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EXCHANGE OF TRACE GASES BETWEEN THE TERRESTRIAL BIOSPHERE AND THE ATMOSPHERE IN THE MIDLATITUDES

Keith A. Smith[1], G. Philip Robertson[2] and Jerry M. Melillo[3]

[1]Soil Science Dept., SAC, School of Agriculture
West Mains Road, Edinburgh EH9 3JG, UK
[2]W.K. Kellogg Biological Station and Dept. of Crop and Soil Sciences
Michigan State Univ., Hickory Corners, MI 49060-9516, USA
[3]The Ecosystems Center, Marine Biological Laboratory
Woods Hole, MA 02543, USA

ABSTRACT

Most terrestrial ecosystems of the midlatitudes have been subjected to human influence. Large areas of forests and grasslands have been converted to agriculture; conversely, reforestation is also extensive in many regions. These changes in land use, the use of fertilizers on agricultural land, and high precipitation inputs of nitrogen from industrial emissions to forests and other natural ecosystems, all have consequences for trace gas exchange, and thus for the atmospheric concentrations of trace gases that contribute to climate change.

The study of these interactions, on a comprehensive world-wide basis, is the task of the IGAC Activity Trace Gas Exchange in Midlatitude Ecosystems (TRAGEX). Although TRAGEX as a coordinated program is still at the planning stage, there is a substantial current research effort in progress in North America, Western Europe and Australia that has already provided some insight into the most important processes governing trace gas exchange in the midlatitudes. Some of the highlights of this work, and major outstanding questions, are outlined in this paper.

Soil carbon availability, temperature/moisture interactions, pH, and nutrient dynamics have been identified as key variables in methane emission from temperate wetlands, but measurement and modeling processes of transfer to the atmosphere are in their infancy. Inputs of nitrogen and restrictions in aeration of topsoils have been shown to reduce very substantially the soil's capacity to act as a sink for CH_4 by microbial oxidation to CO_2. High N inputs promote emissions of N_2O, but it has been shown that large effects on fluxes are caused by variations in soil physical conditions, the chemical form of the N, and soil pH. The very high spatial and temporal variability of N_2O and CH_4 fluxes have made representative flux estimates very difficult to make, and have stimulated major research

efforts in development of improved methods of analysis applicable to areas of 10^3 to 10^4 m^2 and ultimately to the km^2 scale. The linking of process models with the Global Information System (GIS) for large-scale integration is a focus of current planning activity.

Increased CO_2 concentrations and N deposition, and the fact that much of the present forest area of the region is in a mid-successional stage, suggest that there may be a substantial vegetation-related sink for CO_2 in the midlatitudes. Changes in soil organic matter in areas of reforestation and in response to changing agricultural practices may also represent an important contemporary sink for atmospheric CO_2. Feedbacks associated with CO_2 uptake by vegetation, its release during decomposition, and the nitrogen cycle processes that affect plant and microbial growth—including trace gas production—must be better understood to appreciate fully the significance of changing CO_2 sink strengths.

High priority must now be given to three areas: (1) the establishment of flux measurement networks in important mid-altitude regions, especially in those such as the former Soviet Union, China, temperate South America, where comparatively little data have been obtained; (2) the development of adequate process-based models; and (3) the scaling-up of the models to predict fluxes over large regions. This should lead to a substantial improvement in the quality of the input to climate models.

INTRODUCTION

Increasing atmospheric concentrations of gases such as carbon dioxide (CO_2), chlorofluorocarbons (CFC), methane (CH_4) and nitrous oxide (N_2O) are believed to be causing climate change: global warming, increased UVB flux to the earth's surface due to depletion of stratospheric ozone, and changes in precipitation patterns (Houghton *et al.*, 1990, 1992). Efforts such as those by the Intergovernmental Panel on Climate Change to assess the likely extent of these changes by means of climate models are heavily dependent on the quality of information regarding the fluxes of these trace gases as model inputs.

Apart from chlorofluorocarbon compounds, which are entirely industrial in origin, significant proportions of the other main "greenhouse gases" CO_2, CH_4 and N_2O come from processes occurring naturally in the biosphere, and their net fluxes to the atmosphere are increased by activities associated with land use. Additionally there are indirect contributions to the greenhouse effect from carbon monoxide (CO), non-methane hydrocarbons (NMHC), and NO_x (all of which have natural and anthropogenic sources), through their role as precursors of tropospheric ozone.

These biospheric contributions to total emissions are of particular significance in the midlatitudes (30° to 60°). A large proportion of the world's population and the most industrialized societies are concentrated in this zone, mainly in the northern hemisphere. The impacts on the natural terrestrial environment have been correspondingly heavy. They include deforestation, the cultivation of virgin grassland, irrigation and wetland drainage, increasingly intensive use of mineral fertilizers, large-scale production of livestock (and livestock waste), urban waste disposal in landfills, and large contributions of nitrogen and sulfur compounds to atmospheric deposition. All of these processes affect the emissions of greenhouse gases (Andreae and Schimel, 1989).

Currently we have a modest understanding of the principal processes contributing to fluxes of trace gases between the land and atmosphere in the midlatitudes. In recent years we have been able to quantify fluxes under some conditions, to develop models of processes, and to begin the task of scaling flux measurements on a small areal scale to larger scales. However, even in those regions where work has been most intensive, we are not yet able to quantify fluxes well enough to satisfy fully the requirements of the climate models. In addition, there are large regions of the midlatitudes, including most of the former Soviet Union, China, and temperate South America, for which very little information on trace gas fluxes is available. Perhaps most importantly, we do not understand sufficiently well how global change will affect gas fluxes.

In this paper we highlight on going work in some of these fields, including the development of the IGAC-TRAGEX program. TRAGEX is intended to fill these gaps in our knowledge of trace-gas fluxes in midlatitude ecosystems, and to improve our capability of predicting changes in these fluxes.

CO_2 FLUXES

In the last century, the principal cause of atmospheric CO_2 increases was conversion of virgin forest and grassland, much of it in the midlatitudes, to agricultural use (Wilson, 1978). Today, the principal biogenic source is deforestation in the tropics; ecosystems in the midlatitudes appear to be having either no net effect on atmospheric CO_2 concentrations, or are acting as a weak sink. Different modeling approaches, dealing with the difficult problem of resolving relatively small differences between two large numbers—the terrestrial source of and sink for CO_2—have produced conflicting results for midlatitude ecosystems and much discussion.

The *potential* strength of a midlatitude CO_2 sink is, however, less contentious, although its magnitude will depend on the extent of climate change and the as yet unquantified effects of elevated CO_2 and nitrogen deposited from the atmosphere on forest growth and soil carbon storage. It will also depend on rates of reforestation in some regions, and increases in soil carbon storage brought about by changing agricultural practices, which may even today represent a net contemporary sink for CO_2.

N and CO_2 Fertilization of Midlatitude Ecosystems

The present-day response of carbon stores in midlatitude ecosystems to elevated CO_2 and land management changes is a topic of intense debate. Atmospheric boundary models (e.g., Tans *et al.*, 1990) suggest a major terrestrial sink for CO_2 in the northern hemisphere. This sink appears to total 2.0 to 3.4 Pg of C per year, depending on C sources in regions to the south and north of the midlatitudes. However, the mechanism for this sink is unknown, and a decade of terrestrial modeling has converged on present estimates of a 0.4 to 2.6 Pg *source* of atmospheric C globally (Dale *et al.*, 1991). With some significant exceptions (e.g., Harmon *et al.*, 1990; Melillo *et al.*, 1988) little of this contemporary source can be traced to the temperate latitudes of Europe and North America (Houghton *et al.*, 1987), but neither do these regions appear at present to be a significant net sink: even when accounting for upward revisions in estimates of recent midlatitude forest growth (e.g., Kauppi *et al.*, 1992 for European forests), both North America and Europe appear to be within 0.1 Pg yr^{-1} of no net emission (Figure 1).

While terrestrial models suggest no present sinks for CO_2 in the midlatitudes, there is general agreement among most models that *future* changes in atmospheric CO_2 concentration and climate will increase forest C storage. Rastetter *et al.* (1992), for example, predict a 4% increase in C storage in vegetation and soil in a deciduous forest of the eastern U.S., after 50 years of doubled atmospheric CO_2. Smith *et al.* (1992) predict a 0.4 to 9.5% increase in vegetation and soil C storage worldwide under different GCM climate scenarios based on Holdridge Life Zones (Holdridge, 1947); this increased storage derives mainly from northward shifts in the extents of tropical and boreal forests and in the midlatitudes from a decrease in the extent of deserts.

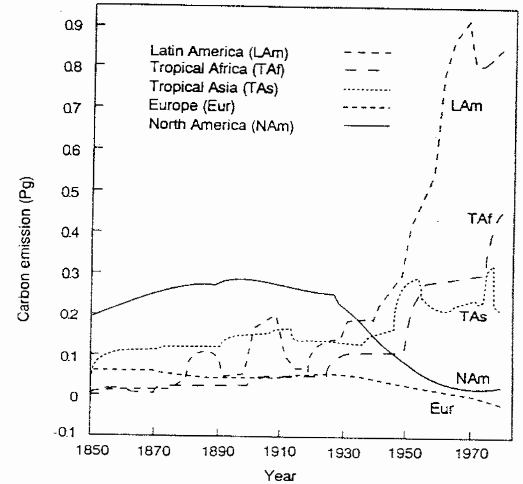

Figure 1. Estimated annual carbon emissions to the atmosphere from different continental regions due to land use changes (redrawn from Dale *et al.*, 1991).

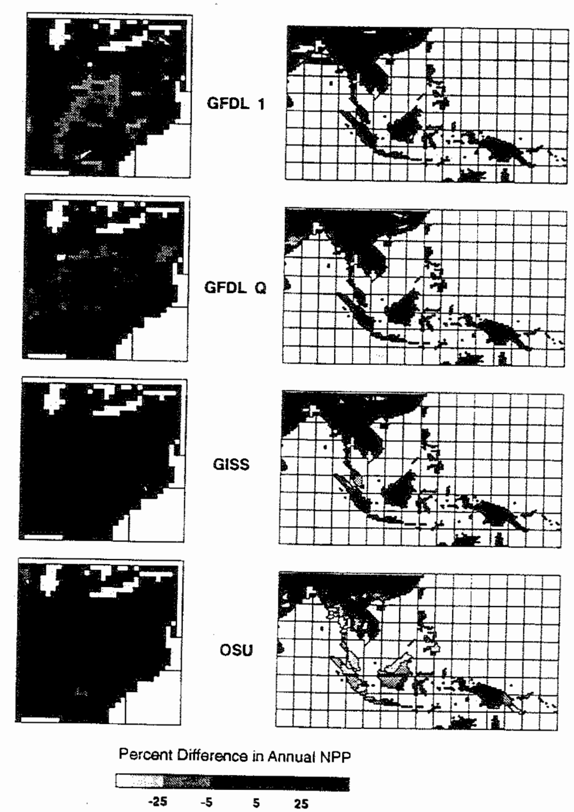

Figure 2. Percent difference in annual net primary production (NPP) between contemporary climate at 313 ppmv CO_2 and four GCM climates at 625 ppmv CO_2 as predicted by the Terrestrial Ecosystem Model for eastern North America (left) and southeast Asia (right) (Melillo *et al.*, 1993).

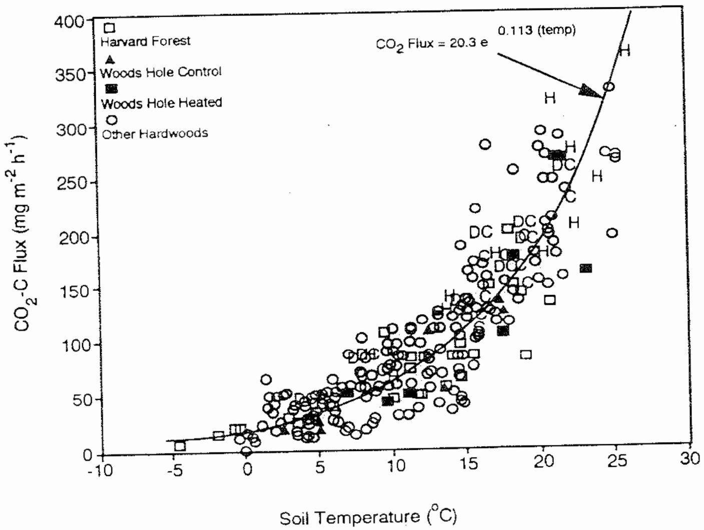

Figure 3. Relationship between CO_2 flux and soil temperature for hardwood forests around the world. Letters refer to Harvard Forest experimental plots (C = control; DC = disturbance control; H = heated plots) (redrawn from Peterjohn *et al.*, 1993).

Melillo *et al.* (1993) have used a process-level ecosystem model to estimate global patterns of net primary production (NPP) and soil nitrogen cycling following a doubling of CO_2 and associated climate changes. Doubling of CO_2 without climate change is predicted to result in a global NPP increase of 16.3%, while changes in both CO_2 and climate increase global NPP by 20 to 26% (Table 1). Predicted responses differ widely between different vegetation/climatic zones (Figure 2); in general, responses for northern and temperate ecosystems are less than 10%. The responses in tropical and dry temperate ecosystems, in their model, are dominated by the effects of elevated CO_2, but those in northern and moist temperate ecosystems reflect primarily the effects of temperature as it influences nitrogen availability and rates of CO_2 release by decomposition from soil organic matter. As Figure 3 shows, the latter effect is marked.

In the last few decades, many regions in the midlatitudes have experienced increased N deposition from the atmosphere as a result of industrial and agricultural emissions of NO_x and ammonia. About 18 Tg yr^{-1} of anthropogenic N is presently added to North American and European ecosystems from this source (Melillo *et al.*, 1989). This inadvertent fertilization will almost certainly increase carbon storage in these systems by stimulating net primary productivity, and may well affect the fluxes of other greenhouse gases such as CH_4 and N_2O (this is discussed further in following sections).

Carbon Storage in Agricultural Soils

In the mid-19th century, agriculture rapidly expanded across virgin grassland and forest regions of North America, temperate South America, Australia, New Zealand, and southern Africa. Native soil fertility and various social factors during this period discouraged the use of crop rotations, manuring, ley cropping and other cultural practices that might otherwise have maintained soil C stores. This led to a large pulse of CO_2 entering the atmosphere prior to 1900. $\delta^{13}C$ analysis suggests that this pulse may have been as high as 110 Pg (Wilson, 1978).

Table 1. Comparison of annual NPP (10^{15} g C) by vegetation type for experiment involving two levels of atmospheric CO_2 and five levels of climate. Climate scenarios correspond to contemporary climate and predictions of four separate global circulation models (GCMs) from the Geophysical Fluid Dynamics Laboratory (GFDL; models 1 and Q), the Goddard Institute for Space Studies (GISS), and Oregon State University (OSU) (Melillo *et al.*, 1993).

CO_2 Scenarios:	312.5 ppmv					625.0 ppmv				
Climate Scenarios†:	C	1	Q	G	O	C	1	Q	G	O
Polar desert/alpine tundra	0.4	0.4	0.4	0.4	0.4	0.5	0.5	0.5	0.5	0.5
Wet/moist tundra	0.6	0.7	0.7	0.7	0.7	0.6	0.8	0.7	0.7	0.7
Boreal woodland	1.1	1.4	1.3	1.3	1.3	1.1	1.6	1.4	1.4	1.4
Boreal forest	2.9	3.8	3.6	3.6	3.5	2.9	4.4	4.0	3.7	3.7
Temperate coniferous forest	1.1	1.1	1.1	1.1	1.1	1.2	1.3	1.3	1.4	1.3
Desert	0.6	0.6	0.5	0.6	0.6	0.9	1.0	0.9	1.0	1.0
Arid shrubland	1.8	1.8	1.8	1.9	1.9	2.3	2.5	2.5	2.7	2.6
Short grassland	1.0	1.2	1.2	1.2	1.1	1.1	1.4	1.3	1.4	1.2
Tall grassland	1.2	1.4	1.4	1.5	1.3	1.3	1.5	1.5	1.6	1.4
Temperate savanna	2.2	2.3	2.4	2.6	2.4	2.5	2.9	2.9	3.1	2.9
Temperate deciduous forest	2.2	2.0	2.1	2.5	2.3	2.3	2.4	2.6	2.8	2.6
Temperate mixed forest	3.3	3.4	3.5	3.7	3.6	3.6	4.0	4.1	4.2	4.0
Temperate broadleaf evergreen forest	2.2	2.3	2.2	2.2	2.2	2.6	2.8	2.8	2.8	2.7
Mediterranean shrubland	0.5	0.5	0.5	0.5	0.5	0.6	0.6	0.6	0.7	0.6
Tropical savanna	5.3	5.7	5.6	6.0	6.0	5.6	6.3	6.3	6.7	6.6
Xeromorphic forest	2.9	2.7	2.7	2.7	2.9	3.7	3.7	3.8	3.8	4.1
Tropical deciduous forest	3.8	3.4	3.5	3.3	3.5	4.5	4.5	4.6	4.5	4.6
Tropical evergreen forest	18.0	16.4	16.3	15.6	14.3	22.0	21.9	21.8	21.3	19.3
Total‡	51.0	51.1	50.8	51.5	49.8	59.3	64.3	63.8	64.2	61.2

†Column headings: C = Contemporary climate, 1 = GFDL-1 model, Q = GFDL-Q model, G = GISS model, O = OSU model.

‡Ecosystem-based estimates may not sum to totals because of the effects of rounding in reporting those estimates.

Farm mechanization and the intensive use of agrochemicals in the midlatitudes since World War II have further depleted soil C stores by the widespread replacement of mixed or animal-based farming systems with continuous monocultures. On the other hand, the higher productivity of intensively fertilized systems may be increasing soil C stores depleted by initial (pre-1900) cultivation where animal-based rotations were not previously important, e.g., most of the former North American prairie (Paustian *et al.*, 1993).

In most agroecosystems, annual cropping provides little or no scope for long-term C storage in vegetation, so soil C storage offers the principal potential for a C sink. Three mechanisms can increase soil C storage in these systems: (1) increased C inputs, (2) decreased decomposition rates, and (3) reduced amounts of CO_2 produced per unit of organic matter decomposed. Thus, where intensification of farming has resulted in higher productivity, C storage can be increased by the associated increased C inputs, so long as decomposition rates remain constant and crop residues are left on the field. Long-term soil C experiments bear this out (Paustian *et al.*, 1993), as do simulation models that show 20 to 30% increases in soil C with 40% increases in net primary production. In fact, Paustian *et al.* have shown that a modest increase in primary production (from increased yields or from incorporation of cover crops into a rotation) results in far more impact on soil C storage in soil than does a 2°C climate change (Figure 4).

Carbon storage may also increase in agricultural soils as a result of decreased decomposition rates, which may result from conservation tillage practices. Soil aggregation which protects organic matter from decomposition by restricting microbial access to substrates (Elliott *et al.*, 1980; Tisdale and Oades, 1982), is typically reduced by tillage. However, aggregate restabilization under conservation or reduced-tillage practices may—together with other effects of conservation tillage—lead to higher C storage under no-till systems (Figure 5). Given the widespread adoption of conservation tillage in North America (Crosson, 1981), this increase in C storage may be significant. Simulation modeling suggests that under optimal circumstances—high productivity, complete residue return, and no-till cultivation—soil carbon stores can largely regain and perhaps exceed pre-cultivation soil C levels under steady-state conditions (Figure 4).

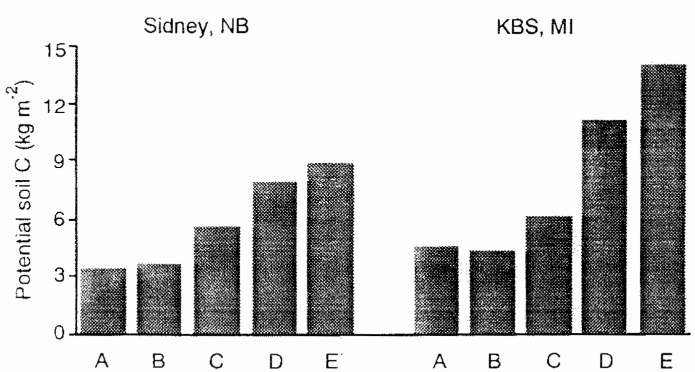

Figure 4. Simulated effects of agricultural management strategies under different climate scenarios on potential carbon storage in soil at a wheat fallow site in Nebraska, U.S., and at a maize site in Michigan, U.S. A = current management; B = A plus a 2°C temperature increase; C = B plus a 40% yield increase; D = C plus conservation tillage; E = D plus a 20% increase in residue lignin content (redrawn from Paustian *et al.*, 1993).

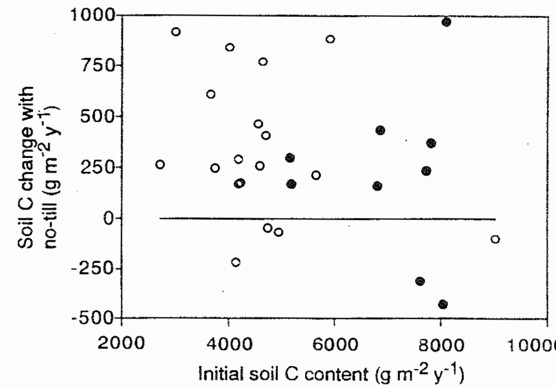

Figure 5. Effects of conservation tillage on soil carbon stores in paired long-term no-till (NT) versus conventional tillage (CT) row crop ecosystems of North America. Open circles represent light textured soils (e.g., sand and silt loams), closed circles represent soils of heavier texture (e.g., clay loams). Note positive effect of no-till on soil C even in soils of high OM content (redrawn from Paustian *et al.*, 1993).

Potential for Managing Carbon Storage

Present global inputs of C as CO_2 to the atmosphere from fossil fuels are circa 5.5 Pg C yr^{-1}. Even the most optimistic reforestation and soil organic matter management scenarios do not envisage the net capture of more than 1 to 2 Pg C yr^{-1} under present climate regimes (Houghton *et al.*, 1990), and once equilibrium is achieved (50 to 150 yr, depending on rates of tree growth and soil organic matter accumulation) this figure decreases to zero. Only under climate change scenarios that allow extensions of high productivity ecosystems into areas now occupied by ecosystems of lower productivity, e.g., the replacement of desert areas by grasslands and the extension of tropical and boreal forests polewards (Smith *et al.*, 1992) will substantial C capture significantly attenuate atmospheric loading from fossil fuels.

Any net effect of climate change on C storage will largely depend, then, on the differential responses of vegetation and soil organic matter to changes in temperature versus precipitation. Jenkinson *et al.* (1991) show via simulation modeling that increasing temperature in the absence of change in precipitation will accelerate soil organic matter decomposition, regardless of its effects on vegetation growth. Soil C stores will remain steady or increase only if: (1) precipitation increases sufficiently to allow enhanced vegetation growth to replace or exceed C lost from decomposition, or (2) precipitation decreases in the midlatitudes sufficiently to retard decomposition in mid-summer.

CH_4 FLUXES

Midlatitude CH_4 Emissions

Important biogenic sources of CH_4 from the midlatitudes include wetlands, paddy rice cultivation, landfills and sewage disposal sites, and ruminant livestock. Recent global estimates of the CH_4 balance (e.g., Tyler, 1991; Mathews and Fung, 1987; Cicerone and Oremland, 1988) suggest that midlatitude wetlands and paddy rice cultivation contribute little to the global flux relative to fluxes from high latitude and low-latitude regions.

However, wetland studies are continuing, and are providing information useful for process-level modeling and new measurement techniques that should also be applicable in other regions. For example, quadrupole mass spectrometry is being used to study the distribution of CH_4 with depth in flooded peatlands in the UK, in relation to the distribution of C sources, other nutrients, redox potential, pH, and temperature (Figure 6). Also, eddy accumulation, flux gradient and eddy correlation methods using a tunable diode laser, and aircraft-based methods are being used to obtain fluxes at different scales (Gallagher *et al.*, 1993; D. Fowler, private communication).

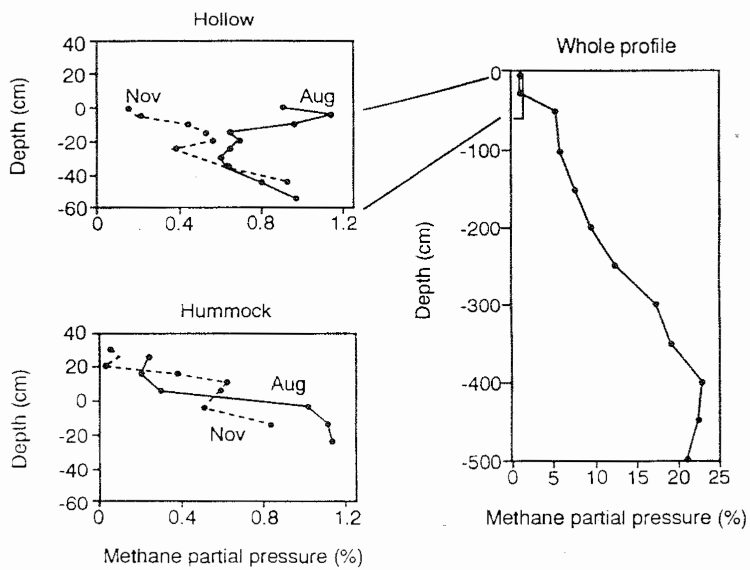

Figure 6. Measurements of methane partial pressure in waterlogged peat by quadrupole mass spectrometry (Clymo *et al.*, 1993).

In contrast with emissions from natural wetlands, CH_4 from midlatitude landfills may comprise a much higher proportion of the contemporary global landfill flux, estimated to be circa 30 Tg yr^{-1} (Houghton *et al.*, 1992; Bogner, 1992). This estimate is a reduction from previous estimates that ranged to 70 Tg yr^{-1} (Bingemer and Crutzen, 1987) because of the recognition of the very high oxidation rates occurring in landfill cover soils. Likewise, the midlatitudes are likely the most important global source of CH_4 from animal and poultry manures, which may produce an additional 25 Tg yr^{-1} CH_4 (Casada and Safley, 1990).

As for biogenic emissions of CO_2 from soils, the contemporary CH_4 flux from midlatitude systems and its contribution to current atmospheric loading rates should be balanced against historical fluxes. Region-wide, it is likely that biogenic methane emissions from soils today are less than emissions occurring prior to widespread wetland drainage in North America and elsewhere, and prior to the disappearance of large ruminant populations in midlatitude savanna regions. These potentials are, however, poorly estimated (Schimel *et al.*, 1992) and deserve much further evaluation.

The Soil as a Sink for Methane

The soil sink for CH_4, resulting from the microbial oxidation of CH_4 to CO_2, is estimated to be 30±15 Tg yr^{-1} (Houghton *et al.*, 1990; Whalen and Reeburgh, 1990). Although this value is only about 6% of total emissions, it is very similar to the estimated annual increase in the atmosphere (32±5 Tg; Houghton *et al.*, 1992).

Inputs of nitrogen have been shown to reduce this soil sink very substantially. The effect has been demonstrated in deciduous and coniferous forests (Steudler *et al.*, 1989; Figure 7) and in short-grass prairie (Mosier *et al.*, 1991) in the U.S. Results from a cropping systems experiment in the U.S. Midwest (Robertson *et al.*, 1993) suggest that the attenuation of CH_4 oxidation in cropping systems may be more related to soil disturbance and changes in organic matter dynamics than to changes in nitrogen availability (N fertilization) per se. Hansen *et al.* (1993) have further shown that compaction by farm equipment can also reduce CH_4 uptake.

The deposition of N from the atmosphere onto forests and natural grasslands and the use of fertilizers in agricultural ecosystems have increased greatly in the midlatitudes over the last few decades (Melillo *et al.*, 1989). If later results bear out early trends with respect to nitrogen attenuation of CH_4 oxidation, N deposition and land use changes may be reducing the CH_4 sink strength by more than 50% in areas already affected.

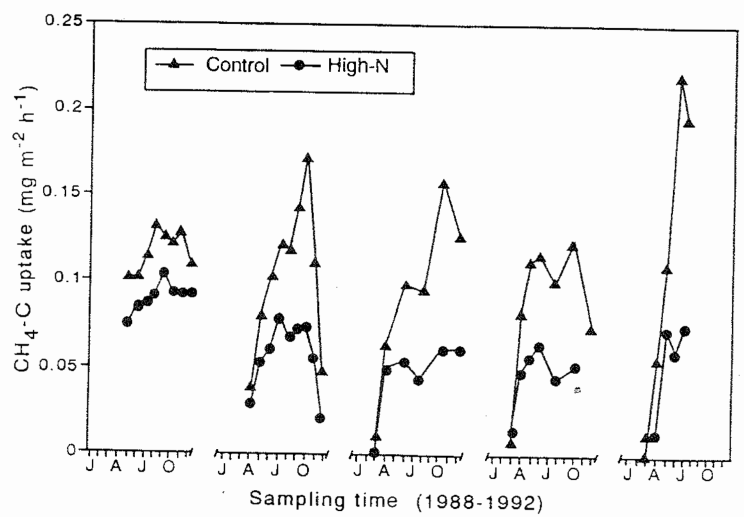

Figure 7. Effect of N fertilization on methane uptake in a red pine stand. Harvard Forest, MA, U.S. (J. Melillo, private communication).

N_2O FLUXES

Globally, soils are the major source of atmospheric N_2O (Houghton *et al.*, 1992; Robertson, 1993). Of the total emission rate of 8 to 18 Tg N yr^{-1}, 2.5 to 5.7 Tg are attributed to tropical soils, whereas the uncertainty ranges are much wider for temperate soils. Of particular note is the uncertainty associated with estimates of midlatitude agricultural activities.

Effects of N Inputs to Agricultural Land

In spite of the IPCC assessment, there is much indirect evidence that the flux of N_2O from agricultural land—much of which is in the midlatitudes—is in fact a major contributor to global emissions (Robertson, 1993). The emissions increase with increasing inputs of nitrogen in the form of mineral fertilizers, organic manures, and also N fixed biologically by legumes and other symbiotic systems.

The relationship between N inputs and emissions is not clearcut, however. In temperate grassland there is a variable but generally large response to N fertilizer, with short-term increases in flux ranging from factors of 2 to more than 10 when fertilizer is applied (Ryden, 1981; Bouwman, 1990). The chemical form of the N, as well as the amount applied, is important (Eichner, 1990; Figure 8). On the other hand, based on reported emissions from arable land (studies carried out predominantly in the northern midlatitudes), Bouwman (1990) derived the relationship

$$N_{em} = 1.879 + 0.0042N_f \quad (1)$$

where N_{em} is the N_2O emission and N_f is the fertilizer N applied, both in kg $ha^{-1}yr^{-1}$. Bouwman's regression suggests that an unfertilized arable field emits on average about 1.9 kg $ha^{-1}yr^{-1}$, and this only increases by about 0.4 kg ha^{-1} when 100 kg N ha^{-1} is applied.

Nitrogen fertilization should probably be regarded as a process that gives a major stimulus to the N dynamics of the system and consequently promotes N_2O production over a prolonged period, rather than simply as the addition of a transient source of mineral N which can serve directly as a substrate for N_2O production. Thus the high fluxes from unfertilized arable land may be due to the increased turnover of nutrients resulting from high net primary production (and attendant residue return) and soil disturbance by cultivation. Such factors increase the rates of N mineralization, nitrification, and denitrification compared with undisturbed environments, thus promoting N_2O production. The variability of the capacity of unfertilized grassland to emit N_2O may reflect management practices that influence these turnover processes.

The total area of cultivated land in the midlatitudes is of the order of 10^9 ha, giving rise, on the basis of Equation 1, to a *background* emission of circa 1.9 Tg N_2O–N yr^{-1}. Eichner (1990) suggests a smaller value based on a literature analysis of fertilizer-response experiments. The effects of fertilizer N inputs on global N_2O emission may, however, be far greater than either estimate. Robertson (1993), for example, points out that very little of the 80 Tg N added as fertilizer to croplands stays in these soil-plant systems very long; in general about 50% is either leached or denitrified within 6 months of application, 25% taken off as yield to end up eventually as sewage, and most of the 25% remaining in crop residue decomposes and is similarly lost over the next several years (soil organic matter rarely builds up under cultivation). Most of this 80 Tg N, then, will be likely eventually to be denitrified in order to close the global N cycle. Only a small portion of this fixed N will need to be denitrified to N_2O rather than to N_2 to have a substantial impact on the global N_2O budget.

The contribution of legumes to N_2O emissions has been little studied compared with the research devoted to conventionally fertilized systems. However, such evidence as there is suggests strongly that this source may be very significant. Duxbury *et al.* (1982) found annual emissions from alfalfa in New England about two and a half times those from a grass site. Similar observations have been made on leguminous pastures in Australia (I. Galbally, private communication); and recent work in Edinburgh showed a comparable increase for a clover/grass sward over a grass-only sward, with emissions an order of magnitude higher when the swards were plowed (Figure 9).

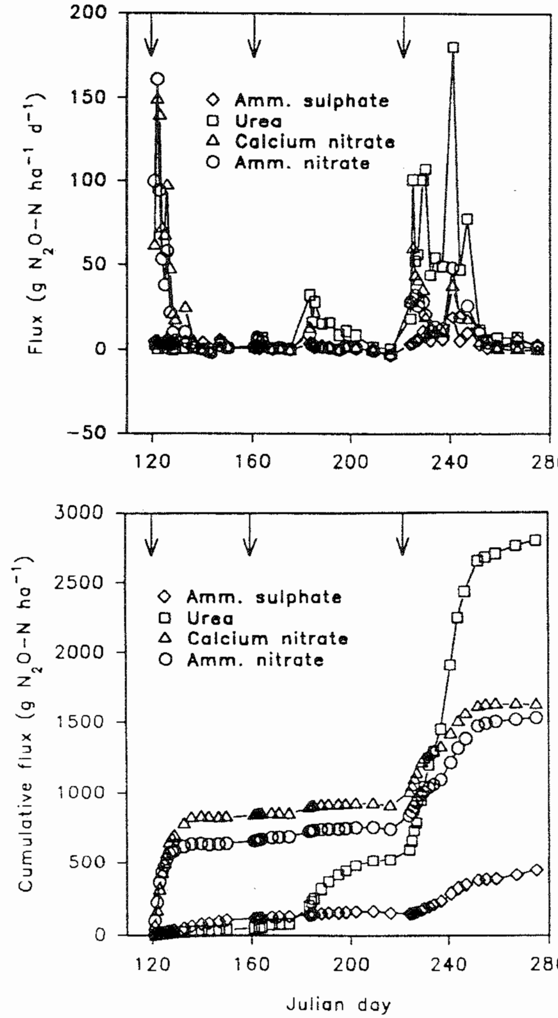

Figure 8. Temporal variations in fluxes (top), and cumulative fluxes (bottom) of N_2O from a grassland site near Edinburgh, fertilized with 4 forms of N fertilizer, 1992. Arrows indicate times of fertilizer applications (120 kg N ha^{-1} on each occasion) (H. Clayton, I.P. McTaggart and K.A. Smith, private communication).

The agricultural systems of temperate South America and Australasia are predominantly based on leguminous inputs of N. There is also a substantial movement towards greater use of leguminous green manures in the other continents, and the total global input of N from biological fixation is estimated to be roughly equal to that from industrially fixed N, circa 80 Tg N yr^{-1} (Isermann, 1993). The contributions from these sources to total global emissions of N_2O require further investigation.

A major challenge to future experimental programs is to obtain representative flux data for agricultural ecosystems not merely over whole annual cycles, but also over the lifetime of characteristic cropping rotations. This is necessary if we are to establish the total fluxes arising from the system as a whole and also to distinguish between the contributions directly associated with fresh N inputs and those from other processes. Such information is required if we are to have satisfactory predictive models to estimate the consequences of changes in land use practices on total emissions.

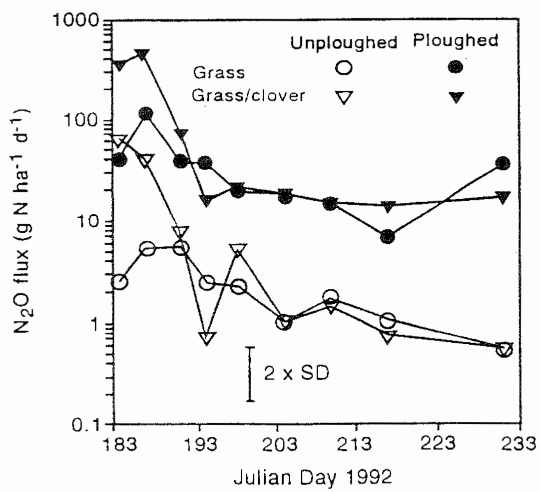

Figure 9. Effect of presence of clover and of plowing on emissions of N_2O from grassland (K.A. Smith, S. Bergos and M. Davies, private communication).

Effects of N Deposition

A major contributory factor to N_2O release in the midlatitudes is the deposition of N onto natural and near-natural ecosystems such as forests and moorlands. An acid beech forest soil receiving atmospheric N inputs of 40 to 60 kg ha^{-1} has been shown to emit 5.6 kg N_2O–N $ha^{-1}yr^{-1}$ (Brumme and Beese, 1992). This rate is of the same order as those recorded from the most intensively fertilized agricultural land, and in fact additional fertilization of the beech soil only increased emissions by 40% (Figure 10). In contrast, in environments receiving less N input, additional fertilization can cause a dramatic increase in emissions (Figure 11). A significant part of N deposition in Europe is in the form of NH_4^+, and its acidifying effects may be a contributory factor to emissions (Nagele and Conrad, 1990); liming reduces the emissions considerably (Brumme and Beese, 1992; Figure 10).

PRECURSORS OF TROPOSPHERIC OZONE: NO_x, CO, NMHC

Tropospheric ozone is predicted to increase with increasing emissions of nitrogen oxides (NO_x), and with increasing emissions of carbon monoxide (CO), CH_4, and non-methane hydrocarbons (NMHC) when the atmospheric abundance of NO_x is greater than 20 to 30 pptv. These species also have complex effects on atmospheric OH, thus affecting the lifetime of other greenhouse gases (Houghton *et al.*, 1992).

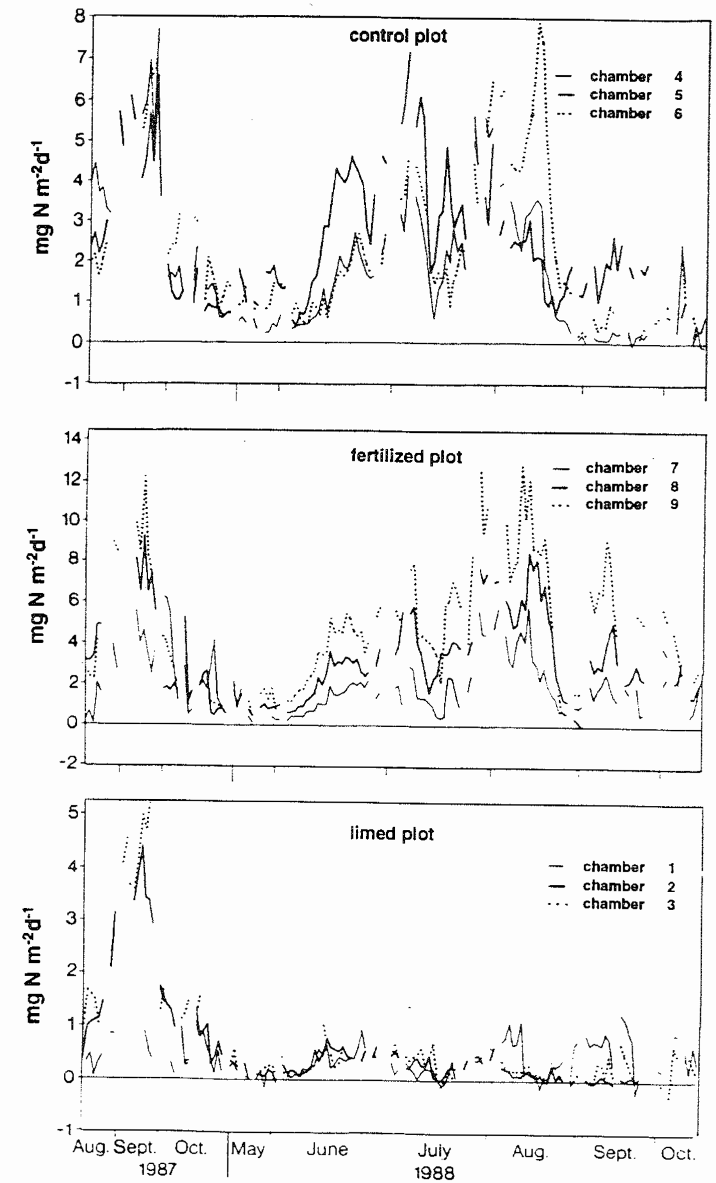

Figure 10. Daily mean values of N_2O emission from control, fertilized, and limed plots (data from three individual automated chambers per plot) (Brumme and Beese, 1992).

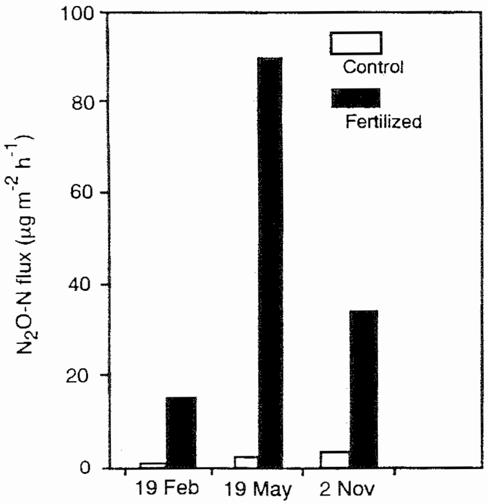

Figure 11. Effect of N fertilization on N_2O emission from soil in a slash pine plantation, Florida, U.S. in 1991 (Castro and coworkers, private communication).

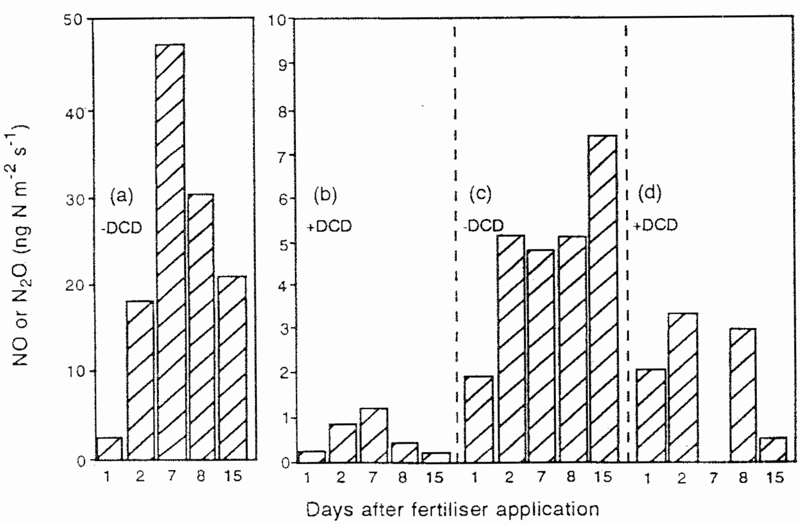

Figure 12. Emissions of NO (a, b) and N_2O (c, d) from sandy loam soil, following addition of ammonium sulfate with (a, c) and without (b, d) dicyandiamide (DCD). Note different y-axis (Skiba *et al.*, 1993).

Nitrogen oxides, CO and NMHC all have significant natural and anthropogenic sources, but budgets remain uncertain. NO is mainly emitted from industrial sources in the developed countries, but it is also a product of nitrification and denitrification. Its emissions by the former process can exceed those from N_2O by a substantial factor (Skiba *et al.*, 1993). Increasing use of ammonium-forming fertilizers and acid deposition is likely to increase NO release, but it has been shown that inhibition of nitrification is highly effective in reducing the emission (Figure 12).

The soil appears to be more important as a sink than as a source for CO, but too few data exist to produce a reliable estimate of the size of this sink. It may well be that the sink will be affected significantly by factors such as N inputs, as occurs for CH_4. More work is needed in this area.

NEW TECHNOLOGY FOR FLUX MEASUREMENT

Several encouraging developments have occurred recently in methods for trace gas flux measurement. We appear now to be on the verge of being able to obtain high-quality large-area flux measurements more or less routinely. If this prospect becomes reality, it should have a major impact on the quality of information being put into climate models.

Micrometeorological Methods

The flux gradient and the eddy correlation micrometeorological methods (Fowler and Duyzer, 1989) and the more recent eddy accumulation (or "conditional sampling") method (Businger and Oncley, 1990) permit flux measurements on the ha-km^2 scale, and thus overcome local spatial variability which is a problem with chamber methods. The conditional sampling method is relatively new and not widely tested, but seems very likely to find widespread application in the medium term. It depends on the determination of the difference in trace gas concentrations in sample bags used to collect air during updraughts and downdraughts. The sampling is controlled by high-speed valves linked to a 3-dimensional sonic anemometer. These need to be located at a height of at least 3 m above the ground. This corresponds to a fetch of 300 to 600 m, and the method integrates over areas of the order of 10 ha. For smaller areas the gradient method, using lower sampling heights, is more suitable (K. Hargreaves, private communication).

Tunable diode laser absorption spectroscopy (TDLAS) techniques now have sufficient sensitivity to permit the measurement of very small fluxes of CH_4, N_2O, and CO. The technique appears extremely promising, but it is recognized that its development for general use by non-specialists will occur only in the medium term, and that in the immediate future the technique will find application mostly in short-term intensive studies. The same is true for FTIR absorption spectroscopy (Galle *et al.*, 1993). These methods have been applied to the measurement of N_2O by the flux gradient method (Wienhold *et al.*, 1993; Galle *et al.*, 1993). Gas chromatography has also been used in this way (Hutchinson and Mosier, 1979), but the sensitivity is much lower than with the spectroscopic methods. It can be increased, however, by multiple replicate analysis of bulk gas samples, and thus used over a wider range of conditions (Arah *et al.*, 1993a; Hargreaves *et al.*, 1993). Figure 13 shows an intercomparison of these three analytical techniques applied to determination of N_2O fluxes by the gradient method, indicating good agreement between them.

Techniques which can directly measure concentration differences, such as dual path optical methods or the optical subtraction FTIR method under development in Sweden (L. Klemedtsson, private communication), and those such as TDL which can be adapted to make sequential analyses from large bag samples, are now being applied to the conditional sampling technique. Their use extends the range of fluxes measurable down to levels far lower than those attainable by GC.

Another integrating micrometeorological method which has only low technological requirements for gas measurement is that which exploits concentration buildups under nighttime inversions. For large relatively homogeneous source regions the method may augment enclosure studies. However, for both this and for micrometeorological measurements in general, it must not be forgotten that measurements of the depth of the boundary layer have to be made, requiring specialized equipment. Here, as in some of the other methods, there is a strong case for collaboration between scientists from several disciplines to provide the required expertise.

Recognizing the resource requirements for such flux measurement experiments, it has been recommended (IGAC, 1992) that certain of the TRAGEX network sites should be designated as lead sites where special resources could be deployed, and which could also serve as sites for intercalibration of instruments and methods.

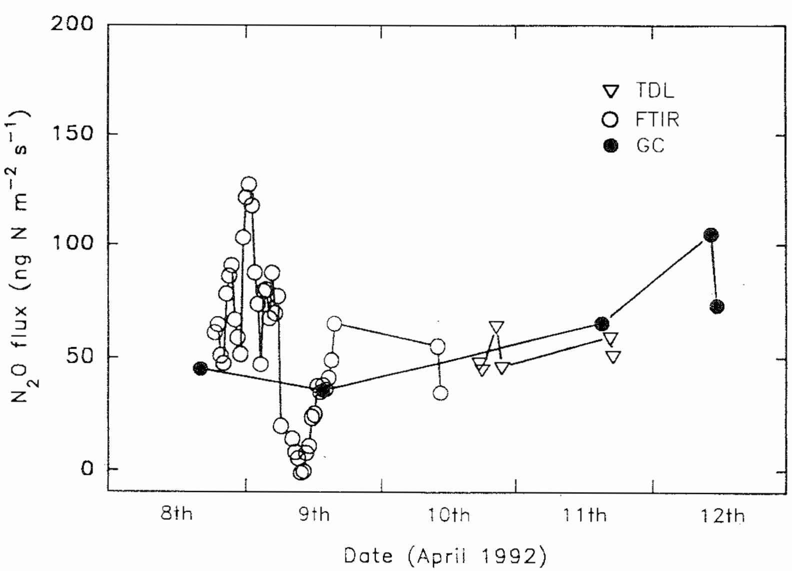

Figure 13. Measurements of N_2O flux from fertilized grassland, Stirling, Scotland, 1992, by flux gradient method, using three different analytical systems (Hargreaves *et al.*, 1993). TDL: tunable diode laser absorption spectrometry; FTIR: Fourier-transform infrared spectrometry; GC: automated gas-chromatography.

Chamber Methods

Large chambers. Where land use practices produce a small-scale mosaic of different crops, woodland and/or inputs, micrometeorological methods are difficult to apply, because of the resulting small fetch. This constraint applies especially to agricultural regions of Europe and Asia. For these regions the continued development of chamber methods is essential. One such development is the use of very large chambers together with long-path infrared absorption spectroscopy (Smith *et al.*, 1993). Such systems have two advantages:

they average emissions over areas 2 to 3 orders of magnitude larger than conventional small chambers, while still allowing flux measurements to be made at sites where the effect of different treatments on fluxes is being investigated, where micrometeorological methods could not be used.

Automatic (small) chambers. The recognition that longer time series of data are required to improve annual flux estimates has led to the development of several systems involving automation of chamber closure, gas sampling, and GC analysis. One such system (Brumme and Beese, 1992) is deployed in the measurement of CO_2 and N_2O fluxes in a temperate forest in Germany, and is being extended to include CH_4. Robertson (private communication) has a comparable system in a series of four cropping systems in Michigan, U.S. In Edinburgh, Scotland, automated chambers have been fitted to large soil monoliths to facilitate manipulation studies (Smith and Thomson, 1993) and a portable battery-powered system of automated field chambers and associated gas samplers has been developed (Smith and Scott, private communication) to take samples over a period at a remote site for return to the laboratory for automated analysis.

These developments reduce the labor requirement for field sampling and analysis enormously, and make much more feasible both long-term measurements and intensive short-term investigations required for process-related studies.

MODELING AT DIFFERENT SCALES

Carbon Dioxide

The modeling of CO_2 emission and uptake processes is more advanced than for the trace gases CH_4 and N_2O. For the assessment of CO_2 fluxes in the midlatitudes, modeling has a larger role to play than direct measurement. Some of the models for predicting changes in NPP and carbon storage in soil and biomass are referred to above in the section on CO_2 fluxes, and will not be considered further here.

Process Models for CH_4 and N_2O

The modeling of N_2O emissions is relatively well advanced at the process level, but scaling to field, landscape and regional levels is not so well developed. Firestone and Davidson (1989) have neatly represented N_2O formation/emission by a "hole-in-a-pipe" model, and Groffman *et al.* (1988) and Robertson (1989) have devised a conceptual model of the controls affecting nitrification and denitrification at different geographic scales. These models improve our capacity to evaluate the relative importance of controlling environmental variables for specific environments. They also serve to identify the key variables which need to be measured or recorded at experimental sites, in association with flux measurements, in order to make quantitative predictive flux models.

Parton *et al.* (1988) have developed a model of N_2O fluxes from nitrification, in which they have simply related fluxes to the rates of mineralization and nitrification occurring in soils of different texture in the semi-arid conditions of Colorado, U.S. The simulations were generally satisfactory in describing the fluctuations in soil mineral N, and accounted for some but not all of the fluctuations in N_2O emissions. This approach could be used to predict fluxes from other, generally well aerated soils in other regions, where nitrification is likely to be the dominant mechanism of production.

In wetter environments, such as northwest Europe and eastern and midwestern U.S., there is much evidence that denitrification is the dominant mechanism for N_2O production. Models of denitrification in soil have been developed that have been at least partly validated by experiment (Smith, 1980; Parkin and Tiedje, 1984; Arah and Smith, 1989; Smith and Arah, 1992; Arah *et al.*, 1993b). The fraction of the denitrified N emitted from the soil as N_2O varies dramatically with environmental factors, and modeling N_2O flux is thus even more of a problem than modeling denitrification per se.

The biochemical and physicochemical processes involved in CH_4 production, transport and oxidation in flooded soils are now being modeled. These processes are essentially similar in both natural wetlands and in rice paddies (cf., Neue and Sass, this volume), and models whose development may have arisen only in the context of one of these environments should be useful in both. The growing interaction between scientists involved in natural wetlands and rice research programs can be expected to promote this cross-fertilization. It is likely that we will see the outcome of present efforts within the next few years.

Scaling Up To Larger Areas

The capacity to extrapolate from process-based models applicable only to a tiny area or volume to landscape or catchment-sized areas and beyond is a major priority. Work in this area is still in its early stages, but it seems likely that the momentum will increase, with at least part of the stimulus coming from the TRAGEX program.

Scaling to larger areas requires a linkage between the controlling variables included in the process models and information contained in spatial databases, such as the Global Information System (GIS) database. Such linkages are most advanced for CO_2 fluxes. For example, Peterjohn *et al.* (1993) have shown that the flux from temperate forest soils is a function of soil temperature, which in turn is linearly related to air temperature. The latter parameter, but not the former, has already been incorporated into GIS, and Kicklighter *et al.* (1993) have modeled regional CO_2 fluxes using this information.

For the more complex relationships driving N_2O emissions, comparable "surrogate" parameters will need to be found. It should be possible to use information on soil texture, structure and water content, and air temperature, as an alternative to direct estimates of denitrification or nitrification. Similar considerations apply to CH_4 emission and uptake. It may thus be possible to score soils subjectively into a small number of classes in relation to each key factor, using soil and vegetation maps together with climatic data: such scoring, combined with an emission/uptake factor for each score class, will permit the aggregation of fluxes over larger areas with more confidence than can be achieved by direct extrapolation from point measurements.

THE IGAC-TRAGEX ACTIVITY

The need for a coordinated program to provide the necessary information is evident. Even where trace gas fluxes have been most intensively studied, data are rarely as comprehensive as modelers would desire. For example, in only a few instances have the estimates from a particular site been based on frequent measurements made over an entire year. As indicated above, there are whole regions for which there is little or no information, even from studies of short duration. Also, most studies have focused on small areas (e.g., individual fields or forest plots). As a result, many estimates of trace-gas fluxes in the temperate region are not very reliable at the regional scale.

The TRAGEX Activity (IGAC, 1992) is intended to remedy this situation; its purpose is:

- to document the contemporary fluxes of CO_2, CH_4, N_2O and the tropospheric ozone precursors, and
- to determine the factors controlling these fluxes and improve our ability to predict future fluxes.

Achievement of these objectives will involve three types of research activities:

- measurement of trace gas fluxes at an international network of representative sites;
- manipulation of what are perceived to be the controlling variables, to determine their impact on fluxes; and
- modeling of the processes responsible for trace gas fluxes.

Sampling networks are being planned to include the main temperate zone ecosystems in each continent. The networks are intended to produce the data necessary to quantify critical variables controlling fluxes of trace gases across each region. These networks must necessarily include the main ecosystem types, both natural and managed, within each geographical component of the temperate zone. Most importantly, the networks must embrace areas where current fluxes are not well known.

In this last category are several large midlatitude agricultural and forested areas of special importance, for which little data are available. These include areas of intensive agriculture in eastern Europe, the former USSR, and China, and the large region of temperate South America where there is a less intensive agricultural system based to an unusual degree on leguminous nitrogen inputs. Two major wetland areas that have been little studied, the Pripet marshes of Eastern Poland, Ukraine, and Belarus and the peatlands of western Siberia, are an additional priority.

The network also needs to include sites representative of the large areas of the midlatitudes that have been affected by atmospheric deposition of nitrogen, sulfur and acidity. The consequences for trace-gas fluxes of loading ecosystems with nutrients are just beginning to be understood, but more quantitative information is a priority within this program.

Human-driven biogenic fluxes that are unique to the temperate zone, such as those from landfills and agricultural wastes in anaerobic lagoons, also need to be quantified. The quantification of emissions from ruminants is not a specific remit of the TRAGEX Activity, but in view of the importance of this source it is an issue which needs to be addressed somewhere within the overall IGBP-IGAC Project.

Main Site Networks

The broad outline of a plan for a midlatitudes program was drawn up at the initial planning workshop of the Activity, held in Boulder, Colorado, U.S. in the autumn of 1991. The basic design calls for sites representing the major vegetation/land use types of the midlatitude zone: forests, rangeland, and agricultural land (Table 2). In addition, there is a need for data from a small number of specific temperate wetland types that are poorly characterized (Melillo *et al.*, 1992).

The first detailed plan for a network in one region, building on the outline produced at Boulder, was that for the continental U.S. (excluding Alaska). This was devised at a workshop at Pingree Park, Colorado in October 1992 (Ojima *et al.*, 1993). The initial site

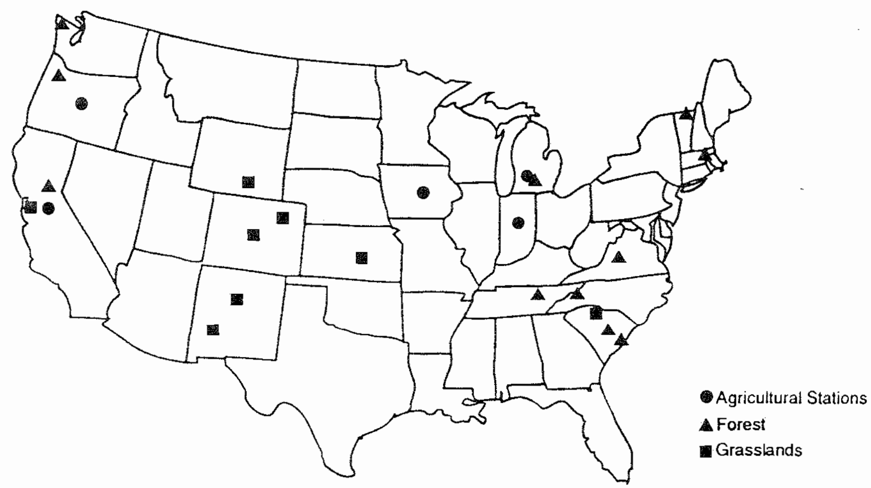

Figure 14. Locations of midlatitude sites for proposed U.S. trace-gas sampling network (Ojima *et al.*, 1993).

network planned for the U.S. is shown in Figure 14. Since then, a similar workshop to plan a network for Europe has been held in Munich, Germany in April 1993. Future meetings of the TRAGEX committee will have to address the problems associated with developing plans for other regions. It must be expected that outside Europe and North America the relative lack of scientific resources, infrastructure problems, and political and economic climates are all likely to make the task of developing and implementing a comprehensive plan a good deal more difficult.

Data Requirements

The Pingree Park meeting established protocols specifying the data that should be collected at each site (Ojima *et al.*, 1993). This had the primary objective of ensuring that the data obtained would all be compatible for modeling purposes, especially the extrapolation to larger regions. This approach is likely to set a precedent for the procedures to be adopted in other regions.

Many of the data collection activities within the monitoring program will need to be long-term, encompassing a wide range of temporal variation that will be helpful for understanding how fluxes will respond to climate change. Data acquired from manipulation experiments should complement those obtained from monitoring, and both will be used for the development of trace gas flux models incorporating the diverse factors that control these exchanges. These models will form the nucleus of a larger modeling activity capable of predicting trace gas exchanges at ecosystem scales. The linking of ecosystem models to atmospheric chemical transport models, and ultimately to general circulation models, will permit the necessary elucidation of trace gas fluxes at regional and global scales.

Table 2. Proposed locations of sampling site networks for midlatitude trace gas fluxes (IGAC, 1992).

Continent	Ecosystem/ Land Use Type	Key Ecosystems for Study	Existing Site Network
North America	Forest	Temperate rainforests (northwest coast)	LTER
		Mixed hardwoods (northwest, midwest)	LTER
		Southern coniferous	
		Montane coniferous	
	Rangeland	Desert	LTER
		Semi-arid shrubland	LTER
		Shortgrass steppe	LTER
		Tallgrass prairie	LTER
	Agricultural	Western irrigated	
		Western dryland	
		Corn belt	LTER
		Southeast	
		Intensive animal-based	LTER
Europe/ North Africa	Forest	Scots pine	NITREX
		Spruce	NITREX, TERN
		Beech	NITREX, TERN
	Rangeland	Desert (North Africa)	
		Semi-arid shrubland (Mediterranean)	
		Shortgrass steppe	
		Tallgrass prairie	
	Agricultural	Intensive pasture	TERN, TIGER
		Conventional arable	TERN, TIGER
		Intensive irrigated (horticultural)	
		Semi-arid	
Asia	Forest	Mixed hardwood	
		Coniferous	CERN
	Rangeland	Desert	
		Semi-arid shrubland	CERN
		Shortgrass steppe	CERN
		Tallgrass prairie	
	Agricultural	Irrigated	CERN
South America/ Australasia	Forest	Southern beech (*Nothofagus*)	
	Rangeland	Desert	
		Semi-arid shrubland	
		Shortgrass steppe	
		Tallgrass prairie	
	Agricultural	Temperate arable (including legume-based)	
		Intensive pasture (including legume-based)	

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