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Abstract

The use of energy accounts for a major fraction of all anthropogenic emissions of greenhouse gases (IPCC, 1995) , and in most industrialized countries the use of transportation fuels and electricity accounts for a major fraction of all energy−related emissions. In the transportation sector alone, emissions of carbon dioxide (CO2) from the production and use of motor−vehicle fuels account for as much as 30% of CO2 emissions from the use of all fossil fuels (DeLuchi, 1991). The production and use of fuels for transportation and electricity also results in emissions of other greenhouse gases, including methane (CH4) and nitrous oxide (N2O). In light of this, and in the face of growing concern about global climate change, analysts have started to evaluated energy strategies for their potential impact on global climate (EIA, 1991a; IEA, 1989; OTA, 1990; Victor, 1992).

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1. INTRODUCTION

The use of energy accounts for a major fraction of all anthropogenic emissions of greenhouse gases (IPCC, 1995), and in most industrialized countries the use of transportation fuels and electricity accounts for a major fraction of all energy-related emissions. In the transportation sector alone, emissions of carbon dioxide (CO_2) from the production and use of motor-vehicle fuels account for as much as 30% of CO_2 emissions from the use of all fossil fuels (DeLuchi, 1991). The production and use of fuels for transportation and electricity also results in emissions of other greenhouse gases, including methane (CH_4) and nitrous oxide (N_2O). In light of this, and in the face of growing concern about global climate change, analysts have started to evaluate energy strategies for their potential impact on global climate (EIA, 1991a; IEA, 1989; OTA, 1990; Victor, 1992). The "Climate Change Action Plan" proposed by President Clinton and Vice President Gore in 1993 calls on the "National Economic Council, the Office on Environmental Policy, and the Office of Science and Technology Policy to co-chair a process...to develop measures to significantly reduce greenhouse gas emissions from personal motor vehicles, including cars and light trucks" (Clinton and Gore, 1993, p. 30).

It is a complex task to evaluate energy strategies for their potential impact on global climate. In the first place, there are many primary energy resources (e.g., fossil fuels, nuclear power, biomass, hydropower, wind power, and direct solar energy), many energy production technologies (e.g., oil refining, biomass gasification and synthesis into liquid fuels, and photovoltaic electricity production), and, in the case of transportation, many energy end-use technologies (e.g., otto-cycle spark-ignition engines, diesel-cycle compression-ignition engines, gas turbines, and electric motors) to consider. In each energy pathway, from the recovery of the primary energy source through energy production to energy end use, there are many sources of greenhouse gases: the use of auxiliary or "process" energy to recover, transport, and produce primary energy and end-use fuels; leaks or releases of greenhouse gases from production fields, pipelines, and soils; and end-use combustion of fuels. Each source can produce several kinds of greenhouse gases: CH_4, N_2O, ozone (O_3) precursors (carbon monoxide [CO], nonmethane hydrocarbons [NMHCs], and nitrogen oxides [NO_x]), and chlorofluorocarbons (CFCs). Finally, each greenhouse gas must be "weighted" according to its relative expected contribution to global warming or the economic damage therefrom.

The intent of this paper is to present and analyze much of the information needed to evaluate the impact of greenhouse-gases other than CO_2 (hereafter referred to as "non-CO_2 greenhouse gases"), for various transportation-fuel and electricity options. Although there are general inventories of global or U.S. emissions of non-CO_2 greenhouse gases (EIA, 1995b; IPCC, 1995; U.S. EPA, 1995c), recent detailed inventories of national emissions of a single non-CO_2 greenhouse gas (U.S. EPA, 1993), and workbooks for estimating emissions of non-CO_2 GHGs (U.S. EPA, 1995b), there is no published detailed analysis of emissions of *all* major non-CO_2 greenhouse gases from *all* emission sources in the production and use of traditional and alternative fuels for transportation fuels and electricity generation.

This paper presents such an analysis. It also shows the contribution of all individual greenhouse gases, including CO_2, to total, fuelcycle, CO_2-equivalent emissions.

The paper focuses on non-CO_2 greenhouse gases because emissions of CO_2 from fuel combustion are easy to estimate: they can be approximated as the carbon content of the fuel multiplied by 3.667 (the ratio of the molecular mass of CO_2 to the molecular mass of carbon), on the assumption that virtually all of the carbon in fuel oxidizes to CO_2 (for data and discussions pertaining to estimating CO2 emissions from energy use see EIA, 1995b; Grubb, 1989; IEA, 1991; Marland and Pippin, 1990; OECD, 1991). In contrast, combustion emissions of all the other greenhouse gases are a function of many complex aspects of combustion dynamics (such as temperature, pressure, and air-to-fuel ratio) and of the type of emission control systems used, and hence cannot be derived from one or two basic characteristics of a fuel. Instead, one must use published emission factors for each combination of fuel, end-use technology, combustion conditions, and emission control system. Likewise, non-combustion emissions of greenhouse gases (for example, gas flared at oil fields, or N_2O produced and emitted from fertilized soils), which can contribute significantly to the overall global warming impact of an energy cycle, also cannot be derived from basic fuel properties, and instead must be measured and estimated source-by-source and gas-by-gas.

The analysis presented here includes a wide range of fuels, feedstocks, and energy-conversion technologies (Table 1). The boundaries of the analysis are drawn widely, to include emissions from the recovery and transport of primary energy feedstocks, the production of fuels from feedstocks, the distribution of fuels to end users, and the end use of fuels in vehicles or power plants. We refer to all these stages together as a "fuel cycle"[1].

To estimate total fuel-cycle emissions of greenhouse gases, we use CO_2-equivalency factors (CEFs) to convert mass emissions of the non-CO_2 greenhouse gases into the mass amount of CO_2 that would have an equivalent climatic or economic impact. Converted emissions plus actual CO_2 emissions are equal to total fuel-cycle CO_2-equivalent emissions. It is important to note that the equivalency factors, while quite useful, also are very uncertain, and may be revised in the future, perhaps substantially. (The implications of uncertainty about the CEFs are discussed more below). The paper is organized by greenhouse gas, beginning with methane.

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[1] A complete fuel and materials cycle also includes emissions from the servicing and maintenance of transport modes, the building of major energy facilities (in the cases where such emissions are likely to be important), and the manufacture of materials for motor vehicles and the assembly of motor vehicles. The complete GHG model used in this analysis actually includes emissions from all these additional stages and activities (DeLuchi, 1991).

2. METHANE (CH_4)

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, 1995). 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, 1995). In 1992-1993, anomolously 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, 1995). 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, 1995). 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.

2.1 Methane as a greenhouse gas

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 (over a 100-year period), although it also has a much shorter lifetime. 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)[2].

The CEFs for CH_4 are shown in Table 2. Note that the "global warming potentials" (GWPs) recently have been 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 is due to two factors: first, a new estimate for the chemical removal rate that is 11% faster than the estimate previously used, and second, the inclusion of CH_4 uptake by soils (IPCC, 1995).

While individual CH_4 sources are not well quantified, most CH_4 is known to come from the anaerobic fermentation of organic matter in rice paddies and swamps, and from the fermentation of mammalian organic excrement (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 following reaction: $CH_4 + OH^\bullet \dashrightarrow CH_3^\bullet + H_2O$. There is a positive feedback loop for accumulations of CH_4 in

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[2] However, because CH_4 is much less reactive than are most other hydrocarbons, it contributes less to ozone formation than do other hydrocarbons.

the atmosphere as additions of CH_4 to the troposphere can deplete OH levels, thereby reducing the rate of CH_4 removal (IPCC, 1995). Increases in carbon monoxide levels can also deplete OH (via $2CO + 2OH^\bullet \rightarrow 2CO_2 + H_2$), 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, 1995).

2.2 Methane emissions from motor vehicles

Methane is emitted from gasoline, diesel, methanol, ethanol, LPG, and natural gas internal-combustion-engine (ICE) vehicles. Methane emissions (in grams per mile, g/mi) are a function of the type of fuel used, how the engine is designed and tuned the type of emission control system, the age of the vehicle, and other factors. Table 3 is a compilation of reported measurements of CH_4 emissions from petroleum- and alternative-fuel vehicles (AFVs), along with the relevant key attributes of the vehicles.

2.2.1 Gasoline LDVs Emissions by model year. The data of Table 3 show that gasoline light-duty vehicles (LDVs) with 3-way catalytic converters (which oxidize CO and NMHCs to CO_2 and H_2O and reduce NO_x to N_2 and O_2) emit between 0.02 and 0.2 g/mi CH_4, with values for recent model-year cars centering around 0.08-0.10 g/mi. The U.S. Environmental Protection Agency's (EPA's) emission-factor computer model, MOBILE, which estimates on-road emissions from a complete fleet of motor vehicles, in any given year, gives a similar range for CH_4 emissions. The emissions database used by EPA to develop an early version of the emissions model, MOBILE3, indicates that gasoline LDVs emit 0.1 g/mi at low-altitude (Chun, 1988; U.S. EPA, 1985). MOBILE5, the most recent version of the model, estimates that the LDV fleet will emit 0.12 g/mi in the year 1990, 0.06 g/mi in the year 2000, and 0.04 g/mi in the year 2020 (Table 3). (The model projects a decline in emissions because beginning in 1994 vehicles will have to meet the lower NMHC emission standards called for in the Clean Air Act Amendments of 1990 (U.S. EPA, 1991), and the technology used to control NMHC emissions will to some extent control CH_4 emissions.)

Relation to NMHC emissions. Methane emissions per se are not regulated in the U.S. The systems used to control emissions of NMHCs and total hydrocarbons (THCs) do to some extent control CH_4 emissions, but not as much as they do NMHCs, because CH_4 is difficult to oxidize catalytically. Thus, whereas controlled NMHC emissions can be an order of magnitude less than uncontrolled emissions, CH_4 emissions from vehicles with HC controls might be about 3 times less than CH_4 emissions from vehicles with no controls. The EPA's study for MOBILE3 found that vehicles without a catalytic converter emit 0.3 g/mi CH_4, compared with 0.1 g/mi for vehicles equipped with a catalytic converter. Thus, methane emissions are larger fraction of NMHC emissions from older, high-emitting cars than from new, tightly controlled cars. Because of this, we do not estimate methane emissions as a fraction of NMHC emissions.

Catalyst age. One would expect methane emissions to increase somewhat as the engine and the emission-control system age and deteriorate. The data of Table 3 do suggest that for most fuels -- nonpetroleum fuels as well as petroleum fuels -- CH_4 emissions increase with the age of the catalyst[3]. The few tests that couple modern vehicles and fuels (i.e. 1992 vans using reformulated gasoline) show emission levels of about 0.05 g/mi when new, rising to about 0.15 g/mi with significant catalyst age. Older three-way catalyst equipped vehicles exhibit somewhat higher rates, ranging from perhaps 0.1 g/mi when new to 0.3 g/mi or more when older. We note, however, that there are virtually no data on methane emissions from very old vehicles.

On the basis of the data of Table 3, we assume that CH_4 emissions increase at the rate of 0.0015 g/mi per thousand miles of catalyst, so that at 100,000 miles, emissions are 0.15 g/mi higher than when the vehicle is new.

Vehicle efficiency. It is worth noting that data (not shown here) on the efficiency of the vehicles in Table 3 indicate that CH_4 emissions are not a function of vehicle efficiency. This latter finding is not particularly surprising, because tailpipe emissions in general are not strongly related to fuel economy (because the standards are in terms of grams per mile, not grams per energy unit) (DeLuchi, et al., 1992).

Ambient temperature. Methane emissions, like NMHC emissions, appear to be higher at lower ambient temperatures (Stump, et al., 1989; 1990), because before the engine is warmed up the temperature of the fuel going into the engine is close to the ambient temperature, and at lower temperatures the fuel does not vaporize as completely, and hence does not burn as completely.

Drive cycle. Recent measurements show that g/mi methane emissions are higher in "bag 1" (cold-start mode) of the Federal Test Procedure (FTP) than in the other bags, and higher over the whole FTP cycle than over a high-speed, high-power cycle called the REP05 (Auto/Oil, 1996).

Because of deficiencies in the MOBILE emissions model, these two "drive-cycle" effects (higher emissions in FTP bag-1 than in other bags, and higher emissions over the FTP than the REP05 cycle) suggest that MOBILE's estimates of methane emissions might be in error. First, the MOBILE model overestimates the average trip length and hence underestimates the average fraction of time spent in the cold-start mode, when emissions are highest (German, 1995; U.S. EPA, 1995d). Because of this, MOBILE tends to underestimate drive-cycle methane emissions. But the MOBILE model also in effect assumes that vehicles drive slower and accelerate less rapidly than they actually do (Ross, et al., 1995). Because of this, MOBILE tends to overestimate methane emissions, which as noted above are lower in high-speed, high-power driving. In the absence of actual analysis, we assume that these two drive-cycle effects cancel.

Our assumptions. On the basis of the estimates of MOBILE5, and the data of Table 3, we assume that the zero-mile methane emission rate decreases over time,

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[3] On the other hand, the EPA's (1985) analysis for MOBILE3 indicated that CH_4 emissions from gasoline vehicles did *not* deteriorate with age. However, the EPA's findings can be downweighted because they do not include current vehicles.

as emission standards become tighter, but that for any given model year, methane emissions rise slowly with the age of the catalyst. Formally, we estimate methane emissions in a target year with the following function:

$$EM_{TY} = ZM_{BMY} \cdot \left(1 + \frac{DZ}{100}\right)^{MY-BMY} + DA \cdot \frac{MI_{TY}}{1000}$$

$$MY = TY - AGE$$

$$AGE = f(AMS, MI_{TY})$$

where:

EM_{TY} = emissions from gasoline LDVs in target year TY (g/mi)

ZM_{BMY} = zero-mile emissions from a base-model-year vehicle (0.08 g/mi in 1993; Table 3)

DZ = the annual percentage change in the zero-mile emission rate (-3.5%/year; based on our analysis of MOBILE5 estimates)

MY = model year of the vehicle (calculated on the basis of the vehicle mileage in the target year)

BMY = base model year for setting emission factors (1993)

DA = the deterioration rate in emissions with vehicle mile (0.0015 g/mi per 1000 miles; see discussion above)

MI_{TY} = total mileage on the vehicle in the target year TY (miles)

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

AGE = the age of the vehicle (years)

AMS = the annual mileage accumulation schedule (U.S. EPA, 1993; U.S. EPA, 1985)

*2.2.2 **Natural-gas LDVs*** Because CH_4 is the primary component of natural gas, one would expect that vehicles using natural gas would emit considerably more CH_4 than do gasoline LDVs. The data of Table 3 confirm this, showing that CH_4 emissions from natural-gas vehicles (NGVs) range from 0.6 to 4 g/mi for dual-fuel vehicles (which carry and use two fuels, gasoline and natural gas), and between 0.4 and 3 g/mi for dedicated vehicles (which carry and use only natural gas).

Vehicle technology. Most of the NGVs of Table 3 are retrofitted or rebuilt gasoline vehicles. Only one set of vehicles, three 1992 Dodge 5.2 liter V8 vans, has been completely designed and built for maximum performance and lowest emissions on natural gas (Battelle, 1995). The much lower emissions of these optimized Dodge vans, relative to older NGVs and similar model but non-optimized NGV vans from other manufacturers, suggest that CH_4 emissions from future, advanced NGVs will likely be under 0.5 g/mi for new vehicles, and perhaps around 1.0 g/mi for vehicles with some catalyst age. The need to meet relatively tight NMHC standards, such as the "ultra-low-emission-vehicle" (ULEV) standard promulgated by the California Air Resources Board (*California Code of Regulations*, Title 13, Section 1960.1) also may reduce CH_4 emissions somewhat, although the need to meet tight NO_X standards might require a fuel-rich air/fuel mixture (to allow the reduction catalyst to reduce NO_X emissions), which would tend to increase emissions of unburned fuel. The cleanest NGVs tested to date, the optimized Dodge vans mentioned above, have exceptionally low emissions. Methane emissions averaged 0.44 g/mi averaged over three vehicles, and as low as 0.28 g/mi in one test (Battelle, 1995).

Age of catalyst. Methane emissions from NGVs, like methane emissions from gasoline vehicles, increase with the age of the catalyst. This is best shown by the Battelle (1995) data from tests of nine 1992 CNG vans from three manufacturers. This is one of the few controlled studies with repeated tests of the same vehicles at different mileage intervals, and the data collected show a consistent increase in emissions from 5,000 to 15,000 miles and from 15,000 to 25,000 miles. Unfortunately, no data are available yet regarding emissions at very high mileage.

Ambient temperature and drive cycle. Whereas ambient temperature does influence CH_4 emissions from gasoline LDVs, it does not strongly influence CH_4 emissions from NGVs (Gabele, et al., 1990a), mainly because CH_4 is a gas at all ambient temperatures and hence does not have to be vaporized -- a process that is dependent on temperature. However, the recent tests by the Auto/Oil Program (1996) do show that methane emissions from NGVs depend on the drive cycle, in the same way that methane emissions from gasoline vehicles do: methane emissions are somewhat higher in "bag 1" of the FTP than in the other bags, and considerably higher over the whole FTP cycle than over the high-speed, high-power REP05 cycle (Table 3).

Fuel composition. One might expect that methane emissions from NGVs would be related to the methane content of the natural gas. However, tests by the Auto/Oil Program (1996) suggest that there is no strong relationship between methane content and methane emissions:

methane content of natural gas	86%	90%	94%	97%
methane emissions from vehicles (REP05 cycle/FTP cycle)	0.47/0.91	0.50/0.93	0.48/0.96	0.49/0.92

Summary. The data of Table 3, and considerations discussed above, suggest that methane emissions from NGVs, like methane emissions from gasoline vehicles, decrease with model year (later models emit less) and increase with vehicle mileage, and generally are about an order of magnitude higher than methane emissions from gasoline vehicles of similar technology and age. Therefore, we assume that methane emissions from NGVs are 10 times methane emissions from gasoline vehicles.

2.2.3 Methanol LDVs Methanol LDVs vehicles definitely emit less CH_4 than do comparable gasoline vehicles. Table 3 shows that dedicated methanol vehicles generally emit less CH_4 than do dedicated gasoline vehicles of the same model, and Table 4 shows that CH_4 emissions from "flexible-fuel vehicles" (FFVs), which can use any mixture of gasoline and methanol, tend to decrease when the gasoline content of the fuel mixture is decreased. Also, the upper end of reported CH_4

emissions from methanol LDVs is not as high as the upper end for gasoline LDVs. Taken together, the data suggest that dedicated M100 vehicles emit about half as much CH_4 as dedicated gasoline vehicles, and M85 vehicles (which use a mixture of 85% methanol and 15% gasoline) about two-thirds as much. Data plots of emissions of methanol dedicated and dual fuel LDVs as a function of catalyst age show slowly rising emissions with increased age, but with very few data points from older vehicles. Based on these data, the model used here assumes that CH_4 emissions from gasoline/methanol mixtures are equal to the M100 emission rate (which is half the gasoline emission rate) multiplied by the methanol fraction, plus the gasoline emission rate multiplied by the gasoline fraction of the mixture[4].

2.2.4 Ethanol LDVs There have been only a handful of recent emission tests of late-model ethanol, LPG, and hydrogen vehicles. The California Air Resources Board (CARB) (1991) has tested one of its Flexible-Fuel Crown Victorias (FFVs) -- which are designed to run on any mixture of alcohol and gasoline, but are optimized for *methanol* and gasoline -- on 85% and 95% ethanol (Table 3). As shown, the FFV emitted a relatively large amount of CH_4 when it was run on ethanol -- much more than it did when it was run on methanol or gasoline. However, as noted above, the vehicle was not designed to burn ethanol, and CARB is not confident of the results. It probably is best to assume that CH_4 emissions from ethanol vehicles are similar to CH_4 emissions from methanol vehicles, because ethanol is similar to methanol, rather than to use what probably are unrepresentative test results.

2.2.5 Liquefied petroleum gas (LPG) LDVs The data of Table 3 indicate that LPG vehicles emit about as much CH_4 as do gasoline vehicles. This is not entirely unexpected, because the species profile of HC emissions from any vehicle tends to reflect the HC composition of the fuel[5], and LPG is mostly propane (C_3H_8), which is similar in many respects to the main components of gasoline (e.g., octane, C_8H_{18}). Although LPG, like natural gas, is a gaseous fuel, it does not contain CH_4, and hence would not be expected to produce as much CH_4 as does natural gas (which typically is at least 90% CH_4). Similarly, LPG is not an alcohol and does not have the properties responsible for the relatively low CH_4 emissions of methanol vehicles. Thus, we use the MOBILE5 estimates of emission rates of gasoline LDVs from 1990-2020 for LPG LDVs.

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[4] Note that it is important to compare same or similar methanol and gasoline vehicles. As shown in Table 3, methane emissions from both gasoline and methanol vehicles can range from near zero to over 0.1 g/mi, and so if one picks randomly a few test results for gasoline vehicles and a few for methanol, one may find that methanol vehicles emits a lot more methane than do gasoline, or vice-versa. But, because automobile manufacturers will build either a gasoline vehicle or a methanol version of the *same* vehicle, depending on economics, environmental regulations, and so on, the correct emissions comparison is between vehicles with the same basic engine, fuel injection, mileage, emission control equipment, vehicle weight, and so on.

[5] The main component of the organic emission from any vehicle is unburned fuel: gasoline components from gasoline vehicles, methane from NGVs, methanol from methanol vehicles, propane from LPG vehicles, and so on .

2.2.6 Hydrogen LDVs Theoretically, hydrogen vehicles could emit trace amounts of CH_4 from the combustion of lubricating oil. However, CARB (1989) found no CH_4 in the oil-related HCs from a hydrogen truck, even though the vehicle burned an unexpectedly large amount of oil. Therefore, one probably can assume that hydrogen vehicles do not emit any CH_4.

2.2.7 Heavy-duty vehicles (HDVs) Table 3 shows the results of several emission tests of gasoline and diesel-fueled HDVs, and compares the results with the CH_4 emissions estimated by MOBILE5. Compared to the test results reported here, MOBILE5 predicts higher CH_4 emissions from diesel HDVs, and lower CH_4 emissions from gasoline HDVs. However, because the MOBILE5 emissions factors are based on a larger data base, it probably is better to use the results of MOBLE5, rather than the emission tests shown in Table 3.

Table 3 also shows CH_4 emissions from methanol and natural-gas HDVs. In order to meet the stringent 1994 NO_x emission standard for HDVs, natural-gas HDVs either will use lean-burn engines, or three-way catalytic converters. According to the few tests conducted so far (Table 3), such vehicles probably will emit between 2 and 4 g/mi CH_4. Unfortunately, the few CH_4 emissions data for methanol HDVs cover a wide range, from near zero to over 1 g/mi. Because there are so few emissions results for methanol HDVs, and because what few there are do not agree, it probably is better to ignore the data and assume instead that, just as methanol LDVs emit less CH_4 than do gasoline LDVs, methanol HDVs emit less CH_4 than do diesel HDVs (we assume half as much).

2.3 *Natural gas losses from production, transmission, and distribution*

Small amounts of natural gas are lost from production fields, transmission lines, and distribution systems. Natural gas leaks from joints and compressor stations throughout the natural-gas system. Gas also leaks, or is vented, from instruments that operate on gas, from valves opened to drain liquids from pipelines, and on occasion from overpressure valves[6]. Gas may be vented from buildings during construction, or emitted during the purging of pipelines (AGA, 1989a). (Intentional venting probably accounts for a minority of losses.) These emissions of natural gas contribute to global warming because natural gas comprises mainly CH_4, a radiatively active greenhouse gas.

In the past few years, as part of efforts to account for sources and sinks of greenhouse gases, several organizations and researchers have estimated leakage from natural gas systems.[7] Many of these are summarized in Table 5. Most of the

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[6] Some gas permeates through the walls of plastic pipes, but the rate, 0.26 ft^3/day-mile (Spriggs, 1988), is insignificant.

[7] The Energy Information Administration publishes estimates of "unaccounted for" gas, but this is an account-balancing item, not an estimate of actual gas losses to the atmosphere. "Unaccounted for" gas is the imbalance between the sum of the components of gas supply and the sum of the components of gas disposition. The imbalance which is created by variations in reporting practices, metering, meter adjusting, and other factors, including gas losses. Because it is strictly an accounting term, unaccounted for" gas can be and actually has been *negative* (EIA, 1989c; this simply means that the estimated

studies done to date agree on two key points. First, that total gas loss to the atmosphere from wellhead to end-use consumer, in modern, well-maintained systems, is probably less than 2% of total gas throughput.[8] Second, losses in old or poorly maintained systems can be quite a bit higher. For example, Hogan et al. (1991) believe that leakage in the [former] Soviet Union is about 6%,[9] and estimates in Mitchell et al. (1990) indicate even higher loss rates for older systems in Britain designed originally to carry city gas (which is mostly carbon monoxide and hydrogen) rather than natural gas (which is mostly CH4). There is disagreement, however, on the total leakage from existing national and regional systems, which comprise parts and subsystems of varying ages and quality. As indicated in Table 5 most gas companies believe that total losses are relatively low, but others (e.g., Mitchell, et al., 1990) believe that the older parts of the gas networks can raise the average loss rate substantially. Generally, there is little reliable information on not only leaks from older systems, but the amount of gas carried in older systems.

Table 5 summarizes several recent estimates of gas loss from production, transmission, and distribution systems. The U.S. EPA study (1993) for the U.S. Congress estimated emission factors for "model," or representative facilities at each stage of the natural gas system (for example, natural gas processing plants, or transmission pipelines), then multiplied these emission factors by "activity factors," (e.g., gas throughput, or miles of pipeline), and aggregated the results for the entire U.S. system. They also made emission projections for the years 2000 and 2010.[10] The results of the study, which are broadly consistent with other recent (but generally less disaggregated studies), also are shown in Table 5.

However, a recently completed, multi-year, detailed study by Radian Corporation, for the Gas Research Institute (GRI) and the EPA (U. S. EPA/GRI, 1996)

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disposition exceeded estimated supply). Obviously, the amount of gas actually lost to the atmosphere cannot be negative.

[8] See Table 5. Staff at the U. S. Energy information Administration (EIA) believe that gas leaks are less than 0.5% total of total deliveries (McCarrick, 1990). The International Workshop on Methane Emissions from Natural Gas Systems, Coal Mining, and Waste Management Systems (1990) comes to essentially the same conclusion.

[9] However, the Alphatania Group (1989) believes that only 0.5% to 1.2% of gas throughput leaks from the gas systems in the former Soviet Union.

[10] In the evaluation of energy policy, we are interested in the greenhouse consequences of future energy choices, which means that we must estimate not where and how gas currently is produced and distributed, but rather where and how *extra* gas demanded as a result of the new policy will be produced and distributed. As Mitchell et al. (1990) note, "...it is clearly still necessary to determine exactly what sort of leakage rate is applicable to marginal increases (or decreases) in gas supply" (p. 817). On the one hand, increased gas consumption may result in higher supply pressures, which will tend to increase leakage. On the other, increased gas supply may have dedicated new lines, which will have very low leakage rates. To have reasonably precise estimates, one would have to evaluate specific policies for specific systems. The U.S. EPA (1993) does try to consider how some of the factors that affect gas emissions might change in the future. For example, they assume that plastic pipes, which have a relatively low leakage rate, will continue to replace steel pipes in distribution systems.

estimates somewhat higher leakage rates than do earlier studies (Table 5). Radian concludes that CH4 emissions from the NG industry were 307 BCF in 1992 (U.S. EPA/GRI, 1996). This figure corresponds to about 1.4% of gross NG production, and suggests that NG systems contributed about 19% to the total anthropogenic CH4 emissions in the U.S. in 1992 (U.S. EPA/GRI, 1996). The study separately estimates "fugitive," "vented," and "combusted" gas from the "production," "gas processing," "transmission and storage" and "distribution" segments of the fuelcycle. The study examines individual sources of emissions in each category, such as pneumatic devices, dehydrator glycol pumps, and "blow and purge" as important sources of vented CH4 emissions. The study concludes that about 48% of total NG industry emissions are the result of fugitive losses during the transmission and storage segment and the distribution segment (U.S. EPA/GRI, 1996). Each of these two categories accounted for nearly 75 BCF of CH4 emitted in 1992, and the only other category with comparably high emissions was identified as the 47 BCF of CH4 vented during the production segment (U.S. EPA/GRI, 1996). See Table 5 and note for a complete summary of the results of this study.

*2.3.1 **Assumptions of CH_4 emissions from the natural gas industry*** The greenhouse-gas emissions model used here disaggregates NG losses into losses from gas production (including gas lifting, gathering, and processing), losses from transmission systems, and losses from distribution systems. We use the U.S. EPA and GRI's (1996) recent estimates of loss rates at these three stages, for a base year of 1992.[11] All three of these kinds of losses -- from production, transmission, and distribution systems -- result from the use of NG for motor vehicles, heating, and cooking. Production and transmission losses, but not distribution losses, result from the use of NG to make methanol or generate electricity, because large gas-using facilities such as methanol plants and power plants often are located near to gas-production fields and typically are connected to large-volume transmission lines.

We assume that the baseline EPA/GRI leakage rates (for 1992) decline slightly (-0.5%/year for distribution systems, -0.25%/year for transmission and production, in relative terms) on the presumption that new systems leak less than the 1992 average system.

The estimates of Table 5, including the U.S.EPA/GRI (1996) estimates, do not include emissions from venting or flaring of NG at gas-producing wells, or from refueling stations that provide compressed or liquefied gas to NGVs. Both of these sources are discussed next.

*2.3.2 **Venting or flaring of natural gas at gas-producing wells*** If the gas extracted from an oil or gas well cannot be marketed, it must be reinjected, vented to the atmosphere, or flared. This typically occurs at oil wells where some gas is drawn up with the oil, but there is no market for the gas. Greenhouse-gas emissions from

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[11] We aggregate the categories gas "production" and "processing," as reported by U.S. EPA/GRI, 1996, into one category that we call production. Also, as indicated in the notes to Table 5, we exclude from the leakage rates emissions of methane from incomplete combustion of natural gas used as a fuel by pipeline compressor engines or natural-gas processing plants. These incomplete-combustion emissions are accounted for the by the CH_4 emission factors for use the use of natural gas as a process fuel in natural-gas engines (Table 12).

venting or flaring at these oil wells should be assigned to the production and use of oil, not to the use of natural gas, because oil is the only product.

However, there apparently is a small amount of venting or flaring (probably not routine) at wells that produce marketable gas. Greenhouse gas emissions from this venting or flaring should be estimated and assigned to the use of natural gas. Such venting and flaring apparently is not included in the loss estimates of Table 5; for example, the EPA's (1993)(1993) estimate of CH_4 emissions attributable to the NG system includes "routine venting associated with the operation of equipment in the production, processing, transmission, and distribution of gas" (p. 7-14), and "system upsets," such as emergency blowdowns, but not venting or flaring of gas in the field. Hence, we must determine if these emissions are important.

Data from the U.S. Minerals Management Service (MMS) (Nixdorff, 1991) indicate that 0.04% of the gas produced from Federal offshore gas wells in the Gulf of Mexico is vented or flared. Most of this "vented or flared" gas is likely to have been flared. Similarly, information in *Options for Reducing Greenhouse Gas Emissions* (1990) indicates that 0.01% of total production is flared, and that no more than 0.005% of total gas production in the U.S. is vented. A venting rate of 0.005% is two orders of magnitude smaller than the NG leakage rate from pipelines and distribution systems, and a flaring rate of 0.04% is two orders of magnitude smaller than the percent of natural gas used as a pipeline fuel (NG used as pipeline fuel has the same effect on global warming as NG burned in a flare). Hence, venting and flaring at gas-producing wells can be ignored.

2.3.3 NG leaks from compressors, liquefiers, and storage systems. In the case of motor-vehicle use of natural gas, one also must account for gas losses from compressors or liquefiers and refueling lines at NG refueling stations. Unfortunately, there are no data on gas leaks at CNG stations. Clearly, though, gas does leak at service stations: one can smell the gas, especially after the compressor turns on. Gas probably leaks from the station compressor. When the refueling nozzle is disconnected from the vehicle, compressed gas does escape from the short space between the check valve on the refueling nozzle and the actual end of the nozzle. The magnitude of these and any other leaks is not known. (It is unlikely that gas leaks from static storage tanks, lines, and joints.) We assume that the leakage rate from CNG stations is similar to the leakage rate from distribution systems: about 0.2% of total throughput.

Leakage from a properly functioning LNG station should be minimal. LNG dispensers are fully automatic and self-sealing, and have a vapor return line that sends vaporized fuel back to the liquefier or gas pipeline. We assume that there is no significant gas leakage from leakage from LNG stations.

If an LNG vehicle is idle for one to three weeks, vaporized LNG will begin to be vented from vehicular storage tanks (DeLuchi, et al., 1987). Tanks vent at about 14 g/hour. However, if the vehicle is driven at least once a week -- and most vehicles are -- the tank will draw off the vaporized LNG and will not vent. Consequently, we assume that very little LNG is lost to boil off. (See Appendix B of DeLuchi, 1991, for more discussion.) We assume also that CNG tanks and fuel systems on board vehicles do not leak any gas.

2.3.4 The composition of NG, and its relevance to global warming Pipeline-quality NG contains CO_2 and NMHCs, as well as CH_4. These greenhouse gases have different warming potentials per gram, and consequently the precise total warming effect of gas leaks depends on the proportions of these individual compounds in the gas. We assume the composition of pipeline gas estimated in Appendix C of DeLuchi (1991), and apply separate CO_2-equivalency factors (see below) to each gas to calculate the total CO_2-equivalent emissions of greenhouse gases.

2.3.5 Synthetic natural gas (SNG) systems A synthetic natural gas (SNG) can be manufactured from coal or biomass, and used in most natural-gas engines, appliances, etc. This analysis examines greenhouse-gas emissions from the use of biomass-derived SNG in motor vehicles.

SNG from biomass is composed of H_2, CO_2, CO, CH_4, and small amounts of NMHCs (Feldman, et al., 1988; Flanigan, et al., 1988). The total climatic impact of leaks of SNG is equal to the sum of the CO_2-equivalent of each of the components of SNG, less a CO_2 credit for each mole of carbon emitted in any compound. This credit is given because all of the carbon in SNG comes originally from atmospheric CO_2, via photosynthesis. For this analysis, we assume a composition of 20% H_2, 20% CO_2, 40% CO, 13% CH_4, and 7% NMHCs (Flanigan, et al., 1988). We also assume that the pipelines that will deliver this medium-Btu SNG will leak at the same rate as the pipelines that deliver fossil NG.

2.4 Vented and flared associated gas attributable to oil production

Many fields contain both natural gas and crude oil. Some of these fields contain mostly crude oil, and are developed in order to sell the oil. When the oil at these fields is produced, small amounts of the associated natural gas are produced also. If the gas cannot be collected and sold economically, it must be disposed of somehow. There are three ways to dispose of unmarketable associated gas: reinject it into the producing field, burn it (called "flaring"), or simply vent it to the atmosphere. Reinjected gas does not enter the atmosphere and so is of no concern in an analysis of emissions of greenhouse gases. However, venting the gas releases it in its original state, as CH_4, higher alkanes, CO_2, N_2, and H_2S, and flaring the gas produces CO_2 and unburned hydrocarbons, including some CH_4[12]. As discussed above, the emissions of CO_2, CH_4, and other gases that result from the venting and flaring of associated gas should be assigned to the use of petroleum fuels.

Several sources, including the United Nations, the Organization of Petroleum-Exporting Countries, the energy agencies of national governments, and major international gas companies, estimate the amount of gas vented and flared locally, regionally, and worldwide. The U.S. Energy Information Administration (EIA) analyzes the quality of these data and publishes its "best-estimates" (EIA, 1995a; EIA, 1994a; EIA, 1992b; EIA, 1992a; EIA, 1991). From 1983 to 1992, 3 to 4 trillion cubic feet

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[12] The composition of raw, in-the-ground gas can be estimated if one knows the composition of pipeline gas (reported in Weaver [1989]), and the amount and composition of nonhydrocarbon gases (e.g., CO_2 and H_2S) and natural-gas liquids (e.g., ethane, propane, and butane) removed in the processing of raw natural gas (reported in EIA [1989d]).

(TCF) of gas were vented or flared worldwide every year (EIA, various years). In 1983, 3.85 TCF of gas -- about 7% of dry gas production -- were vented or flared, and in 1992 the figure was 3.83 TCF, or 5.1% of dry gas production (EIA, 1994a). It appears that some of the previously vented and flared gas now is being reinjected: the quantity of gas reinjected has increased from 5.41 TCF in 1983 to 8.90 TCF in 1992. Worldwide, venting and flaring probably will decline as natural gas increases in value and is reinjected, used domestically, or exported. The United Nations projects that in the year 2010 venting and flaring will be half of what it was in 1987 (United Nations, n.d.)[13]. Major gas-producing developing countries are expected to use associated gas to help fuel industrial development (ICF, 1990a; IEA, 1989)

The EIA data indicate that the amount of venting or flaring varies considerably around the world. In order to estimate venting or flaring emissions attributable to U.S. *consumption* of petroleum (as opposed to U.S. *production* of crude oil), one must know the rate of venting or flaring in the all the countries that produce oil used directly (as crude oil) or indirectly (as petroleum products) by the U.S. One way to calculate this rate is to divide the world into regions, estimate the amount of gas vented or flared per unit of oil produced in each region, and multiply each of the regional rates by the amount of oil that the U.S. gets, directly or indirectly from the region. Table 6 shows the results of such an analysis for the year 1992. The estimates of Table 6 also separate flaring from venting[14], a separation which is discussed below.

2.4.1 Emissions of vented or flared gas not included in the EIA statistics The EIA collects its venting and flaring data from state agencies in the U.S., and from foreign governments. The state offices report venting and flaring from all onshore oil wells, and venting and flaring from State but not Federal offshore oil wells. The EIA's estimates of venting and flaring in the U.S. are based entirely on the state data[15], and therefore do not include venting and flaring from Federal offshore oil wells (McCarrick, 1990).

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[13] For example, Nigeria (West Africa), the largest flarer of gas in the world, "has stated policies encouraging alternatives to flaring," including using the gas domestically or exporting it. Similarly, Algeria (Mediterranean) is planning to increase its reinjection of gas to enhance oil recovery (ICF, 1990a).

[14] It is possible that the EIA's estimates of venting or flaring, which we assign to oil production, actually include venting or flaring at wells that produce marketable gas as well as oil. (It is clear that the EIA's estimates do not include venting or flaring at wells that produce *only* gas, because the EIA *defines* vented or flared gas as associated gas, and detailed state-by-state breakdowns in the EIA's *Natural Gas Annual* (1989d) data show that no gas was vented or flared in states that produced *only* natural gas.) In this analysis, venting or flaring at gas-producing wells already has been assigned to natural-gas use, and hence should not be assigned to oil use. However, the amount undoubtedly is negligible, because as discussed above venting or flaring at all gas-producing wells is negligible, and venting or flaring at wells that produce both oil and gas must be less than venting or flaring at all gas-producing wells.

[15] The state data may have serious shortcomings: they apparently are based not on measurements or engineering calculations of the amount of gas actually vented or flared, but rather on estimates of unaccounted for gas or of some other quantity (Harrison, 1992). If this is true, then amount of gas

The EIA estimates can be supplemented with data from the MMS on venting and flaring from Federal offshore oil wells. In 1990, in the Gulf of Mexico, 48 SCF of gas were vented or flared from oil wells per barrel of crude oil produced (Nixdorff, 1991). Since production from Federal offshore oil wells in the Gulf of Mexico typically is more than 90% of total Federal offshore oil production (Minerals Management Service, 1992), one can assume that this 48 SCF/bbl rate applied to all Federal offshore oil wells in 1990. From 1970 to 1991, Federal offshore production was 10% to 12% of total U. S. oil production (Minerals Management Service, 1992). With these two statistics -- the amount of gas vented or flared per bbl of offshore production, and the offshore production as a fraction of total production -- we can calculate total venting and flaring from offshore oil production. We have included this amount in Table 6.

It is possible, but not likely, that a large amount of associated gas is vented underwater and not reported. Sackett and Barber (1988) state that in the 1970s, it was common to vent natural gas underwater at offshore oil-producing platforms. Sea Technology (1974) and Brooks et al. (1977) cite large estimates of vented and flared gas in 1973 and 1974 (about 150 SCF/bbl), and Brooks et al. (1977) argue that most of this was vented underwater. However, the MMS data discussed above show much less venting and flaring in 1990 (48 SCF/bbl). The question, then, is this: has offshore venting and flaring declined dramatically since the early-to-mid 1970s, or are the MMS data not as complete as the data sources cited in Sea Technology (1974) and Brooks et al. (1977)? The answer appears to be the former, because Sea Technology (1974) cites the Department of the Interior (probably the U.S. Geological Survey), and Brooks et al. (1977) cite a personal communication from the U.S. Geological Survey, and the MMS took over the task of collecting venting and flaring data from the Geological Survey (Nixdorff, 1991). Therefore, all the data probably come from the same source, and show that venting and flaring have declined. (Note, too, that the MMS data come from oil companies, who are supposed to report *all* venting and flaring.)

In sum, it probably is reasonable to assume that the EIA data and the MMS data together cover nearly all of the sources of vented or flared gas in the U.S[16]. Unfortunately, the picture is much less clear for the rest of the world. The EIA has reported that it does not know if the sources it uses to estimate venting and flaring emissions in other countries are reliable or complete. We suspect that in many cases the data are not reliable, and that venting and flaring is significantly under-reported in many parts of the world. As shown in Table 6, we have used our judgment to adjust for this likely under-reporting.

2.4.2 Vented vs. flared To calculate the greenhouse effect of vented and flared gas, the aggregate measure "vented or flared" must be disaggregated to the amount vented (venting releases mainly CH_4, and smaller amounts of NMHCs, and CO_2)

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actually vented or flared might be quire different than the amount reported by the EIA . Unfortunately, no other estimates are available.

[16] But see the notes to Table 6.

and the amount flared (flaring produces mainly CO_2, with smaller amounts of CH_4 and NMHCs). Although none of the documents cited above separate the proportion vented from the proportion flared, it is widely believed that most "vented or flared" gas actually is flared. Staff at the Office of Oil and Gas at the EIA expect that at least 95% of all vented or flared gas is actually flared. They point out that the term "vented or flared" is something of an anachronism, dating from a time several years ago when a fair amount of gas really was vented (McCarrick, 1990). In fact, unmarketable, unreinjected associated gas must be flared, to destroy toxic compounds in the raw gas and to prevent the accumulation of an explosive concentration of natural gas. Generally, gas can be vented only when a very small amount is released in a remote location with strong winds, and these situations are relatively rare. The EPA (1993) cites a draft report by Radian Corporation that estimates that at least 96% of the total reported venting and flaring actually is flared rather than vented.

However, there are several other factors to consider. First, as mentioned above, Sackett and Barber (1988) and Brooks et al. (1977) believe that in the 1970s, a large fraction of offshore waste gas was vented underwater. Brooks et al. (1977) state that "the [offshore] petroleum industry considers venting preferable" to flaring, for several reasons (p. 378), and note that the U.S. Geological Survey estimated that 70% of offshore vented or flared gas actually was vented, in 1974. In support of this, Brooks et al. (1977) also found a high concentration of CH_4 and other hydrocarbons in Gulf waters.

Second, in addition to venting and flaring emissions, there also are fugitive gas emissions from oil production fields. The EPA (1993) estimates that fugitive CH_4 emissions could have been as high as 0.022 Tg (about 1.2 billion cubic feet [BCF]) in 1990, or roughly 1% of reported venting and flaring emissions in the U.S. However, the EPA's best guess is that fugitive emissions were much less than this. They also note that fugitive emissions have been declining.

Third, industry sources have reported that flares often are not nearly 100% effective, and thereby emit a significant amount of unburned gas. For example, industry sources cited in Barns and Edmonds (1990) believe that total CH_4 emissions due both to venting and to incomplete combustion in flares are 20% of total reported venting or flaring emissions. Unfortunately, there are no data on the average effectiveness of flares, or the amount of unburned CH_4 emitted from flares.

We handle these issues as follows. First, we assume that 4% of the EIA-reported venting or flaring emissions from onshore oil production is vented rather than flared. Next, we assume that fugitive emissions are equivalent to an additional 1% of reported venting and flaring, as venting. Then, we assume that flares are 95% effective, so that an additional 5% (0.96 x [1-0.95]) of onshore venting or flaring emissions effectively is "vented," as unburned gas. Therefore, the total effective venting rate for onshore oil production is 4% (direct venting) + 1% (fugitive emissions) + 5% (unburned gas from flares) = 10% of reported venting or flaring.

Regarding underwater venting from offshore oil production, there are two possibilities: either the fraction of vented gas has declined considerably, as implied by the EIA, or else there is still a large amount of perhaps clandestine venting underwater. We take a middle ground, and assume that underwater venting has declined somewhat, from the 70% level estimated by the USGS for 1974, to 30% in 1987. We also assume that gas vented underwater eventually enters the atmosphere.

The onshore vented fraction and the offshore vented fraction must be combined. In 1987, 21 BCF of gas was vented or flared from offshore State and Federal oil leases (based on 48 SCF/offshore-bbl, from above, and 437 million bbl produced from state and Federal offshore leases, according to the MMS [1989]), and 121 BCF was vented or flared from onshore fields (124 BCF from onshore + State offshore production, as reported by the EIA, less my estimate of 3 BCF from the offshore State leases, which are included in the 21 BCF estimate). If 10% of onshore gas was effectively vented, and 30% of offshore gas was vented, then overall, about 13% of total (onshore plus offshore) vented or flared gas was effectively vented, in the U.S. in 1987.

2.4.3 Vented vs. flared in other countries This 13% venting rate applies to crude oil *produced* in the U.S.; it does not necessarily apply to all the oil *consumed* in the U.S., because the U.S. imports roughly half of the oil that it uses, and the percentage of gas that is vented rather than flared will vary from country to country. In Table 6, as part of the calculation of the average venting or flaring rate attributable to U.S. oil use, we assume venting percentages for each of the regions of the world. We assume that in industrialized countries the percent of vented or flared gas that is vented (or not burned in flares) is the same as in the U.S. However, we assume that in other countries, particularly in Africa, the percentage vented is higher -- perhaps as high as 20% -- because of looser regulations and enforcement and poorly functioning equipment. With these assumptions about the fraction vented or incompletely flared, and with separate CEFs assigned to each compound or class of compounds emitted (CH_4, CO_2, and NMHCs), one can calculate the CO_2-equivalent impact of gas emissions associated with the use of oil in the U.S.

2.5 Methane emissions from coal mining

The processes that produce coal -- called "coalification" -- also produce methane and other gases. Some of this coalbed gas is stored in the coal bed itself. However, coalification produces much more gas than the coal itself can store. This excess gas migrates into the surrounding rock and sand strata, forming the "traditional" natural gas deposits mined by the natural gas industry. The formation of a ton of anthracite may generate 6,000 cubic feet of CH_4, and the formation of a ton of very high rank coal may generate up to 27,000 cubic feet (Ayers and Kelso, 1989; Thakur, et al., 1996).

The gas retained in coalbeds ranges from a negligible quantity up to about 900 cubic feet per ton. The rate of gas release from coal mines. as a result of mining, depends on several factors, including: the age, depth, and structure of the coalbed; the mining technique; and the rank and quality of the coal. Gas production increases with the depth of the mine and the rank of the coal (the higher the fixed carbon content of the coal, the higher the CH_4 content) (Deul and Kim, 1988; Kuuskra and Brandenburg, 1989). Underground mines produce an order of magnitude more gas per ton of coal than do surface mines.

Most of the gas in mined coal is released when coal is depressurized to atmospheric pressure as it is exposed. Most of the remaining gas is released when the coal is cleaned, crushed, and prepared for final use. A small amount of gas remains in the prepared coal and is burned with the coal.

In order to estimate the impact on climate of gas produced as a result of coal mining, we must know the rate of release of coalbed gas per ton of coal mined, and the fate of the released gas. Released coalbed gas may be:

i) mixed with air and vented to the atmosphere;
ii) collected, drained, and flared; or
iii) collected, drained, and sold as a fuel[17].

Vented gas has a greater impact on climate than does flared gas (because a mole of CH_4 has a greater warming potential than does a mole of CO_2), and gas recovered and used as a fuel has no net impact on climate at all if some other fossil natural gas would have been used in lieu of the coalbed gas. Thus, we estimate the CO_2-equivalent impact of emissions from coal mining as follows:

$$CBG_{CO2} = (CBGR - CBGC - CBGF) \cdot \left(\sum_i F_i \cdot CEF_i \right) + CBGF \cdot F_C \cdot 3.667$$

$$CBGR = CBGRT_U \cdot P_U + CBGRT_S \cdot P_S$$

Let:

$$CBGC = CBGR \cdot K1$$

$$CBGF = CBGR \cdot K2$$

where:
CGB_{CO2} = the CO_2-equivalent impact of atmospheric emissions of coalbed gas
CBGR = coalbed gas released by coal mining (but not necessarily emitted to the atmosphere)
CBGC = released coalbed gas that is captured and used as a fuel (see the discussion below)
CBGF = released coalbed gas that is flared rather than simply vented to the atmosphere (assumed, in the absence of any actual data, to be 5% of CBGR; i.e., K2 = 0.05)

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[17] A small amount of the methane released by a coal-producing mine can be captured and used as a fuel at the mine. However, statements and data in Deul and Kim (1988), ICF (1990b), and DeLuchi (1991) indicate that the amount of methane used as fuel at the mine is a minuscule fraction of the total amount produced and vented.

F_i = the mass fraction of gas i in coalbed gas (see the discussion of the composition of coalbed gas, below)

CEF_i the CO_2-equivalency factor for gas i (see Table 2 for CH_4 and NMHCs)

F_c = the carbon weight fraction of coalbed gas (calculated from data on the composition of coalbed gas, discussed below)

3.667 = the ratio of the weight of CO_2 to C

$CBGRT_u$ = the rate of release of coalbed gas per ton of coal from underground mines (see the discussion below)

$CBGRT_s$ = the rate of release of coalbed gas per ton of coal from surface mines (see the discussion below)

P_u = coal production from underground mines (estimated from EIA historical data and projections for coal production (EIA, 1996b; EIA, 1994c))

P_s = coal production from surface mines (estimated from EIA historical data and projections for coal production (EIA, 1996b; EIA, 1994c))

In the following, we will discuss coalbed-gas releases from underground and surface mines, the fate of released gas, and the composition of the gas.

2.5.1 Coalbed gas releases from coal mining Detailed estimates of total CH_4 releases from coal mining have been made only recently. Most estimates made in the 1980s, including those by Bolle et al. (1986), Crutzen and Gidel (1983) and Rasmussen and Khalil (1984) were based on articles by Ehhalt and Schmidt (1978) and Ehhalt (1974), which, in turn, referred to a NASA study by Hitchcock and Weschler (1972). The NASA report used an estimate from Koyama (1963), which appears to be an original work. However, Koyama (1963) was concerned mainly with CH_4 from paddy fields; he estimated CH_4 emissions from coal mining in a one-sentence calculation in which he assumed that coal fields produce CH_4 at a rate of 21 cm^3/g of coal mined (p. 3973). This unreferenced, unelaborated, 1963 assumption was propagated through the literature for nearly three decades.

Recently, however, several original and detailed estimates have been made. (Note that most of the following are estimates of emissions to the atmosphere, not releases due to coal mining; as presented above, emissions are equal to releases less amounts captured and used as a fuel.) In 1990, ICF (1990b) estimated emissions from coal mining and use for every state in the U.S., as a function of the amount of coal mined, the mining technique, and the CH_4 content of the coal. The CH_4 content of the coal was estimated using data on the CH_4 content of various classes of coal, and the amount of coal production by class of coal. They also estimated emissions worldwide. They estimated that in 1987 coal mining resulted in emissions of 368 BCF of CH_4 in the U.S., and 2494 BCF worldwide, or about 400 SCF-CH_4/ton-produced in the U.S., and 492 SCF-CH_4/ton-coal-produced worldwide. They estimated that CH_4 emissions from coal mining worldwide will increase from 3788 to 4262 TCF by the year 2000, due partly to increasing coal production, but also due to an assumed increasing average mine depth, a factor which results in higher emissions per ton of coal produced.

In 1993, the EPA updated and expanded the ICF work, to estimate releases and emissions of coalbed gas in 1988. They used general assumptions to estimate releases from surface mines, but actually estimated releases from each underground mine in the U.S. They also estimated that 12 BCF of the released coalbed gas was captured and sold to gas companies in 1988. We have divided their estimates of total CH_4 emissions by their estimates of total coal production, to produce a national average emission factor, shown in Table 7.

Using an approach based on direct analysis of the CH_4 in 137 coal samples, Kirchgessner et al. (1993b) estimated 1989 global CH_4 emissions from underground coal mines by developing two sets of regression equations. The first set of equations relates the total residual and desorbed CH_4 in a coal sample to its heating value, depth, moisture content, and fuel ratio (i.e. the ratio of fixed carbon to volatile matter). Two separate equations were estimated: one for coals with a heating value below 34,680 J/g and another for coals with a heating value above this figure (the figure was chosen based on a clear break-point in a graph of CH_4 content versus heating value). These equations are as follows:

$$(HV<34{,}680\ J/g)\quad IS = 0.0159\ D + 2.2781/M^2 - 2.228$$
$$(HV>34{,}680\ J/g)\quad IS = 0.0136\ D + 0.0015\ HV + 2.6809\ FR - 56.4901$$

where:
HV = coal heating value in J/g
IS = in-situ residual + desorbed gas (cubic meters CH_4/tonne of coal)
D = depth in meters
M = percent moisture content
FR = fuel ratio (fixed carbon/volatile matter)

The estimation of these two equations produced R-squared values of 0.56 and 0.71 respectively (only variables that could be retained with 95% or greater confidence were included). The remaining step in estimating the total in-situ CH_4 content of the coal was to add a factor to account for the CH_4 "lost" between the time the coal was sampled and when it was placed in the sampling canister. Using data from various studies, lost gas factors were estimated for each of seven coal ranks; these factors ranged from 0.05 for high-volatile bituminous, to 0.20 for medium-volatile bituminous. The total in-situ CH_4 content of the coal was then taken as the sum of residual, desorbed, and lost CH_4 (Kirchgessner, et al., 1993b).

The second regression equation estimated by Kirchgessner et al. relates total mine shaft and gob well emissions of CH_4 to annual coal production, total CH_4 content of unmined coal, and a dummy variable based on the product of coal production and in-situ CH_4 content. This equation is as follows:

$$ME = 1.08\ 10^{-7}\ (CP \times IS) + 31.44 - 26.76\ (DV)$$

where:

ME = total emissions of CH_4 from mine shafts and gob wells (10^6 meters per year)
CP = annual coal production in tons per year
IS = total CH_4 content of unmined coal (cubic meters CH_4/ton of coal)
DV = dummy variable (1 if coal production x in-situ CH_4 content < 7.6×10^6, 0 if coal production x in-situ CH_4 content > 7.6×10^6)

The R-squared value for this equation is 0.59, which indicates that nearly 60 percent of the variation in mine CH_4 emissions can be explained by these independent variables. Using these regression equations and the assumptions that 6.9 Tg/year of CH_4 are emitted globally from surface mines and 2.7 Tg/yr. are emitted globally from coal handling operations (assumptions based, respectively, on a single study of surface mine emissions and the unsupported assumption that 25 percent of the CH_4 in mined coals is released after the coal leaves the mine), a final estimate for 1989 global CH_4 emissions from surface and underground mining operations of 45.6 Tg (or 63,469 10^6 m^3) was obtained (Kirchgessner, et al., 1993b). The estimate for U.S. underground mines is shown in Table 7.

The EIA's (1995b) analysis of emissions of greenhouse gases in the United States uses data from the EPA (1993) report and from other more recent sources to estimate CH_4 emissions from coal mining. They estimate releases from surface mines and underground mines, and the amount of the release gas that is captured and used. Table 8 presents details from their analysis. Table 7 shows the EIA estimate for 1988, for comparison with the EPA's estimate. The EIA estimate for 1988 falls between the EPA's low and high estimates for 1988.

Finally, in 1992 the Coal Industry Advisory Board (CIAB) estimated global emissions of CH_4 from coal mining , based on 1990 production data (see Table 7 for the estimate for U.S. mines). The study estimated total CH_4 emissions from mining activity in ten countries, as well as the amount of coalbed CH_4 that can potentially be recovered. Thakur et al. (1996) report the results of this study, as well as estimating the amount of CH_4 actually recovered and used in 1994. According to the CIAB study, coal mining operations in China released the most CH_4, with emissions of over 405 BCF, compared to about 190 BCF emitted in the U.S. (Thakur, et al., 1996). Meanwhile, actual recovery and use rates varied substantially among countries in 1994, with Germany, Austria, and Czechoslavakia having the highest use rates (relative to total emissions) of about 20-25%, the U.S. next with about 18% of total emissions recovered and used, and other countries following with usage rates of 5% to 10% (Thakur, et al., 1996).

Our assumptions. We use the most recent EIA (1995b) estimates of methane releases from coal mines, because they distinguish underground-mine releases from surface mine releases (Table 8), and clearly distinguish releases from emissions (i.e., provide separate estimates of the parameters CBGR and CBGC in our equation above). We start with their estimates of CBGRTu and CBGRTs in 1993, and then assume that CBGRTu increases 0.5%/year, on account of underground mines becoming deeper as they shallower coal is exhausted. (Recall that releases increase with depth.) We assume that CBGRTs remains constant.

2.5.2 *Fate of methane releases from coal mines.* Most released coalbed gas simply is mixed with air and ventilated to the surface. By law, the vented gas must contain less than 1% CH_4 (Kim, 1990), a concentration that is too dilute to burn. This coalbed gas may be counted as a net GHG emission from coal mining.

However, as noted above, some released coalbed gas is recovered and used as a fuel. This recovered coalbed gas fuel presumably simply displaces other fossil natural gas, and hence does not contribute to global warming. Thus, it is important to estimate the amount of released gas that is recovered and used as a fuel, and deduct the amount from gross releases to arrive at net emissions to the atmosphere.

Table 8 shows the amount of released coalbed gas the EIA (1995b) estimates to have been recovered through 1993. The amount of gas recovered has increased in recent years, and is expected to continue to increase. We assume that the amount of released gas that is recovered and used as a fuel increases by 3%/year[18].

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[18] U.S. coalbeds contain about 400 TCF of CH_4, of which at least 90 TCF are recoverable (Ayers and Kelso, 1989; Black, 1994; Black, 1990). Under ideal conditions, 60 to 70 percent of the CH_4 from a coalbed can be recovered and used, but under more typical conditions the usable percentage is in the range of 30 to 40 percent (Thakur, et al., 1996). However, some CH_4 that is recovered cannot be economically utilized, even for cogeneration or on-site heating, and is therefore vented after recovery. Thakur et al. (1996) suggest that while CH_4 recovery could reach 30% to 40% of total emissions, perhaps only 50% of the recovered CH_4 will actually be utilized.

Coalbed CH_4 research and development has grown considerably in recent years, and several large CH_4 recovery projects are operating, near-operating, or planned (Ayers and Kelso, 1989; Kuuskra and Brandenburg, 1989; Schraufnagel, et al., 1990). In 1988, only 28 BCF of CH_4 were recovered from U.S. coalbeds and marketed, mostly in Alabama, Colorado, and New Mexico (EIA, 1989d). In 1989, 80 BCF were recovered and marketed, again mainly in Alabama, Colorado, and New Mexico (EIA, 1990a). However, in 1992 550 BCF, or over 3% of total natural gas supply, were recovered (Black, 1994), and in 1994 over 900 BCF were recovered (EIA, 1995c), some of the increase owing to methane recovery at four extremely gassy mines in Virginia (U.S. EPA, 1995c). By the year 2000, U.S. production could reach 1500 BCF (Petroleum Energy & Intelligence Weekly, 1992).

Still, there are several obstacles to large-scale development of coalbed methane. First, it is difficult to find highly permeable, productive spots in coal fields (Schraufnagel, et al., 1990). Second, gas recovery is expensive. The recent rapid development of coal-bed methane was spurred by a 90cents/CF tax credit, which expired after 1992. The expiration of this credit apparently has dampened interest in producing coalbed methane, because fewer wells are being drilled in most basins. High gas prices or another public subsidy would spur interest again (Black, 1994). Third, environmental regulations -- for example, regarding the disposition of coproduced water -- might be restrictive (Schraufnagel et al., 1990). For these and other reasons, coal companies are reluctant to get involved . Petroleum & Energy Intelligence Weekly (1992) sums up the situation best: "...worldwide interest heralds a rich future for CBM, though it likely will take several more years before the true potential of many of these areas can be accurately gauged. And almost every region sports unique problems -- including scant finances, creaky infrastructure, and political uncertainty. But with the political status of gas rising steadily throughout the world, expect a bigger CBM push on an even more far-ranging basis" (p. 15).

An important clarification: the amount of "recoverable" coalbed gas discussed in this footnote is not the same as the amount of *released* gas that is recovered and used as a fuel. The latter refers to the recovery of gas that is released as a result of mining, whereas the former refers to the recovery of any gas from any coalbed, whether mined or not. The estimates in this footnote include recovery of gas from

*2.5.3 **Composition of coalbed gas**.* In most analyses of greenhouse-gas emissions, coalbed gas is assumed to be 100% CH_4. However, coalbed gas does contain small amounts of other compounds, such as C_2H_6 and CO_2, which have different CEFs from that of CH_4. Typically, about 80 to 95 percent of coalbed gas is CH_4; the remainder being trace quantities of ethane, propane, butane, carbon dioxide, hydrogen, oxygen, nitrogen, and helium (Thakur, et al., 1996). In this analysis we estimate the composition of coalbed gas, based on data in Deul and Kim (1988 they show the composition of coal gas from 7 coalbeds), and multiply the mass of each compound emitted by its CEF.

*2.6 **Methane emissions from other sources in energy fuel cycles***

Methane is emitted from a variety of energy-using and energy-manufacturing equipment and facilities, including: the equipment used to lift and process oil, natural gas, and coal (e.g., scrapers, haulers, diesel engines, and well equipment); the modes used to transport energy feedstocks and products (e.g., trains, ships, and pipeline compressors); and the facilities that manufacture end-use energy from energy feedstocks (e.g., petroleum refineries, electricity-generating plants, and methanol production plants). Generally, one must find or estimate CH_4 emission factors for each of these sources. (Methane emission factors usually are expressed as the weight of CH_4 emitted per unit of fuel or feedstock input, or per unit of product output). A good reference source on emission factors for a variety of pollutants (often including CH_4) from a large number of sources is the EPA's *Compilation of Air-Pollutant Emission Factors*, volumes I (stationary sources) and II (mobile sources), known as "AP-42." The fifth edition of Volume I was published in 1995.

Table 12 compile estimates of CH_4 emissions from several emission sources. As indicated in the notes to those tables, many of the estimates are based on data in AP-42[19]. AP-42 rarely reports CH_4 emissions per se, but often it reports emissions of

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recovery of gas that is released as a result of mining, whereas the former refers to the recovery of any gas from any coalbed, whether mined or not. The estimates in this footnote include recovery of gas from beds that either are unminable (because the coal is too deep, as in the San Juan Basin of Colorado), or are so gassy that they will not be mined unless the gas is removed. Hence the estimates of recoverable coalbed gas greatly exceed the estimates of the amount of gas released by mining and then recovered.

[19] The AP-42 emission factors for fuel combustion are not *net* of the ambient concentration; that is, the background ambient concentration of pollutants (in the input air) has *not* been netted out from the published emission factors (McSorley, 1993). This netting out could be important: Watanabe and Matsuura (1992) measured the [gross] concentration of CH_4 in the flue gas of power plants in Japan to be 0.07-0.615 ppm for oil-fired plants, 0.27-1.37 ppm for LNG-fired plants, and 1.60 ppm for a coal-fired plant, all of which are *less* than the average ambient CH_4 concentration of 1.8 ppm. This means that power plants, at least in Japan, on balance actually *consume* atmospheric CH_4. In the Japanese study, 1.8 ppm corresponds to about 0.41 ug/kJ, or 0.44 $g/10^6$ Btu. As shown here, the AP-42 emission rates for coal and oil-fired power plants are higher than this, and are net of the ambient concentration. American power plants, apparently, produce net CH_4.

hydrocarbons (HCs) and emissions of nonCH_4 hydrocarbons (NMHCs)[20]; the difference between the two presumably is CH_4 emissions. If for a particular emission source AP-42 reports only HC or NMHC emissions (but not both), one can estimate CH_4 emissions based on CH_4/HC or CH_4/NMHC ratios for the same fuel used in similar applications. We have done this here.

One can readily see that, with a few exceptions, CH_4 emissions from the sources of Table 12 are relatively minor, compared both to other CH_4 sources (such as coal mining), and to emissions of other greenhouse gases (especially CO_2) from the same source. For example, emissions of 2.0 g/10^6 Btu are a very minor portion of total CO_2-equivalent emissions from the gasoline LDV fuelcycle -- on the order of 0.1% of total fuelcycle CO_2-equivalent emissions.

2.6.1 Other minor sources of methane. In 1988, Sackett and Barber (1988) proposed that CH_4 emissions from road asphalt might be an important, overlooked source of atmospheric fossil-fuel-derived CH_4 (CH_4 with no ^{14}C present). However, in a detailed experimental analysis and calculation, Tyler et al. (1990) concluded that global CH_4 emissions from asphalt were at most 0.01 Tg/year, and therefore a tiny fraction of the roughly 500 Tg of CH_4 emitted annually. Tyler et al. (1990) also estimated that CO emissions from road asphalt in Denver, Colorado were a small fraction -- about 0.03% of CO emissions from motor vehicles.

Pressurized-water nuclear reactors produce about 900 curies/year of a reduced form of CH_4, $^{14}CH_4$ (Wahlen, et al., 1989). However, at 4.6 curies/gram of carbon-14, the total CH_4 production is infinitesimal.

Hydroelectric power generation can produce CH_4, as inundated soils and organic matter degrade and their carbon content becomes mineralized (to CO_2 as well as to CH_4). These emissions are analogous to emissions of CO_2 and CH_4 from natural processes in pristine lakes and wetlands. Unfortunately, very few studies have been conducted on the magnitude of these emissions. One study conducted on hydroelectric reservoirs in the boreal region of northern Quebec revealed that, over a two year period, emission fluxes of CH_4 to the atmosphere ranged from 5 to 10 mg per m^2 per day, and emission fluxes of CO_2 ranged from 500 to 1100 mg per m^2 per day (Duchemin, et al., 1995). Interestingly, emissions were found to be independent of the nature of the flooded substrate and the amount of time elapsed since inundation. Emissions of CH_4 were judged to be primarily governed by oxidation and advection processes in the water column. The authors conclude that the measured emission levels are significant, but much lower per unit of energy produced than those from thermal power plants[21]. Clearly, before any definitive

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[20] All hydrocarbons contribute to the formation of tropospheric ozone, but at different rates. Methane reacts so much more slowly than do most other hydrocarbons that often it is considered to be effectively nonreactive, and thus irrelevant to emissions estimates concerned with urban air pollution.

[21] A back-of-the-envelope calculation suggests that this conclusion might not be correct. Emissions from hydropower plants depend greatly on the ratio of the output of the hydropower plant to the area inundated. The available data suggest that this ratio can vary enormously, from as little as 1 kW/ha to as much as 1000 kW/ha or more (Moreira and Poole, 1993; Ogden and Nitsch, 1993). The historical

conclusions are drawn regarding overall greenhouse gas emissions from hydroelectric reservoirs, more research is needed on emissions from reservoirs in different latitudes, on emissions from areas with different soil and fauna characteristics, on relative emissions from inundated versus non-inundated areas, and on seasonal variations in emissions.

The transoceanic shipping of LNG in cryogenic vessels and the offloading of LNG at terminals probably results in negligible additional emissions of CH_4. About 2.5% of the loaded LNG boils off during a 15-day transoceanic trip, but this boiloff is used by the LNG tanker as fuel (Chem Systems, 1988). More LNG boils off during offloading at the terminal, but this presumably is reliquified or used as fuel. It does not seem likely that more than about 0.2% of the LNG cargo actually is lost to the atmosphere. The AGA (AGA, 1985) projects that the U.S. will import at most $1 \cdot 10^{12}$ ft^3 of LNG in the year 2010. The associated $2 \cdot 10^9$ ft^3 of atmospheric emissions from the transport of this amount of LNG would be equivalent to roughly 1% of total venting or flaring in the U.S., and less than 5% of the lower estimates of leakage from NG systems.

Finally, there are other minor emissions of CH_4 associated with the use of oil. About 15% of the total evaporative or displacement emissions of HCs from marine-vessel operations, crude-oil storage tanks, and crude-oil treatment facilities is CH_4 (U.S. EPA, 1985). Based on DeLuchi et al.'s (1992) projection of emissions of *non*methane hydrocarbon from the production, transfer, and storage of crude oil in

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average in Brazil is 22 kW/ha (Moreira and Poole, 1993). Combining this power/area rate with the emission factors of 50 to 100 g/ha/day CH_4 and 5,000 to 11,000 g/ha/day CO_2, and assuming the U. S. average of 10-kWh/day/kW (EIA, 1996b) (a capacity factor of about 40%), we calculate emissions of 0.005 to 10 g-CH_4/kWh and 0.5 to 1,100 g-CO_2/kWh. How do these ranges compare with emissions from fossil-fuel power plants? The data of Table 8 indicate that CH_4 emissions from thermal power plants range from 0.1 to 11 g/10^6-BTU fuel input, or about 0.001 to 0.110 g-CH_4/kWh assuming a 34% generating efficiency. Emissions of CO_2 from thermal plants range from 500 to 1000 g/kWh. Thus, in g/kWh:

	Hydropower			Fossil-fuel power
	low emissions, high output	average Quebec emissions, Brazil average output	high emissions, low output	
CH_4	0.005	0.3	10	0.001 - 0.1
CO_2	0.5	40	1,100	500 - 1,000

This quick comparison suggests that CH_4 emissions from hydropower plants might be comparable to or even greater than CH_4 emissions from fossil-fuel plants (per kWh), but that CO_2 emissions from hydropower plants will be less than CO_2 emissions from fossil-fuel plants. However, this conclusion must be tempered by at least three considerations: i) emissions/area and output/area might not be independent, as we have assumed here; ii) one should deduct from the "gross" hydropower emission rate, shown in the table above, the "background" rate of emissions that would have been generated by un-inundated soil and riverbed; and iii) emissions from an existing hydropower plant are independent of the amount of power actually generated.

the year 2000, we estimate that CH_4 emissions from these processes will be about 0.65 g per 10^6 Btu-refinery product. This is a relatively small amount -- for example, it is about 5% of tailpipe emissions of CH_4 from most vehicles -- and so probably can be ignored. We have not included these emissions in the model or the results presented here.

2.7 Comparing our estimates of fossil CH_4 emissions with estimates based on atmospheric ^{14}C concentrations.

The emission factors discussed above for "fossil" CH_4 result in total global emissions consistent with the amount of emissions implied by measurements of "dead" (mostly fossil-fuel-derived) CH_4 in the atmosphere.

Methane derived from recent organic material has a ^{14}C-content very close to that of wood, whereas methane derived from fossil fuels and "old" biological sources has essentially no ^{14}C, because the ^{14}C originally present in the organic material has decayed. Studies of the radiocarbon content of atmospheric methane indicate that 15-30% of the methane in the atmosphere has no ^{14}C, and therefore is not of recent biological origin (Cicerone and Oremland, 1988; Ehhalt and Schmidt, 1978; IPCC, 1992; Lowe, et al., 1988; Wahlen, et al., 1989). Based on this, Cicerone and Oremland (1988) estimate that fossil-fuel use results in emissions of about 80tg/yr. of CH_4, out of total methane emissions of 540 tg/yr.

The emission factors and estimates presented above result in a total very close to the 80 tg/year back-calculated from atmospheric ^{14}C concentrations. The data discussed above suggest that coal mining was responsible for about 45 tg of CH_4 emissions in the mid 1980s. In 1986, the world consumed 63 TCF of natural gas (EIA, 1995). If 2% of this was lost, and the lost gas was 90% methane by weight, then about 24 tg was emitted from natural-gas production, distribution, and use. Worldwide, oil-producing wells vented or flared 3.5 TCF of co-produced (associated) but unmarketable natural gas in the mid 1980s. If unprocessed natural gas straight from the ground was 22.92 grams/SCF and 68% CH4 by weight (DeLuchi, 1991; raw gas also contains appreciable amounts of higher alkanes, CO_2, N_2, and H_2S), and if 15% of the total gas that was either vented or flared actually was actually vented or escaped unburned from flares (estimate based on data of Table 6), then about 8tg of CH_4 were released in 1989, due to oil production alone. An additional 2 tg or so of CH_4 was emitted from other oil or coal-related activities (petroleum refining, crude oil storage and transfer, and oil, gasoline, and coal combustion). The total then is 45 (coal mining) + 24 (natural gas) + 8 (oil production) + 2 (miscellaneous) = 79 tg/year.

2.8 Credit for gas that otherwise would have leaked to the atmosphere

Gas leaks naturally from natural gas formations, coal mines, and landfills. Leakage from landfills alone may account for 10-20% of global CH_4 emissions (Bingemer and Crutzen, 1987). If one does nothing about these leaks, they will continue to contribute to global warming. On the other hand, if one develops these gas sources and puts the gas to work as fuel, one not only gets useful work, one actually reduces emissions of greenhouse gases, because burning the gas transforms the CH_4 into CO_2 and H_2O. Hence, one can argue that the amount of gas that would have escaped anyway, had the gas source never been developed for production, should be counted as a credit against any CO_2 and CH_4 emissions from the gas production-and-use system.

Thomas Gold (1988) has advanced this sort of argument, claiming that in some cases the production of gas "demonstrably" reduces the rate of gas loss to the atmosphere. However, he does not say by how much. For lack of data, we assume in this analysis that any such credits are relatively small, so that the "net" leakage rate -- total leakage from NG production, less the amount that would have leaked had the gas field not been developed -- is about equal to the gross leakage rates reported and used here. Clearly, though, this issue should be investigated further.

3. NITROUS OXIDE (N_2O)

3.1 Nitrous oxide as a greenhouse gas

The concentration of tropospheric 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, 1995; 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, and total anthropogenic emissions are 3 to 8 Tg N per year (IPCC, 1995). 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, 1995). The main sink for N_2O is photolysis in the stratosphere, a process that accounts for about 90% of N_2O destruction (Watson et al., 1990) (Kramlich and Linak, 1994).

Nitrous oxide is a potent greenhouse gas, with a per molecule radiative forcing over 200 times that of CO_2 (Watson, et al., 1990). The CEFs for N_2O are shown in Table 2. N_2O also is involved stratospheric O_3 chemistry. 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).

3.2 Nitrous oxide emissions from motor vehicles

3.2.1 Gasoline LDVs. General. N_2O is emitted directly from vehicles (Table 10). N_2O emissions from catalyst-equipped gasoline LDVs depend significantly on the type and temperature of catalyst, rather than total NO_x levels or fuel nitrogen content. Gasoline contains relatively little nitrogen, and so fuel NO_x and fuel N_2O emissions from autos are low. The high temperatures and pressures of the ICE are sufficient to form NO_x thermally, but evidently are inefficient for production of

N_2O (Hao, et al., 1987). Thus, cars without catalytic converters produce essentially no net N_2O (Table 10). On the other hand, Weiss and Craig (1976) predicted N_2O exhaust concentrations of up to 400 ppmv from autos with platinum reduction catalysts only, according to the reactions:

$$6NO + 4NH_3 \dashrightarrow 5N_2O + 6H_2O \quad \text{and}$$
$$2NO + H_2 \dashrightarrow N_2O + H_2O$$

Seinfeld (in Pierotti and Rasmussen, 1976) suggests that N_2O production in 3-way catalysts (those that oxidize HC and CO, and reduce NO) may occur by a different route:

$$NH_3 + 2O_2 \dashrightarrow N_2O + 3H_2O$$

Alternatively, the NO can be reduced by CO, rather than H_2 (and Dasch, 1992; See also Prigent and Soete, 1989; Ryan and Srivastava, 1989):

$$2NO + CO \dashrightarrow N_2O + CO_2$$

N_2O formed in this last way also may be reduced back to N_2 (Dasch, 1992), via:

$$N_2O + CO \dashrightarrow N_2 + CO_2$$

Temperature. Prigent and De Soete (1989) conclude that the N_2O is formed at relatively low catalyst temperatures; at higher temperatures, the N_2O itself is destroyed. They show a graph in which N_2O formation, via reduction by CO over a metal catalyst, peaks at 120 ppmv at 500 K and drops to zero on either side, at 400 K and 650 K. Prigent (in Ryan and Srivastava, 1989) shows N_2O formation across a 3-way platinum-rhodium catalyst peaking at 180 ppmv at about a 635 K catalyst inlet temperature (which implies a slightly cooler temperature across the catalyst). The dependence of N_2O formation on temperature causes higher emissions from a cold-start test than a hot-start test, and relatively high emissions during catalyst light-off (Prigent and De Soete, 1989). For example, Lindskog (in Ryan and Srivastava, 1989) has found that after a cold start the concentration of N_2O in the exhaust pipe, after the catalyst, is two times higher than the concentration before the catalyst. This has important implications: electrically heated catalysts, which might be used in the future to reduce cold-start HC emissions, probably will reduce N_2O emissions too.

Although lower *catalyst* temperatures almost certainly increase N_2O emissions (Ryan and Srivastava, 1989; Dasch, 1992), colder ambient temperatures may increase or decrease N_2O emissions (see note j, Table 10). This occurs because once a vehicle is warmed up, the temperature of the catalyst is not directly related to the air temperature, but rather is determined by the temperature of the exhaust gases, which are determined by engine load and combustion conditions.

Drive cycle. The type of drive cycle appears to influence N_2O as well, probably because it affects the temperature across the catalyst. N_2O emissions are consistently lower in the highway driving cycle, when the catalyst is warmed up, than in the FTP (Table 10; Dasch, 1992).

Based on tests of four gasoline vehicles and one diesel vehicle, Sasake and Kameoka (1992) show that for gasoline LDVs N_2O emissions decrease with higher vehicle speeds (over the range from 60 km/hr to 100 km/hr) and higher incline gradients. While variations in vehicle speed and incline gradient had no significant effect on N_2O emissions from the diesel vehicle tested, one new gasoline vehicle equipped with a 3-way catalyst demonstrated a reduction in N_2O emissions from 2.6 mg/mi to 0.3 mg/mi with an increase in vehicle speed from 37 mph to 62 mph. This same vehicle produced 1.3 mg/mi of N_2O when operated at 37 mph on a 2% incline gradient, and only 0.6 mg/km of N_2O on a 4% gradient. Over the Japanese "11-step" driving cycle (which includes a cold start), a vehicle with a 1000 km odometer reading produced 26.7 mg/mi of N_2O.

In section 2.2.1, we note that the FTP, the drive cycle used in many N_2O emission tests, overestimates average trip length and hence underestimates the fraction of total trip mileage in cold-start mode in real driving, but also underestimates the average vehicle speed. Therefore, in the real world as compared to the FTP, N_2O emissions will be higher on account of the additional cold starts, but lower on account of the higher speeds. It is not clear what the net effect is.

Age of catalyst. While relatively few studies have investigated the potential effect of catalyst age on N_2O emissions, the available data suggest that N_2O emissions can increase significantly as catalyst age accumulates. Based on a study of eight new or laboratory aged catalysts, De Soete (1993) reports that N_2O emissions from vehicles equipped with catalysts aged to an equivalent of about 15,000 miles of use are on average 3.9 times higher than emissions from vehicles equipped with new catalysts, when tested on the European Urban Driving Cycle. On the ECE cycle, average N_2O emissions from the aged catalyst equipped vehicles were 4.43 times higher than those from the new catalyst vehicles. On the ECE 15-04 driving cycle, which includes a cold-start, N_2O emissions from the aged catalyst vehicles were 2.06 times higher than those from the new catalyst vehicles.

Sasake and Kameoka (1992) found that older vehicles produced much higher N_2O emissions than newer vehicles with similar emission control equipment. While this study was not controlled in such a way as to isolate the effect of catalyst age, the oldest vehicle tested (which had an odometer reading of 52,000 km) produced several times the N_2O emissions of the newest vehicle (with only 1000 km of use). The exact magnitude of the emissions difference between the two vehicles varied by driving cycle, but averaged across the 3 Japanese driving cycles studied the older vehicle produced 8.4 times the N_2O emissions of the new vehicle.

A 1989 study by Lindskog (in De Soete, 1993) reports that N_2O emissions from a vehicle with a catalyst aged 15,000 km were 1.3 times (Swedish cold-start Driving Cycle) and 1.5 times (Swedish hot-start Driving Cycle) higher than emissions with a new catalyst. Prigent and De Soete (also in De Soete, 1993) report similar findings, with emissions increases of 1.51 and 1.45 times, respectively, on the ECE-15 (cold start) and EUDC (hot start) cycles, for vehicles using a catalyst bench aged for 150 hours relative to a new catalyst. Also, a recent study of nine "clean fleet" vans operating on reformulated gasoline in Los Angeles revealed that emissions of N_2O at 15,000 miles were on average 1.93 times higher than emissions at 5,000 miles, and that N_2O emissions at 25,000 miles were on average 2.78 times higher than emissions at 5,000 miles (Battelle, 1995).

These few studies are not sufficient to provide a complete understanding of the effect of catalyst age on N_2O emissions, but without exception they do indicate that N_2O emissions increase with catalyst age. This effect can be readily explained with the understanding that peak N_2O formation occurs at higher temperatures with aged catalysts. While the peak quantity of N_2O formed over aged catalysts is lower at these higher temperatures than the peak for newer catalysts at lower temperatures, actual vehicle emissions are higher for vehicles with aged catalysts because a greater proportion of the driving cycle occurs in the "window" of relatively high N_2O formation (De Soete, 1993).

Failure of emission-control equipment. The failure of emission controls can increase or decrease N_2O emissions: if the EGR system is disconnected, emissions increase substantially (see note c, Table 10), but if the oxygen sensor is disable, N_2O is eliminated (Table 10).

Future emission controls. Electrically heated or close-coupled catalysts, which might be used to meet the new NMHC tailpipe emission standards, will reduce the amount of time that the catalyst is cold, and so will reduce N_2O emissions. Also, the possible N_2O formation mechanism, $2NO + CO \dashrightarrow N_2O + CO_2$, indicates that when CO emissions are reduced, N_2O emissions may be reduced. For these reasons, it is likely that future vehicles will have lower emissions than have the vehicles summarized in Table 10.

Summary. The discussion above, and the results of Table 10, suggest that N_2O emissions are a function of the type of emission control equipment (N_2O emissions from cars with 3-way catalysts are uniformly higher than from cars without catalysts), drive cycle, and vehicle speed (N_2O emissions are relatively high from vehicles operated over test cycles that include a cold-start), catalyst age (emissions tend to increase as the catalyst ages), and other factors.

The data of Table 10 indicate that low-mileage 1980's/early 1990s model-year vehicles with a 3-way catalytic converter, or a 3-way catalytic converter plus an oxidation catalyst, emit 20 to 100 mg/mi N_2O, with an average of about 60 mg/mi. Vehicles with around 100,000 miles of catalyst age produce significantly higher emissions, averaging around 200 mg/mi N_2O. For the reasons discussed above, emissions from future vehicles probably will be lower.

With these considerations, we specify our emissions-factor model (used above for CH_4 emissions) as[22]:

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[22] Because different fuel and engine combinations produce different exhaust gas temperatures, and hence different N_2O emission rates, one might first estimate the temperature of exhaust gas, as a function of fuel type, engine design, engine operation, and catalyst location, and then estimate N_2O emissions as a function of temperature. However, this method does not account for the effect of different catalyst

$$EM_{TY} = ZM_{BMY} \cdot \left(1 + \frac{DZ}{100}\right)^{MY-BMY} + DA \cdot \frac{MI_{TY}}{1000}$$

$$MY = TY - AGE$$

$$AGE = f(AMS, MI_{TY})$$

where:
EM_{TY} = emissions from gasoline LDVs in target year TY (g/mi)
ZM_{BMY} = zero-mile emissions from a base-model-year vehicle (0.030 g/mi in 1993; see discussion above)
DZ = the annual percentage change in the zero-mile emission rate (-2.0% per year)
MY = model year of the vehicle (calculated on the basis of the vehicle mileage in the target year)
BMY = base model year for setting emission factors (1993)
DA = the deterioration rate in emissions with vehicle mile (0.0016 g/mi per 1000 miles; see discussion above)
MI_{TY} = total mileage on the vehicle in the target year TY (miles)
TY = target year of the analysis (specified by the user)
AGE = the age of the vehicle (years)
AMS = the annual mileage accumulation schedule (U.S. EPA, 1993; U.S. EPA, 1985)

3.2.2 ***Heavy-duty vehicles.*** Table 10 shows that emissions from heavy-duty diesel vehicles are similar to emissions from passenger cars equipped with catalytic converters, but that emissions from gasoline-powered heavy trucks can be higher. A slight increase in emissions with vehicle age seems to occur in diesel HDVs, but the data are too sparse to draw any firm conclusion. We assume a constant emission rate of 60 mg/mi for both diesel and gasoline HDVs.

3.2.3 ***NO_x and N_2O.*** One way to get around a lack of N_2O emissions data for alternative fuels is to assume a generic relationship between NO_x and N_2O, because NO_x emissions are almost always known. Dasch (1992) argues that the N_2O formation route $2NO + CO \rightarrow N_2O + CO_2$ suggests that N_2O emissions increase as NO_x emissions increase. However, there does not seem to be a strong theoretical or empirical basis for assuming that N_2O emissions are directly proportional to NO_x emissions. The data of Table 10 show clearly that N_2O emissions, and the proportion of NO_x to N_2O, can range over an order of magnitude. Even the for any particular vehicle, a change in operating conditions that increases NO_x emissions may decrease N_2O emissions. Indeed, the high-temperatures that favor NO_x

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materials, and while practical for a single vehicle would be difficult for many vehicles of many different designs.

formation may destroy N_2O emissions. Data reported by Laurikko and Nylund (1993) show that the ratio of N_2O to NO_x varies greatly during the first 505 seconds of the FTP (the "cold transient" phase of the FTP). With regard to N_2O emissions from power plants, Sloan and Laird (1990) write that "the observed correlation between N_2O and NO_x was...caused wholly by artifact production" (p. 1205). For these reasons, we do not use a NO_x:N_2O ratio, and assume that N_2O emissions per mile are the same for LDVs with the same pollution control equipment, regardless of the fuel used. This assumption undoubtedly will be shown to be incorrect, as more data become available, but for now it appears to be the only supportable assumption.

3.2.4 Artifactual formation of N_2O. As discussed below, researchers recently have discovered that N_2O may form in the containers of the gas samples waiting to be analyzed. However, this artifactual formation probably does not occur with the analysis of automobile exhaust gases, for four reasons. First, a high concentration of sulfur is necessary to promote the formation of N_2O, and gasoline does not contain enough sulfur to generate the threshold sulfur concentration required. The threshold appears to be about 600-1000 ppm; in the gasoline-vehicle tests by Prigent and de Soete (1989), the SO_2 concentration was two or three orders of magnitude lower. Similarly, Dasch (1992) states that the SO_2 concentration in vehicle exhaust is 1 about 1 ppm. Second, if the N_2O observed in vehicle tests was primarily a result of artifactual formation, and not of gas reactions across a catalytic converter, there probably would not be a significant difference between N_2O production with and without catalytic converters. Third, the production of N_2O across 3-way catalysts can be explained (see above). Fourth, Prigent and De Soete (1989) also measured N_2O on-line (i.e., without storing the samples), and the results were consistent with the results of the vehicle/chassis dynamometer tests.

3.2.5 N_2O emissions from alternative-fuel vehicles (AFVs). Based on the preceding analysis, one would expect that N_2O emissions from AFVs would be related to the operating temperature and composition of the catalytic-converter emission-control system. Virtually all AFVs built or converted to date use stock gasoline-vehicle catalytic control systems (or systems very similar to gasoline systems), and, as we shall see, emit about as much N_2O as do gasoline vehicles. Advanced catalytic-control systems made specifically for AFVs may affect N_2O emissions. Such catalysts are beginning to be developed for methanol and CNG vehicles. However, there are so few advanced, optimized AFVs, and even fewer optimized emission control systems, that at present it is impossible to estimate N_2O emissions from fully optimized AFVs.

Tests by Ford Motor Company (1988a) on two flexible-fuel Escorts and two flexible-fuel Crown Victorias (flexible-fuel vehicles can use any mixture of methanol and gasoline) show no striking relationship between N_2O emissions and the methanol content of the fuel (Table 10, and other emission results not shown here). The data do suggest that N_2O emissions increase with the age of the catalyst, but there are so few data that one cannot draw a firm conclusion. As shown in Table 10, the emission rates from the FFVs spanned the range of emissions typically measured from gasoline LDVs: The Escort emitted over 100 mg/mi, and the Crown Victoria 10-15 mg/mi. Because of this, and because N_2O emissions were not affected by the amount of methanol or gasoline fuel, we assume that methanol vehicles -- both flexible-fuel and dedicated -- emit the same amount of N_2O as gasoline LDVs (50 mg/mi). We also assume that methanol HDVs emit as much N_2O as diesel HDVs.

On the basis of the few data available (Table 10), we assume that ethanol and LPG vehicles emit as much as do gasoline vehicles, but that CNG vehicles emit less. A recent study of 36 gasoline, LPG, methanol, and CNG vans produced by Chevrolet, Dodge, and Ford demonstrates (see Table 10) that emissions of N_2O from all four vehicle types are comparable (Battelle, 1995). Only the Dodge CNG vans, which were among the first CNG vehicles produced with catalyst systems tailored for CNG exhaust, emitted significantly less N_2O than the other vehicles. These vehicles produced only 9 mg/mi of N_2O on average when new, but emissions clearly increased with catalyst age to the point where, at 15,000 miles, an average emission rate of 39 mg/mi of N_2O was measured. We assume that N_2O emission factor for CNGVs is 75% of the factor for gasoline vehicles.

We assume that hydrogen LDVs emit no N_2O, because hydrogen vehicles probably will not have catalytic converters.

Finally, we also assume that all HDVs emit at the same level.

3.3 N_2O Emissions from power plants

Power plants also emit N_2O. The power plant combustion chemistry of N_2O is quite complex, but several general trends are apparent. Higher N_2O emissions are generally associated with lower combustion temperatures, higher rank fuels, lower fuel oxygen to nitrogen ratios, higher levels of excess air, and higher fuel carbon contents (Kramlich and Linak, 1994). The exact mechanisms responsible for these relationships are still not fully understood, and in fact are somewhat controversial. Explanations for the temperature dependence of N_2O formation include lower catalytic decomposition activity at lower temperatures, lower availability of NCO (needed for one route of N_2O formation) at higher temperatures due to oxidation to NO, and higher rates of removal of N_2O through reaction with atomic hydrogen at higher temperatures (Kramlich and Linak, 1994). The dependence of N_2O formation on the other variables listed above is less strong than the dependence on temperature, and competing explanations are at least as controversial. Very likely several mechanisms are, to greater or lesser degrees, responsible for these relationships. Table 11 shows emissions of NO and N_2O as a function of type of fuel, type of combustion, combustion temperature, sulfur content, and sampling protocol.

Early tests, especially those by Hao et al. (1987), indicated that coal-fired power plants emitted relatively large amounts of N_2O. Weiss and Craig (1976) and Pierotti and Rasmussen (1976) also measured relatively large amounts of N_2O from fossil-fuel-fired power plants. However, more recent research has shown that these earlier, high N_2O values obtained for power plants are be the result of artifactual

N_2O formation. Muzio and Kramlich (1988) report that "the storage of moist combustion products containing SO_2 and NO for periods as short as 2 hours can lead to the formation of several hundreds of parts-per-million of N_2O in the sample containers where none originally existed" (p. 1369). Drying the stored flue gas reduced but did not eliminate this artifactual in-sample N_2O formation; removing the sulfur eliminated it. The presence of sulfur in the sample flue gas increased the concentration of N_2O by two orders of magnitude, from less than 5 to several hundred ppm. The mechanism for this artifactual formation of N_2O is now believed to involve the dissolution of NO_2 and SO_2 into condensed water, which then produces N_2O through a complex series of additional liquid-phase reactions (Kramlich and Linak, 1994). The effects of moisture, sulfur, and storage time on N_2O formation are illustrated in Table 11.

Importantly, there appears to be an "activation" level of SO_2. In one experiment, the concentration of N_2O in the sample remained quite low, about 15 ppm or less, with less than 600 ppm SO_2 in the furnace. At 1000 ppm SO_2, the concentration of N_2O had jumped to about 260 ppm (Muzio and Kramlich, 1988). This suggests that a low-sulfur fuel will not produce enough SO_2 to cause the artifactual, in-sample formation of N_2O[23].

Artifactual formation of N_2O can be avoided by measuring the concentration of N_2O in the flue gas in real time, without storing the sample (called "on-line" sampling), by storing the flue-gas samples for only a very short time before analyzing it, or by removing sulfur and water from stored flue-gas samples. Tests done in this way consistently find that the concentration of N_2O from high-temperature combustion systems is fairly low, typically less than 5 ppm, even for coal-fired plants. At this level, N_2O emissions from power plants contribute on the order of 1% of fuel-cycle CO_2-equivalent emissions from the use of electric vehicles.

However, it is possible that N_2O is formed in the atmosphere indirectly from direct NO emissions, possibly by the same mechanism as in artifactual in-sample formation. For example, the primary reaction responsible for artifactual N_2O formation,

$$2NO + SO_2 \xrightarrow{H_2O} N_2O + SO_3$$

might occur in the plumes of power plants and produce N_2O. However, Sloan and Laird (1990) point out that the concentration of the reactants in plumes is much less than in the flue gas samples which generate artifactual formation -- and the reaction

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[23] The discovery of the artifactual formation of N_2O, and the implication that direct N_2O emissions may not be as high as previously thought, is actually consistent with "a priori" expectations, based on combustion kinetics and experiments, about how much N_2O should be formed. In fact, explaining the earlier, higher levels was proving to be something of a problem, since N_2O formed early in the flame zone should be destroyed rapidly before it reaches the exhaust (Lyon, et al., 1989).

in the samples is already quite slow -- so that in plumes in the atmosphere, the reaction likely will be much to slow to generate much N_2O. Kokkinos (1990) agrees. Linak et al. (1990) cite previous research that suggests that such indirect N_2O formation is unlikely, but they believe that additional research is needed.

Other artifactual formation routes may exist. Prigent and De Soete (1989) have demonstrated that with combustion in the presence of sulfur (in the form of $CaSO_4$, as might result from emission control devices), the concentration of N_2O dropped with increasing temperature up to about $1200^{o}K$, then increased to a maximum of about 4 ppm at $1275^{o}K$, and then declined again. This demonstration of N_2O formation at high temperatures may reestablish the possibility of significant N_2O formation from the combustion of fossil fuels.

De Soete also suggests that in fluidized bed combustors, N_2O may be formed from reduction of NO and the oxidation of reduced sulfur compounds, via a path similar to that suspected of generating artifactual N_2O at room temperature in samples with sulfur and NO:

$$2NO + CaSO_r \text{ --> } N_2O + CaSO_4$$

The particular sulfur compound he investigated, $CaSO_4$, is reduced (and thus "prepared", in a sense, for oxidation by NO), by CO and C, above $1200^{o}K$. Above about $1400^{o}K$, N_2O is destroyed. With other sulfur compounds, or different concentrations of oxygen, N_2O formation as a function of temperature may be different.

Nevertheless, in the absence of specific evidence to the contrary, we assume that real stack gas becomes too dilute too quickly to permit significant formation of N_2O via reactions involving sulfur species.

3.3.1 Fluidized-bed combustion. Modeling and experimental results indicate that fluidized-bed combustors, which operate at lower temperatures than do utility boilers, and so produce less NO_x, may produce considerably more N_2O than boilers (Amand and Andersson, 1989; Kokkinos, 1990; Lu, et al., 1992; Makansi, 1991; Mann, et al., 1992; Ryan and Srivastava, 1989). Indeed, tests of fluidized-bed combustion that use on-line sampling or are otherwise free of artifactual N_2O formation, indicate substantially higher N_2O emissions than higher-temperature combustion processes (Amand and Leckner, 1991; Mann, et al., 1992). N_2O emissions from fluidized-bed combustors are a function of the type of fuel (coal produces more N_2O than does gas), the operating temperature (lower temperatures produce more N_2O), the excess air level (more excess air is associated with higher N_2O emissions), char particle size for coal combustion (N_2O emissions peak with particle sizes of about 1 mm), and gas residence time within and after the fixed bed (N_2O yield increases and then plateaus with increased residence time)(Lu, et al., 1992; Mann, et al., 1992; Miettinen, et al., 1995). Mann et al. (1992) conclude that N_2O emissions from fluidized-bed combustors typically are between 30 and 120 ppm, and De Soete (1993) reports a range from 30 to 150 ppm with 3% oxygen. The data shown in Table 11 confirm that these ranges encompass most data collected to date.

Fluidized-bed combustion is used at one stage in the production of gasoline: when the fluidized-bed catalytic cracker, which breaks the large hydrocarbon molecules of crude oil into the smaller molecules of gasoline, becomes coated with coke residue from the crude oil, the coke is burned off the catalysts by fluidized bed combustion (called in this case "regeneration") (Cooper and. Emanuelsson, 1992). Hence, this step in the refining process may produce non-trivial amounts of N_2O (Lyon, et al., 1989). The one test of which we are aware measured 3-26 ppm N_2O and about 400 ppm NO from a fluidized-bed catalytic cracker with a zeolite catalyst, in a modern Swedish refinery (Cooper and Emanuelsson, 1992). This concentration is lower than the N_2O concentration measured in other fluidized-bed combustors (Table 11); the authors speculate that this is due to differences in catalysts. The N_2O concentrations measured from the catalytic cracker indicate an emission rate of 0.6 to 5.0 grams N_2O per barrel of oil (Cooper and Emanuelsson, 1992; see Table 12 here). Given 0.32 barrels per day of catalytic cracking capacity per 1.00 barrel per day of crude-oil distillation capacity in the U.S as of January 1, 1990 (Thrash, 1990), 29.66 quads of petroleum products produced by U.S. refineries in 1988 (DeLuchi, 1991), and 4.848 billion barrels of crude oil input to refineries in 1988 (EIA, 1989b), we calculate that, if the Swedish emission rates are representative, 0.03 to 0.3 grams of N_2O were emitted from catalytic crackers for every 10^6 Btu of petroleum product produced in the U.S. in 1988. Assuming that other areas of refineries (aside from those that provide process heat, which are considered separately in the greenhouse-emission model used here) produce some N_2O as well, we assume a refinery-wide emission rate of 0.50 $g/10^6$ Btu-product. (It turns out that an emission rate between 0 and 1.0 has no significant effect on life-cycle CO_2-equivalent emissions.)

3.3.2 The effect of emission controls Some NO_x control strategies may increase N_2O emissions, while others may reduce them, but in general the effects are small. Laboratory investigations have shown that the sum of fuel-N conversion to N_2O and NO is quite constant over a range of temperatures in fluidized-bed combustors, despite the strong variations in the formation of each species with temperature (Pels, et al., 1995). Thus, to the extent that NO_x emissions or N_2O emissions can be reduced through temperature control measures alone, one tends to be reduced at the expense of the other. This fact has lead to the investigation of other measures than temperature control to reduce NO_x and N_2O emissions.

Linak et al. (1990) reported that combustion modifications to control NO_x emissions either did not have a significant effect on N_2O emissions, or else slightly increased them (Table 11). Nimmo et al. (1991) found that combustion-air staging in low-NO_x fuel-oil burners changed the concentration of N_2O as a function of distance from the fuel injector (and apparently slightly increased the concentration of N_2O in the flue gas), but that in both staged-and unstaged burners the final flue-gas concentration was less than 2 ppm. Lu et al. (1992) reported that combustion air staging slightly increased the concentration of N_2O from pressurized fluidized-bed combustors. Kokkinos (1990) noted that urea injection into an oil-fired boiler may have increased N_2O emissions, and Lu et al. (1992) find that NH_3 injection into a pressurized fluidized-bed combustor definitely did (Table 11). However, Sloan and Laird (1990) found no significant difference in N_2O emissions between low-NO_x and conventional burners (Table 11). Also, the use of selective catalytic reduction (SCR) appears to slightly reduce N_2O emissions (Ryan and Srivastava, 1989).

The relatively high emission rate of N_2O from fluidized-bed combustion has attracted recent attention, and research efforts have examined various methods of reducing these emissions. Two primary N_2O emission reduction strategies have been investigated: "afterburning," whereby the temperature of the cyclone is increased by the addition of extra fuel in the upper part of the combustion chamber, and "reversed air staging," which involves supplying more air than usual to the bottom part of the combustion chamber and less to the top part (Lyngfelt, et al., 1996). Both of these methods seek to reduce N_2O emission by selectively altering combustion conditions in the upper and lower parts of the combustion chamber. Conditions in the upper part are most important to N_2O formation, because any N_2O formed in the lower part is readily destroyed, while conditions in the both the upper and lower parts control NO formation and sulfur capture (Lyngfelt, et al., 1996). Experimental results suggest that afterburning can decrease N_2O emissions by up to 90%, and can also slightly reduce CO emissions, with the cost of increased fuel use and the construction of efficient afterburners. Reversed air staging can reduce N_2O emissions by up to 75%, without significant effects on other emissions, with the only cost being slightly higher power consumption (Lyngfelt, et al., 1996). These two methods could be combined, presumably resulting in even more dramatic N_2O emission reductions.

Shimizu et al. (1992) found that the addition of limestone to control SO_x emissions bubbling fluidized-bed coal combustor caused a slight decrease in N_2O emissions if single-stage air feed was used, and had no effect on N_2O if staged air feed was used. Amand and Andersson (1989) found that injecting ammonia into the combustion zone of circulating fluidized-bed combustor did not affect N_2O emissions. Since we do not know if or when any of these emission reduction measures will be adopted, we assume that N_2O emissions from fluidized-bed combustion will continue at rates typical of today's systems. We note, however, the possibility that these emissions may be reduced in the future.

3.3.3 ***Our assumptions*** Direct N_2O emissions from high-temperature combustors appear to be low. We have assumed that N_2O emissions from fossil-fuel boilers are in the upper-middle end of the range of recent estimates shown in Table 11. We use the upper-middle end to give at least a small weight to the possibility of secondary N_2O formation in post-stack gases, and of elevated emissions from plants with emission controls. We convert ppm to $g/10^6$ Btu[24], multiply by the CO_2

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[24] Based on the data summarized in DeLuchi (1991), we assume that 1 ppm N_2O converts to 1 $g/10^6$ BTU for all fuels. Although in any particular case the true conversion factor may be a factor of two or three different, this uncertainty range is not great enough to in any way change the main result that the contribution of combustion-emissions of N_2O to the CO_2-equivalent emissions of energy-use cycles is quite small.

equivalency factor for N_2O (Table 2), and end up with the CO_2-equivalent emission factors of Table 9.

3.4 N_2O emissions from other combustion sources

The data on N_2O emissions from trains, ships, NG pipeline compressors, farm equipment, loaders, petroleum refineries, well equipment, methanol plants, and other miscellaneous sources are very scarce (AP-42 does not report N_2O emissions). Consequently, for most sources, we assume that N_2O emissions depend only on the kind of fuel burned, so that, for example, the N_2O emission rate, in g/10^6 Btu, from a ship burning residual fuel oil is the same as the known rate from a power plant burning residual fuel oil. We use the residual fuel factors for diesel as well as residual fuel.

As discussed above, we assume a relatively high N_2O emission rate for petroleum refineries, due to the low-temperature FCCUs. We assume there will be N_2O emissions from methanol plants, because the catalysts used to promote methanol synthesis operate at very low temperatures compared to utility boilers, and, as discussed above, low temperatures -- at least in FBC units and across automobile catalysts -- seem to produce more N_2O emissions. However, low temperature is not a sufficient condition for high N_2O emissions, and moreover, there are no data on emissions from methanol facilities. Therefore, to check the importance of these assumptions, we also have zeroed out N_2O emissions from all sources for which there are no real data. The overall results are not appreciably different.

For N_2O emissions from coal-to-methanol plants, corn-to-ethanol plants, wood boilers, and wood gasification, see notes to Table 12. These are basically "guesstimates."

3.5 N_2O from the corona discharge from high-voltage electricity transmission lines

A "corona discharge" can occur as a result of electric fields set up by high-voltage transmission lines. The strength of these electric fields is proportional to the charge density, which tends to be high on isolated conducting surfaces with small radii of curvature, such as power lines. The electric fields will attract ions in the air with charges opposite the conductor's. The attracted ions may be accelerated enough to produce more ions upon collision with air molecules, which makes the air still more conducting, and increases the corona discharge. This process can form atmospheric trace species such as N_2O.

Hill et al. (1984) calculated that in 1980, the corona energy loss from power transmission lines in the U.S. probably created $2.3 \cdot 10^{10}$ grams of N_2O, and at most created $4.4 \cdot 10^{11}$ grams. In 1980, U.S. electric utilities delivered 2094 billion kWh of electricity (EIA, 1990). Using an N_2O-to-CO_2 conversion factor of 355 (Table **2**), the N_2O formed from the corona discharge most likely amounted to 3.9 g-CO_2-equivalents/kWh-delivered, and at most amounted to 75 g-CO_2-equivalents/kWh-delivered. The most likely estimate of 3.9 g/kWh is less than 1% of the emissions from the fossil-fuel-power-plant cycle, and only about 5% of emissions from the nuclear-fuel cycle.

The calculations of Hill et al. (1984) are a first-cut, and they point out that this source needs to better quantified. Fortunately, their most-likely estimate is so small that it does not matter if it is included in the total of greenhouse-gas emissions from electricity generation and use, or not. We have included it here. However, the maximum estimate of 75 g/kWh is of the same order of magnitude as emissions from the nuclear-fuel cycle, and hence cannot be ignored. Therefore, we have included an electric-vehicle scenario analysis using the high-end estimate for N_2O from corona discharge.

3.6 N_2O and NO_x emissions from fertilizer

Fertilizer may be used to grow biomass feedstocks, such as corn, grass and wood, that are used to make fuels such as ethanol and methanol. For example, corn production in the U.S. currently requires about 2.5 lbs of fertilizer (about 1.25 lbs nitrogen) per bushel of corn harvested for grain (DeLuchi, 1991), and short-rotation intensive-cultivation of hybrid poplar might require 50 kg-N/ha/year (Turhollow and Perlack, 1991), as well as lime and other fertilizers. Generally, a small amount of the nitrogen in the fertilizer is released to the atmosphere as N_2O and NO_x (the rest ends up in the crop, in the soil, in water, in microorganisms, or in the air as N_2). The net amount of N_2O and NO_x released depends on many factors, including: the type of biomass being grown; the amount, type, depth, and frequency of application of fertilizer; the temperature, water content, and acidity of the soil; agricultural and harvesting practices; and others (Anderson and Levine, 1987; Bowden, et al., 1991; Brumme and Beese, 1992; Conrad, et al., 1983; Eichner, 1990; Li, et al., 1994).

3.6.1 N_2O emissions N_2O is produced from complex microbial nitrification, denitrification, and decomposition processes in soils. Emission rates of N_2O for both disturbed and undisturbed soils are rather variable, depending especially on soil type and moisture regime but also on several other environmental and management factors. Increases in the amount of N added to the soil typically result in increased N_2O emissions. One study suggests a roughly linear relationship between N lost as N_2O and N input, over a range of 0 to 600 kg of fertilizer N per hectare added to several different soil types (Velthof and Oenema, 1995). Several studies have shown that typical values for the percentage of applied N that is emitted as N_2O-N range from about 0.2% to 3%, for corn, barley, and wheat fields in the U.S. and Europe, and that these emissions may represent increases of from a few to a few hundred percent above background levels (Li, et al., 1994; Mosier, et al., 1986; Velthof and Oenema, 1995). N_2O emissions are higher from saturated than from dryer soils, and peat soils and soils high in NO_2 and $CaCO_3$ content seem to have particularly high N_2O emissions (Bandibas, et al., 1994; Velthof and Oenema, 1995).

In general, researchers have a good understanding of many of the individual factors that regulate N_2O production from soils, but they cannot yet predict how these factors will interact to produce reliable N_2O emissions estimates for specific crop, soil, fertilizer, and management combinations (Mosier, 1994). Thus, the direct (i.e. the actual emission flux of N_2O from the field) and even total emissions of N_2O

(including N_2O lost off-site) from soil fertilization can in principle be quantified, but pending further study there will be significant uncertainty in estimates of both direct and indirect emissions.

Grams of N_2O from fertilizer per 10^6 Btu of fuel made available to end users can be calculated as:

$$N2O_f = N2O_e \cdot F \cdot N \cdot 453.6 \cdot 1.57 \cdot U \cdot L \cdot O \cdot V \cdot S$$

where:

$N2O_f$ = grams N_2O from fertilizer per gallon of fuel produced;

$N2O_e$ = grams N evolved as N_2O per gram N of fertilizer, on-site and during the sampling period;

F = factor to account for N_2O evolved after the sampling periods, and off-site;

N = nitrogen application rate, in lbs-N/unit-biomass (about 1.23 lbs/bushel of corn; see DeLuchi, 1991)

453.6 = gs/lb

1.57 = molecular weight of N_2O/molecular weight of N_2

U = units of biomass (e.g., bushels-corn)/gallon-ethanol

L = liquid loss factor (to account for evaporative losses of liquid fuel; this factor generally is quite small -- less than 1%)

O = own use factor (to account for fuel used as process energy, directly or indirectly, to make more fuel [this probably factor is about 2-3% in the case of gasoline from crude oil]);

V = gallons fuel/10^6 Btu;

S = the share of N_2O emissions that should be assigned to the fuel, as opposed to any other products derived from the biomass feedstock

Taking the calculation two steps further, by dividing $N2O_f$ by mpg of an ethanol or methanol vehicle, and multiplying by the N_2O-CO_2 conversion factor, produces grams-CO_2-equivalent N_2O-emissions per mile. In the case of ethanol from corn, this can be a surprisingly large number. Next, we estimates values for the variables in this equation.

$N2O_e$: Eichner (1990) and Mosier (1994) have reviewed and summarized most of the available data on N_2O emissions from soils, including many studies of N_2O emissions specifically from corn fields. These reviews show that from 0.3-2.1% of the fertilizer N applied to corn fields typically evolves to the atmosphere as N_2O-N. Averaging the results from the studies reviewed by Eichner and Mosier reveals a 1.3% mean loss rate in both cases. Mosier et al. (1986) found that 1.5% of the fertilizer N applied to corn was lost as N_2O-N (compared to 0.4% for barley), and cited an earlier study which estimated a 1.3% loss rate (0.6% for barley). Anderson and Levine (1987) calculated that 1.2% of the fertilizer applied to a corn plot in April and May was lost as N in N_2O in June, July, and August (the corn was harvested in September). These studies, as well as reviews by Eichner (1990) and Mosier (1994),

indicate that the $N2O_e$ for corn is consistently quite a bit higher than $N2O_e$ for grains and grasses, at least for soils predominantly composed of sands and clays (grasslands on peat soil can have relatively high emissions). Grass and grain fields typically emit about 0.2-1.5% of fertilizer nitrogen as N_2O, compared to 0.3-2.1 for corn (Eichner, 1990; Mosier, 1994).

It is not clear if the higher rate for corn can be explained plausibly, or if it is reasonable to expect that further research will bring the rate for corn closer to the rate for grains. It is possible that the high evolution rate for corn is due to the type of fertilizer used or to a higher percentage of excess fertilizer N relative to that needed. It might be possible to reduce the rate of N_2O emissions using less fertilizer, different fertilizers, or by adding compounds that inhibit nitrification to N_2O (Bronson, et al., 1992). Bronson et al. (1992) found that the addition of nitrapyrin to urea fertilizer reduced cumulative N_2O losses from irrigated corn fields by about 50%. Mosier and Schimel (1991) report that the application of nitrification inhibitors with urea fertilizer reduced N_2O emissions from corn and wheat fields by 72% and 58% respectively, without any loss in productivity and measured over a 10 to 18 month time period. However, the authors note that this reduction occurred at the expense of soil CH_4 uptake, which decreased by 50% in the case of the corn field and 78% in the case of the wheat field (Mosier and Schimel, 1991). Based on the available data, and uncertainty about the effect of future mitigation measures, we assume a N_2O-N loss rate of 1.3% for corn fields.

There are fewer data on N_2O emissions from fertilizer applied to woody-biomass systems. Bowden et al. (1991) note that N_2O emissions from forest soils have been related positively to chronic high rates of atmospheric nitrogen deposition (e.g., N_2O-N emissions can be 10% of added N), and to short-term additions of nitrogen (see also McKenney, et al., 1984). To determine the relationship between nitrogen additions and N_2O emissions, Bowden et al. (1991) added low (37 kg-N/ha/yr. in the first year, and 50 in the second) and high (120 kg-N/ha/yr. in the first year, and 150 in the second) amounts of NH_4 and NO_3 to 30x30 meter plots of pine and mixed-hardwood forests, and compared the N_2O emissions with emissions from control plots with no added nitrogen. The difference between N emissions from control plots and fertilized plots indicated that in the hardwood forests only 0.02-0.1% of the added N was emitted as N in N_2O, and in the pine forests, 0.03%-0.3%. Moreover, the differences between the control plots and the fertilized plots were not significant at the 5% confidence level. The authors conclude that low rates of net nitrification were responsible for the low N_2O emissions.

Brumme and Beese (1992) applied 140 kg-N/ha/year (as NH_4SO_4) to a plot in a 145-year-old Beech stand, from 1982 to 1988, and compared N_2O emissions with emissions from an unfertilized ("control") plot, and emissions from a plot that received a one-time treatment of lime 30 tonnes/ha in 1982. All plots receive 35 kg-N/ha/yr. from atmospheric deposition. The differences between the control plots and the fertilized plots indicated that anywhere from 0.2% to 3.3% of the applied nitrogen was emitted as N in N_2O, with an average of 1.6%. (The average increase in N_2O was thus $140 \cdot 0.016 \cdot 1.57 = 3.52$ kg-N_2O/ha, where 1.57 is the ratio of the molecular weight of N_2O to N_2.) The fertilized plot also emitted 2200 kg/ha more CO_2 than the control plot -- a rate of 15.7 kg-CO_2/kg-N-fertilizer applied. The limed (but not fertilized) plot emitted 6.44 kg/ha *less* N_2O than did the control plot, but 3300-kg/ha *more* CO_2. Assuming a CEF for N_2O of 355, we can summarize the results of these experiments as follows:

	Fertilized but not limed	Limed but not fertilized
1. Difference in N_2O emissions vs. control plot (kg/ha)	+3.52	-6.44
2. Difference in CO_2 emissions vs. control plot (kg/ha)	+2200	+3300
3. CO_2-equivalent of difference in N_2O emissions (kg/ha) (2 · CEF)	+1250	-2290
4. Total impact (2+3)	*+3450*	*+1010*

Robertson (1991), faced with the paucity of data pertaining specifically to short-rotation intensive cultivation (SRIC) systems, assumed the same emission rate as with fertilizer applied to agricultural soils. In the absence of specific data, we assume 0.3% (with an enhancement factor of 2.0). We consider higher values in a scenario analysis. For NO, we use the same factor as for corn.

F: Researchers expect that fertilizers continue to produce N_2O after the study period, and further, that some of the applied fertilizer is carried away by drainage and ground water and evolves N_2O off-site, beyond the measuring equipment. Eichner (1990) and the IPCC (Watson et al., 1990) double the measured, on-site N_2O emissions to account for these other avenues of N_2O production. Eichner's doubling is a guesstimate; the IPCC's (Watson et al., 1990) is based on two studies (Conrad, et al., 1983; Ronen, et al., 1988). Data in Ronen et al. (1988) indicate that as, an *upper limit*, emissions of N_2O from the drainage of fertilized land could be twice as high the average emission rate measured for corn (1.3%, as above). The authors do not say what the typical situation would be. Conrad et al. (1983) cite one study of the leaching of nitrogen fertilizer into groundwater, and another showing that groundwater may be supersaturated with N_2O, and then "guesstimate" that the N_2O emission from groundwater containing leached nitrogen fertilizer may be equal to the measured field losses.

If these doubling estimates are correct, then about 2.6% of the nitrogen in fertilizer applied to corn ends up as N_2O-N in the air. However, it must be emphasized that the accounting for off-site evolution of N_2O from groundwater and drainage water is based on virtually no data, and is extremely uncertain.

S: The "share" factor, S, which accounts for the possibility that the use of the byproducts of corn-to-ethanol will eliminate some N_2O emissions, is virtually impossible to estimate accurately. However, the factor is likely to be high; that is, it is likely that the byproducts will not "displace" nearly as much fertilizer-derived N_2O emission as will be produced by the corn farming. To understand this, one must recognize two things: first, that the correct way, in principle, to calculate S is to attribute *all* N_2O emissions from the corn farming to ethanol production, and then to deduct N_2O emissions from any crop-growing foregone because of the availability of the ethanol byproducts; and second, that not much N_2O would be so foregone. The first point follows because we are, ex hypothesis, interested in the effects of an ethanol policy, without which there would be no incremental ethanol production, and hence no incremental N_2O emissions from the soil. In other words, if we choose to make ethanol from corn, there will be N_2O emissions from the fertilizer; if we choose not to, there will be no emissions (except in the case in which ethanol is produced by diverting corn feed to ethanol production, and the feed made up by grass).

The second point follows because only some of the byproducts will substitute for other crops (distillers' dried grains and solubles may displace some soybean production, but fusel oil and ammonium sulfate will not displace crops), and because the growing of these displaced crops (such as soybeans) probably does not release as much N_2O as does the growing of corn. For example, total N_2O emissions per acre from corn fields are on average almost four times higher than total emissions from soybean fields (Eichner, 1990).[25] Further, data in Marland and Turhollow (1991) indicate that the ethanol protein byproduct associated with 1 acre of corn will displace only 0.7 acres of soybeans, so that all told, the N_2O-from-soybean credit, relative to a unit emission of N_2O from corn growing, is equal to $1.0 \times 0.7 \times 0.25 = 0.175$. This means that net (after credit) emissions of N_2O from fertilization of corn fields is about 80% of gross N_2O emission.

In the base-case analysis, we assign 80% of the N_2O emissions to ethanol production (because the byproducts effectively displace 20% of the emissions). In the case in which byproduct credits are assigned based on the energy value of all byproducts, we implicitly assign only 55% of N_2O emissions to alcohol production. The "share factor," S, for N_2O from SRIC systems should be one, because wood-to-fuel systems do not produce byproducts.

N_2O can be produced from nitrogen in atmospheric nitrogen deposited onto soils, as well as from nitrogen in added fertilizer. In fact, there is evidence of a strong relationship between chronic deposition of atmospheric nitrogen onto soils, nitrification, and increased N_2O emissions (Bowden, et al., 1991; Brumme and Beese, 1992). These N_2O emissions can be counted as an indirect global warming effect of NO_x emissions, in addition to the effect of NO_x on tropospheric O_3. Unfortunately, there are few data on the fraction of atmospheric nitrogen that is deposited into soils and then re-emitted as nitrogen in N_2O. In the absence of such data, one might assume that this fraction is the same as the fraction of applied nitrogen fertilizer that is emitted as nitrogen in N_2O (about 1%), even though for a variety of reasons this is not likely to be true. Robertson (1991), in fact, has made

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[25] These are total emissions during the sample period, not just emissions attributable to fertilizer. Corn and soybean sites were not necessarily treated similarly, and emissions were not adjusted to reflect crop output.

such an assumption. He also assumes that 1-2% of the nitrogen deposited onto marine and freshwater systems is emitted as N_2O, as a result of eutrophication.

3.6.2 NO_x There are few data on NO_x emissions from fertilizer use. Anderson and Levine (Anderson and Levine, 1987) found that 0.79% of the fertilizer applied to a corn site was lost as N in NO. Other researchers also appear to be finding that NO emissions are of the same order of magnitude as N_2O (Guenzi, 1991). However, there is considerable uncertainty at this point, and the findings are tentative, to say the least. Hutchinson and Brams (1992) applied 52-kg-N (as $(NH_4)_2SO_4$) to a Bermuda-Grass pasture and found that 0.39% evolved as N in N_2O, and 3.22% as N in NO.

Anderson and Levine (1987) did not measure NO_2, but reported a study in which as much NO_2 as NO was emitted. However, this is insufficient evidence, and so we ignore NO_2. Anderson and Levine also did not measure NO evolved offsite; however, they did find that NO was not emitted from soil saturated with water, which implies that little would be emitted from groundwater or drainage water containing fertilizer. We assume no offsite production of NO.

The calculation of NO emissions per gallon is analogous to the calculation for N_2O.

3.6.3 Other considerations Harvesting practices can affect the nutrient content of the soil, which in turn can affect the use of fertilizer. For example, if corn stover is removed from the field (and is used as an energy source in the corn-to-ethanol process), fewer nutrients will be returned to the soil. Additional fertilizer will be required to balance this loss. The use of additional fertilizer will cause additional emissions of greenhouse gases from fertilizer manufacture, and additional emissions of NO and N_2O emissions from the field. DeLuchi (1991) calculates the affect on fertilizer-related greenhouse-gas emissions of using corn-stover as an energy source in the corn-to-ethanol process rather than leaving it in the field. There may be similar effects to harvesting whole trees in SRIC systems. Hendrickson et al. (1984) note that whole-tree harvesting "has consistently been found to reduce forest floor moisture content" (p. 118), and in their own study found that it "caused significant reductions in forest floor nutrients and mineralization rates" (p. 118). On the other hand, Freedman et al. (1984) did not find significant short-term nutrient depletion after whole-tree harvesting in forest stands in Nova Scotia, but noted that the effects of successive clearcuts in SRIC systems was "unclear." Chatarpaul et al. (1984) conclude that the effects of whole tree harvesting will vary from site to site, but that "sufficient evidence is currently available regarding the detrimental effects of excess residue removal to urge a cautious, experimental approach in applying whole tree harvesting" (p. 124).

4. OZONE (O_3)

4.1 Background

Ozone is an effective greenhouse gas both in the stratosphere and the troposphere (IPCC, 1992). An important source of tropospheric O_3 in the lower atmosphere in urban areas is the complex series of photochemical reactions involving reactive nonmethane hydrocarbons (NMHCs), nitrogen oxides (NO_x), and carbon monoxide (CO). These gases are emitted from highway vehicles and other fossil fuel combustion sources (Bolle, et al., 1986). While several years ago the IPCC (Shine, et al., 1990) put forward tentative net radiative-forcing GWP estimates for O_3 precursors, the panel has since retreated from these estimates, arguing that a better understanding of precursor convection in the troposphere and the complex chemical reactions that form O_3 are necessary before O_3 precursor GWP values can be approximated (IPCC, 1995).

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 absolute increase in the concentration of O_3 in the upper troposphere causes a much greater increase in the surface temperature than does an increase in O_3 in other regions of the troposphere (Johnson, et al., 1992; Shine, et al., 1990; Wang and Molnar, 1985). This suggests that O_3 precursors, such as NO_x, emitted high in the atmosphere (e.g., from air planes), cause more warming than the same amount of emission at the earth's surface. In support of this, Johnson et al. (1992) find that the radiative forcing of surface temperature is about 30 times more sensitive to NOx emissions from aircraft than to surface emissions[26]. Similarly, Liu et al. (in Ramanathan et al., 1985) argued that most photochemical production of O_3 occurs above the boundary or inversion layer, in the "free" troposphere above, and that ground level sources of NO_x are not important in O_3 formation from 2-8 km altitude in the troposphere. If these claims are true, then emissions of O_3 precursors from ground vehicles contribute little to a greenhouse warming. On the other hand, Crutzen and Gidel (1983) claimed that the upward transfer of photochemically active species from the polluted boundary or inversion layer, at the surface, to the free troposphere above, via fast exchange episodes such as cyclones, thunderstorms, fronts, and intertropical convergence, was underestimated by the models, and could contribute significantly to O_3 formation and ultimately climate change.

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[26] No_x emissions from aircraft flying in the stratosphere likely will decrease stratospheric ozone levels, although the magnitude of the decrease is not well known (Bekki, et al., 1992).

Moreover, stratospheric O_3 also affects climate, and the concentration of O_3 in the lower stratosphere has been decreasing, due to emissions of CFCs (IPCC, 1992). This decrease could cause a warming in the surface troposphere system.

The atmospheric chemistry and activity of O_3 is complex (Bolle, et al., 1986; Crutzen and Gidel, 1983; Dickinson and Cicerone, 1986; Penner, 1990; Ramanathan, et al., 1985; Wang and Molnar, 1985; Watson, et al., 1990). 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.

4.2 Ozone from vehicles

Ozone formation in the lower troposphere due to vehicular emissions depends on the quantity of NO_x emissions, the quantity and composition of emissions of reactive (nonmethane) hydrocarbon, the relative location of NO_x and NMHCs, atmospheric conditions, and other factors (Seinfeld, 1989; Seinfeld, 1986). Enough is known about emissions of O_3 precursors from petroleum-fueled vehicles and their atmospheric chemistry to support an estimate of the contribution of gasoline and diesel vehicles to O_3 formation in urban areas in the lower troposphere. Less is known about the effect of methanol and electric vehicles, and still less about the effect of natural gas, hydrogen, ethanol, and LPG vehicles.

In the U.S., gasoline- and diesel-fueled motor vehicles are responsible for about 29% of total NO_x emissions, 27% of total NMHC emissions, and 50% of total CO emissions (U.S. EPA, 1991). Based on this, one might conclude that motor vehicles are responsible for roughly 30% of anthropogenic urban O_3 in the U.S. Alternative fuels are expected to reduce urban O_3 primarily by reducing the mass-amount or O_3-forming reactivity of NMHCs, rather than by reducing NO_x or CO emissions. In confirmation of this, models of single-and multi-day O_3 (smog) episodes have shown that, if methanol were substituted for gasoline, and the quantity and type of NMHC emissions reflected methanol use, but NO_x emissions remained the same, then peak *urban* O_3 levels would be reduced (OECD, 1993). However, the effect of methanol substitution on *global climate* would depend on the extent to which any reduction in ground level urban O_3 corresponded to a reduction in total global average O_3 concentration in the middle and upper troposphere, and on the effect on temperature of changes in the vertical distribution of O_3.

There has been little or no urban-airshed modeling of the effects on O_3 of using natural gas, LPG, or ethanol vehicles. There are only a few emissions tests of state-of-the art, single-fuel NGVs, LPGVs, and ethanol vehicles, and little is known about the long-term performance of emissions-control systems designed specifically for these fuels. The few measurements that have been made of the amounts and kinds of NMHCs in the exhaust of these vehicles, combined with current estimates of the maximum O_3-reactivity factors for NMHCs (grams of O_3 formed per gram of NMHC), indicate that, compared to NMHC emissions from gasoline vehicles, NMHC emissions from NGVs have about 25-50% of the maximum O_3-forming potential; NMHC emissions from LPG vehicles, about 60%; and NMHC emissions from ethanol vehicles, 75-80% (Auto/Oil, 1996; CARB, 1992; Chang and Rudy, 1990). However, these estimates apply only to certain O_3-forming conditions, and do not account for potential differences among AFVs in NO_x emissions. Furthermore, these estimates apply only to O_3 formation near the surface of the earth in urban areas, and do not tell us much about larger-scale changes in the concentration of O_3 at all heights in the troposphere. All told, the difficulty of modeling the relationship between ground-level emissions of O_3 precursors and changes in the global distribution of O_3 makes it very hard to estimate the O_3-related global warming effect of using alternative fuels[27].

For at least two reasons, the use of electric vehicles also could reduce O_3 formation near the surface in urban areas. First, power plants produce fewer NMHCs per mile of vehicle travel than gasoline and diesel vehicles, regardless of the fuel used at the power plant or the type of plant (Wang, et al., 1990). Second, most of the emissions attributable to use of electric vehicles would occur at night, assuming that batteries were recharged overnight, and could be dispersed by nighttime air movement aloft before the irradiation necessary for O_3 formation was most intense. Consistent with these expectations, models of O_3 formation due to substitution of electric vehicles for gasoline vehicles have shown very large decreases in O_3 formation, per electric vehicle or mile of travel (Dowlatabadi, et al., 1990; Hempel, et al., 1989).

However, while nighttime emissions may reduce smog formation in urban areas, they may not reduce O_3 formation in the free troposphere above the surface inversion layer, because the inversion layer descends at night, and emissions from high stacks are more likely to penetrate the inversion layer than are ground-level daytime emissions. And because O_3 is more effective as a greenhouse gas in the upper regions of the troposphere (see discussion above), the use of EVs may not reduce the global-warming impact of ozone as much as it reduces the concentration of ozone near the surface in urban areas.

4.2.1 Summary The contribution of motor vehicle use and electricity production to O_3-caused global warming is very complex, and has not yet been modeled fully. In its 1990 report the IPCC did calculate GWPs for NMHCs, CO, and NO_x (Shine, et al., 1990), but in its most recent work it has disavowed the GWPs and has said that "the calculation of indirect effects for a number of other gases (e.g. NOx,

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[27] For example, emissions of ozone precursors may leave an urban area, and enhance ozone formation outside the area. A fuel that produces slowly reacting NMHC may reduce urban ozone, but may increase exurban ozone, because most of the photooxidation and ozone formation will occur after the NMHCs have left the urban area.

CO) is not currently possible because of inadequate characterization of many of the atmospheric processes involved" (p. 23) (IPCC, 1995). Nevertheless, it does seem likely that options that reduce the concentration of urban O_3 will tend to reduce the global warming impact of tropospheric O_3. As noted above, we do have a rough idea of the relative O_3-forming potential of various transportation fuels and technologies. And, as discussed below, the GWPs estimated by the IPCC in 1990 (and now disavowed) would have to be in error by a very large margin in order to significantly change the results of total fuel-cycle CO_2 equivalent analyses.

4.2.2 Estimating emissions of CO, NO_x, and NMHCs from transportation fuelcycles CO, NOx, and NMHCs are regulated urban-air pollutants, as well as indirect greenhouse gases. Because of this, there is much more information on emission factors for them than for CH_4 and N_2O. The EPA's mobile-source emission-factor model, MOBILE5, estimates in-use emissions of CO, NO_x, and NMHCs from all classes of gasoline and diesel-fueled motor vehicles for any future year, taking into account the latest emissions regulations, emission-control technology, deterioration of and tampering with emission-control systems, fleet turnover, temperature, and other factors[28]. There also is an extensive body of literature on emissions of urban-air pollutants from alternative fuel vehicles; some useful summaries and data are in the Auto/Oil Program (1996), the National Renewable Energy Lab (1996), the U. S. DOE (1995), Battelle (1995), Sperling and DeLuchi (1993), Baker et al. (1992), Heath (1991), Sperling and Shaheen (1995)), and Gushee (1992).

Uncontrolled emissions of CO, NMHCs, and NO_x from a wide variety of stationary sources, including power plants and petroleum refineries, are presented in the EPA's *Compilation of Air-Pollutant Emission Factors, Volume II, Stationary Sources* (1985). We have used these emission factors extensively (e.g., Table 12). To estimate emissions from stationary sources that have emission controls, one must make assumptions about the effectiveness and extent of use of emission controls. The emission rates for power plants are shown in Table 9.

5. WATER VAPOR (H_2O)

The global water cycle and the effect of this cycle on global climate are complex. Although water vapor actually is the dominant greenhouse gas in the atmosphere -- it is much more abundant than CO_2, and controls about 10 times more infrared radiation flux (Campbell, 1986; Dickinson and Cicerone, 1986; Mooney, et al., 1987) -- *additional* water vapor emissions could cause *either* global warming or global cooling, depending on such factors as the amount of water emitted; the amount, type, extent, height, and geographic location of clouds formed;

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[28] The model is available from the Environmental Protection Agency's Office of Mobile Sources in Ann Arbor, Michigan. Evidence accumulated over the past decade suggests that the MOBILE model underestimates emissions by as much as a factor of 2 (German, 1995; Ross, et al., 1995; U.S. EPA, 1995d).

and the feedback effects of climate change on evaporation and cloud formation[29]. Complexities such as these make it difficult to represent the hydrologic cycle satisfactorily in global warming models (Kerr, 1989; Ramanathan, et al., 1989).

Water vapor in the stratosphere is a direct greenhouse gas, and it also has an indirect effect due to its critical role in O_3 formation (IPCC, 1995). The IPCC (1995) anticipates that water vapor levels with increase, both due to increasing CH_4 abundance and to increasing tropopause temperatures. Significant increases in stratospheric water vapor levels at some altitudes have been measured at one site above Boulder, Colorado, during the period from 1981 to 1994 (IPCC, 1995). More research is needed to determine if these findings are typical for other areas as well.

5.1 *Fossil fuels and the hydrologic cycle*

It is worth determining whether fossil-fuel use could be altering the global hydrologic cycle. In 1990, the world produced about 60 million barrels per day (mbpd) of crude oil, 4.8 mbpd of natural gas liquids (NGLs), about 78 trillion cubic feet of natural gas, and 5.2 billion short tons of coal, including lignite and brown coal (EIA, 1992a). Assuming that crude oil has a density of 7.3 barrels/metric tonne (EIA, 1992a) and is 12% hydrogen by weight (DeLuchi, 1991), that NGLs have a density of 10.4 barrels/metric tonne (EIA, 1992a) and contain 18% hydrogen by weight (as in propane and butane), that natural gas is 20.4 grams/standard-cubic-foot and 25% hydrogen by weight (DeLuchi, 1991), and that anthracite coal is 2.9% hydrogen by weight as received (including moisture), bituminous coal 5.5%, and lignite 6.9% (EIA, 1989a), then fossil-fuel use produced about $9.6 \cdot 10^{15}$ grams of water in 1990. This is 0.07% of the $1.4 \cdot 10^{19}$ grams of water in the atmosphere (Ahrens, 1985; Campbell, 1986), and, more pertinently, only 0.002% of the $4\text{-}5 \cdot 10^{20}$ grams of water that evaporate annually (Campbell, 1986; Lutgens and Tarbuck, 1982).

This perturbation to the hydrologic cycle due to the use of fossil fuels seems small, and likely does not significantly affect climate. To check, one can compare the fossil-fuel-caused perturbation to the hydrologic with the fossil-fuel-caused perturbation to the carbon cycle, recognizing that the perturbation to the carbon cycle eventually *will* affect climate (if it has not already). According to the IPCC (1990), the earth's atmosphere now contains about $750 \cdot 10^{15}$ grams of carbon, and each year about $197 \cdot 10^{15}$ grams of carbon are emitted from short-lived biota ($50 \cdot 10^{15}$), soil and detritus ($50 \cdot 10^{15}$), the oceans ($90 \cdot 10^{15}$), deforestation ($2 \cdot 10^{15}$), and fossil-fuel use ($5 \cdot 10^{15}$) (Bolin, et al., 1986; OTA, 1991). Thus, man-made fossil-fuel emissions of carbon are 0.7% (5/750) of the amount of carbon in the atmosphere, and 2.6% (5/190) of the amount of carbon emitted globally every year. The effect of fossil fuel use on

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[29] Water as vapor and clouds absorbs solar radiation and absorbs and emits thermal radiation (Dickinson and Cicerone, 1986), but clouds also reflect about 60% of the shortwave radiation they receive from the sun. Satellite research into the Earth's radiation budget has indicated that at present, on average, shortwave reflection wins out, so that clouds cause a net cooling of the earth (Ramanathan, et al., 1989). However, it does not follow from this that *more* clouds means more cooling. Because the incremental effect of additional clouds depends on the type, extent, height, and geographic location of the clouds (and on other factors), it might not be the same as the average effect.

global emissions of water vapor therefore is three orders of magnitude smaller than the effect on global emissions of carbon dioxide. It probably is unimportant.

Substituting hydrogen for all fossil fuels would increase emissions of water vapor, but not enough to be of much concern. If the 300 quadrillion Btu (higher heating value) of fossil-fuel energy produced and consumed worldwide in 1990 (EIA, 1992a) were replaced by 300 quadrillion Btu of hydrogen[30], then, at 141.2 kj/gram-hydrogen (higher heating value), total water vapor emissions would have been $2 \cdot 10^{16}$ grams. This is twice the amount produced by fossil use, but still tiny compared to total global water vapor emissions of $4\text{-}5 \cdot 10^{20}$ grams[31].

6. CHLOROFLUOROCARBONS (CFCS)

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). If one considers only this direct radiative forcing, then CFCs have a warming potential of thousands of times that of CO_2 (IPCC, 1995), and CFC emissions from automobiles have a warming effect of about half that of all other emissions from the entire gasoline fuel-production-and-use (DeLuchi, 1991). 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, 1995).

Ramaswamy et al. (1992) report that the depletion of stratospheric O_3 by CFCs is cooling some regions of the atmosphere, and that this cooling may counterbalance the direct warming effect of CFCs. However, it is worth noting that while the

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[30] Hydrogen energy would not necessarily substitute for fossil-fuel energy quad-for-quad. In some end uses (for example, in internal-combustion engines) hydrogen could be used more efficiently than can fossil fuels, so that less hydrogen energy than fossil-fuel energy would be needed to accomplish a given task (Ogden and Williams, 1989). Also, it would take different amounts and kinds of energy to produce, distribute, and store hydrogen than to produce, distribute, and store fossil fuels (DeLuchi, 1991). Thus, the simple calculation presented here is meant to be illustrative rather than realistic. However, a more precise calculation would not materially change the conclusions.

[31] Hydrogen *production*, as well as end use, also would affect the hydrologic cycle, although again the global effect probably would be tiny. For example, if hydrogen were produced electrolytically from water, then the amount of water vapor emitted from end use would equal the amount of water consumed for the electrolytic production of hydrogen. In this case, hydrogen use would not result in the net production of water as vapor, but rather in a redistribution of water: the hydrogen would be made from liquid water in one location, then emitted as water vapor in a different location. This perhaps could affect local microclimates.

indirect and direct effects of CFCs on climate may 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). Recent work by Daniel et al. (presented in IPCC [1995]), suggests that the net GWP values for most CFCs are on the order of 15-50% lower than the direct GWP values. For example, the 20 and 100-year GWP values for CFC-12 may each be about 77-88% of the direct values, considering uncertainties in the relative and absolute effectiveness of chlorine and bromine removal of O_3 in the lower stratosphere, and variations in the concentrations of all O_3-depleting gases in the future (IPCC, 1995). Interestingly, Daniel et al. also report that for certain compounds, such as halons, carbon tetrachloride, and methyl chloroform, indirect radiative-forcing effects may overwhelm the direct 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, 1995). It is worth noting, however, that all of these estimates are subject to considerable uncertainty, perhaps up to ±50%, and that while relative GWPs for the O_3-depleting gases will remain relatively constant, the absolute GWP values may well be revised upward, or more likely downward, as more is learned about the direct and indirect radiative-forcing effects of these complex gases.

6.1 CFCs from motor vehicles

The main source of CFCs from highway vehicles is the air conditioning system. CFCs slowly leak from this system during regular operation, and are released entirely if a collision ruptures the system or if the system breaks down in such a way as to vent the system charge. When the coolant is changed or the auto is scrapped, EPA regulations require that the refrigerant be completely reclaimed. The refrigerant most commonly used in automobile air conditioning systems was CF_2Cl_2, or CFC-12, until 1993, when some manufacturers began to switch to CH_2FCF_3, or HFC-134a. HFC-134a is now used in all new vehicles produced in the U.S., Japan, and Europe (see below). Over its life, a new or late model vehicle with air conditioning emits roughly 8.4 mg/mi of HFC-134a, and older vehicles emit similar or perhaps somewhat higher rates of CFC-12 (owing to the larger refrigerant reservoirs of vehicles made before efforts to phase-out CFCs commenced). See Table 12 for further details of refrigerant loss rates from vehicle air conditioning units.

The concentration of CFC-12 has increased from a preindustrial level of zero to about 503 parts-per-trillion volume (pptv) since the late 1970s (IPCC, 1995). The concentration was increasing at about 6%/year during the early 1980's (WMO, 1985), but due to phase-out efforts (see below) the rate of increase began to drop in the early 1990's (from 10 pptv per year in 1992 to 7 pptv per year in 1993) and by mid-1994 had almost reached zero (IPCC, 1995). However, CFC-12 has an atmospheric lifetime of over 100-years, so it will persist for quite some time even if net contributions reach zero (IPCC, 1995).

CFC-12 is currently being phased out as the working fluid in automobile air conditioners due to its strong O_3 depletion effect in the stratosphere. The November 1992 revisions to the Montreal Protocol on Substances that Deplete the Ozone Layer require all CFCs to be phased out by January 1, 1996. However, California has adopted legislation that prohibits the sale of new vehicles with CFC refrigerants after January 1, 1995, and Japanese automakers have announced that they plan to stop all use of CFCs and methyl chloroform by the end of 1994 (Walsh, 1993). The near-term replacement is tetrafluoroethane, HFC-134a (CH_2FCF_3), which does not contain chlorine and hence does not deplete stratospheric O_3. Some German automakers were among the first to replace CFC-12, converting virtually all new production from to HFC-134a in 1992-93, and other manufacturers have since followed (Chemical and Engineering News, 1994; Reichelt, 1993). Still, older vehicles with CFC air conditioning systems will remain on the road for some time. To address this issue, the EPA has enacted rules that require the recovery and recycling of O_3-depleting refrigerants (Walsh, 1993).

HFC-134a has a considerably lesser direct radiative-forcing effect 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. As a result, the net 20-year and 100-year net GWPs for HFC-134a (thought to be about half and about one-fifth, respectively, those for CFC-12 (IPCC, 1995)) are higher than direct radiative-forcing effects alone might lead one to believe (see Table 2 for direct and net GWP values for CFC-12 and HFC-134a and section 8.6 for a more complete description of the GWP values for CFC-12 and HFC-134a). The use of HFC-134a in place of CFC-12 is therefore an improvement from a global climate change perspective, but each HFC-134a molecule still has hundreds to thousands of times the GWP of a CO_2 molecule. Already, automakers are looking for long-term alternatives to HFC-134a (Reichelt, 1993).

Theoretically, the transition from CFC-12 to HFC-134a might involve the additional complication of an associated air conditioner efficiency loss, and the subsequent increase in emissions of fuel-related greenhouse gases. The use of HFC-134a could result in lower efficiency (defined as the product of the amount of air cooled and the magnitude of cooling, divided by the quantity of energy consumed) because, while thermodynamically similar to CFC-12, HFC-134a is not miscible with the mineral oils currently in use and is somewhat less efficient with most substitute lubricants (Fischer and McFarland, 1992a). Thus, HFC-134a would have a lower direct effect on radiative forcing due to its lower GWP, relative to CFC-12, but this benefit could be eroded to some extent by its lower efficiency. In order to assess the net effect of a GWP benefit and an energy efficiency loss, the concept of total equivalent warming impact (TEWI) was developed. The computation of TEWI values includes net GWP effects, the mass of refrigerant needed, and the energy use associated with the use of the refrigerant. One study of the TEWI of HFC-134a relative to CFC-12 suggests that even with the small loss in efficiency, the TEWI value of HFC-134a would be only 16% of that of CFC-12 (17,000 lbs of CO_2 equivalent emissions versus 108,000 lbs), based on 500-year GWP values (Fischer, et al., 1992b).

This analysis used direct rather than net GWP values, so it is reasonable to assume that the 16% figure could be revised upward to about 20%, based on the net GWP values reported by the IPCC (1995).

However, the recent experience of the Ford Motor Company suggests that design improvements in the heat exchanger units and other components of automobile air conditioners have compensated for the difference in the theoretical efficiencies of CFC-12 and HFC-134a systems (Wallington, 1996). As a result of this recent air conditioner system efficiency improvement (about 5% from improved heat exchanger design alone), the same amount of HFC-134a refrigerant (about 2 pounds for an average vehicle) is used now as was used of CFC-12 shortly before the transition, and the energy use difference between new HFC-134a systems and late model CFC-12 systems is not detectable (Wallington, 1996).

Thus, under today's strict regulations for reclamation of CFC and HFC refrigerants, CFC-12 and HFC-134a can only be emitted to the atmosphere in four ways. First, the refrigerant charge can be completely vented in the event of a collision that damages the air-conditioning system. Second, air-conditioning systems can malfunction or fail over time, resulting in partial or complete venting. Third, while regulations require that refrigerant from scrapped vehicles be reclaimed, some amount of illegal scrappage undoubtedly occurs and the refrigerant from these vehicles will eventually be released. Finally, a very small amount of refrigerant is released during the refrigerant reclamation or recharging process itself because the gas in a few inches of hose, between the hose valves and the ends of the connectors, is released when the hoses are disconnected. Thus, some vehicles will never completely vent the refrigerant charge to the atmosphere, while others, with malfunctioning air conditioner systems or that are involved in collisions, may completely vent the refrigerant charge more than once. All vehicles will vent at least trace quantities of refrigerant at various times when their air conditioning systems are recharged, or permanently decomissioned for vehicle scrappage, but with modern reclamation systems these emissions are negligible.

We assume that each new gasoline LDV, on average, vents its refrigerant charge once during its lifetime. This assumption results in an HFC-134a emission rate of 8.4 mg/mi (Table 12), which, when multiplied by the CO_2 equivalency factor assumed in Table 2, results in CO_2 equivalent emissions of only 17 g/mi -- nearly 20 times less than CO_2 emissions from fuel combustion from a typical current-model car.

This 8.4 mg/mi emission rate is, in theory, a function of the type of coolant used, charging and maintenance practices, and the life of the vehicle, but not, presumably, of the type of fuel used in an LDV. Thus, all AFVs using the same type of cooling system will be responsible for the same amount of refrigerant-caused global warming.

7. AEROSOLS

Aerosols are not greenhouse gases, but rather are small particles and droplets in the atmosphere. Aerosols may be composed of dust particles, or of other particles and droplets with a wide variety of different chemical compositions, and 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), and 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. 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, 1995).

7.1 Aerosols and climate change

While some aerosols tend to warm the earth's surface, the net climatic effect of anthropogenic aerosols is believed to be a negative radiative forcing. However, considerable uncertainty still surrounds some aspects of aerosol behavior, particularly with regard to indirect effects on cloud properties. The radiative forcing due to aerosols depends on the chemical composition, size, and shape of aerosol particles, and on the spatial distribution of the aerosol (IPCC, 1995). Due to the inhomogenous spatial distribution of aerosols, and the importance of the details of this distribution to their climatic effects, it must be remembered that globally averaged radiative forcing estimates (such as those provided below) are of even more limited utility for aerosols than for greenhouse gases.

Considerable progress has recently been made in understanding the behavior of tropospheric aerosols, and, based on new calculations of the spatial distributions of soot and fossil fuel combustion aerosols, the IPCC (1995) reports that the total direct radiative forcing for anthropogenic aerosols (including sulfates, soot, and organic compounds from biomass burning) is approximately -0.5 Wm^{-2}, with a likely range from -0.25 to -1.0 Wm^{-2}. The largest component of this negative, direct forcing is sulphate aerosols from fossil fuel combustion and smelting activities, with an IPCC (1995) estimated forcing of -0.4 Wm^{-2}. Other estimates for the magnitude of this direct sulfate aerosol forcing range from -0.3 to -0.9 Wm^{-2} (Mitchell, et al., 1995). The direct radiative forcing from particles produced by biomass burning is believed to be about -0.2 Wm^{-2} (with a range of-0.07 to -0.6 Wm^{-2}), and the direct forcing from fossil fuel produced soot aerosols is, believed to be slightly positive, at 0.1 Wm^{-2} (with a range of 0.03 to 0.3 Wm^{-2})(IPCC, 1995). 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, 1995).

The indirect effects of aerosols on cloud properties are not well understood, but they are believed to result in a negative radiative forcing in the range of 0 to -1.5 Wm^{-2} (IPCC, 1995). 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 (1995) uses an average value of -0.8 Wm^{-2} for aerosol indirect effects in some scenario analyses.

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). 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).

8. CO_2 EQUIVALENCY (CEF) CALCULATIONS

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 -- CH_4, CO, N_2O, NMHCs, NO_x, CFC-12, and HFC-134a -- are converted into the mass amount of CO_2 emissions that would have the same impact on climate or the economy. The first CO_2-equivalency factors (CEFs) developed equated emissions solely on the basis of global warming (in terms of degree-years), and hence were called "global warming potential" factors, or GWPs. More recently, researchers have estimated equivalency factors that equate emissions on the basis of the economic impacts of the warming (in terms of the present dollar value of impacts). These factors have been dubbed "economic damage indices," or EDIs. Table 2 summarizes the most recent estimates of GWPs and EDIs, and shows the assumptions used in this analysis[32].

The EDIs potentially are a big step beyond GWPs because they incorporate the present value of the economic damages of future global warming, whereas the GWPs reflect only the future warming. However, the EDIs developed thus far do have some shortcomings: those of Reilly (1992) are based on now-outdated estimates of climate effects, and those of Hammit et al. (1996) do not include the indirect effect of CH_4 or halocarbons on H_2O or O_3, or the effects of CO_2 fertilization. As a result, one still must refer to the GWPs in order to estimate CEFs for the other GHGs. For this reason, and because the issues involved in the calculation of GWPs also are

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[32] To convert the GWPs or EDIs from a mass basis to a mole basis, multiply 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.

involved in the calculation of EDIs (because GWPs in essence are partial EDIs), we will discuss the GWPs in detail.

8.1 The calculation of GWPs.

To calculate a GWP, one needs to know, for both CO_2 and non-CO_2 gases, the relationship between equilibrium surface temperature and equilibrium 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 (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: since one is equating "degree-years" of warming over a period of time, the equation will depend on the length of time chosen, and this choice is important.

Sophisticated estimates of GWPs are available in several recent papers (IPCC, 1995; 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 O_3 depletion potentials for CFCs. The IPCC distinguishes 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 (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:

$$GWP_i = \frac{\int_0^t a_i(t) \times c_i(t) / m_i \, dt}{\int_0^t a_{co_2}(t)dt \times c_{co_2}(t) / 44.01 \, dt}$$

where:

GWP_i = the global warming potential for gas i, relative to CO_2 (on a mass basis)

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

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

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

44.01 = the molecular mass of CO_2 (g/mol)

$a_{co_2}(t)$ = the radiative forcing of CO_2

$c_{co_2}(t)$ = the average ambient concentration of CO_2 over time

t = the length of time (years into the future) considered

All papers express c(t) as an exponential decay function; in Lashof and Ahuja (1990) this function is:

$$c(t) = e^{\left(\frac{-t}{r_i}\right)}$$

where:
t = defined as above
t = defined as above
r_i = the *average* residence time of gas i, in years (note that this is the average residence time, not the actual residence time of every unit of emission)

To obtain a total GWP, which accounts for indirect warming effects as well as direct effects, one either can include in the expression above the indirect warming effects (e.g., one can include in the radiative forcing term, ai, the indirect as well as the direct forcing, as in Lashof and Ahuja, 1990), or calculate indirect effects separately and add them to the direct effects.

Not surprisingly, there is disagreement about several of the key parameters in the calculation of total GWPs, including: the effective lifetime of CO_2 and other greenhouse gases, the relative radiative forcing of CH_4 and other gases, 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. All of these problems are discussed in Shine et al. (1990); the last two, which probably are the most serious, are reviewed briefly here.

8.2 Indirect global warming effects of gases

The production, alteration, and destruction of direct and indirect greenhouse gases can affect the concentration, distribution, and lifetime of several direct greenhouse gases, including CO_2, CH_4, H_2O, and O_3. Unfortunately, the many interactions between species such as NO_x, CO, NMHCs, OH^-, O_3, and H_2O are incompletely understood and difficult to model. Current calculations of GWPs consider only a few such interactions, and generally only simply. Because of the difficulties in accurately modeling these interactions and their effects on global warming, the IPCC (1992) has stated 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). However, this does not mean that *in principle* there are no GWPs for the non-CO_2 greenhouse gases; rather, it means that some of the GWPs are uncertain, and that the earlier IPCC estimates of the GWPs may or may not turn out to be right. Moreover, it turns out, as discussed below, that for CO and NMHCs the uncertainty in the indirect GWP would have to be quite large in order to significantly affect calculated total CO_2-equivalent emissions for a fuelcycle.

8.3 GWP values for carbon monoxide (CO)

The IPCC's 1990 GWP values for CO consisted of two parts: one part that accounted for the eventual oxidation of CO to CO_2, and another that accounted for the role of CO in O_3 formation. The part of the GWP that accounted for oxidation to CO_2 was not uncertain, and over long time horizons it would be the larger of the two parts. The part of the GWP that accounted for the effect on O_3 was small absolutely, but more uncertain. The IPCC estimated in 1990 that this indirect GWP was close to zero over 500 years, and equal to only 1 to 2 over 100 years (Shine, et al., 1990). Due to the uncertainty in this GWP component, however, the IPCC soon disavowed these early estimates.

More recently, Martin and Michaelis (1992) have estimated an even lower indirect GWP for CO (Table 2). But even if these estimates of the indirect GWP of CO are too low by a factor of two, the effect still will be relatively minor. To confirm this, we recalculated some of the base-case emissions totals reported in DeLuchi (1991), assuming now that the O_3-related warming effect of CO is twice as big as estimated by the IPCC in 1990. In most cases, total gram/mile CO_2-equivalent emissions were within 1% (in a few cases, 4-6%) of the results based on the 1990 GWP. In every case, the *percentage change* relative to petroleum was within 1 percentage point of the percentage change calculated in DeLuchi (1991). Eliminating entirely the indirect GWP for CO had an even smaller effect. Thus, unless the ozone part of the GWP for CO has been terrifically underestimated, the true GWP or CEF for CO is not likely to be different enough from the value assumed in Table 2 to have a significant effect on estimated CO_2-equivalent emissions.

In its most recent report, the IPCC concludes that the characterization of the atmospheric processes involved in tropospheric O_3 formation is at this time inadequate to allow for the calculation of the indirect GWP for CO (IPCC, 1995).

8.4 GWP values for non-methane hydrocarbons (NMHCs)

The GWPs for NMHCs are similar to those for CO, in that they consist of one straightforward component that accounts for oxidation of the carbon to CO_2, and a second uncertain component that accounts for indirect effects of NMHCs on the production of O_3. Performing the same sort of sensitivity analysis as with CO (above), we found that even if the indirect-effect part of the CEF for NMHCs is 50% higher than the indirect GWP estimated by the IPCC in 1990 -- or, if the indirect GWP is set equal to zero -- the standing of all the nonpetroleum fuel cycles relative to the petroleum cycles was virtually identical to the relative standing based on the 1990 GWP. Therefore, unless the ozone component of the 1990 GWP for NMOCs is much, much too low, the true GWP or CEF for CO is not likely to be different enough from the value assumed in Table 2 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. Note that Martin and Michaelis' (1992) estimate of the total 50-year GWP for NMHCs is considerably *less* than the IPCC's (Shine, et al., 1990) 100-year estimate (Table 2).

The IPCC in 1990 gave generic GWPs for NMHCs; it did not provide specific factors for individual hydrocarbons, and probably did not intend that the NMHC factor apply as such to emissions of alcohols. In order to have a GWP/CEF that includes alcohols and can distinguish, crudely, between individual hydrocarbons, we have estimated a GWP/CEF on a carbon-weight basis. There are two reasons for this: first, part of the warming effect of NMHCs and alcohols (indeed, of any organic compound) is due to the eventual oxidation of the carbon to CO_2; hence, part of the mass-based warming effect of any organic compound fully oxidized in the atmosphere will be a function solely of the carbon content. Second, it appears that, at least for some NMHCs, the higher the carbon number, the higher the ultimate O_3-forming potential. Seinfeld (1989) notes that higher-carbon alkanes consume more NO, which shifts the NO, O, O_2, O_3, NO_2 equilibrium in favor of O_3. Also, within any family of compounds, reactivity (an indicator of ozone-forming potential) increases with carbon number.

To convert the IPCC's 1990 GWP for NMHCs to a carbon (C) basis, we assume that the IPCC based its GWPs for NMHCs on the current mix of ambient hydrocarbons, which the IPCC says is mostly terpenes, isoprenes, and alkenes (Shine, et al., 1990). For this mix, the average carbon weight content is about 85%. Thus, to estimate our carbon-based CEF for NMHCs, we divide the GWPs for NMHCs by 0.85. The most important effect of expressing the NMHC GWP on a carbon basis is to exclude the weight of oxygen in methanol and ethanol emissions.

As with CO, the IPCC now concludes that the short atmospheric lifetime, unknown spatial distribution, and uncertain source/sink relationships of NMHCs makes the calculation of indirect GWP values (and thus net GWPs) impossible at this time (IPCC, 1995). Thus, the methodology described above is employed until revised GWP values for NMHCs are published, perhaps based on future climate models with higher degrees of spatial resolution.

8.5 GWP values for methane (CH_4)

The IPCC's most recent estimates (1995) of GWP values for CH_4 (See Table 2) are net estimates that include CH_4's indirect role in O_3 formation in the troposphere and water vapor formation in the stratosphere. In 1990 the IPCC estimated direct GWP values, and then estimated that the indirect effects was slightly more than 100% of the direct effects (Shine, et al., 1990). In 1992, the IPCC did not re-estimate the indirect-warming component, and stated only that it could be "comparable in magnitude to the direct value" (p. 15). Also in 1992, the 1990 estimate of the direct-warming GWPs for CH_4 were revised upward by 10-20% (IPCC, 1992). The end result of these re-estimates of both the direct and indirect radiative forcing effects of CH_4, and also a recent downward revision in the atmospheric pulse adjustment rate for CH_4 due to the inclusion of CH4 uptake by soils and an 11% faster than previously thought chemical removal rate (resulting in a mean value of 12 years compared to the old estimate of 14.5 years), is that the 1995 IPCC CH_4 net GWP values are on average about 16% lower than the 1990 net GWP estimates. The 1995 20-year GWP value is 11% lower, the 100-year value is unchanged, and the 500-year value is 38% lower (IPCC, 1995).

Despite these important revisions, however, the GWP values for CH_4 are still uncertain (the IPCC currently estimates they are within about ±35%). Given the importance of CH_4 as a greenhouse gas, and the variation in CH_4 emissions among vehicle types, this uncertainty should be kept in mind when considering the CO_2-equivalent results presented here.

8.6 GWP values for nitrous oxide (N_2O) and oxides of nitrogen (NO_x)

NO_x. NO_x contributes indirectly to global warming in at least two ways: it participates in the photochemistry of ozone, which is a direct greenhouse gas, and it deposits onto soils and denitrifies to N_2O, which also is a direct greenhouse gas. The difficulty of estimating the contribution of NO_x emissions to global warming via the production of O_3 and N_2O make the GWP values for NO_x among the most uncertain of the GWPs. Because the uncertainty is relatively large, and because most fuelcycles produce substantial amounts of NO_x, the uncertainty regarding NO_x GWP values has a non-trivial effect on estimates of fuel-cycle CO_2-equivalent emissions.

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 2 shows the corrected values for NO_x.) At the same time, the IPCC (1992) noted that NO_x emissions have indirect cooling effects. Consistent with this, Martin and Michaelis (1992) estimated a very low 50-year GWP for NO_x (Table 2). Note, though, that neither the corrected estimate by the IPCC nor the estimate by Martin and Michaelis account for the deposition of atmospheric nitrogen onto soils and the re-emission of the nitrogen as N_2O.

Our estimate of the GWP, shown in Table 2, is calculated as follows:

$$GWP_{NO_x} = IOZ + \left(NO_{2(air)} \to N_{soil}\right) \cdot \left(N_{soil} \to N_2O_{air}\right) \cdot \frac{44}{46} \cdot GWP_{N_2O}$$

where:

GWP_{NOx} = the global warming potential for NO_x (as NO_2) emissions, on a mass basis

IOZ = the GWP of the indirect effect of NO_x on ozone (we adopt the estimates of Martin and Michaelis [Table 2])

$NO_{2(air)} \to N_{soil}$ = the fraction of N in ambient NO_2 that is deposited onto soils (we assume 0.50; see discussion below)

$N_{soil} \to N_2O_{(air)}$ = the fraction of the deposited N that is re-emitted to the atmosphere as N_2O (we assume 0.015; see discussion below).

44/46 = the ratio of the molecular weight of N_2O to that of NO_2

GWP_{N2O} = the global warming potential for N_2O on a mass basis (Table 2)

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) (U.S. EPA, 1996). Given that ambient NO_X levels are much higher over land than over oceans, it is reasonable to assume that about 50% of the emitted nitrogen in NO_X eventually is deposited on soils, rather than on oceans. Hence, we assume that the parameter $NO_{2(air)} \rightarrow N_{soil}$ is equal to 0.50.

As discussed above, approximately 0.1% to 3.0% of the nitrogen in applied commercial fertilizer eventually is re-emitted as nitrogen in N_2O, depending on the soil, vegetation, fertilizer, weather, and other factors. Because the nitrates deposited from the atmosphere are similar to or the same as the nitrates in applied commercial fertilizer, it is reasonable to assume that the fraction of deposited N that is re-emitted as N_2O is the same as the fraction of applied fertilizer N that is re-emitted as N_2O. Hence, we assume that the parameter $N_{soil} \rightarrow N_2O_{(air)}$ is equal to 0.010.

N_2O. The 1995 IPCC GWP values are slightly lower than the 1994 IPCC estimates, and somewhat higher than the 1990 estimates.

8.7 GWP values for CFCs and replacements

As described in section 6, the refrigerant used in new automobile air conditioners was CFC-12 for many years, but is now HFC-134a, due to the provisions of the Montreal Protocol. However, all vehicles produced before 1993 were built to use CFC-12, and very few of these older vehicles have been retrofitted to use the new refrigerant. Thus, the majority of vehicles on the road today still use CFC-12, but by 2000 a much greater percentage, perhaps a majority, of vehicles will be using HFC-134a.

The IPCC (1995) has recently developed net GWP values for O_3-depleting gases, including CFC-12. In earlier work, the IPCC presented only the direct GWPs for O_3-depleting gases, thereby describing the gases' effective absorption of infrared radiation only. Of course, these gases also deplete stratospheric O_3, which produces a negative radiative forcing, particularly for O_3 loss near the tropopause (Ramaswamy, et al., 1992). Thus, a complete calculation of GWP values for O_3-depleting gases should include a consideration of both direct and indirect effects.

The GWP values for CFC-12 presented in Table 2 reflect indirect as well as direct effects. The first values shown are the GWPs for direct effects only, which are 7800 for the 20-year time horizon and 8100 for the 100-year time horizon. Shown below these values, for both 20-year and 100-year time horizons, are ranges of values for the net GWPs. The ranges are necessary to illustrate the uncertainty in two important parameters related to the indirect effects of the O_3-depleting gases. These two uncertain parameters 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). Of these two parameters, the uncertainty in the former has a smaller effect on the net GWP values for CFC-12, producing a range of 20-year GWP values of from 6400 to 6800, while the uncertainty in the latter has a somewhat greater effect, producing a range of 6000 to 6800 (IPCC, 1995). The corresponding values for the 100-year time horizon are a range of from 6600 to 6800 due to the uncertainty in bromine versus chlorine O_3 removal efficiency, and from 6200 to 7100 for uncertainty in lower stratosphere cooling (IPCC, 1995). Finally, it is important to note that in calculating these values, Daniel et al. (1995) 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.

Many other complex pathways may exist for O_3-depleting gases to have indirect effects. For example, altered levels of ultraviolet radiation, resulting from O_3 loss, 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, 1995). Unexplored effects such as these suggest that the net GWPs for O_3-depleting gases may well be lower than the current values. Due to uncertainties in the direct effects of O_3-depleting gases, however, the net GWPs may also be larger than indicated. This is considered less likely by the IPCC, but the overall level of uncertainty is believed to be on the order of ±50% (IPCC, 1995). 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.

The GWP values for HFC-134a, also shown in Table 2, reflect direct effects only because HFC-134a does not deplete stratospheric O_3.

8.8 An approximate GWP formula for anthropogenic, tropospheric aerosols

As discussed in Section 7.1, considerable uncertainty still exists for both the direct and indirect climatic effects of aerosols emissions. 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. Such properties as aerosol scattering efficiency and upscatter fraction depend in turn on particle size, composition, and relative humidity (Pilinis, et al., 1995). The complex and incompletely understood nature of these relationships suggests that recent estimates of direct forcing effects from anthropogenic sulfate aerosols (such as those provided in Section 7.1) 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, 1995; Nemesure, et al., 1995). With regard to the indirect effects of aerosols on cloud properties, the uncertainty in climatic effect is even more profound. 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, 1995). 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. At this time, the IPCC (1995) concludes that the indirect effects of aerosols result in a zero or negative radiative forcing, in the range of 0 to -1.5 Wm^{-2}, but also indicates that the understanding of these effects is sufficiently poor to preclude a mid-range estimate.

Thus, due to the above uncertainties, no precise estimate of the net effect of anthropogenic aerosols on global climate is possible at this time. We expect that reasonably sophisticated estimates of GWPs for aerosols will be available relatively soon. Until they are, though, it might be useful to estimate rough, order-of-magnitude GWPs on the basis of a simple scaling relationship:

$$GWP_i = \frac{\frac{R_i}{C_i} \cdot I_i \cdot L_i}{\frac{R_{CO_2}}{C_{CO_2}} \cdot I_{CO_2} \cdot L_{CO_2}}$$

where:
GWP_i = the approximate global warming potential for one type of tropospheric aerosol
GWP_{CO_2} = the global warming potential for CO_2, normalized to 1.0
R_i = the direct radiative forcing of the tropospheric aerosol
I_i = multiplier effect for indirect effects of the aerosol
L_i = the average lifetime of the aerosol
C_i = the average ambient concentration of the aerosol, in $\mu g/m^3$
R_{CO_2} = the direct radiative forcing of CO_2
I_{CO_2} = multiplier for the indirect effects of CO_2
L_{CO_2} = the average lifetime of CO_2
C_{CO_2} = the average ambient concentration of CO_2.

Of these parameters, the most uncertain are R_a, I_a, and C_a. The parameter L_a, the average lifetime of tropospheric aerosols, is variable but falls within known limits. This lifetime is generally rather short compared to the residence times of greenhouse gases, on the order of 4 to 5 days for sulfate aerosols for example, 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 a 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 Pilinus 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. Based on particle size distribution data, a generalized formula in Jaenicke (1993) that relates residence time to particle radius, and studies of sulfate aerosols, we assume a mean residence time of seven days for tropospheric aerosols.

The direct radiative forcing of aerosols is believed to be on the order of -0.4 Wm^{-2} for sulfate aerosols and 0.1 Wm^{-2} for soot aerosols (IPCC, 1995). The uncertainty in the sulfate aerosol parameter is believed to be on the order of a factor of two, and for soot aerosols the uncertainty factor is believed to be about three. For aerosols from biomass burning, the other primary tropospheric aerosol type addressed by the IPCC, direct radiative forcing is believed to be on the order of -0.2 Wm^{-2} with an uncertainty factor of three (IPCC, 1995). Arriving at a net direct radiative forcing estimate for all anthropogenic, tropospheric aerosols is complicated by several factors. First, these direct forcing estimates are based on partial derivative analyses of individual aerosol components, as if each aerosol component has an individual effect that is separate from the other components (IPCC, 1995). In fact, aerosol components are likely to interact and to produce net effects that may not be the sum of the effects of individual components. Second, for some aerosols, such as those from biomass burning, there is uncertainty about the relative contribution of anthropogenic and biogenic sources. Thus, sorting out the direct forcing resulting from anthropogenic sources alone is difficult. Third, in addition to the primary anthropogenic aerosols discussed above, aerosols can also form through secondary processes, such as the formation of nitrate aerosols from NO_x emissions (IPCC, 1995).

These complications notwithstanding, we estimate two different GWP values for tropospheric aerosols using the above formula. The first value is for sulfate aerosols, and the second is for soot aerosols. Our very preliminary estimates of the lifetime, radiative forcing, indirect effect, and concentration result in a relatively small negative GWP for sulfates, and a relatively small positive GWP for soot. This suggests that the effect on climate of a gram of particulate matter emission is minor compared to the effect of a gram of CO_2.

We do not develop a GWP value for aerosols from biomass burning because of the uncertainty in the relative contributions of anthropogenic and biogenic sources, and because biomass burning is not a significant factor in any of the fuelcycles assessed here (with the possible and minor exception of burning agricultural residues in the biomass to ethanol processes).

8.9 The time period considered in the estimation of a 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 CO_2. However, over a much longer period, the greenhouse gas will have been gone from the atmosphere for most of the time (or will 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 2, 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). Elsewhere, 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. We suspect 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. On the other hand, the use of an EDI instead of a GWP requires the use of a discount rate rather than a time cutoff.

In any case, if one uses GWPs (rather than EDIs) as CEFs, we recommend a 100-year horizon because the case for a relatively long time horizon is obvious: global warming is a very long-term problem, and will affect many generations to come. For example, the deep ocean, which stores CO_2, turns over on a time scale of centuries. Therefore, to account for most of the climatic effects of current emissions of greenhouse gases, one should extend the analysis for decades at least; otherwise, the warming effects of some emissions will be ignored.[33]

8.10 An economic damage index as an alternative to the GWP index

Recently, Hammitt et al. (1996) have proposed an economic damage index (or EDI) as an alternative approach to determining the relative importance of different greenhouse gases. In addition to accounting for the relative lifetimes and radiative forcing potencies of different greenhouse gases, the EDI goes a step further to compare 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 for important gas effects that are unrelated to forcing – such as stratospheric ozone depletion from CFCs or CO_2 fertilization of crops.

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[33] 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 (or, logically, must be) 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 average rate of warming is just a back-calculated statistic; 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 in any way 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. In principle EDIs do this, and hence are superior to GWPs.

The EDI, as defined by Hammitt et al. (1996), is the reduction in emissions of a standard 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) define the EDI equation as follows:

$$EDI_i = \frac{\frac{\partial}{\partial e_i} W[C(t)]}{\frac{\partial}{\partial e_o} W[C(t)]} = \frac{\int_0^\infty \Delta C_i(t) \cdot \lambda_i(t) dt}{\int_0^\infty \Delta C_o(t) \cdot \lambda_o(t) dt}$$

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. The damage function is as follows:

$$W[C(t)] = \int_0^\infty \left(\frac{1}{1+r}\right)^t \cdot \alpha \cdot GDP(t) \cdot D[\Delta T(t)]\, dt$$

where:
$W[\Delta T(t)]$ = economic damages from a change in average global temperature
r = the discount rate
α = a scaling constant
GDP(t) = gross world product
$D[\Delta T(t)]$ = economic damage function, related to magnitude of temperature change
$\Delta T(t)$ = 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 ΔT, 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 the 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 ΔT. 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 they 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 2. 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., 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). 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. In general, the "middle case" suggested by the authors produces a CH_4 index that is about one-half the 100-year GWP, while the EDI values for N_2O and CFC-12 are comparable to the GWP values, suggesting that the general effect of the EDI index is to diminish the importance of the shorter-lived gases.

8.11 Summary of our assumptions.

We use Hammitt et al.'s 'middle-case' estimates for CH4, N2O, and CFCs, and our own rough estimates of EDI values for ozone precursors, tropospheric soot and sulfate aerosols, and HFC-134a. We use some extreme EDI values in scenario runs, and these results are discussed below.

9.0 THE CONTRIBUTION OF NON-CO_2 GREENHOUSE GASES TO FUELCYCLE CO_2-EQUIVALENT EMISSIONS

Presented in Delucchi (1996).

TABLE 1: FEEDSTOCKS, FUELS, EMISSION SOURCES, AND GREENHOUSE-GASES CONSIDERED IN THE ANALYSIS

Feedstocks	*End-use fuels*	*Sources of gases*	*Greenhouse gases*
petroleum	gasoline	feedstock production	CH_4
natural gas	diesel fuel	feedstock transport	N_2O
coal	residual fuel oil	fuel manufacture	CO
uranium	CNG or LNG	fuel distribution	NMHCs
corn	methanol	fuel end-use	NO_x
woody biomass	ethanol		H_2O
solar energy	LPG		CFCs
	hydrogen		
	electricity		

CNG = compressed natural gas; LNG = liquefied natural gas; LPG = liquefied petroleum gases (mostly propane).

TABLE 2: ESTIMATES OF TOTAL GLOBAL WARMING POTENTIALS (GWPS) AND ECONOMIC DAMAGE INDICES (EDIS) FOR NON-CO_2 GREENHOUSE GASES (MASS BASIS)

GWPs	CH_4	N_2O	CFC-12	HFC-134a	CO	NMHC	NO_2
IPCC[a]							
20-year horizon	56	280	7,800 (direct) 6,400-6,800 6,000-6,800	3,400	7	31	30[b]
100-year horizon	21	310	8,100 (direct) 6,600-6,800 6,200-7,100	1,300	3	11	7[b]
500-year horizon	6.5	170		420	2	6	2[b]
Martin & Michaelis (1992)[c]							
50-year horizon	26.5	270			3	8.8	3
Bruhl (1993) *(1993)*[d]							
50-year horizon	10-13						
100-year horizon	6-8						
EDIs							
Reilly (1993)[e]							
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)[f]							
Middle case	11.0	354.8	9,067				
Damage exponent = 1	27.21	354.7	9,279				
Damage exponent = 3	5.10	340.1	8,527				
'Hockey stick' damages	6.07	319.4	7,910				
Low climate sensitivity	10.03	353.4	9,028				
High climate sensitivity	12.33	356.6	9,142				
Discount rate = 1%/yr.	3.73	322.2	7,950				
Discount rate = 5%/yr.	23.70	366.2	9,596				
IS92c emission/GDP	22.16	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	49.69	296.7	7,286				
Maximum	2.92	403.6	10,507				
Our equivalency factors[g]	22	355	8,000	2,000	2	5	4

[a] The results for CH_4, N_2O, CFC-12, and HFC-134a are from IPCC (1995). The estimate for CH_4 includes the indirect effects of tropospheric O_3 production and stratospheric water vapor production. The first GWPs shown for CFC-12 are for the direct radiative forcing effects only, while the ranges shown below reflect the inclusion of indirect cooling effects, subject to uncertainty in the magnitude of CFC-induced cooling of the lower stratosphere (estimated at ±30%), and uncertainty in the globally averaged relative efficiency of bromine in removing O_3 relative to chlorine (assumed to be between 40 and 200). The first range reflects uncertainty in the bromine/chlorine ozone removal efficiency, and the second range reflects uncertainty in the magnitude of cooling in the lower stratosphere. These net GWP estimates are thought to be accurate only to ±50%, and while uncertainties exist in both the direct (positive) radiative forcing effects and the indirect (negative) effects, the uncertainty is believed to be greater for the indirect effects. Thus, the GWPs for CFC-12 are unlikely to be much higher than the values shown, but they could be somewhat lower (see section 8.6 for a more complete discussion). The estimates for CO, NMHCs, and NO_2 are from the original IPCC report (Shine, et al., 1990), and represent early, preliminary estimates of total direct-plus-indirect GWPs. Recently, the IPCC (1995) has essentially disavowed these earlier estimates of total GWPs, on the grounds that it is not yet possible to estimate indirect effects accurately for these relatively short-lived and poorly mixed gases.

[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). We 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, NMHCs, and NO_x emissions on CH_4 concentrations; and it distinguishes between emissions of NO_x at ground level and emissions at higher levels.

[d] Bruhl (1993) modeled the GWP under two scenarios, one in which emissions increase over time, and another in which the concentration and lifetime of CH_4 is fixed. The former yields higher GWPs, because increasing concentrations of CH_4 (in the face of a relatively constant amount of the scavenger, OH) result in a longer average lifetime for CH_4. Bruhl explains that his estimate of the indirect GWP of CH_4 is lower than the IPCC's estimate of the indirect effect because the IPCC probably overestimated the production of O_3 due to CH_4.

[e] Reilly (1992) bases his estimates on a highly 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.

[f] 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 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 (see Hammitt et al. [1996] for details). 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.

[8] In principle, EDIs are better than GWPs, because EDIs are estimated in terms of the present dollar value of the impacts of global warming, whereas GWPs are estimated just in terms of global warming. However, as of this writing EDIs have been estimated only for CH_4, N_2O, and CFC-12. Moreover, the Hammit et al. (1996) EDIs for CH_4 and CFC-12 are incomplete: their EDI for CH_4 does not account for the indirect effect of CH_4 on tropospheric O_3 and H_2O, and their EDI for CFC-12 does not account for the indirect cooling effect of CFCs. Given this, we have estimated or assumed our own equivalency factors as follows:

CH_4: Hammit et al. (1996) estimate that the middle-case EDI for the direct effects of CH_4 is 11.0. According to the IPCC, the indirect warming effects of CH_4 are about equal to the direct effects. Thus, we double the middle-case EDI.

N_2O: We adopt the middle case of Hammit et al. (1996).

CFC-12: The middle-case EDI, which includes only the direct warming effects of CFC-12, is higher than the GWP of the direct effects. Thus, we assume that a complete EDI that accounted for indirect cooling would be higher than the GWP that includes indirect cooling.

HFC-134a: We assume a value somewhat above the middle of the range of GWPs estimated by the IPCC (1995), on the assumption that an EDI for HFC-134a would be higher than the comparable GWP.

NMHCs: Our assumption is based on the estimates of Martin and Michaelis (1992) and the IPCC (Shine, et al., 1990). Note that we apply the GWP for NMHCs to the carbon content of NMHCs only. Thus, NMHCs with a higher carbon content have a higher GWP. This is reasonable because NMHCs with a higher carbon content generally cause higher O_3 levels, and when oxidized always produce more CO_2.

CO: Our assumption is based on the estimates of Martin and Michaelis (1992) and the IPCC (Shine, et al., 1990).

NO_x: This is the sum of a factor of 2 due to O_3 production (our assumption based on the estimate of Martin and Michaelis (1992)), and a factor of 2 due to N_2O emissions from the deposition of atmospheric nitrogen in NO_x (our estimate; see the text for discussion).

TABLE 3: METHANE EMISSIONS FROM HIGHWAY VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	CH$_4$ emissions (g/mi)	Reference
Gasoline LDVs				
Ford F250 truck (see CNG)*	none	4,000-9,000	0.20	BC Research (1986)
16 1975-'78 passenger cars	various	various	0.19-0.32	Sigsby et al. (1987)
1978 Olds Cutlass[a]	OC, EGR	NS	0.06-0.08	Urban and Garbe (1979)
Chevy S-10 truck (see CNG)*	OC, EGR	4,000-9,000	0.02	BC Research (1986)
4 1978-'80 passenger cars[b]	3WY, EGR; 2 cars w/OC	low	0.09-0.39	Braddock (1981)
4 1978-'79 passenger cars[c]	3WY; 2 cars w/OC	low	0.03-0.11 [0.07]	Smith and Black (1980)
30 1979-'82 passenger cars	various	various	0.14-0.18	Sigsby et al. (1987)
Dodge 600es truck*	3WY	4,000-9,000	0.04	BC Research (1986)
1981 Rabbit (see MeOH car)*	3WY	25,000	0.03	CARB (1985)
1981 Escort (see MeOH car)*	3WY	38,000	0.23	CARB (1985)
1984 Ford Mustang (see MeOH car)*	3WY	100	0.14[d]	Gabele et al. (1985)
1984 Chev. Cavalier (see MeOH car)*	3WY	4500	0.04[d]	Gabele et al. (1985)
9 1984-1987 4-cylinder passenger cars	various	3,000-62,000	0.08-0.15[e]	Stump et al. (1989)
11 1985-1987 4-8 cylinder cars	various	7,000-64,000	0.13-0.20	Stump et al. (1989)
3 1992 Ford 4.9 liter 6-cyl. vans	3 WY	5,000, 15,000, 25,000	0.09, 0.11, 0.13[f] (RFG) 0.11, 0.11, 0.12[f] (RF-A)	Battelle (1995)
3 1992 Dodge 5.2 liter V8 vans	3 WY	5,000, 15,000, 25,000	0.05, 0.07, 0.08[f] (RFG) 0.08, 0.08, 0.08[f] (RF-A)	Battelle (1995)
1992 Chev. pickup, 1993 Ford Crown Victoria, 1992 Dodge wagon (see CNG)*	3 WY (one dual)	5,000 (but 3-WY aged to 50,000)	0.06 FTP, 0.03 REP (RFG) 0.07 FTP, 0.04 REP (RF-A)	Auto/Oil (1996)
3 1992 Chevrolet 4.3 liter V6 vans	3 WY	5,000, 15,000, 25,000	0.06, 0.07, 0.07[f] (RFG) 0.05, 0.07, 0.08[f] (RF-A)	Battelle (1995)
CARB Lifetime Average Factor	*3WY*	*5,000*	*0.056*[g]	*CARB* (1986)
EPA Lifetime Average Factor (1981+)	*3WY*	*over life*	*0.10*	*U.S. EPA* (1985)
EPA Lifetime Average Factor (1992+)	*3WY*	*over life*	*0.05*	*U.S. EPA* (1985)
Mobile5 Emission Factor (1990-2020)	*fleet average*	*fleet average*	*0.04-0.12*	*simulation runs of MOBILE5*[W]

TABLE 3 (CONT'D): METHANE EMISSIONS FROM HIGHWAY VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	CH_4 emissions (g/mi)	Reference
Gasoline HDVs				
Ford truck	NS	11,000	2.13	Dietzmann et al. (1981)
International Harvester truck	NS	15,000	0.28	Dietzmann et al. (1981)
5 trucks, 1973-1980	NS	35,000-105,000	0.4-1.0	Black et al. (1984)
EPA Lifetime Average Factor (1987+)	*None*	*over life*	*0.18*	*U.S. EPA (1985)*
MOBILE5 Emission Factor (1990-2020)		*fleet average*	*0.18-0.28*	*simulation runs of MOBILE5*[h]
Diesel HDVs				
1979 Caterpillar 4-stroke	NS	7,000	0.05	Dietzmann et al. (1981)
1979 Mack 4-stroke	NS	69,000	~0	Dietzmann et al. (1980)
1979 Cummins 4-stroke	NS	26,000	~0	Dietzmann et al. (1980)
1977 DDT 2-stroke	NS	60,000	~0	Dietzmann et al. (1980)
CARB Lifetime Average Factor	*None*	*150,000*	*0.16*[g]	*CARB (November 1986)*
EPA Lifetime Average Factor (1987+)	*None*	*over life*	*0.10*	*U.S. EPA (1985)*
MOBILE5 Emission Factor (1990-2020)		*fleet average*	*0.10-0.12*	*simulation runs of MOBILE5*[h]

TABLE 3 (CONT'D): METHANE EMISSIONS FROM HIGHWAY VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	CH_4 emissions (g/mi)	Reference
Natural gas dual-fuel LDVs				
Ford F250 truck	none	4,000-9,000	1.8-3.0^{i}CNG; 0.20 G	BC Research (1986)
13 1977-1981 passenger cars	NS	NS	0.60-3.13 [1.44] CNG 0.02-0.18 [0.07] G	Aerospace (1982)^{j}
1986 Chevrolet C30 van	OC, AP	120,000	7.31	Gabele et al. (1990b)
1980 Diplomat	OC, EGR, AP	low	1.18 CNG; 0.11 I; 0.09 I	Peninga (1981)^{k}
1979 Impala	OC, EGR, AP	low	0.67 CNG; 0.06 I; 0.06 I	Peninga (1981)^{k}
Chevy S-10 truck	OC, EGR	4,000-9,000	1.7-2.5^{i} CNG; 0.02 G	BC Research (1986)
Dodge 600es truck	3-WY	4,000-9,000	0.6-1.4^{i} CNG; 0.02 G	BC Research (1986)
1985 Ford Ranger	3WY	NS	2.19-4.38	Overby & Regdon (1987)
1984 GM Delta 88	3WY, EGR	high	2.37-2.46^{l} CNG; 0.08 G	Bruetsch (1988)
1986 Mercury Marquis	3WY?	NS	2.63-3.59	NGV Coalition (1989)
1986 Buick Park Avenue	3WY?	25,000	1.80	CARB (1989)
1987 Ford Crown Victoria	3WY, EGR	low	3.03-3.55^{l} CNG; 0.11 G	Bruetsch (1988)
1987 GM Celebrity	3WY, EGR	low	1.41-1.50^{l} CNG; 0.02 G	Bruetsch (1988)
1989 Buick LeSabre	3WY, EGR	2,500	1.51 CNG; 0.013 I	CARB (1991)
1990 Ford Taurus	3WY, EGR	1,500	1.82 CNG; 0.04 I	CARB (1991)
1990 Dodge Dynasty	3WY, EGR	4,100	1.75 CNG; 2.11 CH_4; 0.05 I	CARB (1991)
1991 Ford Taurus (Impco mixer)	3WY?	NS	1.23	CARB (1992)
1991 Ford Taurus (S & S mixer)	3WY?	NS	0.81	CARB (1992)
1990 Chevrolet Astrovan (truck)	3WY, EGR	11,000	2.08	CARB (1992)
Natural gas dedicated LDV:				
1983 Ford 3.8l V-6	none	low	0.9-2.5^{m}	Swain et al. (1983)
1984 Ford Ranger Pickup	OC, EGR	low	1.06	Adams (1985)
1984 Ford Ranger Pickup	OC, EGR	NS	1.17-1.31^{l}	Bruetsch (1988)
1989 Dodge Ram Van	3WY/none^{l}	4,000	1.47/1.49^{m}	Gabele et al. (1990a)
3 1992 Ford 4.9 liter 6-cyl. vans	3 WY	5,000; 15,000; 25,000	1.78; 2.64; 3.34^{f}	Battelle (1995)
3 1992 Dodge 5.2 liter V8 vans	3 WY, CNG optimized	5,000; 15,000; 25,000	0.44; 0.74; 1.0^{f}	Battelle (1995)
3 1992 Chevrolet 5.7 liter V8 vans	3 WY, Engelhard CNG	5,000; 15,000; 25,000	1.69; 2.58; 3.29^{f}	Battelle (1995)
1992 Chev. pickup, 1993 Ford Crown Victoria, 1992 Dodge wagon	3 WY (one dual)	5,000 (but 3-WY aged to 50,000)	0.92 FTP 0.49 REP (CG1) 0.91 FTP, 0.47 REP (CG4)	Auto/Oil (1996)

TABLE 3 (CONT'D): METHANE EMISSIONS FROM HIGHWAY VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	CH_4 emissions (g/mi)	Reference
Natural gas dual-fuel MDVs				
1989 Ford Club Wagon	CC, EGR/air	13,000	2.81	CARB (1991)
1990 Ford F-350 XLT	CC, EGR/air	800	0.27 I	CARB (1991)
Natural gas HDVs				
Diesel dual-fuel pilot	NS	NS	27.2[o]	BC Research (1987)
GMC 454 CID V-8 bus engine	3WY	low	0.6; 2.4[p]	Jones et al. (1988)
GMC 454 CID V-8 bus engine	none	low	6.4[q]	Alson et al. (1989)
Cummins L-10 lean-burn engine	none	NS	4.0[r]	Lawson (1988)
Methanol dual-fuel LDVs				
Ford Crown Victoria	no catalyst	NS	0.039 M100; 0.72 M85/G	Ford (1988a)
Ford Crown Victoria	3WY	3,000 on catalyst	0.037 M100; 0.031 M85/G	Ford (1988a)
Ford Escort	no catalyst	NS	0.031 M100; 0.034 M85/I	Ford (1988a)
Ford Escort	3WY	5,000 on catalyst	0.020 M100; 0.025 M85/I; 0.037 I	Ford (1988a)
7 1987 Ford Crown Victorias	3WY	0- 16,000	0.02-0.06 [0.04] M85/G; 0.05-0.10 [0.07] M50/1 0.08-0.15 [0.10] I	CARB (1988)
1987 FFV Ford Crown Victoria	2-CC, 2-UF, EGR	22,000	0.023 M100; 0.046M85/I; 0.116 I 0.145 E85/I; 0.171 E95/I	CARB (1991)
1987 FFV Ford Crown Victoria	2-CC, 2-UF, EGR	43,700	0.056 M85/I; 0.139 E85/I; 0.110 I	CARB (1991)
1987 FFV Ford Crown Victoria	2-CC, 2-UF, EGR	15,000	0.049 M85/I; 0.084 M25/I; 0.086 I	CARB (1991)
1987 FFV Ford Crown Victoria	2-CC, 2-UF, EGR	61,000	0.079 M85/I; 0.0192 I	CARB (1991)
1988 VFV Chevrolet Corsica	3WY	4,000	0.003 M100; M50/I	CARB (1988)
1988 VFV Chevrolet Corsica	3WY-HC	2,300	0.014 M85/US	CARB (1991)
1988 VFV Chevrolet Corsica	3WY	4,500	0.010 M100; 0.029 M50/I; 0.031 I	Gabele (1990b)[s]
2.5-liter GM VFV	3-WY?	NS	0.0024 M100; 0.036 G	Williams et al. (1990)
1989 FFV Toyota Corolla	3WY, EGR	4,700-11,300	0.049 M85/I; 0.183 I	CARB (1991)
1988 FFV Nissan Stanza	3WY, EGR	15,700	0.023 M85/US; 0.027 US	CARB (1991)
1989 VFV Chevrolet Corsica	3WY, EGR,	21,000	0.050 M85/I; 0.064 M50/I; 0.072 I	CARB (1991)
1990 GTMV Plymouth Voyager	3WY	1,900-3,200	0.028 M85/US; 0.079 US.	CARB (1991)
1990 FFV Plymouth Voyager	3WY	2,000-2,500	0.014 M85/US; 0.046 US	CARB (1991)
3 Ford 4.9 liter 6-cyl. vans	3 WY	5,000; 15,000; 25,000	0.04 M85, 0.05 M85, 0.06 M85[f]	Battelle (1995)

TABLE 3 (CONT'D): METHANE EMISSIONS FROM HIGHWAY VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	CH_4 emissions (g/mi)	Reference
Methanol dedicated LDVs:				
1981 Rabbit	HCC^t	NS	0.01 M100	Blair & Piotrowski (1988)
2 1981 VW Rabbits	3WY	4,500	0.01-0.02 M95	CARB (1985)
2 1981 VW Rabbits	3WY	22,000	0.02-0.03 M90	CARB (1985)
2 1981 VW Rabbits	3WY	56,000- 66,000	0.03-0.13 M85/G	CARB (1988)
2 1981 Ford Escort Wagons	3WY	5,000- 50,000	0.07 M90-95	CARB (1985)
1981 Ford Escort Wagon	3WY	85,000-115,000	0.09 M90	CARB (1985)
1982 Chevrolet Citation	3WY	30,000-40,000	0.03 M85-90	CARB (1985)
8 1983 Ford Escort Wagons	3WY	0- 20,000	0.01-0.14 [0.06]^u M90	CARB (1985)
1983 Pontiac Phoenix	3WY	2,400	0.02 M88	CARB (1983)
1983 Ford Escort	3WY	1,500	0.06 M90	Gabele et al. (1985)
unspecified developmental vehicle	NS	NS	0.076 M100	Williams et al. (1990)
1985 Toyota Camry	3WY	0-26,000	0.02 M85/G	CARB (1988)
1985 Toyota Camry	none	0-26,000	0.02 M85/G	CARB (1988)
1986 Toyota Carina	3WY	0- 9,000	0.02 M85/G	CARB (1988)
2 1986 Ford Crown Victorias	3WY	2,500-15,000	0.037 M85/I	CARB (1991)
2 1986 Ford Crown Victorias	3WY	29,000-49,000	0.057 M85/I	CARB (1991)
1989 Toyota Corolla	2 CC, 2-UF, EGR	4,000-15,000	0.029 M85/I	CARB (1991)
1990 DI turbo CI VW Jetta	pt. cat., EGR	2,300	0.07 M100	Bruetsch & Hellman (1991)
Methanol HDVs				
MAN spark-ignited 6-cyl. engine 1	OC	new?	0.002^v M100	Ullman and Hare (1986)
MAN spark-ignited 6-cyl. engine 2	OC	28,300	0.04; 0.12^w M100	Ullman and Hare (1986)
DDAD 6V-92TA spark-assisted	2-stroke	8900	1.17; 0.72^w M100	Ullman and Hare (1986)

TABLE 3 (CONT'D): METHANE EMISSIONS FROM HIGHWAY VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	CH_4 emissions (g/mi)	Reference
Other kinds of vehicles				
1979 Hydrogen pick-up truck	NS	23,000	0.00	CARB (1989)
1987 FFV Ford Crown Victoria	2-CC, 2-UF, EGR	22,000	0.023 M100; 0.046M85/I; 0.116 I 0.145 E85/I; 0.171 E95/I	CARB (1991)
1987 FFV Ford Crown Victoria	2-CC, 2-UF, EGR	43,700	0.056 M85/I; 0.139 E85/I; 0.110 I	CARB (1991)
1988 Dual-fuel LPG Chev. 1500 truck	NS	14,000	0.046	CARB (1989)
1989 Dual-fuel LPG Oldsmobile 88	3WY, EGR	22,700	0.064 LPG; 0.047 US	CARB (1991)
1989 Dual-fuel LPG Pontiac 6000 LE	3WY, EGR	31,300	0.042 LPG; 0.037 I	CARB (1991)
1991 LPG Chevrolet Lumina	3WY	4,000	0.022 (FTP) 0.149 (NYCC)	Gabele (1992)
3 Ford 4.9 liter 6-cyl. LPG vans	3 WY	5,000; 15,000; 25,000	0.12; 0.14; 0.16^f	Battelle (1995)
3 Chevrolet 5.7 liter V8 LPG vans	3 WY	5,000; 15,000; 25,000	0.09; 0.11; 0.13^f	Battelle (1995)

NS = not specified; cat. = catalytic converter; 3WY = three-way catalytic converter (one that oxidizes CO and NMHCs, and reduces NO_x); OC = oxidation catalytic converter; HC = heated catalytic converter; CC = close-coupled catalytic converter; UF = under-floor catalytic converter; EGR = exhaust-gas recirculation; LDV = light-duty vehicle; HDV = heavy-duty vehicle; GTMV = gasoline-tolerant methanol vehicle, designed to run on methanol, but "tolerant" of gasoline; FFV = flexible-fuel vehicle; VFV = variable-fuel vehicle; NGV = natural gas vehicle; I = indolene; US = U.S. average gasoline; DI = Direct injection; CI = compression ignition; VW = Volkswagon; CARB = California Air Resources Board; NYCC = New York City Cycle; HFET = Highway Fuel-Economy Test; REP = REP05, the EPA's high-speed, high-load driving cycle used to measure "off-cycle" emissions; RFG = reformulated gasoline; RF-A = industry average unleaded gasoline; CG1 = industry-average natural gas (94% methane); CG4 = natural gas with relatively low methane content (86%)

All emissions results for LDVs were obtained over the Federal Test Procedure (FTP), unless noted otherwise. All emissions results for HDVs were obtained from engine tests over the Heavy-Duty Transient Cycle (HDTC), unless otherwise noted. (The chassis version of the HDTC [Dietzmann et al., 1980] tests the whole chassis, not just the engine.) The EPA results are net of the background ambient concentration.

Emissions estimates in brackets [ ] are averages. Abbreviations following emissions (e.g., M85/I) indicate the fuel used in multi-fuel vehicles (85% methanol and the rest, 15%, indolene). Multiple gram/mile results separated by a semicolon are results for different fuels tested on the particular vehicle in the dual-fuel or multi-fuel configuration.

[*] See the results for the same kind of vehicle tested on an alternative fuel, in this Table. The 1981 Rabbit and the 1981 Escort (tested by CARB, 1985) are production-line gasoline vehicles, and should be compared to the 1981 dedicated methanol Rabbits and Escorts tested by CARB (1985). The 1984 Mustang and the 1984 Cavalier (tested by Gabele et al., 1985) also are production-line gasoline vehicles, and should be compared to the dedicated methanol Escort tested by Gabele et al. (1985; see footnote "f" to this Table). The trucks tested by BC Research (1986) are the same trucks tested as dual-fuel NGVs, except that the results shown under "gasoline LDVs" were obtained prior to the installation of the CNG dual-fuel conversion kit. The Auto/Oil (1996) program tested three dedicated OEM CNG vehicles (1992/93 model years) and their gasoline counterparts.

^The same vehicle as the one immediately above (i.e., not a different vehicle of the same model). Vehicles of the same description but not marked with a "^" are same models but different vehicles.

[a] Methane emissions were slightly higher when the engine malfunctioned. Emissions were much higher with rich idle: 0.52 g/mi.

[b] Emissions were around 0.10 g/mi for 3 of 4 vehicles, with summer fuel and at 78°F ambient temperature, but were over 0.20 g/mi with winter fuel and at 55°F ambient temperature.

[c] Methane missions varied moderately with type of gasoline, and generally increased slightly from 0 to 15,000 miles. Methane emissions in the NYCC were higher, and in the HFET lower, than in FTP.

[d] Methane emissions were measured for the Mustang only. We assume that CH_4 was the same % of HC exhaust from Cavalier as from Mustang. The Cavalier is more similar in weight and power to the Escort than is the Mustang.

[e] The range represents emissions at different ambient temperatures. Methane emissions were lowest at 21°C, which is typical of FTP test conditions, and highest at the lowest temperature (-6.7°C).

[f] Tests were performed on vehicles operated over the FTP, and using an on-line FTIR analyzer. The three numbers correspond to the three vehicle mileage figures listed, and each number is a mean value for tests of three different vans by each manufacturer. Note that the Dodge CNG vans are among the first to use a catalyst system designed and optimized for CNG vehicles.

[g] CARB estimates that 7.6% of total organic gases (TOG) from gasoline LDVs are nonreactive; we assumed that all these nonreactive gases are CH_4, and that no CH4 is counted as reactive. We then multiplied the lifetime average TOG emission factor for LDVs by 0.076. For HD diesels, CARB assumes that 4.4% of TOG are nonreactive.

[h] Methane emissions are the difference between exhaust THC and exhaust NMHC.

[i] Emissions varied with the conversion kit.

[j] The researchers actually *measured* CH_4 emissions from one car only, a 1978 Ford Fairmont. They *assumed* that CH_4 was 80% of total HCs from the 1977-model-year NGVs, 87% from later year NGVs, and 12% from gasoline vehicles. The authors did not specify the driving test cycle over which emissions were measured. The emission results on gasoline are prior to conversion to dual-fuel operation.

[k] The first emission result on indolene (I) is for the stock, unmodified gasoline configuration; the second is for the dual-fuel configuration, optimized for CNG, but running on gasoline (indolene).

[l] The authors reported total HCs and 4 different ways of measuring NMHCs. The range shown here is their HC minus their high highest (of the four) calculated NMHC to their HC minus their lowest NMHC.

[m] Emissions varied with the spark advance and the air-to-fuel ratio. The test cycle was an approximation of the EPA-CVS (the FTP).

[n] The first emission result is for the FTP test, with the vehicle's 3-way catalytic converter in place. The second is the for the FTP test but with no catalyst. Emissions did not vary appreciably with ambient temperature (20° F to 105° F). Methane emissions were 3 g/mi in the NYCC, and 0.90 g/mi in the HFET (with the 3-way catalyst in place).

[o] The result was reported in the reference as 13 g/bhp-hr and 85-90% CH_4. We assumed 2.31 bhp-hr/mi (U.S. EPA, 1985). The engine was tested over the SAE 13-mode test.

[p] We assume that CH_4 was 85% of the total reported HCs, and 2.31 bhp-hr/mi (U.S. EPA, 1985). The first test result shown was obtained by the manufacturer; the second was obtained by the EPA (see also Parker, 1988). In the tests reported by Jones et al. (1988), the HDTC was modified to reflect transit bus applications. See also Alson et al. (1989) results for other EPA tests on the GMC engine.

[q] We assume 2.31 bhp-hr/mi (U.S. EPA, 1985). The engine was tested over the HDTC.

[r] We assumed that CH_4 was 85% of the total reported HCs, and 2.31 bhp-hr/mi (U.S. EPA, 1985). The emissions results were obtained over the SAE 13-mode test.

[s] Emissions were about twice as high at 40^{o} F. Emissions at 90^{o} F were similar to FTP (75^{o}) emissions.

[t] The vehicle was equipped with a resistively heated monolithic catalyst designed to reduce cold-start HC and HCHO emissions.

[u] New vehicles emitted about 0.03 g/mi; older vehicles emitted around 0.06 g/mi, and the average was around 0.06 g/mi. Three of the vehicles had electronic fuel injection.

[v] The authors reported 70 mg CH_4 in the cold-start transient test, 0 in the hot-start test, and about 9.3 kw-hr work in both tests. We used their formula to convert these to mg/hp-hr, and then assumed 2.31 bhp-hr/mi (EPA, 1985) The engine was tested over the HDTC.

[w] The first value was emitted over bus transient cycle; the second over the central business district transient cycle.

TABLE 4: FTP MG/MI METHANE EMISSIONS FROM FFVS, AS A FUNCTION OF METHANOL CONTENT OF FUEL

	Percent methanol in fuel					
	100	85	50	25	0	Reference
1988 VFV Chev. Corsica (40° F; 4500 mi)	NA	41	48	68	82	Gabele (1990b)
1988 VFV Chev. Corsica (75° F; 4500 mi)^	9	20	26	25	34	Gabele (1990b)
1988 VFV Chev. Corsica (90° F; 4500 mi)^	8	14	26	36	42	Gabele (1990b)
VFV General Motors	2	14	20	16[a]	36	Williams et al. (1990)
1988 VFV Chevrolet Corsica (21,000 mi)		50	64		72	CARB (1991)
FFV Ford Escort 1 (4,000 mi on catalyst)	30	31	28		48	Ford (1988a)
FFV Ford Escort 2 (50,000 mi on catalyst)	20	25	23		37	Ford (1988a)
1988 FFV Nissan Stanza (16,000 mi)		19	25	23	27	CARB (1991)
1987 FFV Ford Crown Victoria 1 (15,000 mi)		49	56[b]	84	86	CARB (1991)
1987 FFV Ford Crown Victoria 2 (20,000 mi)	23	49	74	92[c]	71[d]	CARB (1991)

CARB = California Air Resources Board; FFV = flexible fuel vehicle; VFV = variable fuel vehicle. The vehicle mileage readings shown in parentheses have been rounded to the nearest 1000 miles.

^The same vehicle as the one immediately above (i.e., a different vehicle of the same model type). Vehicles of the same description but not marked with "^" are the same model but different vehicles.

[a] 15% methanol.

[b] Emissions on M50 were measured at 8400 and 8800 miles. Emissions on the other fuels were measured at around 15,000 miles. The M50 fuel used unleaded gasoline; the other mixtures used indolene.

[c] I estimated this by interpolating between 70 mg/mi at 15,100 miles and 147 at 32,500 miles.

[d] At 22,000 miles the emission rate was 138 mg/mi.

TABLE 5: RECENT SURVEYS, STUDIES, OR ESTIMATES OF THE RATE OF LEAKAGE OF NG TRANSMISSION AND DISTRIBUTION SYSTEMS

Organization (Reference)	NG lost, % of delivered NG			Comments
	Prod.	*Trans.*	*Dist.*	
Canadian Gas Association (1990)	0.25	0.018-0.082	0.03	Estimate for Canadian companies
Alphatania Group (1989)[a]	0 to 0.20	0 to 0.13	0.03 to 0.30	Survey of 28 companies worldwide
Arthur D. Little (1989)	0.53			Estimate for worldwide production
European gas companies[b]		0.005	<1.00	Higher loss in old dist. lines
Germany (Okken, 1990)	0.50			
PG&E (Cottengim, 1989)[c]		0.14		Mostly distribution losses
SoCal Gas (Mehskati, 1993)[d]		0.12		Mostly distribution losses
American Gas Association (1989a)[e]		0.0-6	0.28	Survey of U.S. NG companies
British Gas (Wallis, 1991)			<1.00	Mainly from old cast-iron pipes
Swedegas (in Svensson, et al., 1991)	<1.00			
Mitchell et al. (1990)		0-0.5		Post-1969 NG pipes in Britain
Mitchell et al. (1990)		1.9-10.8		All NG pipes in Britain
U.S. EPA (1993)[f]	0.32	0.28	0.09	Detailed estimate for U.S. in 1990
U.S. EPA (1993)[f]	0.31	0.22	0.07	Year-2000 projection, high-gas use
Radian Int'l LLC for GRI/EPA (1996)[g]	0.52	0.54	0.39	Detailed estimate for U.S. in 1992
Gas Research Inst. (in Lamb, et al., 1995)	1.0 to 2.0			For U.S. facilities

Notes: see next page.

[a] The Alphatania Group (1989), a consulting firm to the natural gas industry, asked for information on CH_4 leaks from "41 selected companies and organizations closely concerned with natural gas operations worldwide" (p. 3). They received responses from 28. The figure for distribution losses is for new systems; they estimated that up to 1% of throughput leaks from old distribution systems. This study is cited by the Canadian Gas Association (1990) and Wilson (1990).

[b] Communication from personnel at gas companies to Okken and Kram (1989).

[c] Cottengim et al. (1989) call this the "most comprehensive analysis of unaccounted-for gas ever undertaken". The study investigated leakage (both intentional and unintentional), theft, metering inaccuracies, and accounting problems, for the PG&E transmission and distribution system in 1987. Intentional losses, such as from purges and valve operations, were determined from historical records and field surveys. Unintentional leaks from distribution systems were estimated for different categories of distribution pipe by field tests of different kinds of leaks. The transmission system was assumed to leak at "the highest conceivable rate". Losses from unintentional ruptures were estimated by multiplying an average (apparently historical) loss rate per rupture by the number of ruptures in 1987. Unintentional losses from distribution systems were 0.06% of the total; unintentional losses from transmission systems were 0.005%; losses from ruptures of any kind of system were 0.01%; losses from instrument usage, facility blow and purge gas, gas sampling, drip operations, relief valve operations, and miscellaneous operations were 0.065%. Losses were only 9% of all unaccounted for gas; the biggest source of unaccounted for gas was inaccuracies in orifice meters.

[d] SoCal Gas estimates the following "unaccounted for" (that is, not accounted for in normal gas accounting) gas losses to the atmosphere in 1991 (all units in MCF [one thousand cubic feet]): 15,580 from major and minor leakage from transmission systems; 58,039 from leakage from compressor packing seals on reciprocating and rotary compressors; 59,912 from pneumatic instruments, gas sampling and analysis, facility blow and purge gas, drip purging operations, and turbine engine starts; 622,160 from underground leaks from distribution systems; and 182,502 from unreported damage to distribution systems, for a total of 938,193 MCF (Meshkati, 1993). In addition, there were accounted-for losses from the transmission system and from major damage to the distribution system, and unaccounted for losses from natural-gas storage fields. We estimate that these additional losses were about 30% of the 938,193 estimated "unaccounted for" gas losses. The grand-total gas lost to the atmosphere was thus about 1.2 BCF, or 0.12% of the 1.048 TCF of sales by SoCaL Gas in 1991.

[e] The transmission companies claimed to be confident with their estimates; the distribution companies were less confident. (Leaks in high-pressure transmission lines are easier to identify than leaks in low-pressure distribution lines.) Shortly before this study, the AGA was estimating total leakage to be in the range of 0.2 to 0.3% (AGA, 1989b).

[f] The column "Production" in this table includes leakage from field production, gas processing plants, and storage systems, among which leakage from field production is by far the largest emission source. Engine exhaust is not included here.

[g] The complete results of this study, in BCF natural gas emitted in 1992 in the U.S., are as follows (from GRI/EPA, 1996):

Natural Gas Emission Type	Production Segment (BCF)	Gas Processing Segment (BCF)	Transmission and Storage Segment (BCF)	Distribution Segment (BCF)	Total Natural Gas Industry (BCF)
Fugitive	24.0	24.4	72.1	74.7	195.3
Vented	47.0	5.5	33.0	2.2	87.6
Incomplete combustion	6.6	6.4	11.4	N/A	24.4
Total	77.5	36.3	116.5	77.0	307

In the GRI/EPA study, fugitive emissions are unintentional releases of methane from equipment leaks at sealed surfaces, and from underground pipes. Vented emissions are intentional releases of methane from pneumatic devices, dehydrators, chemical injection pumps (a minor source), and blowing and purging. Combustion emissions result from incomplete combustion of methane in burners, flares, and engines (mainly engines). In our estimation of the percentage leakage rate (discussed next), we exclude emissions from incomplete combustion, because we account for these emissions separately with an emission factor for CH_4 from compressor engines. (The total emissions implied by our CH_4 emission factors for natural gas engines, in Table 12, are consistent with GRI/EPA's estimates of emissions from incomplete combustion.)

The authors state that their final 1992 U.S. loss estimates of 1.4% of gross and 1.6% of net natural gas produced are believed accurate to within ±0.5%.

In 1992, the U. S. consumed 19,540 BCF of natural gas (EIA, 1996a). Thus, we estimate that leakage from the production and processing segments, which we combine into a single production category, was 101/19540 = 0.52% of combustion; leakage from transmission and storage (which we call transmission), 105/19540 = 0.54%; and leakage from distribution, 77/19540 = 0.39%.

TABLE 6: VENTING AND FLARING EMISSIONS ASSOCIATED WITH CRUDE OIL USED BY THE U.S., 1992

	10^9 SCF reported vented or flared[a]	Under-reporting factor[b]	10^3 bbl/day crude oil[a]	fraction assumed flared[c]	SCF vented/ bbl[d]
USA -- domestic production including Federal offshore wells	182[e]	1.05	7,171	0.870	73
Crude oil imports to U. S. from:					
Canada	86	1.05	1,065	0.870	232
Mexico	49	1.10	2,669	0.850	55
Northern Europe (United Kingdom, Norway)	17	1.05	4,054	0.870	12
OPEC					
Venezuela	123	1.10	2,371	0.830	156
North Africa (Algeria)	141	1.20	1,214	0.830	382
West Africa (Nigeria, Gabon)	946	1.25	2,241	0.800	1,446
Indonesia	217	1.10	1,504	0.850	435
Persian Gulf (Saudi Arabia, Kuwait)	431	1.10	9,380	0.830	138
Other Middle East (Oman, Yemen)	22	1.10	922	0.830	72
Other Latin America (Colombia, Trinidad and Tobago)	85	1.10	570	0.830	449
Other Africa (Angola)	45	1.25	526	0.800	293
Other Asia (China)	0	1.25	2,845	0.830	0
Total world	*3,828*	*n.e*	*60,213*	*n.e.*	*n.e.*

n.e. = not estimated.

[a] From EIA's *International Energy Annual 1993* (1995a). The venting and flaring figures are the amounts reported to or estimated by the EIA.

[b] It is likely that the amount of venting and flaring is underreported. For example, in the U.S., six states do not report venting and flaring emissions to the EIA (1995c). We estimate that venting and flaring emissions in these states are about 2% of reported venting and flaring in all other states. On the assumption that the states that do report venting and flaring might under-report slightly, we assume that the true venting and flaring emissions in the U.S. are 5% higher than the amount reported to the EIA.

We assume that underreporting is higher in South America and the Middle East, and highest in Africa and Asia.

[c] Assumes that 48 SCF of NG is vented or flared per bbl of crude oil from Federal offshore wells, and that crude oil production from Federal offshore wells is 11% of total U. S. crude oil production. See the discussion in the text for a bit more explanation.

[d] Equal to the amount of gas vented (not flared) divided by total crude oil produced.

[e] Includes gas vented and flared from U.S. Federal offshore oil platforms. See text for discussion.

TABLE 7: ESTIMATES OF METHANE EMISSIONS FROM U.S. COAL MINING

	10^9 CF[a]	10^6 ton	CF/ton	Year
U.S. EPA (1993) low	172.3	950[b]	181.3	1988
U.S. EPA (1993) high	271.4	950[b]	285.7	1988
U.S. EPA (1993) low	193.1	1,125[b]	171.7	2000
U.S. EPA (1993) high	339.3	1,375[b]	246.8	2000
Kirchgessner et al. (1993b)	193.2[c]	356[c]	542.7[c]	1989
CIAB (in Thakur et al. (1996))	190.9	931[d]	205.1	1990

n.e. = not estimated; CF = standard cubic foot of gas; CIAB = Coal Industry Advisory Board; ton = English short ton (2000 lbs). The estimates here are of emissions, which do not necessarily equal releases. (In principle, emissions are equal to releases less quantities captured and used as a fuel.)

[a] The U.S. EPA (1993), the EIA (1993), and Kirchgessner et al. (1993b) report emissions in 10^{12} grams (teragrams). To obtain cubic feet of CH_4 we used the EPA's (1993) conversion factor of 52.2 billion cubic feet per teragram (19.2 g/SCF). Thakur et al. (1996) report emissions in millions of tons, which we then convert to cubic feet using 1,016,000 grams per ton and 19.2 grams per cubic foot.

[b] These amounts were taken from a graph in the EPA (1993) report.

[c] These figures are for underground mine production only. Kirchgessner et al. (1993b), lacking sufficient data for a sophisticated analysis of emissions from surface mines, make the assumption that average emissions of CH_4 from surface mines are 1 cubic meter per ton of coal, or about 35 cubic feet per ton. Multiplying this emission factor by surface mine production and adding to the total shown for underground mines produces a result similar to the EPA (1993) 'High' estimate.

[d] Thakur et al. (1996) report a 1990 U.S. coal production breakdown of 384 Mt from underground mines and 547 Mt from surface mines, for a total of 931 Mt.

TABLE 8. EIA ESTIMATES OF METHANE RELEASES AND ATMOSPHERIC EMISSIONS FROM COAL MINING 1987-1993

	1987	1988	1989	1990	1991	1992	1993
Methane released from underground mines (10^{12} g)	3.86	4.07	4.13	4.42	4.19	4.22	3.53
Methane released from surface mines (10^{12} g)	0.42	0.43	0.45	0.47	0.46	0.46	0.46
Methane recovered and sold (10^{12} g)	-0.25	-0.25	-0.25	-0.25	-0.25	-0.37	-0.48
Methane recovered and sold (10^{9} SCF)	-12.95	-12.95	-12.95	-12.95	-12.95	-19.17	-24.86
Methane recovered as fraction of gas released	-0.058	-0.056	-0.055	-0.051	-0.054	-0.079	-0.120
Production from underground mines (10^{6} short tons)	372.9	382.2	393.8	424.5	407.2	407.2	351.1
Production from surface mines (10^{6} short tons)	545.9	568.1	586.9	604.5	588.8	590.3	594.4
Release rate from underground mines (SCF/ton)	536.2	551.6	543.3	539.4	533.0	536.8	520.8
Release rate from surface mines (SCF/ton)	39.9	39.2	39.7	40.3	40.5	40.4	40.1
Average release rate (SCF/ton)	227.2	231.7	228.7	233.6	228.8	223.8	192.3

From the EIA (1995b). SCF = standard cubic feet.

TABLE 9: EMISSIONS FROM FUEL COMBUSTION AT UTILITY POWER PLANTS, WITH CONTROLS (G/10^6 BTU OF FUEL INPUT TO THE PLANT)

	Coal[a]	Fuel oil[b]	NG (boiler)	NG (turbine)	Methanol	Hydrogen	Biomass
CH_4	0.86	0.85	0.13	10.89	0.70	0.00	0.65
N_2O[c]	4.00	2.00	2.00	2.00	2.00	2.00	4.00
Aldehydes (as HCHO)	0.051	0.13	n.e.	n.e.	n.e.	0.00	n.e.
NMOC	1.29	2.30	0.64	1.92	4.00	0.00	1.52
CO	10.74	15.15	18.14	49.90	15.00	0.00	2.00
NO_x (as NO_2)	233.03	152.27	124.74	99.79	100.00	115.00	43.00
SO_x[d]	403.99	244.05	0.28	0.28	0.30	0.00	0.00
PM[e]	21.91	9.33	1.36	19.01	2.00	0.00	0.00
PM_{10}	4.94	6.61	1.36	19.01	n.e.	0.00	n.e.
$PM_{2.5}$	1.29	4.82	1.36	19.01	n.e.	0.00	n.e.

N. G. = natural gas; n.e. = not estimated; n.a. = not applicable. The numbers shown are equal to uncontrolled emission factors multiplied by the following control factors:

	Coal	Fuel oil	NG (boiler)	NG (turbine)	Methanol	Hydrogen	Biomass
CH_4	1.00	1.00	1.00	1.00	1.00	n.a.	1.00
N_2O	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Aldehydes	1.00	1.00	1.00	1.00	1.00	1.00	1.00
NMOC	1.00	1.00	1.00	1.00	1.00	n.a.	1.00
CO	1.00	1.00	1.00	1.00	1.00	n.a.	1.00
NO_x	0.50	0.75	0.50	0.50	0.50	0.50	1.00
SO_x	0.50	0.50	1.00	1.00	1.00	n.a.	1.00
PM	0.010	0.25	1.00	1.00	1.00	n.a.	1.00
PM_{10}	0.010	0.25	1.00	1.00	1.00	n.a.	1.00
$PM_{2.5}$	0.010	0.25	1.00	1.00	1.00	n.a.	1.00

The uncontrolled emission factors for coal, NG, and fuel-oil plants are from the EPA's emission-factor handbook, AP-42 (U. S. EPA, 1995), except as noted. The uncontrolled emission factors for methanol, hydrogen, and biomass plants are our assumptions; see DeLuchi (1991) for further discussion. Note that the background ambient concentration of the pollutants (in the input air) has *not* been netted out from the results shown here (McSorley, 1993). The control factors show the fraction of emissions remaining after control (hence, a factor of 0.0 means complete elimination of emissions), and are our assumptions; see DeLuchi (1991) for further discussion.

[a]Emission factors for dry-bottom boilers firing pulverized bituminous coal.

[b]Emission factors for utility boilers using "normal" firing, with #6 oil.

[c]Our estimates, based on the data of Table 11.

[d]Uncontrolled SO_x emissions are a function of the sulfur content of the fuel. The emission factors for fuel oil include SO_3 as well as SO_2.

[e]We have multiplied the original AP-42 emission factors by 1.02 to account for condensable PM, which is excluded from the original factor.

TABLE 10: N_2O EMISSIONS FROM MOTOR VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	N_2O Emissions (mg/mi)	Ratio, NO_x/N_2O^a	Reference
Gasoline LDV	*no cat. converter*				
NS	none	NS	0^b	NS	Pierotti and Rasmussen (1976)
NS	none	NS	20	NS	Robertson (1991)
1974 Chevy Impala	none	62,700	16 (15)^c	179	Warner-Selph & Harvey (1990)
1977 AMC Pacer	none; AP	NS	5^d	400	Urban and Garbe (1979)
1977 Volvo	lean operation	200	0	--	Bradow and Stump (Bradow and Stump, 1987)
NS	unspecified cat.	NS	up to 21	NS	Pierotti and Rasmussen (1976)
NS	unspecified cat.	NS	200	NS	Robertson (1991)
	ox. cat.				
1977 Mercury Marquis	OC, AP	81,700	10 (13)^c	167	Warner-Selph & Harvey (1990)
1978 Olds Cutlass	OC, EGR	NS	18^d	45	Urban and Garbe (1979)
1978 Olds Cutlass	OC	NS	23^e	NS	EPA unpublished data
1977 Olds Cutlass	OC	NS	47^e	NS	EPAunpublished data
1978 Malibu	OC, AP	NS	8^d	86	Urban and Garbe (1979)
1978 Malibu	OC, no AP	NS	66^d	18	Urban and Garbe (1979)
1978 Granada	OC, AP	NS	34^d	58	Urban and Garbe (1979)
1978 Mustang	OC, no AP	NS	43	38	Urban and Garbe (1979)
NS 660 cc	OC, EGR	300	1.4, 0.3, 1.0, 0.3, 0.3^f	NS	Sasaki and Kameoka (1992)
	3-wy plus ox. cat.				
8 1978 and 2 1979 cars	7 w/OC, 3 w/3WY	38,000-68,000	74/57^g	28/29^g	Smith and Carey (1982)
1978 Ford Pinto	3WY, OC, EGR	low	12-128^h	9-29^h	Urban and Garbe (1980)
1978 Ford Pinto	3WY, OC, EGR	0-15,000	34-35^i	19-26^i	Smith and Black (1980)
1979 Mercury Marquis	3WY, OC, EGR	low	17-141^h	6-238^h	Urban and Garbe (1980)
1979 Mercury Marquis	3WY, OC, EGR	0-15,000	36-60^i	27-35^i	Smith and Black (1980)
1980 Chevrolet Caprice	3WY, OC, EGR	low	63, 69^j	16, 17^j	Braddock (1981)
1978 Ford Pinto	3WY, OC, EGR	low	84, 84^j	12, 11^j	Braddock (1981)

TABLE 10 (CONT'D): N_2O EMISSIONS FROM MOTOR VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	N_2O Emissions (mg/mi)	Ratio, NO_x/N_2O[a]	Reference
Gasoline LDV (cont'd)	*3-wy cat.*				
1977 Volvo	3WY	200	64[k]	14	Bradow and Stump (1987)
1978 Saab	3WY	low	5-37	10-31	Urban and Garbe (1980)
1978 Saab	3WY	0-15,000	16-35[i]	10-34	Smith and Black (1980)
1978 Pontiac Sunbird	3WY, EGR	low	6-44	21-44	Urban and Garbe (1980)
1978 Pontiac Sunbird	3WY, EGR	0-15,000	32-48[i]	20-40	Smith and Black (1980)
1980 Buick Century	3WY, EGR	low	101, 137[j]	8, 10[j]	Braddock (1981)
1980 Lincoln Continental	3WY, EGR	low	72, 37[j]	24, 38[j]	Braddock (1981)
1983 Buick Regal	3WY, AP	83,000	231 (239)[c]	8	Warner-Selph & Harvey (1990)
2.2 liter Renault	3WY, EFI	NS	50-55[l]	NS	Prigent and De Soete (1989)
1989-1990 U. S. cars	3WY	NS	13-78 [42][m]	5-29[n]	Dasch (1992)
1990 Chevy Lumina van	3WY	NS	89	NS	Dasch (1992)
1990 Chevy Lumina	3WY, TBI	5,300	42 (45)[c]	6	Warner-Selph & Harvey (1990)
1990 Ford Probe	3WY, MPFI	11,500	74 (86)[c]	15	Warner-Selph & Harvey (1990)
Ford Taurus	3WY	low	46	24	Ford (1988b)
Ford Topaz	3WY	low	43	27	Ford (1988b)
1986 Ford Tempo	3 WY, EGR, EVP	76,000	211, 136[o]	NS	Ballantyne et al. (1994)
1986 Oldsmobile Calais	3 WY, EGR, EVP	81,000	120, n/a[o]	NS	Ballantyne et al. (1994)
1987 Chrysler Lebaron	3 WY, EGR, EVP	118,000	192, 66[o]	NS	Ballantyne et al. (1994)
1987 Toyota Pickup	3 WY, EGR, EVP	56,000	93, 30[o]	NS	Ballantyne et al. (1994)
1988 Ford Taurus Wag.	3 WY, EGR, EVP	93,000	114, 53[o]	NS	Ballantyne et al. (1994)
1989 Honda Accord	3 WY, EGR, EVP	87,000	72, 44[o]	NS	Ballantyne et al. (1994)
1989 Honda Civic	3 WY, EVP	107,000	88, 46[o]	NS	Ballantyne et al. (1994)
1989 Volkswagen GTI	3 WY, EVP	94,000	85, 66[o]	NS	Ballantyne et al. (1994)
1990 Mazda 323	3 WY, EVP	34,000	126, 46[o]	NS	Ballantyne et al. (1994)
1992 Honda Civic LX	3 WY, EVP	4,000	75, 25[o]	NS	Ballantyne et al. (1994)
1992 Suzuki Swift	3 WY, EVP	4,000	21, 11[o]	NS	Ballantyne et al. (1994)
1992 Pontiac Sunbird	3 WY, EGR, EVP	4,000	126, 65[o]	NS	Ballantyne et al. (1994)
1992 Chevrolet Pickup	3 WY, EGR, EVP	4,000	264, 47[o]	NS	Ballantyne et al. (1994)

TABLE 10 (CONT'D): N_2O EMISSIONS FROM MOTOR VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	N_2O Emissions (mg/mi)	Ratio, NO_x/N_2O^a	Reference
Gasoline LDV (cont'd)	*3-wy cat.*				
1992 Chevrolet Astro	3 WY, EGR, EVP	4,000	209, 70^o	NS	Ballantyne et al. (1994)
1989 Volvo 740	3 WY	NS	72, 13, 271^p	NS	Jobson et al. (1994)
NS 1800 cc	3 WY, EGR	600	26.7, 22.1, 47.0, 13.8, 2.7^q	NS	Sasaki and Kameoka (1992)
NS 1500 cc	3 WY, EGR	20,000	34.3, 7.4, 54.7, 20.6, 12.2^q	NS	Sasaki and Kameoka (1992)
NS 2000 cc	3 WY, EGR	32,000	197, 116.6, 77.9, 17.4, 25.1^q	NS	Sasaki and Kameoka (1992)
3 1992 Ford 4.9 liter vans	3 WY	5,000, 15,000, 25,000	14, 30, 44^r (RFG)	NS	Battelle (1995)
3 1992 Dodge 5.2 liter vans	3 WY	5,000, 15,000, 25,000	22, 54, 86^r (RFG)	NS	Battelle (1995)
3 1992 Chevy 4.3 liter vans	3 WY	5,000, 15,000, 25,000	254, 301, 326^r (RFG)	NS	Battelle (1995)
Effect of Catalyst Age on Gasoline LDVs					
NS 2200 cc	3 WY	new	37.7, 26.6, 15.7^s	3.5, 0.94, 2.87^s	De Soete (De Soete, 1993)
NS 2200 cc	3 WY	15,000	77.5, 104.5, 69.2^s	3.25, 1.28, 1.54^s	De Soete (1993)
NS 2300 cc	3 WY	new	392.8, 265.6^t	NS	Lindskog (1989) in De Soete (1993)
NS 2300 cc	3 WY	9,300	502.3, 397.7^t	NS	Lindskog (1989) in De Soete (1993)
NS 2100 cc	3 WY	new	34.9, 22.2^u	NS	Prigent and De Soete (1992)
NS 2100 cc	3 WY	150 hours	52.8, 32.2^u	NS	Prigent and De Soete (1992)
3 1992 Ford 4.9 liter vans	3 WY	5,000, 15,000, 25,000	14, 30, 44^r	NS	Battelle (1995)
3 1992 Dodge 5.2 liter vans	3 WY	5,000, 15,000, 25,000	22, 54, 86^r	NS	Battelle (1995)
3 1992 Chevy 5.7 liter vans	3 WY	5,000, 15,000, 25,000	254, 301, 326^r	NS	Battelle (1995)
Diesel LDV					
1.9 liter Citroen	NS	NS	50-58^l	NS	Prigent and De Soete (1989)
NS	NS	NS	50	NS	Robertson (1991)
NS 1800 cc	EGR	17,500	10.5, 10.8, 13.7, 10.8, 7.9^q	NS	Sasaki and Kameoka (1992)
Gasoline HDV					
1979 Ford	NS	11,000	96	133	Dietzmann et al. (1981)
1979 Int'l Harvester	NS	15,000	48	272	Dietzmann et al. (1981)

TABLE 10 (CONT'D): N_2O EMISSIONS FROM MOTOR VEHICLES

Type of vehicle	Emission control equipment	Odometer (miles)	N_2O Emissions (mg/mi)	Ratio, NO_x/N_2O^a	Reference
Diesel HDV					
1977 DDT 2-stroke	NS	60,000	$68\text{-}85^v$	494-571	Dietzmann et al. (1980)
1979 Caterpillar 4-stroke	NS	7,000	$22\text{-}35^v$	545-785	Dietzmann et al. (1980)
1979 Mack 4-stroke	NS	69,000	$50\text{-}58^v$	500-565	Dietzmann et al. (1980)
1979 Cummins 4-stroke	NS	26,000	$35\text{-}47^v$	517-773	Dietzmann et al. (1980)
Diesel truck engine	NS	NS	2000	NS	Robertson (1991)
Flexible-fuel methanol/gasoline vehicles					
Ford Escort-1	3 WY	$4{,}000^w$	61 M100; 105 M85/I; 99 I	4.3 M100; 4.1 M85/I; 4.4 I	Ford $(1988a)^x$
Ford Escort-2	3 WY	$50{,}000^w$	119 M100; 111 M85/I; 147 I	4.6 M100; 5.9 M85/I; 5.6 I	Ford $(1988a)^x$
Ford Escort-2	no catalyst	NS	-3 M100; -2 M85/I	--	Ford $(1988a)^x$
Ford Crown Victoria-1	3 WY	$0/8{,}000/16{,}000^w$	6/17/16 M85/G	90/34/53 M85/G	Ford $(1988a)^x$
Ford Crown Victoria-2	no catalyst	NS	0 M100; 3 M85/G	--	Ford $(1988a)^x$
Ford Crown Victoria-2	3 WY	$3{,}000^w$	14 M100; 12 M85/G	37 M100; 59 M85/G	Ford $(1988a)^x$
3 Ford 4.9 liter 6-cyl. vans	3 WY	5,000, 15,000, 25,000	61 M85, 65 M85, 70 $M85^r$	NS	Battelle (1995)
CNG vehicles					
3 1992 Ford 4.9 liter vans	3 WY	5,000, 15,000, 25,000	$26, 36, 44^r$	NS	Battelle (1995)
3 1992 Dodge 5.2 liter vans	3 WY, CNG opt.	5,000, 15,000, 25,000	$9, 24, 39^r$	NS	Battelle (1995)
3 1992 Chevy 5.7 liter vans	3 WY, Engelhard CNG	5,000, 15,000, 25,000	$47, 50, 54^r$	NS	Battelle (1995)
Propane vehicles					
3 1992 Ford 4.9 liter vans	3 WY	5,000, 15,000, 25,000	$99, 74, 68^r$	NS	Battelle (1995)
3 1992 Chevy 5.7 liter vans	3 WY	5,000, 15,000, 25,000	$69, 87, 101^r$	NS	Battelle (1995)

Notes: see next page.

See Dasch (1992) for a similar table.

EPA testing protocol requires that all vehicular emissions be corrected for background concentration (i.e., that the ambient concentration be subtracted from total measured emissions). We therefore assume that all reported emissions are net of background, unless it is clear that they are not (as in Weiss & Craig, Pierotti & Rasmussen). LDV = light-duty vehicle; HDV = heavy-duty vehicle; 3WY = three-way catalytic converter; OC = oxidation catalytic converter; AP = air pump; EGR = exhaust gas recirculation; EFI = electronic fuel injection; TBI = throttle-body fuel injection; MPFI = multipoint fuel injection; NS = not specified; FTP = Federal Test Procedure; NYCC = New York City Cycle; HFET = Highway Fuel Economy Test; RFG = reformulated gasoline.

All LDVs except those not identified (noted as "NS" under "vehicle" column) were tested over the FTP. All HDVs except the "diesel truck engine" (Robertson, 1991) were tested over the chassis version of the 1983 Heavy Duty Transient Cycle (HDTC).

^The same vehicle as the one immediately above (i.e., not a different vehicle of the same model). Vehicles of the same description but not marked with "^" are the same model but different vehicles.

[a] This is the ratio of g/mi emissions, not the ratio of ppm, except as noted.

[b] A gross concentration of 0.1-0.2 ppm was measured in the exhaust. The background N_2O is 0.3 ppm.

[c] The emission value in parentheses is the result when the vehicle was run on gasoline containing 16.4% MTBE.

[d] The mg/mi figure shown is the reported average of several tests with the emission control system functioning properly. Disabling the EGR increased N_2O emissions by a factor of 1.5-3.0. (Prigent and de Soete [1989] also found that N_2O emissions increased when EGR was disconnected). Other malfunctions were relatively unimportant.

[e] The emission rates reported in test cycles other than the FTP were similar to the FTP emission rates.

[f] The five numbers represent, respectively, results for the Japanese "11-step" driving cycle (with cold start), the Japanese "10-step" driving cycle, urban driving cycle at 10 km/hr, urban driving cycle at 20 km/hr, and urban driving cycle at 50 km/hr. Tests were performed using a chassis dynamometer, sample collection in a heated (393 K) steel tank, and gas chromatography analysis at 543 K.

[g] The first number is the average for the 10 cars as received; the second number is the average after the cars were tuned up. The 10 vehicles were: a 1978 Buick Regal (OC, EGR), 2 1979 Mercury Marquis (3WY, EGR, AP); a 1978 Ford Granada (OC, EGR, AP); a 1978 Volvo 245 DL (3WY); a 1978 Oldsmobile Cutlass (OC); a 1978 Chevrolet Malibu (OC, EGR, AP); a 1978 Chevrolet Monte Carlo (OC, EGR); a 1978 Ford Fiesta (OC, EGR, AP); and a 1978 Chrysler New Yorker (OC, EGR). The Malibu was the only vehicle that had been previously tested (Urban and Garbe, 1979; the model with the air pump [AP]); most of the others were the same model as previously tested vehicles (see entries in this table), but not the same actual vehicle.

[h] The vehicles were tested with a variety of malfunctions, including: a disabled oxygen sensor, disabled EGR, 12% misfire, and high oil consumption. The very high emissions from the Pinto and the Marquis were the result of a disabled EGR system.

[i] The first emission number is at zero miles; the second is at 15,000 miles. Emissions were consistently higher in the NYCC and lower in the HFET than in the FTP. N_2O emissions did not vary appreciably with the type of gasoline.

[j] The first number is the result when the FTP was run at normal temperature (78° or 81° F); the second number is the result at low temperature (55°, 58°, 60°, or 61° F). Laurikko and Nylund (1993) found that N_2O emissions were higher in a +20°C cold-start test (FTP) than in a -20°C cold-start test.

[k] When the oxygen sensor was disconnected, N2O emissions disappeared.

[l] The SO_2 content of the gas was reported to be 12 ppm for undiluted samples, and ten times less for diluted samples. This is well below what appears to be the concentration that actuates artifactual N_2O formation.

[m] The number in the brackets is the average value.

[n] This range includes the NO_x/N_2O ratios measured for the 1978 Pontiac Sunbird, the 1978 Saab, the 1980 Lincoln Continental, and the 1980 Buick Century.

[o] The first number is the composite result from the three phases of the Urban Dynamometer Driving Schedule (UDDS). The second number is the result from the Highway Fuel Consumption Test (HWFCT).

[p] The three numbers represent, respectively, tests taken over the FTP75 cycle with no catalyst, a new catalyst, and a "severely deteriorated" catalyst.

[q] The five numbers represent, respectively, results for the Japanese "11-step" driving cycle (with cold start), the Japanese "10-step" driving cycle, urban driving cycle at 10 km/hr, urban driving cycle at 20 km/hr, and urban driving cycle at 50 km/hr. Tests were performed using a chassis dynamometer, sample collection in a heated (393 K) steel tank, and gas chromatography analysis at 543 K.

[r] Tests were performed using an on-line FTIR analyzer. The three numbers correspond to the three vehicle mileage figures listed, and each number is a mean value for tests of three different vans by each manufacturer. Note that the Dodge CNG vans are among the first to use a catalyst system designed and optimized for CNG vehicles.

[s] The three numbers represent, respectively, tests performed on the ECE 15-04 Driving Cycle (with cold start), the European Urban Driving Cycle (EUDC), and the ECE Driving Cycle. As in note a, the ratio of NOx to N_20 is based on mass and not ppm.

[t] The emissions data have been converted from units of grams of N_20 per kilometer. The first number represents emissions based on the Swedish driving cycle with cold start, and the second number represents emissions on the Swedish driving cycle with hot start.

[u] The emissions data have been converted from units of grams of N_2O per kilometer. The first number represents emissions based on the ECE 15 (cold start) driving cycle and the second number represents emissions based on the EUDC.

[v] Emissions varied with the quality of the diesel fuel used.

[w] Miles accumulated on the catalytic converter.

[x] Ford reported detailed speculated FTIR data for two flexible-fuel Escorts and two flexible-fuel Crown Victoria, at different methanol/gasoline mixtures, and with different catalysts and catalyst ages. The Escort was tested on indolene, the Crown Victoria on gasoline.

TABLE 11. N_2O EMISSIONS FROM COMBUSTION SYSTEMS

Combustor type	Fuel	Sampling method	Temp. (K)	NO_x (ppm)[a]	N_2O (ppm)	Reference
Coal combustion						
6 full-scale utility boilers	medium-S bituminous coal	on-line GC	NS	386-559	<4	Linak et al. (1990)
29 kW DF tunnel furnace	4 bituminous coals	on-line GC	NS	553-753	2.2-4.2	Linak et al. (1990)
Coal furnace	bituminous coal	on-line GC	NS	216-1121	1.28-4.45	Linak et al. (1990)
7 175-700 MW, LNB	coal (S removed from gas)	tedlar bags, S removed	NS	NS	0.5	Yokoyama et al. (1991)
171 MW CF drum boiler	bituminous coal, 0.6% S	on-line GC	NS	386	3.3	Ryan and Srivastava (1989)
50 kW coal combustor	coal	on-line IR	NS	2000	30	Ryan and Srivastava (1989)
~450 MW WF boiler	coal	on-line ND IR	NS	625-735	11-16[b]	Sloan and Laird (1990)
490 MW CF boiler, LNB	coal	on-line ND IR	NS	270-300	2-3[b]	Sloan and Laird (1990)
OF utility boiler	coal	on-line ND IR	NS	662-982	2-11	Kokkinos (1990)
TF utility boiler	coal	on-line ND IR	NS	325-498	1	Kokkinos (1990)
IGCC power plant	coal	on-line ND IR	NS	19	1	Kokkinos (1990)
500 MW CF boiler, LNB	coal	on-line IR	NS	NS	2.2-3.0	Laird and Sloan (1993)
500 MW CF boiler, LNB	coal	off-line GC[c]	NS	270-330	0.6-4.0	Laird and Sloan (1993)
500 MW CF boiler	coal	off-line GC[c]	NS	470-530	1.9-7.1	Laird and Sloan (1993)
500 MW CF boiler	coal	on-line IR	NS	470-530	7-13	Laird and Sloan (1993)
500 MW WF boiler	coal	off-line GC[c]	NS	512-538	0.2-1.4	Laird and Sloan (1993)
660 MW OF boiler, LNB	coal	on-line IR	NS	360-420	2.5-7.1	Laird and Sloan (1993)
660 MW OF boiler	coal	on-line IR	NS	640-740	3.7-4.3	Laird and Sloan (1993)
500 MW WF boiler, LNB	coal	off-line GC[c]	NS	390-450	1.6-2.6	Laird and Sloan (1993)
500 MW WF boiler	coal	off-line GC[c]	NS	690-750	4.2-5.4	Laird and Sloan (1993)
500 MW WF boiler, LNB	coal	off-line GC[c]	NS	390-450	0.2-1.2	Laird and Sloan (1993)

TABLE 11 (CONT'D): N_2O EMISSIONS FROM COMBUSTION SYSTEMS

Combustor type	Fuel	Sampling method	Temp. (K)	NO_x (ppm)^a	N_2O (ppm)	Reference
Fuel-oil combustion						
FF utility boiler	oil	on-line ND IR	NS	268	1	Kokkinos (1990)
21 156-1000 MW boilers	oil (S removed from gas)	tedlar bags, S removed	NS	NS	0.3	Yokoyama et al. (1991)
733 kW boiler	#5 fuel oil (236 ppm S in gas)	on-line GC	NS	189 NO	1.3	Linak et al. (1990)
733 kW boiler	#2 fuel oil (58 ppm S in gas)	on-line GC	NS	105 NO	0.30	Linak et al. (1990)
588 kW boiler, LNB	#2 fuel oil (130 ppm S in gas)	on-line GC	NS	64/536 NO^d	<0.24/0.27^d	Linak et al. (1990)
588 kW boiler, LNB	#5 fuel oil (270 ppm S in gas)	on-line GC	NS	60/682 NO^d	0.26/0.73^d	Linak et al. (1990)
320 MW OF	#6 fuel oil, 3% S	on-line GC	NS	220-480	n.d.-1	Ryan and Srivastava (1989)
3 500 MW WF boilers	oil	off-line GC	NS	550-650	0.4-0.9	Laird and Sloan (1993)
Natural-gas combustion						
FF utility boiler	NG	on-line ND IR	NS	120	2	Kokkinos (1990)
Combustion turbine	NG	on-line ND IR	NS	19	4	Kokkinos (1990)
733 kW boiler	NG (0 ppm S in gas)	on-line GC	NS	62 NO	<0.24	Linak et al. (1990)
588 kW boiler, LNB	NG (4-6 ppm S in gas)	on-line GC^c	NS	50/638 NO^d	<0.24/0.72^d	Linak et al. (1990)
15 175-1000 MW, LNB	LNG (S removed from gas)	tedlar bags, S removed	NS	NS	0.1	Yokoyama et al. (1991)
35 MW turbine, no water inject.	NG	NS	NS	NS	0.5-1.9	De Soete (1993)
35 MW turbine, water injection	NG	NS	NS	NS	0.75-1.65	De Soete (1993)

TABLE 11 (CONT'D): N_2O EMISSIONS FROM COMBUSTION SYSTEMS

Combustor type	Fuel	Sampling method	Temp. (K)	NO_x (ppm)[a]	N_2O (ppm)	Reference
Fluidized-bed combustion						
100-kW FBC	coal	on-line IR	NS	NS	100	Ryan and Srivastava (1989)
CFBC	coke	aluminum bags, stored dry	1180	32	124^e	Amand and Anderson (1989)
CFBC	coke	aluminum bags, stored dry	1073	32	170^e	Amand and Anderson (1989)
CFBC	NS	on-line ND IR	NS	NS	20-100	Kokkinos (1990)
CFBC	coal	NS	NS	NS	80-315	Makansi (1991)
1 MW CFBC	lignite coal, 0.7% S	on-line ND IR	1125/1020	NS	$22/125^{f,b}$	Mann et al. (1992)
1 MW CFBC	bituminous coal, 2.3% S	on-line ND IR	1200/1085	NS	$50/265^{f,b}$	Mann et al. (1992)
130 kW PFBC	peat, 0.2% S	on-line IR	1073/1123	$140/175^f$	$95/37^f$	Lu et al. (1992)
130 kW PFBC	bituminous coal, 1.2% S	on-line IR	1073/1193	$70/105^f$	$59/4^f$	Lu et al. (1992)
FBCC	petroleum	aluminum bags, S removed	923-1033	~400	3-4	Cooper and Emanuelsson (Cooper and Emanuelsson, 1992)
FBCC	petroleum	aluminum bags, dried	923-1033	~400	12-26	Cooper and Emanulesson (1992)
7 CFBCs 30-160 MW	coal	NS	850-910	NS	20-132	De Soete (1993)
bubbling FBC	coal, 0.4% S	on-line ND IR	1123	200-800	25-90	Shimizu et al. (1992)
Effect of sulfur and temperature						
Fixed-bed laboratory reactor	graphite, no S	on-line GC	1000/1275	$1000/1000^f$	$4/0^f$	De Soete (1989)
Fixed-bed laboratory reactor	graphite plus $CaSO_4$	on-line GC	1275	700	4	De Soete (1989)
Entrained-flow reactor	peat, 0.14% S	on-line IR	1000/1123	710/870	$\sim 380/125^g$	Aho and Rantanen (1989)
1 MW CFBC	lignite coal, 0.7% S	on-line ND IR	1125/1020	NS	$22/125^{f,b}$	Mann et al. (1992)
1 MW CFBC	bituminous coal, 2.3% S	on-line ND IR	200/1085	NS	$50/265^{f,b}$	Mann et al. (1992)
130 kW PFBC	peat, 0.2% S	on-line IR	1073/1123	$140/175^f$	$95/37^f$	Lu et al. (1992)
130 kW PFBC	bituminous coal, 1.2% S	on-line IR	1073/1193	$70/105^f$	$59/4^f$	Lu et al. (1992)

TABLE 11 (CONT'D): N_2O EMISSIONS FROM COMBUSTION SYSTEMS

Combustor type	Fuel	Sampling method	Temp. (K)	NO_x (ppm)^a	N_2O (ppm)	Reference
Effect of emission controls						
160-kW air-staged burner	oil (S removed from gas)	SS probes, S removed	NS	$210/280^h$	$< 2/<2^h$	Nimmo et al. (1991)
588-kW boiler, LNB	#2 fuel oil (130 ppm S in gas)	on-line GC	NS	$64/536^d$	$<0.24/0.27^d$	Linak et al. (1990)
588-kW boiler, LNB	#5 fuel oil (270 ppm S in gas)	on-line GC	NS	$60/682^d$	$0.26/0.73^d$	Linak et al. (1990)
588-kW boiler, LNB	NG (4-6 ppm S in gas)	on-line GC	NS	$50/638^d$	$<0.24/0.72^d$	Linak et al. (1990)
130 kW PFBC w/air staging	peat, 0.2% S	on-line IR	NS	$110/55^i$	$70/90^i$	Lu et al. (1992)
130 kW PFBC w/air staging	bituminous coal, 1.2% S	on-line IR	NS	$70/25^i$	$59/72^i$	Lu et al. (1992)
130 kW PFBC w/NH_3	peat , 0.2% S	on-line IR	NS	$195/70^i$	$60/90^i$	Lu et al. (1992)
Front-fired boiler w/urea	oil	on-line ND IR	NS	117	23	Kokkinos (1990)
175-MW boiler w/SCR	coal (S removed from gas)	tedlar bag, S removed	NS	NS	$1.9/1.3^j$	Yokoyama et al. (1991)
700-MW boiler w/SCR	coal (S removed from gas)	tedlar bag, S removed	NS	NS	$1.2/0.7^j$	Yokoyama et al. (1991)
175-MW boiler w/FGD	coal (S removed from gas)	tedlar bag, S removed	NS	NS	$1.5/1.4^j$	Yokoyama et al. (1991)
Effect of sample storage:						
29-kW DF tunnel furnace	bituminous coal	SS container wet, 0/1/4/150 hrs	NS	757	$\sim 5/145/155/180^k$	Linak et al. (1990)
29-kW DF tunnel furnace	bituminous coal	SS container dry, 0/1/4/140 hrs	NS	757	$\sim 5/5/5/15^k$	Linak et al. (1990)
165-MW utility boiler	bituminous coal, medium-S	SS container wet, 0/1/4/150 hrs	NS	354	$\sim 2/35/50/65^k$	Linak et al. (1990)
450-MW WF boiler	coal	plastic bag wet, 3/8/15 days	NS	$735/725/645^b$	$53/184/212^b$	Sloan and Laird (1990)
450-MW WF boiler	coal	plastic bag dry 3/15 days	NS	$700,720/625^b$	$12,18/15^b$	Sloan and Laird (1990)

Notes: see next page.

IGCC= integrated-gasification combined-cycle; LNB = low-NO_x burner; CFBC = circulating fluidized-bed combustor; FBC = fluidized-bed combustor; PFBC = pressurized fluidized-bed combustor; FBCC = fluidized-bed catalytic cracker; LNG = liquefied natural gas; DF = down-fired; TF = tangentially fired; OF = opposite-fired; CF = corner-fired; WF = wall-fired; FF = front-fired; SCR = selective catalytic reduction; S = sulfur; FGD = flue-gas desulfurization; GC = gas chromatograph; IR = infrared spectrometer; ND = non-dispersive; SS = stainless steel; MW= megawatt; ND = none detected; NS = not specified.

See Ryan and Srivastava (1989) for a tabulation of results obtained prior to 1989, and also (U.S. DOE, 1991), Appendix B.

[a] Reported as NO in Cooper and Emanuelsson (1992); Nimmo et al. (1991); Linak et al. (1990), Aho and Rantanen (1989), and de Soete (1989), and as NO_x in the others.

[b] All gas concentrations were measured at 3% oxygen, dry. In all cases, N_2O emissions measured by on-line IR were between 11 and 17 ppm.

[c] Off-line gas chromatography analyses, using electron capture detection, were conducted using either 5.5 liter Saran (polyvinylidene chloride) or 10 liter polyethylene/aluminum gas sampling bags. GCs were installed on-site, and time from sampling to analysis was less than 10 minutes.

[d] The first emission result is with combustion air staging; the second result is without.

[e] The results were reported as 73 mg-N_2O/MJ-coke and 100 mg-N_2O/MJ-coke, with a conversion factor of 1.7 ppm/(mg/MJ).

[f] The first concentration number corresponds to the first temperature, the second to the second temperature.

[g] The first emission measurement is at the first temperature, the second is at the second temperature. The N_2O concentration was reported at 1073^oK, per 10% CO_2. They also reported the percentage of the nitrogen in the fuel that was converted to N_2O at 1000, 1073, and 1123^oK. We estimated the N_2O ppm concentration at 1000 and 1123^oK by assuming that the ratio of ppmv concentrations at two temperatures is equal to the ratio of fuel conversion percentages at the same two temperatures.

[h] This is the NO concentration with/without combustion-air staging, with 3% excess oxygen (wet).

[i] The first result is without NH_3 injection or combustion-air staging; the second result is with NH_3 injection or combustion-air staging. Measured at 6% oxygen, dry. The emission results with NH_3 injection are at a 4:1 NH_3:NO_x molar ratio; the emission results with combustion-air staging are at a secondary air level of 20 volume percent, and with 25% excess air in the case of peat, and 29% in the case of bituminous coal.

[j] The first number is the concentration before the SCR or FGD inlet; the second number is the concentration at the outlet.

[k] Emission measurements separated by slashes correspond to different sample storage times, in hours, separated by slashes. NO_x concentration shown is initial concentration. In the wet sample taken from the 29-kW tunnel furnace, the SO_2 concentration declined from 800 ppm at 0 hours to near zero after only 1 hour.

TABLE 12: EMISSION FACTORS FOR ENGINES, BOILERS, AND FUEL PRODUCTION FACILITIES (G/10^6 BTU-FUEL-INPUT TO BOILER, OR FEEDSTOCK INPUT TO PLANT, EXCEPT AS NOTED)

Fuel-production facility:	Oil refinery	Oil refinery or corn-to-ethanol	Oil refinery	Oil refinery	Oil refinery	Oil refinery	Natural-gas pipeline	Natural-gas pipeline
source of emissions :	*process areas*	*coal boiler*	*petroleum coke boiler*	*natural gas boiler*	*refinery gas boilers*	*fuel-oil boiler*	*natural-gas turbine*	*natural-gas engine*
CH_4	2.0	0.9	0.6	1.3	0.5	3.0	23.1	524.7
N_2O	0.5	4.0	4.0	2.0	2.0	2.0	2.0	2.0
NMHC	6.5	1.3	3.0	1.2	2.0	0.8	0.9	49.1
CO	8.9	10.8	30.1	15.4	14.9	15.2	77.1	267.6
NO_x	3.0	468.0	225.9	55.4	81.3	150.0	115.7	616.1

Fuel-production facility:	Well equipment	Industrial engine	Industrial engine	Natural-gas-to-methanol plant	Coal-to-methanol plant	Wood to-methanol plant	Wood-to-ethanol plant	Corn-to-ethanol plant
source of emissions:	*diesel fuel*	*gasoline fuel*	*diesel fuel*	*all sources*	*all sources*	*wood gasifier*	*wood FBC*	*process areas*
CH_4	4.5	142.9	15.9	6.0	9.7	3.3	10.0	0.2
N_2O	2.0	10.0	2.0	0.5	1.6	0.7	2.0	0.0
NMHC	40.9	1263.3	179.2	0.3	96.9	42.8	128.0	432.0
CO	367.4	28440.7	430.9	4.2	8.4	10.0	30.0	0.0
NO_x	1124.9	739.4	2000.4	31.2	32.3	12.4	37.0	0.0

Notes: see next page.

DeLuchi (1991) shows emission factors for more sources.

Oil refinery, process areas: Shown here are all emissions *other* than those from fuel combustion (which are calculated separately, using emission factors for natural gas and refinery gas, coal, and oil boilers). The CO, NO_x, and NMHC emissions are based on DeLuchi et al.'s (1992) estimates of controlled emissions from refineries in the year 2000. The NMHC-emission factor includes fugitive emissions, and assumes that all NMHC emissions are controlled. The CH_4 emission factor was estimated on the basis of two sources of data. First, we divided total annual CH_4 emissions from several individual refineries in California and Texas (South Coast Air Quality Management District, 1990; Texas Air Control Board, 1990) by the total 10^6 BTU output of the same refineries (EIA, various years). The result was 0.24 - 2.4 g-$CH_4/10^6$ BTU-product-output. Second, the EIA (1995b) cites an estimate by Radian corporation that fugitive emissions and emissions from flares and tank farms at refineries are 15.6 g CH_4 per barrel of refinery capacity, or about 2.4 $g/10^6$ BTU fuel output. On the basis of this, we assume 2.0 $g/10^6$ BTU fuel output.

The N_2O emission factor is based on data discussed in DeLuchi (1991).

Catalytic crackers can be a large source of CO. In this analysis we assume that CO emissions from catalytic crackers are controlled. However, the control of these CO emissions produces CO_2; we have counted these CO_2 emissions.

The emission factors are in units of $g/10^6$ BTU of products *output* from the plant.

Oil refinery or corn-to-ethanol plants, coal: The CH_4, NMHC, CO, and NO_x emission factors are EPA's AP-42 (1995a) factors for industrial coal boilers. For N_2O we assumed the same emission rates as estimated here for full-size utility coal boilers.

Oil refinery, petroleum-coke: The CH_4, NMHC, CO, and NO_x emission factors are from the third edition of EPA's AP-42 (U.S. EPA, 1977), for boilers. (The fifth edition of AP-42 does not have emission factors for petroleum coke.) For N_2O we assumed the same emission rates as estimated here for full-size utility coal boilers.

Oil refinery, natural gas: The CH_4, NMHC, CO, and NO_x emission factors are EPA's AP-42 (1995a) factors for industrial boilers, except that we assume a 10% reduction in NO_x emissions. For N_2O we assumed the same emission rates as estimated here for full-size utility natural-gas boilers.

Oil refinery, refinery gas: The emission factors are calculated on the basis of the assumed composition of the refinery gas. In essence, there is a separate set of emission factors for each component of refinery gas (CH_4, LPG, H_2S, and H_2). The factors for each component are weighted by the energy share of the component (so that if methane is 40% of refinery gas on an energy basis, then the methane emission factors get a weight of 0.40), and the weighted factors are summed for all of the constituents to produce a weighted-average emission factor. Each set of emission factors (one set for each of the components, CH_4, LPG, H_2S, and H_2) is estimated as $NG_P \cdot K_{P\text{-}C}$, where NG_P is the emission factor for pollutant P from natural-gas-fired industrial boilers, and $K_{P\text{-}C}$ is emissions of P from component C (say, LPG) relative to emissions of P from natural-gas combustion. Thus, all emission factors are estimated relative to the natural-gas factors. Sulfur emissions are calculated on the basis of the sulfur content of the gas, due to H_2S

Oil refinery, fuel oil: The CH_4, NMHC, CO, and NO_x emission factors are EPA's AP-42 (1995a) factors for industrial boilers, except that we assume a 10% reduction in NO_x emissions. For N_2O we assumed the same emission rates as estimated here for full-size utility fuel-oil boilers.

Natural-gas pipeline, turbines: The CH_4, NMHC, CO, and NO_x emission factors are from EPA's AP-42 (1995), "Criteria Emission Factors for Uncontrolled Natural Gas Prime Movers," except that we assume a 25% reduction in NO_x. The N_2O emission factor is the same as for natural gas boilers. The PM emission factors are the sum of solid and condensable PM emissions reported for "Stationary Gas Turbines for Electricity Generation," which are not the same as the turbines included as "Uncontrolled Natural Gas Prime Movers," above.

Natural-gas pipeline, engines: The CH_4, NMHC, CO, and NO_x emission factors are from EPA's AP-42 (1995), "Criteria Emission Factors for Uncontrolled Natural Gas Prime Movers," except that we assume a 50% reduction in NO_x emissions from engines. The factors shown here are the weighted-average of the factors for 2-cycle lean burn (2/3 weight), 4-cycle lean burn (1/6 weight), and 4-cycle rich burn (1/6 weight). The 2/3 weight is based on EPA's (1995) statement that "for reciprocating engines, 2-stroke designs contribute approximately two-thirds of installed capacity" (p. 3.2-1). We assume that the remaining 1/3 is split evenly between the two types of four-stroke engines. The N_2O emission factor is the same as for natural gas boilers. The PM emission factors are the sum of solid and condensable emissions for a 2-stroke lean-burn engine.

Well equipment, diesel fuel: From EPA's AP-42 (1995), "Gaseous Emission Factors for Uncontrolled Stationary Diesel and All Stationary Dual-Fuel Engines." HCHO factor is all aldehydes. $PM_{2.5}$ emission factor is equal to filterable $PM_{3.0}$ plus condensable PM.

Industrial engines, gasoline or diesel fue: From EPA's AP-42 (1995), "Emission Factors for Uncontrolled Gasoline and Diesel Industrial Engines". HCHO factor is all aldehydes. EPA reported PM_{10} but stated that "all particulate is assumed to be ≤ 1.0 μm in size" (3.3-3).

Natural gas-to-methanol, all sources: These estimates are based on the data of Table 14 (especially the estimates therein by Intech (1990) and Heath (1991)).

Coal-to-methanol, all sources: The NMHC and NO_x emission factors are estimated on the basis of the data of Table 15 (we believe that the lower ends of the ranges shown in Table 15 are more reasonable than the upper ends). The CO emission factor is from Chadwick et al. (1987), and pertains to Texaco gasification. To estimate N_2O emissions from fuel combustion in coal-to-methanol plants, we assumed that 20% of the input coal feed goes to the boilers (Paul, 1978; Salmon, 1986), and then use the N_2O emission factor for coal-fired industrial boilers (this table). We then assumed that N_2O emissions from non-fuel combustion processes are equal to emissions from fuel combustion. CH_4 emissions are estimated to be 10% of NMHC emissions. (This might be too low, because CH_4 emissions from coal-burning power plants are more than 10% of NMHC emissions (U.S. EPA, 1995)).

These emission factors are per unit of coal feedstock into the plant (that is, *all* coal feedstock; not just that used to provide process heat).

Wood-to-methanol, wood gasifier: On the basis of the studies in Table 15, we assumed 20g-$NO_x/10^6$-BTU-methanol-output, and then converted this to g/10^6-BTU wood input. To estimate all the other emission factors, we assumed that the ratio of emissions of each of the other pollutants to NO_x emissions, for wood gasification, was the same as the pollutant/NO_x ratio for wood boilers.

These emission factors are per unit of all wood feedstock into the plant.

Wood-to-ethanol, wood FBC: In the enzymatic hydrolysis process, which is the most promising, the lignin is burned to raise steam for process heat and to generate electricity (more electricity is produced than is needed internally; this excess is sold). We use emission factors for fluidized combustion (FBC) of wood (Table 14) to approximate emissions from lignin combustion. The N_2O factor is our estimate (see the text for some discussion of fluidized-bed combustion).

These emission factors are per unit of lignin into the boiler.

Corn-to-ethanol, process areas: I assume that there are three sources of emissions from a corn-to-ethanol plant: 1) on-site boilers; 2) off-site boilers generating electricity purchased by the plant; and 3) other process areas within the plant. Emissions from on-site boilers and off-site electricity generation are estimated elsewhere in this model. We assume that other process areas (the third source) emit CH_4 (from distillation and dehydration [Table 15]) and non-methane hydrocarbons (ethanol vapor [Table 15]), but not CO, N_2O, or NO_x.

These emission factors are per unit of product *output* from the plant.

TABLE 13: ESTIMATES OF G/MI EMISSIONS OF REFRIGERANTS FROM MOBILE AIR CONDITIONERS

Total CFC-12 emissions due to cars		**Cars with a/c**	**Emission rate**
Region and year	10^9 *grams*	10^9 *miles*[a]	*grams/mile*[b]
USA in 1985	31.5[d]	1200	0.026
USA/1986	46.0[c]	1200	0.038
California/1990	4.3[e]	170	0.025
Calculations based on emissions from a single car[f]			
Emissions of CFC-12, 1980s vehicle (Shiller, 1989)			0.0147
Emissions of HFC-134a or CFC-12, 1991 MY and later (Wallington, 1996)			0.0084

a/c = air conditioning.

[a] These are rough estimates based on vehicle miles of travel reported in FHWA (1991; 1988) and a/c use data in Hammerle et al. (1988). We have assumed greater use of air conditioning in California than nationally.

[b] Equal to total grams, from the first column, divided by total miles from the fourth column.

[c] From Ford (Hammerle, et al., 1988).

[d] From the EIA (1989e). The estimate includes manufacturing losses of CFCs.

[e] From CARB (CARB, 1990). The estimate includes manufacturing losses of CFCs.

[f] Prior to the recent transition from CFC-12 to HFC-134a, average CFC refrigerant use declined from about 4 lbs., before concern about the effect of CFCs on stratospheric ozone, to a little under 3 lbs, in response to planned phase-outs of CFCs (Shiller, 1989). We assume that a 1980s vintage vehicle has a 3.5-lb charge of CFC-12, which on average it will lose once, completely, over its 108,000-mile life, on account of collisions (which in many cases damage the a/c system), malfunction, and illegal scrappage. (Of course, in reality some vehicles will never completely vent a single charge to the atmosphere, and some will more vent more than one charge.)

HFC-134a is somewhat less efficient than CFC-12. During the transition to HFC-134a, a/c units were completely redesigned so that the loss in efficiency was made up by better heat exchanger efficiency (about a 5% improvement) and better design. Thus, according to the Ford Motor Company (Wallington, 1996), the amount of refrigerant used in average-sized vehicle a/c systems has remained about 2 lbs., in spite the transition from CFC-12 to HFC-134a. We assume, again, that on average this charge is lost once over the 108,000-mile life of an LDV. This results in an emission rate of 8.4 mg/mi HFC-134a for post 1993-1994 vehicles, and 8.4 mg/mi CFC-12 for 1991-1993 vehicles.

TABLE 14. EMISSIONS FROM FUEL PRODUCTION PROCESSES, GRAMS PER 10^6-BTU FEEDSTOCK INPUT TO PLANT OR 10^6-BTU FUEL INPUT TO BOILER

Reference	Product/process	Feedstock	HCs	CO	NO_x	SO_x	PM	CH_4
Intech (1990)	methanol/steam reforming plant	natural gas	0.2	1.4	68.7[a]	n.e.	n.e.	
Mueller (1990)	methanol/steam reforming plant	natural gas	0.2	5.5	53.7[a]	n.e.	n.e.	
Heath (1991)[b]	methanol/steam reforming plant	natural gas	neg.	25.6	30.9	neg.	neg.	
Ecotraffic AB (1992)[c]	methanol/steam reforming plant	natural gas	15.0	15.0	82.4[a]	<1.0	n.e.	3.0
Darrow (1994)[d]	methanol/steam reforming plant	natural gas	0.3	3.6	3/14.3	0.1	0.1	
Texas Air Board (1990)[e]	methanol/steam reforming plant	natural gas	n.e.	n.e.	n.e.	n.e.	n.e.	1-10
U.S. DOE (1988)	steam/uncontrolled industrial boiler	wood	16-118	15-2000	6-104	0.5-24	104-1360	n.e.
U.S. DOE (1988)	steam/controlled industrial boiler	wood	n.e.	n.e.	n.e.	n.e.	23	n.e.
Tellus (1993)[f]	steam/boilers	agricultural wastes	n.e.	1710	90	n.e.	n.e.	15
U.S. EPA (1995)	steam/stoker boilers	wood and waste	7[g]	686	76[h]	4	2-444[i]	2[g]
U.S. EPA (1995)	steam/fluidized-bed combustion	wood and waste	n.e.	71	101[h]	4	n.e.	n.e.
Ismail and Quick (1991)[j]	steam/fluidized-bed combustion	wood	128	30	37	n.e.	n.e.	n.e.

Notes: see next page.

n.e. = not estimated.

[a] These NO_x emission factors seem relatively high. However, the high temperature of steam reforming, about 1500° F, could cause relatively high NO_x emissions. Low-temperature processes, or processes using pure oxygen, would have lower emissions.

[b] Heath (1991) cites a 1989 study that estimates emissions from eight different sources in a methanol plant. Her estimates were expressed per unit of methanol output; we converted to emissions per unit input assuming a 65% (HHV) conversion efficiency.

[c] The estimates by Ecotraffic AB (1992) appear to be based on emission factors (cited in Swedish studies) for heaters and flares used in the recovery of crude oil. Ecotraffic expressed its estimates per unit of methanol output; we converted to emissions per unit input, using Ecotraffic's estimated 70% (LHV) conversion efficiency. Ecotraffic's estimate of CH_4 emissions is based on an assumed 0.1% gas leakage rate. Its estimate of HC emissions is a "hydrocarbon equivalent," in which any methanol emissions are multiplied by 0.19 (methanol's O_3-forming potential relative to gasoline's).

Ecotraffic AB (1992) estimates N_2O emissions of less than 1.0 $g/10^6$-BTU for NG to methanol.

[d] Darrow's (1994) estimates are based on emission factors for gas boilers. The low NO_x-emissions estimate assumes emission controls in the year 2000; the high estimate assumes no controls today.

[e] Data on CH_4 emissions from plants that produce methanol *and* other products (Texas Air Control Board, 1990), combined with data on the production capacity of methanol facilities (U.S. Department of Commerce, 1985), indicate that CH_4 emissions may be on the order of 1-10 grams per 10^6 BTU of methanol. However, it is not clear how CH_4 emissions should be allocated among the multiple products. Note, though, that this range for methanol (1-10) is consistent with the range estimated for petroleum refineries (0.24-2.4), because methanol plants process natural gas, whereas methanol plants process crude oil, and one would expect higher CH_4 emissions from a facility that process natural gas.

[f] The Tellus (1993) estimates are from an EPA data base.

[g] The most recent version of AP-42 (U.S. EPA, 1995) reports emissions of total organic compounds, but does not differentiate between CH_4 and non-methane emissions. However, an earlier version of AP-42 does differentiate. We have applied the methane/total and non-methane/total ratios from this earlier version to the most recent estimate of emissions of all organics. (Incidentally, the earlier version reported much higher total organic emissions; viz.: 8.1 g-methane/10^6-BTU input, for "wood and bark combustion in boilers.") Data in Dahlberg et al. (1988) indicate that CH_4 emission from the combustion of wood chips is almost 100 times higher than from combustion of fossil fuels -- 300 ppmv in effluent gas vs. 5 ppmv.

[h] Note that the EPA's NO_x emission factors are well below the lowest New Source Performance Standards for any industrial-steam-generating unit (136 $g/10^6$ BTU).

[i] The low factor is emissions from wood-and-bark-fired boiler with an electrostatic precipitator; the high end is uncontrolled emissions from a wood-fired boiler.

[j] Ismail and Quick's (1991) data pertain to a plant in Fresno, California. Wood-fired FBC power plants in Maine must meet a 68 g/10^6 BTU standard for NO_x, CO, and NMHCs.

TABLE 15. EMISSIONS FROM FUEL PRODUCTION PROCESSES, G/10^6-BTU FUEL OUTPUT

Reference	Product/process	Feedstock	HCs	CO	NO_x	SO_x	PM	CH_4
Sperling (1988)	methanol/gasification and synthesis	coal (0.4-0.6% sulfur)	100-500[a]	n.e.	15-150[a]	30-200	1-25	n.e.
Sperling (1988)	ethanol/fermentation	corn	5-140	10-170	100-830	37-1500	45-370	n.e.
U.S. DOE (1983)	ethanol/fermentation	corn	432[b]	n.e.	174	227	76[c]	n.e.
U.S. DOE (1988)	ethanol/fermentation: distillation and dehydration only	corn	n.e.	n.e.	n.e.	n.e.	n.e.	0.22
U.S. DOE (1983)	methanol/gasification and synthesis	wood	n.e.	n.e.	18	n.e.	n.e.	n.e.
Sperling (1988)	methanol/gasification and synthesis	wood	n.e.	n.e.	10-200[a]	neg.	0-30[a]	
Ecotraffic AB (1992)[d]	methanol/gasification and synthesis	wood	8	19	11	3	n.e.	n.e.
U.S. DOE (1988)	ethanol/gasification and fermentation	biomass	n.e.	n.e.	53	104	3	n.e.
Ecotraffic AB (1992)[e]	ethanol/conversion of lignocellulose	tree residues	28	81	93	11	n.e.	32
Ecotraffic AB (1992)[d]	ethanol/conversion of lignocellulose	SRIC trees	121	258	137	42	n.e.	n.e.
NREL (1992)[f]	ethanol/acid hydrolysis and fermentation	switchgrass, wheat grass	6	36	25	9	16	n.e.
NREL (1992)[f]	ethanol/acid hydrolysis and fermentation	cottonwood and alder	9	53	30	4	20	n.e.

Notes: see next page.

n.e. = not estimated; SRIC = short-rotation intensive cultivation; NREL = National Renewable Energy Laboratory. Use of controls is mentioned if known.

[a] According to Sperling (1988), the upper bounds are "suspect."

[b] Ethanol.

[c] Includes fugitive dust.

[d] Ecotraffic AB (1992) assumes that emissions from the methanol and ethanol conversion processes arise from the combustion of biomass (lignin) for process heat. They calculate these emissions by multiplying emission factors for lignin combustion (g/BTU-lignin) by lignin-use factors (BTU-lignin/BTU-fuel). The emission factors for lignin combustion are undocumented assumptions, and are the same for both processes. However, the lignin-use factor is much higher for the ethanol process (1.63:1) than for the methanol process (0.12:1). Hence, the considerable difference in g/BTU-fuel emission rates is due entirely to the considerable (unexplained) difference in assumed lignin usage rates.

[e] Ecotraffic AB (1992) assumes that emissions from this process arise from the combustion of biomass (lignin) and biogas for process heat. They calculate these emissions by multiplying emission factors for lignin combustion (g/BTU-lignin) by lignin-use factors(BTU-lignin/BTU-fuel) and emission factors for biogas (g/BTU-gas) by biogas-use factors (BTU-gas/BTU-fuel). The emission factors for lignin combustion are undocumented assumptions. Their emission factors for biogas combustion are from the product specifications of an engine.

[f] This study includes emissions from all operations at the plant site, including evaporative emissions from storage tanks, emissions from diesel loading equipment, fugitive emissions from vents, emissions from on-site utilities, and more. It is by far the most detailed study of emissions from biomass conversion that we have seen.

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