GLOBAL BIOGEOCHEMICAL CYCLES, VOL. 7, NO. 4, PAGES 717-733, DECEMBER 1993

THE BIOSPHERE AS AN INCREASING SINK FOR ATMOSPHERIC CARBON: ESTIMATES FROM INCREASED NITROGEN DEPOSITION

D. W. Schindler
Department of Zoology and Department of
Botany, University of Alberta, Edmonton
Alberta, Canada

S. E. Bayley
Department of Botany, University of Alberta
Edmonton, Alberta, Canada

Abstract. Estimates of carbon uptake and storage based on global nitrogen deposition, C:N ratios for typical terrestrial ecosystems, and recent ecosystem-scale nutrient studies indicate that 1.0-2.3 Gt C yr^{-1} of carbon storage may be stimulated by anthropogenically caused increases in nitrogen deposition in the past century. Sixty four to eighty four percent of global nitrogen uptake appears to occur on northern continents, with the remainder largely in northern coastal oceans. Increased nitrogen input by terrestrial ecosystems causes increased accumulation of carbon as plant tissue, with C:N ratios generally 50 to 200:1. Calculations suggest that northern continents are a major sink for carbon and that nitrogen-stimulated carbon uptake may more or less balance global carbon losses to the atmosphere from deforestation and agriculture. Much of the uptake appears to occur in aggrading forests, and the question of how long it can continue has important consequences for global carbon budgets.

Paper number 93GB02562.
0886-6236/93/93GB-02562$10.00

INTRODUCTION

Despite strong evidence that burning of terrestrial biomass is a significant contributor to atmospheric CO_2 [Houghton et al., 1990], several recent models and empirical studies indicate that carbon uptake by the terrestrial biosphere must be almost equal to that lost via biomass burning, that is, that the terrestrial carbon budget is more or less in balance. For example, Quay et al. [1992] compared changes in atmospheric CO_2 with stable carbon isotope ratios in the oceans and the atmosphere from 1970 to 1990. They concluded that while the decreased ^{13}C in the oceans reflected fossil fuel consumption and deforestation, there had been no net release of CO_2 from the biosphere during that period. This implies that a large terrestrial carbon sink is balancing the documented loss of carbon from tropical biomass burning. Recent models of the global carbon budget suggest that the missing sink lies in the northern hemisphere and that an uptake of 1.6 to as much as 4.7 Gt C yr^{-1} is required to balance the global carbon cycle [Houghton et al., 1990; Tans et al., 1990]; (reviewed by Sarmiento and Sundquist [1992]; see also Watson [1992]).

Other recent studies have decreased the magnitude of the required terrestrial sink slightly. Broecker and Peng [1992] provide evidence that deepwater transport of carbon from the North

Atlantic to the South Atlantic, which is not accounted for in the above models, may reduce the magnitude of the terrestrial influx of carbon needed to balance the global budget. In addition, estimates of oceanic CO_2 influx have increased by 0.7 Gt yr^{-1} as the result of the recent recognition that the surface film of the ocean averages 0.3° C. cooler than general surface waters [Robertson and Watson, 1992]. Nevertheless, a significant terrestrial carbon sink is still needed to balance recent global carbon models.

Recent evidence from several sources suggests to us that both terrestrial and marine ecosystems in the northern hemisphere have increased their carbon uptake and storage in the 20th century, stimulated by the enhanced deposition of nitrogen. As a result, the biosphere may now be a larger sink for carbon than it was in preindustrial times. Our calculations indicate that the greater carbon storage caused by increased nitrogen deposition may be roughly equivalent to amounts lost in the destruction of forest ecosystems. A similar calculation was done by Peterson and Melillo [1985], but data available at that time were few. They concluded that only 0.2 Gt of the then estimated total CO_2 emissions of 7.8 Gt yr^{-1} was returned to the biosphere as the result of stimulation by nitrogen. Most notably, estimated increases in nitrogen deposition from human activity were lower then, due both to higher assumed background values and to the poorly known role of dry deposition. Studies in the late 1980s and early 1990s on coastal sedimentation and on the C:N ratios of biological tissues (reviewed below) also have shown increases in estimated carbon storage. On the basis of recent studies of nitrogen deposition, effects of forest fertilization and decline, eutrophication and long-range transport, we believe that Peterson and Melillo's calculated value is much too small, and that nitrogen-stimulated carbon uptake in the biosphere may be an increasingly important sink for atmospheric carbon.

NITROGEN-STIMULATED CARBON UPTAKE AND STORAGE BY THE OCEANS

Past models of global carbon budgets have assumed that oceans currently operate much as they did under pristine conditions. Broecker [1992] correctly argues that it is unlikely that we can directly measure the changes caused by human release of carbon on the carbon cycle for the global oceans, for rates of change are small compared to major fluxes and pool sizes. However, it is now known that human industrial activities have greatly increased nitrogen deposition over both the North Atlantic [Galloway and Whelpdale, 1987] and North Pacific oceans [Prospero and Savoie, 1989]. Altogether, anthropogenic activity accounts for 40-70% of the atmospheric inputs of nitrogen to northern oligotrophic oceans [Sharp, 1991]. Owens et al. [1992] also show that the nitrogen input to the surface ocean in the northern hemisphere has been increased substantially by anthropogenic emissions, largely via episodic events. They calculate that increased nitrogen deposition could have caused a substantial increase in the productivity of northern oceans.

Similarly, Walsh [1992] used recent sediment trap data to show that anthropogenically caused increases in nitrogen budgets of continental margins in the northern hemisphere have resulted in increasing losses of carbon and nitrogen from coastal areas to the deep sea, despite high rates of denitrification in coastal areas [Seitzinger, 1988] (reviewed by Sharp [1991]). Similarly, 40 years of detailed studies of the Baltic Sea also show that inputs of nitrogen have increased severalfold [Rosenberg et al., 1990], causing increases in production and standing crop of phytoplankton, and accumulation of organic matter in bottom sediments [Cedarwall and Elmgren, 1990].

Because most of the increase in nitrogen emissions to the atmosphere has occurred since the turn of the century, northern oceans could be a larger sink for carbon than they were under pristine conditions. Legendre and Gosselin [1989] recommended that the role of atmospheric nitrogen deposition in the ocean's carbon budget should be reconsidered.

NITROGEN-STIMULATED CARBON UPTAKE AND STORAGE ON THE CONTINENTS

Calculations of nitrogen-stimulated carbon storage for terrestrial ecosystems are difficult, due to the extreme diversity of ecosystem types, C:N ratios and turnover times for carbon and nitrogen pools. Below, we outline some of the more severe problems, but proceed to make the calculations in any case, for they illustrate that nitrogen-stimulated terrestrial carbon uptake is important, if almost any reasonable assumptions are used.

Linking the Terrestrial Carbon and Nitrogen Cycles

While deforestation and forest decline have released carbon to the atmosphere in some areas [Houghton et al., 1990; Kauppi et al., 1992], evidence is accumulating that carbon storage in the terrestrial carbon pool of the northern hemisphere has increased in the past several decades. The changes appear to involve small increases in carbon flux over relatively large areas. For reasons similar to those voiced by Broecker for the oceans, it is therefore nearly impossible to directly measure large-scale rates of change in the terrestrial carbon pool or flux. However, assuming that the changes occur over large areas, and that many northern forests and wetlands are nitrogen limited, or at least strongly retain entering nitrogen [Vitousek and Howarth, 1991] (also discussed below), we believe that increased carbon storage can be estimated from anthropogenic changes to the terrestrial nitrogen cycle. Many of the assumptions that must be made in this calculation have been tested by recent research.

One of the present difficulties is linking studies of the terrestrial nitrogen and carbon cycles. Although there are a number of high-quality studies of either element (for example, the numerous European studies of critical nitrogen loading discussed below and papers in Woodwell [1984]), investigators, sites, sampling and analytical methods, times of study and units often differ so greatly that interfacing budgets for the two elements is very difficult. For example, often, investigators have examined the effects of nitrogen on photosynthesis, while ignoring effects on net long-term carbon storage, redistribution, or recycling.

Wet deposition of nitrogen in Europe and North America has been thoroughly studied as a component of the acid precipitation problem, and estimates are quite good. Estimated losses of nitrogen of anthropogenic origin to the oceans via river systems are less precise, with recent estimates varying by a factor of 5 (reviewed by Duce et al. [1991]). The most recent total estimate [Wollast, 1983] is about the average of all other estimates at 0.021 Gt yr^{-1}, and we use it in our calculations below. The concentrations of nitrate exported is largely a function of human population densities in individual basins [Peierls et al., 1991]. Other components of the terrestrial nitrogen cycle are still rather poorly known. For example, releases from the biosphere due to denitrification and uptake by nitrogen fixation have been studied at only a few sites. Still other components are very difficult to estimate: for example, deposition of nitrogen via dry deposition and from fog and dew are still only known for a few sites [e.g., Johnson and Lindberg, 1992; Sirois and Barrie, 1988; Weathers et al., 1988; Malanchuk and Nilsson, 1989; G. Lovblad and J.W. Erisman, unpublished manuscript, 1992]. Nevertheless, continental nitrogen budgets are much more precisely known than those for carbon.

Evidence for Increasing Nitrogen Deposition to the Continents

Atmospheric inputs of nitrogen to the continents have increased dramatically since the industrial revolution. Nitrogen oxides and ammonium released to the atmosphere by high-temperature combustion, fertilizer manufacturing and application, intensive livestock culture, and a number of smaller activities have increased the concentrations of ammonium and nitrate in 20th century rain and snow in Europe and much of North America severalfold from prehistoric values [Schindler et al., 1981; Malanchuk and Nilsson, 1989]. In addition, fluxes of organic nitrogen, dry deposition, and in some cases direct uptake of gaseous ammonium and nitrogen oxides by plants are all important fluxes that have been increased significantly by human activity in the past several decades. At most intensively studied sites, these mechanisms increase the total deposition of nitrogen by at least twofold over wet deposition values [Lindberg et al., 1986; Driscoll et al., 1989].

Recent European studies at several sites directly illustrate the importance of dry nitrogen deposition. At a number of locations in northern, central and southern Europe, transparent "roofs" have recently been installed just below the canopies of experimental catchments, in order to catch "throughfall" and "stemflow" [Dise and

Wright, 1992; Rasmussen, 1990; Beier et al., 1993]. These studies also show that capture of gases, particles, or aerosols by plant canopies can cause total nitrogen inputs to forests to be severalfold higher than from wet deposition alone (see also Hauhs et al. [1989] and Dise and Wright [1992]).

Changes in land use in the 20th century have also stimulated the deposition of nitrogen, by enhancing the ability of the biosphere to remove nitrogen. In general, reforestation of areas cleared earlier for pastures and agriculture has contributed to increased nitrogen deposition in Europe and northeastern North America [e.g., Drablos et al., 1980; Johannes et al., 1986]. Both coniferous and deciduous forests are generally more efficient than grasslands at capturing dry and gaseous forms of nitrogen and other substances [Rasmussen et al., 1993; Hultberg et al., 1993]. Also, in dissected landscapes where forests and fields are intermixed, uptake of nitrogen at the edge of forests can be much higher than in the center of forest plots, due to "edge effect" removal of atmospheric pollutants [Grennfelt and Hultberg, 1986; Weathers et al., 1992]. In some areas, fertilization of forests and agricultural lands with nitrogen produced by anthropogenic fixation of atmospheric nitrogen would supplement these estimates [Rosenberg et al., 1990].

Inputs of nitrogen to land at high elevation have probably been undersetimated. At high elevations, fog and cloud inputs can enhance deposition as rain and snow, measured as wet deposition. For example, Lovett et al. [1982], Weathers et al. [1988], and Norton et al. [1993] found total nitrate deposition at high-elevation catchments in eastern North America to be up to 6X higher than in wet deposition at the sites. Hicks [1989] reviews the topic.

Retention of Nitrogen Input by Terrestrial Ecosystems

Recent detailed studies at many locations show that in relatively pristine areas, much of the nitrogen entering terrestrial ecosystems from the atmosphere is retained, or, more correctly, that little of the nitrogen input escapes via streamflow [Likens et al., 1977; Dise and Wright, 1992; Bayley et al., 1992; Schindler et al., 1976, 1980; Dillon and Molot, 1990; Swank and Waide, 1988; Wright et al., 1993]. Even at severely polluted sites, nitrogen retention is often more than 70% of total input. In the 40 catchments at 19 locations in 10 countries studied as part of the European ENCORE (European Network of Catchments Organised for Research on Ecosystems) project, retention of nitrogen was over 75% at all but a few sites. Sites with detectable forest decline generally retained slightly lower percentages of nitrogen input than other sites [Hornung, 1993]. At European NITREX (Nitrogen Saturation Experiments) sites, retention efficiency at sites receiving deposition of over 35 kg of inorganic N $ha^{-1}yr^{-1}$, decreased to less than 50%. At deposition rates of up to 20 kg of inorganic N ha^{-1} yr^{-1}, retention at most NITREX sites was over 90% [Dise and Wright, 1992]. In general, young, aggrading forests retain nitrogen more efficiently than old, mature stands, and retention efficiency is highly variable in the latter [Bormann and Likens, 1979]. Johnson [1992] found that 19 of 30 sites with mature forest stands had retention efficiencies of over 40%.

Even when heavy doses of fertilizer are applied to aggrading forested sites in areas with high nitrogen input from the atmosphere, retention of nitrogen can be very high. In Swedish forests fertilized with 60 kg N ha^{-1} yr^{-1} for 17 years as urea or NH_4NO_3, Tamm and Hogberg [1993] found nearly complete retention of fertilizer. In the Black Forest of southern Germany, retention of nitrogen from fertilized forest catchments in 1986-1991 were over 80% of inputs in one set of catchments and 96% in another set [Feger, 1993]. At Klosterhede, Denmark, with deposition of 140 meq N m^{-2} yr^{-1}, nearly 100% of input was retained by forest ecosystems [Gundersen et al., 1991]. Subarctic watersheds in Norway where nitric acid was applied at 50 meq m^{-2} yr^{-1} for 8 years still retained over 85% of input, despite very thin, stony soils [Wright et al., 1993]. At Bear Brook, Maine, nitrogen retention was 99% of inputs in the reference catchment, and 90% in a catchment fertilized for 2 years [Norton et al., 1993]. In Japan, several years of application of sewage water to forests resulted in retentions of nitrogen of over 90% [Iwatsubo and Nagayama, 1993]. Johnson [1992] found that some nitrogen was retained by forested ecosystems, even at fertilizer applications of 1500 kg ha^{-1}.

Wetland catchments appear to be even more efficient than forests at retaining nitrogen from precipitation [Bayley et al., 1992; Urban and Eisenreich, 1988; Hemond, 1983], providing they do not receive large volumes of water as runoff or stream input. Despite the potential of wetlands for denitrifying large amounts of nitrate, most measurements to date have revealed that denitrification rates are low, especially in nutrient-poor bogs and fens. The subject is reviewed by Urban and Bayley [1988] and Verhoeven [1992].

Similarly, several years of nitric acid applications to an ombrotrophic bog in the boreal forest and to a subalpine upland catchment did not significantly lower retention of nitrogen [Bayley et al., 1992; Wright et al., 1993].

In many of the above studies, retention by terrestrial catchments was calculated by subtracting the loss of nitrogen measured in streamflow from input measured in rain and snow. If all inputs were corrected for gaseous and dry deposition, as discussed above, calculated percentage retention would be even higher. Our calculations assume that inputs to ecosystems from nitrogen fixation and losses from denitrification are insignificant. Schlesinger [1991] estimates that the former usually ranges from 1 to 5 kg $ha^{-1}yr^{-1}$ and the latter average 2 kg $ha^{-1}yr^{-1}$ at terrestrial sites, although data are few and spatial variability is high.

Altogether, it appears that retention of anthropogenically released nitrogen by continents has been substantial in the 20th century. An approximation of the total retention of anthropogenically produced nitrogen on land can be made by subtracting the sum of (1) riverine losses from the continents and (2) atmospheric inputs of nitrogen to the oceans from the sum of (3) global emissions of oxidized and reduced forms of nitrogen to the atmosphere and (4) fertilizer production. Fortunately, Duce et al. [1991] provide recent estimates of 1, 2, and 3. Schlesinger [1991] estimates that fertilizer production is nearly as large as continental nitrogen deposition, but most fertilizer is applied to agricultural fields where there is negligible long-term carbon storage. We therefore ignore carbon uptake resulting from fertilization and nitrogen fixation on agricultural lands in our calculations.

Evidence that Nitrogen Stimulates Terrestrial Carbon Uptake

A high proportion of the increased deposition of nitrogen has occurred over areas covered primarily by temperate mixed or boreal forests and associated lakes and wetlands. Most of the studies of nitrogen deposition and retention reviewed above were done in such areas. They illustrate that retention of nitrogen by terrestrial ecosystems is associated with increased growth of woody plants. Growth in most northern forest tree species is stimulated by increased inputs of nitrogen, at least at rates similar to those resulting from atmospheric deposition [Mitchell and Chandler, 1939; Tamm, 1990]. In a nitrogen-limited forest ecosystem, nitrogen fertilization stimulated primary productivity more than decomposition [Hunt et al., 1988]. Nitrogen fertilization increased the N content of the living plant material and forest litter [Hunt et al., 1988]. A recent review of nitrogen and carbon dynamics in North American vegetation types shows that primary production of temperate, boreal, and tundra ecosystems in many areas is moderately to severely nitrogen limited and that coupled carbon-nitrogen dynamics must be accounted for in order to properly model terrestrial carbon fluxes [McGuire et al., 1992].

Forest biomass appears to be increasing, even at many sites where air pollution is causing damage to some species. Kauppi et al. [1992] have recently argued that there has been a 25% increase in stem volume of European trees between 1971 and 1990, despite forest decline caused by air pollution in some areas. Where measured, rates of forest growth have increased even more, averaging 30% for Finland, France, and Sweden. In contrast, they estimate that areas of forest decline are approximately 8000 km^2, about 0.5% of the total forested area of Europe.

Reports from several intensive studies at the May 1992 Conference on Experimental Manipulation of Ecosystems in Copenhagen, Denmark, support the view that north temperate forests and wetlands have responded and will respond in the future to increased inputs of nitrogen by adding atmospheric carbon to long-term terrestrial carbon sinks such as soils, tree stems, and peat [Rasmussen et al., 1993]. Tamm and Hogberg [1993] found that annual fertilization with NH_4NO_3 increased the stem growth of young

Swedish pine and spruce plantations by from 30 to 300% over 17-20 years. Iwatsubo and Nagayama [1993] reported that sewage nitrogen sprayed on forests in Japan for 13 years increased growth of trees by sixfold. In EXMAN (EXperimental MANipulations of forest ecosystems) plots in the Netherlands, fertilization with nitrogen also caused substantial increases in growth of trees at most sites. Near Skogaby, Denmark, application of $(NH_4)_2SO_4$ to forest plots increased growth of trees as basal area by from 22 to 37% [Beier et al., 1993]. Abrahamsen [1993] reported that biomass in Norwegian forests has increased by 30% in the last 20-30 years, due to a combination of increased nitrogen deposition, better forestry, and denser stands.

Nitrogen fertilization experiments at bog and tundra sites caused a severalfold increase in growth of plant species. Experimental addition of 5 kg N $ha^{-1}yr^{-1}$ as HNO_3 to a boreal bog caused increased growth of Sphagnum mosses [Rochefort et al., 1990; S.E. Bayley, unpublished data, 1993]. Aerts et al. [1992], Ferguson et al. [1984], and Gardetto [1987] report similar results from manipulation experiments in wetlands. Shaver et al. [1992] and Shaver and Chapin [1980, 1986, 1991] report 50 to 150% increases in biomass and carbon accumulation in Alaskan tundra plots fertilized with N, P, and K, with nitrogen usually the limiting element.

Calculating the Increased Global Carbon Storage Stimulated by Increases in Nitrogen Deposition

The increased amount of carbon stored in the oceans is relatively simple to calculate for oceanic regions, if we assume that C:N in sedimenting plankton is in Redfield proportions of 7:1 by weight, that most anthropogenic inputs of nitrogen are consumed by plankton, and that sedimented nitrogen and carbon retain the Redfield ratio (reviewed by Owens et al. [1992]). Using the sum of nitrogen inputs from precipitation and river inputs to marine systems given by Duce et al. [1991], oceans would be expected to store 0.36 Gt of C yr^{-1} more today than they did under pristine conditions. This assumes that the anthropogenic nitrogen inputs have not caused compensating losses of nitrogen from surface oceans in coastal areas. Recent estimates of global carbon burial on continental slopes and rises based on the few available data indicate similar values, less than 0.7 Gt yr^{-1} [Reimers et al., 1992].

For terrestrial ecosystems, the calculation is more difficult, because the C:N ratios and turnover times of various terrestrial ecosystem components vary greatly. New leaves or shoots and active soil layers generally have rather low C:N ratios of 15-20 by weight, but range from 10 to 50 (Table 1), with turnover times ranging from months to a few years. Such components are relatively unimportant reservoirs of increased carbon storage. In contrast, dead or inactive plant material such as the boles and roots of trees have C:N ratios of 150-300. Ratios in peat range from 28 to 180 (Table 1). Such materials probably contain most of the carbon storage stimulated by increased nitrogen deposition. Turnover times range from decades for woody forest material to millenia for long-term peat deposits. The C:N ratio in the upper meter of forest soil usually averages 10-30 [Post et al., 1985] (Table 1), and carbon turnover times appear to be several years at the few sites where they have been measured [Harrison et al., 1993].

TABLE 1. Carbon:Nitrogen Ratios by Weight of Major Storage Components of Biomes in the Temperate, Boreal, and Arctic Parts of the Northern Hemisphere

Location/Compartment	C:N Ratio	Reference
	Tundra	
General (from model)		
vegetation	41.6	Rastetter et al. [1991]
soils	17.5	Rastetter et al. [1991]
Sedge meadow Devon Island CN		
vegetation	52	Van Cleve and Alexander [1981]
soils	14.6	Van Cleve and Alexander [1981]

Table 1. (continued)

Location/Compartment	C:N Ratio	Reference
Toolik Lake Alaska		
tussock plants	49	Shaver and Chapin [1991]
shrub plants	55	Shaver and Chapin [1991]
Toolik Lake Alaska		
alpine tundra vegetation	70	McQuire et al. [1992]
wet/moist tundra	50	McQuire et al. [1992]
moist tundra soils	16	McQuire et al. [1992]
Wet meadow Barrow Alaska		
vegetation	76.6	Van Cleve and Alexander [1981]
soils	20.6	Van Cleve and Alexander [1981]
	Boreal Forest	
Black spruce Fairbanks Alaska		
plants	217.1	Van Cleve and Alexander [1981]
soils	34.7	
Aspen forest Fairbanks Alaska		
plants	224	Van Cleve and Alexander [1981]
soils	17.7	
Birch forest Fairbanks Alaska		
plants	217.1	Van Cleve and Alexander [1981]
soils	815.8	
Birch forest Finland		
plants	186	Van Cleve and Alexander [1981]
Boreal woodland Schefferville CN		
vegetation	91	McQuire et al. [1992]
Boreal forest Bonanza Cr Alaska		
vegetation	449	McQuire et al. [1992]
soils	30	McQuire et al. [1992]
Northern conifer		
total biomass	256	Vitousek et al. [1988]
leaf	49	Vitousek et al. [1988]
branch	137	Vitousek et al. [1988]
bole	433	Vitousek et al. [1988]
root	289	Vitousek et al. [1988]
	Peatlands	
Mire Stordalen Sweden		
plants	57	Van Cleve and Alexander [1981]
peat	37	Van Cleve and Alexander [1981]
Bog Ontario CN		
Sphagnum plants	45	S. E. Bayley (unpublished data, 1993)
peat (0-10cm)	49	S. E. Bayley (unpublished data, 1993)
Bog after N fertilization with HNO3 @ 50 kg N ha^{-1} yr^{-1}		
Sphagnum plants	28	S. E. Bayley (unpublished data, 1993)
peat (0-10cm)	37	S. E. Bayley (unpublished data, 1993)
Bog Minnesota		
Sphagnum plants	50	Urban and Eisenreich [1988]
Bog eastern Ontario CN		
peat (0-10cm)	45	Riley [1987], Riley and Michaud [1989]
peat (whole column)	29	Riley [1987], Riley and Michaud [1989]
Bog northern Sweden		
peat (0-10cm)	100-180	Malmer and Holm [1984]
peat (10-20cm)	100-150	Malmer and Holm [1984]
Bog southern Sweden		
peat (0-10cm)	100	Malmer and Holm [1984]
peat (20 cm)	60	Malmer and Holm [1984]

Table 1. (continued)

Location/Compartment	C:N Ratio	Reference
	Temperate Coniferous Forest	
H.J. Andrews forest		
vegetation	604	McQuire et al. [1992]
soils	50	McQuire et al. [1992]
	Deciduous Forest	
General (from model)		
plants	159.5	Rastetter et al. [1991]
soils	13.2	Rastetter et al. [1991]
woody tissue	300	Rastetter et al. [1991]
Hubbard Brook New Hampshire		
leaf material	20	Likens et al. [1981]
living biomass	137	
forest floor	22	
Aspen forest Wisconsin		
vegetation	207	Pastor and Bockheim [1984]
forest floor	41	
mineral soils	11.6	Pastor and Bockheim [1984]
	Deciduous forest (cont.)	
Southern deciduous forest Coweeta North Carolina		
plants	109	Monk and Day [1988]
litter	36.8	Monk and Day [1988]
soils	3.9	Monk and Day [1988]
Harvard Forest Massachusetts		
deciduous forest vegetation	417	McGuire et al. [1992]
deciduous forest soils	20	McGuire et al. [1992]
mixed forest vegetation	408	McGuire et al. [1992]
mixed forest soils	20	McGuire et al. [1992]
Temperate broad-leafed deciduous forest		
total biomass	155	Vitousek et al. [1988]
leaf	25	Vitousek et al. [1988]
branch	106	Vitousek et al. [1988]
bole	236	Vitousek et al. [1988]
root	107	Vitousek et al. [1988]

Because most of the increased nitrogen deposition has occurred in the past hundred years [Husar, 1986], in forested areas it is probably still associated largely with biological reservoirs such as tree trunks and roots, which have C:N ratios of 150 or more [Schlesinger and Hartley, 1991] (Table 1). Johnson [1992] also found that nitrogen added to fertilized forest plots caused biomass to accumulate largely in trunks and roots. C:N values in such reservoirs do not appear to decline as nitrogen inputs increase.

In boreal wetlands, most stored carbon is in peat, rather than in living plants. Because average peat accumulation rates average only 0.5 mm yr^{-1} [Gorham, 1991], carbon uptake stimulated by increased nitrogen deposition will be stored in the upper 10-20 cm of peat deposits. These have C:N ratios of 37-180 (Table 1).

While it is impossible to specify a biospheric average C:N ratio for terrestrial material sequestered in the past century, it is probably in the range of 50 to 150, when the C:N ratios of various carbon pools, their turnover times and their locations with respect to areas of increased nitrogen deposition are considered. To illustrate the possible importance of nitrogen-stimulated carbon uptake, we have chosen values of 50, 100, and 150 for C:N ratios, which we believe bracket the likely average C:N ratio of several decade storage products in temperate and boreal forests and wetlands. Overall results of our calculations are set out in Table 2.

The calculations illustrate that a good part of the carbon indicated as "missing" by recent models, 1.0 to 2.3 Gt yr^{-1}, could be accounted for by increased uptake and storage in the biosphere, resulting from stimulation by anthropogenically enhanced nitrogen deposition. Of the increase in carbon stimulated by nitrogen fertilization, our estimates suggest that the terrestrial biosphere accounts for from 64% to 84% of the total, with the remainder in the oceans. This nitrogen-driven terrestrial sink for carbon has probably developed largely in aggrading European and eastern North American forests within the past century, and its future magnitude will depend on the human emissions of nitrogen to the biosphere, and the degree of saturation of "receptor" ecosystems.

Our calculations have ignored the effect of fertilization on agricultural soil. Indeed, carbon and nitrogen may have been lost to the atmosphere due to degradation of agricultural soils in the 20th century [e.g., Jenny and Raychaudhuri, 1960]. However, fertilizer nitrogen is undoubtedly an important component of nitrogen losses to the sea via river flow [Wollast, 1983; Peierls et al., 1991]. In brief, exclusion of fertilizer nitrogen may tend to cause our estimates of terrestrial carbon uptake to be slightly low, but would not affect our estimates for the oceans.

In general, our calculations support the results of stable carbon budgets for the ocean [Quay et al., 1992] and O_2/N_2 for the atmosphere [Keeling and Shertz, 1992] (see also Broecker and Severinghaus [1992]) in the conclusion that there has been little net change in the carbon storage in

TABLE 2. Anthropogenically Enhanced Nitrogen Inputs and Associated Carbon Uptake by the Biosphere

Line	Parameters	Gt Yr $^{-1}$
	Anthropogenic Sources and Sinks of Nitrogen	
1	Total nitrogen emissions	0.064
2	N deposited to oceans	0.030
3	N carried to oceans by rivers	0.021
4	Net N deposition in oceans (lines 2+3)	0.051
5	Net nitrogen deposition on land (lines 1-(2+3))	0.013
	Sinks of Carbon Based on Various C:N Ratios	
6	Net C deposition in oceans (line 4) x C:N = 7	0.36
7	Net C deposition on land (line 5) x C:N ratio	
	assuming a C:N ratio of 50:1	0.65
	assuming a C:N ratio of 100:1	1.30
	assuming a C:N ratio of 150:1	1.95
8	Total biospheric carbon uptake (based on line 6 + line 7)	
	assuming a C:N ratio of 50:1	1.01
	assuming a C:N ratio of 100:1	1.66
	assuming a C:N ratio of 150:1	2.31

the terrestrial biosphere taken as a whole, that is, that carbon uptake stimulated by anthropogenically caused increases in nitrogen deposition more or less balances the losses from deforestation and agriculture.

NITROGEN DEPOSITION: A FAUSTIAN BARGAIN?

If anthropogenic increases in nitrogen deposition have indeed helped to minimize carbon accumulation in the atmosphere, the benefits of enhanced carbon removal and storage in decreasing the potential for climatic warming must be weighed against the negative effects of increased nitrogen deposition, which include forest dieback, eutrophication, acidification and formation of toxic air pollutants. It therefore seems useful to briefly summarize the negative effects of allowing high nitrogen emissions from human activity to continue.

Forest dieback in Europe is closely linked to nitrogen deposition in many areas, due to a combination of factors, including soil acidification caused by nitrification [e.g., Johnson et al., 1991] and micronutrient deficiencies induced by overfertilization with nitrogen and leaching of acidified soils. Ozone, peroxyacetyl nitrate, and other phytotoxic air pollutants are also formed by reaction of nitrogen emissions in the atmosphere [e.g., Schulze, 1989; Schulze and Freer-Smith, 1991].

In some areas where nitrogen deposition has increased, small declines in retention efficiencies of nitrogen by terrestrial catchments have caused small, but important increases in nitrate concentrations in surface waters. Logically, decreased rates of uptake of nitrogen by forests are expected as vegetation matures, but the time required for complete saturation is still not known. In softwater lakes and streams, nitric acid can cause acidification, particularly during snowmelt in the spring [Galloway et al., 1987]. Henriksen and Brakke [1988] reported that nitrate concentrations in Norwegian lakes had doubled in less than 10 years, becoming a major concern with respect to lake acidification. Even where acidification is not a threat, excess nitrogen can cause changes in the nutrient availability to aquatic plants. For example, Bennett [1986] documented a severalfold increase in nitrate in Lake Superior in this century.

Streams in several areas also carry increased concentrations of nitrate. Streams in the Catskill Mountains appear to have increased in nitrate for several decades [Stoddard and Murdoch, 1991]. At Dicke Bramke and Lange Bramke in Germany, nitrate has increased fivefold or more in 15 years [Hauhs, 1990]. Rudd et al. [1990] found that nitrogen saturation of freshwater lakes caused a dramatic increase in midsummer concentrations of nitrate. Uptake and sedimentation of nitrogen by algae were limited by phosphorus availability. Denitrification, a weak and seasonal process, was insufficient to remove high inputs of nitrate, serving as a "bottleneck" to normal air-water nitrogen cycling. Kelly et al. [1990] demonstrated that many North American lakes and streams appear to have reached the point where high nitrogen deposition has caused excess inorganic nitrogen to be present in summer. However, even where evidence of "leakage" of nitrogen to surface waters is evident, the efficiency of retention by terrestrial catchments is very high, implying that efficient nitrogen storage can be expected for some time in the future.

Recently, several scientific workshops have addressed the estimation of "critical loads" of nitrogen for European ecosystems, which should not be exceeded if forest damage and lake acidification are to be prevented [Nilsson and Grennfelt, 1988; Malanchuk and Nilsson, 1989; G. Lovblad and J.W. Erisman, unpublished manuscript, 1992].

Increased inputs of nitrogen to ecosystems also often encourage the growth of less desirable species that those that would otherwise be grown. Gradual replacement of species that are evolutionarily adapted to low-nitrogen conditions with those that respond quickly to enhanced nitrogen may occur in all terrestrial ecosystem types [Tamm, 1990]. In brief, there are many negative aspects of high nitrogen deposition that make one hesitate to recommend it as a mechanism for decreasing the rate of accumulation of CO_2 in the atmosphere.

In marine systems, enhanced nitrogen inputs are known to have caused eutrophication problems in most coastal bays and estuaries adjoining populous areas [Peierls et al., 1991; Howarth, 1993]. Larger examples include the Baltic [Rosenberg, et al. 1990] and Chesapeake Bay [Fisher et al., 1988].

CRITICAL RESEARCH QUESTIONS

As mentioned at the beginning, the recent concern over deposition of strong acids has stimulated interest in obtaining better estimates of nitrogen inputs and studies of the terrestrial nitrogen cycle. Similarly, concern for climatic warming has stimulated studies of the degree to which higher atmospheric CO_2 can stimulate carbon uptake has been the subject of recent climate-related research. It is unfortunate that these studies have not been better linked. There is little reason to expect that the cycles would or could operate independently, for the C:N ratio in plant tissues must certainly be among the most conservative properties of the biosphere. Without consideration of linkages between the two cycles, important questions regarding how long and to what extent higher than normal nitrogen deposition can continue to stimulate carbon storage in the biosphere cannot be addressed. The question of whether there are lags between the responses of nitrogen-driven carbon uptake and decomposition that may affect duration of nitrogen-stimulated storage is also critical, and slight redirection to coordinate ongoing studies of carbon and nitrogen could greatly improve the accuracy of estimated nitrogen-driven carbon uptake.

It is also necessary to consider linkages with other nutrient cycles. For example, phosphorus, potassium, and occasionally other elements are known to limit carbon uptake in some systems. If a nutrient other than nitrogen becomes more limiting, it could decrease the efficiency of nitrogen inputs.

Linkages to nonnutrient elements may be important. For example, increased deposition of nitric acid and ammonium can lead to high rates of soil acidification in areas with poorly buffered soils. Soil acidification can in turn disrupt some of the key nitrogen cycling processes in soils, for example, by eliminating soil fauna that is important in mineralization, or by disrupting nitrification. Tamm [1990] discusses the differences expected due to such disruptions in various soil types, but data are too few to allow generalizations over large areas.

Recent predictions of the patterns and rates of nitrogen releases in the 21st century suggest that changes in the global patterns of deposition may occur, so that it is critical to understand C:N relationships in the tropics and southern hemisphere. Galloway et al. [1993] predict a doubling of nitrogen emissions and deposition of oxidized nitrogen species between 1980 and 2020, but most of the increases are predicted to occur in the tropics and southern hemisphere, where enormous population increases and modest industrial growth are expected. Nitrogen deposition to both marine and terrestrial systems is expected to increase by fourfold or more south of the equator. In the northern hemisphere, nitrogen deposition over continental land masses is predicted to decrease throughout most developed countries, while increases in deposition of 1.5-fold to threefold are predicted for eastern Asia and the North Pacific [Galloway et al., 1993]. Because most of the increased deposition is expected in areas that are not presently subjected to high nitrogen deposition, but where mature forests have recently been cut at high rates, the terrestrial biosphere may continue to act as a carbon sink. Unfortunately, few studies have been made of the degree to which nitrogen additions can stimulate carbon uptake in terrestrial ecosystems of the southern hemisphere, although there is some evidence that tropical forests are not nitrogen limited [Vitousek and Howarth, 1991]. On the other hand, studies of tropical soils suggest that increased nitrogen inputs could enhance carbon sequestration in tropical forest soils for hundreds of years [Jenny et al., 1948; Jenny and Raychaudhuri, 1960]. In brief, more information is needed before it can be assumed that nitrogen-induced carbon uptake in the tropical terrestrial biosphere is of long-term significance.

There may also be both positive and negative feedbacks between nitrogen-driven carbon storage and climatic change, with no resolution of the relative importance of different mechanisms. In general, terrestrial ecosystems retain much more of incoming nitrogen during the growing season than they do in winter, so that increasing the length of growing seasons should enhance carbon storage. Warmer temperatures may also increase rates of recycling of organic nitrogen is soil [Jenny and Raychaudhuri 1960], making more available for tree growth or loss to streamflow. In mature forest plots where soils were experimentally heated 8°-10°C, artificially warming and extending the growing season, nitrogen contents of soil and tree needles were enhanced, but no evidence of higher tree growth was observed [Van Cleve et al., 1990]. However, in central Europe, droughts experimentally imposed upon forests under roofed catchments showed lowered retention of nitrogen [Beier et al., 1993]. One result of increased nitrogen supplies, whether from deposition or increased recycling rates, is that a higher proportion of carbon and nitrogen becomes stored in the roots and boles of trees than under pristine conditions (see also Hunt et al. [1988]). This would further enhance the carbon storage by terrestrial forests, because these components generally have higher C:N ratios than soils (Table 1).

In summary, predicting the role of increasing nitrogen deposition in stimulating carbon storage is not a straightforward process, and it requires much additional study.

Acknowledgments. We are grateful to W.S. Broecker for a series of stimulating arguments that caused us to pursue these calculations. G. E. Likens further stimulated our interest by providing estimates of nitrogen deposition under pristine conditions. Eville Gorham kindly sent us unpublished data for C:N ratios in mosses and peats of northern wetlands (Gorham in press), a number of rare references. J. N. Galloway, L. Rasmussen and R.F. Wright allowed us access to pre-publication copies of their manuscripts. Broecker, Likens, Gorham, R.W. Howarth, and an anonymous reviewer provided insightful criticisms of the manuscript. R. Vet sent us recent manuscripts and reports dealing with increased nitrogen deposition and critical loads. Work on the manuscript was supported by operating grants from the National Science and Engineering Research Council of Canada to D. W. Schindler (grant 89673) and S. E. Bayley (grant 89899).

REFERENCES

Abrahamsen, G., Long-term acidification experiments in forest stands, in *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* edited by L. Rasmussen et al., pp. 192-194, Commission of European Communities, Brussels, 1993.

Aerts, R., B. Wallen, and N. Malmer, Growth-limiting nutrients in Sphagnum-dominated bogs subject to low and high atmospheric nitrogen supply, *J. Ecol., 80,* 131-140, 1992.

Bayley S.E., D.W. Schindler, K.G. Beaty, B.R. Parker, and M.P. Stainton, Effects of multiple fires on nutrient yields in streams draining boreal forest and fen watersheds: Nitrogen and phosphorus, *Can. J. Fish. Aquat. Sci., 49,* 584-596, 1992.

Beier, C., et al., Effects of changing the atmospheric input to forest ecosystems-results of the "EXMAN" project, in *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* edited by L. Rasmussen et al., pp. 138-154, Commission of European Communities, Brussels, 1993.

Bennett, E.B., The nitrifying of Lake Superior, *Ambio, 15,* 272-275, 1986.

Bormann, F. H., and G. E. Likens, *Pattern and Process in a Forested Ecosystem,* Springer-Verlag, New York, 1979.

Broecker, W.S., Keeping global change honest, *Global Biogeochem. Cycles, 5,* 191-192, 1992.

Broecker, W.S., and T.-S. Peng, Interhemispheric transport of carbon dioxide by ocean circulation, *Nature, 356,* 587-589, 1992.

Broecker, W.S., and J.P. Severinghaus, Diminishing oxygen, *Nature, 358,* 710-711, 1992.

Cedarwall, H., and R. Elmgren, Biological effects of eutrophication in the Baltic Sea, particularly the coastal zone, *Ambio, 19,* 109-112, 1990.

Dillon, P.J., and L.A. Molot, The role of ammonium and nitrate retention in the acidification of lakes and forested catchments, *Biogeochemistry 11,* 23-43, 1990.

Dise, N.B., and R.F.Wright (Eds.), The NITREX Project (Nitrogen Saturation Experiments), *Rep. XX,* Air Pollut. Res., Comm. of Eur. Commun., Brussels, 1992.

Drablos, D., I. Sevaldrud, and J.A. Timberlid, Historical land-use changes related to fish status development in different areas of southern Norway, in *Ecological Impact of Acid Precipitation,* edited by D. Drablos and A. Tollan, pp. 367-369, SNSF (Acid precipitation - Effects on Forest and Fish) Project, Oslo, Norway, 1980.

Driscoll, C.T., D.A. Schaefer, and J.L. Malanchuk, Discussion of European and North American data, in *The Role of Nitrogen in the Acidification of Soils and Surface Waters,* edited by J. L. Malanchuk and J. Nilsson, pp. 6-1 to 6-45, Nordic Council of Ministers, Stockholm, 1989.

Duce, R. A., et al., The atmospheric input of trace species to the world ocean, *Global Biogeochem. Cycles, 5*, 193-259, 1991.

Feger, K. H., The ARINUS Project: Effects of whole-watershed nitrogen and sulfur additions on biogeochemical cycling in spruce forests, in *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* edited by T. Rasmussen et al., pp. 12-22, Commission of European Communities, Brussels, 1993.

Ferguson, P., R.N. Robinson, M.C. Press, and J.A. Lee, Element concentrations in five Sphagnum species in relation to atmospheric pollution, *J. Bryol., 13,* 107-114, 1984.

Fisher, D., J. Ceraso, T. Mathew, and M. Oppenheimer, *Polluted Coastal Waters: The Role of Acid Rain,* Environmental Defense Fund, New York, 1988.

Galloway, J.N., and D.M. Whelpdale, WATOX-86 overview and Western North Atlantic Ocean S and N atmospheric budgets, *Global Biogeochem. Cycles, 1,* 261-281, 1987.

Galloway, J.N., G.R. Hendrey, C.L. Schofield, N.E. Peters, and A.H. Johannes, Processes and causes of lake acidification during spring snowmelt in the west-central Adirondack Mountains, New York, *Can. J. Fish. Aquat. Sci., 44,* 1595-1602, 1987.

Galloway, J.N., H. Levy II, and P.S. Kasibhatla, Year 2020: Consequences of population, growth and development on deposition of oxidized nitrogen, *Ambio,* in press.

Gardetto, P. E., Nutrient limitation and the accumulation of added nitrogen and phosphorus in Sphagnum fuscum, M.Sc. thesis, Univ. of Minn., Minneapolis, 1987.

Gorham, E., Northern peatlands: Role in the carbon cycle and probable responses to climatic warming, *Ecol. Appl., 1,* 182-195, 1991.

Gorham, E., The biogeochemistry of northern peatlands and its possible responses to global warming, in *Biotic Feedbacks in the Global Climate System: Will the Warming Speed the Warming?,* edited by G. M. Woodwell, Oxford University Press, New York, 1993.

Grennfelt, P., and H. Hultberg, Effects of nitrogen deposition on the acidification of terrestrial and aquatic systems, *Water Air Soil Pollut., 30,* 945-963, 1986.

Gundersen, P., Nitrogen circulation in three spruce forests in Denmark, paper presented at the First European Symposium on Terrestrial Ecosystems, Florence, Italy, May 20-24, 1991.

Harrison K., W. Broecker, and G. Bonani, A strategy for estimating the impact of CO_2 fertilization on soil carbon storage, *Global Biogeochemical Cycles, 7,* 69-80, 1993.

Hauhs, M., Lange Bramke: An ecosystem study of a forested watershed, in *Acidic Precipitation,* vol. 1, *Case Studies,* edited by D. C. Adriano and M. Havas, pp. 275-305, Springer-Verlag, New York, 1990.

Hauhs, M., K. Rost-Siebert, G. Raben, T. Paces, and B. Vigerust, Summary of European data, in *The Role of Nitrogen in the Acidification of Soils and Surface Waters,* edited by J. L. Malanchuk and J. Nilsson, pp 5-1 to 5-37, Nordic Council of Ministers, Stockholm, 1989.

Hemond, H.F., The nitrogen budget of Thoreau's Bog, *Ecology, 64,* 99-109, 1983.

Henriksen, A., and D. F. Brakke, Increasing contributions of nitrogen to the acidity of surface waters in Norway, *Water Air Soil Pollut., 42,* 183-201, 1988.

Hicks, B. B., Overview of deposition processes, in *The Role of Nitrogen in the Acidification of Soils and Surface Waters,* edited by J. L. Malanchuk and J. Nilsson, pp. 3-1 to 3-21, Nordic Council of Ministers, Stockholm, 1989.

Hornung, M., ENCORE: European network of catchments organized for research on ecosystems, in *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* edited by L. Rasmussen et al., pp. 37-45, Commission of European Communities, Brussels, 1993.

Houghton, J., G. Jenkins, and J. Ephrams (Eds.), *Climate Change: The IPCC Scientific Assessment,* 358 pp., Cambridge University Press, New York, 1990.

Howarth, R. W., Eutrophication and airborne pollutants, in *Exposure and Effects of Airborne Contaminants,* Great Waters Program, U.S. Congress, Washington D. C., in press.

Hultberg, H., B.I. Anderson, and F. Moldan, The covered catchment - an experimental approach to reversal of acidification in a forest ecosystem, in *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* edited by L. Rasmussen et al., pp. 46-54, Commission of European Communities, Brussels, 1993.

Hunt, H.W., E.R. Ingham, D.C. Coleman, E.T. Elliott, and C.P.P. Reid, Nitrogen limitation of production and decomposition in prairie, mountain meadow and pine forest, *Ecology, 69,* 1009-1016, 1988.

Husar, R., Emissions of sulfur dioxide and nitrogen oxides and trends for eastern North America, Ch. 2 in *Acid Deposition: Long-Term Trends,* National Academy Press, Washington, 1986.

Iwatsubo, G., and Y. Nagayama, Effects of sewage-water spraying on the mineral cycling in a forest ecosystem, in *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* edited by L. Rasmussen et al., pp. 78-85, Commission of European Communities, Brussels, 1993.

Jenny, H., and S. P. Raychaudhuri, in *Effect of Climate and Cultivation on Nitrogen and Organic Matter Reserves in Indian Soils,* Indian Council of Agricultural Research, New Delhi, 126 pp., 1960.

Jenny, H., F. Bingham, and B. Padilla-Saravia, Nitrogen and organic matter contents of equatorial soils of Colombia, South America, *Soil Sci., 66,* 173-186, 1948.

Johannes, A. H., Y. L. Chan, K. Dackson, and T. Sulecki, Modeling of throughfall chemistry and indirect measurement of dry deposition, *Water Air Soil Pollut., 30,* 211-216, 1986.

Johnson, D.W., Nitrogen retention in forest soils, *J. Environ. Qual., 21,* 1-12, 1992.

Johnson, D. W., and S. E. Lindberg, *Atmospheric Deposition and Forest Nutrient Cycling,* 707 pp., Springer-Verlag, New York, 1992.

Johnson, D.W., M.S. Cresser, S.I. Nilsson, J. Turner, B. Ulrich, D. Binkley, and D.W. Cole, Soil changes in forest ecosystems: Evidence for and probable causes, *Proc. R. Soc. Edinburgh, Ser. B, 97,* 81-116, 1991.

Kauppi, P. E., K. Mielikainen, and K. Kuusela, Biomass and carbon budget of European Forests, 1971-1990, *Science, 256,* 70-74, 1992.

Keeling, R. F., and S. R. Shertz, Seasonal and interannual variations in atmospheric oxygen and implications for the global carbon cycle, *Nature, 358,* 723-727, 1992.

Kelly, C.A., J.W.M. Rudd, and D.W. Schindler, Lake acidification and nitric acid: Future considerations, *Water Air Soil Pollut., 50,* 49-61, 1990.

Legendre, L., and M. Gosselin, New production and export of organic matter to the deep ocean: Consequences of some recent discoveries, *Limnol. Oceanogr., 34,* 1374-1380, 1989.

Likens, G.E., *The Ecosystem Approach: Its Use and Abuse,* 166 pp. Ecology Institute, Oldenburg, Germany, 1992.

Likens, G. E., F. H. Bormann, R. S. Pierce, J. S. Eaton, and N. M. Johnson, *Biogeochemistry of a Forested Ecosystem,* Springer-Verlag, New York, 1977.

Likens, G.E., F.H. Bormann, and N.M. Johnson, Interactions between major biogeochemical cycles in terrestrial ecosystems, in *Some Perspectives of the Major Biogeochemical Cycles,* SCOPE 17, edited by G. E. Likens, pp. 93-112, John Wiley, New York, 1981.

Lindberg, S. E., G M. Lovett, D.D. Richter, and D. W. Johnson, Atmospheric deposition and canopy interactions of major ions in a forest, *Science, 231,* 141-145, 1986.

Lovett, G. M., W.A. Reiners, and R. K. Olson, Cloud droplet deposition in subalpine balsam fir forests: Hydrological and chemical inputs, *Science, 218,* 1303-1304, 1982.

Malanchuk, J. L., and J. Nilsson (Eds.), *The Role of Nitrogen in the Acidification of Soils and Surface Waters,* Nordic Council of Ministers, Stockholm, 1989.

Malmer, N., and E. Holm, Variation in the C/N quotient of peat in relation to decomposition rate and age determination with Pb 210, *Oikos, 43,* 171-182, 1984.

McGuire, A.D., J.M. Melillo, L.A. Joyce, D.W. Kicklighter, A. L. Grace, B. Moore III, and C.J. Vorosmarty, Interactions between carbon and nitrogen dynamics in estimating net primary productivity for potential vegetation in North America, *Global Biogeochem. Cycles, 6,* 101-124, 1992.

Mitchell, H. L., and R. F. Chandler, Jr., The nitrogen nutrition and growth of certain deciduous trees of northeastern United States, *Black Rock For. Bull. No. 11,* 94 pp., Cornwall-on-the-Hudson, New York, 1939.

Monk, C.D., and F.P. Day, Jr., Biomass, primary production and selected nutrient budgets for an undisturbed hardwood watershed, in *Forest Hydrology and Ecology at Coweeta,* edited by W.T. Swank and D.A. Crossley, Jr., pp. 151-159, Springer-Verlag, New York, 1988.

Nilsson, J., and P. Grennfelt (Eds.), Critical loads for sulphur and nitrogen, Miljorapport 1988:15, Nordic Council of Ministers, Copenhagen, 1988.

Norton, S.A., J.S. Kahl, I.J. Fernandez, T.A. Haines, and J. Lee, Effects of the addition of

$(NH_4)_2SO_4$ to a whole catchment at Bear Brook Watershed, Maine, U.S.A., in *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* edited by L. Rasmussen et al., pp. 55-63, Commission of European Communities, Brussels, 1993.

Owens, N.J.P., J.N. Galloway, and R.A. Duce, Episodic atmospheric nitrogen deposition to oligotrophic oceans, *Nature, 357,* 397-399, 1992.

Pastor, J., and J.G. Bockheim, Distribution and cycling of nutrients in an aspen-mixed hardwood-spodosal ecosystem in northern Wisconsin, *Ecology, 65,* 339-353, 1984.

Peierls, B. L., N. F. Caraco, M. L. Pace, and J. J. Cole, Human influence on river nitrogen, *Nature, 350,* 386-387, 1991.

Peterson, B.J., and J.M. Melillo, The potential storage of carbon caused by eutrophication of the biosphere, *Tellus, Ser. B, 37,* 117-127, 1985.

Post, W.M., J. Pastor, P.J. Zinke, and A.G. Stangenberger, Global patterns of soil nitrogen storage, *Nature, 317,* 613-616, 1985.

Prospero, J. M., and D. L. Savoie, Effect of continental sources on nitrate concentration over the Pacific Ocean, *Nature, 339,* 687-689, 1989.

Quay , P. D., B. Tilbrook, and C. S. Wong, Oceanic uptake of fossil fuel CO_2: Carbon-13 evidence, *Science, 256,* 74-79, 1992.

Rasmussen, L., Study on acid deposition effects by manipulating forest ecosystems, *Rep. 24,* Air Pollut. Res., Commission of the European Communities, Brussels, 1990.

Rasmussen, L., T. Brydges, and P. Mathy (Eds.), *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* Commission of European Communities, Brussels, 1993.

Rastetter, E.B., M.G. Ryan, G.R. Shaver, J.M. Melillo, K.J. Nadelhoffer, J.E. Hobbie, and J.D. Aber, A general biogeochemical model describing the responses of the C and N cycles in terrestrial ecosystems to changes in CO2, climate and N deposition, *Tree Physiol., 9,* 101-126, 1991.

Reimers, C. E., R. A. Jahnke, and D. C. McCorkle, Carbon fluxes and burial rates over the continental slope and rise off central California with implications for the global carbon cycle, *Global Biogeochem. Cycles, 6,* 199-224, 1992.

Riley, J.L., Peat and peatland resources of northeastern Ontario, *Open-File Rep., 5631,* 261 pp., 1987.

Riley, J.L., and L. Michaud, Peat and peatland resources of northwestern Ontario, *Ont. Geol. Surv. Miscell. Pap., 144,* 175 pp., 1989.

Robertson, J. E., and A. J. Watson, Thermal skin effect of the surface ocean and its implications for CO_2 uptake, *Nature, 358,* 738-740, 1992.

Rochefort L., D.H. Vitt, and S.E. Bayley, Growth, production and decomposition dynamics of Sphagnum under natural and experimentally acidified conditions, *Ecology, 71,* 1986-2000, 1990.

Rosenberg, R., R. Elmgren, S. Fleischer, P. Jonsson, G. Persson, and H. Dahlin, Marine eutrophication case studies in Sweden, *Ambio, 19,* 102-108, 1990.

Rudd, J.W.M., C.A. Kelly, D.W. Schindler, and M.A. Turner, A comparison of the acidification efficiencies of nitric and sulfuric acids by two whole-lake acidification experiments, *Limnol. Oceanogr., 35,* 663-679, 1990.

Sarmiento, J.L., and E.T. Sundquist, Revised budget for the oceanic uptake of anthropogenic carbon dioxide, *Nature, 356,* 589-592, 1992.

Schindler, D.W., R.W. Newbury, K.G. Beaty, and P. Campbell, Natural water and chemical budgets for a small Precambrian lake basin in central Canada, *J. Fish. Res. Board Can. 33,* 2526-2543, 1976.

Schindler, D.W. et al., *Atmosphere-Biosphere Interactions: Toward a Better Understanding of the Ecological Consequences of Fossil Fuel Combustion,* National Academy Press, National Academy of Science, Washington, D. C., 1981.

Schindler, D.W., R.W. Newbury, K.G. Beaty, J. Prokopowich, T. Ruszczynski, and J.A. Dalton, Effects of a windstorm and forest fire on chemical losses from forested watersheds and on the quality of receiving streams, *Can. J. Fish. Aquat. Sci., 37,* 328-334, 1980.

Schlesinger, W.H., *Biogeochemistry: An Analysis of Global Change,* Academic, San Diego, Calif., 1991.

Schlesinger, W.H., and A.E. Hartley, A global budget for atmospheric NH_3, *Biogeochemistry, 15,* 191-211, 1991.

Schulze, E.-D., Air pollution and forest decline in a spruce (Picea abies) forest, *Science, 244,* 776-783, 1989.

Schulze, E.D., and P.H. Freer-Smith, An evaluation of forest decline based on field observations focussed on Norway spruce, Picea abies, *Proc. R. Soc. Edinburgh, Ser. B, 97,* 155-168, 1991.

Seitzinger, S.P., Denitrification in freshwater and coastal marine ecosystems: Ecological and geochemical significance, *Limnol. Oceanogr., 33,* 702-724, 1988.

Sharp, J.H., Review of carbon, nitrogen, and phosphorus biogeochemistry, *U.S. Natl. Rep. Int. Union Geod. Geophys. 1987-1990, Rev. Geophys., 29,* 648-657, 1991.

Shaver, G.R., and F.S. Chapin III, Response to fertilization by various plant growth forms in an Alaskan tundra: nutrient accumulation and growth, *Ecology, 61,* 662-675, 1980.

Shaver, G.R., and F.S. Chapin III, Effect of NPK fertilization on production and biomass of Alaskan tussock tundra, *Arct. Alp. Res., 18,* 261-268, 1986.

Shaver, G.R., and F.S. Chapin III, Production/biomass relationships and element cycling in contrasting arctic vegetation types, *Ecol. Monogr., 61,* 1-31, 1991.

Shaver, G.R., W.D. Billings, F.S. Capin III, A.E. Giblin, K.J. Knadelhoffer, W.C. Oechel, and E.B. Rastetter, Global change and the carbon balance of arctic ecosystems, *Bioscience, 42,* 433-441, 1992.

Sirois, A., and L. A. Barrie, An estimate of the importance of dry deposition as a pathway of acidic substances from the atmosphere to the biosphere in eastern Canada, *Tellus, Ser. B, 40,* 59-80, 1988.

Stoddard, J.L., and P.S. Murdoch, Catskill Mountains, in *Acidic Deposition and Aquatic Ecosystems: Regional Case Studies,* edited by D.C. Charles, pp. 237-271, Springer-Verlag, New York, 1991.

Swank, W.T, and J.B. Waide, Characterization of baseline precipitation and stream chemistry and nutrient budgets for control watersheds, in *Forest Hydrology and Ecology at Coweeta,* edited by W.T. Swank and D.A. Crossley, Jr, pp. 57-79, Springer-Verlag, New York, 1988.

Tamm, C. O., *Nitrogen in Terrestrial Ecosystems,* 116 pp., Springer-Verlag, New York, 1990.

Tamm, C. O., and P. Hogberg, Critical loads of nitrogen as derived from long-term experiments with annual additions of nitrogen to forest ecosystems, in *Experimental Manipulations of Biota and Biogeochemical Cycling in Ecosystems,* edited by L. Rasmussen, et al., pp. 272-276, Commission of European Communities, Brussels, 1993.

Tans, P.P., I.Y. Fung, and T. Takahashi, Observational constraints on the global atmospheric CO_2 budget, *Science, 247,* 1431-1438, 1990.

Urban, N., and S. E. Bayley, The relative importance of denitrification and nitrate assimilation in midcontinental bogs, *Limnol. Oceanogr., 33,* 1611-1617, 1988.

Urban, N.R., and S.J. Eisenreich, Nitrogen cycling in a forested Minnesota bog, *Can. J. Bot., 66,* 435-449, 1988.

Van Cleve, K., and V. Alexander, Nitrogen cycling in tundra and boreal ecosystems, in Terrestrial Nitrogen Cycles, edited by F.E. Clark and T. Rosswall, *Ecol. Bull., 33,* 375-404, 1981

Van Cleve, K., W. C. Oechel, and J. L. Hom, Response of black spruce (Picea mariana) ecosystems to soil temperature modification in interior Alaska, *Can. J. For. Res., 20,* 1530-1535, 1990.

Verhoeven, J. T. A., *Bogs and Fens in the Netherlands: Vegetation, History, Nutrient Dynamics and Conservation,* 491 pp., Kluwer, Dordrecht, The Netherlands, 1992.

Vitousek, P. M., and R. W. Howarth, Nitrogen limitation on land and in the sea: How can it occur?, *Biogeochemistry, 13,* 87-115, 1991.

Vitousek P.M., T.Fahey, D.W. Johnson, and M.J. Swift, Element interactions in forest ecosystems: succession, allometry and input-output budgets, *Biogeochemistry, 5,* 7-34, 1988.

Walsh, J.J., G.T. Rowe, R.L. Iverson, and C.P. McRoy, Biological export of shelf carbon is a sink of the global CO_2 cycle, *Nature, 291,* 196-201, 1981.

Walsh, J. J., Importance of continental margins in the marine biogeochemical cycling of carbon and nitrogen, *Nature, 350,* 53-55, 1992.

Watson, A., Conveying that sinking feeling, *Nature, 356,* 561-562, 1992.

Weathers, K. C., et al., Cloudwater chemistry from ten sites in North America, *Environ. Sci. Technol., 22,* 1018-1026, 1988.

Weathers, K.C., G.M. Lovett, and G.E. Likens, The influence of a forest edge on cloud deposition, in *Precipitation Scavenging and Atmosphere-Surface Exchange.* vol. 3,-- *The*

Summers Volume: Applications and Appraisals, edited by S.E. Schwartz and W.G.N. Slimm, pp. 1415-1423, Hemisphere Publishing Corporation, Washington D.C. 1992.

Wollast, R., Interactions in esturaries and coastal waters, in *The Major Biogeochemical Cycles and their Interactions, SCOPE 21,* edited by B. Bolin and R.B. Cook, pp. 385-407, John Wiley, New York, 1983.

Woodwell, G. M. (Ed.), *The Role of Terrestrial Vegetation in the Global Carbon Cycle,* John Wiley, New York, 1984.

Wright R.F., and N.B. Dise, Integration, The NITREX Project (Nitrogen Saturation Experiments), *Report XX,* edited by N.B. Dise and R.F. Wright, pp. 74-76, Air Pollut. Res., Commission of European Communities, Brussels, 1992.

Wright, R.F., E. Lotse, and A. Semb, Experimental acidification of alpine catchments at Sogndal, Norway: Results after 8 years, *Water Air Soil Pollut.,* in press, 1993.

S.E. Bayley, Department of Botany, B414 Biological Sciences Center, University of Alberta, Edmonton, Alberta, Canada T6G 2E9

D.W. Schindler, Department of Zoology and Botany, CW312 Biological Sciences Center, University of Alberta, Edmonton, Alberta, Canada T6G 2E9

(Received March 5, 1993;
revised July 16, 1993;
accepted September 8, 1993.)