Hydrology and Earth System Sciences, 5(3), *299–310* (2001) © EGS

Hydrology & Earth System Sciences

Trends in nitrogen deposition and leaching in acid-sensitive streams in Europe

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Abstract

Long-term records of nitrogen in deposition and streamwater were analysed at 30 sites covering major acid sensitive regions in Europe. Large regions of Europe have received high inputs of inorganic nitrogen for the past 20-30 years, with an approximate 20% decline in central and northern Europe during the late 1990s. Nitrate concentrations in streamwaters are related to the amount of N deposition. All sites with less than 10 kgN ha^{-1} yr^{-1} deposition have low concentrations of nitrate in streamwater, whereas all sites receiving > 25 kgN ha^{-1} yr^{-1} have elevated concentrations. Very few of the sites exhibit significant trends in nitrate concentrations; similar analyses on other datasets also show few significant trends. Nitrogen saturation is thus a process requiring many decades, at least at levels of N deposition typical for Europe. Declines in nitrate concentrations at a few sites may reflect recent declines in N deposition. The overall lack of significant trends in nitrate concentrations in streams in Europe may be the result of two opposing factors. Continued high deposition of nitrogen (above the 10 kgN ha^{-1} yr^{-1} threshold) should tend to increase N saturation and give increased nitrate concentrations in run-off, whereas the decline in N deposition over the past 5–10 years in large parts of Europe should give decreased nitrate concentrations in run-off. Short and long-term variations in climate affect nitrate concentrations in streamwater and, thus, contribute "noise" which masks long-term trends. Empirical data for geographic pattern and long-term trends in response of surface waters to changes in N deposition set the premises for predicting future contributions of nitrate to acidification of soils and surface waters. Quantification of processes governing nitrogen retention and loss in semi-natural terrestrial ecosystems is a scientific challenge of increasing importance.

Keywords: Europe, acid deposition, nitrogen, saturation, recovery, water

Introduction

During the 1900s large regions of Europe received elevated deposition of nitrogen (N) compounds. Emissions of oxidised N species from combustion of fossil fuels and emissions of reduced N compounds from agriculture increased dramatically in Europe during the 1900s to reach peak levels by about 1980. Since then emissions have levelled off and decreased slightly in most countries (Tarrason and Schaug, 2000).

Excess N deposition has long been viewed as a threat to the nutrient balance and health of forests and semi-natural terrestrial ecosystems. In the absence of significant N deposition, N is usually the growth-limiting nutrient in these ecosystems (Tamm, 1991). Chronic excess N deposition can lead to N saturation, defined by Aber *et al.*, 1989) as "the availability of ammonium (NH_4) and nitrate (NO_3) in excess of total combined plant and microbial nutritional demand". By this definition N saturation is manifest by increased leaching of inorganic N (generally NO_3) below the rooting zone. Since NO_3 is a strong acid anion, increased leaching of NO_3 enhances acidification of soils and surface waters. Increased concentrations of inorganic N in run-off (streamwater) thus indicate N saturation of terrestrial ecosystems, assuming that there are no significant sources of N in the catchment (such as fertilisers, municipal and industrial wastewater).

Long-term records of deposition and run-off of N are available for streamwaters in several acid-sensitive areas in Europe. Many of these data have been collected as part of national monitoring programmes and are included in international co-operative programmes under the auspices of the UN-ECE Convention on Long Range Transboundary Air Pollution, such as EMEP (Co-operative Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe), ICP-Waters (International Co-operative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes) (Kvæven *et al.*, 2001) and ICP Integrated Monitoring (Lundin *et al.*, 2001). Trends in deposition and run-off from such data provide a basis for assessing both the role of N in recovery of surface waters from acidification and the status and change in N saturation status of terrestrial ecosystems. EMEP (Barrett *et al.*, 2000a) and ICP-Waters (Stoddard *et al.*, 2001) have previously conducted such analyses based on emission and deposition data and surface water data, respectively, from the 1980s and 1990s.

The European research project RECOVER:2010 is designed to assess the impact of acid deposition on sensitive freshwater ecosystems and to predict the recovery by the year 2010 (Ferrier *et al.*, 2001). One objective of RECOVER:2010 is to analyse systematically long-term records of deposition and surface water chemistry at acid-sensitive sites characteristic of the major types of acidified waters in Europe. RECOVER:2010 includes sites in Scandinavia (Norway, Sweden and Finland), the British Isles (UK), central Europe (Germany, Czech Republic, and Slovakia) and the southern Alps (Italy). Here we present the long-term trends in inorganic N concentrations in deposition and streamwater at 30 stream sites in Europe (Table 1, Fig. 1). In a companion paper, Prechtel *et al.* (2001) present trends in sulphur (S) for these sites. Lakes are not addressed here, because of the complicating factor of in-lake retention of N. Trends in N concentrations in Scandinavian lakes are presented by Skjelkvåle *et al.* (2001a), trends in Czech lakes are presented by Kopácek *et al.*, (1998) and trends in four Italian lakes are presented by Rogora *et al.* (2001).

Materials and methods

SITES AND RUN-OFF DATA

All of the 30 sites are running waters with catchments ranging in size from a few hectares (Gårdsjön F1 in Sweden) to over 100 km^2 (Cannobino River, Italy) (Table 1). The four sites in Norway (Birkenes, Storgama, Langtjern and

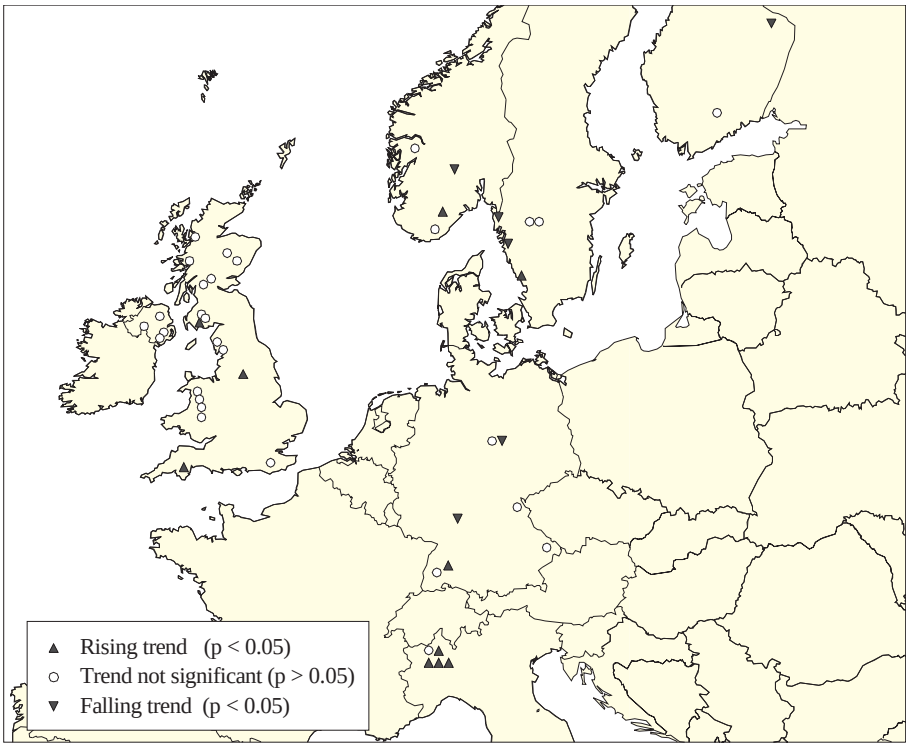

Fig. 1. *Map of Europe showing RECOVER:2010 stream sites used in analysis of long-term trends in N. Triangles up indicate significant increasing trends in NO_3 concentrations. Triangles down indicate significant decreasing trends. Circles indicate sites with no significant trend.*

Table 1. Location and characteristics of streamwater sites included

Catchments	*Lat. N*	*Long. E*	*Altitude m a.s.l.*	*Area km²*	*Vegetation*	*Bedrock geology*	*Soil*	*Reference*
Germany								
Lange Bramke	51°52'	10°26'	535-700	0.76	Norway spruce	sandstone	spododystric cambisols	Alewell *et al.*, 2000
Lehstenbach	50°09'	11°52'	694-871	4.2	Norway spruce	granite deeply weathered	dystric cambisols and podzols	——"——
Markungsgraben	48°57'	13°25'	890-1355	1.1	Norway spruce and mixed woodland	granite, gneiss	cambisols	——"——
Metzenbach	49°54'	9°26'	385-586	2.4	Deciduous forest, mostly beech	red sandstone	cambisols	——"——
Schluchsee	47°49'	8°06'	1150-1290	0.11	Norway spruce	granite	haplic podzol	——"——
Villingen	48°03'	8°22'	810-945	0.463	Norway spruce	sandstone	dystric cambisols and dystric planosols	——"——
Italy								
Cannobino	46°04'	8°42'	193-2193	110.4	Mixed deciduous	orthogneiss and micaschist (60%)		Boggero *et al.* 1996
Pellino	45°47'	8°04'	290-942	17.5	Mixed deciduous	white granite and granodiorite (50%), orthogneisses and micaschists (30%)		——"——
Pellesino	45°48'	8°04'	290-1491	17.5	Mixed deciduous	white granite and granodiorite (80%)		——"——
Norway								
Birkenes	58°23'	8°15'	200-300	0.41	Mixed coniferous	granite	podzols	Lydersen 1994
Storgama	59°01'	8°32'	580-690	0.6	Individual trees, heathland	granite	podzols	——"——
Langtjern	60°22'	9°39'	510-750	4.8	Mixed coniferous	gneiss	podzols	——"——
Kaarvatn	62°47'	8°53'	200-1375	25	alpine heathland	gneiss, quartzite	podsols	——"——
Sweden								
Gårdsjön F1	58°03'	12°01'	113-170	0.037	Norway spruce	gneiss	Podzols	Moldan *et al.* 2001b
Lommabäcken Nedre	58°42'	14°38'	190-230	1.04	pine/spruce, mire	granite	thin podzols, peat	——"——
Ringsmobäcken	58°59'	11°45'	180-245	1.4	pine/spruce, heathland, wet forrest	gneiss	thin podzols, peat	——"——
Pipbäcken Nedre	57°03'	12°47'	75-165	0.93	spruce/pine/decid, mire	gneiss	moraine, podzols, peat	——"——
Finland								
Hietajärvi	63°10'	30°43'	165-214	4.64	pine/spruce/decid	granitoids	histisols, podzols	Moldan *et al.* 2001b
Valkea-Kotinen	61°14'	25°04'	150-190	0.30	Norway spruce and deciduous	gneiss	dystric cambisols, transitions to podzols	——"——
United Kingdom								
Allt a'Mharcaidh	57°07'	3°50'	325-1111	9.98	98% moorland, 2% pine	granite	alpine & peaty podsols, blanket peat	Monteith and Evans, 2000
Allt na Coire nan Con			10-756	7.9	42% conifers, 54% moorland	schists, gneiss	peaty podsols, peaty gleys, peats	——"——
Dargall Lane			225-716	2.1	100% moorland	greywackes, shales, mudstones	podsols, peaty gleys, blanket peat	——"——
River Etherow			280-633	13	100% moorland	millstone grit	peaty podsols, blanket peat	——"——
Old Lodge			94-198	2.2	70% heathland, 15% deciduous, 15% conifers	sandstone	podsols	——"——
Narrator Brook			225-456	2.53	98% grassland, 2% deciduous	granite	iron pan stagno-podsols, brown podsols	——"——
Afon Hafren			355-690	3.58	50% conifers, 50% moorland	sedimentary	podsols, peat	——"——
Afon Gwy			440-730	3.89	100% moorland	sedimentary	podsols, peat	——"——
Beagh's Burn			150-397	3.03	100% moorland	schist	blanket peat	——"——
Bencrom River			140-700	2.16	100% moorland	granite	blanket peat	——"——
Coneyglen Burn			230-562	13.11	95% moorland, 5% conifers	schist	blanket peat	——"——

Kaarvatn) are all included in the Norwegian monitoring programme for long range transboundary air pollution (Skjelkvåle and Tørseth, 2001) and are described by Lydersen (1994). Monitoring began in the 1970s and is reported annually (Aas *et al*., 2000; SFT, 2000). The Swedish site at Gårdsjön is part of several research projects (NITREX: Moldan and Wright, 1998; roof catchment: Moldan *et al*., 2001a) as well as a monitoring site included in ICP Waters and ICP Integrated Monitoring. The other three Swedish sites are part of the national monitoring programme and operated by the Swedish University of Agricultural Sciences. The Finnish sites are operated by the Finnish Meteorological Institute. Further details on these sites are given in Moldan *et al.* (2001b). The UK sites are the 11 streams included in the UK Acid Waters Monitoring Network (AWMN) (Monteith and Evans, 2000) and have been sampled since 1988. The German sites are included in the ICP-Waters programme and are described by Alewell *et al*. (2001), with methods given by LfW (1994) (Markungsgraben, Metzenbach and Lehstenbach) and Armbruster (1998) (Schluchsee and Villingen). Hauhs (1990) describes the Lange Bramke catchment (Harz, Germany). The three Italian rivers (Pellino, Pellesino and Cannobino) are also part of ICP-Waters (Mosello *et al*., 2000) and are described by Boggero *et al*. (1996).

The catchments of the 30 streams are in coniferous and deciduous forests or heathlands (moorlands), on bedrock and soils resistant to weathering (with the exception of the three Italian rivers) and with cambisols, podsols or peaty soils (Table 1). There are no major sources of pollution from within the catchments and, thus, they are well suited for examining the role of atmospheric deposition on water chemistry.

DEPOSITION DATA

Routine and systematic measurement of deposition of acidifying pollutants in Europe has been conducted since the 1950s. Measurement stations are operated as part of national monitoring programmes and since 1978 have been formally organised as EMEP as part of the UN-ECE Convention on Long-Range Transboundary Air Pollution. EMEP entails systematic measurement of wet and dry deposition of N compounds at > 50 sites in Europe (Barrett *et al.*, 2000a,b). EMEP data comprise both measured and regional deposition modelled by an emission-transport-deposition model. The modelled deposition has been used as the basis for negotiations of emissions protocols under the convention (Bull *et al*., 2001).

Results reported here are from three sources: bulk precipitation collected at the sites (six sites in Germany, four sites in Norway and one site in Sweden), measured deposition at EMEP or other stations located in the same region as the streamwater sites (Barrett *et al*., 2000a) and modelled deposition by country (Tarrason and Schaug, 2000).

TREND ANALYSES

Trend analyses on deposition and streamwater data were carried out using the seasonal Kendall test. Details are given by Evans *et al*., 2001.

Results

DEPOSITION

Deposition of both oxidised and reduced N in Europe increased during the 1900s and plateaued in the 1970s and 1980s. Levels have decreased somewhat during the late 1990s in central and northern Europe (Tarrason and Schaug, 2000) (Fig. 2a,b). Trend analyses on the EMEP data conducted by Barrett *et al*. (2000a,b) indicate approximately 20% decline in deposition of N in central Germany and southern Scandinavia during the period 1989-1998. This is interpreted as a response to declining emissions of N compounds during the 1990s.

Measured wet deposition of N compounds at EMEP sites, and sites included here from RECOVER:2010, show relatively large year-to-year variations (Fig. 2b), probably mainly due to natural variations in meteorological conditions such as the amount of precipitation. This "noise" in the record means that trends in N deposition must be relatively large before they become statistically significant. This probably explains why there are no significant trends in N deposition during the 1990s at many of the EMEP and RECOVER sites.

STREAMWATER

Mean concentrations of NO_3 in streamwater were lowest in northern Scandinavia and northwestern UK and highest in central Europe (Table 2). This is the same geographic pattern as N deposition. Ammonium concentrations at all these sites are very low, often below the analytical detection limit. (Detection limit for NH_4 varies between laboratories, and has changed over time, but is generally 0.5-1.0 µeq L^{-1}).

Mean concentrations of NO_3 in streamwater at the Scandinavian catchments were < 10 µeq L^{-1} with a clear gradient of increasing concentrations from north to south (Table 2). The records begin in the 1970s at the Norwegian sites, the mid-1980s at the Swedish sites and 1988 at the Finnish Sites. Only the sites Pipbäcken Nedre in

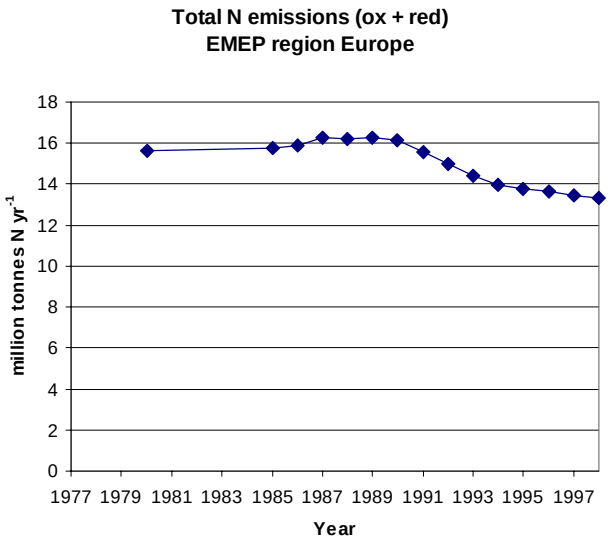

Fig. 2a. *Emissions of N compounds to the atmosphere (oxidised plus reduced) in the EMEP region of Europe (data from Tarrason and Schaug, 2000). The data indicate a 20% reduction in emissions during the 1990s.*

southernmost Sweden and Storgama and Birkenes in southernmost Norway exhibited signs of elevated NO_3 concentrations in streamwater (Figs. 3a,b).

The 11 stream sites in the UK also showed pronounced geographic gradient, with low concentrations (< 5 µeq L^{-1}) at sites on the periphery (northern Scotland, western Northern Ireland, southwestern and southeastern England) and higher concentrations (> 10 µeq L^{-1}) at sites in southern Scotland, central England, central Wales and southernmost Northern Ireland (Table 2, Fig. 4). Again the highest NO_3 concentrations are found at sites receiving highest N deposition, and in the most polluted regions approach concentrations observed in central Europe.

With the exception of Villingen in the Black Forest, all the German sites had very high concentrations of NO_3 in streamwater with mean concentrations for the 12 year period 1987–99 exceeding 50 µeq L^{-1} at Lehstenbach (Fichtelgebirge) and Markungsgraben (Bavarian Forest) (Table 2, Fig. 5). These sites also receive very high N deposition (> 25 kg ha^{-1} yr^{-1}). The three streams in northern Italy all had high concentrations of NO_3 (> 40 µeq L^{-1}) (Table 2, Fig. 6).

Only nine of the 30 sites had significant trends over time in NO_3 concentrations in streamwater (Table 2). All three Italian rivers had significant positive trends amounting to an increase of about 1-2% per year. Two sites in the UK (River Etherow and Narrator Brook) also had positive trends. The only sites with negative trends were Metzenbach (central Germany), Storgama (southern Norway) and Gårdsjön F1 (west-coast Sweden). The latter two sites had low mean concentrations of NO_3 (< 10 µeq L^{-1}) and the

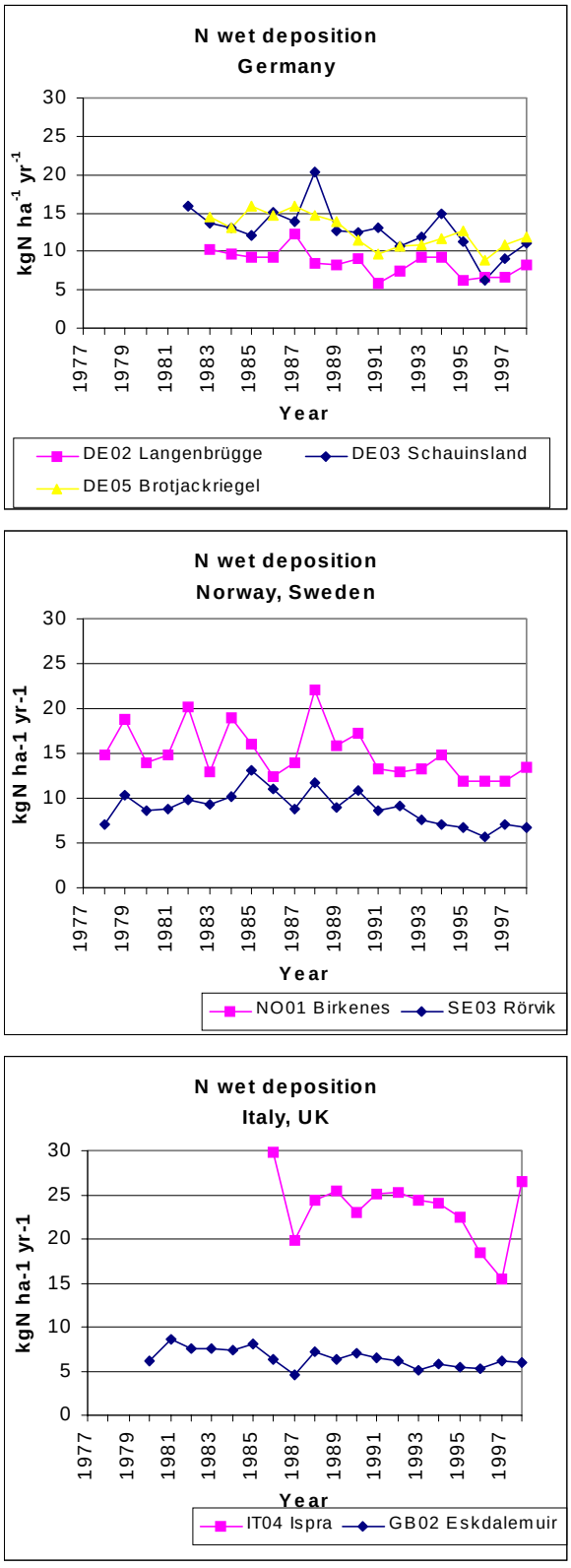

Fig. 2b. *Measured wet deposition of N (NO_3 plus NH_4) at several EMEP stations in Europe. For the 10-year period 1989-1998 none of the trends in annual deposition were significant at the p=0.05 level except for the site SE03 Rörvik in southern Sweden (data from EMEP Chemical Co-odinating Centre http://www-nilu.no/projects/ccc/ default.htm; trend analyses from Barrett* et al.*, 2000a).*

Table 2. Results of trend analyses for nitrate and ammonium concentrations in streamwater. Level of significance p< 0.05. NS=not significant. ISD= insufficient data. Test: Seasonal Kendall. Also shown are stages of nitrogen saturation based on nitrate concentrations in run-off by the criteria of Stoddard and Traaen (1995).

						μeq L^{-1} NO_3			μeq L^{-1} NH_4		Nitrogen saturation	
Catchment	*Start year-mo*	*End year-mo*	*Frequency*	*N*	*mean*	*trend*	*sign.*	*mean*	*trend*	*sign.*	*Stage*	*Year*
Germany												
Lange Bramke	1969-08	1999-12	weekly	937	38.9	NS[a]			ISD		1	1999
Lehstenbach	1987-04	1999-11	2/month	330	66.7	NS		2.7	NS		2	1999
Markungsgraben	1987-03	1999-11	2/month	289	114.9	NS[b]		2.1	NS		3	1995
Metzenbach	1987-02	1999-11	1/month	283	46.6	-1.6	0.004	2.5	0.2	0.005	2	1999
Schluchsee	1987-03	1996-10	1/week	509	31.3	NS		0.2	NS		2	1996
Villingen	1987-03	1996-10	1/week	504	4.8	NS		0.2	NS		1	1996
Italy												
Cannobino	1971-10	1999-11	1/month	300	44.2	0.5	0.003	1.8	-0.1	0.000	2	1999
Pellino	1984-01	1999-11	1/month	206	101.5	2.0[c]	0.002	1.0	0.0	0.009	3	1999
Pellesino	1986-01	1999-11	1/month	178	102.3	1.8	0.006	2.1	NS		3	1999
Norway												
Birkenes	1972-07	1999-12	1/week	2028	7.5	NS			ISD		0	1999
Storgama	1974-07	1999-12	1/week	1311	8.8	-0.1	0.064		ISD		0	1999
Langtjern	1972-09	1999-12	1/week	1489	1.6	NS			ISD		0	1999
Kaarvatn	1978-02	1999-12	1/week	1042	1.7	0.0	0.021		ISD		0	1999
Sweden												
Gårdsjön F1 Control	1979-10	2000-03	1/week	236	2.2	-0.1[d]	0.000	1.3	0.1	0.001	0	1999
Lommabäcken Nedre	1985-01	1999-12	1/month	159	2.7			1.6			0	1999
Ringsmobäcken	1985-01	1999-12	1/month	160	3.0			1.3			0	1999
Pipbäcken Nedre	1985-01	1999-12	1/month	160	9.3			1.7			1	1999
Finland												
Hietajärvi	1988-01	1998-12	1/month	117	2.5			4.7			0	1998
Valkea-Kotinen	1988-01	1998-12	1/month	128	1.5			0.9			0	1998
United Kingdom												
Allt a'Mharcaidh	1988-06	2000-03	1/month	142	1.5				ISD		0	1999
Allt na Coire nan Con	1988-08	2000-03	1/month	140	4.4				ISD		0	1999
Dargall Lane	1988-07	2000-03	1/month	141	10.7				ISD		1	1999
River Etherow	1988-07	2000-02	1/month	142	46.8	1.1	0.022		ISD		2	1999
Old Lodge	1991-04	2000-03	1/month	108	7.4				ISD		0	1999
Narrator Brook	1991-06	2000-03	1/month	106	7.0	0.3	0.032		ISD		1	1999
Afon Hafren	1988-08	2000-02	1/month	140	20.7				ISD		2	1999
Afon Gwy	1991-04	2000-02	1/month	108	10.0				ISD		1	1999
Beagh's Burn	1988-07	2000-03	1/month	141	3.3	NS			ISD		0	1999
Bencrom River	1988-07	2000-03	1/month	141	26.8	NS			ISD		1	1999
Coneyglen Burn	1990-08	2000-03	1/month	116	3.0				ISD		0	1999

[a] Before 1984 t=3.07, sig=0.013, after t=-1.74, sig=0.000
[b] NO_3: after 1995 t=6.5, sig=.000
[c] Before 1993 t=2.31, sig=0.000, after t=4.52, sig=0.000
[d] NO_3 strongly influenced by high con. 1983-85

negative trends are not significant with respect to streamwater acidity.

The seasonal pattern of NO_3 concentrations was quite pronounced with higher levels during the dormant seasons and lower levels during the growing season. At a few sites concentrations of NO_3 were moderate to high all year round. These seasonal patterns correspond to various stages of N saturation where stage 0 represents the pristine, unpolluted stage, and stage 3 represents the fully N saturated stage (Stoddard, 1994). Stoddard and Traaen (1995) suggested

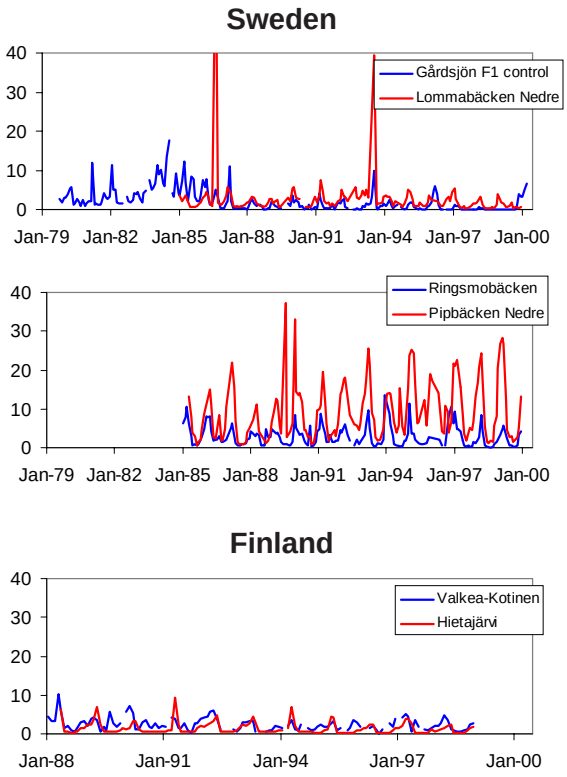

Fig. 3a. *Concentrations of NO_3 (µeq L^{-1}) in streamwater at four catchments in Sweden (upper 2 panels) and two catchments in Finland (bottom panel) over the period 1985-1999 (data from Moldan* et al.*, 2001b)*

criteria for classification of streams based on NO_3 concentrations during the growing and dormant seasons (Table 3, Fig. 7). Based on these criteria, 14 of the catchments were classed as stage 0 (Scandinavian and some UK sites), 7 as stage 1 (German and UK sites), 6 as stage 2 (German and UK sites) and 3 as stage 3 (German and Italian sites) (Table 2). At Markungsgraben the strong increase in concentrations and disappearance of seasonal pattern during the 1990s is probably due to bark beetle attack in the forest (Alewell *et al.*, 2001).

Only one of the sites (Lange Bramke) showed clear change in stage over the period of data. Lange Bramke moved from stage 1 in 1970 to stage 3 in 1985 but then back to stage 1 in 1999 (Fig. 8). These changes in stage, of course, parallel the general trends in NO_3 concentrations at Lange Bramke. Two adjacent catchments (Dicke Bramke and Steile Bramke) both also show the same decline in NO_3 concentrations beginning in about 1985 (Alewell *et al.*, 2001). These changes are not explained by changes in N deposition, climate or management practices.

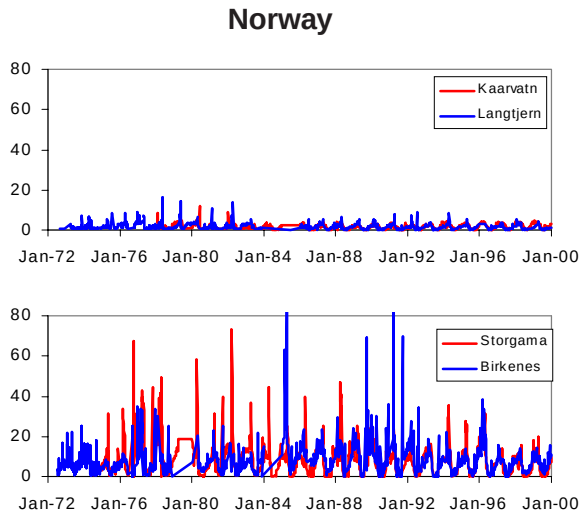

Fig. 3b. *Concentrations of NO_3 (µeq L^{-1}) in streamwater at four catchments in Norway over the period 1972-1999 (data from annual reports of the Norwegian monitoring programme; SFT, 2000).*

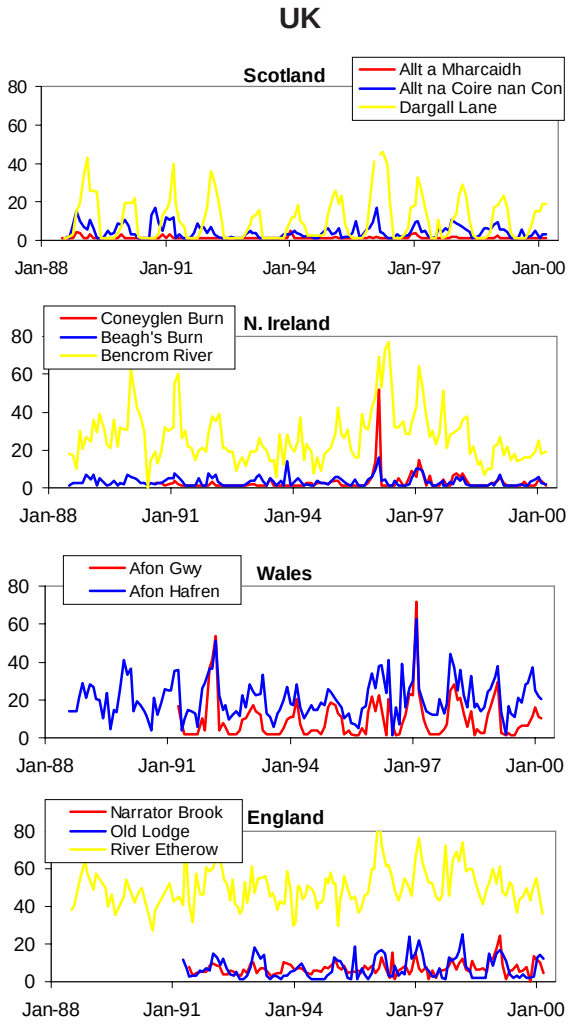

Fig. 4. *Concentrations of NO_3 (µeq L^{-1}) in streamwater at 11 catchments in the UK over the period 1988-1999 (data from Monteith and Evans, 2000; Evans and Monteith, 2001)*

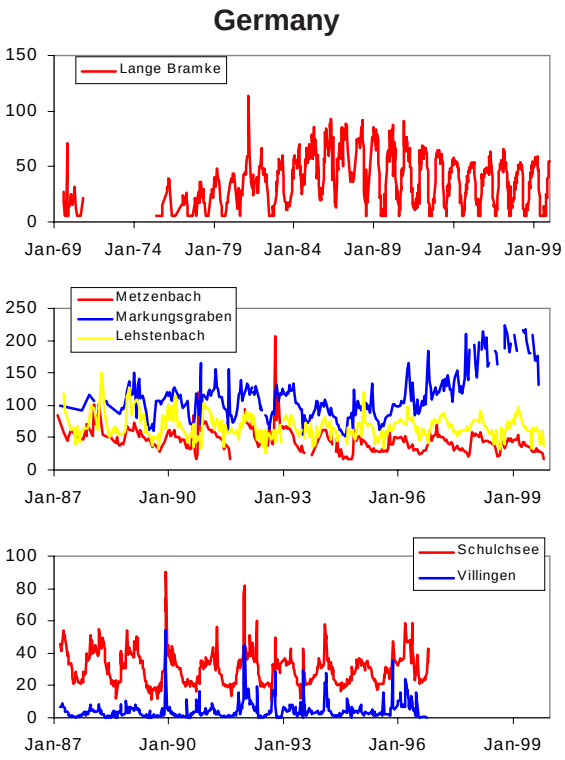

Fig. 5. *Concentrations of NO_3 (µeq L^{-1}) in streamwater at catchments in Germany. Upper panel: Lange Bramke north-central Germany (Harz Mountains). Middle panel: three catchments in central and southern Germany. Bottom panel: two catchments in southwestern Germany (Black Forest). Data from Alewell* et al., *2001).*

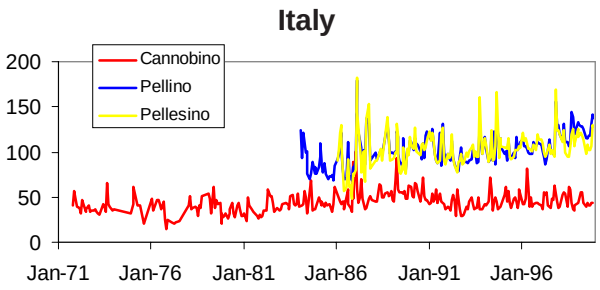

Fig. 6. Concentrations of NO_3 (µeq L^{-1}) in streamwater at three catchments in northern Italy over the period 1972 (or 1984 or 1986)-1999 (data from Rogora et al., *2001)*

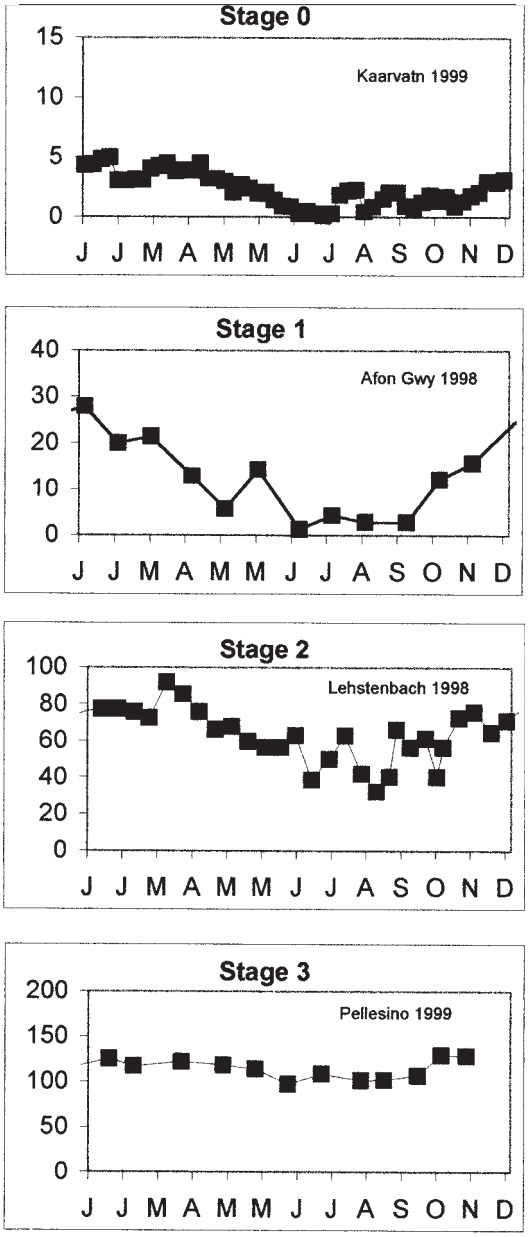

Fig. 7. *Seasonal patterns of NO_3 concentrations (µeq L^{-1}) in streamwater at four sites in Europe, illustrating the four stages of N saturation (after criteria of Stoddard and Traaen, 1995)*

Table 3. Criteria for stage of nitrogen saturation based on nitrate concentrations in run-off (Stoddard and Traaen, 1995)

Stage	*Lowest value months with conc. below 3 µeq L^{-1}*		*Highest value conc. µeq L^{-1}*	*months*
0	more than 3	and	less than 20	0
1	more than 3	and	more than 20	more than 3
2	1-3	and	less than 50	more than 3
3	0	and	less than 50	less than 3

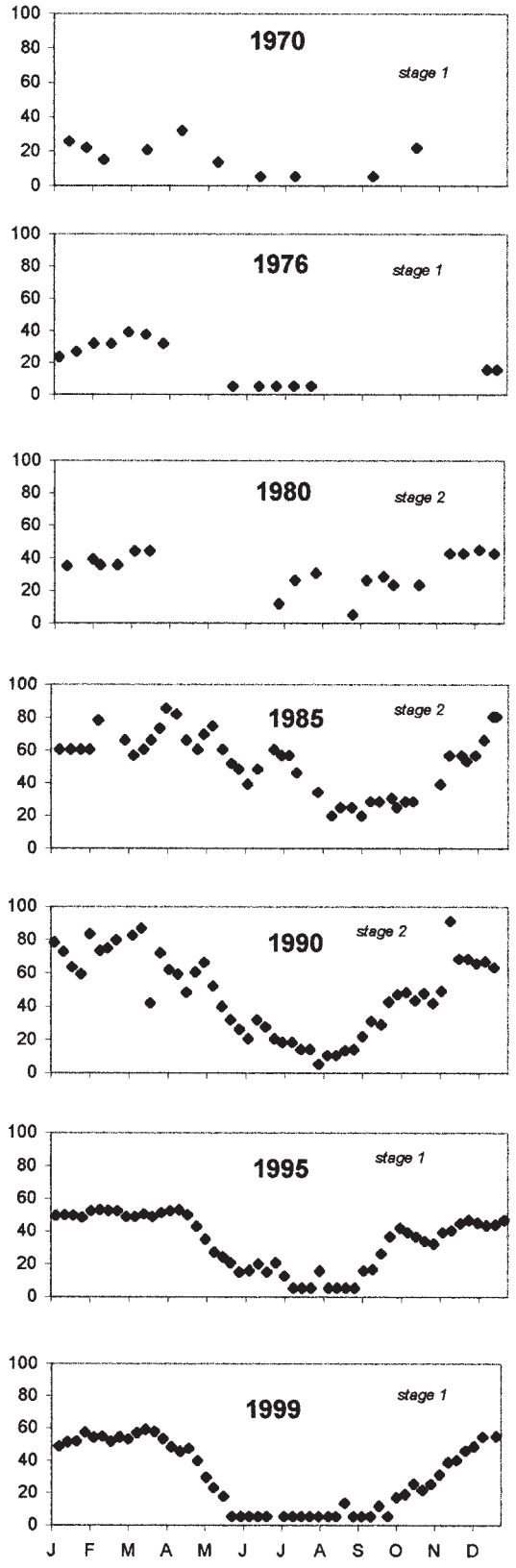

Fig. 8. *Seasonal patterns of NO_3 concentrations (µeq L^{-1}) in streamwater at Lange Bramke, Germany, at five year intervals over the period 1970 to 1999. Stage of N saturation (after criteria of Stoddard and Traaen, 1995) indicated.*

Discussion

The data from these 30 stream sites in Europe fit well into the general picture of N deposition and N concentrations in surface waters. Ammonium concentrations at all sites are very low, as is usually the case in most streamwaters (except, of course, those receiving NH_4 pollution from direct sources within the catchment). Nitrate concentrations are clearly related to N deposition; average concentrations are below 5 µeq L^{-1} in areas receiving N deposition below about 10 kgN ha^{-1} yr^{-1}, variable in areas receiving 10-25 kgN ha^{-1} yr^{-1} and above 10 µeq L^{-1} at sites receiving more than 25 kgN ha^{-1} yr^{-1} (Fig. 9). These same two thresholds emerge from several previous data compilations such as those from forested sites in Europe (the ENSF or ECOFEE dataset) (Dise and Wright, 1995), the ICP-Waters sites in Europe and North America (Skjelkvåle *et al*., 2001b), lakes in Norway (Skjelkvåle *et al*., 1996) and upland surface waters in the UK (Allott *et al*., 1995). Chronic N deposition at levels above about 10 kg N ha^{-1} yr^{-1} is thus a necessary but not sufficient factor for elevated concentrations of NO_3 in surface waters.

The general absence of trends in NO_3 concentrations in the 30 streams addressed here is consistent with several other analyses of N trends in surface waters in Europe. Skjelkvåle *et al*. (2001c) conducted trend analyses for the period 1989–98 on the ICP-Waters data set. They found very few significant trends in NO_3 concentrations at these sites. An earlier analysis of the ICP data by Stoddard *et al*. (1999) showed significant trends in the region north/central Europe with increases in the 1980s followed by decreases in 1990–95.

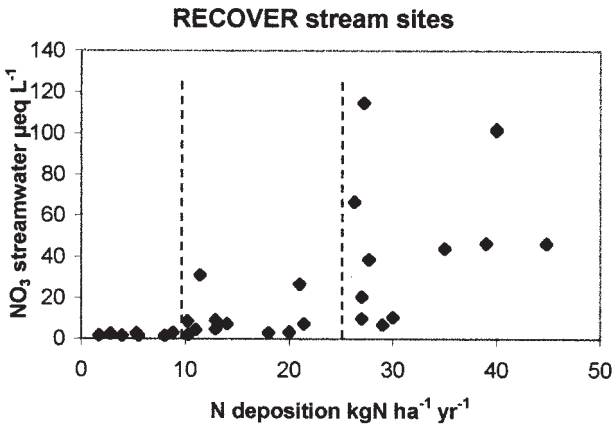

Fig. 9. *Mean concentration of NO_3 (µeq L^{-1}) in stream waters at 30 sites in Europe and inorganic N deposition (throughfall or bulk). Time periods covered by the data differ from site-to-site (see Table 2). The vertical dotted lines indicate minimum threshold of 10 kgN ha^{-1} yr^{-1} below which all streams have very low concentrations of NO_3 and the maximum threshold of 25 kgN ha^{-1} yr^{-1} above which all streams have elevated concentrations of NO_3 (thresholds from Dise and Wright, 1995).*

The concept of N saturation implies that terrestrial catchments have a finite capacity to store N deposited from the atmosphere. A larger and larger fraction of incoming N will be leached as the capacity is gradually depleted. Thus increasing trends in NO_3 concentrations in surface waters are expected in regions receiving excess N deposition.

The trend analyses presented here from other studies, however, do not show consistent regional patterns of increasing concentrations during the past 15-30 years. These empirical data from many sites in Europe show that the process of N saturation is very slow. This in turn implies that ecosystems not currently suffering from N saturation are under no immediate threat of increased NO_3 leaching.

This does not, however, mean that N saturation does not occur over the long term. Synoptic empirical data for forest stands in Europe compiled by Gundersen *et al*. (1998), Dise *et al*. (1998) and Gundersen and Kristensen (2001) show that stands currently receiving high deposition of N (above 10 kgN ha^{-1} yr^{-1}) have statistically significantly different characteristics relative to stands receiving less than 10 kg N ha^{-1} yr^{-1}. Among other factors, the C/N ratio of the forest floor (uppermost soil layers) is significantly lower at sites receiving high N deposition. There is also a significant correlation between C/N ratio and N leached from these sites.

Measures of pools and fluxes of N within temperate and boreal forest ecosystems shows that by far the largest amount of N from deposition is stored in the soil, at least over the short-term (Nadelhoffer *et al*., 1999). The implication is that as a site is exposed to chronic elevated N deposition, the N retained goes mostly to the soil and leads to a lower C/N ratio in forest floor. As the C/N ratio declines, less N is retained, and more goes to run-off, and appears as NO_3 in streamwater. But the trend data presented here clearly show that this evolution from N-limited to N-saturated requires at least several decades at N deposition levels typical for Europe.

N saturated sites leach significant amounts of NO_3 only if they receive substantial N deposition. Large-scale experiments with roofs to exclude acid deposition all show immediate and large decreases in NO_3 leached. Such experiments have been conducted at Risdalsheia, Norway (RAIN project: Wright *et al*., 1993; Wright and Jenkins, 2001), Ysselsteyn and Speuld, the Netherlands (NITREX project: Boxman *et al*., 1998) and Solling, Germany (NITREX project: Xu *et al*., 1998). The roof catchment experiments at Klosterhede, Denmark (Beier *et al*., 1998) and Gårdsjön, Sweden (Moldan *et al*., 2001a) are on N-limited systems. Prior to treatment these ecosystems all exhibited symptoms of N saturation with low C/N ratios in the forest floor and high concentrations of NO_3 in leachate. With exclusion of N deposition, the ecosystems stopped leaching significant amounts of NO_3, despite the fact that the systems were still full of N. Nitrogen deposition is thus a necessary factor in accounting for high NO_3 concentrations in surface waters. Together, the NITREX and roof experiments show that terrestrial ecosystems exhibit extreme hysteresis in run-off NO_3 response to N deposition; increased NO_3 concentrations come first after many decades of high N deposition, but decreases in concentrations come immediately following decreases in deposition.

In light of the quick response of the roof experiments, the recent 20% decline in N deposition in central and northern Europe should result in a commensurate decline in NO_3 concentrations in surface waters. Yet trend analyses at the 30 sites gave significant declining trends at only three sites, Metzenbach in central Germany, Gårdsjön F1 control in Sweden and Storgama in southernmost Norway. The recent compilation of trends in the 1990s from the ICP-Waters programme showed significant negative trends at only eight of 96 sites (Skjelkvåle *et al*., 2001b).

The overall lack of significant trends in NO_3 concentrations in streams in Europe may be the result of two opposing factors. Continued high deposition of N (above the 10 kgN ha^{-1} yr^{-1} threshold) should tend to increase N saturation and give increased NO_3 concentrations in run-off, whereas the decline in N deposition over the past 5–10 years in large parts of Europe should give decreased NO_3 concentrations in run-off.

Nitrate concentrations in streamwater often show large regular seasonal variations, with low concentrations during the growing season and high concentrations during the dormant season. The large intra-annual variations in the data contribute substantially to the "noise" from which the long-term trend "signal" is to be separated. Furthermore, the climatic factors that influence biological activity in the terrestrial ecosystem also affect processes involved in retention and release of N in the ecosystem. Thus, the record of NO_3 concentrations in streamwater will also be affected by short-term and long-term variations in climate and these variations will also mask long-term trends caused by changes in N deposition. Examples of short-term climatic variations are the cold winters followed by unusually high concentrations of NO_3 in streamwater in the spring; this occurred in 1996 at sites in the UK Acid Waters Monitoring Network (Monteith *et al*., 2000), and in 1990 at sites in the eastern United States (Mitchell *et al*., 1996). The CLIMEX experiment at Risdalsheia, Norway, a N-saturated site, showed that a permanent increase in temperature of 3-5°C results in a long-term increase in NO_3 concentrations in run-off (Wright, 1998; Wright and Jenkins, 2001).

The empirical data for geographic pattern and long-term

trends in response of surface waters to changes in N deposition such as the RECOVER:2010 sites described here, set the premises for predicting future concentrations of NO_3 and thus future contributions of NO_3 to acidification of soils and surface waters. Process-oriented acidification models such as MAGIC (Cosby *et al*., 2001) require such constraints so that the range of possible forecasts can be restricted. As SO_4 deposition and SO_4 concentrations in acidified surface waters continue to decline, the role of NO_3 in acidification will become greater. Quantification of processes governing N retention and loss in semi-natural terrestrial ecosystems is thus a scientific challenge of increasing importance.

Acknowledgements

The synthesis and analysis of results reported here was carried out as part of the RECOVER:2010 project (the Commission of European Communities EVK1-CT-1999-00018). The work was financially supported in part by the German Ministry of Education and Research (grant no. PT BEO 51-0339476), the Nordic Council of Ministers, the Research Council of Norway, and the Norwegian Institute for Water Research and the Natural Environment Research Council (UK).

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