6   Fluxes of Nitrous Oxide and Other Nitrogen Trace Gases from Intensively Managed Landscapes: A Global Perspective

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Concentrations of N_2O in the global atmosphere have been rising over the last 20 yr at about 0.8 parts per billion volumetric (ppb_v) or 0.25% yr^{-1} to the 310 ppb_v present in today's atmosphere (Fig. 6–1; Elkins and Rossen, 1989). Ice core data that indicate concentrations of around 280 ppb_v in the preindustrial atmosphere (e.g., Pearman et al., 1986) suggest that this rise reflects a long-term trend (Fig. 6–2; IPCC, 1990). In fact, this 0.8 ppb_v increase represents a 3.5 Tg addition of N_2O–N to the global atmosphere each year, and if one assumes that the present photodissociation rate of N_2O in the stratosphere (11 Tg yr^{-1} N) is typical of preindustrial steady-state loading rates, then the preindustrial loading rate of N_2O–N was 11 Tg yr^{-1}—suggesting that we are today adding another 40% (3.5 Tg N) of N_2O to the atmosphere each year than was the case 100 yr ago (Cicerone, 1987; Robertson et al., 1989).

This loading rate is significant primarily for two reasons. First, N_2O is one of the major greenhouse gases, accounting for 6 to 8% of the present greenhouse forcing rate ascribed to anthropically derived gases (CO_2, CFC's, CH_4, and N_2O; Hansen et al., 1990; IPCC, 1990). On a molar basis N_2O is about 250 times more potent than CO_2 as an absorber of infrared radiation—in part due to its molecular structure but largely due to the fact that it absorbs in a portion of the infrared transmission window that in our present atmosphere is relatively clean (Duxbury et al., 1993). Second, N_2O is the major natural regulator of stratospheric O_3, which effectively controls the earth's ultraviolet-B (UV_B) radiation balance. Through a series of reactions elucidated over the last 20 yr, the oxidation of N_2O to NO via reaction with photolytically produced atomic oxygen $O(^1D)$ in the upper stratosphere (>25 km) produces NO that in turn reacts with O_3 to form
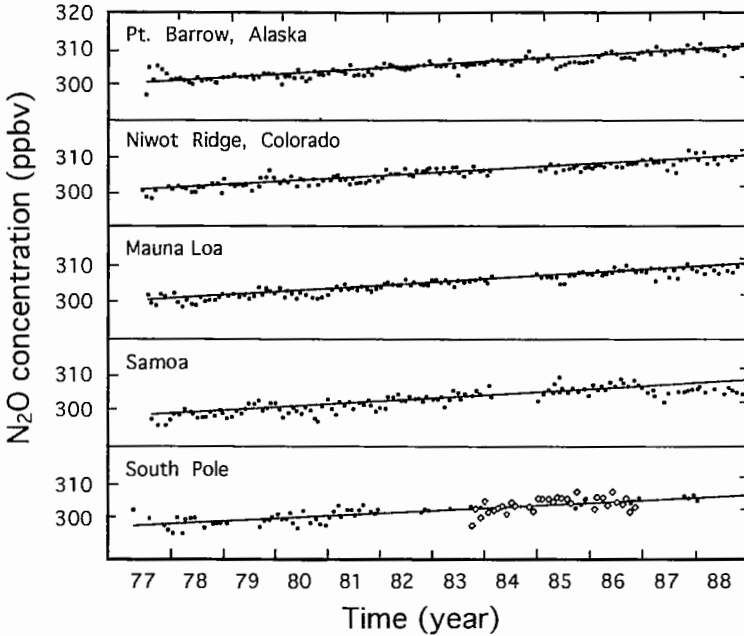
Fig. 6–1. Atmospheric measurements of N_2O from the NOAA–Geophysical Monitoring for Climate Change network (Elkins & Rossen, 1989; redrawn from IPCC, 1990).

NO_2 and O_2 (Hahn & Crutzen, 1981). Under steady-state (preindustrial) conditions, this set of reactions balances the formation of O_3 in the stratosphere. Because these reactions represent the only major atmospheric sink for N_2O, N_2O has an atmospheric lifetime of about 150 yr. This contrasts with a lifetime for CH_4 of about 10 yr, implying that the effects of present N_2O loading rates will be especially long-lasting.
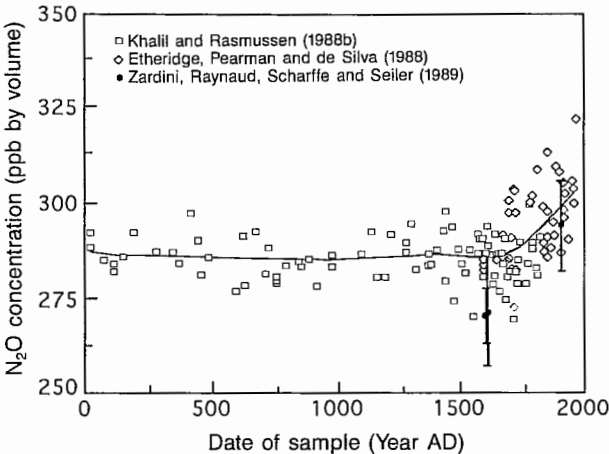

Fig. 6–2. Nitrous oxide concentration in ice core samples (redrawn from IPCC, 1990).

NO_x, on the other hand, is not an effective greenhouse gas at atmospheric concentrations, nor is biogenically produced NO_x an important player in stratospheric gas reactions. Rather, NO_x (primarily NO and NO_2) is important for its role in the chemistry of the troposphere, where it regulates the photochemical production of tropospheric O_3 and the abundance of hydroxl [OH] radicals, primary oxidants for a number of tropospheric trace gases (Jacob & Bakwin, 1990). Under daylight conditions the transformation of tropospheric O_3 to NO_2 and O_2 via reaction with NO is balanced in a 1:1 manner by the photolysis of NO_2 back to NO and O_3; at night there is no reaction of NO_2 back to NO and O_3 so NO_2 may accumulate as O_3 is depleted. Likewise, the reaction of NO with HO_2 in the troposphere is a major regulator of OH concentrations, the primary oxidant for the photochemical destruction of atmospheric CH_4 and other gases. Further, the oxidation of NO_x to HNO_3 in the atmosphere is a major contributor to acid precipitation (NAS, 1986), and may be contributing significantly to low-level N fertilization of large portions of the terrestrial biosphere (Melillo et al., 1989).

Neither the global N_2O budget nor the global NO_x budget is presently balanced with precision—in fact our knowledge of the global N_2O cycle is especially off-track with almost 50% (6.5 Tg N) of the sources needed to balance the known sinks (14.1 Tg N) unidentified (see below). The situation for NO_x is not so bad perhaps only because sinks for NO_x are not as easily quantified as are sinks for N_2O.

In the pages that follow is a discussion of our present state of knowledge with respect to the global budgets of these important trace gases. Of particular note are the roles of agricultural and other intensively managed landscapes as contributors to global fluxes; our knowledge of fluxes in these environments is particularly weak in light of their likely contribution to increased global emission rates.

THE GLOBAL NITROUS OXIDE BALANCE

Of all of the radiatively important trace gases (CO_2, CH_4, CFC, N_2O), none is more poorly understood with respect to sources than N_2O. Unknown sources of N_2O account for almost twice the current global atmospheric loading rate of 3.5 Tg N_2O-N yr^{-1}. Less than 5 yr ago this was not thought to be the case, i.e. the global N_2O budget appeared to be in approximate balance (McElroy & Wofsy, 1986), with 14.1 ($\pm$3.5) Tg yr^{-1} N of sinks about balanced by 15.3 ($\pm$6.7) Tg yr^{-1} of sources (Table 6-1). What has happened since then to shrink our estimates of source–strengths to only 7.5 Tg yr^{-1}? Primarily refinements of existing estimates—and especially refinements of flux estimates from industrial- and managed-landscape sources.

Combustion

Until 1989, global N_2O budgets considered industrial combustion a substantial net source of N_2O to the atmosphere, with an estimated annual con-

Table 6-1. The global N_2O cycle: major sinks and sources (except as noted, references in text).

Sinks/sources	1986†	1992
	—— Tg yr^{-1} N ——	
Sinks (stratosphere)		
Photolysis and 0('D) reactions	10.6	
Atmospheric accumulation (0.25% yr^{-1})	3.5	
Total sinks	14.1	
Sources		
Open ocean	2.0	2.0
Combustion		
Industrial	4.0	0.0
Biomass burning	0.7	0.2 (0.1–0.3)
Manufacturing (nylon)		0.4
Agriculture		
Direct fertilizer emissions	0.8	0.3
Emissions from groundwater		0.2
Indirect via NO_3^- loss		?
Native communities		
Temperate forests, grasslands	0.2–0.6	0.4 (02.–0.6)
Tropical forests	7.4	
Humid forests		2.4
Pasture conversion		0.7
Seasonally dry forests		1.0
Total sources	15.3	7.6
Balance	+1.2	−6.5

† From McElroy and Wofsy (1986).

tribution of approximately 4 Tg N via coal and oil burning. With the discovery (Muzio & Kramlich, 1988; Muzio et al., 1989) that this source estimate is largely a sampling artifact caused by gas reactions in sample flasks prior to N_2O analysis, the strength of this source has been revised downwards almost three orders of magnitude. The present estimate of <0.01 Tg yr^{-1} N_2O–N from industrial combustion reflects this new knowledge (Table 6-1).

Biomass burning—primarily during annual land-clearing operations in tropical savannah and rainforest regions—represents a different combustion source that appears to make a minor though significant contribution to the global N_2O flux. Crutzen and Andreae (1990) estimate a flux of 0.1 to 0.3 Tg yr^{-1} N from tropical sources based on our current knowledge of land clearing rates and N_2O emissions from relatively low-temperature fires. Almost all of this land clearing—certainly most that occurs in extensive savannah regions burned annually or biennially—is associated with agricultural production. In fact, these fires are a very significant source of many other important gases in addition to N_2O; e.g., Crutzen and Andreae (1990) have coined the phrase pyrodenitrification to describe the heretofore unrecognized role of biomass fires in N_2 production.

Row-Crop Agriculture

Row-crop agriculture is assumed in most global assessments of N_2O fluxes to contribute most heavily to the global flux primarily through effects

Table 6-2. Relative effect of different sources of fertilizer on N_2O emissions from fertilized agricultural systems. Control refers to unfertilized crop (from Eichner, 1990).

Treatment (fertilizer type)	Ratio of treatment flux to control flux	Fertilizer-derived N_2O as % of total N_2O emission
Anhydrous ammonia	6.1	82
Ammonium nitrate	2.5	57
Ammonium (Cl, SO$_4$)	1.4	29
Urea	1.5	30
Nitrate (Ca, K, Na)	1.1	12

of fertilizer inputs. In the mid-1980s approximately 70 Tg of N fertilizer were applied to crops worldwide (FAO, 1985); analysis of the proportion of fertilizer N emitted as N_2O following application (Eichner, 1990) suggests that, on average, about 0.5% (0.35 Tg) of fertilizer inputs is lost from this land use. This estimate is based on evidence that the N_2O flux from fertilized agriculture is most intense following fertilization (references in Eichner, 1990), and is weighted by fertilizer type to account for the fact that N_2O emission rates from fertilizers appear to be strongly related to fertilizer source. For example, direct N_2O fluxes from fields treated with anhydrous ammonia appear to be some six times higher than from fields treated with equivalent amounts of NO_3^- salt (Table 6-2, Fig. 6-3).

Direct fluxes of N_2O from agricultural sources may also result from the hydrologic transport of dissolved N_2O through groundwater to surface waters—where atmospheric equilibration will result in its release to the atmosphere—and to aquifers, where N_2O may be stored as a dissolved gas for long periods (Ronen et al., 1988). The IPCC (1990) global N_2O assess-
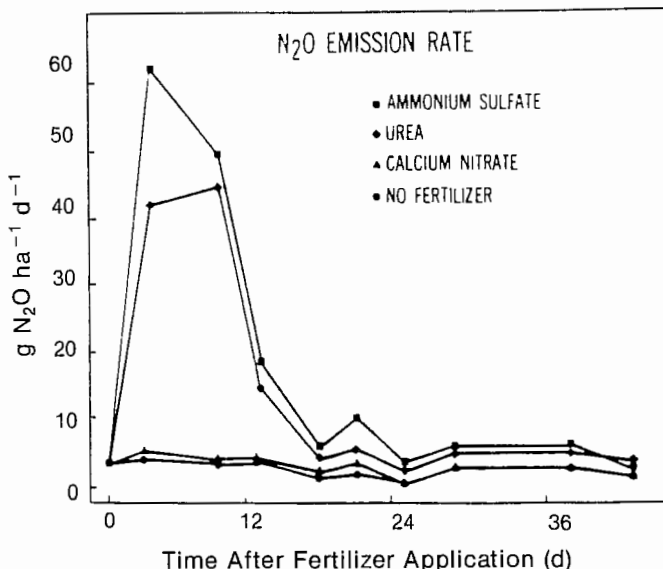

Fig. 6-3. Nitrous oxide emissions from an agricultural field treated with equivalent amounts but different types of N fertilizer (from Breitenbeck et al., 1980).

ment suggested that up to 1.1 Tg N may be added each year to the atmosphere from this source.

The effect of fertilized agriculture on N_2O loss from agricultural landscapes is probably not, however, limited solely to direct losses of N_2O to air and water. Of the more than 70 Tg of fertilizer N added to cropping systems annually, most is not taken off with the crop yield and very little of what remains is retained by the soil–plant system—if it were there would be little need to fertilize annually. Numerous fertilizer-^{15}N studies dating from the mid-1950s (e.g., Allison, 1955; Robertson et al., in review) have demonstrated that less than 50% of the fertilizer N applied to most crops is taken up by the crop. And if one considers that less than 50% of the N that *is* taken up is taken off with the grain yield, and then considers that the N that remains in the residue contributes little to long-term soil organic matter buildup in most tillage systems, then one is left with the conclusion that more than 75% of the N fertilizer added to most cropping systems (50% of that applied + 50% of that taken up by plants) is lost from the residue–soil system on a multiyear basis. At a global level this is a staggering amount of N—approximately 50 Tg yr^{-1}, with most if not all of this N eventually denitrified to close the global N cycle. Only a small portion of this N would need to be denitrified to N_2O rather than to N_2 to have a major impact on the global N_2O budget.

The effect of fertilized agriculture on the global N_2O balance is probably far greater, then, than that projected by current analyses. To evaluate this effect will, however, require analyses of N_2O fluxes at landscape rather than field scales—present studies at field scales, though relatively few, suggest that direct emissions of fertilizer-derived N_2O from agricultural fields are too short-lived to contribute much to annual global losses (e.g., Fig. 6–3). Rather, fertilizer N emissions are likely being expressed primarily at landscape or even regional-level "control points"—habitats such as soil–stream interfaces and near-coastal marine environments where conditions favor the transformation of solute N (NO_3^-, DON, NH_4^+) either directly or indirectly to N_2O and N_2. Too little N_2O flux work has been undertaken along hydrologic flow paths draining agricultural watersheds to adequately evaluate these sources today.

Forests and Grasslands

Estimates of N_2O fluxes from temperate forests and grasslands have changed little since McElroy and Wofsy's (1986) estimates of 0.2 to 0.6 Tg N yr^{-1}. Although it is possible that chronic low-level N fertilization of large regions due to elevated NO_3^- concentrations in rainwater (Melillo et al., 1989) may eventually result in enhanced potentials for N_2O loss from these regions, in the near term it is more likely that the low levels of added N will be immobilized in plant biomass and soil organic matter than be emitted as N_2O (Brumme & Beese, 1992).

Estimates of N_2O fluxes from tropical forests, on the other hand, were probably significantly overestimated in previous budgets. Matson and Vitou-

Table 6-3. Nitrous oxide fluxes from moist and wet lowland tropical forests (from Matson & Vitousek, 1990).

Soil type	Area	N_2O flux	Total flux
	10^6 km^2	kg ha^{-1} yr^{-1}	Tg yr^{-1}
High fertility	1.66	2.6	0.43
Intermediate fertility	9.38	1.9	1.80
Low fertility	1.09	0.5	0.05
Flooded	1.70	0.1	0.02
Other	0.99	0.25	0.03
Total	14.8		2.4

sek (1990) stratified fluxes from intact tropical forest on the basis of soil type, basing an estimate for a given type largely on a generalized relationship between soil N availability (mineralization potential) and N_2O flux for a series of tropical sites (Matson & Vitousek, 1987). This stratification (Table 6-3) leads to a total estimate of 2.4 Tg N_2O-N from moist and wet lowland tropical forests. If one includes Matson and Vitousek's (1990) estimate for additional flux due to conversion of tropical forests to pasture (0.7 Tg N), plus a flux of 1.0 Tg N for fluxes from seasonally dry tropical forests (Vitousek et al., 1989), one can estimate tropical forest fluxes at 4.1 Tg N_2O-N annually. This contrasts with an earlier estimate of 7.4 Tg N for these regions (Table 6-1).

Even this flux may need downward revision, however, when additional measurements of N_2O flux from successional forests become available. Davidson et al. (1991) report higher N_2O fluxes following dry forest conversion to agriculture (*Zea mays*) only for the first 2 yr following clearing; thereafter, following conversion to pasture, N_2O fluxes were similar to the lower fluxes in intact forest. Robertson and Tiedje (1988) found high denitrification rates in lowland tropical rainforests recently cleared of midsuccessional vegetation, but only for the first 6 mo following clearing. After 6 mo and in older midsuccessional sites, rates were substantially lower than in either the recently cut or in uncut primary rainforest sites, apparently due to the rapid depletion of labile soil C stores—initially elevated by clearing—together with the depletion of soil inorganic N due to rapid plant regrowth. Such evidence suggests that tropical forest conversion may lead ultimately to lower rather than to higher N_2O fluxes following deforestation.

Nitrous Oxide

Summing these best estimates of N_2O source strengths, including a newly discovered contribution of 0.4 Tg N from the manufacture of Nylon (Thiemens & Trogler, 1991), yields a total global source strength for N_2O of 7.9 Tg N (Table 6-1). This contrasts with a sink strength of 14.1 Tg N, leaving 6.5 Tg N_2O-N yr^{-1} unidentified as to source.

What is the missing source? Inverse modeling of the global atmosphere using observed interhemispheric N_2O ratios (Prinn et al., 1990) suggests that about one-half of the present atmospheric accumulation rate (3.5 Tg N_2O-

N) ought to be coming from tropical sources and about one-half from temperate. Examination of the global budget presented in Table 6-1 suggests that identified tropical sources are about three times greater than identified temperate sources. This suggests—surprisingly—that our temperate region sources are more poorly known than our tropical sources. Perhaps much of this missing source will be traced to the >70 Tg of manufactured fertilizer N added each year to the biosphere.

THE GLOBAL NITROGEN OXIDES
(NITRIC OXIDE, NITROGEN DIOXIDE) BALANCE

Logan's (1983) global budget for NO_x lists seven sources for atmospheric NO_x that toal to 25 to 99 Tg N yr^{-1} (Table 6-4). Major sinks for NO_x sum to 24 to 64 Tg N. This level of imprecision makes it difficult to evaluate the net global NO_x balance, though one could argue that it is not apparently out of balance.

The large degree of uncertainty in these ranges stem largely from measurement uncertainty, but it also stems from a relatively poor understanding of atmospheric processes affecting NO_x. Unlike N_2O, which is basically unreactive in the lower atmosphere, NO_x reacts with a wide variety of canopy-generated compounds near the ground and is produced elsewhere in the atmosphere by lightning and the oxidation of NH_3. Deposition sinks—in particular dry deposition onto heterogeneous surfaces such as plant canopies—are equally difficult to quantify. The global NO_x budget is correspondingly imprecise.

Of the major biogenic sources of NO_x, soil microbial activity is one of the most important. In a recent revision of Logan's (1983) values for soil NO_x sources, Davidson (1991) revised the earlier estimates of 8 Tg yr^{-1} of emitted NO_x-N to 20 Tg N on the basis of the number of NO_x flux studies

Table 6-4. Nitrogen oxides (NO_x) budget (from Logan, 1983, except as noted).

Sinks/sources	NO_x-N
Sinks	
Wet precipitation	12–42
Dry precipitation	12–22
Total	24–64
Sources	
Combustion	
Industrial	21 (14–28)‡
Biomass burning	12 (4–24)
Lightning	8 (2–20)
Soil (microbial activity)	20†
Atmospheric NH_3 oxidation	6 (1–10)
Oceans	<1
Stratosphere	<1
Total	68 (43–104)

† From Davidson (1991).
‡ Values in parentheses indicate likely range.

published in the interim years. Davidson's analysis suggests that cropland is the most intensive NO_x-N emittor, with average flux rates >6 ng cm^{-2} yr^{-1} that extrapolate to about 30% of the 20 Tg total flux from soils. The 23 flux estimates cited in his analysis represent a major improvement of our understanding of NO_x fluxes from terrestrial ecosystems [a single study was available to Logan (1983)], but the database remains lean; as Davidson (1991) points out, we are in great need of reliable NO_x fluxes from a number of systems and our understanding of the effects of soil and ecosystem disturbance of these fluxes remains weak.

Part of the difficulty in assessing NO_x fluxes from soil stems from the complex reactions that NO_x undergoes in the plant canopy (Jacob & Bakwin, 1991; Johansson, 1989): NO emitted from soils (see Hutchinson & Davidson, 1993) is converted rapidly to NO_2, which can then either be photolyzed back to NO, vented to the atmosphere above the canopy, converted to peroxyacetylnitrate (PAN), or deposited to foliar surfaces where it is reduced to nitrite and then assimilated via nitrite reductase to plant N. The conversion of soil-derived NO to NO_2 is mediated by O_3 and peroxy radicals produced from the decomposition of plant-produced hydrocarbons such as isoprene and terpenes. The conversion of NO_2 to PAN is also mediated by the oxidation of plant-produced hydrocarbons.

The NO_x emissions from terrestrial ecosystems—be they native communities or agricultural communities—is thus a complex function of NO emission rates from soil microbes, plant photosynthetic activity and hydrocarbon emission rates, canopy venting rates, incident light, and tropospheric O_3 concentrations. Understanding the NO_x cycle and its global balance—and especially its capacity for affecting important gas reactions in the troposphere via the regulation of OH and O_3 concentrations—at this point suffers from a paucity of emission data. One hopes that the next 5 yr show as much progress in this area as have the previous 5 yr.

FLUX EXTRAPOLATIONS TO GLOBAL SCALES

A major problem facing those who attempt to use local flux measurements to estimate global fluxes is that of extrapolation. Nitrogen gas fluxes are extremely heterogeneous at both field (e.g., Folonoruso & Rolston, 1984; Robertson et al., 1988) and landscape (e.g., Matson & Vitousek, 1987; Groffman & Tiedje, 1989) scales. Evaluating the appropriate sampling scales for within-field estimation and subsequent extrapolation of these rates to biome/disturbance types worldwide is not an exercise for the faint of heart. It is not unusual for coefficients of spatial variation within individual communities to exceed 100% for chamber-based flux estimates, nor it is unusual for specific types of plant communities (e.g., northern hardwood forest or tropical pasture) to express different annual fluxes in different parts of a landscape.

Problems of extrapolation are best addressed via sampling efforts at a variety of scales, from approaches based on 0.5-m^2 chambers (see Hutch-
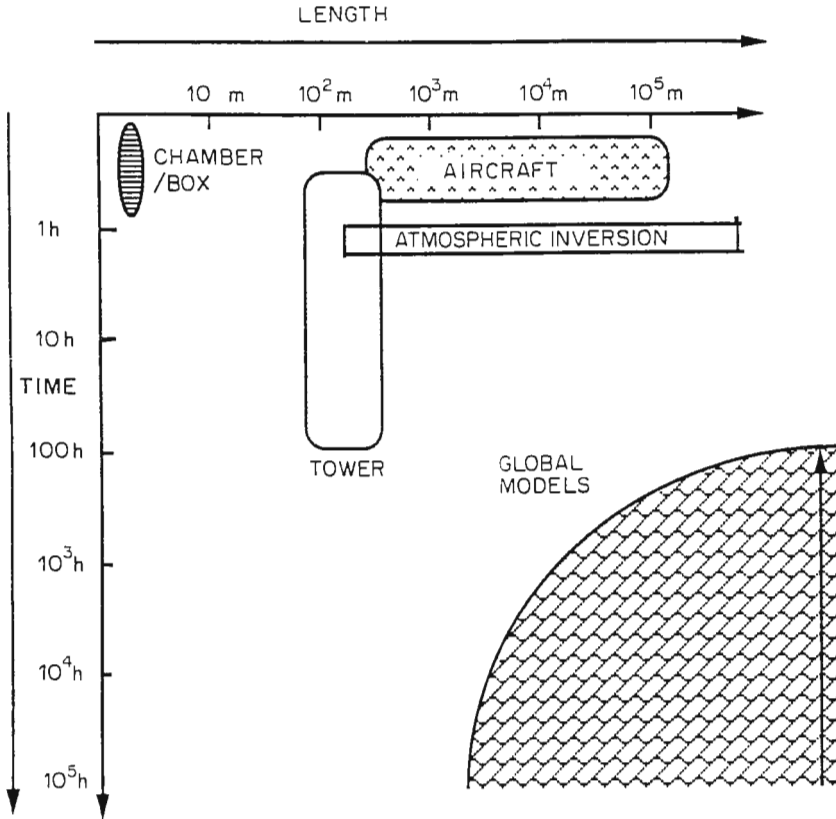
LENGTH

Fig. 6-4. Comparison of the scales that different measurement techniques capture in space and time (from Stewart et al., 1989).

inson & Livingston, 1993) to those employing towers and aircraft with kilometer square and greater footprints (Fig. 6-4; and Desjardin et al., 1993). For N_2O, however, present technology limits sampling strategies primarily to chamber-based approaches: fluxes of N_2O from soils are generally low relative to the 310 ppb_v concentration in today's atmosphere, and this for all practical purposes makes the use of open-air techniques based on concentration gradients (e.g., eddy-correlation and FTIR techniques) all but impossible. We are thus limited to chamber-based approaches for quantifying N_2O fluxes from most ecosystems, which means that our choice of within-system sampling strategies as well as our choice of within-biome habitats will be critical for an adequate assessment of worldwide fluxes—as well as for our predictions of changes in fluxes in response to potentially widespread disturbance such as changes in physical and chemical climate (Robertson et al., 1989).

   Up to this point our lack of data from N_2O and NO_x fluxes for major biomes has justified expeditionary-style sampling campaigns to strategic regions. Such campaigns have provided and will continue to provide needed

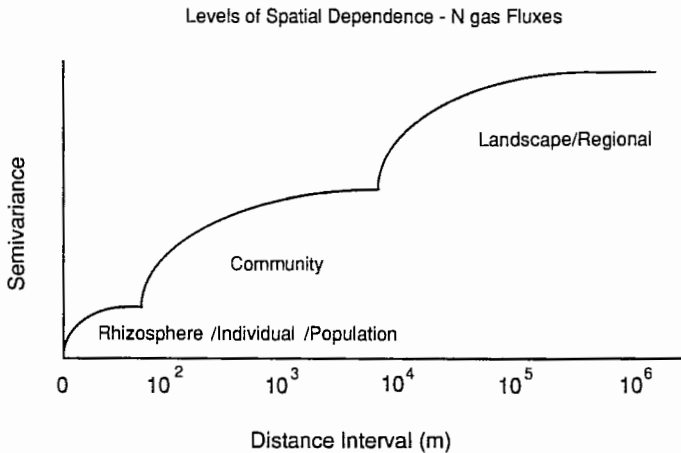Levels of Spatial Dependence - N gas Fluxes

Fig. 6-5. Potential levels of spatial dependence (semivariance) for N gas fluxes across several spatial scales. In this representation >80% of the variance in total global fluxes occurs at the community and landscape/regional scales rather than at the scale of individual plants to populations.

initial assessments of the likely importance of specific biomes and habitat types to regional and global fluxes. Indeed, these assessments form most of the basis for our global flux estimates (Tables 6-1 and 6-4).

We are now close to the point, however, at which much-needed improvements in the precision of our estimates will not come from examination of fluxes in unknown regions, but from refinements of our understanding of fluxes across high-flux landscapes. But these refinements must be sufficiently detailed and must provide sufficient information about mechanisms underlying these fluxes to allow the development of testable gas-flux models. Such models (e.g., Mosier et al., 1983) are necessary for generalizing across both landscapes and disturbance regimes.

Choice of spatial scale is critical for optimizing sampling efforts. One means for making reasonable choices is embedded in geostatistical approaches to assessing spatial variability. In particular, semivariance analysis (e.g., Webster, 1985) provides a means for defining appropriate sample intervals. If, e.g., one were to examine the spatial autocorrelation of a flux across a geographic regions, one would likely find that variance in flux rates changes at definable geographic intervals corresponding to changes in major controls on fluxes.

This can be diagrammed conceptually as in Fig. 6-5, where a stepped semivariance function provides information on the scale at which major controls change (Burrough, 1983). For example, at very small sample intervals control is exerted by rhizosphere or soil aggregate influences as they affect the availability of labile C (provided by root exudates and soil organic matter breakdown), inorganic N supplies, and O_2 availability. Different plant species may have differnet influences on these factors, so that another change in control may appear at scales approximating plant community changes. At greater distances geomorpholocial factors may play a deciding role as soil

textures and types change, and at still greater scales climate effects—corresponding to biome changes—will be expressed.

Understanding how variation in fluxes is stepped through this range of scales should give one the opportunity to sample a known proportion of landscape-level variation by sampling at appropriate geographic intervals. For example, landscapes in which 90% of flux variability is expressed at geomorphic scales can be effectively sampled via intense sampling at these larger scales (e.g., Groffman & Tiedje, 1989); landscapes where most flux variability occurs at smaller scales, on the other hand, will require substantially more intensive sampling at the smaller scales.

It is too early to know which fluxes in which landscapes may benefit most from this type of analysis; the formalization of what is now done largely intuitively will probably, however, have the greatest payoff in very heterogeneous landscapes such as those typical of many temperate agricultural regions. Such landscapes are usually made up of a mosaic of very different habitats (field–forest–wetland), with most of the agricultural fields managed very differently from one another with respect to crop rotation and tillage and fertilization regimes.

CONCLUSIONS

1. Global budgets of N_2O and NO_x are important for quantifying the existing impact of anthropic activity on atmospheric processes that affect both physical climate and precipitation chemistry.

2. Current global budgets for N_2O suggest large unknown sources totaling 6.5 Tg N—known sources account for only 7.9 Tg of the 14.1 Tg sink for N_2O–N in the atmosphere.

3. Best estimates of fluxes suggest that managed landscapes—in particular temperate-region landscapes with high fertilizer inputs and tropical landscapes undergoing large-scale land conversion—are the most likely sources behind unknown fluxes.

4. Current global budgets for NO_x are too imprecise to conclusively judge their balance; nevertheless, the impact of soil-produced NO_x likely has a major effect on the OH and O_3 chemistry of the troposphere and on the N content of wet and dry deposition.

5. There appears to be no easy solutions to identifying unknowns in the current N_2O and NO_x budgets. Comprehensive sampling programs must take into consideration large-scale spatial variation (mainly spatial but implicitly temporal) that has been documented for many important habitats.

ACKNOWLEDGMENTS

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