approximate mean value of 0.1 µm.

Estimates of the lifetime of particulates are given in the following.

Gas	**Lifetime**	**Source**	**Comments**
sulfate	2.8 days for SO_4	IPCC (2001, App. II)	based on ratio of atmospheric burden (0.52 Tg S in SO_4) to annual emissions (69.0 Tg S / yr.)
OC aerosol	7.3 days	IPCC (2001, App. II)	based on ratio of atmospheric burden (1.52 Tg) to annual emissions (81.4 Tg / yr.)
OC aerosol	15 days	Bond et al. (2002)	Results from Model for Atmospheric Transport and Chemistry 4.1; based on ratio of emissions to burden
BC aerosol	7.7 days	IPCC (2001, App. II)	based on ratio of atmospheric burden (0.26 Tg)[10] to annual emissions (12.4 Tg / yr.)

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[9] The LEM calculates lifecycle CO_2-equivalent emissions for emissions in any user-specified year from 1970 to 2050. The LEM looks up the concentration (burden parameter *B* in eq. D.12) corresponding to the specified target year in the concentration / scenario tables.

[10] The IPCC (2001) estimate of an atmospheric burden of 0.26 Tg BC is consistent with estimates in Jacobson (2002). In Jacobson's (2002) simulation, the global loading of fossil-fuel (f.f.) BC plus organic matter (OM) varied from 0.25 to 0.60 Tg, or 0.08 to 0.20 Tg BC alone (one third of the total was BC, two thirds was OM). Elsewhere, Jacobson (2002) says that f.f. BC constitutes half of the total anthropogenic burden of BC, which therefore implies a total anthropogenic BC burden of 0.16 to 0.40 Tg, consistent with the IPCC estimate.

BC aerosol	4.6 days (troposphere)	Wang (2002)	based on 3-D aerosol-climate model to study the impact of black carbon on climate
BC aerosol	4 – 8 days	Wang (2002)	range given in three other studies cited by Wang (2002)
BC aerosol	10 days	Bond et al. (2002)	results from Model for Atmospheric Transport and Chemistry 4.1; based on ratio of emissions to burden
PM < 1.0 µm	~ 8 days	Delucchi and McCubbin (1996)	for PM in lower troposphere; see discussion below

(BC = black carbon, OC = organic carbon, PM = particulate matter.)

Other sources indicate a lifetime of about 8 days for small particles. Bond et al. (2003) cite several references in support of the statement that the lifetime of BC in the atmosphere is about 1 week. Delucchi and McCubbin (1996) use a formula in Wiman et al. (1990) to estimate the residence time of particles in the lower and upper troposphere as a function of their aerodynamic diameter:

Particle size	**lower troposphere (below 1.5 km)**	**mid troposphere to tropopause**
µm	*days residence*[a]	*days residence*[a]
0.1	7.6	18.5
0.2	7.9	20.0
0.5	7.9	20.0
1.0	7.5	18.0
1.5	7.0	15.4
2.0	6.4	12.8
3.0	5.2	8.7
5.0	3.2	4.3
10.0	1.1	1.3

Delucchi and McCubbin (1996) also review the literature on the size distribution of different types of particles, and assume the following mean mass-weighted aerodynamic diameters:

PM$_{2.5}$ and PM$_{10}$ exhaust from motor vehicles = 0.1 to 0.6 µm (best estimate of 0.2 µm)

PM$_{2.5}$ dust paved roads = 1.0 to 1.8 µm

PM_{10} dust paved roads = 4.0 to 7.0 µm.

Assuming a value of 0.2 µm for particles from gasoline and diesel combustion, the residence time would be 7.9 days in the lower troposphere, and 20 days in the upper troposphere. Larger particles, such as dust particles, would have a shorter residence time.

The lifetime of CO_2. The lifetime of CO_2 is a function of the concentration of CO_2 and of the time horizon being considered. (I explain both of these relationships below). Because the lifetime is a function of the concentration, I will use a variant of eq. D.12 to estimate the effective perturbation lifetime of an incremental increase in CO_2. The variant for CO_2 is:

$$PT_B = PT_{B*} \cdot \left(\frac{B}{B*} \right)^S \qquad \text{eq. D.13}$$

where:

PT_{B*} = perturbation lifetime for CO_2 at a reference concentration B* (a function of the time horizon being considered; discussed below)

Other terms are defined above.

This equation is obtained by substituting eq. D.9 into eq. D.12. In the case of CO_2 I have expressed the perturbation lifetime PT as a function of a reference perturbation lifetime PT_{B*} rather than as a function of a reference average burden lifetime LT_{B*} because I am able to derive an estimate of the former (PT_{B*}) but not of the latter.

Thus, our formula for the lifetime of CO_2, given in eq. D.13, requires an estimate of the perturbation lifetime at a reference atmospheric burden (PT_{B*}), the associated reference concentration (burden, B*), and the sensitivity parameter S that relates changes in the lifetime to changes in the concentration. I consider the perturbation lifetime at a reference atmospheric burden first, using data from the IPCC 2001). This reference lifetime turns out to be a function of the time horizon TH.

Reference perturbation lifetime PT_{B*}. The IPCC (2001) does not report a single e-folding lifetime PT for CO_2. This is because CO_2, unlike most non-CO_2 gases, is characterized by multiple distinct decay patterns, due to multiple distinct removal mechanisms. (By contrast, most non-CO_2 gases have one main removal mechanism and hence a single decay rate represented by a single e-folding parameter PT.) Some CO_2 is removed from the atmosphere quickly, in a matter of years; some remains for decades, and some remains for centuries (IPCC, 2001, 1996a).

In its calculations of GWPs, it appears that the IPCC (2001) estimates the parameter $X_{CO2}(t)$ – the amount of CO_2 remaining in the atmosphere, as a function of time – explicitly on the basis of multiple exponential decay functions. To represent this, I assme that there are *i* distinct fractions of CO_2, each with its own distinct lifecycle and removal mechanism. Each distinct fraction is Xi % of the total CO_2 emission and has an e-folding time of Li. Evaluating the exponential decay integral for each fraction and multiplying by the appropriate percentage for each CO_2 fraction (Xi), we can calculate the total amount of CO_2 remaining after TH years:

$$X_{CO2}(t = TH) = \frac{\sum_i Xi \cdot Li \cdot \left(1 - e^{\left(\frac{-TH}{Li}\right)}\right)}{TH} \qquad \text{eq. D.14}$$

The IPCC (2001) does not provide values for the parameters Li and Xi. As a result, we will have to take a different approach to estimating $X_{CO2}(t)$: we will estimate the single effective e-folding parameter – call it L^ -- that produces the same integrated decay over a given period of time TH as do the actual multiple decay rates (represented by Li) over the same period TH:

$$X_{CO2}(t = TH) = \frac{L^\wedge \cdot \left(1 - e^{\left(\frac{-TH}{L^\wedge}\right)}\right)}{TH} \qquad \text{eq. D.15}$$

The IPCC's (2001) discussion of the estimation of GWPs provides all of the information necessary to calculate L^. Specifically, referring to equation D.4 here, the IPCC provides all parameter values *except* the value of L_{CO2}^(t), for several gases. This means it is possible to back-calculate the values of L_{CO2}^(t) for different time horizons. Because equation D.4 cannot be solved easily for $L_{CO2}(t)$, we use trial and error to find the values of L_{CO2}^(t) that reproduce the IPCC GWPs given all of the other parameter values. As explained in the notes to Table D-3, the following values reproduce the IPCC GWPs exactly:

Time horizon (TH) yrs	*effective single CO_2 e-folding time (L_{CO2}^(t))*
20	23.0
100	53.0
500	150.0

If we were interested only in the IPCC time horizons, then the back-calculated values presented above would suffice. However, we may want to be able to estimate L^ for values of TH other than 20, 100, or 500 years. In order to this, we will have to estimate L^ as a function of TH. We can do this by infilling estimates of L^ for values of TH other than the three shown above, and then regressing L^ against TH to develop a simple function of the form L^ = K0 + K1·TH^{K2}, where K0 and K1 will be parameters estimated in the regression.

To generate values for this infilling, we first express L^ as a function of TH, Li, and Xi. We set eq. D.14 equal to eq. D.15 and solve for the single equivalent e-folding time L^ (although we still end up with L^ on both sides):

$$\frac{L^\wedge \cdot \left(1-e^{\left(\frac{-TH}{L^\wedge}\right)}\right)}{TH} = \frac{\sum_i Xi \cdot Li \cdot \left(1-e^{\left(\frac{-TH}{Li}\right)}\right)}{TH}$$

$$L^\wedge = \frac{\sum_i Xi \cdot Li \cdot \left(1-e^{\left(\frac{-TH}{Li}\right)}\right)}{1-e^{\left(\frac{-TH}{L^\wedge}\right)}} \qquad \text{eq. D.16}$$

Eq. D.16 can be solved for L^ (the single equivalent e-folding time) at different values of TH, given values of Li and Xi, by iterative calculations in a spreadsheet. First, though we must find Xi and Li. We do this by trial and error, trying different combinations of Xi and Li until we find those that reproduce the combinations of L^ and TH derived from the IPCC GWPs (as explained above). The following values of Xi and Li reproduce L^ = 23 when TH = 20, L^ = 53 when TH = 100, and L^ = 150 when TH = 500:

parameter Xi	*parameter Li*
56% of CO_2	e-folding time of 10 years
16% of CO_2	e-folding time of 200 years
28% of CO_2	e-folding time of 1000 years

These values also are consistent with the IPCC's (1996a) statement that 40-60% of the CO_2 that is released into the atmosphere is removed within 30 years, and with the modeling assumption in West et al. (1997) that 20% of emitted CO_2 remains in the atmosphere indefinitely. They also are broadly consistent with the diagram in Jain et al. (2000) that shows that the Bern Carbon Cycle model (which the IPCC [2001] uses) predicts that about 50% of CO_2 remains after 25 years, 38% remains after 50 years, 32% after 100 years, 28% after 150 years, and 22% after 500 years.

With these values of Xi and Li, we used equation D.16 to generate values of L^ at different time horizons TH. We then regressed the estimated series of L^ values against TH (transformed by some power), and produced the following estimate of L^(TH), which also is our estimate of the perturbation time PT_{B*}:

$$L_{CO2}{}^{\wedge}(TH) \equiv PT_{B*} = K0 + K1 \cdot TH^{K2} \qquad \text{eq. D.17}$$

where K0 = 8.477, K1 = 1.708, and K2 = 0.71.

This function reproduces the values of L^ that we back-calculated are used in the IPCC GWPs to within about 0.5%. It also produces reasonable values of the single e-folding time L^ for THs outside the IPCC range (years):

Time horizon (TH) →	*10*	*20*	*50*	*100*	*250*	*500*	*1000*
L_{CC2}^ estimated by eq. D.17	17.2	22.8	35.9	53.4	94.6	149.3	239
L_{CC2}^ derived from GWPs	n.e.	23.0	n.e.	53.0	n.e.	150.0	n.e.

*The reference concentration B**. In the preceding section, we essentially back-calculate the IPCC estimate of the perturbation lifetime of CO_2 implicit in the IPCC estimates of GWPs. In this section, we associate this estimate with a reference CO_2 burden (concentration) – parameter B* in eq. D.13. Unfortunately, the IPCC (2001, 1996a) gives few details regarding its estimate of the lifetime of CO_2 as part of its GWP calculations. However, for three reasons, I assume that the IPCC estimate of the lifetime of CO_2 (which it uses in its calculations of GWPs, and which we use as the basis of our estimate of PT [eq. D.17]) is associated with the year-2000 concentration of CO_2. First, the IPCC estimates of the lifetime of the non-CO_2 GHGs (used in the same GWP calculations that use the estimate of the lifetime of CO_2 derived above) clearly are based on current (ca. year 2000) concentrations of the non-CO_2 gases, and consistency would demand that the lifetime of CO_2 be estimated on the same basis. Second, in the its Second Assessment Report (SAR), the IPCC (1996) states that the GWPs are "referenced to the updated decay response for the adopted carbon cycle model…and future CO atmospheric concentrations held constant at current levels" (p. 121). (The Third Assessment Report uses the same carbon-response function used in the SAR [IPCC, 2001, p. 386].) Third (and least significantly), our own highly simplified parameterization of the carbon cycle, discussed in the next section, indicates that the perturbation lifetime of an incremental emission of CO_2 is around 30 years at the 1901 concentration, 50 to 250 years at the 2001 concentration, and over 300 years at the projected 2051 concentration. The result corresponding to the year 2001 (50 to 150 years) is consistent with the estimates of eq. D.17; whereas results corresponding to 1901 or 2051 concentrations are not.

Therefore, I assume that B* for CO_2 in eq. D.13 = 367 ppmv.

The sensitivity parameter S. Finally, we need to estimate the sensitivity parameter S in eq. D.13. As indicated in eq. D.10, this parameter relates changes in the lifetime of a gas to changes in the concentration. In the case of CO_2, S is based on the perturbation lifetime rather than the average burden lifetime. Substituting LT = PT · (1 – S) from eq. D.9 into eq. D.10 gives us S in terms of PT rather than LT:

$$S = \frac{\ln\left(\frac{PT_B}{PT_{B*}}\right)}{\ln\left(\frac{B}{B*}\right)} \qquad \text{eq. D.18}$$

The following shows how the estimated perturbation lifetime PT (years) of CO_2 varies with different values of S and the atmospheric burden (in ppmv):

B ¯ S ®	*0.40*	*0.60*	*0.80*	*1.00*	*1.20*	*1.40*	*1.60*
250	128	119	110	102	94	87	81
300	138	132	127	122	117	113	108
350	147	145	144	142	141	140	138
400	155	157	160	163	166	168	171
450	162	169	176	183	191	199	207
500	169	180	191	203	216	230	245
550	176	190	206	224	243	263	285
600	182	201	221	244	269	297	328
650	188	210	236	264	297	332	373
700	193	220	250	285	324	369	420
750	199	229	265	305	352	406	469
800	204	238	279	326	380	445	520

The IPCC (2001) and other sources discuss factors pertinent to the estimate of the sensitivity parameter, but do not provide a quantitative estimate of S for CO_2. In order to make an approximate estimate of S, we construct a simple, parameterized model of the carbon cycle which we use to estimate how the perturbation lifetime PT changes with the burden (concentration) B. In the appendix to this report (itself an appendix!) we discuss the construction of and results from our carbon-cycle model, and use the results from this model to make an estimate of S.

<Note: eq. D.19 and D.20 are in the appendix.>

The results of the carbon-cycle model documented in the appendix indicate that, for the emissions scenario chosen, the decay of an incremental emission dampens relatively rapidly with increasing concentrations, to the point that by the year 2021 the incremental emission does not even decay to 65% (by 35%) over the projection period. This suggests that emissions are relatively sensitive to concentration – i.e., that the value of S is relatively high. I assume a value of 1.2 (see the table above showing lifetime vs. the sensitivity parameter S and the burden B).

Assumptions regarding lifetime / decay in our analysis. The following summarizes our assumptions regarding the e-folding time in the calculation of CEFs for all of the gases in the LEM:

gas	**lifetime**	**source/comments**
CO_2	eqs. D.13, D.17, and D.18	Consistent with IPCC assumptions used to calculate GWPs. This is an estimate of the effective lifetime of an incremental change in concentration, as a function of concentration.
CH_4	eq. D.12	From IPCC (2001). See comments for CO_2.
N_2O	eq. D.12	From IPCC (2001). See comments for CO_2.
CFC-12	12 years	From IPCC (2001).
HFC-134a	13.8 years	From IPCC (2001).
SF_6	3,200 years	From IPCC (2001, ch. 6). Used in their GWP calculations.
CF_4	50,000 years	From IPCC (2001, ch. 6). Used in their GWP calculations.
C_2F_6	10,000 years	From IPCC (2001, ch. 6). Used in their GWP calculations.
O_3	15 days	IPCC (2001) says 3 – 20 days, but Daniel and Solomon (1998, p. 13253) assume 28 days.
CO	60 days	From IPCC (2001), showing 30-90 days, and Figure 2 and Table 5 in Daniel and Solomon (1998) indicating an e-folding time of about 60 days
BC	8 days	Sources discussed in data review above.
OM	8 days	Assume same as BC.

SOA	5 days	Assume similar to sulfate.
sulfate ($2(NH_4)SO_4$)	4 days	IPCC (2001) and Delucchi and McCubbin (1996), who indicate longer residence time than IPCC (2001).
nitrate (NH_4HNO_3)	3 days	Assumed to be slightly less tan for sulfate, based on data discussed in Delucchi and McCubbin (1996) indicating that nitrate aerosol is slightly larger than sulfate aerosol.
dust	2 days	Based on Delucchi and McCubbin (1996) analysis summarized above, assuming dust is relatively large.

Direct radiative forcing (function F(t))

In this section, we discuss our estimates of unit direct radiative forcing, in $Wm^{-2}ppmv^{-1}$ for all gases except aerosols and ozone, which are in Wg^{-1}. (Also, in the case of aerosols, our estimates here will include indirect radiative forcing.) First we analyze and summarize the available estimates of radiative forcing, then we tabulate our assumptions. Note that in keeping with our method of estimating CEFs for incremental emissions of a gas, our estimate of the $Wm^{-2}ppmv^{-1}$ radiative forcing is an estimate of the forcing of an incremental change in the concentration, not an estimate of the average forcing of a given concentration.

For many gases, and for CO_2, CH_4, and N_2O in particular, the change in radiative forcing *per unit of the gas* (e.g., Wm^{-2} $ppmv^{-1}$) is a function of the concentration. The radiative forcing of a 1 ppmv increase in the concentration of a gas starting from the current (year-2000) concentration is greater than the radiative forcing of a 1 ppmv increase starting from twice the current concentration, because (in loose terms) as the concentration increases the molecules start crowding one another out and “competing” for radiation, and as a result the amount of radiation to be absorbed by each molecule decreases.

CO_2, CH_4, N_2O. The IPCC (2001, Table 6.2) provides formulae that show the change in radiative forcing as a function of a change on concentration, where the concentration change is represented by an initial concentration C_O and a post-change concentration C. These formula are provided for CO_2, CH_4, and N_2O. (There is a formula for CFC-12, but it shows that the unit forcing is constant at 0.32 Wm^{-2} $ppbv^{-1}$.) I use these formula to estimate the radiative forcing for a one-unit increase in concentration ($C= C_O+1$) for values of C_O ranging from the concentration in 1970 (the earliest analysis year in the LEM) to projected far-distant-future concentrations, for CO_2, CH_4, and N_2O (see the subsection on emissions and concentrations scenarios). The result is a projection of the unit radiative forcing (in W m^{-2} $ppmv^{-1}$) over a range of historical, current, and projected concentrations[11].

The IPCC (2001, Table 6.2) equations are as follows:

$$F(C)_{CO2} = 4.841 \cdot \ln[(C_{CO2}+1) / C_{CO2}] + 0.0906 \cdot [(C_{CO2}+1)^{0.5} – C_{CO2}^{0.5}] \qquad \text{eq. D.21}$$

$$F(C)_{CH4} = 0.036 \cdot [(C_{CH4}+1)^{0.5} – C_{CH4}^{0.5}] – [f(C_{CH4}+1, C_{N2O}) – f(C_{CH4}, C_{N2O})] \qquad \text{eq. D.22}$$

$$F(C)_{N2O} = 0.012 \cdot [(C_{N2O}+1)^{0.5} – C_{N2O}^{0.5}] – [f(C_{CH4}, C_{N2O}+1) – f(C_{CH4}, C_{N2O})] \qquad \text{eq. D.23}$$

$$f(C1, C2) = 0.47 \cdot \ln[1+2.01 \cdot 10^{-5} (C1 \cdot C2)^{0.75}+5.31 \cdot 10^{-15} C1 \cdot (C1 \cdot C2)^{1.52}] \qquad \text{eq. D.24}$$

where:

$F(C)_{CO2}$ = the change in radiative forcing associated with a one-unit change in the concentration of CO_2 (w m^{-2} $ppmv^{-1}$)
$F(C)_{CH4}$ = the change in radiative forcing associated with a one-unit change in the concentration of CH_4 (w m^{-2} $ppbv^{-1}$)
$F(C)_{N2O}$ = the change in radiative forcing associated with a one-unit change in the concentration of N_2O (w m^{-2} $ppbv^{-1}$)
C_{CO2} = the concentration of CO_2, in ppmv
C_{CH4} = the concentration of CH_4, in ppbv
C_{N2O} = the concentration of N_2O, in ppbv

As one would hope, the IPCC formulae reproduce values used by the IPCC to calculate GWPs, and are consistent with other values reported by the IPCC (2001). For example, the IPCC (2001) uses these formula to calculate radiative forcing for small changes around the current concentration of CO_2, CH_4, and N_2O. These current-C radiative-forcing values, in turn, are used in the IPCC calculations of GWPs (see the notes to Table D-3 here). In the case of CO_2, the radiative forcing at the current concentration (about 368 ppmv) is 0.0155 Wm^{-2} $ppmv^{-1}$. The IPCC also estimates that the historical 87 ppmv increase the concentration of CO_2 from the year1750 to present is responsible for 1.46 Wm^{-2} – an average of 0.0168 Wm^{-2} $ppmv^{-1}$ over the period[12]. The

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[11] The IPCC (2001) does not use these formula in its calculations of GWPs and instead assumes a constant unit radiative forcing for each gas presumably because the GWP formula is easier to integrate if the radiative forcing term is constant.

Reilly et al. (2001) use formula similar to the IPCC formulae to estimate radiative forcing as a function of concentration in their estimation of EDIs for CO_2, N_2O, and CH_4.

[12] Jain et al. (2000) estimate an identical value, 0.168 Wm^{-2} $ppmv^{-1}$ for the historical effect of CO_2. However, this is not an independent estimate, because the IPCC (2001) refers to the work of Jain et al. (2000) in its estimates. On the other hand, for other gases the estimates of Jain et al. (2000) differ somewhat from those of the IPCC (2001) (Wm^{-2} $ppmv^{-1}$):

IPCC (2001, Table 6.2) formula for CO_2 used here indicates that the unit radiative forcing starts at 0.0200 at the pre-industrial concentration and declines to 0.0155 today, consistent with the reported value of 0.0168 over the entire period. Similarly, the IPCC estimates that future radiative forcing will average about 0.011 Wm^{-2} $ppmv^{-1}$ over a future near-doubling of the concentration of CO_2; their formula indicates that the unit forcing will decline at 0.0155 today's concentration to 0.009 at the nearly doubled concentration.

Ozone. The IPCC (2001), Harvey et al. (2000), Derwent et al. (2001), Joos et al. (2001) and Daniel and Solomon (1998) present estimates of the radiative forcing of ozone:

Gas	**Radiative forcing**	**Source**	**Comments**
ozone (O_3)	+0.35 Wm^{-2} +0.48 Wg^{-1}.	IPCC (2001, ch. 6)	Central estimate. I calculated Wg^{-1} based on IPCC (2001, p. 261) estimate of 370 Tg O_3 globally.
ozone (O_3)	+0.3 to +0.7 Wm^{-2} in the global mean	Estimate reported in Harvey (2000)	Apparently applies to current (~year 2000) concentrations.
ozone (O_3)	+ 0.6 to 1.2 Wm^{-2} in the global mean	my est. based on Derwent et al. (2001)	Derwent et al. (2001) 3D model gives 0.023 Wm^{-2} $ppbv^{-1}$ change in mean global O_3 in 4.8 to 11.2 km range, which according to them is where ozone changes exhibit greatest radiative forcing. IPCC (2001) says 0.65 DU $ppbv^{-1}$ and 34 DU O_3 globally; if 50% of O_3 but 100% of O_3 forcing is in 4.8 to 11.2 km range, result is 0.6 Wm^{-2}.

	Jain et al. (2000)	IPCC (2001)	% difference
CH_4	0.570	0.459	-19%
N_2O	3.25	3.41	5%
CFC-12	302	319	6%

The Jain et al. (2000) estimate for CH_4 is quite a bit higher than the IPCC (2001) estimate, and as a result Jain et al. (2000) estimate a significantly higher 100-year GWP for methane (28 vs. 23 for the IPCC). I use the IPCC (2001) estimates because the IPCC (2001) reviewed the work of Jain et al. (2000).

ozone (O$_3$)	+0.38 Wm^{-2}	Joos et al. (2001)	Current forcing.
ozone (O$_3$)	+0.6 Wm^{-2}	Daniel and Solomon (1998)	They assume 30 ppbv increase in O$_3$ and 0.02 Wm^{-2}ppbv^{-1}.

Particulates (aerosols). The following table summarizes a number of estimates of the direct and indirect radiative forcing of sulfate, nitrate, black carbon (BC), and organic carbon (OC) aerosols. We pay special attention to the estimates of Jacobson (2001, 2002, 2003), because these appear to be the most sophisticated available.

Gas	**Radiative forcing**	**Source**	**Comments**
sulphate aerosol (direct forcing)	-0.3 to –0.8 Wm^{-2} (IPCC picks –0.4) –110 to –460 W(g-sulphate)$^{-1}$	Estimates tabulated in IPCC (2001, ch. 6)	Present (year 2000) forcing. Covers most of the studies in Harvey (2000) plus some more recent one. Most estimates around –200 W(g-sulphate)$^{-1}$
sulphate aerosol (direct forcing)	-0.3 to –0.7 Wm^{-2}	Estimates tabulated in Harvey (2000)	Present day forcing.
sulphate aerosol (direct forcing)	-0.35 Wm^{-2} -140 W(g-sulphate)$^{-1}$	Jacobson (2002, 2003a)	Jacobson (2002) reports Wm^{-2} direct forcing; Jacobson (2003a) says total atmospheric loading of sulfate was 1.3 Tg.
sulphate aerosol	-0.4 to –1.6 Wm^{-2}	Webster et al. (2002)	Projected forcing year 2100. Presumably direct effect only.
sulphate aerosol (indirect forcing)	-0.4 to –5 Wm^{-2}	Estimates tabulated in IPCC (2001, ch. 6)	The IPCC does not express the indirect effects in Wg^{-1}, but assuming that the gm^{-2} values used in the analysis of direct effects apply, the indirect effects would be on the order of –400 W(g-sulphate)$^{-1}$.
sulphate aerosol (indirect	-0.4 to –2.2 Wm^{-2}	Estimates tabulated in Harvey (2000)	Present day forcing. Most estimates around –1.0 Wm^{-2}. See also Mitchell et al. (1995).

forcing)			
nitrate aerosol (direct forcing?)	-0.02 to –0.2 Wm^{-2} –30 to –350 W(g-NO$_3$)$^{-1}$	IPCC (2001, ch. 6)	IPCC (2001) says that the lower estimate assumes most nitrate aerosol is coarse, but that this might not be true. Data in Delucchi and McCubbin (1996) indicate that most nitrate is between 1.0 µm and 2.5 µm.
dust aerosols (direct forcing)	-0.60 to + 0.40 Wm^{-2}	IPCC (2001, ch. 6)	IPCC states that the net forcing is likely to be negative, but that a positive forcing cannot be ruled out.
BC f.f. combustion (direct forcing)	0.20 Wm^{-2} 1400 Wg^{-1}	IPCC (2001, ch. 6)	Central estimate. I calculate W g^{-1} assuming 0.14 mg / m^{-2}.
OC f.f. combustion (direct forcing)	– 0.02 to –0.24 Wm^{-2} –36 to –340 Wg^{-1}	IPCC (2001, ch. 6)	Range reported in three studies.
BC from all sources (total forcing)	+0.37 Wm^{-2} (726 Wg^{-1})	Bond et al. (2002)	Results from Model for Atmospheric Transport and Chemistry 4.1.
OC from all sources (total forcing)	-0.22 Wm^{-2} (-80 Wg^{-1})	Bond et al. (2002)	Results from Model for Atmospheric Transport and Chemistry 4.1.
BC from f.f. + b.m. combustion (direct forcing)	+0.55 Wm^{-2}	Jacobson (2001)	see Jacobson (2002) for more recent estimates.
BC+OM from f.f. combustion (direct	+0.25 Wm^{-2} (200 to 500 Wg^{-1})	Jacobson (2002)	I derive Wg^{-1} from reported Wm^{-2} given 0.25 to 0.60 Tg BC+OM.

forcing)			
BC+OM from f.f. combustion (total forcing)	+0.53 Wm^{-2} (450 to 1100 Wg^{-1})	Jacobson (2002)	I derive Wg^{-1} from reported Wm^{-2} given 0.25 to 0.60 Tg BC+OM.
BC from all sources (total forcing)	+2860 to +3370 Wg^{-1}	Based on Jacobson (2002, 2003)	My derivation based on values reported in Jacobson; see discussion below.
OC from f.f. combustion (direct forcing)	–43 to –64 Wg^{-1}	Jacobson (2002)	Low value accounts for UV absorption by organics.
OM from all sources (total forcing)	-470 to -740 Wg^{-1}	Based on Jacobson (2002, 2003)	My derivation based on values reported in Jacobson; see discussion below.
all aerosols (indirect effects)	0 to –2.0 Wm^{-2}	IPCC (2001, ch. 6)	includes “1st” and “2nd” indirect effects for all aerosols.

(BC = black carbon, OC = organic carbon, OM = organic matter; f.f. = fossil fuel, b.m.= biomass, DU = Dobson Unit.)

In a series of steps we can derive from the work of Jacobson (2002, 2003) estimates of the total Wg^{-1} forcing of BC aerosol and OM aerosol. Formally, we set up two equations, which will have two unknowns (the Wg^{-1} forcing of BC and the Wg^{-1} forcing of OM), and solve for the unknowns. The two equations state that the total global radiative forcing due to f.f. or b.m aerosols is equal to product of the atmospheric loading and the radiative forcing per unit mass, for each major constituent. In Jacobson (2002), f.f. aerosols comprise BC and OM; in Jacobson (2002, 2003), b.m. aerosol comprise BC, OM, and other constituents, such as K^+ and Na^+. Hence:

$$\begin{aligned} &AL_{ffBC} \cdot TF_{BC} + AL_{ffOM} \cdot TF_{OM} = TGF_{ffBC+OM} \\ &AL_{bmBC} \cdot TF_{BC} + AL_{bmOM} \cdot TF_{OM} + AL_{bm*} \cdot RTF_{*} \cdot TF_{OM*} = TGF_{bmPM} \end{aligned} \qquad \text{eqs. D.25, D.26}$$

where:

AL_{ffBC} = the atmospheric loading of f.f. BC (0.14 Tg; Jacobson [2002] reports that the loading of f.f. BC+OM varied between 0.25 and 0.60 Tg, and that 1/3 of this was BC; I assume a mid-range value of 0.42 Tg BC+OM [which is consistent with the IPCC [2001, Appendix II] estimates], and hence 0.14 Tg f.f. BC)
TF_{BC}= the total radiative forcing of BC (Wg^{-1}; an unknown to be solved for)
AL_{ffOM} = the atmospheric loading of f.f. OM (0.28 Tg; see f.f. BC loading, above)
TF_{OM}= the total radiative forcing of OM (Wg^{-1}; an unknown to be solved for)
$TGF_{ffBC+OM}$= the total global forcing of f.f. OM+BC (270 W; Jacobson [2002] reports a total forcing of 0.53 Wm^{-2} at the tropopause, and I estimate an earth surface area of 510 Tera-m^{2})
AL_{bmBC} = the atmospheric loading of b.m. BC (0.12 Tg; Jacobson [2002] reports that his simulations assumed emissions of 5.1 Tg/yr f.f. BC and 4.5 Tg/yr b.m. BC, and Jacobson [2003a] states that the same inventory was used in Jacobson [2002] and Jacobson [2003]; assuming that the lifetime of b.m. BC is the same as the lifetime of f.f. BC, then the atmospheric loading of b.m. BC is 88% [4.5/5.1] of the 0.14 Tg loading of f.f. BC)
AL_{bmOM} = the atmospheric loading of b.m. OM (0.96 Tg; Jacobson [2002, 2003] assumes 8:1 OM:BC ratio for b.m. aerosols)
AL_{bm*} = the atmospheric loading of b.m. other constituents (0.24 to 0.38 Tg; Table 3 of Jacobson [2003] indicates that the other constituents of b.m aerosols are 25-40% of OM)
RTF_{*} = the total radiative forcing of other b.m. aerosol constituents *relative to* the forcing of OM (unit less scalar; I do calculations for values of 1.0 [same forcing as OM] and 2.0 [twice the forcing of OM])
TGF_{bmPM} = the total global forcing of biomass aerosols (-460 W; Jacobson [2003] reports -0.84 Wm^{-2} average total forcing over 10 years of simulations, and –0.6 to –1.2 Wm^{-2} total forcing in years 5 to 10; I assume –0.90 Wm^{-2} and 510 Tera-m^{2} earth surface area)

We solve the eq. D.25 for the radiative forcing of BC:

$$TF_{BC} = \frac{TGF_{ffBC+OM} - AL_{ffOM} \cdot TF_{OM}}{AL_{ffBC}} \tag{eq. D.27}$$

Then we substitute this into equation D.26 and solve for the radiative forcing of OM, in terms of known parameters:

$$TF_{BC} = \frac{TGF_{ffBC+OM} - AL_{ffOM} \cdot TF_{OM}}{AL_{ffBC}}$$

$$AL_{bmBC} \cdot \frac{TGF_{ffBC+OM} - AL_{ffOM} \cdot TF_{OM}}{AL_{ffBC}} + AL_{bmOM} \cdot TF_{OM} + AL_{bm*} \cdot RTF_{*} \cdot TF_{OM*} = TGF_{bmPM}$$

$$AL_{bmBC} \cdot \left(TGF_{ffBC+OM} - AL_{ffOM} \cdot TF_{OM}\right) + AL_{bmOM} \cdot TF_{OM} \cdot AL_{ffBC} + AL_{bm*} \cdot RTF_{*} \cdot TF_{OM*} \cdot AL_{ffBC}$$
$$= AL_{ffBC} \cdot TGF_{bmPM}$$

$$-AL_{ffOM} \cdot TF_{OM} \cdot AL_{bmBC} + AL_{bmOM} \cdot TF_{OM} \cdot AL_{ffBC} + AL_{bm*} \cdot RTF_{*} \cdot TF_{OM*} \cdot AL_{ffBC}$$
$$= AL_{ffBC} \cdot TGF_{bmPM} - AL_{bmBC} \cdot TGF_{ffBC+OM}$$

$$TF_{OM} = \frac{AL_{ffBC} \cdot TGF_{bmPM} - AL_{bmBC} \cdot TGF_{ffBC+OM}}{-AL_{ffOM} \cdot AL_{bmBC} + AL_{bmOM} \cdot AL_{ffBC} + AL_{bm*} \cdot RTF_{*} \cdot AL_{ffBC}} \qquad \text{eq. D.28}$$

With these equations and the parameter values above, we find:

TF_{OM} = - 470 W / g
TF_{BC} = 2860 W / g

when AL_{bm*} = 0.38 Tg and RTF_{*} = 2.0, and

TF_{OM} = - 720 W / g
TF_{BC} = 3370 W / g

when AL_{bm*} = 0.24 Tg and RTF_{*} = 1.0

Note that the main report discusses estimates of emissions of BC and OM aerosol for the LEM. In that report, we account for the effect of the other constituents of biomass aerosols by scaling the effect of OM from biomass combustion.

Deriving CO_2 forcing in Wg^{-1}. In the case of the CEF for aerosols and ozone, we estimate the quantity Wg^{-1} rather than the quantity W $ppmv^{-1}$ m^{-2}. (We do this because aerosol concentration data are given in a mass rather than a volume basis, and the radiative forcing estimates for aerosols are in Wg^{-1}.) We estimate this quantity by dividing the calculated [W $ppmv^{-1}$ m^{-2}] by [g $ppmv^{-1}$ m^{-2}]. This requires an estimate of the quantity g $ppmv^{-1}$ m^{-2} for CO_2. We get this by estimating the density of the atmosphere as a function of height:

$$COL_{CO2} = PPM_{CO2} \cdot MW_{CO2} \cdot DN_{AVE/SUR,H} \cdot DN_{SUR} \cdot H \qquad \text{eq. D.29}$$

$$DN_{SUR} = \frac{P_{SUR}}{R \cdot T_{SUR}}$$

$$DN_{AVE/SUR,H} = \frac{\int_0^H DN_{AVE/SUR}(H)\,dH}{H}$$

$$DN_{AVE/SUR}(H) = e^{-\frac{H}{H_{REF}}}$$

$$DN_{AVE/SUR,H} = \frac{H_{REF} \cdot \left(-e^{-\frac{H}{H_{REF}}} + 1\right)}{H}$$

where:

COL_{CO2} = the column density of CO_2 (g / m^2 / ppmv)
PPM_{CO2} = one ppmv CO_2
MW_{CO2} = the molecular mass of CO_2 (44.01 g / mole)
$DN_{AVE/SUR,H}$ = the average density of the atmosphere (in mole / L) from the surface to the assumed height H of the atmosphere, *relative* to the density at the surface
DN_{SUR} = the density of the atmosphere at the surface of the earth (mol / L; calculated from the ideal gas law)
$DN_{AVE/SUR}(H)$ = the average density of the atmosphere relative to the density at the surface as a function of the height H (a simple exponential relationship)
P_{SUR} = the pressure at the surface of the earth (H=0) (1 atm.)
R = the gas constant (0.082057 L atm. K^{-1} mol^{-1})
T_{SUR} = average temperature at the surface of the earth (H=0) (assumed to be 278^{o} K)
H = the height of the atmosphere assumed to contain CO_2 (various sources appear to assume 8 -12 km for this sort of analysis, but the atmosphere extends beyond this; I will do the analysis for 20 km)
H_{REF} = a reference height used in the density vs. height equation (8 km)

For the parameter values above, COL_{CO2} = 9.8 to 14.2 g ppmv^{-1} m^{-2} for atmospheric heights of 8 to 20 km. This result of eq. D.29 can be compared with values derived from the IPCC (2001, ch. 3) and Jacobson (2002). The IPCC (2001) estimates that the in the 1980s the atmosphere contained 730 Pg of C, or 2680 Pg of CO_2. (Similarly, Jacobson [2002, p. 16-10] estimated a forcing of 1.6 Wm^{-2} due to 182 Pg-C of anthropogenic CO_2([13]). If the anthropogenic CO_2 is 25% of the total, then the total implied by Jacobson is 730 Pg C.) Dividing by an earth surface area of $5.09 \cdot 10^{14}$ m^2 (based on a radius of the earth of 3955 miles) and a 1980s concentration of 350 ppmv results in 15.0 g $ppmv^{-1}$ m^{-2}, which is consistent with the result estimated for a 20 km column. This seems reasonable, because remarks in the IPCC (2001) indicate that they consider the entire height of the atmosphere.

Assumptions regarding radiative forcing in our analysis. The following shows our assumptions for the parameter $F_i(t)$:

gas	**forcing (F)**	**source/comments**
CO_2	eq. D.21	See discussion above. Consistent with IPCC assumptions used to calculate GWPs.
CH_4	eq. D.22	See discussion above. Consistent with IPCC assumptions used to calculate GWPs.
N_2O	eq. D.23	See discussion above. Consistent with IPCC assumptions used to calculate GWPs.
CFC-12	320 Wm^{-2} $ppmv^{-1}$	From IPCC (2001). (Value used in IPCC's GWP calculations.)
HFC-134a	150 Wm^{-2} $ppmv^{-1}$	From IPCC (2001).
SF_6	520 Wm^{-2} $ppmv^{-1}$	From IPCC (2001, ch. 6). Used in IPCC's GWP calculations.
CF_4	80 Wm^{-2} $ppmv^{-1}$	From IPCC (2001, ch. 6). Used in IPCC's GWP calculations.
C_2F_6	260 Wm^{-2} $ppmv^{-1}$	From IPCC (2001, ch. 6). Used in IPCC's GWP calculations.
O_3	+0.40 Wm^{-2} 0.55 Wg^{-1}	Based on studies cited above.
CO	0.32 Wm^{-2} $ppmv^{-1}$ (direct)	The IPCC (2001) cites a study in which an increase in the concentration of CO from 25 to 100 ppbv causes a direct radiative forcing of 0.024 Wm^{-2}. Indirect effects calculated in a separate section.
BC	+2600 Wg^{-1}	Total forcing, including "indirect." Based on data summarize above, with extra weight given to Jacobson

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[13] The paper doesn't state that the units are C instead of CO_2, but in a personal communication Jacobson has confirmed that the units are C.

		(2002).
OM	- 350 Wg^{-1}	Total forcing, including “indirect.” Based on data summarize above, with extra weight given to Jacobson (2002, 2003).
SOA	-250 Wg^{-1}	Assume same as nitrate, based on studies of direct forcing (see above).
sulfate	-550 $W(g\text{-}SO_4)^{-1}$	Total forcing, including “indirect.” On basis of studies above, assume 0.40 Wm^{-2} direct, 1.0 Wm^{-2} indirect forcing.
nitrate	-250 $W(g\text{-}NO_3)^{-1}$	Total forcing, including “indirect.” Based on data summarize above.
dust	-200 Wg^{-1}	Total forcing, including “indirect.” Based on data summarize above.

Note that we have included the direct effects of CO, even though they are small.. Using the parameter values given above, the IPCC 100-year GWP for CO would be 0.2. Although this is two orders of magnitude lower than the GWP for CH_4, one should keep in mind that global emissions of CO are about 5 times larger than are global emissions of CH_4, and that some of the indirect effects of CO (discussed below) are not much larger than the estimated direct effect of CO.

Indirect radiative forcing (function I(t))

In this section we discuss in the indirect radiative forcing of CH_4, O_3, and CFC-12. (The indirect forcing of aerosols is included in the previous subsection on direct forcing, and the indirect effects of CO NO_x, SO_x CO, and NMOCs are discussed in a separate major section below.)

Methane (CH_4.) Over the past decade, estimates of the indirect effects of methane have become more sophisticated. In its first estimate, in 1990, the IPCC estimated that the indirect effects were slightly more than 100% of the direct effects (Shine, et al., 1990). In 1992 the IPCC (1992) revised its estimate of the direct effect upward by 10-20%, and stated that the indirect-warming effects could be “comparable in magnitude to the direct value” (p. 15). In 1993, Bruhl (1993) estimated considerably lower GWPs than had the IPCC (10-13 for a 50-year time horizon, and 6-8 for a 100-year time horizon). Bruhl (1993) modeled the GWP under two scenarios, one in which emissions increase over time, and another in which the concentration and lifetime of CH4 is fixed. The former yielded higher GWPs, because increasing concentrations of CH4 (in the face of a relatively constant amount of the scavenger, OH) result in a longer average lifetime for CH4. Bruhl explained that his estimate of the indirect GWP of CH4 was lower than the IPCC’s estimate of the indirect effect because the IPCC probably overestimated the production of O3 due to CH4.

In the second IPCC assessment (IPCC, 1996), the GWPs were revised to include the indirect effects of CH_4 on tropospheric O_3 and stratospheric water vapor production, and to reflect a somewhat lower atmospheric lifetime than was previously used (12±3 versus 14.5±2.5 years). This revised CH_4 lifetime was due to two factors: first, a new estimate for the chemical removal rate that was 11% faster than the estimate previously used, and second, the inclusion of CH_4 uptake by soils (IPCC, 1996). The end result of these re-estimates of both the direct and indirect radiative forcing effects of CH_4 was that the 1995 IPCC 20-year GWP value was 11% lower than the 1990 GWP, the 100-year value was unchanged, and the 500-year value was 38% lower (IPCC, 1996).

Kandlikar (1996) estimated an EDI for the indirect as well as the direct effects of methane (Table D-3 here). In his analysis, the indirect effect, which accounted for the effect of CH_4 on O_3, H_2O, and OH, was about 1/2 of the direct effect and hence about 1/3 of the total EDI.

In its TAR, the IPCC (2001) discusses four indirect effects of methane:

i) on the production of tropospheric ozone (they estimate this to be 25% of the direct-forcing GWP);

ii) on the production of stratospheric water vapor (they estimate this to be 5% of the direct-forcing GWP);

iii) on its own lifetime, via destruction of OH (they include this effect in their estimate of the perturbation lifetime); and

iv) on the production of CO_2 via the final oxidation of the carbon in CH_4 (the IPCC does not include this in its GWP on the grounds that the conversion of the C in CH_4 to C in CO_2 is already counted in national inventories of CO_2 that assume that all fuel carbon is converted ultimately to CO_2).

Finally, Derwent et al. (2001) use a sophist acted 3-D climate model to estimate that the indirect effect of methane on tropospheric ozone has a 100-year GWP of 3.3. This is somewhat lower than the IPCC (2001) value of about 4.4 (25% of the direct radiative forcing GWP of 17.5). Derwent et al. (2001) do not estimate the effect of methane on stratospheric water vapor.

Our assumptions for methane. The IPCC (2001) multiplies the estimated direct forcing by 1.30 to account for the effects of methane on ozone and water vapor. The recent analysis by Derwent et al. (2001) suggests that this factor might be a little too high. We therefore multiply our estimated direct forcing by a total factor of 1.25 to account for the indirect forcing due to the effect on tropospheric ozone and stratospheric water vapor.

The effect of methane on its own lifetime is already accounted for in our estimate of the lifetime of methane.

Lastly, we include the discounted present value of the final oxidation of the carbon in CH_4 to carbon in CO_2 (because in the LEM the fate of carbon is tracked explicitly). There are two aspects to this. First, when a gram of methane is oxidized, it ends up as 2.75 grams of carbon dioxide, because oxygen molecules are heavier than the hydrogen they replace. However (second), the oxidation to CO_2 occurs L_{CH4} years after the initial emission of CH_4, where L_{CH4} is the average lifetime of CH_4. This means that, compared with the damages from a gram of CO_2 emitted today, the damages from CO_2 effectively emitted L_{CH4} years from now are discounted by a factor DF, which is a function of the discount rate (which itself is a function of time) as well as of L. Formally:

$$CEF_{CH4/CO2} = \frac{MW_{CO2}}{MW_{CH4}} \cdot DF\Big|^{t=L_{CH4}}$$ eq. D.30

where:

$CEF_{CH4/CO2}$ = the CO_2-equivalency of the effect of the final oxidation of CH_4 to CO_2

MW_{CO2} = the molecular mass of CO_2 (44.01 g / mole)

MW_{CH4} = the molecular mass of CH_4 (16.04 g / mole)

DF = the discount factor, a function of time and the discount rate (see the subsection on discounting), evaluated for time = L_{CH4}

L_{CH4} = the atmospheric lifetime of CH_4 (see discussion elsewhere in this major section)

O_3. Ozone has at least two indirect but related effects on the carbon cycle and climate: an increase in the concentration of ozone reduces the carbon content of soils (Loya et al., 2003) and reduces the net primary productivity (NPP) of plants (Felzer et al., 2003). Both effects recently have been quantified[14].

Soil carbon. Loya et al. (2003) measured the total soil carbon (excluding roots and coarse material) in forest soils exposed for 3.5 years (May 1998 to October 2001) to four different concentrations of O_3 and CO_2, and found the following:

Treatment	*Soil (g-C/m²)*	*? w.r.t. control*	*% change*
control (347 ppmv CO_2, 37 ppbv O_3)	5,385	0	0
high O_3 (347 ppmv CO_2, 52 ppbv O_3)	5,237	-148	-2.7%
high CO_2 (560 ppmv CO_2, 37 ppbv O_3)	5,683	+298	5.5%
high O_3, CO2 (560 ppmv CO_2, 52 ppbv O_3)	5,114	-271	-5.0%

With these data and a number of assumptions, we can make an order-of-magnitude estimate of the effect of O_3 on the carbon content of the soil, and then translate this into a measure of equivalent fossil-fuel CO_2 emissions. In the low case I

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[14] In their model of global N deposition and associated C sequestration, Holland et al. (1997, p. 15862) discuss the effect of O_3 on vegetation, but do not explicitly model O_3 damage.

assume that the 15 ppbv increase in O_3 is associated with a 148 g-C / m^2 reduction in the C content of all soils globally; in the high case, I assume that the 15 ppbv increase is associated with a 5% decrease in the C content of all soils globally. (The low case assumes the current concentration of atmospheric CO_2, and that the absolute change in C content rather than the percentage change from Loya et al. [2003] is relevant; the high case assumes the high future concentration of CO_2, and that the percentage change in the C content is relevant.) Given an average of as much as 13,400 g-C / m^2 in all soils globally and $1.5 \cdot 10^{14}$ m^2 of soils globally (IPCC, 2001, p. 192), we calculate a low ozone effect estimate of $1.5 \cdot 10^{15}$ g-C / ppbv-O_3, and a high estimate of $6.8 \cdot 10^{15}$ g-C / ppbv-O_3, over 3.5 years. Given 7.1 Tg-O_3 / ppbv-O_3, based on IPCC (2001, p. 261), the estimates become 210 and 950 g-soil-C-reduced / g-atmospheric-O_3, over 3.5 years of exposure.

The foregoing ozone-effect measures are not CEFs. Two adjustments to the foregoing measures are required to derive a CEF, which equates a gram of O_3 addition (in effect an "emission") to the atmosphere with a gram of net fossil-fuel CO_2 emission. These adjustments translate the ozone quantity in the denominator and the carbon quantity in the numerator into equivalent effective *emissions* unit. Consider the ozone quantity first. The figure 210 g-C / g-atmospheric-O_3 means that 210 g of C are lost from the soil for every gram of O_3 in the atmosphere for 3.5 years. If ozone has a mean lifetime of 10 - 20 days (see above), then in order to maintain one gram of ozone in the atmosphere for 3.5 years, about 65 – 130 grams of ozone would have to be added to the atmosphere (in effect, "emitted") over 3.5 years – one gram every 10 to 20 days over about 1300 days. Thus, when the ozone effect measure is expressed per gram of O_3 added (or emitted), the result is 2 – 15 g-C / g-O_3-added (emitted).

The final step is to translate the change in soil carbon over 3.5 years to an equivalent effective net emission of fossil-fuel CO_2. This step goes in several parts. To begin here, we recognize that all of the carbon "lost" due to the elevated O_3 (i.e., the difference between the carbon content off the soil without elevated O_3 and the carbon content with elevated O_3) was in effect an emission of C-CO_2 during the experiment due to the elevated CO_2 – an emission (or reduced uptake) that would not have occurred had O_3 not been elevated. This effective increased emission of CO_2 occurred as either an accelerated oxidation of soil carbon or a reduction in the formation of soil carbon (Loya et al., 2003), where the latter is tantamount to a reduction in uptake of atmospheric CO_2, (which in turn is identical to an emission [addition] of CO_2 to the atmosphere). However, in the "control" or non-elevated-O_3 case, the C-CO_2 that was *not* lost during the experiment (relative to the elevated-O_3 case) – i.e., the soil C-CO_2 that was not oxidized, or the C-CO_2 that was incorporated into the soil ultimately from the atmosphere – *would* have been lost eventually from the decomposition of soil organic matter. Thus, the net effect of elevated O is a reduction in the average lifetime of soil carbon. The fossil-fuel CO_2 emission-equivalent representation of this is simply the present value of the emission under elevated O_3 (relative to a value of 1.0 today) less the present value of the emission without elevated O_3 (relative to a value off 1.0 today).

The present value relative to a value of 1.0 today is simply the discount factor, equal to $1/(1+r)^L$, where r is the effective discount rate, discussed above, and L is the time of the effective emission. We will assume that the CO_2 emission from soils due to elevated ozone occurs today, which means that the discount factor is 1.0. Thus, what remains is to determine when the soil carbon would have been emitted had ozone not been elevated.

The results of Loya et al. (2003) indicate that major effect of ozone fumigation is an accelerated decomposition of organic matter with a relatively rapid turnover time, with a secondary effect being a reduction in the formation of soil carbon with a relatively long turnover time. I assume a mean effective lifetime of 15 years, which has an discount factor of around 0.65 (in our formulation, the discount factor does not change much between 10 and 30 years).

With these assumptions, the fossil-fuel CO_2-emission equivalent of the accelerated emission of soil carbon caused by ozone is 1.0 –0.654 = 0.35 grams of fossil-fuel equivalent. Hence, our estimate of 2-15 g-C of soil lost per g of O_3 added (from above) becomes about 0.7 to 5.0 g-C-fossil-fuel-equivalent emitted per g of g of O_3 added, or about 2.6 to 19 grams of CO_2 equivalent per g O_3. This value is quite large compared with the CEF for the direct radiative forcing of O_3, which is on the order of 0.50. In the LEM, we use values closer to the lower end of the range.

Plant NPP. Felzer et al. (2003) simulated the impact of ozone control policies on global NPP. Ozone control typically increased NPP by 5-6% by the end of the simulation period (the year 2100). Baseline global NPP was about 50 Pg-C / year. It is not possible from the information provided in their report to determine the reduction in the global ozone burden associated with their control scenarios. Therefore, I simply assume that the burden (370 Tg, as reported elsewhere here) is reduced by 25% at the end of the simulation period. I also assume that only half of the reduction in NPP is *in addition* to the reduction in soil carbon formation estimated in the previous section. (Recall that one off the effects of ozone on soil carbon, as estimated above, is a reduction in the formation of soil carbon, which is tantamount to a reduction in NPP.) Finally, I assume that the carbon in the lost NPP would have had a lifetime in the biosphere of 50 years before being re-emitted to the atmosphere. With these assumptions, the CO_2 equivalent impact of ozone on NPP is about 0.8, significantly less than that associated with changes in soil carbon.

CFC-12. The ozone-depleting gases, such as CFC-12, have a direct radiative-forcing effect on climate and an indirect cooling effect due to the depletion of stratospheric ozone. In its first assessment, the IPCC presented only the direct GWPs for the ozone-depleting gases, but in its second assessment the IPCC estimates included the indirect component (Daniel et al., 1995; presented in IPCC, 1996). Table D-3 shows the estimated net (direct + indirect) GWP for CFC-12. For most CFCs, the net GWPs are approximately 15 to 50% lower than the direct GWP values. Interestingly, Daniel et al. (in IPCC, 1996) also report that for certain compounds, such as halons, carbon tetrachloride, and methyl chloroform, indirect effects may overwhelm the direct radiative-forcing effects, producing net GWPs that are slightly negative to very negative. For example, the direct 20-year GWP of the halon, H-1301, is estimated at 6,100, while the net 20-year GWP is thought to be in the range of -14,100 to -97,600 (IPCC, 1996).

The estimation of the indirect cooling effect is complicated. Two key uncertainties are:

- the relative efficiencies of bromine and chlorine in removing O_3; and
- the magnitude of cooling in the lower stratosphere (due to uncertainties in the O_3 loss profile).

The uncertainty in the O_3 removal efficiency has less of an effect than does the uncertainty in the lower stratosphere cooling, which is estimated at ±30%. As shown in

Table D-3, the IPCC (2001) in its most recent (third) assessment estimates that the indirect effect of CFC-12 is about –19% of the direct effect.

The ozone-depleting gases might have indirect climatic effects not included in the GWP estimates discussed above. For example, altered levels of ultraviolet radiation, resulting from the loss of ozone, could affect OH levels in the troposphere. Particularly given the importance of OH to aerosol formation, the resulting effects could turn out to be important components of the overall impact of O_3 loss on climate change, and could therefore alter the GWP values for O_3-depleting gases (IPCC, 1996). Unexplored effects such as these suggest that the net GWPs for O_3-depleting gases may be lower than the current values.

Overall, the net GWPs for ozone-depleting gases are thought to be accurate only to ±50%, mainly on account of uncertainty in the indirect effects. Given this level of uncertainty, net GWP values for O_3-depleting gases are likely to be revised in the future as more becomes known about the complex chemistry of O_3 in the upper atmosphere, the relative strength of chlorine and bromine in removing O_3, and the actual magnitude of cooling that results from O_3 removal.

HFC-134a has a considerably lesser direct radiative-forcing effect than does CFC-12, but then does not have any compensating indirect global-cooling effect. As shown in Table D-3, the 20-year and 100-year GWPs for HFC-134a are about 40% and 15% of those for CFC-12 (IPCC, 1996). Consequently, the use of HFC-134a in place of CFC-12 greatly reduces but does not entirely eliminate the impact of automotive refrigerants on global climate.

Our assumptions. We follow the IPCC (2001), and assume that the indirect effect of CFC-12 is –19% of the direct effect. Thus, we estimate the total radiative forcing effect of CFC-12 by multiplying the direct effect, calculated using the formula presented above, by 0.81.

Temperature as a function of radiative forcing (function T[F(t),I(t)])

In this section, we estimate changes in temperature as a function of changes in radiative forcing.

The IPCC (2001, p. 354) defines a "climate sensitivity parameter," λ which is the ratio of the change in global mean surface temperature response ? T_S to the change in radiative forcing ?F:

$$?T_S / ?F = \lambda$$
$$?T_S = \lambda \cdot ?F$$

Allowing that in general the climate sensitivity parameter might be different for different gases, and different for indirect than for direct effects, and normalizing the parameter to a value of 1.0 for that for CO_2, we have:

$$T_i\left[F_i(t), I_i(t)\right] = l_{i,F} \cdot F_i + l_{i,I} \cdot I_i \qquad \text{eq. D.31}$$

where

$\lambda_{i,F}$ = the climate sensitivity parameter for direct radiative forcing of gas i (long-run run ["stabilized," or "equilibrium"] change in global mean surface temperature per unit change in direct radiative forcing, relative to the direct-forcing sensitivity parameter for CO_2)

$\lambda_{i,I}$ = the climate sensitivity parameter for indirect radiative forcing of gas i (long-run ["stabilized," or "equilibrium"] change in global mean surface temperature per unit change in indirect radiative forcing, relative to the direct-forcing sensitivity parameter for CO_2)

The climate sensitivity parameter depends on the spatial and temporal nature of the radiative forcing: it will be different for gases whose radiative forcing varies significantly in time and space (ozone and aerosols) compared with gases whose forcing is relatively uniform over the globe over a year (CO_2, CH_4, and N_2O). With regards to the well-mixed gases (CO_2, CH_4, and N_2O), the IPCC (2001) notes that "although the value of the parameter λ can vary from one model to another, within each model it is found to be remarkably constant for a wide range of radiative perturbations...The invariance of λ has made the radiative forcing concept appealing as a convenient measure to estimate global, annual mean surface temperature response..." (p. 354). Elsewhere, however, the IPCC (2001,396) emphasizes that the climate sensitivity parameter may be different for inhomogeneous gases such as ozone and aerosols, because the radiative forcing from these gases (unlike the forcing from the well-mixed gases) varies widely over time and space. Indeed, the IPCC (2001) makes it clear that at present it is an act of faith to assume that a unit of radiative forcing estimated for ozone and aerosols will have the same effects on global climate as will a unit of radiative forcing estimated for CO_2. Unfortunately, the IPCC does not indicate how the climate sensitivity parameter might be different for different gases.

However, Jacobson (2002, 2003) does provide model results that allow us to estimate a climate sensitivity parameter for some aerosols, albeit crudely. His global climate model reports the following changes in radiative forcing and long-run mean global temperature:

Gas	**Scenario**	**? T (^{o}K)**	**F (Wm^{-2})**	**Comment and source**
CO_2	With versus without current atmospheric mixing levels of anthropogenic CO_2 (based on 6 years of with and without runs).	0.90	1.6 (direct forcing)	New long-run temperature equilibrium. I assume that the total forcing is the same as the direct forcing. Temperature and forcing from Jacobson (2002).
CH_4	With versus without current	0.27	0.47	New long-run temperature equilibrium.

	atmospheric mixing levels of anthropogenic CH_4 (based on six years of with and without runs).		(direct forcing)	Temperature and forcing from Jacobson (2002).
SO_2	With versus without emissions of f.f. SO_2; "short term" cooling	-0.70	-0.35 (direct forcing) -1.23 (my est. of total forcing)	The year or period to which the temperature change applies is not specified in Jacobson (2002), but Jacobson (2003a) says that it is year 5 after 5 years of with and without simulations. I assume that the long-run temperature change is the same as the year-5 value. Jacobson (2002) reports –0.35 W^{-2} direct forcing. On the basis of the studies cited in the section above on radiative forcing, and consistent with my assumptions there, I assume that the total forcing is 3.5 times the direct forcing.
f.f. BC+OM	With versus without 5 years of emissions of f.f. BC+OM.	0.42 (my extra-polation)	0.53 (total forcing)	Jacobson (2002) reports a temperature change of –0.35 K at year 5 of the simulation, but his Figure 1 shows that the temperature had not stabilized by year 5, and suggests that had the with and without simulations continued for 5 to 10 more years, the mean global temperature difference would have been 0.40 to 0.45° K.[15] Total forcing from Jacobson (2002).
b.m. aerosol	With versus without 10 years of emissions of b.m. aerosols and gases.	-0.35	-0.90 (total forcing)	Temperature change at year 10, from Jacobson (2003). Figure 16 in Jacobson (2003) shows how the temperature changes over the first 10 years of the simulation, and the trend in this figure suggests that the temperature would not have changed much had the simulation continued for many more years. Total forcing based on Jacobson (2003); see discussion of radiative forcing, above.

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[15] This is confirmed by more recent simulations in which 10 years of with and without emissions of f.f. BC+OM resulted in a temperature change of 0.36 K in year 10, but with a significantly *lower* f.f. BC+OM emissions inventory than the one that generated the results in Figure 1 of Jacobson (2002) (Jacobson, 2003a).

With these, we can calculate values for λ, based on the long-run temperature change and the total forcing, and normalized to CO_2 = 1.0. To calculate sensitivities for BC and OM separately, given the data for f.f.BC+OM and b.m. aerosols, we first calculate ?T/Tg for each component, using the same method used to calculate W/g (e.g., eq. D.28, with ?T substituted for total forcing), and then calculate ?T/(Wm^{-2}) from ?T/Tg, TW/Tg (same as W/g) and 510 tera-m^2 earth surface area. The results of these calculations are:

	Absolute K/(w/m^2)	*Relative to CO_2*
f.f. BC+OM	0.79	1.41
CO_2	0.56	1.00
CH_4	0.57	1.02
SO_2	0.57	1.02
b.m. aerosols	0.39	0.69
BC separately	0.68 - 0.76	1.26 - 1.36
OM separately	0.52 - 0.70	0.93 - 1.25

Given this, the following shows our assumptions for λ for all GHGs:

gas	**l_F (direct)**	**l_i (indirect)**	**source/comments**
CO_2	reference	n.a.	Taken to be reference value.
CH_4	1.0	1.0	IPCC (2001). Assume indirect same as direct.
N_2O	1.0	n.a.	IPCC (2001). (See discussion above.)
CFC-12	1.0	1.0	Assume same as CO_2.
HFC-134a	1.0	1.0	Assume same as CO_2.
O_3	1.0	1.0	Working assumption in absence of data.
CO	n.a.	1.0	Working assumption in absence of data.
BC	1.3	1.3	Based on total forcing and long-run temperature changes summarized above, for f.f. BC+OM. Assume λ direct and λ indirect are the same as λ calculated based on total forcing.
OM	1.1	1.1	Based on total forcing and long-run temperature changes summarized above, for b.m. aerosols.

			Assume λ direct and λ indirect are the same as λ calculated based on total forcing.
SOA	1.1	1.1	Assume same as OM.
sulfate (SO_4)	1.0	1.0	Based on total forcing and long-run temperature changes summarized above. Assume λ direct and λ indirect are the same as λ calculated based on total forcing.
nitrate (NO_3)	1.0	1.0	Assume same as sulfate.
dust	1.1	1.1	Assume same as OM.

n.a. = not applicable.

Damages as a function of temperature (function D{T})

In this section we present our estimates of damages as a function of radiative forcing. We will use a simple exponential relationship:

$$D_i\{T_i[F_i(t), I_i(t)]\} = \{T_i[F_i(t), I_i(t)]\}^K \qquad \text{eq. D.32}$$

where K is the exponent that determines the nature of the relationship between temperature change and damages:

K = 0 --> damages are constant (independent of temperature)
K = 1 --> a factor change of X in temperature results in a factor change of X in damages (damages are linear with temperature)
K > 1 --> a factor change of X in temperature results in more than a factor change of X in damages
0 < K < 1 --> a factor change of X in temperature results in less than a factor change of X in damages.

What is a reasonable value of K? In their estimates of EDIs, Hammit et al. (1996) assume a range of values, including K=1 (linear), K=2 (as part of a quadratic), and K=3 (cubic). Kandlikar (1996) assumes linear and quadratic formulations. On the other hand, Reilly et al. (2001) assume only that damages are linear with radiative forcing (K=1). I think it is reasonable to assume that damages are very slightly nonlinear with temperature. I assume a value of K = 1.05.

The discount factor and the discount rate (function DF[r(t)])

The discount rate is used to calculate a discount factor that is applied to some future quantity (e.g., temperature change or damages) to generate the present worth of the quantity. In financial analyses the discount factor is calculated as $1/(1+r)^t$, where the time step is a year (i.e., *r* is an annual discount rate, and time *t* is in years). (As discussed in the section above on the theory of the discount rate, the discount factor also may be estimated as e^{-rt}.)

Earlier we presented evidence and theory that indicates that the discount rate ought to decline with time. Therefore, in our calculation of the CEFs, we calculate a year-by-year discount factor that incorporates a discount rate that declines as a function of time:

$$DF = \frac{1}{(1 + r(t))^t} \qquad \text{eq. D.33}$$

$$r(t) = r * \cdot t *^{1-K} \cdot t^{K-1} \qquad \text{eq. D.34}$$

where:

r* = reference annual discount rate, at time t*
t* = the reference time, at which the reference discount rate applies (years)
t = time (years)
K = exponential parameter

With this formulation, r(t) declines exponentially towards zero from r* as time exceeds t*, and rises exponentially to infinity from r* as time approaches zero. The asymptotic decline of the discount rate to zero as time approaches infinity is appropriate, but the asymptotic rise of the rate to infinity as time approaches zero is problematic, because there is no evidence or theory to support it. Indeed, it seems more reasonable to assume that there is some fixed, relatively high short-term rate that applies from time zero to some near-term future, and that the rate then starts its exponential decline beyond this short term. Formally, then, we will model the discount rate as follows:

for 0 = < t <= t* $\qquad$ r(t) = r*

for t > t* $\qquad$ $r(t) = r * \cdot t *^{1-K} \cdot t^{K-1}$

I assume that the discount rate is 7% for the first 5 years, and then declines gradually after that. (The lower the value the exponent K, the more gradual the decline.) Thus, in this analysis

r* = 0.07
t* = 5.0

K = 0.2

These parameters generate the following results:

time (yrs)	*1*	*2*	*5*	*10*	*20*	*50*	*100*	*200*	*500*	*1000*
DF	0.93	0.87	0.70	0.67	0.63	0.57	0.53	0.48	0.42	0.36
r(t)	7.0%	7.0%	7.0%	4.0%	2.3%	1.1%	0.64%	0.37%	0.18%	0.10%

With this formulation, the discount rate declines from 7% in years 1 to 5 to 4% in year 10, 1% in year 50, 0.5% in year 100, and close to 0% beyond year 500. This is consistent with the theoretical and empirical work discussed above, and seems reasonable. By comparison, in their estimates of EDIs, Reilly et al. (2001) assume a scenario in which the discount rates starts at 5% in year 2000 and declines 0.1%/year to 0% by the year 2050.

CO_2 EQUIVALENCY FACTORS (CEFS): OUR METHODS AND PARAMETER VALUES FOR GASES WITH INDIRECT EFFECTS ON CLIMATE

The “indirect” climate-change gases: CO, NMOCs, H_2, SO_x, and NO_X

The production, alteration, and destruction of “indirect” climate-change gases, such as CO, H_2, NMOCs, SO_x and NO_X, can affect the concentration, distribution, and lifetime of the “direct” climate-change gases CO_2, CH_4, N_2O, O_3, H_2O, and aerosols. However, the many interactions between species such as NO_X, CO, NMOCs, OH^-, O_3, and H_2O are incompletely understood and difficult to model. Most calculations of GWPs for these gases consider only a few such interactions. Although in its first assessment in 1990 the IPCC did estimate GWPs for NMOCs, CO, and NO_X (Shine, et al., 1990) , it disavowed them in 1992, stating that “most of the [IPCC’s earlier estimates of the] indirect GWPs...are likely to be in substantial error, and none of them can be recommended” (p. 15; brackets added). In its second assessment, the IPCC continued to maintain that “the calculation of indirect effects for a number of other gases (e.g. NOx, CO) is not currently possible because of inadequate characterization of many of the atmospheric processes involved” (p. 23) (IPCC, 1996). However, in its recent TAR, the IPCC (2001) discusses estimates of GWPs for CO and NO_X, and publishes a table (6.9) showing estimates of indirect GWPs for CO.

Table D-3 here shows several estimates of indirect GWPs for CO, NO_X, and NMOCs, including most of the studies discussed in the IPCC (2001), and a more recent study by Derwent et al. (2001), not included in the IPCC (2001) reviews. The recent study by Derwent et al. (2001) may be of particular importance because it is one of the first to use a 3-D rather than a 2-D model, and whatever resistance the IPCC (2001) still has to estimating an official GWP for CO appears to be due in part to the lack of 3-D studies available at the time.

In the following sections we make our own estimates of indirect CEFs for CO, H_2, NMOCs, SO_x and NO_X.

The CEF for carbon monoxide

Carbon monoxide (CO) can affect climate in at least four ways. First, it has a minor direct radiative forcing impact. Second, it reacts with OH and hence reduces the OH available to oxidize methane, thereby increasing the lifetime and concentration of methane. Third, it is a precursor of tropospheric ozone, which is a climate-change gas. Finally, within a few months or less, most or all of it ends up completely oxidized to CO_2 (Johnson and Derwent, 1996; Fuglestvedt et al., 1996; Daniel and Solomon, 1998). In essence, the reaction of CO with OH eventually leads to more O_3, more CH_4, and more CO_2.

All of these effects start with the main removal mechanism for CO:

$$CO + OH \dashrightarrow H + CO_2 \qquad \text{eq. D.35}$$

Reaction D.35 shows the consumption of OH (which affects the lifetime of CH_4) and the production of CO_2. The consumption of OH and the production of H also leads to the production of ozone, via an analog of the series of reactions D.61 to D.65 presented in the section “Ozone forming potential of organic compounds”(substitute equation D.35 for equation D.61, then substitute “H” for “R” in equations D.62 to D.65).

Our estimate of the CEF for CO accounts for all four impacts:

$$CEF_{CO} = CEF_{CO/RF} + CEF_{CO/CH4} + CEF_{CO/O3} + CEF_{CO/CO2} \qquad \text{eq. D.36}$$

$$CEF_{CO/CH4} = EE_{CO/CH4} \cdot CEF_{CH4} \qquad \text{eq. D.37}$$

$$CEF_{CO/O3} = MP_{CO/OH} \cdot MP_{O3/OH} \cdot \frac{MW_{O3}}{MW_{CO}} \cdot CEF_{O3} \qquad \text{eq. D.38}$$

$$CEF_{CO/CO2} = \left(MP_{CO/OH} + COOX \cdot \left(1 - MP_{CO/OH}\right)\right) \cdot \frac{MW_{CO2}}{MW_{CO}} \cdot DF\Big|^{t=L_{CO}} \qquad \text{eq. D.39}$$

where:

CEF_{CO} = the total CO_2-equivalency factor for CO

$CEF_{CO/RF}$ = the CEF of the minor direct radiative forcing impact of CO (estimated using the methods and parameter values documented in the major section on direct radiative forcing)

$CEF_{CO/CH4}$ = the CO_2-equivalency of the effect of CO on the concentration of CH_4

$CEF_{CO/O3}$ = the CO_2-equivalency of the effect of CO on the concentration of O_3

$CEF_{CO/CO2}$ = the CO_2-equivalency of the effect of CO oxidizing to CO_2

$EE_{CO/CH4}$ = the effect of a unit of CO emission on the mass concentration of CH_4 (grams CH_4 concentration change per gram of CO emission; see discussion below)

CEF_{CH4} = the CO_2-equivalency factor for CH_4 (estimated using the methods and parameter values given above)

$MP_{CO/OH}$ = moles of CO reacted with OH per mole of CO emitted (I assume 0.85; the IPCC [2001, p.257] says that 80 -90% of CO is removed from the atmosphere by reaction with OH; Daniel and Solomon [1998, p. 13252] say 80% [the remainder is removed by surface deposition and soils or is lost to the stratosphere])

$MP_{O3/OH}$ = moles of O_3 produced per mole of OH consumed by CO (discussed below).

MW_{O3} = the molecular mass of O_3 (48 g / mole)

MW_{CO} = the molecular mass of CO (28 g / mole)

MW_{CO2} = the molecular mass of CO_2 (44 g / mole)

CEF_{O3} = the CO_2-equivalency factor for the direct-forcing effect of O_3 (estimated using the methods and parameter values given above)

COOX = of the CO that is taken up by soils or lost to the stratosphere, the fraction that is oxidized to CO_2 (I assume 0.95; the remainder is assumed to be permanently removed as CO)

DF = the discount factor, a function of time and the discount rate (see the subsection on discounting), evaluated for $t = L_{CO}$

L_{CO} = the atmospheric lifetime of CO (see discussion in major section on direct radiative forcing)

The effect of CO on CH_4 ($EE_{CO/CH4}$). Derwent et al. (2001) have a table that gives estimates of $GWP_{CO/CH4}$, $GWP_{NOx/CH4}$, $GWP_{H2/CH4}$, and GWP_{CH4}. One can calculate EE values from these by dividing the reported estimates of $GWP_{CO/CH4}$, $GWP_{NOx/CH4}$, and $GWP_{H2/CH4}$ by the reported estimate for GWP_{CH4}. Derwent et al. (2001) also present a graph of the parameters $EE_{CO/CH4}$, $EE_{NOx/CH4}$, and $EE_{H2/CH4}$. The values of EE in this graph are the same as the values I back calculate from the Derwent et al. (2001) GWPs. The graph and the back-calculation indicate that $EE_{CO/CH4} = 0.039$. The simpler photochemical box model of Daniel and Solomon (1998), with full chemistry, produces a similar value of about 0.045 (based on their Figure 3 that shows 0.39 ppbv increase in CH_4 resulting from a 5.0 ppbv pulse of CO).

Note that I do not use the Derwent et al. (2001) estimate of $GWP_{CO/CH4}$ directly, but rather estimate the product of $EE_{CO/CH4}$ and CEF_{CH4}, where the parameter $EE_{CO/CH4}$

comes from Derwent et al. (2001) and CEF_{CH4} is my own estimate. This more disaggregated method ensures that the same value of CEF_{CH} is used throughout my analysis. (If I used the Derwent et al. estimate of $GWP_{CO/CH4}$ I would be using their estimate of GWP_{CH4}, which is slightly different from my estimate of CEF_{CH4} and hence inconsistent with the rest of my calculations.)

The effect of OH on O_3 ($MP_{O3/OH}$). The ozone chemistry described above and in reactions O1 to O5 suggests that the loss of a molecule of OH leads to the production of a molecule of O_3 whenever there is sufficient NO available. It appears that most of the time, sufficient NO is available: Daniel and Solomon (1998) cite another reference that indicates that "NO_x is believed to be large enough in most regions of the troposphere so that increases in HO_2 from a CO pulse should lead to more additional ozone production than ozone loss" (p. 13252). I will assume that sufficient NO is available 80% of the time, which means that $MR_{O3/OH} = 0.80$.

The effect of CO on CO_2 ($CEF_{CO/CO2}$). After no more than a few months, most CO oxidizes in the atmosphere to CO_2, which has a lifetime of decades. Presumably any CO taken up by soils also is oxidized to CO_2 relatively quickly. (CO lost to the stratosphere may not be oxidized to CO, but I assume that this fate is relatively rare [see parameter COOX in eq. D.39].) Hence the discount factor DF in eq. D.39 is close to 1.0 and the CO_2-equivalent effect of the oxidation of CO to CO_2 ($CEF_{CO/CO2}$) is just the ratio of the weight of CO_2 to CO, which is 1.57.

Comparison of our estimates with others. Table D-3 summarizes other estimates of GWPs and EDIs for CO. These estimates indicate that the GWP of the indirect effect of CO on O_3 and CH_4 -- $CEF_{CO/CH4} + CEF_{CO/O3}$ in eq. D36, computed as GWPs -- is in the range of 2-3. This seems roughly reasonable, since the ozone-forming potential of CO, at least as regards ozone at the surface, is quite low (Carter, 1998). The most recent and most sophisticated estimate, by Derwent et al. (2001), is that $GWP_{CO/CH4}$ is about 1.0 and $GWP_{CO/O3}$ is 0.6, for a 100-year time horizon. Our parameter values from the preceding major section result in a 100-year GWP for O_3 of 0.4, which when combined with the parameter values in this subsection results a value of 0.5 for $CEF_{CO/O3}$ on a 100-year GWP basis. This is close to the Derwent et al. (2001) value, and for the most part is calculated independently. (Our estimate of $CEF_{CO/CH4}$ on a 100-year GWP basis also is close to the Derwent et al. (2001) value, but it is not independent of it.)

Our assumptions for the MP parameters result in an emission of one mole of CO producing 0.85 x 0.80 = 0.68 moles of O_3, or 1 gram of CO emission producing 1.2 grams of O_3. The model of Joos et al. (2001) assumes 0.0011 Dobson Units of O_3 per year per Tg of CO emitted, or 0.012 Tg of O_3 per year Tg of CO emitted (given 10.9 Tg per DU [IPCC, 2001, p. 261]). Given an O_3 turnover time of 3-20 days (see above), 0.12 to 1.5 Tg O_3 would have to be formed over the course of a year (from the 1.0 Tg CO) in order to maintain 0.012 Tg O_3 in the atmosphere. Our estimate falls within this range.

The part of the CEF that accounts for oxidation to CO_2 is not uncertain. However, the effects on O_3 and CH_4 are quite uncertain, and this uncertainty has a nontrivial effects on the results. For example, if the sum $CEF_{CO/CH4} + CEF_{CO/O3}$ is changed from 2.5 to 0.0 or 5.0, CO_2 equivalent emissions change by about 5%, and the *percentage changes* relative to the petroleum baseline change by one or two percentage points.

The CEF for hydrogen

Although hydrogen is not a direct greenhouse gas, it can affect concentrations of CH_4 and O_3 via its reactions with OH (IPCC, 2001; Derwent et al., 2001). The IPCC (2001) remarks that while it does not consider H_2 emissions in its analysis, “in a possible fuel-cell economy, future emissions may need to be considered as a potential climate perturbation” (p. 256). Acknowledging that hydrogen is “a candidate future fuel,” Derwent et al. (2001) estimate a CEF for hydrogen, and conclude that “hydrogen emissions appear to generate an indirect radiative forcing in contradiction with the commonly held view that switching to hydrogen-based fuels will eliminate global warming” (p. 482).

The climatic chemistry effects of hydrogen are similar to the effects of CO. Hydrogen can react with and consume OH via the reaction: $H_2 + OH = H_2O + H$. Because OH also reacts with and removes CH_4, consumption of OH by H_2 reduces the OH sink for CH_4 and thereby increases the lifetime and radiative forcing of CH_4. The consumption of OH and the production of H also leads to the production of ozone, via the series of reactions O1 to O5 presented in the section “Ozone forming potential of organic compounds” (substitute “H” for “R” in those reactions).

Given this, a total CEF for hydrogen can be estimated simply as the sum of its effects on methane and ozone:

$$CEF_{H2} = CEF_{H2/CH4} + CEF_{H2/O3} \qquad \text{eq. D.40}$$

$$CEF_{H2/CH4} = EE_{H2/CH4} \cdot CEF_{CH4} \qquad \text{eq. D.41}$$

$$CEF_{H2/O3} = MP_{H2/OH} \cdot MP_{O3/OH} \cdot \frac{MW_{O3}}{MW_{H2}} \cdot CEF_{O3} \qquad \text{eq. D.42}$$

$CEF_{H2/CH4}$ = the CO_2-equivalency of the effect of H_2 on the concentration of CH_4

$CEF_{H2/O3}$ = the CO_2-equivalency of the effect of H_2 on the concentration of O_3

$EE_{H2/CH4}$ = the effect of a unit of H_2 emission on the mass concentration of CH_4 (grams CH_4 concentration change per gram of CO emission; Derwent et al. [2001] estimate that $EE_{H2/CH4} = 0.13$.)

CEF_{CH4} = the CO_2-equivalency factor for CH_4 (estimated using the methods and parameter values given above)

$MP_{H2/OH}$ = moles of H_2 reacted with OH per mole of H_2 emitted (0.30 – according to the IPCC [2001, p.256], 1/3 of H_2 is removed from the atmosphere by reaction with OH; the remainder is removed by soils)

$MP_{O3/OH}$ = moles of O_3 produced per mole of OH consumed by H_2 (discussed in the section on the CEF for CO).

MW_{O3} = the molecular mass of O_3 (48 g/mole)

MW_{H2} = the molecular mass of H_2 (2 g/mole)

CEF_{O3} = the CO_2-equivalency factor for the direct-forcing effect of O_3 (estimated using the methods and parameter values given above)

Derwent et al. (2001) estimate that $CEF_{H2/O3}$ = 2.4. The parameter values used here result in $CEF_{H2/O3}$ = 2.6.

In the LEM, the CEF for hydrogen is applied to leaks of hydrogen from vehicles, dispensing equipment, compressors, liquefiers, and pipelines.

The CEF for NMOCs

The CEF for NMOCs also is similar to that for CO, in that it consists of one straightforward component that accounts for oxidation of the carbon to CO_2, and another, uncertain component that accounts for the indirect effects of NMOCs on the production of O_3 and the lifetime of CH_4. In addition, anthropogenic NMOCs can form secondary organic aerosols (SOAs), which as indicated above are believed to have a slight cooling effect.

The relationship between emissions of NMOCs and the formation of ozone and the slowed destruction of methane depends in part on what is known as the “ozone creation potential” of the NMOC. The amount of CO_2 formed from oxidation depends of course on the carbon content of the fuel. Both of these parameters -- the ozone creation potential, and the carbon content -- vary from one NMOC compound to the next. For example, alcohols have a lower carbon content and a lower ozone-forming potential -- and hence a considerably lower CEF -- than have some hydrocarbon compounds. Because emissions of the various NMOCs can vary considerably from one fuelcycle to the next, an equitable analysis of the climate-change impact of fuelcycles will distinguish the carbon content and ozone forming-potential of different compounds or classes of compounds.

Consequently, we estimate CO_2-equivalent emissions separately for each NMOC source *s* (e.g., ethanol combustion), on the basis of the carbon content and the ozone-forming potential relative to that of gasoline (which is my arbitrary referent) of the NMOCs. The carbon content is multiplied by the ratio of the molecular mass of CO_2 to the molar mass of carbon (3.664), and the ozone-forming potential relative to that of

NMOCs from gasoline is multiplied by the CEF for the effect of gasoline NMOCs on O_3 and CH_4.

We also include the effect of SOAs, although it turns out to be relatively minor. The IPCC (2001, p. 3000) states that the formation of SOA from NMOCs is related to the aromatic content of the NMOC. We use gasoline-combustion NMOCs as a baseline, and then assume that the aromatic content of any NMOC relative to that of gasoline is the same as the compound's ozone forming potential relative to gasoline (ROFP). We use ROFP as a proxy for relative aromatic content because ROFPs are readily available, there is some correlation between ROFP and relative aromatic content, and the SOA CEF effect is too minor to warrant further specific elaboration.

Formally:

$$CEE_s = ME_s \cdot \left(\left(CEF_{GNMOC-O3/CH4} + SOAC \cdot CEF_{SOA} \right) \cdot ROFP_s + CF_s \cdot \frac{MW_{CO2}}{MW_C} + \right) \qquad \text{eq. D.43}$$

where:

CEE_S = CO_2-equivalent emissions of NMOCs from source *s* (e.g., NMOC emissions from the combustion of ethanol)
ME_S = mass emissions of NMOCs from source *s* (discussed in various sections of this report)
$CEF_{GNMOC-O3/CH4}$ = the CO_2-equivalency factor for changes in O_3 and CH_4 due to emissions of NMOC from the combustion of gasoline (discussed below)
SOAC = grams of secondary organic aerosol (SOA) formed per gram of gasoline NMOC emitted (0.01; the IPCC [2001, p. 300] estimates 0.6% for all fossil-fuel NMOCs, I assume gasoline NMOCs are slightly higher than this average)
CEF_{SOA} = the CO_2-equivalency factor for SOA; estimated using CEF data and formulae presented above
$ROFP_S$ = the ozone-formation potential (OFP) of NMOCs from source *s*, relative to the OFP of NMOCs from the combustion of conventional gasoline (Table D-4 discussed in a separate section below)
CF_S = the carbon weight fraction of NMOCs from source *s* (Table D-4)
MW_{CO2} = the molecular mass of CO_2 (44.01 g / mole)
MW_C = the molar mass of carbon (12.01 g / mole)

The CEF for changes in ozone and methane due to emissions of gasoline NMOC is in principle difficult to estimate, because tropospheric ozone chemistry, and the relationship between tropospheric ozone and global climate, are complex. We do not attempt an original estimate of this parameter ($CEF_{GNMOC-O3/CH4}$ in eq. D.43), but rather use the values from the estimates of Table D-3.

The GWP estimates by Martin and Michaelis (1992), Johnson and Derwent (1996), and perhaps Shine et al. (1996) (Table D-3) apparently do not include the effect of the final oxidation to CO_2, and so apparently are estimates of the GWP of the effect of NMOCs on O_3 and CH_4. Johnson and Derwent (1996) estimate this GWP for several different kinds of NMOCs. The average value is around 4.0, which is a bit lower than the estimates by Martin and Michaelis (1992) and Shine et al. (1990). This also appears to be the value of the kinds of NMOCs produced by gasoline combustion. I assume, then, that $CEF_{GNMOC-O3/CH4} = 4$. This value is consistent with our independent estimate of the *maximum* value of the ozone-forming component alone: multiplying the "maximum incremental ozone reactivity" of NMOCs from gasoline (3.8 g-O_3/g-NMOC-gasoline – see discussion in section "Ozone formation potential of organic compounds" by our estimated 100-year GWP for ozone (0.40, as discussed above) yields a value of 1.5.

Because the lifetime of the NMOCs is days, whereas the lifetime of the CO_2 formed by their final oxidation is decades, we may ignore the "lag" between the emission of an NMOC and the production of CO_2 (i.e., in eq. D.43, we do not include an NMOC counterpart to eq. D.39).

The method of D.43 estimates the direct CO_2 contribution of NMOC evaporative and combustion emissions exactly, and accounts reasonably well for the ozone-forming potential of different NMOC emissions. Thus, to a large extent, it appropriately differentiates the overall effect of low-carbon, low-ozone-forming NMOCs, such alcohols, from the effect of high-carbon, high-ozone-forming NMOCs, such as from some petroleum compounds. It still suffers, however, from the uncertainty in the modeling of ozone formation and global climate. In fact, as noted in Table D-3, the IPCC believes that the short atmospheric lifetime, unknown spatial distribution, and uncertain source/sink relationships of NMOCs make it impossible today to calculate the GWP (IPCC, 1996).

We believe, however, that the most recent estimates published in the peer-reviewed literature are serviceable at present -- especially because it appears that the uncertainty in the CEF for NMOC has only a minor affect on CO_2-equivalent emissions. Performing the same sort of sensitivity analysis as with CO, we find that if the parameter $CEF_{GNMOC-O3/CH4}$ is changed from 4.0 to 0.0 or 8.0, CO_2 equivalent emissions change by only 1%, and the *percentage changes* relative to the petroleum baseline change by less than a percentage point. Therefore, the true CEF for NMOC is not likely to be different enough from the value assumed here to have a significant effect on estimated CO_2-equivalent emissions -- because both mass emissions of NMOCs and the O_3 part of the GWP are relatively small.

The CEF for SO_2

The combustion of sulfurous fuels produces SO_2 and other sulfur compounds. In the atmosphere, some of the SO_2 is converted to sulfuric acid via gas-phase and aqueous-phase reactions. Some of this sulfate, in turn, is partly or fully neutralized by ammonia to particulate ammonium bisulfate or particulate ammonium sulfate. This particulate sulfate has direct and indirect effects on climate, as discussed above.

Our estimate of the CEF for SO_2 emissions embodies these three components: conversion of SO_2 to sulfate, neutralization of sulfate to particulate sulfate, and the climate-change impact of particulate sulfate:

$$CEF_{SOx} = FSO4 \cdot FPMS \cdot \frac{MW_{PMS}}{MW_{SO2}} \cdot CEF_{PMS} \qquad \text{eq. D.44}$$

where:

CEF_{SOx} = the CO_2-equivalency factor for SO_x (as SO_2) emissions, on a mass basis
FSO4 = of emitted SO_2, the fraction that is oxidized to sulfate (SO_4) (0.25; Delucchi and McCubbin, 1996)
FPMS = of the sulfate formed, the fraction that is neutralized by ammonia to particulate sulfate ($[NH_4]_2SO_4$) (assumed to be 1.0; Delucchi and McCubbin, 1996)
MW_{PMS} = the mass of sulfate (SO_4, 96 g / mole)
MW_{SO2} = the molecular mass of SO_2 (64 g / mole)
CEF_{PMS} = the CO_2-equivalency factor for particulate sulfate, on a mass basis (Table D-3; see discussion elsewhere in this appendix)

Note that the CEF is calculated on the basis of the ratio of the weight of SO_4 to the weight of SO_2, rather than on the basis of the weight of the particulate sulfate ($2NH_4$-SO_4, with a formula mass of 132 g / mole). This is because the radiative forcing estimates discussed above are with respect to SO_4; that is, they are in units of W $(g\text{-}SO_4)^{-1}$. However, I assume that sulfate has its effect only as part of an ammonium sulfate molecule – hence the factor, in the formula above, that accounts for the fraction of sulfate that is neutralized by ammonia to particulate sulfate.

The CEF for NO_x emissions

Anthropogenic emissions of nitrogen oxides to the atmosphere can disturb the complex global nitrogen cycle and ultimately have a wide range of environmental impacts, including eutrophication of lakes and coastal regions, fertilization of terrestrial ecosystems, acidification of soils and water bodies, changes in biodiversity, respiratory disease in humans, ozone damages to crops, and changes to global climate (Mosier et al, 2002; Jenkinson, 2001; Erisman et al., 1998; Galloway, 1998; Vitousek et al., 1997; Smith et al., 1999; Galloway et al., 1995). Nitrogen oxide emissions contribute to climate change through complex physical and chemical pathways that affect the concentration of ozone, methane, nitrous oxide, and carbon dioxide:

i) it participates in a series of atmospheric chemical reactions that involve CO, NMOCs, H_2O, OH^-, O_2, and other species, and which affect the concentration of tropospheric ozone, a "direct" climate-change gas;

ii) in the atmospheric chemistry mentioned in i), it affects the concentration of the hydroxyl radical, OH, which oxidizes methane and thereby affects the lifetime of methane;

iii) as nitrate, it deposits onto soils and oceans and then denitrifies or nitrifies into N_2O (a strong, long-lived direct climate-change gas) and NO (which oxidizes back to the indirect GHG NO_2 that was the source of the deposited N in the first place), and also affects soil emissions of CH_4;

iv) as nitrate, it fertilizes terrestrial and marine ecosystems and thereby stimulates plant growth and carbon sequestration in nitrogen-limited ecosystems;

v) it forms particulate nitrates, which as aerosols probably have a net negative radiative forcing (see the section on aerosols)

vi) as deposited nitrate, it can increase acidity and harm plants and thereby reduce CO_2 sequestration;

Our estimate of the CEF combines the all of the components listed above:

$$CEF_{NOx} = \frac{\begin{array}{c} CEF_{NOx/O3} + CEF_{NOx/CH4} + CEF_{ND/CH4} + CEF_{ND/N2O} \\ + CEF_{ND/C} + CEF_{NOx/PM} + CEF_{NOx/DN} \end{array}}{1 - NORF} \qquad \text{eq. D.45}$$

$$NORF = \sum_{E} [NO2_{E} / NO2_{air}] \cdot [N(NO)_{P} / ND_{E}] \cdot [N(NO)_{AIR} / N(NO)_{P}] \qquad \text{eq. D.46}$$

$$[NO2_{E} / NO2_{air}] = [NO2D_{E} / NO2_{air}] + [NO2L_{E} / NO2_{air}]$$

$$[N(NO)_{P} / ND_{E}] = [N(NO)_{P} / ND_{E,NL}] \cdot NLF_{E,TY} + [N(NO)_{P} / ND_{E,NNL}] \cdot \left(1 - NLF_{E,TY}\right) \qquad \text{eq. D.47}$$

$$NLF_{E,TY} = \frac{1}{1 - e^{b(E)\cdot(TY-1990)}} \cdot \frac{1 - NLF_{E,1990}}{NLF_{E,1990}} \qquad \text{eq. D.48}$$

$$[NO2L_E\ /NO2_{air}] = \sum_{OE} TR_{OE\rightarrow E} \cdot [NO2D_{OE}\ /\ NO2_{air}] \qquad \text{eq. D.49}$$

$$TR_{OE\rightarrow E} = TR_{OE,NL\rightarrow E} \cdot NLF_{OE,TY} + TR_{OE,NNL\rightarrow E} \cdot \left(1 - NLF_{OE,TY}\right) \qquad \text{eq. D.50}$$

$$CEF_{NOx/CH4} = EE_{NOx/CH4} \cdot CEF_{CH4} \qquad \text{eq. D.51}$$

$$CEF_{ND/CH4} = CEF_{CH4} \cdot \sum_{E} NO2_E\ /\ NO2_{air} \cdot \frac{MW_N}{MW_{NO2}} \cdot gCH4\ /gND_E \qquad \text{eq. D.52}$$

$$CEF_{ND/N2O} = CEF_{N2O} \cdot \sum_{E} NO2_E\ /NO2_{air} \cdot \frac{MW_N}{MW_{NO2}} \cdot N(N2O)_{air}\ /\ ND_E \cdot \frac{MW_{N20}}{MW_{N2}} \qquad \text{eq. D.53}$$

$$CEF_{ND/C} = \sum_{S}\sum_{E} NO2_E\ /\ NO2_{air} \cdot NT_{S,E} \cdot NLF_{E,TY} \cdot CNR_{S,E} \cdot \left(1 - ACF_E\right) \cdot \left(1 - DF\right) \cdot \frac{MW_N}{MW_{NO2}} \cdot \frac{MW_{CO2}}{MW_C}$$

eq. D.54

$$DF = f\left[r(t)\right]^{t=ECL_{S,E}} \qquad \text{eq.D.55}$$

$$ECL_{S,E} = LC_{S,E} \cdot NCF_{S,E} \qquad \text{eq.D.56}$$

$$CEF_{NOx/PM} = CEF_{PMN} \cdot FNO3 \cdot FPMN \cdot \frac{MW_{PMN}}{MW_{NO2}} \qquad \text{eq. D.57}$$

where:

CEF_{NOx} = the CO_2-equivalency factor for NO_x (as NO_2) emissions, on a mass basis

$CEF_{NOx/O3}$ = the CO_2 equivalency of the indirect effect of NO_x on tropospheric O_3 (discussed below)

$CEF_{NOx/CH4}$ = the CO_2 equivalency of the indirect effect of NO_x on the lifetime of atmospheric CH_4

$CEF_{ND/CH4}$ = the CO_2 equivalency of the indirect effect of N deposition on CH_4 emissions from soils

$CEF_{ND/N2O}$ = the CO_2 equivalency of the indirect effect of N deposition on N_2O emissions from soils

$CEF_{ND/C}$ = the CO_2 equivalency of the indirect effect of N deposition on carbon sequestration

$CEF_{NOx/PM}$ = the CO_2 equivalency of the indirect effect of the conversion of NO_X to particulate nitrate

$CEF_{NOx/DN}$ = the CO_2 equivalency of the effect of deposited acid nitrate on carbon sequestration by plants (discussed below)

NORF = the fraction of deposited nitrogen that is returned to the air as NO or NH_3

$NO2_E / NO2_{AIR}$ = the fraction of ambient NO_2 (from fossil-fuel combustion) that ends up in ecosystem E, either by direct deposition or transfer of direct deposition from another ecosystem

$NO2D_E / NO2_{AIR}$ = the fraction of ambient NO_2 (from fossil-fuel combustion) that is directly deposited onto and taken up by ecosystem E (discussed below)

$NO2L_E / NO2_{AIR}$ = the fraction of ambient NO_2 (from fossil-fuel combustion) that is deposited onto ecosystems adjacent to ecosystem E and leached into ecosystem E

$N(NO)_P / ND_E$ = the fraction of the deposited N in ecosystem E that is converted to N in NO or NH_3

$N(NO)_{AIR} / N(NO)_P$ = the fraction of the NO-N produced that escapes past the canopy to the atmosphere (the remainder is assumed to be captured by the vegetative canopy); my assumptions are:

Tropical Forest	*Boreal Forest*	*Grassland*	*Agriculture*	*Other*
0.50	0.60	0.75	0.75	1.00
Hall & Matson (1999); Davidson and Kingerlee (1997)		my assumptions		

$N(NO)_P / ND_{E,NL}$ = the fraction of the deposited N in nitrogen-limited ecosystem E that is converted to N in NO or NH_3 (Appendix C)

$NLF_{,E,TY}$ = the fraction of ecosystem type E that is nitrogen limited in the target year TY

$N(NO)_P / ND_{E,NNL}$ = the fraction of the deposited N in non-nitrogen-limited ecosystem E that is converted to N in NO or NH_3 (Appendix C)

b(E) = exponent that determines rate of change of the N-limited fraction of ecosystem E (A positive value means that the N-limited fraction decreases with time; a higher value means that it decreases at a greater rate. I assume a value of 0.02 for all ecosystems, which results in a relatively modest decline in the N-limited fraction with time)

TY = target year of analysis (specified by model user)

1990 = base year for estimate of fraction of ecosystems nitrogen limited

$NLF_{,E,1990}$ = the fraction of ecosystem type E that is nitrogen limited in the base year of 1990 (Appendix C)

$TR_{OE \rightarrow E}$ = of the amount of deposition onto originating ecosystem OE, the fraction that is transferred to (leached into) ecosystem E

$NO2D_{OE} / NO2_{AIR}$ = the fraction of ambient NO_2 (from fossil-fuel combustion) that is directly deposited onto ecosystem (same as $NO2D_E$)

$TR_{OE,NL \rightarrow E}$ = of the amount of deposition onto originating nitrogen-limited ecosystem OE, the fraction that is transferred to (leached into) ecosystem E (discussed below)

$TR_{OE,NNL \rightarrow E}$ = of the amount of deposition onto originating non-nitrogen-limited ecosystem OE, the fraction that is transferred to (leached into) ecosystem E (discussed below)

$NLF_{,OE,TY}$ = the same as $NLF_{,E,TY}$

$EE_{NOx/CH4}$ = the effect of a unit of NO_X emission on the mass concentration of CH_4 (grams CH_4 concentration change per gram of NOx emission) (discussed below)

CEF_{CH4} = the CO_2-equivalency factor for CH_4 on a mass basis (discussed elsewhere in this appendix; see Table D-3)

$gCH4/gND_E$ = grams of CH_4 emitted per gram of nitrogen deposited in ecosystem E (discussed briefly below; see Appendix C)

MW_N = the molecular mass of nitrogen in NO_2 (14.01 g / mole)

MW_{NO2} = the molecular mass of NO_2 (46.01 g / mole)

CEF_{N2O} = the CO_2-equivalency factor for N_2O on a mass basis (discussed elsewhere in this appendix; see Table D-3)

$N(N2O)_{AIR} / ND_E$ = the fraction of the deposited N in ecosystem E that is re-emitted to the atmosphere as nitrogen in N_2O (discussed briefly below; see Appendix C)

MW_{N2O} = the molecular mass of N_2O (44.01 g / mole)

MW_{N2} = the molecular mass of N in N_2O (28.01 g / mole)

$NT_{S,E}$ = of the amount of N deposited (directly or indirectly) into ecosystem E, the fraction that is taken up by sink S (discussed below)

$CNR_{S,E}$ = the carbon:nitrogen sequestration ratio of sink S (discussed below)

ACF_E = the "accelerated carbon fixation" factor: the fraction of carbon fixed by added N that would have been fixed eventually anyway, but at a later date (discussed below)

DF = the discount factor, a function of the discount rate (which is a function of time – see the subsection on discounting, this appendix), evaluated for t = $ECL_{S,E}$

MW_{CO2} = the molecular mass of CO_2 (44.01 g / mole)

MW_C = the molar mass of C (12.01 g / mole)

$ECL_{S,E}$ = the effective lifetime carbon in sink S in ecosystem E – the amount of time that *any* C in sink S remains associated with an additional unit of N in ecosystem E (years)

r(t) = the discount rate as a function of time (see subsection on discounting, this appendix)

$LC_{S,E}$ = the "once-through" lifetime of carbon in sink S in ecosystem E (years) (discussed below)

$NCF_{S,E}$ = the nitrogen cycling factor: the number of C lifetimes that N is recycled within ecosystem E before the N goes to a fate in which it is no longer able to sequester C (discussed below)

CEF_{PMN} = the CO_2-equivalency factor for nitrate PM on a mass basis (Table D-3; see discussion elsewhere in this appendix)

FNO3 = of emitted NO_2, the fraction that is oxidized to nitrate (NO_3) (0.06; Delucchi and McCubbin, 1996)

FPMN = of the nitrate formed, the fraction that is neutralized by ammonia to particulate nitrate (NH_4NO_3) (I assume 0.80, on the basis of Delucchi and McCubbin, 1996)

MW_{PMN} = the mass of nitrate (NO_3, 62 g/mole)

MW_{NO2} = the molecular mass of NO_2 (46 g/mole)

subscript E or OE = ecosystems: oceans, freshwater, forest biomass, forest soils, agricultural and pastoral biomass, agricultural and pastoral soils, other biomass, other soils, other (stratosphere, man-made)

subscript S = biome sinks for N: soil, woody biomass, or non-woody biomass (e.g., leaves)

The factor NORF, expanded in eq. D.46, accounts for the impact of recycling NO_2: some of the N deposited from atmospheric NO_2 (or NH_3) is re-emitted to the atmosphere as NO (which eventually becomes NO_2) or NH_3, and hence is available again for all of the reactions and indirect effects considered here. The factor 1/(1-NORF) is derived as follows: Define CEF_{NOX}' to be equal to CEF_{NOX} without the factor 1/(1-NORF). To account for the first round of NO returned to the atmosphere, we multiply CEF_{NOX}' by NORF, and add it to CEF_{NOX}'. To account for the second round, we multiply the first round quantity, $CEF_{NOX}' \cdot NORF$, by NORF again, and add it. We do this repeatedly, and we have

$$CEF_{NOX} = CEF_{NOX}' + CEF_{NOX}' \cdot NORF + CEF_{NOX}' \cdot NORF^2 + CEF_{NOX}' \cdot NORF^3 + \ldots \qquad \text{eq. D.58}$$

which simplifies to:

$$CEF_{NOX} = CEF_{NOX}' \ (1 + NORF + NORF^2 + NORF^3 + \ldots). \qquad \text{eq. D.59}$$

But the series in parentheses is equal to 1 / (1-NORF), so we end up with

$$CEF_{NOX} = CEF_{NOX} / (1\text{-}NORF). \qquad \text{eq. D.60}$$

Nitrogen deposition by type of ecosystem (eq. D.46). We first distinguish land, freshwater, coastal, and marine ecosystems, and then further disaggregate land systems by biome type. (We also will distinguish later between soil and biomass "sinks" for N.)

Data and analyses reviewed in the IPCC (1996) indicate that 62.5% of N in emitted NO_X eventually deposits on terrestrial ecosystems (50% on the ground, 10% on plants, and 2.5% on freshwater) and 37.5% deposits on marine ecosystems. Mackenzie et al. (2002) cite an estimate that in 1860, 60% of the N in NO_x emitted from fossil-fuel combustion was "returned to the terrestrial realm while the rest was deposited onto coastal marine surface waters" (p. 16). Data in Furiness et al. (1998) imply considerable long-range transport of atmospheric N. A detailed model of emissions and deposition of N in the United States found that 53% of the emitted N was deposited onto non-urban areas of the U. S. (Holland et al., 1999).

Galloway et al. (1995) perform an accounting of the fate of anthropogenic N globally. Their analysis indicates that about 70% of anthropogenic NO_Y emissions and about 80% of anthropogenic NH_X emissions are returned to the continents via deposition, with the balance going to the oceans.

Holland et al. (1997) report results from five three-dimensional models of N emissions, transport, and deposition. These models indicated the following distribution of deposited N by ecosystems globally:

	Fossil-fuel NO_X *(5 models)*	*All NO_x* *(5 models)*	*All NH_3* *(1 model)*
Ocean/land split			
Oceans	45%	30 – 63%	44%
Land	55%	37 – 70%	56%
Breakdown of land			
forests	9%	12 – 15%	13%
cropland	52%	43 – 51%	49%
ice or desert	8%	4 – 8%	5%
other (unforested)	31%	31 – 36%	32%

("All NO_X" comprises fossil-fuel, biomass-burning, lightning, and soil sources.) These figures from Holland et al. (1997) are remarkably consistent across models and sources of N considered.

Galloway et al. (1996, Figure 3, p.12) show deposition of N (as NO_Y and NH_X) across the Northern and Western hemisphere, at 45^O and 5^O north latitude and various longitudes from 175 W to 25 E. Their estimates indicate the following distribution of N deposition:

at 45^O N latitude: 3% North Pacific, 39% North America, 5% North Atlantic, and 53% Europe.

at 5^O N latitude: 22% North Pacific, 16% Central and South America, 13% North Atlantic, and 50% Africa.

It is not clear however how much of the deposition to North America and Europe falls on coastal waters rather than land. Elsewhere, Galloway et al. (1996) present a summary of N fluxes for the watershed, estuary, shelf, and open ocean of the North Atlantic, and show that deposition to estuaries and shelves exceeds that to watersheds.

On the basis of the preceding data summaries I assume the following initial distribution of N deposition globally:

Terrestrial	*Lakes*	*Rivers/Coasts*	*Marine*
0.600	0.030	0.150	0.220

Next we break down the "terrestrial" category by ecosystem (agricultural, forest, and other soil and plants). The IPCC's (2000) special report on land use estimates the distribution of land use types over the earth:

- tropical, temperate, boreal forests, 28%
- tropical savannas, 15%
- croplands and grasslands, 19%
- desert, semi-desert, and tundra, 36%
- wetlands, 2%

However, the amount of deposition a region receives is a function of its proximity to major NO_x and NH_3 sources, and it appears that most such sources are far from deserts, semi-deserts, tundra, and tropical savannas, and relatively close to forests, grasslands, and croplands. In their analysis of the fate of N in the North Atlantic region (including the eastern U. S., and western Europe), Howarth et al. (1996) estimate that 70% of the land in the region is forested, and hence that 70% of all terrestrial deposition in the region occurs on forests.

Smil (1999) uses a variety of data sources to calculate that the world's agricultural land receives at least 1/3 of all the reactive N that is deposited on land, and says that the large share is due to agricultural regions being close to non-agricultural sources of NO_X emissions, particularly in Europe, eastern North America, and east Asia (p. 649).

Our assumptions. Assuming that NO_X emission sources are closest to croplands, and furthest from deserts and tundra, I assume the following breakdown of the "terrestrial" deposition category:

Tropical Forest	*Temp. Forest*	*Grassland*	*Agriculture*	*Arid*	*Urban*
0.100	0.200	0.200	0.300	0.100	0.100

In Appendix B, I use these estimates as a basis for estimating country-specific distributions.

Nitrogen transfers between ecosystems (eq. D.49 and D.50). As indicated by eqs. D.49 and D.50, the LEM crudely tracks transfers of atmospherically deposited N between ecosystems. In this subsection we review estimates pertinent to estimating the transfer coefficients in eq. D.49 and D.50. (See also the discussion of leaching of fertilizer N in Appendix C.)

In the LEM, nitrogen initially is input to an ecosystem by direct atmospheric deposition. This directly input N then can be transferred to other ecosystems by leaching into groundwater, rivers, coastal flows, and erosion. Eventually this N reaches its final "sinks," which essentially remove it from the N cycle and thus end the chain of climate-relevant impacts. Galloway et al. (1995) identify the following sinks:

- storage in terrestrial soils and sediments;
- active N cycling in subcompartments of natural ecosystems;

- storage in marine sediments (this is relatively minor)[16];
- storage in groundwater (effecting a permanent increase in N);
- accumulation in the deep ocean as NO_3 or dissolved N_2; .
- conversion to N_2 and emission to the atmosphere (Galloway et al. [1996] write that this can be viewed as a permanent sink, because the residence time of atmospheric N_2 is on the order of 1 million years [p. 14]);
- conversion to N_2O and emission to the atmosphere (N_2O has a lifetime of about 120 years and is relatively inert);

Galloway (1998) estimates that in 1990 about 140 Tg of reactive N (mainly in fertilizers or from fossil-fuel combustion) was created as a consequence of human action. He then outlines the fate of this reactive N. First, he cites one estimate that 16 Tg/yr dissolved inorganic N ends up being discharged from rivers to the oceans, and another estimate 20 Tg/yr of dissolved N and 20 Tg/yr of particulate N from anthropogenic sources are transported by rivers to coastal areas. Reactive nitrogen discharged to coastal areas may be stored in sediments, denitrified to unreactive N_2O or N_2 and emitted to the atmosphere, or transported to the open ocean. He speculates that most riverine N is denitrified in coastal regions.

Howarth et al. (1996) note that very little is known about the fate of organic N inputs to estuaries and coastal, and state that “a critical question is to determine how much is biologically available and how much is simply buried”(p. 92).

Nixon et al. (1996) estimate that in the North Atlantic region about 29% of the total N discharged in rivers is buried in sediment in deltas or continental shelves.

Galloway (1998) estimates that about 80 Tg/yr N remains on the continents. This N can be stored in long-term reservoirs (e.g., buried organic matter, groundwater, or biomass) or denitrified.

Howarth et al. (1996) indicate that storage of N in groundwater or wetlands is relatively minor. They also estimate that N storage in rivers is only 5-20% of inputs, because of relatively low residence times. Retention in lakes is much higher, because of longer residence times.

My assumptions. On the basis of the information discussed above and the studies summarized in Appendix C on leaching of fertilizer N, I assume generally that most non-N-limited ecosystems transfer 25% of their deposition input to rivers, 10% to marine/coastal areas, and 3% lakes. I also assume that non-N-limited grasslands and agriculture areas transfer 5% of their deposition input to each other or to temperate or tropical forests. I assume that N transfers out of N-limited ecosystems are much lower than N transfers out of non-N-limited systems, because N-limited systems retain and

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[16] Galloway et al. (1995) distinguish three sinks of N inputs to coastal oceans: storage in coastal sediments, denitrification to N_2, or export to open ocean, which is considered to be relatively minor. They also identify three sinks of N inputs to the open oceans: accumulation in the deep ocean as nitrate or N_2, denitrification to and emission to the atmosphere of N_2, and deposition in sediments (estimated to be less than 10%).

use a greater fraction of deposited N (Appendix C). Formally, I assume that N-limited systems transfer out one-fifth as much N as do non-N-limited systems.

Indirect effect on atmospheric zone (O_3) and methane (CH_4). In 1990, the IPCC estimated GWPs for the indirect effect of NO_X on ozone production. These GWPs were quite high, and gave NO_X a surprisingly important role in the global warming impact of most fuel cycles (DeLuchi, 1991). However, shortly after the original 1990 GWPs were published, Johnson et al. (1992) reported that the model used to calculate the ozone-GWP of NO_X contained an error that overestimated the GWPs by a factor of five. (Table D-3 here shows the corrected values for NO_X.)

Martin Michaelis re-estimated the effect of ground-level effects of NO_X, accounting for the impact of NO_X on CH_4, as well as on ozone. Their estimate for a 50-year time horizon is relatively low (Table D-3); converted to a 100-year basis, their estimate would be even lower. Derwent et al. (2001) and Johnson and Derwent (1996) estimate separate GWPs for the Southern Hemisphere and the Northern Hemisphere (Table D-3); in the work of Johnson and Derwent (1996), the GWP for the Southern Hemisphere actually is negative. Derwent et al. (2001) and Fuglestvedt et al. (1996) also estimate GWPs for NOx from aircraft; these are quite large, on account of the strong effect of high-level NOx emissions on ozone.

The estimates of (Table D-3) show that climatic impact of NOx emissions depends greatly on the location of the emissions: northern vs. southern hemisphere, ground-level versus high-altitude emissions. The LEM distinguishes emissions in the southern hemisphere from emissions in the northern hemisphere. I use the parameter values of Derwent et al. (2001): $CEF_{NOx/O3} = 13$ (NH) or 39 (SH) and $EE_{NOx/CH4} = -0.33$ (NH) or –0.93 (SH).

Indirect effect of N deposition on CH_4 (eq. D.52). Deposition of N, like the addition of fertilizer N, accelerates emissions of NO, N_2O, NH_3, and CH_4 (IPCC, 2001; Appendix C here). We consider all of these effects. Our assumptions regarding the amount of methane emitted per gram N deposition are discussed in Appendix C. (The LEM formally distinguishes leaching from deposition input, and N-limited from non-N-limited ecosystems, but as indicated in Appendix C the available data indicate that the emission factors are the same for all circumstances.) This impact turns out to be relatively minor.

Indirect effect of N deposition on N_2O (eq. D.53). Ambient NO_X eventually is deposited at the surface of the earth as nitrate (e.g., nitric acid, HNO_3, or ammonium nitrate, NH_4NO_3) (EPA, 1996c; Erisman et al., 1998) . This deposited nitrate, like artificial nitrogen fertilizer, can nitrify or denitrify to produce N_2O. The LEM has different N2O / N emission factors not only for different ecosystems, but for leached versus deposited N, and for N-limited versus non-N-limited ecosystems. The emission factor shown in eq. D.53 thus is the composite of four basic factors: deposition / N-limited, deposition / non-N-limited, leaching / N-limited, and leaching / non-N-limited, by type of ecosystem. Each of these four is weighted by an appropriate shares (e.g., deposition N fraction of total N input and N-limited fraction of all ecosystems) to obtain the composite factor. Appendix C reviews studies pertinent to the estimation of the four basic emission factors and provides base-case estimates.

Indirect effect of N deposition on C sequestration (eq.D.54). Nitrogen from atmospheric NO_X deposits on and is taken up by terrestrial and marine ecosystems and thereby can enhance primary productivity and carbon storage (Schindler and Bayley, 1993; Townsend et al., 1996; IPCC, 1996, 2000). Although added nitrogen stimulates growth only in nitrogen-limited ecosystems, and there is a threshold beyond which additional nitrogen reduces primary productivity and cause tree death and carbon loss (IPCC, 1996; Asner et al., 2001; Emmett, 1999)[17], "it is clear that rates of plant production and of the accumulation of biomass in whole ecosystems are limited by N supply over much of Earth's surface...particularly in temperate and boreal regions, and equally clear that human activity has increased N deposition substantially over much of this area" (Vitousek et al., 1997, p. 740).

As shown in eq. D.50, the amount of carbon sequestered is a function of a number of factors: the amount of nitrogen deposited directly or indirectly to the ecosystem, whether or not the ecosystem is N-limited (systems that are not N limited generally do not take up the excess deposited N), the fate of the nitrogen within the ecosystem (soil, woody biomass, or non-woody biomass), the C:N ratio of the components of the ecosystem that sequester C, and the length of time that *any* carbon remains associated with (sequestered by) deposited N. In this subsection, we discuss the fate of N within ecosystems, the C:N ratios, and the effective lifetime that C remains sequestered by additional N within the ecosystem.

The fate of N within ecosystems. For each terrestrial ecosystem type, we distinguish between N that ends up in woody biomass, non-woody (n.w.) biomass, and soil, because each of these has radically different C:N ratios (see discussion elsewhere). Regarding first the split between biomass and soil: Asner et al. (2002), Nadelhoffer et al. (1999), Emmett (1999), the IPCC (1996), Fenn et al. (1998), and Galloway et al. (1995) all point out that most deposited N remains in the soil. The IPCC (1996) reports that "recent studies of the fate of nitrogen added to forest ecosystems suggest that between 70 and 90% ends up in the soil...In addition, much atmospheric nitrogen is in reality deposited on grasslands and agricultural lands where storage occurs in soils with low average C:N ratios" (p. 456). Nadelhoffer et al. (1999) used N-tracer studies to determine the fate of N deposited in forest ecosystems, and found that only 20% of the added N went into tree biomass; the bulk of the added N was incorporated into soil or

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[17] Emmett (1999) discuss several studies in which added N had either a neutral or a negative effect on tree growth. They suggest that initially N deposition results mainly in N accumulation in soils (as a result of suppression of decomposition, abiotic N fixation, and N assimilation by certain fungi), but that over the long run the accumulation of N in the soil results in increasing nitrification, which in turn is associated with a decrease in the retention efficiency of deposited N and an increase in leaching of deposited N. They state that "this reduced efficiency ..may result in a decline of tree growth on some acidification-sensitive soils" (p. 72).

lost by leaching or gasification (Nadelhoffer et al. [1999] assumed 10% was lost off site). Asner et al. (2002) confirm this, citing Nadelhoffer et al. (1999) and several other long-term fertilization and tracer studies in support of the proposition that in eastern U. S. and European forests most added N does not enter vegetation and hence does not stimulate C sequestration. Emmet (1999) cites Nadelhoffer et al. and two other studies that show that less than 20% of N input to forest is taken up by trees (one of the two studies estimated a figure of 15% for Finnish forests). Galloway et al. (1995) also cite studies that show that added N is retained primarily in the soil organic pool (p. 245). Finally, Fenn et al. (1998) summarize a number of tracer studies of the fate of N added to forest ecosystems, and find that 4-33% of N ends up in vegetation, 2-33% leaches, and 19-86% ends up in soils (p.712). The vegetation share of vegetation+plus soil ranged from 8% to 44%, with an average of 23%, which indicates a 77%/23% soil/vegetation split, consistent with the studies cited above.

Turning now to woody vs. non-woody biomass, Townsend et al. (1996) assume generally a 50-50 split between woody and non-woody biomass. However, the tracer studies of Nadelhoffer et al. (1999), mentioned above, indicate that most of the added N goes to non-woody biomass: in their study of temperate forests, 17% of the added N went to non-woody biomass and only 3% went to woody biomass, the remainder being lost or stored in the soil.

Considering these data, I assume the following fractions of deposited N to woody and non-woody (n.w.) biomass, by ecosystem:

	Tropical Forest	*Temp. Forest*	*Grass*	*Ag.*	*Arid*	*Urban*	*Lakes*	*Rivers/ Coasts*	*Marine*
N to n.w. biomass	0.160	0.160	0.120	0.120	0.040	0.040	0.000	0.000	0.000
N to woody biomass	0.040	0.040	0.010	0.020	0.010	0.010	0.000	0.000	0.000

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The estimates of N to n.w. biomass are meant to include any N captured from NO evolved from N deposited onto soils. The amount of N that remains in the soil is calculated as the difference between the total N input and the sum of N to n.w. biomass, N to woody biomass, N-NO losses, and N-N_2O losses. (In the case off lakes, rivers/coasts, and marine, the “soil” category becomes “aquatic organisms”.)

C:N sequestration ratios. The general assumption in most estimates of C sequestration due N deposition (including this one) is that additional N sequesters C at relatively constant C:N ratios – that is, that the addition of N does not change the C:N ratio. This seems generally to be the case. For example, Ma et al. (2001) studied the effects of nitrogen application on the carbon content and concentration of switchgrass and found that added nitrogen increased the biomass of switchgrass but not the carbon concentration.

In the following paragraphs I review data pertinent to estimating C:N ratios for non-agricultural ecosystems, agriculture, and oceans.

Nadelhoffer et al. (1999) calculate C uptake by forest plants worldwide as a result of N deposition. Their calculation assumes the following C:N ratios:

C:N ratio woody biomass	500
C:N ratio n.w. biomass	15
C:N ratio soils	30 (they cite a range of 10 – 30)

Asner et al. (2001) use the following parameter values in their TerraFlux model of biogeochemical processes (ranges, with base-case values in parentheses):

	Lowland tropical forest	*Semi-arid grassland*
NO_3 deposition	0 – 10 kg / ha / yr	0 – 10 kg / ha / yr
NH_4 deposition	--	0 – 10 kg / ha / yr
C:N foliar	20 – 40 (30)	40 – 60 (49)
C:N fine root	20 – 40	40 – 60
C:N wood	(80)	--
C:N soil microbial	8 – 12 (10)	8 – 12 (10)
slow SOC C:N	10 – 20 (15)	10 – 20 (20)

In their comprehensive global calculation of carbon storage due to nitrogen deposition, Townsend et al. (1996) use the following parameter values:

	Trop. forest	*Temp. forest*	*Grass*	*Ag.*	*Arid*
C:N n.w. biomass	50	50 – 70	50 – 55	n.e.	60
C:N woody	150	250 – 300	n.a.	n.e.	180
C:N soil microbial	14	14	10	n.e.	14
C:N soil detrital	120	120	60	n.e.	120
C:N soil slow	25	25	20	n.e.	25

Crop systems have relatively low C:N ratios and rapid turnover times, and therefore low potential to sequester C (Asner et al, 1997). In their model of global C sequestration in response to global N deposition, Holland et al. (1997) ignore N-stimulated carbon storage in crops, because crops “have no perennial tissue in which to store the carbon and most cultivated lands already receive large inputs of nitrogen through fertilization (p. 15849), and because the lifetime of C in crop biomass is so short.

Assuming a typical nitrogen content of 1-3% (see the main text documentation) and a typical carbon content of 40%-50% (see the main text documentation), the C:N ratio for crop biomass ranges from about 10:1 to 50:1. Regarding agricultural soils, data discussed in Appendix C to the LEM model documentation indicate that the use of commercial nitrogen fertilizers may slightly increase carbon sequestration (by slightly reducing carbon oxidation) in agricultural soils, perhaps at the rate of 1.0 g-C / g-N.

Finally, we estimate the C:N sequestration ratios pertinent to carbon sequestration in aquatic biomass. Nitrogen deposited over the oceans appears to increase the sequestration of carbon in the deep ocean (IPCC, 1996, Karl et al., 2002). The nitrogen is a nutrient for marine phytoplankton, which convert carbon to organic and structural molecules (Takeda et al., 1995; Michaels et al., 1996). The organic matter thus formed at the surface of the ocean eventually sinks or is transported by currents to the deep ocean, where it is buried or converted by bacteria into dissolved inorganic carbon. As long as the surface of the ocean remains depleted in nitrate, additions of nitrate will continue to sequester carbon in the deep sea (Karl et al., 2002, p. 83).

Galloway et al. (1995) state that most of the reactive N delivered to the open ocean is quickly incorporated into organic matter at the Redfield C:N ratio and then transported and held below the surface. Much of this nitrogen is remineralized in the main thermocline (p. 248). Similarly, the IPCC (1996) states that “carbon is considered to be taken up by phytoplankton during primary production and released during remineralisation or organic matter in constant proportion to the major nutrients, referred to as Redfield ratios” (p. 492). IPCC data and experimental results in Anderson and Sarmiento (1994) indicate that the C:N ratio is about 7:1 per mole, or about 6:1 per gram.

However, for a variety of reasons, the relationship of C to N can vary away from the classic Redfield ratios (Michaels et al., 1996). Karl et al. (2002) assume a median C:N ratio of 11:1 for remineralisation in their calculation of carbon transport to the ocean. Similarly, Nixon et al. (1996) assume a C:N molar ratio of 10:1 in their calculation of the burial of nitrogen (in organic matter) in the marine continental shelves. Michaels et al. (1996) suggest that a ratio of 15:1 is more appropriate for the one of the main N-fixing diazotrophs in the open ocean.

Nitrogen deposition also may stimulate primary productivity in freshwater ecosystems (Smith et al., 1999). Hessen et al. (1997) suggest that most freshwater systems are P-limited or both P- and N-limited (p. 319), and that the C:N ratio is lower for freshwater than for marine systems.

Considering these data, the LEM assumes the following C:N ratios (negative values are used to indicate C sequestration as opposed to emission):

	Trop. Forest	*Temp. Forest*	*Grass*	*Ag.*	*Arid*	*Urban*	*Lakes*	*Rivers/ Coasts*	*M*
Soil (or aquatic biomass)	-25	-25	-20	-1	-25	-25	-7	-9	-

non-woody biomass	-40	-60	-50	-30	-60	-50	0	0
woody biomass	-150	-275	0	-150	-180	-200	0	0

Ignoring sequestered C that represents accelerated growth rather than additional final mass. Schlesinger (2001) points out that “the growth enhancement from nitrogen deposition may simply allow forests to attain maximum biomass more rapidly, rather than at higher final values” (p. 4). In a similar vein, the IPCC [2001] notes that increases in atmospheric CO_2 concentration merely may cause plants to grow more rapidly rather than cause plants to attain a greater maximum mass. If in fact the C fixed by N deposition would have been fixed anyway, but just at a somewhat later date, then the C sequestration benefit of N deposition is *not* the value of fixing additional C that otherwise would not have been fixed (which is what we wish to represent here), but rather the relatively small difference between the present value of sequestering a fixed amount of carbon at a normal (non-N-enhanced) rate and the present value of sequestering at a slightly N-enhanced rate. This difference in general will be small compared with the difference between sequestering or not sequestering at all a given amount of C.

We wish to include in our estimate of the CEF for NO_2 only C that would not have been fixed by N deposition, and to exclude (because of its small effect) C whose fixation merely has been accelerated by N deposition. Accordingly, I assume that some fraction of C fixed by deposited N is accelerated rather than additional C -- i.e., is C that would have been fixed anyway, but at a later date -- and that this accelerated C has no net carbon sequestration benefit. This accelerated C-fraction is represented by factor ACF in eq. D.54.

We do not have data on the accelerated-C fraction. In the absence of data, I assume a value of 30% for the parameter ACF for soil C, non-woody biomass C, and woody biomass C, in all ecosystems. This of course has the effect of reducing the estimated C-sequestration parameter by 30%.

The length of time that any C remains associated with a gram of added N. Nitrogen does not sequester carbon in ecosystems permanently. Eventually, sequestered C is oxidized and returned to the atmosphere as CO_2, and the N with which it was associated – perhaps after several cycles of sequestering C – goes to a sink in which it no longer is able to fix C. Thus, the CO_2- removal effect of N deposition is only temporary. To account for this, the discounted present value of the “final” release of sequestered C as CO_2 (where the “final” release is the one that occurs after the N is no longer available to sequester C) must be deducted from the original C sequestration. This discounted deduction is accomplished by the term 1-DF in eq. D.54. If the discount rate is zero, then a merely temporary sequestration has no value: the present value of the final release cancels the value of the original sequestration. On the other hand, if the discount rate is very large, or the time of sequestration is very long (with a non-zero discount rate), then the final release of the sequestered carbon has essentially no value, and the carbon sequestration is effectively permanent[18].

It is difficult to estimate the length of time that any carbon is effectively fixed by nitrogen fertilization in different ecosystems. The main difficulty lies in determining the fate of the nitrogen, which might be available for carbon fixation again after the release of the "initially" fixed carbon. As Holland et al. (1997) point out, deposited N is "continually recycled in terrestrial ecosystems by the release or mineralization of N during decomposition and subsequent plant and microbial uptake of that N" (p. 15850). In the "first" cycle, atmospherically deposited N may sequester a certain amount of C in plant biomass. After the plant dies and the biomass decomposes, the C oxidizes to CO_2 and is released to the atmosphere, but the N may be made available for another cycle of C sequestration. This will continue until the N ends up in one of its permanent sinks, (as N_2, N_2O, N sequestered in soils of sediments, or increased nitrate concentration in aquatic systems). From the standpoint of determining the C-sequestration CEF for NO_2 – which is our purpose here – the relevant factor is the length of time that *any* C remains associated with an additional gram of N deposition.

As indicated in eq. D.56, in the LEM the effective lifetime of sequestered C is estimated as a multiple of the "once-through" lifetime of C, where the once-through lifetime is the time from fixation to oxidation, and the multiplier accounts for the possibility that after the oxidation of the "first" C, another C may be fixed by the *same* N molecule that fixed the first C. In this section we estimate the once-through lifetime of C (from fixation to oxidation) and the number of C lifetimes that N remains active in the ecosystem.

We begin with estimates of the once-through lifetime of C in different ecosystems. In their comprehensive global calculation of carbon storage due to nitrogen deposition, Townsend et al. (1996) use the following parameter values for woody and non-woody biomass C:

	Trop. forest	*Temp. forest*	*Grass*	*Ag.*	*Arid*
lifetime woody C (yrs)	150 – 200	100 – 150	n.a.	n.e.	20
lifetime n.w. C (yrs)	1 – 1.2	1 – 2	2 – 3	n.e.	1

I adopt their mid range values here.

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[18] Note that, in order to completely "cancel" an emission of CO_2 from fossil-fuel combustion, the sequestration of carbon in biomass must be permanent since the carbon in fossil fuel presumably would be never be released naturally were it not released by man. The general rule is straightforward: in order to cancel an anthropogenic emission that advances natural emission by X years, the biotic sequestration must last for X years. Were it not burned by us, fossil fuel would oxidize naturally over geologic time, which from our perspective is practically infinite.

Regarding soil, the IPCC (2000) distinguishes three classes of soil organic matter (SOM)[19]: litter, with a life of months to years; "active," with a life of years to decades; and "stable," with a life of centuries to millennia. They state that "active SOM largely consists of soil micro-organisms and their immediate products, with a cycling or residence time of 1-10 years. Stable or slow-release SOM consists of neo-formations of polymeric substances, which can be extremely diverse in composition...it has a residence time of between 10 and 50 years (sometimes more). Inert or 'recalcitrant' SOM...would not be destroyed for up to 500 years" (sec. 2.2.5.5; also Table 4-3). Similarly, Nadelhoffer et al. (2002) distinguish between "light" SOM with a decadal turnover time, and heavier mineralized fractions with multi-decadal to century turnover times. Kuylenstierna et al. (1998) imply that long-term storage of nitrogen in soil organic matter is on the order of at least 50-200 years. They state that this long-term storage will occur in acid soils with a high C:N ratio.

Comins and McMurtrie (1993) assume that "active" soil organic matter decomposes in 2-3 months, that "slow" organic matter decomposes in 10 years, and that "passive" organic matter decomposes in 300 years. Davidson and Hirsch (2001) assume 2 years, 20 years, and 200 years for the three pools.

I assume that if carbon is sequestered according to C:N ratios, it happens mainly (but not exclusively) in the active, light fractions controlled by micro-organisms. I assume a once-through lifetime of 20 years for soil in all ecosystems.

It is difficult to estimate the average lifetime of carbon fixed in the oceans as a result of N fertilization. First, it is not clear what fraction of the organic material formed as a result of nitrogen fertilization is permanently or nearly permanently sequestered in the ocean sediments or as dissolved inorganic carbon. Nixon et al. (1996) report an estimate that 10 – 30% of the primary production of marine continental shelves is buried in sediment. This sequestration presumably is effective for many decades if not centuries. However, the fate of the remaining carbon is not clear.

By contrast, the IPCC (2001, their Figure 3.1c) indicates that less than 1% of the net primary production (NPP) of plankton is buried in marine sediments. According to the IPCC (2001), about 75% of the NPP-carbon is returned as dissolved inorganic carbon (DIC) in the surface layer by heterotrophic respiration, and the remainder is returned as DIC in the middle layers by heterotrophic respiration. The IPCC (2001) figure 3.1c indicates that DIC in the middle layers circulates to the surface layers, and that DIC in the surface layer exchanges with CO_2 in the atmosphere, but unfortunately the IPCC (2001) does not indicate the timescale of this circulation[20].

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[19] The IPCC (2001, sec. 2.2.5.5) defines SOM as all organic compounds in the soil that are not living roots or animals.

[20] The IPCC (2001) says that "heterotrophic respiration at depth converts the remaining organic carbon back to DIC. Eventually, and usually at another location, this DIC is upwelled into the ocean's surface layer again and may re-equilibrate with the atmospheric CO_2" (p. 198).

The IPCC (2001, Figure 3.1c) also indicates that there is a small net accumulation of atmospheric carbon in the ocean. This means that a small fraction of the NPP-C effectively remains in the oceans as DIC forever.

In their model of global carbon sequestration in response to global N deposition, Holland et al. (1997) estimate N deposition over the oceans, but do not estimate the consequent C sequestration, apparently on the grounds that the turnover time for such C sequestered in the oceans in less than a decade (p. 15849).

Micheals et al. (1996) state that 50-80% of the carbon taken up by oceanic organisms as a result of excess nitrate is sequestered in the oceans for multi-year timescales, but that little is sequestered for longer than a decade (p. 216-217). Using their data on the residence time of the nitrate that is associated with the carbon uptake, I calculate a mean residence time of 8 to 18 years.

Given these data, I assume a once-through C lifetime of 2 years for freshwater lakes, 5 years for coastal areas, and 10 years for oceans.

Finally, we consider the last parameter: the number of C lifetimes that N remains available to sequester C. Unfortunately, it is difficuult to find information on this parameter. In the absence of data, I assume a multiple of 3.0 for soil, 1.5 for woody biomass, and 5.0 for non-woody biomass, in all terrestrial ecosystems. For aquatic ecosystems, I assume a multiplier of 3.0 for freshwater systems, 2 for coastal areas, and 3.0 for oceans.

Comparison of our estimate of global effect of N deposition on C sequestration with other estimates. Our estimate of the CEF for the effect of NO_2 deposition on carbon sequestration can be compared with other estimates of the global average N:C sequestration ratio. (The CEF gives the CO_2:NO_2 sequestration ratio, which is close to the C:N ratio.) The IPCC (1996a) reviews estimates of nitrogen deposition and carbon sequestration done and concludes that the 50 to 80 Tg of nitrogen deposited annually worldwide[21] from fertilizers and fossil-fuel burning in the 1980s resulted in carbon sequestration of 500 to 1000 Tg C per year[22]. (The studies reviewed by the IPCC [1996a, Table 9.10 actually estimate that 7 to 30 Tg of N input resulted in 200 to 2000 Tg of C stored, but the IPCC argues that the high end of the estimated C-storage range is unlikely.) Vitousek et al. (1997) state that estimates of net carbon storage due to nitrogen deposition range from 100 to 1300 Tg/yr., the IPCC (2001) cites a low estimate of 200 Tg/yr. (Nadelhoffer et al., 1999, reviewed below) and a high estimate of1400 Tg/yr., and Nadelhoffer et al. (1999) state that estimates in the literature range from 100 to 2300 Tg of C/yr. These estimates indicate that today, the emissions and subsequent

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[21] Assuming that all N emitted to the atmosphere is deposited to the earth, this estimate implies worldwide N emissions of 50-80 Tg-N/yr .in the 1980s. The IPCC (2001, ch. 4) cites estimates that global emissions of NO_x (natural and anthropogenic) were about 45 Tg-N/yr in the 1980s. This in turn implies emissions of about 20 Tg-N/year from other sources.

[22] For a review of deposition of organic nitrogen specifically, see Neff et al. (2002). For model analyses of long-term relationships between the nitrogen cycle and the carbon cycle, see Mackenzie et al. (2002).

deposition of one gram of N results in the global-average sequestration of anywhere from 1 to 50 grams of C, with a best estimate of about 1 to 10. Our estimated CEF of about 7 (or about 9 if we do not deduct accelerated carbon, or discount future emissions of sequestered CO_2) lies within this best-estimate range.

Particulate nitrate (eq. D.57). Note that the CEF is calculated on the basis of the ratio of the weight of NO_3 to the weight of NO_2, rather than on the basis of the weight of the particulate nitrate formed (NH_4NO_3, with a formula mass of 80 g / mole). This is because the radiative forcing estimates discussed above are with respect to NO_3; that is, they are in units of W $(g\text{-}NO_3)^{-1}$. However, I assume that nitrate has its effect only as part of an ammonium nitrate molecule – hence the factor, in the formula above, that accounts for the fraction of nitrate that is neutralized by ammonia to particulate nitrate.

Acid deposition. It is well established that acid deposition and photochemical oxidants can damage plants and reduce primary productivity (e.g., Kley et al. [1999] regarding damage from photochemical oxidants). However, studies reviewed in Delucchi et al. (1998) indicate that crop damage due to acid nitrate and sulfate deposition is relatively minor, and smaller than the nitrogen fertilization effect. I assume, therefore, that the parameter $CEF_{NOx/DN}$ (the CO_2 equivalency of the effect of deposited acid nitrate on carbon sequestration by plants) is zero.

The calculated CEF. With these parameter values, we estimate the components of the total CEF for NO_X as follows (for emissions in the U. S. in the year 2020):

Effect	CEF
Effect on tropospheric O_3	13.0
Effect of NOx on lifetime of ambient CH_4	-5.1
Effect of deposition on CH_4 emissions	0.0
Effect of deposition, leaching on N_2O emissions	3.4
Effect of deposition on C sequestration	-6.6
Effect of particulate nitrate	-2.7
Effect of acidification on C sequestration (by assumption)	0.00
Total (sum of all components)	*2.1*

(These estimates do not account for the 1-NORF factor in equation D.45 and D.46). The single largest component (in absolute value) is the positive (warming) effect on tropospheric ozone. However, the negative (cooling) effects of C sequestration, particulate formation, and methane life-reduction together nearly outweigh the positive effects on tropospheric ozone and N_2O, so that the total CEF (the sum of all components) is relatively small. Of course, all of these indirect effects of NO_2 on climate are difficult to model and hence very uncertain, and as a result the overall CEF for NO_X

is quite uncertain. Because the uncertainty is relatively large, and because most fuelcycles produce substantial amounts of NO_x, the uncertainty regarding NO_X CEF values has a non-trivial effect on estimates of fuel-cycle CO_2-equivalent emissions.

Note on fertilization by agricultural fertilizers . Deposition of atmospheric NO_X from fossil-fuel combustion is not the only source of nitrogen fertilizer; the commercial nitrogen fertilizer that leaches and runs off from agricultural fields also eventually eutrophies freshwater and marine ecosystems. CO_2 sequestration due to commercial nitrogen fertilizers can be estimated on the basis of some of the same data used to estimate carbon sequestration due to nitrogen deposition, except that one needs to know the fraction of nitrogen fertilizer, rather than the fraction of NO_X emissions from fossil fuel combustion, that reaches marine, freshwater, and terrestrial ecosystems. The effect of commercial fertilizers is estimated in the main report.

Ozone-forming potential of organic compounds

Ozone -- an urban air pollutant and “greenhouse gas” in the troposphere -- is not emitted as such by motor vehicles or any other combustion source, but rather is formed in the atmosphere from a series of photochemical reactions that involve NO_X, reactive organic compounds, and other compounds. The reaction rate and equilibrium depends on the relative abundance of the reactants, the characteristics of the organic compounds, temperature, atmospheric mixing, and other factors (National Research Council, 1991). The reactions are complex and highly nonlinear, and there is no simple, universal formula for determining the marginal contribution of each emission source or each precursor pollutant to ozone.

The National Research Council (1991) provides a good summary of the ozone formation process (see also Kley et al, 1999, and Fowler et al., 1999). First, reactive organic compounds (RH), emitted from a variety of sources, react with hydroxyl radicals (OH) to form organic radicals (R):

$$RH + OH \text{-->} R\cdot + H_2O \qquad \text{eq. D.61}$$

The organic radicals combine with oxygen in the presence of an inert third body M to form peroxy radicals (RO_2):

$$R\cdot + O_2 \text{--M-->} RO_2\cdot \qquad \text{eq. D.62}$$

The peroxy radicals react with nitric oxide, which is emitted from combustion and other sources, to form nitrogen dioxide:

$$RO_2\cdot + NO \text{-->} NO_2 + RO\cdot \qquad \text{Eq. D.63}$$

Nitrogen dioxide is photo-dissociated by high-energy solar radiation (hv, Planck's constant h multiplied by frequency v):

NO_2 + hv --> NO + O eq. D.64

The oxygen atoms then combine with molecular oxygen to form ozone (O_3):

$O + O_2$ --M--> O_3 eq. D.65

The foregoing series of reactions also slow the removal of methane by decreasing the OH radical available (eq. D.61) to oxidize methane.

In this simplified representation of the chemistry, there are two main precursor pollutant emissions: reactive organic compounds (RH; henceforth, I will refer to these as nonmethane organic compounds, or NMOCs), and nitrogen oxides (NO_x). As one can infer from even the simplified chemistry, the ozone contribution of the reactive organics depends on the rate of reaction with OH (eq. D.61), and on the quantity and characteristics of the organic reaction products (eq. D.61, D.62, D.63). The effect of NMOCs on methane, via removal of the OH radical, similarly depends on the rate of the initial reaction (eq. D.61), and on the subsequent reactions involving the products of D.61

Some compounds, such as ethane, are relatively unreactive; others, such as certain aromatics, are quite reactive. Because different fuels and in general different emissions sources emit different kinds of organic compounds, and the OH-removing and ozone-forming propensity of compounds can vary over two orders of magnitude, an accurate and fair evaluation of the ozone-impact of different sources of organic compounds ought to consider the OH-reactivity and ozone-forming propensities of the compounds. Because the reactivity of NMOC with OH, and hence the effect of NMOC on methane, is a major part of the overall "ozone forming potential," we will assume that an index of the potential of an NMOC to form ozone -- which is what we discuss below -- also is an index of the potential of an NMOC to slow the destruction of methane.

Maximum Incremental Reactivity (MIR), and Photochemical Ozone-Creation Potential (POCP). To estimate the relative ozone-forming potential, or reactivity, of NMOCs, researchers first construct an atmospheric photo-chemistry model of organic compounds and ozone formation. These models depend heavily on data obtained from laboratory experiments designed to simulate the formation of ambient ozone (Derwent et al., 1998; Carter, 1994). With such a model, investigators can estimate the change in ozone resulting from a change in emissions of NMOCs, under a variety of scenarios. The scenario variables can include the magnitude of the change in emissions of NMOCs, emissions of NO_x, environmental conditions, the measure of ozone (e.g., peak ozone, or integrated ozone exposure), and other factors (Derwent et al., 1998; Carter, 1994). The models themselves can vary greatly with respect to the complexity of the chemistry represented (e.g., the number of compounds and the number of reactions) and complexity of the atmospheric physics (e.g., trajectory modeling versus three-dimensional grid modeling). The result is that there is no single "correct" set of reactivity factors for NMOCs; rather, there are many sets, representing different measures, scenarios, and models.

In the U. S., the most widely used "reactivity"-adjustment factors are the maximum incremental reactivity (MIR) factors developed by Carter (1998, 1997a, 1997b, 1994), on the basis of chemical mechanisms that simulate a variety of experiments with environmental chambers. The incremental reactivity is the change in ozone per unit NMOC added (g-O_3/g-NMOC), and the MIR is the incremental reactivity when NO_x emissions are adjusted so that the base-line NMOC mixture has the maximal incremental reactivity (Carter, 1994). The comprehensive results published in 1994 are based on a mechanism with 120 organic compounds (Carter, 1994). The mechanisms have since been updated (Carter, 1997a), and new MIRs published (Carter, 1998).

In Europe, the "photochemical ozone creation potentials" (POCP) developed by Derwent et al. (1998, 1996) are widely used. Derwent et al. (1998) used a "master chemical mechanism" containing over 2400 chemical species and 7100 reactions to describe the atmospheric degradation of organic compounds and the associated regional scale ozone production, for conditions typical in northwest Europe. For over 120 organic compounds, they estimated the POCP relative to that of ethylene. The predicted ozone concentrations along a five-day trajectory matched those predicted by chemical mechanisms based on smog chamber data, and the estimated POCPs generally correlated well with Carter's MIRs.

Carter's most recent MIRs (Carter, 1998) are reported relative to the MIR of a base NMOC mixture (4.08 g-O_3/g-NMOC). Derwent et al. (1998) report their POCPs relative to that of ethylene (given a POCP of 100). Assuming that exhaust from conventional gasoline vehicles has an MIR of 3.8 g-O_3/g-NMOC (Kelly et al., 1996a, 1996b, 1996c; Black et al., 1998; Auto/Oil, 1996)[23], and a POCP of 0.78 (based on the estimate of

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[23] Kelly et al. (1996a, 1996b, 1996c) applied Carter's (1994) MIRs to the speciated exhaust from vehicles using reformulated gasoline and driven over the FTP, and estimated the specific reactivity, or ozone-forming potential: 4.1 g-O_3/g-NMOC exhaust from 35 Dodge B-250 vans; 3.2 g/g from 69 Dodge Spirits; 4.4 g/g from 18 Ford Econoline vans; and 3.74 from 21 Chevrolet Luminas. The values for vehicles using conventional gasoline and driven over a more realistic driving cycle (with higher speeds and harder accelerations than on the FTP) would be similar, because the ozone-forming potential of emissions from conventional gasoline is a bit higher than that for reformulated gasoline, but the ozone-forming potential of emissions from high-speed, hard-acceleration driving is a bit lower than that of emissions of emissions from the FTP (Ho and Winer, 1998; Black et al., 1998; Auto/Oil, 1996).

Similarly, the Auto/Oil (1996) program applied Carter's (1994) MIRs to the speciated exhaust of vehicles using conventional (industry-average) gasoline (g-O_3/g-NMOC):

	FTP (standard)	REPO5 (high-speed)
4 1989 passenger cars	4.10	3.88
1992 pickup, 1992 wagon, 1993 car	3.71	3.25

Derwent et al., 1996; discussed below), we can re-estimate the most recent Carter MIRs and the Derwent et al. POCPs *relative to the MIR or POCP of exhaust from conventional gasoline:*

Compound	MIR	POCP	MIR / POCP	0.70 MIR + 0.30 POCP
alkanes				
methane	0.004	0.008	0.55	0.01
ethane	0.08	0.16	0.54	0.11
propane	0.15	0.23	0.66	0.17
n -butane	0.31	0.45	0.68	0.35
i -butane	0.34	0.39	0.86	0.36
n -pentane	0.38	0.51	0.75	0.42
i -pentane	0.45	0.52	0.86	0.47
n -hexane	0.31	0.62	0.50	0.40
n -heptane	0.23	0.63	0.37	0.35
n -octane	0.18	0.58	0.31	0.30
alkenes				
ethylene	2.17	1.28	1.70	1.91
propylene	2.88	1.44	2.00	2.45
1-butene	2.77	1.38	2.00	2.35
isoprene	2.44	1.40	1.74	2.13
aromatics				
benzene	0.21	0.28	0.76	0.23
toluene	1.34	0.82	1.64	1.18
o - xylene	2.21	1.35	1.63	1.95
m - xylene	3.70	1.42	2.61	3.02
p - xylene	0.75	1.29	0.58	0.92
aldehydes				
formaldehyde	1.72	0.67	2.58	1.40
acetaldehyde	1.63	0.82	1.99	1.39
alcohols				
methanol	0.17	0.17	1.01	0.17
ethanol	0.45	0.49	0.90	0.46
ethers				
dimethylether (DME)	0.23	0.22	1.05	0.23
MTBE	0.19	0.19	0.98	0.19
ETBE	0.56	0.27	2.05	0.48

As shown in the fourth column of the table above, the alkanes are less reactive, and the alkenes and aldehydes more reactive, in the MIR scheme. Derwent et al. (1998) explain that the long-range transport model used to estimate POCPs "gives an

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These studies consistently indicate that the specific reactivity of NMOC exhaust from vehicles using conventional gasoline and driven in a realistic driving cycle is between about 3.4 and 4.2. I assume an average of 3.8.

increased resolution of the least reactive organic compounds [i.e., alkanes] compared with the MIR scale" (p. 2435).

The differences between the MIR and the POCP values arise from differences in chemical mechanisms, differences in NO_X scenarios, and differences in model conditions: the MIR scale is based on a single-day trajectory model specified for U. S. conditions; the POCP scale is based on long-range transport model specified for European. conditions.[24] The last column of the data table above shows a weighted 0.70 MIR + 0.30 POCP measure, which I use as the basis of some of my assumptions in Table D-4. Note that the term "POCP" refers specifically to the measure of Derwent et al. (1998), whereas the term "OFP" refers to the ozone-weight used in this analysis.

<u>Relative Ozone-Formation Potential (ROFPs) of NMOCs from different sources</u>. Given a set of ROFPs, as above (the weighted relative MIR and POCP values), one can calculate the ROFP-weighted NMOC emission from a particular source (e.g., methanol vehicles) by multiplying emissions of each individual compound by its ROFP, and summing over all compounds emitted. The emissions-weighted source-average ROFP is then equal to ROFP-weighted emissions divided by unweighted emissions:

$$ROFP_s = \frac{\sum_i ROFP_i \cdot EM_{i,s}}{\sum_i EM_{i,s}} \qquad \text{eq. D.66}$$

where:

$ROFP_S$ = the emissions-weighted ozone-creation potential of NMOCs from source *s*, relative to that of NMOCs from the combustion of gasoline
$ROFP_i$ = the ozone-creation potential of NMOC *i* relative to that of NMOCs from the combustion of gasoline
$EM_{i,s}$ = emissions of NMOC *i* from source *s*

In the model, I distinguish several sources of NMOCs (see Table D-4), and then further distinguish combustion emissions from evaporative or leakage emissions. For those emission sources where the NMOC composition is specified in the model (e.g., leaks of gaseous fuels, such as natural gas), eq. D.66 is used in the model to calculate

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[24] Carter (1994, 1998) has several other reactivity scales, in which the impact of NO_X or the measure of ozone production are different than in the MIR scale. In at least one important respect, the difference between the MIR and the other Carter scales is similar to the difference between the MIR and the POCP scale: in these other Carter reactivity scales, as in the POCP scale of Derwent et al. (1998), the alkanes are relatively more reactive than in the MIR scale. It appears to me that, qualitatively, the differences between the MIR and the POCP scales are at least as great as the differences between the MIR and the other Carter scales, so that MIRs and POCPs establish reasonable alternative bounds on the likely reactivity values.

the ROFP of NMOCs from the source. Where the composition is not specified in the model (e.g., exhaust emissions from methanol vehicles), I turn to estimates by others of the ROFP. These are discussed next.

ROFPs for exhaust emissions from alternative fuel vehicles have been estimated. According to Black et al. (1998), CARB uses the following ROFPs, for reformulated gasoline (RFG), 85% methanol/15% gasoline (M85), 85% ethanol/15% gasoline (E85), and compressed natural gas (CNG)[25]:

RFG	M85	E85	CNG
0.94	0.37	0.63	0.43

Kelly et al. (1996a, 1996b, 1996c) calculated the specific reactivity (g O_3/gNMOC) of all NMOC emissions from vehicles using reformulated gasoline, methanol, and ethanol, by summing the product of the specific reactivity and the emissions share. Using Carter's (1994) MIR factors, they estimated the following ROFPs, relative to reformulated gasoline (RFG), for flexible-fuel vehicles (FFVs) and single-fuel vehicles (SFVs) tested over the FTP:

	M85 v RFG in FFV	M50 v RFG in FFV	E85 v. RFG in FFV	E50 v. RFG in FFV	CNG v RFG in SFV
auto	0.36	0.75	0.70	0.87	--
van	0.49	0.81	--	--	0.50

The Auto/Oil (1996) program tested similar vehicles over the FTP, and over high-speed, high-acceleration cycle called the REP05, and used Carter's (1994) MIRs to estimate ROFPs relative to conventional gasoline (CG) and reformulated gasoline (RFG):

	M85 v CG in FFV	M85 v RFG in FFV	E85 v. RFG in FFV	CNG v. CG in SF van	CNG v.RFG, SF van
FTP	0.56	0.48	0.76	0.52	0.54
REP05	0.87	0.80	1.23	0.96	0.98

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[25] The results of detailed photochemical air quality modeling usually imply that alternative fuels are less effective at reducing ozone levels than might be inferred from the ozone-forming potential factors. For example, Dunker et al. (1996) used the Urban Airshed Model, a detailed, three-dimensional grid model that includes transport, dispersion, deposition and atmospheric chemistry, to estimate the effect of alternative fuel formulations on various measures of ozone, and found that "a research test gasoline produced less ozone than M85 cases in Los Angeles and New York and either more or less ozone than M85 in Dallas-Fort Worth, depending on the assumptions" (p. 787).

Generally, under a high-speed, high-acceleration driving cycle (the REP05), compared with the standard cycle (the FTP) the g-O_3/g-NMOC specific reactivity of alternative fuels increases, whereas the specific reactivity of gasoline stays the same or decreases, with the result that the ROFP for alternative fuels is higher under the high-speed, high-acceleration drive cycle. (Black et al. (1998) report similar results.) The upshot is that real-world ROFPs for alternative fuels are likely to be somewhat higher than those estimated for the FTP.

Black et al. (1998) point out that ROCPs for NMOC exhaust from light-duty CNGVs have ranged from 0.18 to 0.87, and than show that this variation is a function of the level of NMOC emissions: high NMOC emissions result from high levels of uncombusted low-C, unreactive alkanes, such as ethane. Accounting for the higher ROFPs under real-world driving than under the FTP, the following function reasonably well describes ROCP vs. NMOC emission data shown by Black et al. (1998):

if $NMOC_{CNG} > 0.75$, ROCP = 0.15;

otherwise:

$$ROCP_{CNG} = 1.2017+1.3086 \cdot NMOC_{CNG}^{-0.11}$$

where $NMOC_{CNG}$ is the NMOC exhaust emission rate for LD CNGVs, in g/mi.

Ho and Winer (1998) tested the effect of gasoline composition and driving cycle on the reactivity of exhaust emissions. Their results indicate that distillation temperature, sulfur concentration, aromatic content, and oxygen content can affect exhaust reactivity, but not in any easily quantifiable way. They did find that reactivity of exhaust decreased slightly in the high-speed, high-acceleration driving cycle, compared with the FTP.

To help us estimate the ROFP of NMOC emissions from other sources, such as diesel vehicles, or power plants burning coal, we can use work by Derwent et al. (1996). Using an early version of the “master chemical mechanism” developed more fully in Derwent et al. (1998), they estimated what they called a “sector-mean POCP” for each major emissions-source sector in the United Kingdom’s emission inventory. The sector mean POCP, for emission sector *s*, was estimates as follows:

$$POCP_s = \frac{\sum_c VOC_{UK-s-c} \cdot POCP_c}{VOC_{UK-s}}$$

where:

VOC_{UK-i-s} = emissions of compound *i* in sector *s* in the U. K.

$POCP_C$ = photochemical ozone-creation potential of organic compound *c* under European conditions

$VOC_{UK\text{-}i}$ = emissions of VOCs in sector *s* in the U. K.

Expressing the sector-mean $POCP_S$ relative to that for their sector "petrol exhaust[26]," we have the following (for $POCP_S / POCP_{petrol}$):

Petrol exhaust	1.00
Petroleum refining and distribution	0.79
Petrol evaporation[27]	0.78
Solvent usage	0.76
Stationary combustion	0.63
Diesel exhaust	0.56
Industrial and residential waste	0.36
Natural gas leakage	0.33
Chemical processes	0.27

Of course, in applying this, I assume that POCPs estimated for European conditions are similar to POCPs for U. S. conditions, and that the mix of VOCs in each source category in the U. K. inventory (e. g., diesel) is similar to the mix in the corresponding category in the U. S. emissions inventory.

Finally, Hayman and Derwent (1997) show that the POCPs of the likely near-term replacements for CFCs (mainly, hydrochlorofluorocarbons [HCFCs] and hydrofluorocarbons [HFCs]) are very low -- on the order of 0.10 (relative to a POCP of 100 for ethylene), which is lower even than the POCP for methane. Hence, we can ignore the contribution of HFCs and HCFCs to tropospheric ozone.

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[26] For reference, $POCP_{petrol}$ = 0.78.

[27] Black et al. (1998) estimated an ROFP of 0.70 for evaporative emissions of reformulated gasoline, relative to exhaust from reformulated gasoline.

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TABLE D-1. THE EFFECT OF POLLUTANTS ON GLOBAL CLIMATE AND ESTIMATED CEFS

Pollutant --> effects related to global climate	CEF (U.S. 1990)	CEF (U.S. 2050)
CO_2 --> +R	1	1
CH_4 --> +R, -OH, +O_3 (t), +CH_4, +H_2O (s), +CO_2	17	14
N_2O --> +R	288	250
CFC-12 --> +R, -O_3 (s)	10,550	9,600
HFC-134a --> +R	1,170	1,060
O_3	6.0	6.0
PM (black carbon) --> +R, clouds	1,630	1,490
PM (organic matter) --> -R, clouds	- 170	-150
CO --> -OH, +O_3 (t), +CH_4, +CO_2	9.2	9.1
H_2 --> -OH, +O_3 (t), +CH_4	37	36
NMOCs --> -OH, ±O_3 (t), +CH_4, +CO_2	3.3 + C	3.4 + C
NO_2 --> -CO_2, +N_2O, ±OH, ±O_3 (t), ±CH_4, +PM	-0.0	4.4
SO_2 --> +PM	- 44	- 40
H_2O --> +R (s), +OH, -CH_4, clouds	n.e.[a]	n.e.[a]

CEF = CO_2-equivalency factor, estimated on a mass basis; R = direct radiative forcing; (t) = troposphere; (s) = stratosphere. “+” = positive effect, “-” = negative effect; n.e. = not estimated. See discussion in the text for details.

[a] Water vapor actually is the main climate-controlling gas. However, H_2O emissions from fossil-fuel use worldwide are a tiny fraction of global evaporation (0.0013%), and hence probably can be ignored.

TABLE D-2: CONCENTRATIONS, RATES OF CHANGE, AND LIFETIMES OF SOME GHGS

	CO_2	CH_4	N_2O	CFC-11	HCFC-23
Pre-industrial conc.	~280 ppmv	~700 ppbv	~270 ppbv	zero	zero
1998 concentration	365 ppmv	1745 ppbv	314 ppbv	268 pptv	14 pptv
Rate of conc. change	1.5 ppmv/yr (0.4%/yr)	7.0 ppbv/yr (0.6%/yr)	0.8 ppbv/yr (0.25%/yr)	-1.4 pptv/yr (0%/yr)	0.55 pptv/yr (5%/yr)
Atmospheric lifetime	50-200 yrs[a]	12 yrs[b]	114 yrs[b]	45 yrs	260 yrs

Source: IPCC (2001).

[a] No single lifetime is defined because of different rates of uptake by different removal processes. (See the discussion in the text.)

[b] This lifetime is defined as an “adjustment time” or “perturbation lifetime” that takes into account the indirect effect of the gas on its own residence time.

TABLE D-3. ESTIMATES OF THE CO_2-EQUIVALENCY OF CLIMATE GREENHOUSE GASES OTHER THAN CO_2

GWPs

	CH_4	N_2O	CFC-12	HFC-134a	CO	NMOC	NO_2
IPCC[a]							
First and second reports							
20-year horizon	56	280	6,000-6,800	3,400	7	31	30[b]
100-year horizon	21	310	6,200-7,100	1,300	3	11	7[b]
500-year horizon	6.5	170		420	2	6	2[b]
Third assessment report							
20-year horizon	62	275	7,100-9,600 (10,200)	3,300			
100-year horizon	23	296	7,300-9,900 (10,600)	1,300			
500-year horizon	7	156	(5,200)	400			
Martin & Michaelis (1992)[c]							
50-year horizon	26.5	270			3	8.8	3
Johnson and Derwent(1996)[d]							
100-year horizon	28.7				2.1	1 to 6	5.0 / -10
Fuglestvedt et al. (1996)[e]							
100-year horizon	30				3		441 aircraft
Derwent et al. (2001)[f]							
100-year horizon	25.7				1.6		4.5 / 15 (277 aircraft)

EDIs

	CH_4	N_2O	CFC-12	HFC-134a	CO	NMOC	NO_2
Reilly (1992)[g]							
Linear damages	21	201	2,140		0.9		
Quadratic damages	74	208	7,309		2.9		
Quadratic damages + CO_2 fertilization	92	260	9,119		3.7		
Hammitt et al. (1996)[h]							
Middle case	11.0	354.8	9,067				
Damage exponent = 1	27.2	354.7	9,279				
Damage exponent = 3	5.1	340.1	8,527				
'Hockey stick' damages	6.1	319.4	7,910				
Low climate sensitivity	10.0	353.4	9,028				
High climate sensitivity	12.3	356.6	9,142				
Discount rate = 1% / yr.	3.7	322.2	7,950				
Discount rate = 5% / yr.	23.7	366.2	9,596				
IS92c emission / GDP	22.2	345.2	8,934				
IS92e emission / GDP	8.01	399.2	10,272				
Emission year 2005	6.78	364.0	9,423				
Emission year 2015	3.96	373.5	9,779				
Minimum	49.7	296.7	7,286				
Maximum	2.9	403.6	10,507				
Kandlikar (1996)[i]							
linear damages, r = 2%	27.8	269					
quadratic damages, r = 2%	17.9	282					
linear damages, r = 6%	42.6						
quadratic damages, r = 6%	31.0						
Tol (1999)[j]	14	348					
Reilly et al. (2001)[k]							
TH=100, r = 5.0%	30	225					
TH=200, r 5% to 0%	~10	~250					

Notes: see next page. TH = time horizon, r = annual discount rate.

[a] First and second assessment reports. The results for CH_4, N_2O, CFC-12, and HFC-134a are from IPCC (1996). The estimate for CH_4 includes the indirect effects of tropospheric O_3 production and stratospheric water vapor production. The ranges shown for CFC-12 reflect uncertainty in the magnitude of CFC-induced cooling of the lower stratosphere (estimated at ±30%), and uncertainty in the globally averaged ozone-removal efficiency of bromine relative to that of chlorine. In calculating these values, Daniel et al. (reported in IPCC, 1996) assumed that O_3-depleting gases can be compared in a globally averaged sense, that future CO_2 levels are constant, and that indirect effects for each gas depend linearly on its contribution to chlorine or bromine release in the stratosphere. The estimates for CO, NMOCs, and NO_2 are from the 1990 IPCC report (Shine et al., 1990), and represent early, preliminary estimates of total direct-plus-indirect GWPs. The second IPCC assessment report (IPCC, 1996) essentially disavowed these earlier estimates of total GWPs, on the grounds that it was not possible to estimate indirect effects accurately for these relatively short-lived and poorly mixed gases.

Third assessment report. The IPCC (2001) calculates direct forcing GWPs for N_2O and HFC-134a, and direct+indirect GWPs for CH_4 and CFC-12. Our table shows the direct+indirect GWP for CH_4, and the range of direct+indirect GWPs for CFC-12 with the direct-only effect shown in parentheses.

For all these gases, the IPCC (2001) calculates direct radiative forcing GWPs using a variant of eq. D.1 presented in the text here, with the following parameter values:

	CH_4	N_2O	CFC-12	HFC-134a	CO_2
molecular weight (g/mole)	16.04	44.01	120.91	102.01	44.01
forcing (W per m^2 per ppmv)	0.370	3.1	320	150	0.01548
lifetime (years)	12	114	100	13.8	varies

The forcing term shown here is the ratio F^*/C in eq. D.3. The fate of CO_2 is not represented by a single decay function, and hence the representative lifetime depends on the time horizon being considered. Using eq. D.4 and the IPCC parameter values presented in this note, I find that the following effective lifetimes for CO_2 (values of "L" in eq. D.4) reproduce all the IPCC GWPs exactly: 23 years for the 20-year horizon, 53 for the 100-year horizon, and 150 years for the 500-year horizon. (See the discussion in the text for more on the lifetime of CO_2.)

The IPCC accounts for the indirect warming effects of CH_4 by multiplying the calculated direct effect by 1.30 (25% additional warming due to production of ozone, and 5% additional warming due production of stratospheric water vapor).

In its TAR the IPCC (2001) discusses GWPs for CO and NO_x, and presents estimates by Johnson and Derwent (1996) and Fuglestvedt et al. (1996), but does not develop its own values.

[b] The GWPs originally published in Shine et al. (1990) were: 150, 40, and 14, for the 20-, 100-, and 500-year time horizons. However, those values were in incorrect due to an error in the calculation of O_3 inventory changes (Johnson et al., 1992). I have shown the corrected values here (for emissions of NO_x at earth's surface) (Johnson et al., 1992).

[c] The results for the 50-year time horizon are from modeling done by Harwell Laboratories of the Energy Technology Support Unit (ETSU) in Great Britain (Martin and Michaelis, 1992). The ETSU work appears to improve upon the early IPCC work in some respects: it re-estimates the global-warming effect of ground level O_3; it accounts for the effects of CO, NMOCs, and NO_x emissions on CH_4 concentrations; and it distinguishes between emissions of NO_x at ground level and emissions at higher levels. The values for CO, NMOCs, and CH_4 probably do not account for their final oxidation to CO_2.

[d] Johnson and Derwent (1996) use a 2-dimensional zonally averaged model to estimate the GWPs for "step" changes in emissions of CO, NMOCs, CH_4, and NO_x. Their GWPs account for the effect of a gas on the life of methane, and on the concentration of tropospheric ozone, but apparently do not account for the final oxidation of the gas to CO_2. The first NO_x value is for emissions in the Northern Hemisphere; the second, for emissions in the Southern Hemisphere. The range shown for NMOCs is the range they estimate for individual hydrocarbons.

[e] Fuglestvedt et al. (1996) use a two-dimensional photochemistry transport model to study the effects of emissions of CH_4, CO, and NO_x on levels of ozone and methane. The value shown here *excludes* the effect of final oxidation of CO to CO_2. (However, Fuglestvedt et al. do acknowledge this effect and discuss it in their text.) Fuglestvedt et al. (1996) estimate a GWP for NOx emissions from aircraft but not for NOx emissions at ground level.

[f] Derwent et al. (2001) use a global three-dimensional Lagrangian chemistry-transport model, called "STOCHEM," to estimate changes in the tropospheric distribution of methane and ozone following the emissions of short-lived pulses of CH_4, CO, NO_x, and H_2. The value shown here *excludes* the effect of final oxidation of CO to CO_2. (However, Derwent et al. do estimate this effect.). By comparison, the IPCC (2001) says that the greenhouse perturbation of 100 g of CO is equivalent to that of 5 g of CH_4, which implies a GWP (presumably for "indirect effects") of about 1.0. Daniel and Solomon (1998), whose modeling work is discussed in the text here, estimate a 100-year GWP for CO of 1.0 for the effect on CH_4 and 0.0 to 3.4 for the effect on O_3.

The first NO_x value is for emissions in the Northern Hemisphere; the second, for emissions in the Southern Hemisphere. Derwent et al. (2001) also estimate a GWP for NOx emitted from aircraft.

[g] Reilly (1992) bases his estimates on a simplified model of greenhouse gas atmospheric behavior. He assumes that a doubling of trace gas concentrations would cause a welfare loss of $38 billion in the agricultural sector, and that damages to all economic sectors would be six times the agricultural-sector losses. Reilly further assumes that the agricultural damages would be double the amount shown if it were not for the beneficial effect of carbon fertilization. Estimates are shown for three cases: i) economic damages rise linearly with trace gas concentrations; ii) economic damages rise as a quadratic function with trace gas concentrations, and iii) economic damages rise as a quadratic function with trace gas concentrations but CO_2 fertilization causes linear benefits with CO_2 concentration. Given that his “linear damages” GWP for CO is lower than the GWP due to oxidation to CO_2, we infer that none of his estimates account for the effect of final oxidation to CO_2.

[h] Hammitt et al. (1996) base their EDI calculations on a simple climate model, the Integrated Science Assessment Model, and calculate several different indices by varying their climate change and economic damage assumptions. Shown first is the index for the 'middle case', which assumes a 3%/yr. discount rate, a damage function exponent of 2, a 2.5 °C temperature rise with a doubling in trace gas concentrations, the IPCC’s “IS92a” emission/GDP scenario, and an emission year of 1995. Other cases vary one of these assumptions while keeping the others constant. The 'hockey stick' damage function is a function that can be varied from a quadratic damage function to a very convex 'catastrophic' type function -- the one assessed here is quite convex. The 'minimum' and 'maximum' indices are the high and low values for 81 different combinations of input assumptions. The EDIs do not include the indirect effect of CH_4 or halocarbons on H_2O or O_3, or the effects of CO_2 fertilization.

[i] Kandlikar (1996) develops a “trace gas index” based on the ratio of the marginal costs of abatement for a non-CO_2 gas to the marginal cost of abatement for CO_2, noting that at the economic optimum this ratio is the equal to the ratio of the marginal damages. His index is a function of the rate of change of damage per change in the temperature rate, the change in radiative forcing per change in concentration, the change in concentration per unit emission, the discount rate, and the lag between the change in forcing and the change in temperature. He presents results for linear, quadratic, and cubic functions; for discount rates of 0%, 2%, and 6%; and for two IPCC emission scenarios. The results shown here are for the IPCC-A scenario and a 100-year time horizon. Kandlikar (1996) *does* include the indirect effects of CH_4 on H_2O, O_3, and OH.

[j] For emissions between 1995 and 2004, with a discount rate of 3%, no equity weighting, and the IPCC IS92a baseline scenario. Tol (1999) does not account for the indirect effect of CH_4 on H_2O or O_3.

[k] Reilly et al. (2001) estimate EDIs as the relative shadow costs of controlling gases in general-equilibrium cost-benefit analysis. Major determinants of the resulting EDI are the time horizon TH and the annual discount rate r. In the first case shown, the time horizon is 100 years, and the discount rate is constant at 5%. In the second case, the time horizon is 200 year, and the discount rate declines from 5% in year 0 to 0% in year 50, at 0.1%(absolute terms) per year. Reilly et al. (2001) assume that radiative forcing decreases with increasing concentrations (due to saturation of absorption bands) but that damages are linear with radiative forcing.

TABLE D-4. THE CARBON CONENT AND OZONE-FORMING POTENTIAL OF NMOC EMISSIONS FROM FUEL COMBUSTION AND EVAPORATION

	Carbon wt. fraction of NMOCs from[a]:		**Relative O_3 potential of NMOCs from[b]:**		**Total CEF[c]**
	Combustion	*Evaporation*	*Combustion*	*Evaporation*	*Combustion*
Conventional gasoline	0.866	0.830	1.00	0.80	5.7
Reformulated gasoline	0.842	0.830	0.95	0.80	5.5
Diesel oil	0.858	0.858	0.60	0.60	4.6
F-T diesel	0.848	0.848	0.60	0.60	4.6
biodiesel (soy oil)	0.778	0.778	0.50	0.50	4.1
Crude, fuel, and lube oil	0.849	0.849	0.50	0.50	4.4
Methanol (pure)[d]	0.400	0.375	0.40	0.15	2.5
CNG or LNG for LDVs	0.800	calculated	f(NMOC)[e]	calculated	f(NMOC)[e]
Gaseous fuels [f]	0.800	calculated	0.20	calculated	3.4
NG to FTD, MeOH, H_2(g)	0.800	calculated	0.20	calculated	3.4
Coal, coke	0.600	n.a.	0.65	n.a.	3.8
Wood	0.500	n.a.	0.65	n.a.	3.5
Ethanol (pure)[d]	0.550	0.522	0.70	0.40	3.8
LPG	0.800	calculated	0.60	calculated	4.4

	C wt.[h]	**ROFP[h]**	**Total CEF[c]**
ethane	0.866	0.80	5.7
propane	0.842	0.80	5.5
butane	0.858	0.60	4.6
pentanes plus[i]	0.848	0.60	4.6

CEF = CO_2-equivalency factor; F-T diesel = diesel made from natural gas by the Fischer-Tropsch process; LPG = liquefied petroleum gases; n.a. = not applicable.

[a] The carbon weight fraction of NMOC emissions from combustion is based on the discussion in DeLuchi (1993), and my judgment. The carbon weight fraction of NMOC

evaporative emission from liquid fuel is equal to the carbon content of the fuel of the itself, except in the case of gasoline, which is assumed to evaporate mainly the lighter, lower-carbon compounds such as pentane. The carbon weight fraction of leaks of gaseous fuel and LPG is calculated on the basis of the quantity and carbon content of the individual compounds in the gas (the main report).

[b] The ozone-creation potentials for the liquid and solid fuels are relative to that for NMOCs from the combustion of conventional gasoline, and are estimated on the basis of data discussed in the text. The ROFPs for evaporative emissions of ethanol and methanol are the weighted-average MIR (Carter et al., 1998) and POCP (Derwent et al., 1998) values discussed in the text. The ROFPs for leaks of gaseous fuels and of LPG are calculated on the basis of the quantity (the main report) and ROFP (second part of this table) of the individual compounds in the gas.

[c] Calculated as:

$$\text{Total CEF} = CF_i \cdot 3.6641 + ROFP_i \cdot CEF_{O3}$$

where:

Total CEF = total CO_2-equivalency factor

CF_i = the carbon weight fraction of compound i

$ROFP_i$ = the relative ozone creation potential of compound i

CEF_{O3} = the CEF for the ozone-creation effect of NMOCs (TableD-1).

[d] The values shown are for 100% methanol or 100% ethanol. In the model, the values for any mixture of alcohol and gasoline (e.g., 85% methanol, 15% ethanol) is calculated as a weighted average of the values for the alcohol, and for gasoline.

[e] Estimated as a function of the NMOC exhaust emission rate. See the discussion in the text.

[f] Gaseous fuels include raw (out-of-the-ground) natural gas, dry marketed natural gas, refinery gas, and coalbed methane. Hydrogen is not included because it does not produce NMOCs. (Emissions from combustion of lubricating oil in H2 engines use the “crude oil, fuel oil, and lube oil” line.)

[g] Emissions from process areas in plants that use NG as a feedstock to make Fischer-Tropsch diesel (FTD), methanol (MeOH), or hydrogen (H_2).

[h] The carbon weight percentages from the main report.

APPENDIX: DEVELOP OF AND RESULTS FROM A SIMPLIFIED MODEL OF THE CARBON CYCLE

Background

Carbon cycles naturally between three main reservoirs: the atmosphere, the oceans, and the terrestrial biosphere (IPCC, 2001). Atmospheric CO_2 is dissolved in seawater or taken up by plants during photosynthesis. The oceans release CO_2 back to the atmosphere, and plants release their CO_2 back to the atmosphere when their organic matter decays and oxidizes. In the absence of anthropogenic disturbance, the air-sea and air-biosphere exchanges are approximately in equilibrium, and there is no net build up of CO_2 in the atmosphere.

An anthropogenic emission of CO_2 as a result of burning fossil fuel instantaneously increases the amount of carbon in the atmosphere by the full amount of the emission. However, this initial increment to the atmospheric reservoir immediately starts to “decay” via air-sea and air-biosphere exchanges. Moreover, these exchange rates are affected by the increased concentration of CO_2: the initial increase in the atmospheric concentration of CO_2 (due to the anthropogenic fossil-fuel emission) increases the rate of photosynthesis for some plants under some conditions. (This is called CO_2 fertilization.) Eventually the rate of decay will be in equilibrium with photosynthesis, but the total amount of carbon in the terrestrial biosphere in equilibrium will be greater than it was before the anthropogenic emission of CO_2.

A similar process occurs with the air-sea exchange, with the result that in the eventual equilibrium, the oceans will hold more carbon than they did before the anthropogenic emission.

Thus, an initial anthropogenic emission of X grams of carbon increases the atmospheric reservoir of C by X instantaneously, but immediately thereafter the oceanic and terrestrial biosphere reservoirs of C start to increase, eventually reaching a new, greater equilibrium. As this happens, the increment to the atmosphere decreases from X to its final equilibrium value, some fraction of X. From this decay, the lifetime of the incremental or perturbation emission can be estimated. We develop a simple carbon-cycle model to represent this decay process and estimate the perturbation lifetime of CO_2 at different initial atmospheric CO_2 concentrations.

Overview of our model

Our model represents exchanges between three reservoirs – atmosphere, ocean, and terrestrial biosphere – and the effect of anthropogenic emissions on these reservoirs. The uptake of CO_2 by oceans and the biosphere is represented as a function of the atmospheric concentration of CO_2.

The model starts with the amount of carbon in three global reservoirs at the beginning of the year 1850, before significant anthropogenic emissions: 600 Pg-C in the atmosphere; 38,000 Pg-C in the oceans; and 2,000 Pg-C in the terrestrial biosphere (based on IPCC, 2001).

Then, beginning mid-year in 1850 and continuing every mid-year thereafter, the model represents CO_2 fluxes between global reservoirs: fossil-fuel emissions to the atmosphere, land-use and deforestation CO_2 emissions to the atmosphere, emissions of CO_2 from the oceans to the atmosphere, emissions of CO_2 to the atmosphere from decomposition of organic matter in the terrestrial biosphere (apart from that associated with anthropogenic land-use and deforestation changes), uptake of atmospheric CO_2 by the oceans, and uptake of atmospheric CO_2 by the terrestrial biosphere, via photosynthesis (also referred to as net primary productivity).

Global emissions of CO_2 from fossil-fuel combustion and cement manufacture are added to the atmospheric reservoir every mid year. Yearly fossil-fuel emissions from 1850 to 2000 are taken from Marland et al. (2003). Fossil-fuel CO_2 emissions from 2000 to 2100 are from the IPCC's (2001) "AIB" emission scenario (see also the *IPCC Special Report on Emissions Scenarios* [Nakicenovic et al. 2000]). (The IPCC projects emissions every decade; I interpolate between their decadal projections.) I assume that emissions beyond 2100 decrease by 2% per year.

Global emissions of CO_2 from anthropogenic land-use change are subtracted from the terrestrial biosphere reservoir and added to the atmospheric reservoir. Yearly land-use change emissions from 1850 to 2000 are taken from Houghton and Hackler (2000), but reduced by us by 20% because other data in the IPCC (2001) indicate that the Houghton and Hackler (2002) estimates may be too high. Land-use change and deforestation CO_2 emissions from 2000 to 2100 are from the IPCC's (2001) "AIB" emission scenario (we interpolate between their decadal projections. I assume that emissions beyond 2100 decrease by 1.5% per year.

Emissions of carbon (as CO_2) from the ocean in year Y (mid year) are equal to the total oceanic C-CO_2 pool in year Y (beginning of year) divided by turnover time (in years) in year Y of the entire pool. The oceanic C-CO_2 pool in year Y is equal to the pool in year Y-1 plus uptake in year Y-1 less emissions in year Y-1. (The initial value in 1850 is given above). The turnover time in 1850 is equal to the C-CO_2 pool in 1850 divided by the C-CO_2 emissions from the oceans in 1850 (values reported above). I assume that the turnover time decreases by 0.10% for ever 1.0% change in the amount of C-CO_2 in the oceans. The basis for this is the assumption that an increasing concentration of CO_2 in the ocean and an increasing global temperatures (associated with increasing atmospheric CO_2, which in turn is associated with increasing oceanic CO_2) both serve to accelerate the sea-air exchange of CO_2.

Emissions of CO_2 from the terrestrial biosphere (apart from emissions related to anthropogenic land-use change and deforestation) are calculated in the same way as are emissions from the ocean. I assume that the turnover time of carbon in the terrestrial biosphere decreases by 0.10% for ever 1.0% change in the amount of C-CO_2 in the atmosphere, because increasing atmospheric CO_2 increases global temperatures which in turn accelerate decomposition of organic matter[28] (Joos et al., 2001; Xiao et al., 1996,

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[28] The turnover time of carbon in the terrestrial biosphere also will be affected by anthropogenic land-use change and deforestation if the turnover time of carbon in the affected lands would have been different (in

1997; Gerber et al., 2003; Cramer et al., 2001). The dynamic global vegetation model (DGVM) of Gerber et al. (2003) estimates that in increase in global temperature of 2.5° C reduces the turnover time of carbon in litter and soil from about 27 years to about 23 years (about 15%), and has no effect on the turnover time of carbon in vegetation. Assuming that the 2.5° C increase in temperature is associated with a doubling of CO_2 (100% increase), and allowing that there is no effect of temperature on C in vegetation, the results of Gerber et al. (2003) are roughly consistent with our assumption that a 100% increase in the concentration of CO_2 reduces the turnover time of carbon in the terrestrial biosphere by 10%. Similarly, the Terrestrial Ecosystem Model (TEM) of Xaio et al. (1996) assumes that the ecosystem heterotrophic respiration rate R_H (i.e., decomposition) is a function of $e^{0.0693T}$, where T is temperature; this relationship implies that a 2.5° C increase in temperature increases the respiration rate by almost 20%. Six DGVMs run by Cramer et al. (2001) indicate that a doubling of current atmospheric CO_2 causes heterotrophic respiration to increase by 27%. Finally, our assumed parameter value (0.1% per 1.0%) produces results consistent with the those of the sophisticated Bern CC model (See the comparison below of our estimates and Bern CC model estimates of terrestrial biospheric carbon uptake with and without the feedback of concentration on carbon turnover time: in the work of Joos et al. [2001], this feedback reduces uptake by 188 Pg-C from 2000 to 2099; in our model, an elasticity of –0.10 reduces uptake by 190 Pg-C from 2000 to 2099.)

Oceanic and terrestrial biosphere uptake of CO_2 in year Y is calculated by multiplying the uptake in year Y-1 by an elasticity that relates percentage changes in uptake to percentage changes in atmospheric CO_2. The series starts with the 1850 uptake values mentioned above. The percentage change in atmospheric CO_2 is the ratio of the year Y atmospheric CO_2 pool to the year Y-1 pool, and the year Y atmospheric CO_2 pool is equal to the year Y-1 pool plus emissions from fossil-fuels, land-use change, oceans, and the terrestrial biosphere, and less uptake by oceans and the terrestrial biosphere. (The series starts with the 1850 value presented above.)

On the basis of information presented below, the uptake elasticity -- the percentage change in the Pg-C / year uptake per 1.0% change in the Pg-C total burden [concentration] of atmospheric CO_2 (eq. D.20) -- is assumed to be positive (so that an increase the concentration of CO_2 leads to increase in the uptake of CO_2 by oceans and the plants), but also to decline with time. In our simple model, the uptake elasticity for the terrestrial biosphere declines from about 0.35 in 1850 to about 0.22 at twice the year 2000 concentration; the elasticity for the ocean uptake declines from about 0.15 to 0.0.

The uptake elasticity for the oceans is positive because an increase in the concentration of CO_2 increases the partial pressure of CO_2 at the surface of the ocean, which in turn increases the amount of CO_2 dissolved in the ocean (Joos et al., 2001;

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the absence of land-use change and deforestation) from the global average. I ignore this effect, and essentially assume that the turnover time of carbon in anthropogenically affected lands would not have been significantly different from the global average turnover time.

IPCC, 2001). The uptake elasticity declines with increasing CO_2 levels because absorption capacity of the ocean decreases with increasing CO_2 content (IPCC, 2001). As shown below, our elasticity function, combined with our estimate of the change in ocean-C turnover time associated with changes in atmospheric CO_2 concentration (discussed above), results in estimates of ocean C uptake consistent with those of Joos et al. (2001) and the IPCC (2001).

The relationship between changes in the concentration of CO_2 and changes in CO_2 uptake (primary productivity) by the terrestrial biosphere

In regards to the uptake elasticity for the terrestrial biosphere, the information in the literature typically pertains to what is called a "fertilization coefficient," which embodies the relationship between changes in CO_2 concentration and changes in the total stock of plant carbon or net primary productivity:

$$b = \frac{\dfrac{NPP - NPP*}{NPP*}}{\ln \dfrac{C}{C*}} \qquad \text{eq. D.19}$$

where:

β = the fertilization co-efficient
NPP = net primary productivity (Pg-C / yr)
NPP* = NPP at a reference concentration C*
C = the concentration of CO_2 (ppmv)
C* = a reference concentration of CO_2 (ppmv)

By contrast, our uptake elasticity is defined as:

$$b\,\hat{}= \frac{\dfrac{NPP - NPP*}{NPP*}}{\dfrac{C - C*}{C*}} \qquad \text{eq. D.20}$$

For small differences between C and C* (which is what we model here), ln(C / C*) is very close to (C – C*) / C*, and hence $b \approx b\,\hat{}$.

The following summarizes information pertinent to our estimate of the uptake elasticity $b\,\hat{}$.

• Some of the most pertinent data come from the "free-air CO_2 enrichment" (FACE) experiments, which compare the productivity of trees subjected to artificially elevated CO_2 with that of similar trees growing under ambient CO levels (Oren et al., 2001; Davidson and Hirsch, 2001; DeLucia et al., 1999). In these experiments, dozens of trees in outdoor stands are fumigated with CO_2 at 200ppmv above ambient levels during the daylight hours of the growing season (Oren et al., 2001, p. 469). These appear to be among the only, if not the only, actual experimental data taken from a large number of trees of significant size grown outdoors, as opposed to modeling results or experimental data based on saplings in laboratories.

In the FACE experiments, a ~ 56% increase in CO_2 every year caused an increase in annual NPP of 34% over the first four years but an increase of only 6% over the next 4 years (Oren et al., 2001). Oren et al. (2001) attribute the decline in increased productivity to nutrient limitations. In support of this, they found that trees grew much more when supplied with extra nutrients and enhanced CO_2 than with enhanced CO_2 only.

Oren et al. (2001) observe that "CO_2 responses of growth in pine forests will be highly variable and depend greatly on site fertility, perhaps to the point of not responding at all on the nutritionally poorest sites"(p. 470). Following up on this, Davidson and Hirsch (2001) conclude that the FACE results "are the clearest evidence yet that nutrients, and perhaps water, may constraint trees' response to CO_2 fertilization in real forests" (p. 432).

- Cramer et al. (2001) run six different dynamic global vegetation models (DGVMs) to illustrate the effect of CO_2 and climate on ecosystem productivity, and find that NPP increases about 27% when the current concentration of CO_2 doubles (β = 0.27).

- The Bern Carbon Model, used in some of the IPCC TAR climate change scenarios (IPCC, 2001), assumes that the CO_2 fertilization coefficient β is 0.287 (www.climate.unibe.ch/~joos/outgoing/ipcc_sres_jan99/; Joos et al., 2002).

- Alexandrov et al. (2003) state that field studies indicate that the fertilization coefficient β is between 0.35 and 0.60, and that most modelers use values between 0.20 and 0.60. Their own process-based model of NPP and global climate is consistent with a value of β of 0.34 for CO_2 concentrations between 290 and 690 ppmv. However, their model does not consider nitrogen limitation, which they acknowledge might considerably reduce the NPP response to increased CO_2. Alexandrov et al. (2003) also point out that increased growth of plants will tend to close the canopy and block sunlight to lower plants, thereby restricting the ability of plants to increase NPP when CO_2 increases.

- The Terrestrial Ecosystem Model (TEM) of Xiao et al. (1996) is set so that a 100% increase in atmospheric CO_2 causes a 37% increase in plant primary production, *if* nitrogen is not a limiting nutrient. According to Xaio et al. (1996, p. 5), nitrogen availability "down regulates" the response of NPP to increased CO_2 in the TEM. There runs of the TEM and a climate model find that an increase in CO_2 from 315 to 522 ppmv, with associated changes in temperature, cloudiness, and precipitation, causes an 18.5% increase NPP. This indicates an elasticity of 0.28 (0.185/0.65), which is similar to that used in the Bern Carbon Model.

• Xiao et al. (1997) emphasize that the effective “fertilization coefficient” will change over time and space because of changes in climate and nutrient availability and the acclimation of plants. Similarly, Gerber et al. (2003) find that the sensitivity of NPP to atmospheric CO_2 decreases with increasing atmospheric CO_2. (Because of this, our estimate of the uptake elasticity declines with increasing CO_2 concentrations.)

• Studies cited by the IPCC (2001) and Jacobson (2003) indicate that a doubling of the atmospheric concentration of CO_2 would increase carbon stored in vegetation by 20-40%. Jacobson (2003). Given this, and estimates of total carbon stored in vegetation globally, Jacobson calculates that 22% of every increment to the concentration of CO_2 is removed by fertilization.

• Interactions between CO_2, nitrogen, and climate are important determinants of NPP (Alexandrov et al., 2003; House et al., 2003; Gerber et al.,, 2003; Cramer et al., 2001; Oren et al., 2001; Davidson and Hirsch., 2001; Joos et al., 2001; IPCC, 2001; Cao and Woodward, 1998; Xiao et al., 1997;Comins and McMurtrie, 1993). For a variety of reasons, the CO_2 fertilization effect diminishes at high concentrations of CO_2, and may be nil above about 800 ppmv CO_2 (IPCC, 2001, p. 196; Gerber et al., 2003). Cramer et al. (2001) use six DGVMs to illustrate the effect of CO_2 and climate on ecosystem productivity, and find that in all models “the rate of NEP begins to level off around 2030 as a consequence of the ‘diminishing return’ of physiological CO_2 effects at high CO_2 concentrations” (p. 357). Similarly, the DGVM of Gerber et al. (2003) found that the fertilization effect decreases with increasing CO_2 concentrations, and might be further limited by N availability. They found that increasing CO_2 from 280 ppm to 490 ppm (and holding climate variables constant) increased carbon storage in all vegetation, soil, and litter from 2690 Gt to 3608 Gt, implying an elasticity of 0.45. Cao and Woodward (1998) coupled a terrestrial biogeochemical model with an atmospheric general circulation model to study the effect of increasing CO_2 on net ecosystem production, and found that the fertilization coefficient declines as the fertilization effect becomes saturated and is diminished by changes in climatic factors (p. 249). Their model results imply that a doubling of atmospheric CO_2 above pre-industrial levels increases NPP by 27%, implying a fertilization coefficient of 0.27. Finally, Comins and McMurtrie (1993) used models of forest productivity and soil carbon and nutrient dynamics to study how plant productivity is affected by nutrient limitations, and found that an instantaneous doubling of CO_2 leads to an immediate 27% increase in productivity, but that the productivity gain declines to less than 10% within 5 years because of nutrient-cycling limitations. They conclude that in order for the nitrogen-cycling constraint on long-term productivity to be relaxed, either the N:C ratios of plant parts or else the rate of production of litter must be reduced (p. 678).

The modeling results summarized in the previous paragraph, which suggest that nutrient availability may limit the response of plants to enhanced CO_2, have been confirmed by the FACE experiments cited above.

• Increases in photosynthesis spurred by higher CO_2 concentrations can cause a plant to achieve its final mass faster, or to achieve a greater final mass. In the former case, the overall rate of litter production increases, and so the stock of soil carbon increases; in the latter case, both below-ground and above-ground carbon stocks increase. The IPCC (2001, p. 195) cites a source stating that both effects have been observed.

• In the IPCC's (2001) scenarios of future climate change, "the CO_2 fertilization factor is adjusted to give a balanced 1980s mean budget…and the model has been tuned to give results that are similar to those of the Bern-CC and ISAM models for a climate sensitivity of 2.5ºC" (p. 554).

These studies suggest that *b* ^ lies between 0.2 and 0.4, and declines with increasing CO_2 concentrations.

Validating our model

As shown by the following, our simple model produces results consistent with historical data and the results of more sophisticated models:

Parameter	*LEM*	*IPCC (2001)*	*Bern CC and Hilda models (Joos et al., 2001)*	*House et al. (2003)*
Atm. CO_2, 1900 (ppmv)	292	295		
Atm. CO_2, 1960 (ppmv)	306	316		
Atm. CO_2, 1980 (ppmv)	328	337		
Atm. CO_2, 2000 (ppmv)	359	368		
Atm. CO_2, 2050 (ppmv)	513	517 (ISAM model)	522 (standard model set up)	
Atm. CO_2, 2100 (ppmv)	712	717 (ISAM model)	703 (standard model set up)	
Net C uptake by terrestrial biosphere, 1980-1989 (Pg-C / yr.)	2.1	-0.3 to 3.8 (1.9 ref. value)		0.3 to 4.0
Net C uptake by oceans, 1980 - 1989 (Pg-C / yr.)	1.9	1.9 ± 0.6		1.8 ± 0.8
Net C uptake by terrestrial biosphere, 1980 -1999 (Pg-C / yr.)	2.3		1.7	2.6 (ave. of 1980s, 1990s
Net C uptake by oceans, 1980-1999 (Pg-C / yr.)	2.1	1.8 (ave. of 1980s,1990s)	2.1	1.9 (ave. of 1980s, 1990s)
Total C uptake by terrestrial biosphere,	232		282 (standard model set up)	

2000-2099 (Pg-C)		
Total C uptake by oceans, 2000-2099 (Pg-C)	436	418 (standard model set up)
Increase in C uptake by terrestrial biosphere (relative to reference case) when there is no feedback of concentration (climate) to carbon turnover time, 2000 to 2099 (Pg-C)	190	188

All of our estimates but one (net carbon uptake by the terrestrial biosphere from 2000 to 2099) are very close to historical data and estimates by the IPCC (2001), Joos et al. (2001), and House et al. (2003). We estimate a lower C uptake by the terrestrial biosphere because Joos et al. (2001, 2002) assume a constant "fertilization coefficient" (analogous to our uptake elasticity), whereas, as discussed above, I assume one that declines with increasing CO_2.

Using the model to determine the sensitivity to concentration of the decay of an increment of CO_2

We can run this model to determine the decay of a small incremental CO_2 emissions at different concentrations. To do this, we run the model for a reference case, and record the year-by-year ambient CO_2 concentrations over the entire run period, from 1850 to 2350. We will call this the "reference CO_2" series. We then run the model with an additional one-time emission of one Tg of fossil-fuel CO_2 in a specific year. The year-by-year ambient CO_2 concentrations are recorded again, as the "incremental CO_2" series. We take the difference between the incremental and the reference CO_2 series, year by year. This difference is zero up until the time of the incremental emissions, then is 1.0 Tg in the year of the incremental emissions, and then declines after that, although not necessarily monotonically. We do this for incremental emissions in 1901, 1981, 2001, 2021, 2051, and 2101. The results are as follows:

Year of incremental emission →	*1901*	*1981*	*2001*	*2021*	*2051*	*2101*
Time to decay to 37% (yrs.) (e-folding time)	31	>320	>300	>280	>250	>200
Time to decay to 50% (yrs.)	18	31	>300	>280	>250	>200
Time to decay to 65% (yrs.)	10	15	20	>280	>250	>200

As mentioned in the text of this appendix, these results indicate that the decay of an incremental emission of CO_2 is very sensitive to changes in $concentration_2$.