

Effect of Increased Carbon Dioxide and Temperature on Runoff Chemistry at a Forested Catchment in Southern Norway (CLIMEX Project)

Richard F. Wright

Norwegian Institute for Water Research, Box 173 Kjelsås, 0411 Oslo, Norway

ABSTRACT

CLIMEX (Climate Change Experiment) is an integrated, whole-ecosystem research project that focuses on the response of forest ecosystems at the catchment scale to increased CO₂ and temperature. KIM catchment (860 m²) is completely enclosed by a transparent greenhouse, receives deacidified “clean” rain, and has elevated CO₂ (560 ppmv) and elevated air temperature (3°–5°C above ambient). The uppermost 20% of the catchment is partitioned off, is not subject to changed CO₂ or temperature, and serves as an untreated control. Fluxes of nitrate and ammonium in runoff from KIM catchment increased from 2 mmol m⁻² y⁻¹ each in the 3 years before treatment to 6 and 3 mmol m⁻² y⁻¹, respectively, in the 3 years after treatment (May 1994–April 1997), despite a 15 mmol m⁻² y⁻¹ decrease in N dry deposition due to the sealing of the walls to the enclosure. N flux in runoff from three reference catchments and the control section did not change. The net loss of inorganic N was thus about 20 mmol

m⁻² treated soil y⁻¹. There were no changes in organic N or total organic carbon in runoff. The ecosystem switched from a net sink to a net source of inorganic nitrogen (N). The increased loss of N may be due to accelerated decomposition of soil organic matter induced by higher temperature. Due to many decades of N deposition from long-range transported pollutants, the ecosystem prior to treatment was N saturated. If global change induces persistent losses of inorganic N on a regional scale, the result may be a significant increase in nitrate concentrations in fresh waters and N loading to coastal marine ecosystems. In regions with acid sensitive waters, such as southern Norway, the increased nitrate release caused by global change may offset improvements achieved by reduced sulfur and N deposition.

Key words: catchment; global change; forest; nitrogen; runoff; acid deposition; Norway.

INTRODUCTION

Emissions of CO₂ and other greenhouse gases to the atmosphere may lead to significant increases in global temperature over the next decades, and the increases are expected to be largest at high latitudes

(Houghton and others 1995; Greco and others 1994). Boreal forest ecosystems are particularly sensitive to temperature, and thus climate change is likely to have a large impact (Kirschbaum and others 1996). Northern ecosystems typically have large stores of carbon (C) and nitrogen (N) in the soil. On the one hand, higher concentrations of CO₂ may lead to increased plant growth and uptake of N, while, on the other hand, increased temperature

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e-mail: richard.wright@niva.no



Figure 1. Photo of the 1200-m² enclosure covering KIM catchment of the CLIMEX project. Insert Map of Norway showing the location of the site.

Table 1. Overview of the Five Catchments at Risdalsheia Included in the CLIMEX Project

Catchment	Area m ⁻²	Enclosure	Rain Quality	Climate Treatment	Monitor Start
KIM	860	Roof	Clean	CO ₂ + air warming	March 1984
EGIL	400	Roof	Acid	Soil warming	March 1984
ROLF	220	No roof	Acid	None	October 1984
METTE	650	No roof	Acid	None	May 1993
CECILIE	380	No roof	Acid	None	May 1993

The first three catchments were run by the RAIN project for 11 years (June 1983–May 1994). Clean rain treatment began in June 1984. CLIMEX treatment began in April 1994. KIM and EGIL were divided in April 1994 to an upper control section (KIM-c, EGIL-c, no climate change) and lower treatment section (KIM-t, EGIL-t, climate change).

may lead to enhanced mineralization of soil N and C. The net result and in particular the long-term effects are largely unknown, yet because of the extent of the boreal forest biome these effects can have major impacts on the global C balance (Kirschbaum and others 1996) as well as the N inputs to freshwater and marine ecosystems (Hessen and Wright 1993).

Large-scale whole-ecosystem experiments provide one of the few available tools by which the ecosystem response can be measured to evaluate global models (Mooney and others 1991; Pastor and Post 1988). CLIMEX (Climate Change Experiment) is an international, cooperative research project that focuses on the response of forest ecosystems at the catchment scale to increased CO₂ and temperature (Jenkins and others 1992; Dise and Jenkins 1995). The CLIMEX project began in December 1992, using the former RAIN (Reversing Acidification in Norway) project facilities and catchments at Risdalsheia, near Grimstad, Norway (Wright and oth-

ers 1993) (Figure 1). CLIMEX comprises two separate but linked experiments. At KIM catchment, a 1200-m² transparent greenhouse completely covers the forest ecosystem in which air temperature is raised 3°–5°C above ambient and CO₂ concentration is raised to 560 ppmv during the growing season (April through October) (Figure 1). At EGIL, catchment soil temperature is raised 3°–5°C by means of electric heating cables. Three adjacent catchments (ROLF, CECILIE, and METTE) serve as untreated references. Treatment began April 1994 (Table 1).

The flux of energy and matter across ecosystem boundaries gives an integrated measure of the whole-ecosystem response to environmental perturbation. For the CLIMEX project, changes in fluxes of water and solutes in runoff provide information on the catchment-scale effects of the experimentally altered CO₂ and temperature regime, and a direct measure of the impact of terrestrial change on aquatic ecosystems. A central objective of CLIMEX is to measure the chemical composition and fluxes of runoff at the two experimental and three reference catchments at Risdalsheia to detect changes induced by the experimental treatments. Here, I report results of 3 years of treatment at KIM catchment (CO₂ + temperature). Results after 2 years at EGIL catchment (soil warming) are reported by Lükewille and Wright (1997).

SITE DESCRIPTION AND METHODS

Site Description

The CLIMEX project is located at Risdalsheia (58°23'N, 8°19'E) near Grimstad, Norway, about 20 km inland from the North Sea. The site is 300 m above sea level and is typical of large areas of upland southern Norway. Risdalsheia has a maritime climate with mean annual precipitation of about 1400 mm, runoff 1200 mm, and mean annual temperature of about 5°C (mean of –3°C in January and +16°C in July). Vegetation is mainly a sparse cover

of pine (*Pinus sylvestris* L.) and birch (*Betula pubescens* L.), with heather (*Calluna vulgaris* L.) and blueberry (*Vaccinium myrtillus* L.) as dominant ground species. Bedrock is biotite granite covered by thin (about 10 cm) and patchy podsollic and peaty soils.

Risdalsheia receives high levels of acid deposition. Mean values of total deposition (wet and dry) for the 8-year period 1984–92 were 226 mmol m⁻² y⁻¹ S, 132 mmol m⁻² y⁻¹ N (59 as NH₄⁺ and 73 as NO₃⁻), and 106 mmol m⁻² y⁻¹ H⁺ (Wright and others 1993).

Experimental Design

In the winter of 1983–84, the forest ecosystem at KIM catchment (860 m²) was covered by a 1200-m² transparent roof above the canopy as part of the RAIN project (Wright and others 1993). Manipulation of the precipitation (termed “clean” precipitation) at KIM catchment began June 1984 and has continued uninterrupted to date. Incoming precipitation is collected from the roofs by gutters, led to storage tanks, filtered, passed through a mixed-bed ion-exchange resin, dosed with natural levels of sea salts (at about 1:8000 during the summer and 1:5000 during the winter), and automatically distributed back out under the roofs through a sprinkler system. During the winter, the sprinkling systems are not operated. From 1984–93 at KIM, catchment snow was made artificially by commercial snow-making equipment. Water from a nearby lake was ion exchanged and sea salts readded. Beginning with CLIMEX in the winter of 1993–94, there has been no snow cover in KIM. Instead, precipitation coming as snow outside is delivered as rain under the roof by the sprinkling system. At all times, care was taken to avoid disturbance of the vegetation and soils within the catchments.

In the winter of 1993–94, the catchments were further modified for the CLIMEX project (Dise and Jenkins 1993). KIM catchment was completely enclosed with airtight transparent walls, and a dividing wall was installed across the catchment such that the uppermost 20% of the catchment (KIM-c) received the clean rain treatment only and not higher CO₂ and temperature. KIM-c served as control area for plant and soil studies that were conducted on small plots of about 4–5 m². Runoff from KIM-c flowed into the treated 80%, and thus runoff from the treated section (KIM-t) contained water from both KIM-c and KIM-t.

Carbon dioxide is added to the air at six points inside the treatment section. The target concentration is 560 ppmv during the growing season (defined as 1 April–31 October) (Figure 2). Measurements are made by a central infrared analyzer.

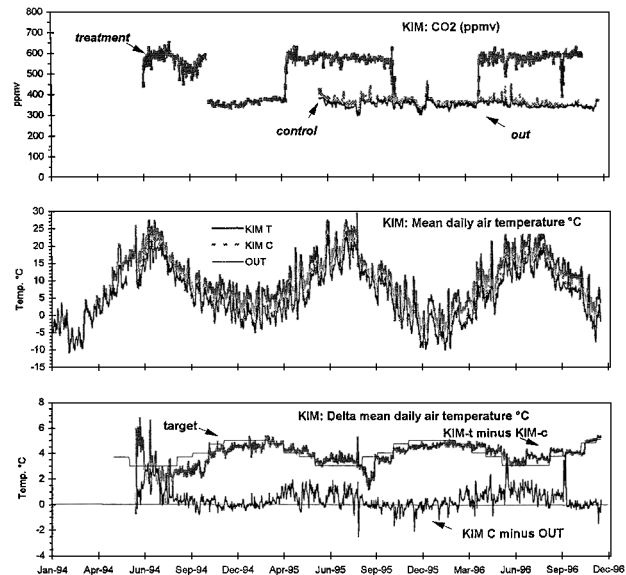


Figure 2. Mean daily CO₂ concentration (ppmv) and air temperature (°C) during the 3 years of CLIMEX treatments at KIM catchment. KIM-t, treatment section; KIM-c, control section.

Maximum dosing rate is 55 kg h⁻¹ (1.3 tons d⁻¹). CO₂ consumption is about 100 metric tons y⁻¹.

The air is warmed in the treatment section by means of a central electric heating system in which heated fluid (maximum 60°C) circulates through three ranks of 6-cm-diameter steel pipes mounted on the insides of the walls around the entire treated section. Temperature is monitored continuously at six points. The target for air warming is +5°C above KIM-c in January and +3°C in July, with intermediate temperatures in the intervening months (Figure 2).

Mixing of the air within both the treated and control sections is accomplished by means of six large fans each with maximum circulation capacity of 1000 m³ h⁻¹. Six fans are hung from the roof in KIM-t and two in KIM-c. Fan speed is adjusted continuously to maintain an even temperature within each section. Ventilation (cooling) is by means of windows mounted along the entire length of the four sides of the structure.

Precipitation

Ambient bulk precipitation is collected by funnel, and volume is measured weekly at the meteorological station located in an open area at Risdalsheia. At KIM catchment, the volume of sprinkled water is metered and read at minimum weekly intervals.

Chemical composition of ambient bulk precipitation is determined on samples collected weekly and refrigerated pending analysis. Samples are sent to

Table 2. Analytical Methods Used for Precipitation (P) and Runoff (R) Samples at the Norwegian Institute for Water Research

Parameter	Sample Type	Method	Detection Limit	Standard Error
pH	P, R	Potentiometry	0.01	±0.05
Na	P, R	Inductive-coupled plasma-emission spectroscopy	0.02 mg/L	±0.06
Ca	P, R	Inductive-coupled plasma-emission spectroscopy	0.02 mg/L	±0.01
Mg	P, R	Inductive-coupled plasma-emission spectroscopy	0.003 mg/L	±0.04
K	P, R	Atomic adsorption spectrophotometry	0.02 mg/L	±0.01
NH ₄ -N	P, R	Automated colorimetry	5 µg N/L	±5
NO ₃ -N	P, R	Automated colorimetry	1 µg N/L	±2
Cl	P, R	Ion chromatography	0.2 mg/L	±0.08
SO ₄	P, R	Ion chromatography	0.2 mg/L	±0.12
Reactive Al	R	Automated colorimetry	10 µg/L	±3
Organic Al	R	Cation exchange, automated colorimetry	10 µg/L	±3
TOC	R	Oxidation, spectrophotometry	0.2 mg C/L	±0.01
Total N	R	Oxidation, automated colorimetry	5 µg N/L	±5
Total P	R	Oxidation, automated colorimetry	1 µg P/L	±0.2
SiO ₂	R	Automated colorimetry	0.1 mg/L	±0.16
F	R	Potentiometry, ion-specific electrode	0.1 mg/L	±0.08

TOC, total organic C.

the Norwegian Institute for Water Research (NIVA) for analysis of pH, electrical conductance, Ca, Mg, Na, K, NH₄, NO₃, Cl, and SO₄ by methods summarized in Table 2.

Dry Deposition

Dry deposition is assumed to equal that estimated by Tørseth (1996) for the nearby EMEP station at Birkenes. These estimates are based on measured daily concentrations in air and assumed deposition velocities for S and N species. Species measured and deposition velocities (cm s⁻¹) are

SO ₂ gas	0.1 (winter) and 0.7 (summer)
SO ₄ particles	0.4
NO ₂ gas	0.3
HNO ₃ + NO ₃	2.0 for HNO ₃ and 0.4 for NO ₃ (25% was assumed HNO ₃ , 75% NO ₃)
NH ₄ + NH ₃	0.4 for NH ₄ and 0.4 for NH ₃ (92% was assumed NH ₄ , 8% NH ₃)

About 20% of dry deposition of S and 25% dry deposition of N are calculated to occur during the winter months (November–April) (Tørseth 1996). Dry deposition of marine aerosol is calculated by difference from the Cl flux in runoff minus that in bulk precipitation at each catchment for each year.

Runoff

Runoff from the catchments is collected at fiberglass dams at the bottom of each catchment, led by hoses to 0.5-m³ tanks that empty automatically when full.

Discharge is measured by logging the number of tanks emptied. Samples for chemical analysis are collected automatically at about weekly intervals from each full tank (or a preset selection of tanks). These systems at Risdalsheia have been in continuous operation since March 1984 (KIM), October 1984 (ROLF), and May 1993 (METTE and CECILIE) (Table 1).

Runoff samples are analyzed at NIVA for pH, electrical conductance, Ca, Mg, Na, K, NH₄, NO₃, Cl, SO₄, aluminum species (reactive Al and organic Al), total organic C (TOC), total N, total phosphorus (P), silica, and fluoride by methods summarized in Table 2. Organic N is defined as the molar difference of total N minus NO₃ minus NH₄. Acid-neutralizing capacity (ANC) is defined as the difference in equivalent sum of strong base cations (SBC: Ca + Mg + Na + K + NH₄) less the equivalent sum of strong acid anions (SSA: NO₃ + Cl + SO₄).

Flux Calculations

Fluxes are calculated for each winter and summer period. Start of winter is set at the date at which the sprinkling system was switched off due to frost (usually in November). Start of summer is set at the date at which snowmelt and any addition of extra precipitation are complete (usually in May). Precipitation inputs of chemical components at KIM catchment are calculated from the amounts of sea salts added assuming that the ion-exchanged water contains no chemicals.

Runoff fluxes are calculated from the chemical composition measured in the weekly samples and the volume of water discharged during the week. An interpolation routine is used by which daily chemical composition is calculated assuming linear change between weekly samples. Discharge during the week was assumed to be evenly distributed among the days.

RESULTS

Clean Rain Period: June 1984–May 1994

The CLIMEX treatment of increased CO_2 and temperature at KIM catchment is superimposed upon the previous 10 years of “clean” rain treatment during the RAIN project. The “clean” rain treatment removes all of the nonmarine sulfate, nitrate, ammonium, and acid from wet precipitation. Since June 1984, atmospheric deposition beneath the roof thus consists only of readded sea salts plus dry deposition of gases and particles beneath the roof.

Runoff chemistry at KIM catchment showed major changes in response to the drastically reduced acid deposition (Wright and others 1993). The largest absolute change was in sulfate concentration, which declined exponentially for about 8 years from about $111 \mu\text{eq L}^{-1}$ prior to treatment to a new level of about $28 \mu\text{eq L}^{-1}$ after about 8–10 years (Figure 3 and Table 3). Sulfate concentrations in runoff at the reference catchments also decreased somewhat in response to the general decrease in sulfate deposition in southernmost Norway, especially during the 1990s (Tørseth 1996).

The most rapid and largest relative changes in concentrations were the declines in nitrate and ammonium concentrations, which both dropped immediately in response to the drastically reduced N deposition (Wright and others 1994) (Figure 4). Nitrate went from $33 \mu\text{eq L}^{-1}$ to $3 \mu\text{eq L}^{-1}$ and ammonium from $17 \mu\text{eq L}^{-1}$ to $2 \mu\text{eq L}^{-1}$.

About 85% of the change in concentrations of strong acid anions (sulfate and nitrate) was balanced by increase in ANC, with the remaining 15% balanced by decrease in base cations (Figure 3). But because these waters have high levels of organic acids (TOC concentrations were 700–1500 mmol/L) (Figure 4), the increase in ANC gave rise to only a modest decrease in H^+ concentrations, from about $89 \mu\text{eq L}^{-1}$ to about $56 \mu\text{eq L}^{-1}$.

Chemical composition of runoff at KIM catchment appeared to have stabilized by about 1991. During the period June 1991–June 1994 (the last 3 years of the RAIN project and prior to onset of CLIMEX treatments), there were no major changes in the volume-weighted annual concentrations of

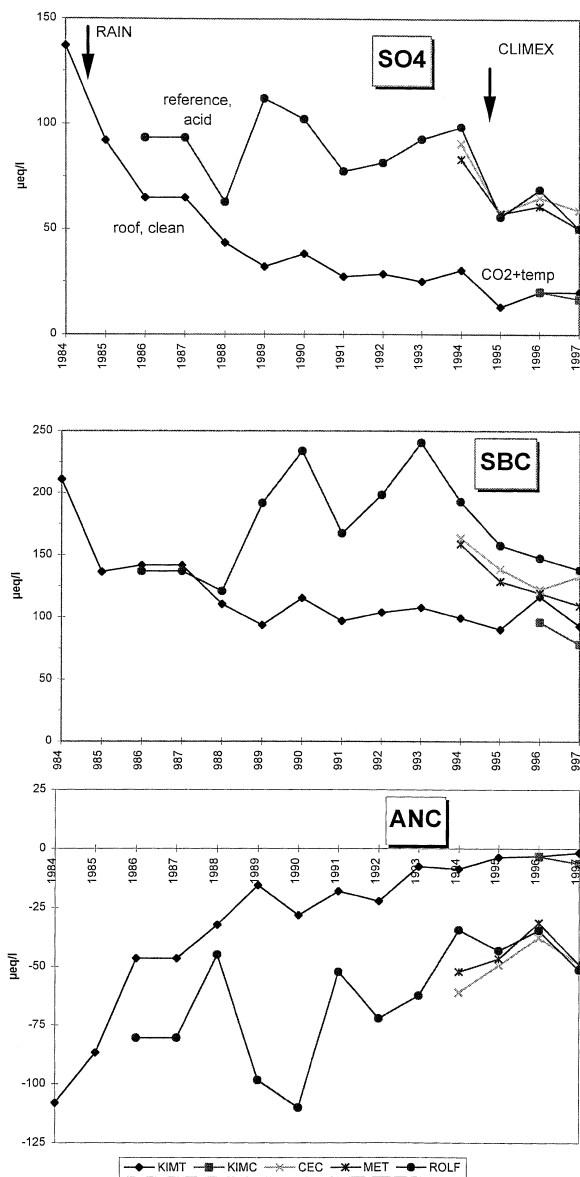


Figure 3. Volume-weighted mean annual concentrations of SO_4 , strong base cations (SBC), and acid-neutralizing capacity (ANC) in runoff from KIM catchment (clean rain, climate treatment), KIM-c (clean rain, ambient climate), and ROLF, METTE, and CECILIE catchments (references). The arrows indicate the start of the clean rain and climate treatments, respectively.

major components in runoff. The input-output budgets indicate that the catchment was still losing a small amount of old sulfate ($7 \text{ meq m}^{-2} \text{ y}^{-1}$), and was still a net sink of N ($14 \text{ meq m}^{-2} \text{ y}^{-1}$ N in and $4 \text{ meq m}^{-2} \text{ y}^{-1}$ out; net retention of 70%). The reference catchments were in approximate balance with respect to sulfate and also retained about 70% of the $85 \text{ meq m}^{-2} \text{ y}^{-1}$ incoming N (Table 3).

Table 3. Fluxes (Mean and Standard Deviation of Annual Fluxes) of Water and Major Chemical Components in Deposition (Wet Plus Dry) and Runoff (Out) at the CLIMEX Catchments

Fluxes		KIM Pre 3 Years (n = 3)			KIM-t Post 3 Years (n = 3)			KIM-c Post 2 Years (n = 2)			ROLF Pre 3 Years (n = 3)			ROLF Post 3 Years (n = 3)			ROLF, METTE, CECILIE Pre 1 Year (n = 3)			ROLF, METTE, CECILIE Post 3 Years (n = 9)						
		In	Out	SD	In	Out	SD	In	Out	SD	In	Out	SD	In	Out	SD	In	Out	SD	In	Out	SD				
H ₂ O	942	92	831	134	1064	287	861	258	901	712	1147	92	961	99	1153	280	1003	226	1144	18	956	59	1152	245	943	213
H ⁺	14	4	47	11	0	0	46	12	0	30	63	7	80	5	49	11	74	18	60	1	72	2	49	10	67	18
Na	61	16	65	12	64	11	60	17	54	47	126	37	139	18	95	36	99	31	83	5	113	18	80	32	86	29
K	1	0	3	1	1	0	3	1	1	2	5	1	9	7	3	1	5	2	4	0	7	1	3	1	5	2
Ca	3	2	6	1	3	1	6	1	2	4	11	3	14	2	9	1	12	3	10	1	13	1	9	1	10	3
Mg	15	4	11	2	14	3	12	2	11	9	29	8	28	6	23	7	24	5	19	1	21	3	19	6	19	6
Al	0	0	3	1	0	0	2	1	0	2	0	0	7	2	0	0	5	1	0	0	7	2	0	0	6	2
NH ₄	5	0	2	1	0	0	3	1	0	1	44	9	12	8	37	9	10	4	50	1	11	4	37	8	7	3
NO ₃	9	0	2	1	0	0	6	3	0	1	56	8	21	10	49	9	23	8	58	1	29	1	48	8	20	7
Cl	72	19	72	19	70	17	67	22	57	52	147	44	147	44	112	42	112	42	96	5	96	5	94	36	94	36
SO ₄	16	3	23	3	7	2	15	1	6	13	82	11	88	17	64	15	58	13	79	2	87	11	62	13	55	13
A ⁻	3	4	39	7	4	6	45	11	6	29	-7	2	34	9	-8	2	36	11	-7	1	33	10	-8	1	31	9
Sum+	100	26	137	28	81	15	132	32	69	95	278	53	291	42	216	60	228	62	227	9	244	27	196	52	200	59
Sum-	100	26	137	28	81	15	132	32	69	95	278	53	291	42	216	60	228	62	227	9	244	27	196	52	200	59
SBC	86	23	86	16	81	15	84	20	69	62	215	47	203	39	167	51	149	43	166	8	165	27	147	44	127	40
SSA	97	22	97	23	77	19	87	21	63	66	285	53	256	46	224	59	193	53	233	8	211	17	204	51	168	51
Alk	-11	1	-11	9	4	6	-3	2	6	-3	-70	5	-53	16	-57	10	-43	13	-67	0	-47	10	-57	9	-42	13
TOC			1008	134			1057	175		717			984	25			965	194			855	160			858	174
Reactive Al			8	2			7	2		5			9	0			8	2			10	1			8	2
Organic Al			7	1			6	2		5			7	1			6	1			7	1			6	1
Total N			26	5			33	6		23			67	4			50	22			63	6			45	14
Organic N			22	7			23	10		21			33	15			17	10			24	1			18	4
Inorganic N			14	1			4	2		2			100	17			33	12			40	5			16	27

SBC, strong base cations; SSA, strong acid anions; TOC, total organic C. For 3 years before treatment (14 June 1991–26 May 1994) and 3 years after treatment (27 May 1994–28 April 1997). Units: meq m⁻² y⁻¹; water mm y⁻¹; total organic C (TOC), reactive Al, organic Al, total N, mmol m⁻² y⁻¹.

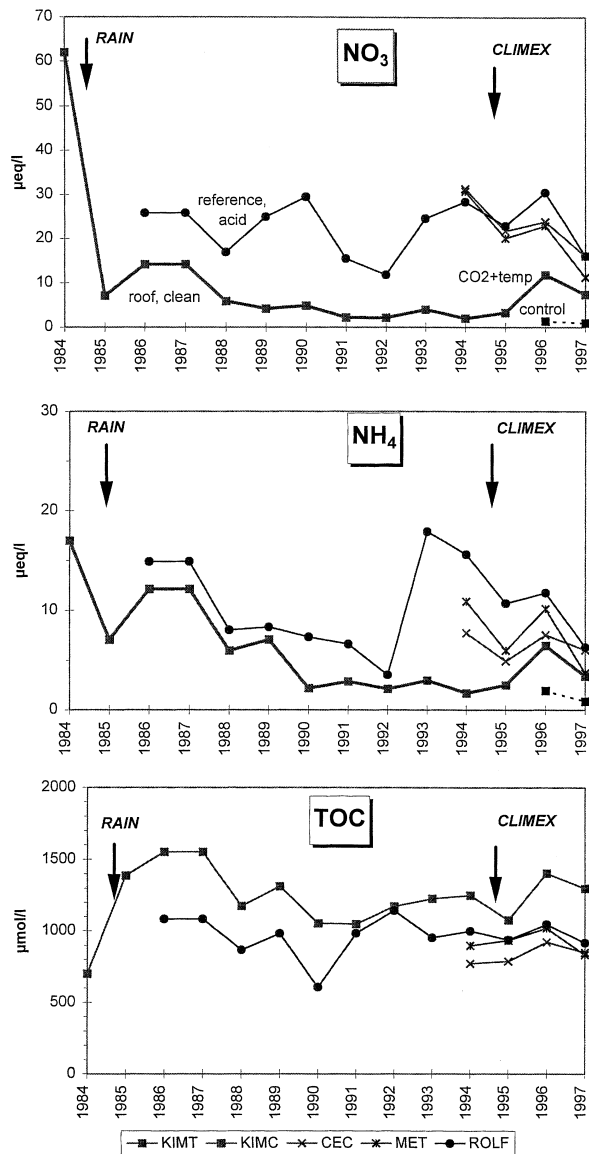


Figure 4. Volume-weighted mean annual concentrations of NO_3 , NH_4 , and total organic C (TOC) in runoff from KIM catchment (clean rain, climate treatment), KIM-c (clean rain, ambient climate), and ROLF, METTE, and CECILIE catchments (references). The arrows indicate the start of the clean rain and climate treatments, respectively.

CLIMEX Period: June 1994–April 1997

Further changes in runoff chemistry started in June 1994 in response to the onset of increased CO_2 and temperature at KIM catchment. Sulfate concentrations and flux in runoff decreased from $23 \text{ meq m}^{-2} \text{ y}^{-1}$ to $14 \text{ meq m}^{-2} \text{ y}^{-1}$ (Figure 3 and Table 3), probably because of the exclusion of dry deposition due to the installation of airtight walls around the greenhouse structure. After June 1994, sulfate depo-

sition consisted almost entirely of sulfate readed as sea salts to the wet precipitation. That the decrease in sulfate concentrations was not due to the CO_2 and temperature was indicated by the fact that sulfate levels in runoff from the untreated control section (KIM-c) were not different from the sulfate levels from the treatment section (KIM-t). Concentrations of nonmarine sulfate in runoff from KIM-c and KIM-t have reached levels typical for remote unpolluted lakes such as those in central and northern Norway (Skjelkvåle and others 1996).

Both nitrate and ammonium concentrations in runoff at KIM catchment increased following onset of the CLIMEX treatment (Figure 4). During the 3 years prior to treatment (June 1991–May 1994) volume-weighted annual average concentrations of NO_3 were $1\text{--}3 \mu\text{mol L}^{-1}$, whereas during the 3 years of treatment (June 1994–May 1997) concentrations were $3\text{--}12 \mu\text{mol L}^{-1}$. There was a significant increase in frequency of samples with NO_3 concentrations above $6 \mu\text{mol L}^{-1}$. Samples with elevated NO_3 concentrations were from all seasons, but higher values were generally measured during the non-growing season. Annual flux of NO_3 in runoff was $2.3 \text{ mmol m}^{-2} \text{ y}^{-1}$ for the 3 years prior to the start of the CLIMEX treatments, and $5.8 \text{ mmol m}^{-2} \text{ y}^{-1}$ for the 3 years of treatment (Table 3). At KIM-c concentrations and fluxes were low (Figure 4 and Table 3). No pretreatment data were available for KIM-c, as the division of the catchment was first made at the onset of the CLIMEX treatment in April 1994.

The concentrations and fluxes of ammonium at KIM catchment also increased after the onset of treatment (Figure 3). At KIM catchment as at all other Risdalsheia catchments, NH_4 concentrations were generally lower than NO_3 concentrations. The observed increase in NH_4 values occurred in samples from all seasons, and the frequency distribution of concentrations was significantly different than the pretreatment period. Annual flux of NH_4 in runoff was $1.9 \text{ mmol m}^{-2} \text{ y}^{-1}$ for the 3 years prior to the start of the CLIMEX treatments, and increased $3.3 \text{ mmol m}^{-2} \text{ y}^{-1}$ for the 3 years of treatment (Table 3). At KIM-c, concentrations were low (Figure 4 and Table 3).

At KIM catchment, during the 10-year RAIN project period (1984–94), the roof and clean rain treatment substantially reduced the input of inorganic N from about $130 \text{ mmol m}^{-2} \text{ y}^{-1}$ to about $14 \text{ mmol m}^{-2} \text{ y}^{-1}$ (mostly dry deposition remaining), and the catchment retained about 70%, with a runoff flux of only about $4 \text{ mmol m}^{-2} \text{ y}^{-1}$. At the start of CLIMEX treatment in April 1994, the walls were closed, and N inputs decreased further as a portion of dry deposition was excluded. The remain-

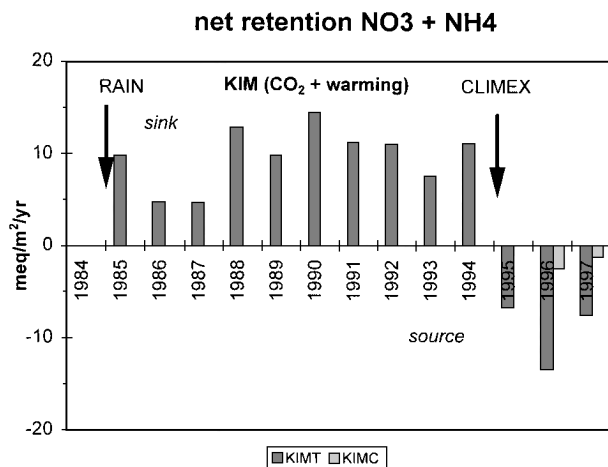


Figure 5. Annual net retention (deposition minus runoff) of inorganic nitrogen ($\text{NO}_3 + \text{NH}_4$) at KIM-t and KIM-c catchment. The arrow indicates the start of the climate treatment.

ing amount of dry deposition is difficult to assess and is assumed to be negligible. Despite the lower inputs, the flux of N was higher during the 3 years of treatment and was also markedly higher relative to KIM-c (Table 3). Input-output budgets indicate that the catchment has switched from a net sink of inorganic N (70% retention) to a net source. Whereas during the 3 years prior to CLIMEX treatment the catchment had net retention of $10 \text{ mmol m}^{-2} \text{ y}^{-1}$ inorganic N, during the 3 years of treatment the catchment showed net loss of $8.5 \text{ mmol m}^{-2} \text{ y}^{-1}$ (Table 3 and Figure 5). KIM-t is downstream of KIM-c, and thus runoff at KIM-t is a mixture of runoff from 170-m^2 untreated and 680-m^2 treated area. The net loss of N per square meter of treated catchment was thus about $10.6 \text{ meq m}^{-2} \text{ y}^{-1}$. Further, since the catchment is about 50% bare rock, the net loss of N per square meter of treated soil was about $20 \text{ meq m}^{-2} \text{ y}^{-1}$.

Together, the net effect of these changes has been a slight increase in ANC in runoff for the 3 years of treatment (Figure 3 and Table 3). Because the decrease in sulfate has more than offset the increase in nitrate, the sum of strong acid anions has decreased. The sum of base cations has not changed.

There have been no other significant changes in concentrations or fluxes of major ions, organic N, total organic C, or total P that can be ascribed to the CLIMEX treatment.

DISCUSSION

The experimental design of CLIMEX (and the previous RAIN project) takes the paired-catchment approach, in which one catchment is manipulated and

one or more otherwise similar catchments were monitored as untreated references. This experimental approach has the clear advantages of (a) large scale, (b) the whole ecosystem is manipulated, and (c) the effects of the treatment can be followed through the ecosystem to the runoff. The latter is essential for assessment of the impacts of runoff and downstream aquatic ecosystems. The paired-catchment approach has clear drawbacks, however, such as the lack of replicates for statistical purposes.

For the climate change experiment at KIM catchment, there were several ways by which changes in the chemical composition of runoff and input-output budgets can be assessed. The posttreatment data can be compared with the 10-year pretreatment data, and the data from KIM-t can be compared with those from KIM-c. There is no true whole-catchment untreated control for the CLIMEX experiment at KIM catchment because KIM is the only catchment that has received clean precipitation since June 1984. The conclusion that the CLIMEX treatment has caused increases in nitrate and ammonium in runoff is supported by the higher volume-weighted annual concentrations in the 3 years of treatment relative to the 3 years prior to treatment (Figure 4) and a shift in the annual N flux from net retention to net sink (Figure 5). Although the differences between pre-treatment and post-treatment data might be due to natural variations in factors such as hydrology, the fact that the changes have persisted over 3 years and have not been seen in the reference catchments strongly suggests that they were related to the CLIMEX treatment.

Support comes also from the comparison between KIM-c, the uppermost 20% of the catchment that has ambient climate, with KIM-t, the downstream 80% of the catchment that has been manipulated. Runoff samples from KIM-c consistently have lower concentrations of both nitrate and ammonium relative to KIM-t (Figure 4), while the concentrations of most other components were quite similar (Figure 3). Unfortunately, there were no samples from KIM-c prior to the onset of the climate change treatment.

The increase in concentrations and runoff fluxes of inorganic N points to an increase in decomposition of soil organic matter due to the $3^\circ\text{--}5^\circ\text{C}$ increase in temperature. Higher temperature generally causes an increase in the rate of decomposition, both of new litter and older soil organic matter (Stanford and others 1973; Edwards 1975). This decomposition could potentially release a number of chemical compounds to soil solution, in particular NO_3 and NH_4 . Laboratory incubations of soils from KIM catchment indicated increased mineralization and

release of nitrate to leachate at increased temperature (P. Verburg personal communication).

In N-limited ecosystems, the N released by increased mineralization would most likely be removed from soil solution through microbial mobilization and uptake by plants. Little or no inorganic N would appear in runoff. In N-saturated systems, the N released should appear, at least partially, in the form of nitrate and ammonium in runoff, but this would come on top of an already high and noisy signal of N concentrations and thus be difficult to detect unless sufficiently large.

KIM catchment is in the unusual position of being a N-saturated ecosystem at which N inputs have been reduced to zero. N output is very low, and yet the ecosystem is still full of N. C/N molar ratio in soil is only about 20. Prior to the start of "clean" rain treatment in 1984, runoff loss of inorganic N was about 30% of N deposition, a fraction similar to that at the reference catchments at Risdalsheia. New sources of N, either as N deposition or increased mineralization, thus give rise to increased N in runoff. But because the pre-treatment N concentrations in runoff were very low, the increased release of N is relatively easy to detect.

That increased temperature results in increased release of N is also indicated by the soil-warming experiment at EGIL catchment (Lükewille and Wright 1997). This experiment has run in parallel with the CO₂ + air-warming experiment at KIM catchment. At EGIL catchment, soil warming has caused statistically significant increases in NO₃ and NH₄ concentrations relative to the reference catchment ROLF. These increases can be explained by increased decomposition of soil organic matter due to the higher soil temperature (Lükewille and Wright 1997; P. Verburg personal communication). The soil-warming experiment at EGIL catchment is one of nine such experiments using heating cables in temperate and boreal ecosystems, although only CLIMEX entails treatment at the catchment scale. At all sites, there were indications of increased mineralization of organic N in the soil (Lükewille and Wright 1997).

The treatment at KIM catchment warmed the air as well as the soil, whereas at EGIL only the below-ground part was affected. At KIM catchment, both the increased temperature and increased CO₂ tend to promote plant growth and thus plant demand for N. The CO₂ enrichment and higher temperatures at KIM-t have resulted in a significant increase in biomass of *Calluna* and *Vaccinium*, as indicated by a greater number of leaves on first-year branches and increased thickness of the branches (W. Arp personal communication). N released from

enhanced decomposition might therefore be in part taken up by plants and not appear in runoff. At EGIL catchment, on the other hand, the air is not affected and plant growth has not increased except for species with bulbs or other below-ground growth points.

The changes in runoff chemistry caused by the CLIMEX treatment at KIM catchment suggest that the internal cycling of nutrients has been moderately affected during the 3 years of treatment. Further years of treatment will reveal whether the increases in chemical fluxes in runoff are permanent or merely transient.

Although the changes in N cycling may be small with respect to the forest ecosystem, the increased concentrations of NO₃ and NH₄ may have significant impact on aquatic ecosystems downstream. At KIM catchment, the fluxes of NO₃ rose from an average 3 mmol m⁻² y⁻¹ during the 3 years before treatment to 7 mmol m⁻² y⁻¹ for the 3 years of treatment. NH₄ rose from 2 to 4 mmol m⁻² y⁻¹ (Table 3). Since the treated area is only 80% of the entire catchment, the actual increases were about 5 and 2.5 mmol m⁻² y⁻¹, respectively. Further, since the catchment is about 50% bare rock, the increase per square meter of soil is about 10 and 5 mmol m⁻² y⁻¹, respectively. The rise in NO₃ represents a significant contribution to the concentrations of nonmarine strong acid anions and offsets to a large extent the decrease in nonmarine sulfate concentrations caused by the exclusion of dry deposition.

That increased temperature results in increased flux of NO₃ to streams and rivers has been shown by comparison of warm years with cold years. The effect may shift the seasonal pattern of NO₃ flux in streams and rivers, as was the case in southern Norway in the late 1980s and early 1990s (Lydersen 1995; Hessen and others 1997). The effects of warm summers and droughts can persist over several years, as was the case at Plynlimon, Wales, in the mid-1980s (Reynolds and others 1992).

Coastal marine waters are the ultimate recipient of N in runoff, and an increased N supply due to climate change can potentially cause a significant increase in N loading to these ecosystems (Hindar and others 1989). Climate change may thus exacerbate N pollution problems caused by acid deposition and emission of effluents from other anthropogenic sources (Gulbrandsen and others 1990; Hindar and others 1989).

If the CLIMEX results are extrapolated in time and space, large regions of southern Norway and other parts of northern Europe and eastern North America potentially can be adversely affected by N release. It is as yet unknown whether a long-term

increase in temperature due to global change will result in widespread rise in N leaching due to increased mineralization of soil organic matter. And it is also unknown whether increased leaching of N will be limited to those ecosystems that receive significant N deposition. Certainly the organic soils typical of the boreal region contain N stores equivalent to many decades of N deposition, and loss of only a small fraction can cause significant effects on soils and waters (Hessen and Wright 1993). To date, the whole-ecosystem manipulations of CLIMEX provide the only catchment-scale experimental evidence bearing on these questions.

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