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Abatement of Nitrous Oxide, Methane, and the Other Non-CO₂ Greenhouse Gases: The Need for a Systems Approach

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Contributions to Global Warming

Carbon dioxide currently accounts for about 49 percent of the radiative forcing of the atmosphere that is attributable to greenhouse gases (3.0 watts per square meter [W m^{-2}]; Houghton et al. 2001). Tropospheric ozone and black carbon are responsible for another 18 percent, and the well-mixed greenhouse gases (GHGs)—principally methane, nitrous oxide, and various halocarbons—are responsible for the remaining 33 percent (Figure 29.1). Changes in well-mixed GHGs and in black carbon, tropospheric ozone, and ozone precursors (NO_x , CO, and NMVOCs), whether engineered or unintentional, could thus have a substantial impact on the radiative forcing of future atmospheres.

All of the well-mixed non-CO₂ GHGs are more potent than CO₂: 100-year global warming potentials (GWPs) range from 23 for methane to >10,000 for a number of the halocarbons (Table 29.1; Prinn, Chapter 9, this volume). Small changes in the net fluxes of these gases can thus have a proportionately larger effect on radiative forcing than similar changes in CO₂ flux. This sensitivity to small changes provides a strong impetus for including the non-CO₂ GHGs in the development of effective mitigation strategies: keeping or removing a given quantity of N₂O from the atmosphere, for example, can have almost 300 times the impact of removing the same mass of CO₂. This impact arises in part from wide differences in atmospheric lifetimes. Short-lived gases such as methane and some halogenated compounds exert most of their radiative influence on decadal or shorter time scales, whereas the influence of N₂O and some halogens persist

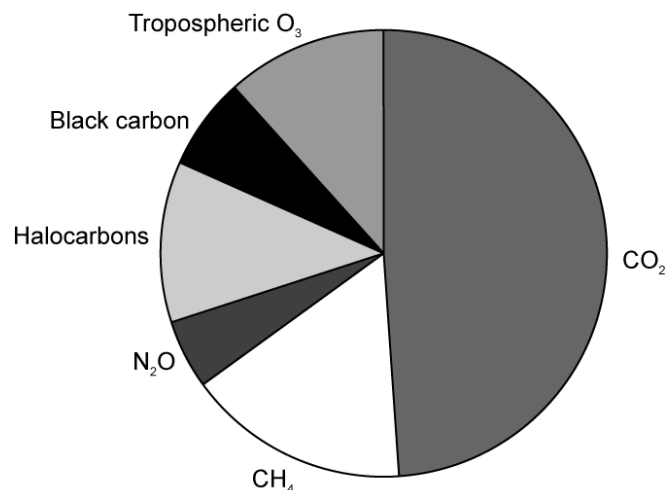


Figure 29.1. Estimated climate forcings (globally averaged) between 1750 and 1998 (data from Houghton et al. 2001)

for centuries, and in the case of a few fully fluorinated species such as sulfur hexafluoride (SF₆), for tens of millennia (Table 29.1).

The importance of the non-CO₂ fluxes are especially apparent when converted to C or CO₂-equivalents using 100-year GWPs (Table 29.2). The total anthropic flux of methane, 344 teragrams of methane per year (Tg CH₄ y⁻¹), for example, is equivalent to 2.2 petagrams of carbon equivalents per year (PgC_{equiv} y⁻¹) and that of N₂O is equivalent to 1.0 PgC_{equiv}. Together these fluxes are similar to the net annual loading of CO₂ to the atmosphere (3.2 PgC; Sabine et al., Chapter 2, this volume).

Until recently, plans for mitigating the buildup of greenhouse gases in the atmosphere have focused primarily on CO₂. Strategies include both reducing CO₂ emissions and capturing emitted CO₂ in biological and other sinks (Caldeira et al., Chapter 5, this volume). More recently the mitigation focus has shifted and grown to include the non-CO₂ gases. There is growing recognition that many of the sources of non-CO₂ gases are manageable and that small changes in flux can be important and have long-term impact. In one analysis, Hansen et al. (2000) argue that the rapid, observable climate warming of recent decades has been driven mainly by the non-CO₂ gases, owing to the offsetting climate forcings of CO₂ and aerosols from fossil-fuel burning. They suggest that mitigating the non-CO₂ gas fluxes, together with reductions in black carbon emissions (Chameides and Bergin 2002), could lead to a faster decline in the rate of global warming than similar reductions in fossil-fuel burning. Although still controversial (Hansen 2002; Schneider 2002; Wuebbles 2002), the argument that non-CO₂ GHGs deserve greater attention is well taken; clearly a combination of mitigation strategies (sensu Hoffert et al. 2002) is warranted.

Table 29.1. Global warming potentials (mass basis) for major greenhouse gases over different time periods

Gas	Atmospheric	Global warming potential by time horizon		
	lifetime (years)	20 years	100 years	500 years
CO ₂			1	1
CH ₄	12	62	23	7
N ₂ O	114	275	296	256
HFCs	1.4-260	40-9,400	12-12,000	4-10,000
PFCs	2,600-50,000	3,900-8,000	5,700-11,900	8,900-18,000
SF ₆	3,200	15,100	22,200	32,400

Source: Houghton et al. (2001).

The impact of non-CO₂ GHG fluxes on radiative forcing also provides a need for understanding how these fluxes will be affected by CO₂ stabilization efforts and by climate change itself. Gases of primarily biological origin—CH₄ and N₂O—are particularly vulnerable to impact. For example, using nitrogen fertilizer to accelerate carbon capture by crops ignores the potential for the additional fertilizer's stimulating N₂O and NO_x production and thereby nullifying most or all of the benefit of additional CO₂ capture and soil C storage (Robertson et al. 2000). The mitigation potential of N fertilizer used to increase productivity is further reduced by the CO₂ costs of its manufacture (Schlesinger 1999). Synergistic interactions are also possible, however. For example, planting cover crops in an annual crop rotation or immediately following harvest of an energy crop such as poplar can both stimulate CO₂ capture and reduce soil nitrogen levels and consequent N₂O production. Understanding the interactions between CO₂ mitigation strategies and fluxes of the non-CO₂ greenhouse gases is crucial for effective CO₂ stabilization policy.

With one possible exception, there are no significant manageable sinks for the non-CO₂ GHGs. The exception, described later in this chapter, is methane oxidation by bacteria in soils abandoned from agriculture. While other sinks are theoretically possible—N₂O reduction to N₂ also occurs in soil and aquatic habitats, and NO_x can be captured by plant canopies—none are known to be particularly manageable or significant vis-a-vis their respective sources. Unlike CO₂, then, mitigation of the non-CO₂ gases depends almost entirely on source attenuation—management intended to reduce the source strength of a given gas. For some gases, notably those with an industrial source such as the halogens, source strength attenuation appears to be working well: the growth rate of the two principal CFCs is near zero and will shortly be negative, owing to Montreal Protocol restrictions on production (Fraser and Prather 1999). For non-CO₂ gases of mainly biologic origin, however, attenuation efforts are untested.

Table 29.2. Annual fluxes of CH₄ and N₂O in carbon-equivalent units (100-year GWP time horizons^a)

<i>Gas/source</i>	<i>Mass flux (Tg y⁻¹)</i>	<i>CO₂-equivalent flux (Pg CO₂-equiv. y⁻¹)</i>	<i>C-equivalent flux (Pg C_{equiv} y⁻¹)</i>
<i>Methane (CH₄)</i>			
Industry	162	3.73	1.02
Energy sector	101	2.32	0.63
Industrial combustion	15	0.35	0.09
Landfills	46	1.06	0.29
Agriculture	182	4.19	1.14
Enteric fermentation	94	2.16	0.59
Rice cultivation	40	0.92	0.25
Biomass burning	34	0.78	0.21
Animal waste treatment	14	0.32	0.09
Total CH ₄ flux	598	13.8	3.8
Anthropic CH ₄	344	7.9	2.2
Non-anthropoc CH ₄	254	5.8	1.6
<i>Nitrous oxide (N₂O-N)</i>			
Industry	1.3	0.61	0.17
Agriculture	6.8	3.16	0.86
Soils	4.2	1.95	0.53
Animal waste treatment	2.1	0.98	0.27
Biomass burning	0.5	0.23	0.06
Total N ₂ O-N	17.7	8.2	2.2
Anthropic N ₂ O-N	8.1	3.8	1.0
Non-Anthropoc N ₂ O-N	9.6	4.5	1.2

Sources: Mosier et al. (1998), Sass et al. (1999), Houghton et al. (2001).

^a See Table 29.1

Points of Intersection: Synergies and Liabilities

The intersection between CO₂ mitigation and the attenuation of non-CO₂ GHGs provides a rich opportunity for exploiting existing synergies. There are CO₂ stabilization strategies that promote GHG abatement that are important to leverage. But there are also strategies that promote non-CO₂ GHG emissions, and it is equally important to avoid these liabilities.

Industry and Transportation

Of the non-CO₂ GHGs, halocarbons are unique in their exclusively industrial origin. They are, however, little affected by CO₂ stabilization technologies. From manufactur-

ing sources--mainly from aluminum smelting, semiconductor production, as replacements for CFCs in refrigerant applications, and from electrical switching equipment (Metz et al. 2001)--there are few direct points of intersection with either fossil-fuel use or engineered sinks for CO₂. Rather, many halogen fluxes intersect with efforts to reduce the production of the stratospheric ozone-depleting compounds (chloro- and bromocarbons) and provide good examples of potential mitigation synergies in general.

Industrial sources of nitrous oxide--mainly adipic and nitric acid production--likewise offer few opportunities for CO₂ stabilization synergies and in any case are declining rapidly because of voluntary abatement (Reimer et al. 2000; Metz et al. 2001). Fossil-fuel and, to a lesser extent, vehicle combustion represent industrial sources of nitrous oxide that are reduced by concomitant reductions in fuel use owing to CO₂ mitigation--a synergistic effect--but these sources represent <5 percent of total global N₂O sources, a small savings.

Of greater importance could be the effects of reduced combustion on NO_x fluxes. These and other ozone precursors (CO and the nonmethane volatile organic compounds [NMVOCs]) exert a greenhouse effect indirectly by affecting concentrations of tropospheric ozone; NO_x is additionally a precursor of nitric acid. Nitric acid is a major constituent of acid rain, which adds nitrate to extensive areas of the Earth's surface at about 25 percent of the global rate of agricultural N fertilizer addition (85 TgN y⁻¹). The fate of this deposited N is unknown, but it is likely that some is denitrified by soil bacteria to N₂O and thus is a potential contributor to the 6 TgN₂O-N y⁻¹ now estimated to be emitted by natural forests and grasslands (Nevison et al. 1996).

Industrial methane sources include emissions from landfills, oil and natural gas processing and transport, coal mining, and wastewater treatment. Methane in landfills is produced by bacteria decomposing organic waste =aerobically. Worldwide, landfills contribute around 40 TgCH₄ y⁻¹ to the troposphere; about twice this amount is contributed by the energy sector (Houghton et al. 2001). Together these sources represent only a modest portion of total global CH₄ emissions--about 17 percent of the 600 Tg annual CH₄ flux--but both of these sources exceed methane's current atmospheric loading rate of 14 TgCH₄ y⁻¹. Thus, relatively minor CO₂ stabilization strategies, such as biogas energy capture and improvements in pipeline and energy production efficiencies, could significantly affect atmospheric CH₄ concentrations. In the United States CH₄ emissions from all industrial sources appear to have declined by 7 percent since 1990 (EPA 2003) owing to the economic value of methane recovery and government incentive and regulatory programs.

Agriculture

Globally, agriculture is responsible for >20 percent of anthropic greenhouse gas emissions; the IPCC (Houghton et al. 2001) estimates that agricultural activities emit 2125 percent of all anthropic CO₂ fluxes, 55-60 percent of total CH₄ emissions, and 6580 percent of total N₂O fluxes. Carbon dioxide emissions are from deforestation and

fossil-fuel use; CH₄ is from enteric fermentation, rice cultivation, biomass burning, and animal wastes; and N₂O is from cultivated soils, animal wastes, and biomass burning. The magnitude of these fluxes and their sensitivity to management makes agriculture an attractive part of many CO₂ stabilization schemes, and because most of these fluxes are interdependent, there are numerous opportunities to exploit synergies; at the same time, there are numerous GHG liabilities to avoid.

CO₂ stabilization strategies for agriculture include

- gains in energy efficiency from improvements in the fuel efficiency of farm machinery, irrigation scheduling, and other farm operations that consume fuel;
- carbon sequestration in soil from changes in tillage, changes in crop residue and animal waste management, and changes in the use of cover crops, fallow periods, and other aspects of crop rotation management;
- the production of biofuels and the emergence of bio-based materials technology to offset the use of fossil fuels for energy production and industrial feedstocks; and
- continued gains in the production or yield efficiencies for grain, livestock, and other agricultural products to defray the need to otherwise open new land for agricultural development and consequent carbon loss.

Each of these strategies affects fluxes of CH₄ and N₂O, sometimes in indirect and unforeseen ways. Examination of the anthropogenic budgets of CH₄ and N₂O (Figure 29.2) provides an indication of likely points of intersection; most occur within three types of production systems: animal industry, lowland rice systems, and upland field crops.

Animal Industry

Animal production is directly responsible for about 31 percent of all anthropic CH₄ emissions and about 26 percent of the anthropic N₂O flux. Most of the CH₄ flux in the animal industry is from enteric fermentation—methane generated by anaerobic bacteria in cattle and sheep rumen. The rate of CH₄ production is nutritional and is thus tightly linked to physiological efficiency: as feed efficiency in developed countries has increased substantially in recent decades, leading to CO₂ savings in production and transportation, methane emissions have correspondingly decreased.

Methane production robs the animal of energy that would otherwise be available for the production of meat, milk, or power; there is thus a financial incentive—based on energy and feed costs—to limit methane emissions. Dietary supplements such as lipids, starches, and ionophores (antibiotic feed supplements) are widely used in feedlots and dairies in the United States to improve feed conversion efficiencies, as are hormones such as bST, and all are known to reduce methane emissions by 8-30 percent per kilogram

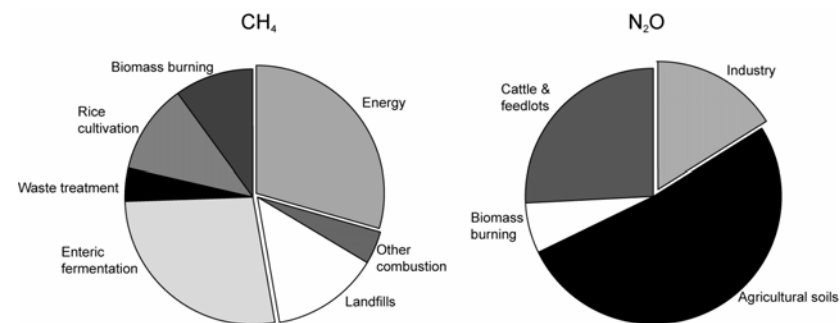


Figure 29.2. Estimated global budgets of the anthropic sources of CH₄ (Tg y⁻¹) and N₂O (Tg Ny⁻¹). The total annual flux of anthropic CH₄ is 344 Tg CH₄ or 2.2 PgC equivalent based on a 100-year time horizon); the total annual flux of anthropic N₂O is 8.1 Tg N₂O-N or 1.0 PgC equivalent (from Mosier et al. 1998; Kroeze et al. 1999; Sass et al. 1999; Houghton et al. 2001). Not included are fluxes from deforestation (see text). Agricultural sources are grouped on the left of each circle.

(kg) of feed. Public resistance—including legislation in parts of Europe—may make this approach difficult to implement elsewhere, however.

Methane production by grazed animals appears more difficult to abate. Production efficiencies from intensive grazing management, which can in theory lead to better utilization of deforested landscapes and thus indirectly contribute to CO₂ stabilization, has not been shown to reduce CH₄ fluxes, although stocking tannin-rich forage has shown promise in New Zealand (Clark et al. 2001). Specific anti-methanogenic vaccines are now under development and may be appropriate for both intensive and extensive grazing systems. Costs, however, will greatly affect their deployment, because they will not necessarily boost feed or forage efficiency. Public resistance may be an additional impediment to adoption.

Animal waste management is responsible for a significant portion of the global CH₄ and N₂O flux. Methanogenesis occurs when waste is stored in anaerobic lagoons or in compost mounds, as occurs in most confined animal feeding operations. Nitrous oxide is produced when the nitrogen in waste undergoes nitrification and denitrification. Two strategies related to CO₂ stabilization can, however, largely abate both sources of these GHGs (CAST 2004). The first strategy is to store waste in capped lagoons or move it to a centralized storage tank that will allow produced methane to be captured and used as an energy source. Such systems are technically feasible but not widely deployed. By keeping the waste anaerobic, any N₂O that is formed from residual nitrate (new nitrate production is inhibited by anaerobic conditions) will be further reduced to N₂. Once

digested, the waste can be spread back onto fields as nitrogen-rich compost, contributing to soil C buildup, although there is a risk that some of the nitrogen added will be subsequently emitted as N₂O.

The second strategy is to immediately spread the waste onto a cropped field or pasture. By keeping the waste aerobic and applying it only when a crop is present, methane production will be minimal and available nitrogen will offset the need for synthetic fertilizer with its high energy and CO₂ cost. Additionally the waste can contribute to soil C accretion. This second strategy is less suitable for intensive centralized confined animal feeding operations, however, owing to the need to transport waste to dispersed fields, and storage will be needed during periods when crop growth is nil, such as wintertime.

Rice Production

Lowland rice accounts for about 12 percent of the anthropic CH₄ budget (Figure 29.2); some 40 TgCH₄ y⁻¹ are emitted by methanogenic bacteria in submerged rice soils (Sass et al. 1999; Houghton et al. 2001). Much of the methane produced is dependent on the rice plant, which provides both substrate (fixed carbon) to the methanogens and a physical conduit to the surface. Like most macrophytes, rice lacks leaf stomates; thus some portion of the methane produced in the rhizosphere is transmitted directly to the atmosphere via aerenchyma in the rice plant. Managing the rice crop thus provides an indirect means to mitigate CH₄ emissions, and recent work (Cotton et al. 2000; van der Gon et al. 2002) has shown a remarkable inverse relationship between CH₄ production and rice yield (Figure 29.3). Well-managed, high-yielding rice crops have substantially lower CH₄ emissions owing to photosynthate-partitioning: higher-yielding plants allocate more C to grain and less C to the rhizosphere where it can undergo methanogenesis. This is an important synergy not yet exploited as increasing rice yields also contributes to CO₂ stabilization by reducing pressure to expand agricultural areas.

Other means for managing CH₄ production in rice fields includes residue management and irrigation scheduling. Incorporating crop litter early in the season significantly diminishes CH₄ release, as does pre-fermentation of added compost (Wassmann et al. 1993); in both cases less labile C is available to methanogens after soil flooding. Likewise, mid-season drainage or alternate flooding and drying markedly decreases the *seasonal* methane flux; in a recent Chinese study (Lu et al. 2000) a five-day, mid-season drainage reduced seasonal CH₄ loss by almost 50 percent.

Rice cultivation also emits N₂O, but because the soil is anoxic during much of the year and nitrification is consequently low, methane dominates the total greenhouse gas flux. In the few studies of total GHG emission in lowland rice (e.g., Abao et al. 2000), N₂O emissions are a small proportion of the total GWP-corrected flux. Where CH₄ emissions are mitigated, however (by adjusting organic matter inputs and the timing and duration of irrigation, for example), N₂O fluxes can dominate and are of a magnitude

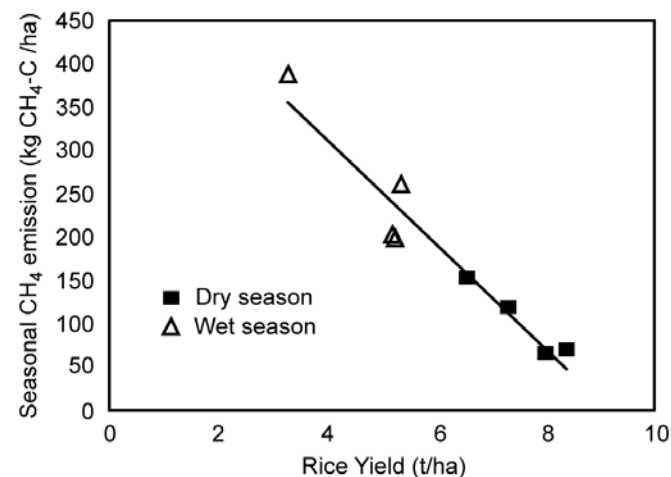


Figure 29.3. Seasonal CH₄ emission in lowland rice as a function of grain yield (from van der Gon et al. 2002, after Corton et al. 2000)

similar to those in other annual cropping systems (Bronson et al. 1997; Wassmann et al. 2003).

Row-Crop Agriculture

Nitrous oxide from agricultural soils accounts for >50 percent of the global anthropic N₂O flux. N₂O is formed during nitrification, the microbial oxidation of NH₄⁺ to NO₃⁻, and during denitrification, the microbial reduction of NO₃⁻ to N₂O and then N₂. Nitrifiers are especially active in well-aerated soil with available NH₄⁺, and denitrifiers in poorly drained soils with available C and NO₃⁻. Denitrifiers are also active in well-aerated soils, particularly following rain events and in anaerobic or partially anaerobic microsites such as the interior of soil aggregates (Robertson 2000).

Nitrous oxide fluxes are highest where inorganic nitrogen is readily available. As a percentage of total nitrogen inputs, N₂O flux appears to be relatively low; for example, the IPCC national inventory methodology calls for calculating total flux as 1.25 percent of N inputs from fertilizer, legumes, compost, and crop residue (Metz et al. 2001). Because modern crop yields are heavily dependent on high nitrogen availability, however, on an absolute basis agricultural N₂O fluxes are very high even where inputs of synthetic fertilizer are low.

Efforts to abate N₂O fluxes in agricultural soils have met with limited success. Specific inhibitors of nitrification (such as nitrapyrin, nitrogen dicyandiamide, and CaC₂) are expensive and work inconsistently. The effects of specific types of fertilizers can be equally inconsistent. Probably the most promise for mitigating field crop N₂O pro-

duction comes from increasing the efficiency of crop N use by manipulating the magnitude, placement, and timing of nitrogen fertilizer. Nitrogen taken up by the crop is not available for microbial uptake; ergo, N₂O production will be low. Effecting this synchrony in most annual cropping systems without affecting crop yields is technically feasible but difficult, and there is little economic payoff with today's low fertilizer cost. For irrigated crops and high-value perennial crops, however, chemigation (the addition of nitrogen and other chemical inputs in irrigation water) and other fertilizer technologies are less difficult and can markedly abate N₂O flux (e.g., Matson et al. 1996).

Because carbon and nitrogen cycling are so tightly linked in most soils, CO₂ stabilization strategies that affect soil C will likely also affect nitrogen cycling and thus N₂O flux. Cover crops that are planted to stabilize or build soil carbon and crop residues that are left in place rather than taken as secondary harvest will potentially immobilize soil nitrogen otherwise available to nitrifiers and denitrifiers. Thus efforts to stabilize CO₂ by sequestering carbon in soil may have synergistic effects on N₂O flux, so long as the soil organic matter in which the nitrogen is stored is not mobilized at a later time, when plant nitrogen uptake is low and N₂O emission potentials high. This potential has not yet been demonstrated experimentally, however, and effects will likely vary by sequestration method. No-till cultivation, for example, does not in theory change nitrogen availability in soil, but the effects of no-till cultivation on N₂O flux are widely variable, and most likely reflect site-specific responses to simultaneous changes in soil aggregate structure, water-filled pore space, and carbon availability (Robertson et al. 2000; Dobbie and Smith 2002). The complexity of this response, coupled with the extreme spatial and temporal variability of soil N₂O fluxes, argues for the development and testing of effective N₂O models (e.g., Li et al. 1997; Del Grosso et al. 2002).

Although theoretically possible, soil methane oxidation is not known to be promoted by any existing agronomic practice. Conversion of forest and savanna soils to agricultural production reduces soil CH₄ oxidation by 80-90 percent (Mosier et al. 1991; Smith et al. 2000). To date, the only documented recovery of a lost CH₄ oxidation capacity is after decades of secondary succession and reforestation (Robertson et al. 2000).

Deforestation and Other Land Use Change

Nitrous oxide fluxes increase following deforestation. Where cessation of deforestation is used to mitigate CO₂ emissions, N₂O emissions will be similarly abated. High emissions persist for only a few years, however, especially under pasture conversion. And since nitrogen gas fluxes prior to deforestation are likely to be higher than subsequent fluxes in either pasture or secondary succession (Robertson and Tiedje 1988; Keller et al. 1993), over the long term, accelerated fluxes during the several years post-clearing may very well be mitigated by the following decades of depressed fluxes.

Likewise, wetland drainage may substantially reduce CH₄ fluxes at the same time

Table 29.3. Sources of greenhouse gas flux in agricultural and forest systems at a U.S. Midwest site

Ecosystem management	Flux (g CO ₂ -equivalents m ⁻² y ⁻¹)						Total net
	CO ₂						
	Soil C	N Fertilizer	Lime	Fuel	N ₂ O	CH ₄	
Annual crop							
Conventional tillage	0	27	23	6	2	4	114
No-till	-110	27	34	12	6	-5	14
Organic	-29	0	0	9	56	-5	41
Energy crop (poplar)	-117	5	0	2	0	-5	-105
Native forest	0	0	0	0	1	-25	-4

that it causes the emission of large quantities of CO₂ from newly aerobic soil organic matter (and possibly large quantities of N₂O). In both of these cases, a long-term, total GHG analysis is needed to evaluate the full effects of land conversion.

The Need for a Systems Approach

The difficulty of assessing the effects of CO₂ stabilization strategies on other parts of a system, and especially on the non-CO₂ GHG fluxes, argues for a systems approach to their implementation. This is especially true in ecosystem settings, where indirect effects can cause counterintuitive, unintentional, and possibly unwanted flux changes. A systems approach can also illuminate synergies not otherwise recognized.

In a U.S. Midwest corn-soybean-wheat rotation, for example, soil carbon sequestration following no-till implementation almost completely mitigated the GHG cost of the cropping system (Table 29.3). Systems analysis showed that substantially more mitigation could potentially be achieved were the system managed to reduce N₂O flux and minimize synthetic fertilizer and agricultural lime use. Similar potentials have been suggested for sugar cane Meier 1998), subtropical wheat (Robertson and Grace 2003), and rice (Wassmann et al. 2003) ecosystems.

Fluxes following land use change can be similarly complex, as noted in the preceding section. Analyses of these systems at a watershed or larger scale could inform landscape management decisions to optimize CO₂ stabilization. In many landscapes it is likely that the value of lower non-CO₂ GHG fluxes will exceed the value of CO₂ mitigation over time, especially for CO₂ strategies that employ sinks that will eventually saturate, such as trees and soil (Caldeira et al., Chapter 5, and Smith, Chapter 28, both

this volume). Even in the absence of CO₂ stabilization per se, including the non-CO₂ gases in mitigation efforts will be highly worthwhile. For many non-CO₂ GHG sources, abatement technology is currently available, and for a number of other sources, new mitigation technology awaits a modest investment in research and development.

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